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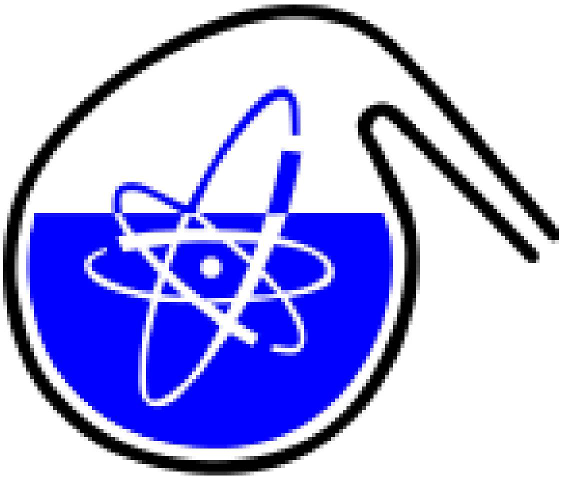

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JRNC Anniversary Session / 870

Fifty years of the Journal of Radioanalytical and Nuclear Chemistry

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Plenary 1 / 704

Superheavy elements of the D.I. Mendeleev's Periodic Table - Present and future

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The synthesis of super heavy elements (SHE) with atomic numbers 113-118 has been achieved in the 48Ca-induced reactions. The seventh period of the Periodic Table has been completed. Unique data on the nuclear-physical properties of isotopes of the new elements have been obtained. Pioneering experiments on the chemical properties of elements 112 (Cn), 113 and 114(Fl) have been carried out 1.
The discovery of an Island of stability of SHE has brought up a whole new series of questions:
• Can there be nuclei heavier than synthesized super heavy nuclei?
• Is this newly discovered "island of stability of SHE" the last one on the map of nuclides?
• Where is a limit of the Periodic Table?
• To what extend the chemical properties of SHE resemble those of their lighter homologues? And many others.
As a response to this challenge, a new project has been launched at FLNR (JINR) –"SHE Factory" 2, comprising: the construction of a new heavy ion accelerator produced stable and radioactive isotopes with masses A = 10–100 and intensities up to 10 pμA. The new laboratory will be equipped with target materials, new separators and detectors for the study of the nuclear, atomic and chemical properties of the new elements.

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Plenary 1 / 871

Towards increased cooperation and coordination of R&I activities for decommissioning

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CEA (French Alternative Energies and Atomic Energy Commission) is both the operator of important nuclear facilities all over the nuclear cycle, in charge of major new built or D&D projects and a R&D group with dynamic policy of technology transfer. The position of CEA in D&D is unique because of the number and the wide diversity of facilities under decommissioning, with some high level of contamination.
Innovative solutions are thus being developed in 6 main axes to protect the operators, to minimize the overall costs and the volumes of waste:

- Investigations in the facilities
- Radiological measurement of waste
- Robotics, laser cutting devices, virtual reality to simulate and choose the best way to operate,
- Decontamination of soils and structures
- Waste treatment and conditioning
- Methods and Information Technology (IT)Tools for project and waste management

CEA developed tools for its own D&D projects when they didn't exist yet on the market: robots, tele-operated equipment, cutting process and software for validation and optimization of intervention scenarios.

Concerning decommissioning and environmental remediation (D&ER), there appears to be a general consensus:

- Significant redundancy exists in current R&D programmes for D&ER being undertaken in different countries, though often no basis for industrial competition exists, e.g. on methods for treatment of exotic waste or for conditioning of intermediate and high activity waste;
- Project managers are often reluctant to use innovative technologies and need confidence from pilot operations with successful results to minimize project risks;
- Individual countries are finding it increasingly difficult to justify expenditure on developments that can require up to 10 years to complete; and
- Many possibilities exist to promote exchanges of information and lessons learnt but, beyond these, there is an urgent need to promote and organize at multinational level, the co-financing of R&D developments by actors with common objectives.

As a forerunner to the establishment of a framework for collaboration on R&D activities related to D&ER, it is proposed firstly to undertake a study to provide the following outputs:

- A mapping of the needs of D&ER projects, R&D programmes, human resources and platforms currently available;
- Identification of candidate topics for international financial collaboration, identification of actors involved with current sources of funding and rough business plans and schedules;
- Choice of financial and organizational model after benchmarking in other fields; and
- Proposal for an implementation schedule for projects that may begin immediately with varying timeframes for implementation, with the main focus being on projects taking place over the next 5-10 years.

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Environmental radioactivity research in Finland

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Nuclear weapons test fallout studies –radioecology competence developed
Environmental radioactivity in Finland is an established research field and has long traditions. Research started at the Department of Radiochemistry, University of Helsinki and at the predecessor of the Finnish Authority for Radiation and Nuclear Safety (STUK) in the last years of 1950s. Also

Finnish Meteorological Institute started monitoring of atmospherich radioactivity at the same time. The most important task at the time was to clarify the effects of radioactive fallout from the atmospheric nuclear weapons tests in the environment and food chains. A major scientific achievement from that period was the study on a critical radiocesium-enriching food chain from lichen via reindeer into reindeer-herding Sami people observed in Finnish Lapland. Fallout studies remained the largest field in environmental radioactivity studies in the 1960s to 1970s and were extended to environmental behavior of transuranium elements in the early 1970s.

Importance of indoor radon becomes evident
Early 1980s the researchers at STUK revealed the importance of indoor radon to radiation doses to humans and in the coming years radon research became more and more important. Today we know that indoor radon in responsible for more than half of the average radiation dose to Finnish population and measures to decrease its intrusion into dwellings have been developed at STUK. The finding was of utmost importance in developing radiation protection strategy and practices.

Fallout of the Chernobyl accident requires good competence
The competence developed in the weapons tests fallout studies became very important in 1986 when Finland was exposed to a radioactive fallout from the Chernobyl accident. In fact Finland received the largest fallout excluding the three closest countries to Chernobyl, Ukraine, Belorussia and Russia. The environmental surveillance and research department of STUK had considerably increased to about 60 persons of which 20 were researchers. Thus they had good resources to react to the accident which they very well did. Department of Radiochemistry, University of Helsinki, focused on the behavior of transuranium elements in the environment and, for example, together with STUK and the Finnish Meteorological Institute unique spatial distribution maps of Pu, Np, Am and Cm in Finland were developed. It is no exaggeration to say that no other country has such a deep knowledge on distribution and behavior of radionuclides from the Chernobyl fallout on their own territory.

National needs to study naturally occurring radionuclides
Through intensive fallout studies we have, however, learned to know that the radioactive pollution has only a minor effect on the radiation doses to humans. Eve in the most critical groups, such as reindeer herders in 1960's and most exposed groups after the Chernobyl accident, the doses from pollution radionuclides have been below that received from indoor radon on average. During the last twenty years or so, the importance of naturally occurring radionuclides has gained more attention in research in Finland. Studies like uranium series radionuclides of U, Ra, Po and Pb in the drinking water and in forest environment have tackled important national problems and considerably increased our knowledge in these areas. Latest achievements are studies on the behavior of naturally occurring radionuclides in the mining processes. Due to increasing mining activities and potential in Finland this field definitely needs further attention.

Studies on geological behavior of nuclear waste radionuclides brings new insight into environmental research
After constructing four nuclear power reactors in Finland in late 1970s and early 1980s geological behavior of radionuclides in relation to final disposal of spent nuclear fuel became a major research field in radiochemistry in Finland. This work, done to support safety analysis of final disposal, has been mainly carried out at the Laboratory of Radiochemistry, University of Helsinki, and the VTT Technical Research Centre, Finland. The radionuclides studied are largely the same as in traditional radioecological research, isotopes of Cs, Tc, Sr, I, TRUs etc., but also new radionuclides characteristic to spent nuclear fuel only, such as isotopes of C, Cl, Se, Mo, Nb, have been covered. Even though bedrock has been the main interest, compared to surface environment in traditional radioecology, the chemical and physical processes studied are largely the same. Main difference is that nuclear waste studies have been carried almost entirely using model experiments while in traditional radioecology actual environmental samples have been used.

Radiochemistry at the University of Helsinki in key role in educating experts
From the very beginning the environmental radioactivity studies have been mainly implemented by physicists and chemists. The chemists working in the field have been mainly educated at the Radiochemisty Department/Laboratory/Unit of the University of Helsinki. The unit has almost 60 years'experience in working in this field, so it has the required traditions and knowledge to educate new experts to the field. It has relevant facilities, including laboratories and instruments, to do work in this field. Radiochemistry education at the University of Helsinki has internationally a unique, extensive radiochemistry teaching program covering handling and measurement of radionuclides and behavior of radionuclides in the environment.
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Quo Vadis, neutron activation analysis?

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The opportunities of measuring the identities and amounts of chemical elements by measuring the activities induced by bombardment with neutrons was conceived more than 80 years ago. During the decades, practioners worldwide have been inspired by the challenges of this neutron activation analysis methodology, by the needs from end-users and by technological innovations in irradiation and measurement techniques. Examples will be given how this resulted in advancement of the development and use of neutron activation analysis at the Reactor Institute Delft .

It has been demonstrated that the methodology of neutron activation analysis is now fully understood, that a complete uncertainty budget can be made and that measured quantity values can be metrological traceable to the S.I. As a result, it has been worldwide increasingly acknowledged as valuable and even indispensable to (inter)national programs for metrology in chemistry; provided the technique is operated by metrological competent practioners.

The world's activation analysis community counts currently about 100 reactor-based neutron activation analysis laboratories and several tens of facilities using other neutron (and particle or photon) sources for activation. The number is decreasing for several reasons such as the lack of succession planning for retired people. Even if there is a succession plan: which challenges in the development and application of neutron activation analysis may inspire students to continue research on further development ; and in which applied fields we may call for awareness to prefer the technique above alternatives?

So the question is legitimate: quo vadis? How to go on, which road to take? Is there a road that inspires by technological or methodological advancement? Will there be –and preferably worldwide- requests for large scale measurement projects in which NAA would be methodology of choice? Or do we have to accept that the only passable path is the on which we already are; which means that we continue what we already were doing, accepting that there no new young generation will being inspired to take over?

Inspirations from the past, and considerations on a strategy to continue will be presented for further discussion during the conference.

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Nuclear Forensics –Where radiochemistry meets police work

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Nuclear forensic science is an evolving discipline which aims at re-establishing the history of nuclear material of unknown origin by using measurable material properties. The main focus of nuclear forensic science is on chemical and radiochemical parameters, such as isotope ratios, chemical composition, chemical impurities or products of radioactive decay for deriving information on the intended use of the material or its place and date of production. Such information may be relevant for non-proliferation, for countering nuclear terrorism, for combating illicit trafficking of nuclear

or other radioactive materials and for nuclear security in general. Obviously, this may involve law enforcement aspects as well as scientific aspects. In order to provide credible hints on the origin and the history of the material, useful parameters or a combination of several parameters (referred to as "nuclear forensic signature") need to be identified. The paper will review recent advances in method development and in the identification of relevant signatures. Moreover, the area of traditional forensic analysis of radioactively contaminated evidence will be discussed. Case studies (from real incidents) will illustrate the wealth of information that can be obtained through nuclear forensic investigations.

Plenary 2 / 683

Structures, properties and dynamics of uranyl peroxide cage clusters

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Since their discovery more than 10 years ago, the family of uranyl peroxide cage clusters has grown to more than 60 published members. These clusters, with diameters in the range of 1.5 to 4 nm, are soluble macro anions in water where they can persist for at least several years. This presentation will provide a brief overview of the structures of this family of polyoxometalate clusters, and then will focus on research conducted over the past two years that emphasizes cluster dynamics and solubility in solution, cluster fate under intense gamma irradiation, and the thermodynamic properties of these clusters. Potential applications in the nuclear fuel cycle will be discussed.

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GENIORS, a European Project on Advanced Fuel Recycling Strategies —Consolidating the Outcome of Preceding Projects

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The current open nuclear fuel cycle utilises only approximately one percent of the energy contained in natural uranium. Spent fuel recycling (as done e. g. in France) results in significantly increased fuel efficiency. Future multi-recycling strategies to be deployed in fast reactors will lead to further improvement, eventually rendering further uranium mining unnecessary if depleted uranium stockpile is available.

In this context and in the continuity of previous EURATOM projects (NEWPART, PARTNEW, EUROPART, ACSEPT, SACSESS), GENIORS addresses research and innovation in fuel cycle chemistry and physics. More specifically, GENIORS carries out research and innovation for developing compatible techniques for dissolution, reprocessing and manufacturing of innovative oxide fuels, potentially containing minor actinides, in a "fuel to fuel"approach, taking into account safety issues under normal and mal-operation. The most promising options developed in prior EURATOM projects are further developed to address the challenges specific to GEN IV. To deliver the full picture of a MOX

fuel cycle, GENIORS works in close collaboration with the INSPYRE project on oxide fuels performance.
By implementing a three-step approach (reinforcement of the scientific knowledge —process development and testing —system studies, safety and integration), GENIORS contributes to the provision of more science-based strategies for nuclear fuel management in the EU. It will support nuclear energy to contribute significantly and sustainably to EU energy independence. In the longer term, it will facilitate the management of ultimate radioactive waste by reducing its volume and radiotoxicity.

TAN 1 / 514

Superheavy Element Chemistry at GSI: Present and Future

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The coupling of chemistry setups to an electromagnetic separator is advantageous as this provides the necessary suppression of the primary beam and of products of multi-nucleon transfer reactions, as it was demonstrated in the first study on the carbonyl complex of Sg at GARIS (RIKEN, Japan) 1 and in experiments on Fl chemistry at TASCA (GSI, Germany) 2. The combination of physical and chemical separation increases the sensitivity in SHE chemistry experiments and allows almost background-free decay-spectroscopy measurements, e.g., with the novel ALBEGA setup [3,4].
Superheavy elements flerovium and nihonium are in the current focus of the chemistry experiments behind TASCA. Recent molecular, cluster, and solid-state relativistic calculations on Cn, Nh, and Fl have been performed and suggest these elements to be more inert than their lighter homologs; however chemical bond formation with gold is still expected [5,6]. In recent experiments, eight Fl atoms were observed in TASCA experiments devoted to the study of the interaction of Fl with SiO2 and Au surfaces. None of them were found adsorbed on the SiO2 covered detector, which was placed upstream to the Au covered detectors for a part of these studies. Five Fl atoms deposited on Au at room temperature, indicating a rather strong interaction with gold. Three Fl atoms, though, were registered at a very low temperature below dew point after transport over the entire gold surface to a thin ice layer. The interpretation of the observed interaction of Fl with Au will be presented.

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NFC 1 / 723

Lipophilic pyridine-2,6-bis(1H-1,2,3-triazol-4-yl) extractant for selective MA(III) extraction in SANEX-like processes

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With the perspective of improving nuclear energy fingerprint by reducing the waste volume and its long-term radiotoxicity, the Partitioning and Transmutation (P&T) strategy is being developed as a viable solution.[1, 2] Nowadays great efforts are constantly dedicated to the development of compact and efficient hydrometallurgical partitioning processes based on CHON compliant hydrophilic or lipophilic ligands. In Europe, the Selective ActiNide EXtraction (SANEX) process, together with its variants, allow to separate Minor Actinides (MA) from the high active raffinate downstream of a PUREX-like process.[3] Recently, different heteroaromatic nitrogen donor ligands, such as bis-triazine-pyridine/bis-pyridine/phenantroline (BTP, BTBP and BTPhen) ligands were developed showing a remarkable An/Ln selectivity, but most of them suffer of kinetic or stability problems in the harsh extraction condition.[4] In the last few years the pyridine-bis-triazole unit was found to be rather effective and selective in An separation from simulated nuclear waste.[3] In the present research work, the main features of the newly synthetized PyTri-Ethyl-Hexyl ligand were investigated in order to evaluate its potentialities in SANEX-like processes. Several liquid-liquid extraction tests were performed with a synthetic aqueous feed spiked with 241Am, in some cases 244Cm, and 152Eu as representatives of MA and Ln families respectively and, in some cases, containing also macro-concentration of stable lanthanides (Ln, about 2.4 g/L). All batch extraction experiments were performed by mixing aqueous and organic phases for one hour, since equilibrium proved to be achieved within 20 minutes. The ligand behavior was investigated as a function of the nitric acid concentration in the aqueous feed, the ligand and the 1-otcanol concentration in the organic solvent, in order to evaluate the impact of these fundamental parameters on the extracting properties. Preliminary information regarding the ligand radiochemical stability were obtained by performing liquid-liquid extraction tests and HPLC-MS analyses on solutions irradiated at 100 kGy by means of a Co-60 source or aged for several weeks, even in contact with 3 mol/L HNO3. The selectivity of the ligand core for MA was demonstrated, as well as its promising loading capacity. The promising results obtained justify further investigations on the ligand behavior, in order to better evaluate the selectivity and the stability towards hydrolysis and radiolysis.

This work has been partially supported by the Italian Ministry of Education, University and Research.

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TAN 1 / 874

Is there a future for liquid-phase super-heavy element chemistry?

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NFC 1 / 766

Investigations of diluent effect on europium extraction behavior with TODGA

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Extraction behavior of trivalent europium from nitric acid with N,N,N',N'-tetraoctyl-diglycolamine (TODGA) in different diluents was investigated. Systematic evaluation of diluent influence on the extraction behavior was performed. Sets of secondary alcohols, ketones and alkanes with increasing carbon chain from 5 to 10 carbons were used as diluents in solvent extraction experiments. Stoichiometry of extracted europium complexes formed in the organic phase was evaluated by slope analysis of the obtained distribution coefficients. Thermodynamic extraction data for extracted complex in chosen diluents were determined and the influence of diluent polarity on the extraction behavior was investigated. A significant increase in distribution coefficients of europium was observed when non-polar diluent was utilized in the experiments when compared to ketones and alcohols. Future work will be focused on the effect of extracted nitric acid into the organic phase on the extraction behavior.

NFC 1 / 624

Carbonate reprocessing of SNF in CARBEX-process

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CARBEX-process, the concept of which was formulated in 2008 1, is assigned to water methods and is an alternative one to the widely used PUREX-process. The developing method of uranium and mixed uranium-plutonium fuel processing is based on going from nitrate media widely used for purification of uranium and plutonium from FP and extraction to carbonate media.The significant difference of the CARBEX-process is carrying out all water processes in the absence of such a strong oxidizer as nitric acid, changing of solubility of all components involved in the process in carbonate media, as well as changing of the selectivity.

At present, the principle flowsheet of CARBEX-process includes the following steps: 1) high temperature voloxidation of SNF in an oxidizing atmosphere in presence of alkali metals carbonates; 2) oxidative (in presence of H2O2) dissolution of uranium, plutonium and some FP in solutions of alkali metals and/or ammonium carbonates; 3) the separation of a solid insoluble residue from the carbonate solution; 4) extraction of U(VI) from carbonate solution using a carbonate of methyltrioctylammonium (MTOA) as the extractant; 5) solid-phase re-extraction of carbonate compounds of U(VI) from saturated organic extracts; 6) re-dissolution of carbonate precipitation of uranium in Na2CO3 or (NH4)2CO3 solution in presence of H2O2; 7) extraction purification of U(VI) from FP, including Pu(IV) from carbonate solutions by the carbonate of MTOA; 8) solid-phase re-extraction of (NH4)4[UO2(CO3)3] from saturated extracts by concentrated solutions of (NH4)2CO3 and/or NH4HCO3; 9) oxidation of Pu(IV) to Pu(VI) in carbonate U(VI) raffinates with a suitable oxidizing agent; 10) extraction of Pu(VI) from carbonate solutions by carbonate of MTOA followed by extraction carbonate refining of Pu(VI) from FP and PuO2 obtaining.

Systematic works on classic of voloxidation of uranium SNF in Russia were carried out at the Radium Institute named by V.G. Khlopin and VNIINM named by academician A.A. Bochvar and published [2,3].

On studing of oxidative dissolution of the SNF simulator in Na2CO3+H2O2 solutions, it was found that uranium completely goes into solution. The following fission products: MoO3, Cs2O, CaO, I, Tc, Re are well soluble, ZrO2, BaO, SrO, Al2O3 are practically insoluble, and Y2O3, Ln2O3 (Ln = La, Nd, Sm, Gd, Dy), CeO2, Pd, Sb2O3, MnO2 poorly soluble in carbonate media. It was also found that the solubility of Pu, Am, Np, Cm oxides strongly depends on their oxidation degree.

In the presence of H2O2 U(IV) is oxidized to U(VI) and a peroxide carbonate compound Na4[UO2(O2)(CO3)2] is formed, whose solubility can reach up to 200 gU(VI)/l. At the same time, Pu(IV) and Np(IV) are not oxidized in the presence of H2O2. The solubility of PuO2 and NpO2 in Na2CO3+H2O2 solution does not exceed 10-50 mg/l. In the presence of other oxidants such as Na2S2O8, oxidation of Pu(IV) to Pu(VI) in Na2CO3 solution with formation of compound Na4[PuO2(CO3)3], take place concentration of plutonium reaches gram values. Oxidized Np(IV) (Np(VI)) is soluble in Na2CO3 solutions, forming a compound Na4[NpO2(CO3)3].
The concentration of REE in the core of dissolution of the oxides in Na2CO3+H2O2 solutions does not exceed 0,3 gLn2O3/l due to formation of the carbonate Na(n-3)[Ln(CO3)n], where n=2-5 and peroxide-carbonate compounds Na(m+n-3)[Ln(O2)m(CO3)n].
In the course of developing of the extraction part of CARBEX-process it was found that it is necessary to carry out two stages of extraction. The first stage –the extraction of U(VI) from carbonate solutions of oxidizing dissolution of uranium SNF, as the result of which U(VI) goes in organic phase together with some impurities of FP. The second stage is extraction purification of U(VI) from co–extracted impurities-extraction carbonate refining. In Table 1 presents data on the values of the coefficients of uranium(VI) purification from impurities simulators of some FP.

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Investigation of the chemistry of U(VI) extraction showed that, organic phase from carbonate solutions such compound were extracted: solvated by molecules of the extractant - (R4N)4[(UO2(O2)(CO3)2]•n(R4N)2CO3 and (R4N)4[(UO2(CO3)3]•n(R4N)2CO3, where R4N is a quaternary ammonium cation, n=1-2; nonsolvated: (R4N)4[(UO2(CO3)3] and (R4N)4[(UO2(O2)(CO3)2], polynuclear: (R4N)6[(UO2)2(O2)(CO3)4] and (R4N)6[(UO2)3(O2)2(CO3)4].
It was found that the effective separation of plutonium from U(VI) is possible for Pu(IV), the separation factor of U(VI)/Pu(IV) was 107. At the same time, Pu(VI) and U(VI) are effectively extracted together, which allows the development of variants of joint extraction of U(VI) and Pu(VI) in the CARBEX-process to prevent the production of pure plutonium dioxide.
Thus, within the framework of the CARBEX-process, it is possible both as separate and joint extraction purification of U(VI) and Pu(VI) from FP, followed by obtaining individual or mixed powders UO2 and PuO2, which can be sent to the production cycle of ceramic nuclear fuel based on UO2 or MOX fuel.
The work was supported by Mendeleev University of Chemical Technology of Russia. Project Number 008-2018.

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TAN 1 / **602**

Chemical properties of lawrencium (Lr, Z = 103): adsorption behavior on tantalum surface

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Recently we have determined the first ionization potential (IP_1) of lawrencium (Lr, $Z = 103$), which is the last member of the actinide elements, using an ISOL (Isotope Separator On-Line) equipped with a surface ion-source coupled to a He/CdI_2 gas-jet transport system at the JAEA tandem accelerator facility 1. A good agreement between the measured IP_1-value and the theoretical one, predicted by state-of-the-art relativistic calculations, strongly indicates that Lr would have a valence electron in the $7\mathrm{p}_{1/2}$ orbital. This results from strong relativistic effects in Lr, even though the 6d orbital is anticipated from the analogy to lutetium (Lu), the lanthanide homolog of Lr, which has the $[\mathrm{Xe}]4\mathrm{f}^{14}5\mathrm{d}6\mathrm{s}^2$ configuration.

It has been discussed that the Lr atom could possibly be much more volatile than other actinides and lanthanides since the valence electronic orbital of Lr is similar to that of group 13 elements 2. Surface ionization used in Ref. 1 involves the adsorption-desorption process of atoms on a surface, which characterizes the chemical stability of the atomic state and its reactivity. In this work, the adsorption behavior of elemental Lr on tantalum has been studied by using the surface ion-source as a vacuum chromatographic column.

Tantalum metal was selected as a working surface for surface ionization process, superimposed by adsorption-desorption processes. The short-lived Lr isotope ^{256}Lr ($T_{1/2} = 27$ s), produced in the reaction of ^{249}Cf with ^{11}B [3], was deliverd to the ion-source by the gas-jet transport system and was surface ionized. Ionized Lr was extracted by an extraction electrode and accelerated for mass-separation. The mass separated ions were collected and measured to determine apparent ionization efficiency (I_{eff}). In case of a sufficiently high temperature in the ion-source, the adsorption retention and subsequent decay loss of atoms on the surface can be neglected. However, at lower temperature, atoms may be retained on the surface according to its adsorption interaction and decay. In order to

deduce how many atoms are lost in the absorbed state inside the ion-source, we compared the measured apparent I_{eff} value with estimated one obtained from the Saha-Langmuir equation 4. The I_{eff} measurements of Lr and lanthanide isotopes were conducted in the temperature range from 2100 K to 2600 K. From the observed temperature dependence of I_{eff} of Lr and various elements, we found that Lr behaves as a reactive element with low volatility similarly to Lu and contrary to possible expectation based on its ground-state electronic configuration.
In the presentation, details of the experiments and the results will be presented.

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NFC 1 / 692

Closed nuclear fuel cycle based on the accelerator driven system

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With rapid growing of nuclear power plants in China, the safety management of spent fuel becomes most important issues. Accelerator Driven Sub-critical System (ADS) takes the spallation neutrons as external neutron source to drive the sub-critical reactor, which will provide the hard and wide neutron spectra, large flux and inherent safety. In order to increase the utilization rate of uranium resources and decrease the disposal of high high-level radioactive waste, the Accelerator Driven Used Fuel Recycle (ADFUR) was proposed by institute of Modern physics, Chinese Academy of Science1. The idea is only to remove the volatile fission products and to separate neutron poison such as rare elements partially from PWR spent fuel, the rest will be refabricated to new nuclear fuel in carbide form and burn in China Initiative Accelerator Driven System (CiADS). This reprocessing procedure will avoid separating magnanimous uranium and plutonium from spent fuel in contrast with the PUREX reprocess.
Disassembling, extraction, and cutting operations of actual spent PWR fuel assembly have been conducted in a laboratory scale for "Direct Use of PWR spent fuel In CANDU reactors"(DUPIC) experiment in KAERI in last two decades2. High temperature oxidation/reduction can pulverize spent fuel material from UO2 pellet to U3O8 powder in an air atmosphere and reduced into UO2 in a hydrogen atmosphere subsequently. The volatile gaseous nuclide such as 3H,14C,Kr, I, Xe and semi-volatile fission products such as Mo, Tc, Ru and Te in oxide form can be released from spent fuel material and selective trapped in a special filter. The rare earth elements which have a large neutron absorption cross-section were also considered to remove from the spent fuel material by means of the dissolution of the oxide of rare earth elements with the carboxy-functionalized ionic liquid. The rest of spent fuel in UO2 will be convert to UC by thermal-carbon reduction reaction and refabricate to the new nuclear fuel in carbide form and burn in China Initiative Accelerator Driven System (CiADS).

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TAN 1 / **744**

Microfluidic studies of SHE homologues in new facility at NPI Rez

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Exploration of the Super-heavy Elements (SHEs) is one of today's research frontiers that recently drew wide public attention after discoveries of four new elements. Although most chemistry investigations on SHEs use the "easier"gas-phase methodology, many fundamental properties can best be studied in liquid phase.
An experimental set-up assembled at the U-120M accelerator at the Nuclear Physics Institute CAS in Rez within its CANAM infrastructure based on the available equipment from the University of Oslo (target chamber and gas-jet (GJ) transfer system) and microfluidic liquid-liquid extraction (LLX) system from the Czech Technical University is utilized.
Aim of the work is to produce suitable isotopes of SHE homologues of interest, such as W, Mo or Zr, transfer them into aqueous phase, find proper and fast extraction agents and study their extraction behavior in various selected chemical systems. In the first experiments, extraction of W and Mo isotopes in the Cyanex 600/nitric acid system was tested.

NFC 1 / **575**

Spectrophotometric analysis of synergic systems to replace tri-n-butyl phosphate (TBP) in used fuel reprocessing

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When irradiated nuclear fuel is dissolved in nitric acid and contacted with tri-n-butyl phosphate (TBP) during PUREX processing, many reactions can occur. There is the potential to form a range of insoluble species, especially when solutions are concentrated by evaporation, as occurs prior to high activity liquor storage or vitrification. By replacing TBP with CHON alternatives, many of the possible drawbacks may be mitigated. N,N-dihexyloctanamide has previously been identified as a potential TBP replacement, although this can be hampered by low uranium distributions symptomatic of low complex solubility in the organic phase. Through the combination of synergic extractants, uranium recovery may be improved to the point that a CHON reprocessing flowsheet is feasible. In this report, the interactions between UO22+ and several small amide-based ligand mixtures in pseudo-aqueous media are investigated to determine whether potentially exploitable uranyl-multi-ligand complexes are generated. N,N-dimethylacetamide (DMAA), malonamide, N1,N1,N3,N3,-tetraethylmalonamide (TEMA), N1,N1,N3,N3,-tetraethyldiglycolamide (TEDGA) and N2,N2,N6,N6-tetraethylpyridine-2,6-dicarboxamide (TEPDCA) were used for this study. Small ligands were used to allow easier solubility in pseudo-aqueous media as well as reduce the currently unnecessary hindrances of sterics. Preliminary UV-visible spectroscopy screening suggests UO2-DMAA-TEMA and UO2-DMAA-TEDGA systems produce ternary complexes, whereas UO2-DMAA-TEPDCA systems do not. Comparison of malonamide and TEMA spectra suggest that ligand chelation plays a significant role in interaction strength. Spectrophotometric titrations were conducted

to determine likely stoichiometry and conditional stability constants of binary complexes of UO2 with DMAA, TEMA and TEDGA. Currently, spectra suggest that UO2-TEMA complexes appear to have two dominant stoichiometries dependent on ligand concentration. Jobs plot data suggests UO2 strongly prefers diamide complexes within UO2-monoamide-diamide systems. This effect has been attributed to the weak adduct formation of DMAA.

TAN 1 / 657

Diamond-based detectors for future chemistry experiments with superheavy elements

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Today's state-of-the-art alpha-decay- and SF-detection systems for artificially produced superheavy elements (SHEs) consist of silicon-based detectors. For recent chemistry experiments with SHEs a thermochromatographic channel comprised of silicon solid-state detectors in a sandwich-like geometry (e.g., Cryo On-Line Detector [COLD], Cryo Online Multidetector for Physics And Chemistry of Transactinides [COMPACT]) have been applied [1-5]. The crucial advantage of this method is that the surface of the detector serves directly as the stationary surface in the chromatographic process. Different detector coverages can be applied, e.g. SiO2, Au, Se. This technique allows for unprecedented detection efficiencies thereby giving access to crucial chemical information. Despite the success of this approach, the silicon-based detector setups can only be used for fairly volatile elements or their compounds with adsorption enthalpies ranging from −ΔHads ≈ 60 to 15 kJ/mol. The limiting factor here is the silicon material itself with its rather low band gap of 1.1 eV. As a result, the upper temperature limit for a reliable spectroscopic performance is encountered at around 40℃[6]. Furthermore, Si-based detectors need to be operated in complete darkness.
Due to its unique properties, diamond is a superior candidate for the replacement of silicon as semiconductor material for radiation detection. Diamond is a wide band gap semiconductor (5.4 eV), which allows diamond-based sensors to be operated theoretically up to 1000 ℃, even under intense UV/vis/IR-irradiation [7]. Currently, the upper temperature limit of diamond-based detectors for alpha-spectroscopic measurements is pinpointed at around 200 −300 ℃ [6, 8].
The application of diamond detectors is a promising prospect for a long sought after extension of the chromatographic range of detector arrays such as the COLD. Using diamond-based detectors with their before mentioned current temperature threshold, the upper limit of the observable adsorption enthalpy −ΔHads could be nearly doubled in comparison to today's detectors. In other words, this would give access to elements and compounds with stronger adsorption interactions with the chromatographic surface, such as to the one of Hg on Au. Such advancement is necessary for future chemical investigations of less volatile SHEs using the efficient method of thermochromatography.
Herein, we present a most recent investigation on heated diamond-based sensors aiming at highest possible operation temperatures for alpha-spectroscopic measurements. The prospects of their application in SHE experiments are discussed. Furthermore, we present a design study of a future COLD like detection system employing diamond-based detectors.

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NAM 1 / 678

Sensitivity of in-beam neutron activation analysis

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In-beam activation analysis, i.e. the combination of prompt gamma activation analysis (PGAA) using neutron beams from research reactors combined with neutron activation analysis (NAA), can in principle be performed at many neutron centers all over the world. Thanks to its strong beam flux (max. 6E10 cm^-2 s^-1), at Heinz Maier-Leibnitz Zentrum (MLZ), Garching, Germany it has been successfully implemented and used for several years now. The flux value almost compares with those in miniature neutron sources, or Slowpoke reactors, while the sample size can be much larger.
The great advantage of the technique is that in many cases the irradiation can be combined with a PGAA measurement yielding the major and minor components of the sample, in other words the composition of the matrix, thus any neutron self-shielding effects can be determined even for components having no or just weak radioactive daughter products (e.g. B, Li, or even Cd). There are components which can typically be determined using both PGAA and NAA and can be used as internal standards (e.g. Na, or Mn), so the method can be performed in a relative way eliminating several error sources. However, the absolute ways has also been performed successfully. Many short-lived nuclides which are hard to determine from the decay spectra, can be detected in the prompt spectra (like F, Al, V, etc.) with reasonable sensitivities. In-beam activations analysis does not compete with INAA for the detection of trace elements, its detection limits may be 1-2 orders of magnitude worse for the typical NAA elements, however it adds a set of nuclides, which are hard to reach with NAA.
At MLZ, the method has successfully been implemented with broadening the analytical capabilities of PGAA with the determination of many trace elements. Their sensitivities are compared with those of other typical facilities for activation analysis. The facility and a few applications are also presented.

SEP 1 / 732

Fluorinated diluents - progress and hope

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Usually extractants have been used as solution in diluents. Diluent influences on properties of extraction mixture very significantly. Hydrocarbons are traditional diluents. Unfortunately, hydrocarbons are low-polar compounds and therefore are not good as diluents for polar extractants.

Fluorinated diluents are possible alternative for hydrocarbons ones. Best studied polar fluorinated diluent: meta- nitrobenzotrifluoride (F-3, MNBTF) was used as diluent for chlorinated cobalt dicarbollide in industrial scale. It was used also for many polar extractants –from carbamoylphosphine oxide to calixarenes and amides of heterocyclic acids. It's disadvantages - rather high solubility in acid solution and toxicity (because it's nitroaromatic compound), were reduced by use of phenyl trifluoromethyl sulfone (FS-13, PMTS).
Fluorinated diluents without aromatic ring are more promising. Different classes of compounds - fluorinated alcohols, formals, esters and ethers of fluorinated alcohols were studied as diluents for crown-ethers, TBP, carbamoylphosphineoxides, amides of diglycolic acid etc.
The goal of the presented work is the review of history of fluorinated diluents use and discussion of possible ways of its future development.

SEP 1 / 608

Alkyl chains effects on uranium(VI) and plutonium(IV) speciation in organic phases containing tetra-alkylcarbamides

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The separation of uranium(VI) and plutonium(IV) by solvent extraction from spent nuclear fuels has already been studied with several families of extractants such as trialkyl phosphate, phosphine oxides or monoamides. A new family of extractant containing the carbamide function (also called urea with the general formula R1R2NC(O)NR3R4) is currently studied in our laboratory. Only few extraction studies of uranium(VI) and plutonium(IV) by carbamide extractants can be found in the literature. They show the high potential of those molecules to extract actinides(IV) and (VI). The objective of this study is to evaluate the effect of alkyl chains length on actinide extraction. Our results show that the chain length (from butyl to octyl chain) does not influence significantly uranium(VI) extraction while it has a strong impact on plutonium(IV) extraction. In order to better understand the relationship between the alkyl chains length and extraction performances, U(VI) and Pu(IV) complexes formed in the organic phase were characterized by spectroscopic techniques. First, to gain information on the coordination sphere of U(VI) and Pu(IV) complexes with carbamide, single crystals were synthesized with short alkyl chain carbamide (R1=R2=R3=R4= iso-butyl). An octahedron complex and a bicapped dodecahedron complex are obtained for UO2(NO3)2L2 and Pu(NO3)4L2 (with L: N,N,N',N'-tetra-iso-butylcarbamide) respectively. Then, the analysis of organic phases after uranyl extraction confirmed the participation of the carbonyl function and the nitrate anions denticity (bidentate) to uranyl coordination, and showed that the coordination sphere of uranyl is not modified by a change in the alkyl chains length of the carbamide. At the opposite, the speciation of plutonium(IV) depends on alkyl chain length of the carbamide and acidity of the solution. UV-Vis-NIR and EXAFS spectroscopy indicate that two different complexes are formed: a neutral complex Pu(NO3)4L2 and an anionic complex Pu(NO3)62- and their proportion varies with the length of the alkyl chain. The organic phase containing the higher proportion of anionic complex is associated to the higher Pu(IV) distribution ratio. These results suggest that the anionic complex is more extracted than the neutral complex and that increasing alkyl chain length enhances its organic phase solubility.

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Improving the Accuracy, Precision and Detection Limits for PGAA

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Prompt gamma-ray activation analysis has long been used for nondestructive elemental analysis, particularly of the light elements (e.g. H, C, N, S, P, Cl). However, the accuracy and precision of the method for elemental determination of hydrogenous materials is limited by the calibration of elemental sensitivities (cps/mg element), which vary with both neutron scattering power (i.e. hydrogen content) and target geometry. In addition, sensitivities for many of the low Z elements (C, N, S, P) are poor, yielding poor counting statistics without extremely long irradiations. We address these issues with these three approaches: 1) A combination of cold and thermal neutron PGAA continues to be used to improve both accuracy and precision of light element measurement in hydrogenous materials. CNPGAA yields higher signals due to the greater neutron capture cross sections for cold neutrons, but the absolute sensitivity varies as sample mass, a complication of a greater neutron scattering cross section. The latter issue is addressed by performing an absolute sensitivity calibration for selected matrix elements at the TNPGAA, where the scattering effect is minimal. This way, we take full advantage of the CNPGAA for efficient and precise determination of the sensitivity RATIOS, and improve the accuracy of the absolute elemental concentration via sensitivity calibration at the TNPGAA. The TNPGAA calibration essentially serves as a correction for scattering effects in CNPGAA. We have previously reported use of this method for determination of sulfur in fuel oil reference materials. More recent measurements include the determination of carbon at 12.5 % mass fraction in dolomite limestone reference material with an expanded uncertainty of 1.3 %, and C, N, S, and other elements in coal SRMs. Using both methods combined avoids the possible analytical bias from neutron scattering in CNPGAA and the poorer uncertainties from counting statistics from TNPAA. 2) The calculation of element sensitivities using tabulated k0 factors or partial capture cross sections and carefully measured detector efficiencies also serves as a useful check on elemental sensitivities measured from standards. CNPGAA measurements of C, H, and N in plastics using cross sections agreed with measurements made using comparator standards to within a few percent, indicating that the cross-section method may be used to verify measurements made using the standards comparison method, or for standard-free PGAA measurements when high precision is not needed. 3) The determination of gamma-ray background can also be a hindrance to detection limits, especially in the measurement of hydrogen. Measurements of ultra-trace amounts of hydrogen in materials require careful measurement of H in packaging materials as well as investigation of possible low-level interference peaks from other methods. Methods for studying these interferences are ongoing.

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Archaeometry with Prompt Gamma Activation Analysis, In-Beam Neutron Activation Analysis and Neutron Tomography at the PGAA instrument of MLZ

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The low-destructive nature of many neutron-based analytical techniques is a main argument for their application in archaeometry. Further advantages of these methods are the little sensitivity for the matrix composition, easy sample preparation and fairly low detection limits. A combination of two or more of these methods broadens the application possibilities. For instance, prompt gamma activation analysis (PGAA) and neutron activation analysis (NAA) are complementary techniques, so that combining them can significantly increase the number of the detectable elements. This is especially useful for studies needing as much information as possible like provenance research. At the PGAA instrument of the Heinz Maier-Leibnitz Zentrum (MLZ) in Garching (Germany) we use this approach e.g. for the provenance analysis of stone material from the UNESCO world heritage

site Lorsch Abbey –a unique remnant of the Carolingian age. For the comparison of the geochemical fingerprints multivariate statistic methods are used, which makes it possible to assign the findings to certain deposits. This information is useful for archaeologists who try to reconstruct ancient trade and transport routes and identifying re-used stones from old Roman (so-called spolia). It makes also sense to combine PGAA and neutron tomography (NT) to get information about both –the inner structure and the elemental composition of archaeological artefacts. It can help to shed light on ancient handicraft techniques, for instance. We recently investigate Celtic burial gifts in this context. In the presentation we will provide information about our recent projects, the results and the developments at the PGAA instrument of the MLZ.

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Simultaneous preconcentration of cadmium, cobalt, copper, manganese, nickel and zinc with 1-(2-Pyridylazo)-2-naphthol by cloud point extraction followed by neutron activation analysis

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A method for the simultaneous preconcentration of six environmentally relevant transition metals, namely cadmium, cobalt, copper, manganese and zinc, by cloud point extraction followed by their determination using neutron activation analysis (NAA) was developed. The method involved the use of a nonionic surfactant, namely PONPE-7.5, and the chelating agent 1-(2-pyridylazo)-2-naphthol commonly known as PAN for the extraction. Factors that can influence the extraction efficiency of metals, such as solution pH and PAN concentration, were evaluated. The irradiations were performed at the Dalhousie University SLOWPOKE-2 Reactor at a neutron flux of 5 x 1011 cm-2 s-1 in the inner pneumatic sites. Samples were irradiated in heat-sealed polyethylene irradiation vials. Two different irradiation-decay-counting (ti, td, tc) schemes were employed considering the half-lives and sensitivities of the elements of interest: 10 min, 1 min, 10 min for Cd, Co, Cu and Mn, and 3h, 1h, 2h for Ni and Zn. Quantitative recoveries (> 96%) were obtained for all the elements in a single extraction step under optimal conditions (pH 8-9, CPAN = 5×10-5 M). A preconcentration factor of 70 was achieved leading to detection limits of the order of nanogram per mL. The method was validated using NIST SRMs. The precision and accuracy of measurements were within ±10%. The CPE method developed here can be used to simultaneously preconcentrate trace metals from aqueous samples in only one-step reducing the possibility of contamination, while the use of NAA allows multielement determinations.

SEP 1 / 650

Production of Lutetium-177 via a solvent extraction based Lu-177m/Lu-177 radionuclide generator

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Lutetium-177 (¹⁷⁷Lu) is a radionuclide with tremendous potential in radionuclide therapy. ¹⁷⁷Lu based pharmaceuticals are applied in neuroendocrine tumour treatment and have been approved for prostate cancer treatment. We have proposed the idea of a ^{177m}Lu/<sup>177 radionuclide generator for 177Lu production based on the separation of the daughter nuclei, ¹⁷⁷Lu from its parent nuclei, ^{177m}Lu 1. It was followed by the proof of concept using a reversed phase chromatographic system 2. In our work, ^{177m}Lu-DOTA-(Tyr3)-octreotate (DOTATATE) complex was retained on tC-18 silica and the bond ruptured ¹⁷⁷Lu ions was collected using a mobile phase flow. Using this method of separation, ¹⁷⁷Lu/^{177m}Lu activity ratios were increased from 0.25 (in equilibrium) to values around 250. The ¹⁷⁷Lu was collected with efficiencies of about 65%. However in order to meet the clinical demands ¹⁷⁷Lu/^{177m}Lu activity ratios close to 10,000 is needed. In our current research we are exploiting the potential of solvent extraction in separating the two isomers.

In solvent extraction, ^{177m}Lu is complexed with different chelating agents such as 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (DOTA), DOTA-(Tyr3)-octreotate (DOTATATE) and others. The aqueous solution of complexed ^{177m}Lu is left at a fixed temperature for a fixed interval of time to allow the accumulation of ¹⁷⁷Lu. After the fixed accumulation period, it is stirred with an immiscible organic phase (dihexyl ether) containing an extracting agent, Di-(2-ethylhexyl)phosphoric acid (DEHPA) to collect the free ¹⁷⁷Lu, as shown in figure 1. Using this method of separation we are able to achieve ¹⁷⁷Lu/^{177m}Lu activity ratios close to 3000, with a ¹⁷⁷Lu collection efficiency close to 50%.

By optimizing the extraction process and by utilizing a microfluidic device we aim to deliver a system able to reach the quality needed for medical applications and to provide with a generator with ease of use that can provide with lutetium-177 for long periods of time on location.

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Geochemical characterization of Quaternary loess–paleosol sediments from Lingtai by instrumental neutron and photon activation analyses

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Quaternary aeolian sediments, particularly loess–paleosol sequences constitute some of the longest and most detailed records of Quaternary climatic change and valuable data archives for paleoenvironmental reconstructions. In the Chinese Loess Plateau in north-central China, the loess horizons represent arid periods of increased influx of wind-blown dust when the dust-transporting winter monsoon was dominant. The paleosol horizons represent semi-arid periods of reduced aeolian deposition and intensified pedogenesis reflecting a reduced strength of northerly winds and an increased intensity of the summer monsoon blowing from south. About 175 m thick section of Quaternary loess–paleosol sediments exposed near Lingtai in the southwestern part of the Chinese Loess Plateau represents a key locality which reveals a succession of 33 loess-paleosol levels overlying the Upper Miocene Red Clay Formation and going back to about 2.6 Ma. A pilot research conducted between 1996 and 2000 by a joint Czech-Chinese team focused on younger Last Glacial age deposits that form the uppermost 15 meters of the section resulted in collection of about 80 loess and paleosol samples and discovery of two distinctive geomagnetic excursions identified as the Laschamp and Mono Lake geomagnetic field excursions.

To reveal changes in paleoclimate which may have been associated with the previously discovered geomagnetic events, detailed geochemical characterization of the Last Glacial loess-paleosol sample suite from the Lingtai section, achieved by combined use of instrumental neutron and photon activation analyses, is presented. The obtained geochemical data provide also a supporting evidence for a recently proposed theory of origin of Australasian tektites (AAT). The theory assumes that AAT and Chinese loess may have had common source materials in the desert area of northwest China, and locates there a hypothetical parent crater for AAT.

The study has been supported by the Czech Science Foundation within the project P108/12/G108, and by the Ministry of Education, Youth and Sports of the Czech Republic within the projects LM2015056 (CANAM - Center of Accelerators and Nuclear Analytical Methods) and LM2015074 (Nuclear Research Reactors LVR-15 and LR-0).

SEP 1 / 489

Determining the stoichiometry of An(III)-DMDOHEMA complexes formed during extraction

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N,N'-dimethyl,N,N'-dioctylhexylethoxymalonamide (DMDOHEMA) is used to separate An(III) and Ln(III) from fission products in several liquid-liquid extraction processes (DIAMEX, EXAm, GANEX)$^{1-2}$ that aim at recycling actinides. One key to understand these processes is the stoichiometry of the complexes formed during extraction. Time-resolved laser fluorescence spectroscopy (TRLFS) poses, in contrast to extraction experiments, a technique to study the inner sphere coordination of fluorescent metal ions (e.g. Cm(III)). Therefore, the stoichiometry of the complexes can be determined. The presented work focuses on the complexation of Cm(III) with DMDOHEMA studied by TRLFS in mono- and biphasic systems. First, stability constants of the Cm(III)-DMDOHEMA complexes are determined in 1-octanol. The formation of $[\mathrm{Cm(DMDOHEMA)}_n]^{3+}$ with $\log\beta'_1$ = 2.63 ± 0.33, $\log\beta'_2$ = 3.99 ± 0.48, $\log\beta'_3$= 4.34 ± 0.52 is confirmed. Fluorescence lifetime measurements indicate the formation of a 1:4 complex. Furthermore, biphasic experiments are performed with Cm(III) and DMDOHEMA in dependence of the DMDOHEMA, nitric acid and nitrate concentration. TRLFS measurements of the different organic phases reveal a predominance of two species: (I) at λ_{max} = 601.3 nm, and (II) 604.2 nm. These are identified as $[\mathrm{Cm(DMDOHEMA)_3(NO_3)(H_2O)}]^{2+}$ (I) and $[\mathrm{Cm(DMDOHEMA)_4(H_2O)}]^{3+}$ (II) by comparison with the monophasic experiments. Species II is dominant at low acid concentrations. This trend is explained by the free DMDOHEMA concentration, which decreases due to the extraction of both nitric acid and water. To support the proposed stoichiometries, vibrational sideband spectroscopy is performed. This laser-based technique allows the observation of vibrations of functional groups coordinated to the fluorescent metal ion. Clear differences are obtained for species I and II in the range of 900 –1400 cm^{-1}. Moreover, vibrational spectra for species I and II calculated by density functional theory (DFT) are in good agreement with the experimental data, confirming the proposed stoichiometries.

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Poster SEP / 445

Physicochemical principles of self-sufficient decontamination arias from radioactive pollutants based on model of competitive adsorption

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A theory of concurrent sorption as an instrument for predicting the conditions for desorption of radionuclides, Sr(II) in particular, from "contaminated solid –water"systems is formulated in this presentation. Typical isotherms of concurrent sorption are presented. Based on the sorption isotherm we analyze the possibility of desorption of traces of Sr(II) and Sr-90 from ion-exchange materials. The role of humic acid (HA) in Sr(II) mass transfer in river water and the possibilities of strontium ion desorption from solids with the help of HA solutions are discussed in detail. We describe finally new competitive sorption techniques based on concurrent sorption of the aimed microelement from the contaminated material to the sorbent placed into a specially made pocketed membrane mini-reactor. The investigation is supported by RFBR, project 18-03-00051, and project of UBRAS 18-3-3-5.

Poster NAM / 439

Natural Radionuclides Analysis Using High Volume Liquid Scintillation Counter

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For the analysis natural radionuclides, gamma spectroscopy, alpha spectroscopy and liquid scintillation counter(LSC) were used. Among them, LSC was widely used because it can be applied alpha, beta and gamma emitting nuclides. But some low activity nuclides are not analyzed because most of the LSC used 20 mL vial. Therefore, some radionuclide, such as H-3, are needed concentration. To improve detection limit of the LSC, high volume LSC was used and performance was studied. LSC-LB7 was a unique detector structure with the vial up to 145 ml, three photomultiplier tubes, and reducing the effects of external radiation by the guard counter, anti-coincidence signal detection and massive layer of lead. Direct measurement is ensured without pre-processing, such as electrolytic enrichment, for ultra-low level H-3 measurement in the environmental samples. The counting efficiency, figure of merit(FOM) and MDA were compared conventional 20 mL vial used LSC. Counting efficiency was decreased due to the high volume but FOM and MDA was increased by using more than 7 times high volume used. The blank background was 2.18 ± 0.12 cpm when 145 mL vial was used. For the analysis of H-3, dead water and UltimGold LLT was used and 4.00 ± 0.29 cpm and 3.42 ± 0.97 cpm background were acquired when 145 mL and 20 mL vial was used.

Poster EDU / 864

New Master Course on FNSPE –Decomissioning of nuclear facilites

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The decommissioning of nuclear facilities (NF) has become more topical, primarily in Europe, but also in the USA and in Japan. This is mainly because the lifespans of the major parts of nuclear power plants are rapidly approaching their end, although in some cases the reasons for decommissioning are different (e.g. political). Another reason is that nowadays "immediate dismantling"is preferred which does not transfer the burdens connected to decommissioning to future generation. The strong demand for qualified experts specialized in decommissioning of nuclear facilities and in radioactive waste management has grown not only in the Czech Republic, but also in Europe.
The Faculty of Nuclear Sciences and Physical Engineering (FNSPE, www.fjfi.cvut.cz) is one of eight faculties of the public university - the Czech Technical University in Prague (CTU in Prague). The FNSPE is traditionally a research centre with activities which are supported by outstanding research staff. One of the strategic aims of the FNSPE is the education of experts for the Czech nuclear industry and for the state authorities which are dedicated to the peaceful employment of nuclear energy. The importance of these issues with which the FNSPE is strongly associated can be proven by the ongoing introduction of new all-faculty five-year specialized branch "Decommissioning of nuclear facilities". This activity is supported by the ESF/ERDF project of the Ministry of Education, Youth and Sports. By creating this new programme the Faculty of Nuclear Sciences and Physical Engineering has been flexible in the reactions to the needs of the labour market in which a critical shortage of experts in these fields has been manifested. It is supposed that the education programme will be closely connected to its field of application, either in the form of study stays or in the form of involvement of experts from industry in lectures.

Poster SEP / 446

Salophen modified biochar fibres derived from Luffa Cylindrica and uranyl binding

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Salophen modified biochar fibres derived from Luffa cylindrica sponges were prepared and subsequently studied as a potential U(VI) adsorbent by batch equilibrium experiments. The material was characterized before and after U(VI) adsorption by spectroscopic and microscopic methods of analysis. FTIR and SEM investigations revealed that the salophen modification of the biochar surface was successful without any significant changes of the textural properties of the material. The experimental data were well fitted by the Langmuir adsorption model (R= 0.99) and resulted in extraordinary high values for the maximum adsorption capacity even under acidic conditions (qmax at pH 3 and 5.5 equal to 714 g·kg-1 and 833 g·kg-1, respectively). Hence, the material could be a very attractive candidate as an effective adsorbent for the selective adsorption of U(VI) from process solutions and industrial wastewaters.

Poster NAM / 461

Study of Gravity Effect on Neutron Spatial Distribution in Cold Neutron beam by HANARO Research Reactor

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The cold neutron beam guide of HANARO research reactor has a length of about 100 m and the cold neutron has a velocity of about 300 m / s. At this time, the cold neutrons are affected by gravity for

about 0.333 seconds. In the Hanaro study, there is no history of measuring the influence of gravity and it is not reflected in the design. In this study, neutron beam images of the neutron energy using the neutron imaging system and the moderator were tried to confirm the gravity effect in the neutron beam.

Poster SEP / 447

Thorium binding by biochar fibres derived from Luffa Cylindrica after controlled surface oxidation

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Adsorption of Th(IV) by Luffa Cylindrica sponges after controlled surface oxidation by nitric acid has been investigated by batch-type experiments as a function of various physicochemical parameters (e.g. adsorption capacity, pH, ionic strength, mass of the adsorbent, temperature and contact time). Fitting of the experimental results reveal that even under strong acidic conditions the relative sorption is above 70% and the maximum adsorption capacity of the biochar fibres for Th(IV) is qmax= 70 g·kg-1, at pH 3. In addition, experimental results related to the effect of ionic strength on the adsorption efficiency and corresponding IR studies clearly indicate that adsorption occurs through the formation of inner-sphere complexes.

Poster NAM / 471

Substituted tetraphenylethene compounds as chemosensors for recognition of Th4+ and UO22+

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Thorium and Uranium are representative elements of actinide metal that have naturally radioactivity and widely distributed in the environment [1,2]. Thorium is used as a material for producing nuclear fuel, and ThO2 is often employed as a catalyst. Also, thorium is applied in high-quality lenses and high-tem-perature ceramics, and the alloys of thorium are frequently used in air craft engines. Uranium is one of the main fuels in nuclear energy generation, and it also has been used in nuclear weapons. With the growing human demand for nuclear energy, the worldwide uranium consumption is continuous increasing.

Considering the widespread use of these two metals and these toxic properties, the development and improvement in analysis methods for the determination of thorium and uranium are vital. Therefore, many techniques have been used for the determination of thorium and uranium. Among these analysis methods, chemosensors detection is a simple, rapid, highly selective, and low-cost method for metal ion determination.

Considering the unusual photophysical property of aggregation-induced emission (AIE) compounds, AIE compounds may as chemosensors and provide good approach for actinide metal ions recognition. Herein, a series of compounds with tetraphenylethene structure are prepared as chemosensors for recognition of thorium and uranium. These sensors have excellent selectivity and anti-interference ability, which can efficient identification and analysis of the target metal ions under complicated conditions. Meanwhile, due to the aggregation-induced emission property, these sensors can realize the target metal detection without expensive instrument. These sensors have potential applications in environmental systems for thorium and uranyl ions detection.

Poster NAM / 475

Tritium speciation in nuclear site metals: An aid to decommissioning

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Insights into the manner in which tritium is associated and retained in metals can be gained by studying total tritium and its progressive loss with incremental heating from 20-900℃. A range of metals from two nuclear sites and one non-nuclear site has been investigated for their tritium content and associated thermal desorption profiles. These metals have had different exposure histories to tritiated water or neutrons and these variations have led to their different tritium content. Irradiated and non-irradiated metals show different tritium thermal desorption profiles and this reflects the relative ease of thermally decomposing hydrated oxides trapped along grain boundaries or the greater difficulty in mobilizing tritium trapped in the metal lattice. Significant 3H was observed in non-irradiated metals (e.g. stainless steel and copper) which implies that exposure of the metal to tritiated water vapour or gases near the nuclear facilities is a significant source of tritium contamination. Tritium contamination into the metals via atmospheric absorption is mainly confined to the surface layer or paint layer. Tritium penetrates into the metal surface by diffusion with a rate controlled by the metal types and the surface condition of the metal (painted, unpainted).

Tritium contamination of metals via neutron activation is not mainly confined to the surface layer. It will be affected by the distribution of some elements (e.g. 6Li or 10B) presented in the metal. Tritium of metal mainly presented two different forms including a weakly bound 3H (inferred to be an HTO form) and a strongly bound form (inferred to be a non-HTO form) that required a high temperature to liberate it. However, the HTO form of the metal has different thermal decomposition behaviour indicating a slow desorption rate at low temperature (~120 ℃). This is significantly different thermal decomposition behavior of HTO isolated rapidly at low temperature (~100 ℃). Therefore, the effective and quantitative extraction of 3H from metals requires prolonged heating at a high temperature (900 ℃ for 4-5 hours).

Poster SEP / 453

The Effect of Surface Properties on the Uranium Adsorption by Mesoporous Ceria

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CeO2 solids of different surface areas have been prepared by sol-gel reverse micelles method using non ionic surfactant Triton X-100. The samples have then characterized by N2 isothermal adsorption, SEM microscopy and FTIR spectroscopy. The effect of the BET surface area/surface groups of the solids on the U(VI) adsorption has been investigated by batch-type experiments and evaluated by correlating the surface parameters with the corresponding maximum adsorption capacity values, which varied between 0.108 and 0.162 mg g-1. The adsorption of U(VI) by CeO2 is well described by the Langmuir isotherm model indicating the predominance of one type's interaction between the U(VI) ions and the surface hydroxyl groups and that the adsorption efficacy strongly depends on the BET surface/surface hydroxyl groups.

Poster NAM / 476

Development and Implementation of a Method for HEU and LEU nuclear material Age-dating

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To identify and characterize illicit trafficking and smuggling nuclear material, information obtained from analysis of unknown samples are important and helpful for authorities (Law Enforcement) to give fast and appropriate response actions.

In the framework of studies and collaboration with safeguard networks, our laboratory is able to carry out the characterization of nuclear material using various analytical methods, including isotopic composition, chemical impurities contents, and material properties. Furthermore, a specific method was developed and improved to give information about the date of production of HE and LE Uranium samples.

Our methodology for dating the last purification of the unknown Uranium material using the parent/daughter chronometer 234U/230Th is presented : after chemical extraction from the bulk material and the chemical preparation by co-precipitation, Thorium amount is determined using ⊠-spectrometry, and Uranium fraction is measured by Isotopic Dilution - Thermal Ionization Mass Spectrometry (ID-TIMS).
A focus is done on the specific chemical preparation of the sample, and several examples of applications of age-dating method are given.

Poster SEP / 454

Studies on the Separation of Ra(II), U(VI) and Eu(III) from Aqueous Solution using MnO2-resin

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The applicability of MnO2-resin for the selective separation of Ra(II) from aqueous solutions containing also U(VI) and Eu(III) has been investigated as a function of solution pH and salinity. The measurements were performed by LSC for radium and by spectrophotometry for uranium and europium, using Arsenazo-III. According to the experimental results the maximum chemical recovery of the MnO2-resin for Ra(II) is observed in the pH range between 5 and 7 and selective separation of Ra(II) from aqueous solutions can be triggered by adjusting solution pH and salinity.

Poster NAM / 844

An experimental approach to the determination of the gross activity of uranium, plutonium, americium and strontium in human biological samples using solid-state scintillation

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Rapid bioassay methods have been mainly proposed for the sequential measurements of actinides in human blood, bone marrow and urine. The method involves sample treatment using microwave digestion techniques followed by direct measurement of digested products using powder scintillator and alkaline solution as a replacement for traditional liquid scintillation cocktail. The selected group of radionuclides was chosen due to their occurrence in the nuclear fuel cycle, nuclear accident, high radiotoxicity, and the possibility of potential misuses, such as dirty bombs. The work aimed at verifying the connection of microwave digestion using alkaline solution with solid-state scintillation using a powder scintillator YAP:Ce together with an alkaline medium.

Poster SEP / 485

Ultrafiltration study of Cs-137, Co-60, Sr-85 speciation in model solution of spent nuclear fuel pool

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Spent fuel is generated during power reactor operation in the form of spent fuel assemblies. Spent fuel assemblies are placed to at-reactor spent fuel pool for cooling, reducing of fission products radioactivity and radiation protection of staff. During storage of spent fuel assemblies radionuclides, both fission and activation origin, get to solution of spent fuel pool. Radionuclides, interacting with products of corrosion, can form radioactive soluble species and suspensions that fall to bottom of spent fuel pool or to hard-to-reach places. Radioactive sediments increase background radiation and staff doses, as well as problems at spent fuel pool decontamination during operation or decommissioning.

Existing treatment installations for spent fuel pool solutions manage with treatment process partially. More effective treatment methods and technologies for these solutions are required. Radionuclide speciation in solution of spent fuel pool is an important information that is necessary for solving this problem. The aim of this study is to establish speciation of major radionuclides in solution of spent fuel pool.

The retention of trace amounts of radionuclides (^{60}Co, ^{137}Cs, ^{85}Sr) in model solutions by regenerated cellulose ultrafiltration membrane with MWCO 1000 Da was studied within this research. The model solutions were: distilled water, 0.1 M KNO_3 and 20 g/L boric acid solution (the composition similar to spent fuel pool medium) at different pH values. Measurements of solutions were carried out by γ –spectrometry method (activity of radionuclides ^{60}Co, ^{137}Cs, ^{85}Sr) and volumetric analysis of boric acid.

The results of radionuclides retention on ultrafiltration membrane depending on solution pH value and solution composition were obtained during of the research. Predictably, boric acid is retained on this membrane slightly, within 5%. Retention of radionuclide ^{60}Co Co reaches 100% at pH 9 and more, and it weakly depends on solution medium. Probably, this is because of generation of hydrolysis forms of cobalt.

The speciation of all investigated radionuclides is similar (Fig. 1) in boric acid solution in weak acid pH region. Rather, equal speciation is explained by formation of pseudocolloid particles of radionuclides. As illustrated (Fig. 1), that speciation of radionuclides ^{137}Cs, ^{85}Sr is uniformly in boric acid solution.

The experiment has shown the possibility of ^{60}Co, ^{137}Cs, ^{85}Sr separation from boric acid solution using membrane methods in the medium of spent fuel pool.

Figure 1

Poster SEP / 564

The development of separation method of iodine from intermediate level radioactive waste

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The aim of this study is develop a procedure to separation of iodine from intermediate level radioactive waste. The total activity of radionuclide 129I is limited in near surface repository Mochovce and the total limit is 4.58*10E+11 Bq. Proposed procedure consists of several separation and preconcentration steps like distillation, extraction chromatography by means CL-Resin and precipitation. This separation method ensures to determine 129I activity and or reach very low values of MDAs. The sample is prepared as a palladium iodide precipitate for measurements.

Poster SEP / 583

The use of various inorganic acids as shelf deposition solutions and their effect on nickel plates

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The present study shows the efficiency of various inorganic acids as shelf deposition solutions and their effects on nickel plates of 99% purity, 25 mm diameter and 0.50 mm of thickness 1. 0.5M, 2M and 0.1M hydrochloric acid solutions in addition to 0.1M and 0.5M solutions of nitric, sulfuric, hydrofluoric and phosphoric acid were studied and compared.
Tap water was used as matrix, polonium analysis performed in 0.5L, while 209Po was used as an internal tracer. The pre-concentration of polonium performed via rotary evaporation at 40mbars/ 45oC. The deposition solutions were kept in a calibrated water bath, ascorbic acid was used as reducing agent and the temperature of each process was monitoring by a calibrated thermometer. The sources were measured by a-spectrometry.
Hydrochloric acid solutions are mostly preferable in polonium deposition. 0.5M hydrochloric acid solution was primarily examined at various temperatures and maximum deposition times of 6h (fig.1), since it is considered one of the most common used deposition solution in polonium analysis 2.

Figure 1. Polonium chemical recoveries on nickel plates in 0.5M HCL, at various deposition times and temperatures.

No damages were observed on nickel surface after the use of 0.5M hydrochloric acid solution at 55oC, 65oC and 75oC. Nevertheless, after 4h of stirring at 85oC potential corrosion was observed at the surface of the nickel plates which was indicated by a dark brown coloration of the deposition solution. Additionally, a reduction in chemical recovery observed after 4h of stirring at 85oC either due to damage on nickel surface or due to volatility of polonium although losses have been found to take place at 100oC [3]. 2M hydrochloric acid solution was afterwards examined. High chemical recoveries were achieved at 55oC for 4h and 6h of stirring however potential corrosion on the metal surface was similarly observed as the deposition solution acquired a dark brown coloration. 0.1M hydrochloric acid solution was also studied at the same conditions with no observed damages on nickel surface.
Nitric acid was rejected due to its corrosive effect on nickel surface even at room temperature. In

contrast, the rest solutions caused no effect on the surface of the plates at the specific temperature (fig. 2).

Figure 2. Effect of various shelf deposition solutions on nickel plates at room temperature.

The deposition conditions for the rest solutions of the study were steadily selected at 55oC and 4hours stirring. The chemical recoveries and the solutions'effects on nickel plates are presented in table 1. The best chemical recoveries achieved via the use of 0.5M and 2M hydrochloric acid solutions, though 2M solution caused damage on nickel surface. Except nitric acid solutions which found to corrode nickel the rest inorganic acids of the study could successfully been used as alternative polonium shelf deposition solutions on nickel plates.

Deposition Temperature Time of Chemical Effects on nickel surface
solution (oC) deposition (h) recovery (100%)

2M HCL 55 4 89 Potential corrosion
2M HCL 55 6 97 Potential corrosion

0.1 M HCL 55 4 68 None
0.1 M HCL 55 6 71 None

0.1M HNO3 Room temp. Corrosion
0.5MHNO3 Room temp. Corrosion

0.5M H2SO4 55 4 80 None
0.1M H2SO4 55 4 66 None

0.5M H3PO4 55 4 75 None
0.1M H3PO4 55 4 56 None

0.5M HF 55 4 74 None
0.1M HF 55 4 50 None

Table 1. Polonium recoveries after the use of various deposition solutions and the solutions'effect on
nickel plates.

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Poster SEP / 644

Nanostructured aluminosilicate sorbents of radionuclides based on clay-salt slimes of JSC "Belaruskali": Physicochemical properties and application

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Solving the problem of ensuring of the radioactive waste safe management from NPP is very relevant in the world due to the intensive development of nuclear energy, as well as environmental pollution because of technogenic accidents at NPP (Chernobyl, Fukushima). Natural aluminosilicates and modified products based on them can be promising materials for purification from radionuclides of technological solutions of nuclear power plants, including liquid radioactive waste, as well as contaminated with radionuclides of natural ecosystems.

One of the possible directions of utilization of clay-salt slimes (CSS) which are industrial wastes of potassium production of JSC "Belaruskali"is obtaining of nanostructured aluminosilicate sorbents of radionuclides on them. As a result of long-term studies of the composition and structure of sorbents based on CSS it was pointed out that they belong to clay minerals. Illite prevails in the composition of clay minerals, the content of which varies in the range of 44–54%, depending on the type of sorbent. The specific surface of sorbent samples varies from 21 to 32 m2/g. Water-acid treatment allows to increase the specific surface of sorbents by almost 2 times, and the content of the sorption-active mineral –illite up to 65% due to the decrease in the content of carbonate minerals and water-soluble salts.

On the basis of studying the sorption kinetics of 137Cs with aluminosilicate sorbents based on CSS at a ratio of solid : liquid = 1 : 100 phases, it has been established that sorption occurs rapidly and within 10 minutes and is about 95% 1. Further increase in the time of interaction of the sorbent with the radioactive solution up to 30 days leads to an increase of sorption degree up to 98%. The sorption isotherm 137Cs, constructed in a wide range of concentrations from 10-10 to 10-1 mol/l, showed that it is possible to distinguish two sites of sorption, which differ in 137Cs distribution coefficient. These sections show that the sorption of 137Cs on samples of sorbents occurs on two types of sorption centers, differing in capacity and selectivity of 137Cs binding. An investigation of the acidity (pH) effect of the radioactive solution on 137Cs sorption by aluminosilicate sorbents showed that in 2–12 pH range the degree of sorption varies from 95 to 98%. It was found that the degree of sorption of 137Cs is significantly influenced by the presence of K+ and NH4+ cations in the solution. In the concentration range of the Na+ cation up to 0.5 mol/l, the 137Cs sorption on the aluminosilicate sorbents practically does not change. Studies of the sorption of Cs(I), Sr(II), Eu(III), Am(III) by sorbents obtained from CSS have been carried out at the Institute of Nuclear Chemistry and Technology (Warsaw, Poland) 2. It was pointed out that the degree of sorption of Cs(I), Sr(II), Eu(III), and Am(III) radionuclides is 99, 99, 91 and 97%, respectively.

The results of the performed studies indicate that the aluminosilicate sorbents based on the CSS of JSC "Belaruskali"are very effective for both sorption of 137Cs and radionuclides of Sr(II), Eu(III) and Am(III). Sorption materials based on CSS can be effective sorbents for purification from radionuclides of liquid radioactive waste from NPP, aquatic environments and natural ecosystems.

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Poster SEP / 755

Synthesis of diglycolamide extraction agents anchored to polyacrylonitrile matrix

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Increasing demand of high-purity and radiopharmaceutical grade radionuclides leads to the research and development of new, fast, simple and cost-effective separation methods. The most frequently used separation methods in nuclear medicine are liquid-liquid extraction or chromatographic methods. For example, the radionuclide ²²³Ra for targeted alpha-particle therapy is exclusively gained from ²²⁷Ac/²²⁷Th/²²³Ra radionuclide system and the separation of ²²³Ra is usually performed by ion-exchange or extraction chromatography. Diglycolamide solid extractants were successfully used in case of extraction chromatog-

raphy 1.
Diglycolamides, ligands forming strong complexes especially with trivalent ions, can find broad use in separation of various radionuclides in many branches of nuclear chemistry, such as reprocessing of used nuclear fuel, nuclear medicine, analytical chemistry or production of high-purity radionuclides.
Most of the diglycolamide solid extractants are based on impregnated silica or polymer matrix or a chromatographic paper. This study focuses on the synthesis of diglycolamides covalently bound to polyacrylonitrile (PAN) beads. It is presumed that the covalent bond with the matrix will eliminate the leakage of the extraction agent from the solid extractant matrix which is an issue in case of impregnated materials.
PAN beads were chosen as a convenient material for its physical, chemical and radiation endurance and its chemical structure 2. The presence of nitrile groups in the molecule of polyacrylonitrile allows the formation of covalent bond with diglycolamide. The synthesis is based on partial surface reduction of nitrile groups of PAN beads to primary amine and subsequent solid-state synthesis of diglycolamide.
PAN beads were partially reduced using multiple systems such as complex metal hydrides metal hydrides with transition metal salts or hydrogen with homogeneous catalysts. Several samples of partially reduced PAN beads were prepared and characterised via FT-IR and NMR spectrometry. The content of amine groups in the prepared samples was determined using acid-base titration.
Subsequently, the best reduction systems were used for the preparation of precursors for solid state synthesis of diglycolamides that has been performed using amide formation via coupling agent. Prepared solid extractants were characterized by FT-IR and NMR spectrometry, scanning electron microscopy and measurement of specific surfaces, and tested by the basic sorption experiments.

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Poster SEP / 640

Activated carbon as an effective pertechnetate adsorbent

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Activated carbon (AC) is pores space bounded by the carbon atoms. Pores of AC are directive for its application. It was shown that, surface modified form of carbon-based materials, are promising sorbents for separation of various radionuclides. In our study 5 AC adsorbents (A-E) are prepared from cellulose wool by treating with different solutions and carbonized at the temperatures of (700-800) ℃ for Tc adsorption. They are characterized by FTIR for organic functional groups determination, HRTEM for microscopic imaging, liquid N_2 sorption for pore size distribution and specific surface area determination, and acid-base titration conduced potentiometrically for surface charge investigation. Surface area of the sample is within the range of (150-510) $m^2 \cdot g^{-1}$. Samples C and E possess also mesopores. AC samples have a positive surface charge and point of zero charge lies in the range of pH 6-8.
Tc is a fission product with a long half-life ^{97}Tc (EC; $4.21 \cdot 10^6$ a), ^{98}Tc (β^-; $4.2 \cdot 10^6$ a), ^{99}Tc (β^-; $2.1 \cdot 10^5$ a) formed in the thermal neutron fission of ^{235}U and ^{99m}Tc is the most commonly used diagnostic tool. One option how to remove or control released Tc in the environment, is sorption process. It has been shown that AC has a relatively high efficiency for Tc retaining ($^{99m}TcO_4{}^-$). Adsorption

equilibrium of TcO_4^- is reached in few minutes except for sample D (60 minutes). Mechanism of TcO_4^- adsorption involves: R-C-OH + TcO_4^- = R-C-OTcO3 + OH^- or if phenolic or carboxylic groups are present: R-CO···H + TcO_4^- = R-C···H···$OTcO_3$. Tc can bind direct to the C as -C-$OTcO_4$ or mechanism involves chemical reduction of the soluble TcO_4^- to TcO_2 and its adsorption on the surface. Adsorption of TcO_4^- on samples A - E occurs at pH of 2.
Presence of anions suppresses adsorption of TcO_4^-. $HCOO^-$, NO_3^-, Br^-, Cl^-, CH_3COO^-, SO_4^{2-}, and ClO_4^- are used to investigate the adsorption competition between TcO_4^- and anions. Competition strongly depends on pH, structure of the anions, and standard absolute molar enthalpy of the hydration. The most significant decrease of TcO_4^- adsorption is observed in presence of NO_3^- and ClO_4^-. It is due to close values of the highest standard absolute molar enthalpies of hydration. Moreover, ClO_4^- has similar tetrahedral structure to TcO_4^-. Following order for sample A shows the suppression of TcO_4^- adsorption from the weakest to the strongest (kJ mol^-): SO_4^{2-} (-1,099) < $HCOO^-$ (-384) < CH_3COO^- (-374) < Cl^- (-359) < Br^- (-328) < NO_3^- (-316) < ClO_4^- (-205).
In order to investigate desorption of TcO_4^- and thus reusability of AC, simple column desorption experiments are carried out. Desorption solutions consist of anions as in the previous paragraph, respectively, at pH 12. Continuous gamma measurements show that 1 M ClO_4^- has the “strongest” ability to remove TcO_4^- from AC. Adsorptions of TcO_4^- on samples with oxidized and reduced surface are also studied. Immediate adsorption of TcO_4^- makes reduced surface sample preferable to oxidized one. The results obtained in this study improve that activated carbon is an effective sorbent for the separation of the pertechnetate. This work was supported by VEGA Project No. 1/0507/17.

Poster SEP / 750

Technetium separation from concentrated molybdate solution issuing recycling of molybdenum from irradiated CerMet nuclear fuel

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In the Partitioning and Transmutation (P&T) technology, minor actinides burning to shorter-lived radionuclides can happen in Accelerator-Driven Transmuters (ADT) or in the GEN IV nuclear reactors. Among the fuel matrices considered to embed minor actinides for burning, inert matrices composed of molybdenum metal (CerMet fuel) or inert ceramic magnesium oxide matrix (CerCer fuel) are high on the list of candidates.
Molybdenum-based CerMet fuels have an excellent thermal conductivity, however they require use of molybdenum with specific isotopic composition. ^{99}Mo is the recommended isotope for ADT. A multi-recycling process of spent Mo-based CerMet fuels should hence include recycling of the isotopically manipulated molybdenum. In addition to separating the bulk transmuted radionuclides, separation of the residual inventory of radionuclidic impurities may be required prior to manufacture of new CerMet fuel from the re-cycled molybdenum. Among these potential impurities, ^{99}Tc was identified.
Therefore, this study is focused on elimination of technetium (TcO_4^-) from concentrated molybdenum solutions. A336-PAN(HNO_3) solid phase extractant has been identified and tested for this purpose. Based on the proposed reprocessing scheme of CerMet fuels, molybdate model solution with Mo concentration 100 g L^{-1} and pH = 9.1 was used.
Although the pH 9.1 is given by dissolving MoO_3 in ammonia solution, it was found that in the range of pH 8.1 –10.2 the extraction percentage varies by only about 0.8 %. In the conditions used, the maximum value of the extraction percentage corresponds to 99.1 %. This fact shows that small pH deviations will not have significant effect on the extraction process. Uptake of TcO_4^- is not very fast, the equilibrium is reached within 30 minutes. The dependence of weight distribution ratio (D_g) on molybdenum concentration in batch experiment revealed that D_g values decrease with increasing molybdenum concentration. This decrease can be ascribed to the competition of NH_4^+ ions from ammonium dimolybdate. The highest D_g was reached for 10 g L^{-1} of Mo. Somewhat surprisingly, the lowest D_g (D_g ~ 0 mL g^{-1}) was found in absence of molybdenum. The probable reason may be associated with the low ionic strength of deionized water. Even at the molybdenum concentration

100 g L^{-1} Mo, the weight distribution ratio of technetium is sufficient for practical application (D_g = 24·10^3 mL g^{-1}). The extraction capacity (Q_m) of A336-PAN(HNO_3) for TcO_4^- was investigated using the perrhenate carrier. The extraction isotherm, determined by batch technique, revealed that the maximum extraction capacity of A336-PAN(HNO_3), corresponding to the solid extractant saturation, is equal approximately to 1 mmol g^{-1}.
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Poster NAM / 571

Preliminary results of analysis of atmospheric aerosols in Bratislava using PIGE technique

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Recently, the CENTA laboratory was established at Faculty of Mathematics, Physics and Informatics, Comenius University in Bratislava. The laboratory comprises the system for production of various ion beams from solid and gaseous samples, the energy and mass separation and the 3 MV tandem accelerator, that is used for acceleration of the ions up to tens of MeV. The beams can be used for several IBA (ion beam analysis) techniques, like PIXE (particle induced X-ray emission), PIGE (particle induced gamma-ray emission), RBS (Rutherford back-scattering). In this work, the PIGE was used to study concentrations of several elements like Al, Na and F in atmospheric aerosols. BEGe detector from Canberra Industries was used for detection of gamma-rays from inelastic scattering of accelerated protons in thin samples. The aerosols were collected in weekly intervals at the university campus and reflect the composition of aerosols in industrial and urbanized area. For the analysis, the preparation of thin samples was developed from the collected aerosols and NIES CRM No. 28 Urban Aerosol reference material was used for the calibration of the system.

Poster NAM / 621

Study on the nuclear forensics analysis of uranium pellet sample

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The study of nuclear forensics can effectively deal with nuclear security incidents and potential nuclear terrorist threats, and prevent radioactive crimes or terrorist attacks. Through the analysis of the nuclear material of nuclear fuel cycle different stage, the initial source of nuclear material and the whole production process could be revealed. Due to the different geological conditions and the industrial process a unique characteristics in nuclear materials and the characteristics can be distinguished. The age of nuclear material, trace impurity and stable isotope composition are important characteristics in the analysis of unclear materials and various analysis indicators of material properties.

In this work, the uranium pellets, the most important product in the nuclear fuel cycle, were used as the analysis objects, and the analysis technology of nuclear forensic is used to trace the source and process conditions of the pellets. The fingerprint information, including physical appearance, micro structure, trace impurity, stable isotopic composition and uranium age etc. were analyzed. Following the analysis principle of lossless analysis fist, the diameter of the pellet is deduced about 8mm. The diameter of the uranium pellet is one of the most important fingerprint information, considering may be used in different size of pellets in different reactors. Combined with the X-ray energy wave analysis and X-ray diffraction analysis, the main ingredients of the sample were uranium and oxygen existing as UO2. By measuring the surface dose of sample, the micromorphological analysis includes the microscopic statistical analysis of the hole and grain size of the sample surface. The surface roughness is analyzed by surface profile analyzer, and the density of the sample is measured by buoyancy method. Those characteristics are closely related to the production process of the uranium pellets.The amount of 235U was 1.8% and with no 236U. The source of the pellet is natural uranium, and the pellet was never irradiated by the reactor. After cleaning, drying and dissolving, most impurities were separated from the sample and measured by Inductively coupled plasma mass spectrometry, and the content of stainless steel components such as Fe、Cr、Ni were high concluding that the UO2 powder was produced by wet process, because the stainless steel container is used in the wet process. Using U-Th to analyze the uranium age in the pellet, the raw uranium material was enriched around December of 2007. Based on the measurement and comprehensive analysis of the information of the above nuclear forensic evidence, and in comparison to the domestic data of nuclear fuel pellets, the sample pellet was produced in the early 2008 and called type AFA-3G nuclear pellet used in the Lingao nuclear power station.

Poster NAM / 673

Instrumental neutron activation analysis in monitoring environmental changes in a heavy traffic area after opening the Blanka tunnel complex in Prague

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The Blanka tunnel complex, a part of the Prague City Ring Road, is with a length of about 5.5 km the longest road tunnel in the Czech Republic and the longest city tunnel in Europe. Designed to relieve the historic center of Prague from heavy traffic, it connects the area west of Prague Castle with the Trója district in the northeast. The construction of Blanka started in 2007. Initially due to open in 2011, the tunnel complex was officially opened to the public in September 2015. Model calculations have shown that starting Blanka operation will significantly increase traffic on a connected main road, the V Holešovičkách street, which passes through a compact populated area in the length of 1.6 km.

To study changes in the dust fallout near the highly exposed V Holešovičkách street, expected to appear with road traffic intensification after opening the Blanka tunnel complex, two sets of dust samples collected in the street vicinity were used: a set collected long before Blanka opening between December 2012 and September 2013, and a set collected immediately before Blanka opening and during its pilot operation between February 2015 and April 2016. The study included characterization of elemental composition of the collected dust samples by means of instrumental neutron activation analysis, and SEM/EDX for further compositional characterization and identification of particles of the particulate air pollution. The relationship of compositional variation in the studied dust samples to the traffic intensification, regarding probable seasonality effects, will be discussed.

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Poster NAM / 691

Performance Prediction of Coincidence-based Prompt Gamma Activation Imaging System using Geant4 Monte Carlo Toolkit

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The prompt gamma activation imaging (PGAI) using the neutron beam has been used in archeology and cultural heritage researches to obtain the two- or three-dimensional elemental distribution of the volumetric sample. To measure the high-energy prompt gammas emitted by the neutron induced nuclear interactions with the sample, a simple measurement system, which consists of an HPGe detector, a single-slit collimator and a scanning system, have been used. Although the measurement system can visualize the elemental distribution of the sample, it required a time-consuming process to scan the sample.

For the practical use of PGAI, a new imaging system, which determines the 2D elemental distribution of the bulk sample by using the coincidence logic, was proposed. The imaging system consists of a calorimeter for the element identification by measuring the energy and a position sensitive detector coupled with a parallel-hole collimator for the position determination. To visualize the 2D distribution, the imaging system selectively measure the coincident events where prompt gammas emitted by one nuclear interaction interact simultaneously with the calorimeter and the position sensitive detector. In the imaging system, one detector is located at the opposite site of the other detector, and the sample is placed at the center of both detectors.

To estimate the feasibility of the proposed imaging system, in the present study, we performed the Monte Carlo simulation using the Geant4 toolkit for the rectangular iron plate implanted with the nickel. To improve the computational speed, the simulation was separated by two parts. First, we calculated prompt gammas emitted by the nuclear interactions between the neutrons and each sample including the number of generated photons and each energy information. After that, for the detector setup of PGAI, the prompt gammas were generated in the sample by using this information, and the coincidence events was recorded. The emission positions were randomly determined by considering the emission yield of prompt gammas for each element, and the emission angles were also randomly sampled. For the coincidence events, we can identify the characteristic gamma-rays' peaks for the iron (352, 7631 and 7645 keV) and the nickel (465, 8533 and 8998 keV). With the energy gate of 460-470, 8528-8538 and 8993-9003 keV for the calrolimeter, we also recognized the shape of implanted nickel from the 2D image obtained by the imaging system. Base on the results, we expects that the proposed imaging system can be utilized in the elemental analysis of volumetric samples by measuring the prompt gammas.

Poster NAM / 717

Comparative characterization of biosorbents by thermodynamic and kinetic functions in processes of radionuclides adsorption

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The current concept of handling low-level liquid radioactive waste (LRW) consists of development of technological schemes providing efficient and cheap LRW treatment. In addition, such technologies ought to lead to compacting of radioactive waste in combination with long-term, reliable and safe isolation from the biosphere. Unfortunately, some types of waste from the nuclear industry still stay in uncontrolled storage condition and represent a serious environmental hazard. All of this often leads to the fact that technically occurred radionuclides come into the environment, changing the natural radioactivity of soils, natural waters and sediments 1.
Processing of any type of LRW and environmental rehabilitation based on the extraction and concentration of long-lived and highly toxic radionuclides from solutions which contain complex chemical and radionuclide compounds. In order to solve the problem of safe handling of waste from radiochemical plants, it is necessary to involve fractional isolation of radioactive components before storage. It is also important to have reliable analytical methods for determining the ultra low level of radionuclides in environment. To implement these processes, various types of sorbents may be widely used. A special feature of inorganic sorbents (IS) is high selectivity with respect to cesium and strontium ions, high chemical, thermal and radiation resistance. However, their usage is limited by the mechanical parameters of these materials. The efficiency of organic sorbents (OS) depends to a large extent on the composition and concentration of salts in solutions, surfactants and types of ligands. But small compaction of used types of IS and OS leads to a growth of in the volume of solid secondary waste and the cost of their disposal 2.

In recent years, development of a new class of sorbents consisting or involving substances of biogenic origin (biosorbents) has been widely applied. The most widespread are polymeric materials based on chitin (CT) and chitosan (CTS), which have a wide raw material base e.g. marine crustaceans shells, fungi, bee and other insects, etc. This supply base is constantly expanding due to the involvement of new alternative sources[3]. The unique properties of CT and CTS are high sorption ability, low ash content, biodegradability to substances that are safe for living matter, the possibility of obtaining sorbents with optimal geometry and a large surface of particles. The presence of amino group in CTS chain allows making functionalization of the biopolymer and producing sorbents with high selectivity and improved capacitance characteristics. Thus, the study of interaction of radionuclides with polymeric materials based on CT and CTS is an actual scientific and practical task [4–7].

Now it is well known the large number of sources containing data on the thermodynamic properties and kinetic characteristics of CT and CTS in the processes of interaction with radionuclides[8]. However, the comparative analysis of sorbents with combination of a complex of physicochemical characteristics in literature has been rarely given.

Mass transfer laws in adsorption processes have been shown how the interaction between radionuclides and sorbent surfaces take place. The mass transfer process is a complex phenomenon, which is the result of internal and external diffusion [9].

Various polymeric sorbents were used in this work to analyze their properties by thermodynamic and kinetic laws. A natural complex of chitin and melanin Mycoton (NCTM-Mycoton) [10] and its modifications by inorganic substances have been used for adsorption of An, Eu and Sr (MDM-15 contains 15% w. $\mathrm{MnO_2}$ or MSB-15 contains 15% w. $\mathrm{BaSO_4}$)[11], Cs (Mycoton-Cs contains 30% w. $\mathrm{K_2Cu[Fe(CN)_6]}$)[10] respectively. Spherical granulated chitosan (SGCTS)[12], chitosan cryogel (CCTS) and chitosan cryogel crosslinked by pyridoxal phosphate (CCTS-PP)[13,14] also have been investigated in radionuclides adsorption.

To determine the rate of the process, experimental data have been used to calculate the diffusion coefficients (D) and Bio number (Bi). Both of them reflects the similarity of the distributed substance transfer through the boundary of the solid and liquid phases, and includes the ratio of the mass transfer coefficient and the coefficient of internal diffusion characterizing the rates of external and internal diffusion.

To evaluate the interaction of sorbate with a sorbent, adsorption isotherm were obtained and maximal capacity (A_∞) were calculated. The interaction of the sorbate with the sorbent was estimated from the value of the Gibbs energy.

Table. Comparative characterization by thermodynamic and kinetic functions of different types of biosorbents and radionuclides

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- by Freundlich equation

The table shows that the calculated values of Bi, for all types of sorbent, are less than 10. This fact allows to conclude that the mechanism of sorption by CT and CTS-containing polymers is rather complicated and it takes place both in the external and internal diffusion zones[9]. However, the main part of the process is the external diffusion. For inorganic modifications, it has been noted the displacement of the mechanism toward the external diffusion region, as a result of preferential sorption on a fine crystalline inorganic modifier.

As it has been shown in the table, for all the sorbents studied, the values of $\Delta \mathrm{G}_0$ have been reached the value less than −40 kJ/mol. Due to this fact the process can be attributed as weak interactions. The increase in $\Delta \mathrm{G}_0$ in case of cesium adsorption on Mycoton-Cs has been shown the decisive contribution of inorganic material in interaction with radionuclides.
Investigations of the sorption processes with using CT and CTS-containing sorbents have been described by Langmuir adsorption model. Only one exception has been obtained. The freshly formed HCTS adsorbs uranium and in this case the sorption processes can be described by Freundlich equation. It was established that the highest values of the capacity (A) for uranium were obtained on CCTS-PP.

Comparison of A_∞ for Cs obtained with Mycoton-Cs and SGCTS-Cs (contains 60% w. $\mathrm{K_2Cu_3[Fe(CN)_6]_2}$ [15] showed that the use of chitosan with a higher modifier content allows to achieve higher values of the capacities. Capacity of modifier is an important factor in the future of its application in decontamination technology of $^{137}\mathrm{Cs}$-containing solutions and analytical practice. Under dynamic conditions in fixed bed column experiments Mycoton-Cs loses the modifier on the surface, in contrast to the SGCTS-Cs where the ferrocyanide is firmly fixed in the volume of the adsorbent. Nevertheless, the equilibrium in Cs sorption has been established faster with usage Mycoton-Cs than applying SGCTS-Cs.

Comparative analysis of thermodynamic data and kinetic characteristics allowed to reveal the factors influencing on interaction efficiency of radionuclides with biosorbents.
1. The addition of highly disperse inorganic matters leads to increase in selectivity, process speed, sorbent capacity when biosorbents interact with strontium and cesium.
2. Changing the structure of chitosan by reducing the degree of crystallinity gives an improvement of sorption characteristics.
3. Forming cryogels from chitosan leads to increament in the capacity and speed of the sorption process and expands the number of the adsorbed radionuclides.
4. Modification of spherical granulated chitosan by inorganic materials allows the creation of strong and efficient sorbents for cesium adsorption from sea water and high salted technology solution

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Poster NAM / 736

Preparation of micro samples using AAA automat for AMS radiocarbon dating

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Radiocarbon dating is an exact dating method and its pioneer was the team of Professor F.W.Libby in the 1950s. This is the most significant and most commonly used 14C determination application. Within the carbon cycle in nature, 14 C is generated in the upper part of the atmosphere by the interaction of 14N with cosmic radiation 14N (n, p) 14C. Radiocarbon is relatively quickly oxidized to 14CO2. Subsequently by photosynthesis 14CO2 is assimilated by plants and transmitted through the food chain. 14C activities in the atmosphere and the tissues of plants and animals are therefore almost identical. If the sample was isolated from the carbon cycle, the activity of 14C decreased due to radioactive conversion. Due to the half-life of 14C decay (T½ = 5730 years), the range of the radiocarbon method is approximately 40 to 50 thousand years.
The AMS radiocarbon dating process is started by selecting the appropriate processing method. The process follows the isolation and purification of datable chemical forms of carbon. The Acido-Alkali-Acid (AAA) method is used for sample pre-treatment. After pre-treatment, the sample is dried. The sample with CuO is sealed under a vacuum in a quartz glass tube and burnt at 900 °C. The prepared CO2 is dried and dosed to graphitization. Reduction is carried out in a closed tube with Zn vapour after the catalytic action of Fe. Accelerator Mass Spectrometry (AMS) makes it possible to convert several percent of the 14C atoms from the sample to the detector during a short-term measurement. Carbon weights from the sample are about 1 mg.
Samples for dating include foreign ingredients that need to be removed. Mobile chemical forms of carbon cause significant influence on the dating result. These include, in particular, humic and fulvo acids, which usually seem to reduce the age of the sample. Furthermore, the presence of a carbonate form of carbon significantly affects the result of dating, when it can be reduced or increased. The AAA method is based on alternate leaching in acid (removal of carbonate forms), neutral and alkaline media (removal of humic and fulvo acids). The AAA method can be used to pre-treatment of

carbon, wood samples, and also to process collagen from bone samples.
For pre-treatment of micro samples using the AAA method, we have assembled an automated computer-controlled unit in our laboratory. The sample is placed in a 10 ml glass single-necked cuvette. The cuvette is sealed with a cap with capillaries for inlet and outlet of solutions. The machine consists of prepared solutions of HCl, NaOH and distilled water, which are guided through the capillaries through the switching valve and the peristaltic pump into the cuvette with the sample according to the currently selected program. After the leaching of the sample is done in the cuvette, the solution is drained into the waste container. Depending on the sample type, the cuvette can be placed in a thermoblock heated to 90 ℃ or left freely in the rack. In the current layout, 6 samples of the same type can be processed on a machine during one cycle lasting approximately 17 to 30 hours.

Poster NAM / 665

Evaluation of Radon Suppression in Low Background Gamma-ray Spectroscopy Based on Monte Carlo Simulation Approach

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This study proposes the approach to remove the contribution of Rn-222 from the measured gamma-ray spectra, which achieved from a HPGe spectrometry. Several scenarios of flushing the central detector volume with nitrogen gas to actively remove the radon isotopes were simulated to evaluate the effect of Radon on the low-background gamma-ray spectra. To quantify the gamma background a set-up was prepared, the MCNPX code was adopted to evaluate the scenarios will be applied to the gamma-ray spectroscopy system consisting of a coaxial ORTEC HPGe detector (model number - GMX40-76) and the energy of events up to ~3 MeV at the NAA lab (KAERI, Republic of Korea). Rn-222 in the air around the detector contributes significantly to the remaining background and attention will be paid to the radon concentration, especially inside the sample chamber. The optimal configuration based on the simulations presented that a factor 4 improvement on the daughter decay peaks of Rn-222 with a ~30% reduction again in the integrated rate between 100-2700 keV to 0.751±0.001, and translating to improved sensitivity of the detector for the U-238 lines. The results also show a slight reduction in the 238 keV energy peak, more than expected from purging radon purely. This could be as a result of thoron reduction inside the main detector volume, which could explain why only the 238 keV peak was reduced and not the 338 and 911 keV peaks. Therefore, it can be concluded that the HPGe spectroscopy system at the NAA lab is fully operational and capable of making reliable measurements of sample activity with competitive sensitivity. It is now ready for use in material screening and selection for the environmental experiment.

Keywords: Radon, HPGe, MCNPX, Gamma-ray.

Poster NAM / 538

Determination of plutonium in water fluids of naval nuclear reactor plants

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A rapid method to determine plutonium in water is to separate it on microfiltration polymer membranes impregnated with antimony, zirconium, and manganese oxides or zirconium, titanium and other phosphates, having the highest efficiency for adsorption of plutonium. In addition, ballast salts are removed. A method was developed for determining the total activity of alpha emitters with preliminary concentration of transuranic elements (TRU) by filtering a sample through the cellulose acetate membrane impregnated with hydrated manganese dioxide. However, this method had a problem with identification of spectra in measurements of TRU activity by alpha spectrometry. Preliminary evaporation of a water sample and consequent adsorption of plutonium onto an ion exchange resin is a preferable choice for separation of TRU and identification of plutonium. The method is to add nitric acid in a water sample, evaporate the sample to almost dryness, dissolve it again, adjust all forms of plutonium to Pu(IV), and filter the sample through a strong-base anion exchanger. The anion exchange resin adsorbs only plutonium, while all other TRU including americium remain in the solution. Plutonium is then eluted from the resin using ammonium iodide dissolved in hydrochloric acid. A sample for counting is prepared by filtering the plutonium sample through the cellulose acetate membrane impregnated with hydrated manganese dioxide. The activity of plutonium precipitated on the membrane is determined by alpha radiometry or alpha spectrometry. Comparison of spectral characteristics shows that electrolytic and membrane methods of preparing samples for alpha counting have a relatively similar efficiency. However, the electrolytic separation takes at least 2 hours, while the membrane filtration time is 0.5 hours. The method for determining the volumetric activity of plutonium nuclides in plant water measures the activity in the range of 5-5·103 Bq/l, the efficiency of removing artificial and natural impurities being higher than 103. The degree of plutonium adsorption can reach 90%.

Poster NAM / 642

Instrumental neutron activation analysis with anticoincidence counting significantly reduces interferences from 82Br and 122Sb to allow reliable measurements of nanogram levels of arsenic in biological materials via 76As

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Neutron activation analysis (NAA) can be conveniently used to measure microgram amounts of arsenic. However, in biological materials major elements such as bromine, chlorine, and sodium can produce high activities resulting in poorer detection limits for arsenic. The 559.1-keV photopeak of 76As (half-life = 25.9 h) is particularly interfered with by the 554.3-keV photopeak of 82Br (35.3 h) and 564.1-keV photopeak of 122Sb (65.3 h). The modern HPGe detectors have good enough resolution for separating these three peaks. However, when bromine content is high, the tailing of 554.3-keV photopeak can mask the 559.1-keV photopeak of 76As and can make the detection of the arsenic peak rather difficult, if not impossible. In practice instrumental NAA (INAA) methods can only be used to measure arsenic down to a few ppm levels in biological materials. The 76As nuclide decays by β-emission and two major gamma-rays, namely 559.1 and 657.0 keV, which are partially coincident. The peak efficiency reduction factors (PERF) of these peaks have been measured as 0.83 and 0.24, respectively. INAA in conjunction with anticoincidence counting technique can be beneficially used under such situations. The concentrations of arsenic in several reference materials were determined by irradiating them at a neutron flux of 5 × 1011 cm-2 s-1 for 7 h, followed by decay for about 50 h and counting for 8 h. The anticoincidence gamma-ray spectrometer used in this work consisted of a HPGe detector and a 10”x10” NaI(Tl) guard detector with a 3”x3” NaI(Tl) plug. The peak-to-Compton plateau ratio of this system is about 590:1. The application of anticoincidence spectrometry was found to reduce the 554.3 keV peak of 82Br, and to suppress the background under the 559.1 keV peak of 76As. The background activities in the anticoincidence spectra were reduced by factors of 4 to16 for the biological reference materials analyzed making the measurement of nanogram amounts arsenic in them possible.

Poster NAM / 535

Leak detection of irradiated fuel assemblies in naval marine plants

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Ex-core monitoring of irradiated fuel rod leakage using general-purpose assembly of fault detection is based on identification of 85Kr release from leaking irradiated fuel assemblies. The sensitivity of this method is defined by minimum volume activity of this nuclide that is authentically measured in air mixture of leak-tight circulation circuit (LTCC) via the radiation monitoring system (RMS). Beta-emitting radioactive gases concentration monitor (UDG-1B) as a component of RMS system lowers the limit of 85Kr detection in 10 times compared with gamma-spectrometric method.

However, along with obvious advantages of UDG-1B monitor there is also a considerable shortcoming –impossibility of reliable determination of 85Kr activity when other beta-emitting gaseous radionuclides exist in LTCC (it was revealed during fault detection of irradiated fuel assemblies). These "unwanted" radionuclides - 14C in the CO2 form mainly - entered the leak-tight circulation circuit. This led to necessity for constant gas sampling from the fault detection monitor for the subsequent gamma-spectrometric analysis in laboratory that followed by increased time of measurements.

In order to solve this problem, the fault detection monitor was improved - a bubbler that provide removal of gaseous beta-emitting 14C radionuclide without 85Kr loss has been added. Besides, to decrease the radioactive emissions and, as a result, population radiation loads, the filter with a Siloxide sorbent (NITI's development) for detecting long-lived 129I has been included into process scheme of LTCC.

The set of the performed measures and researches has allowed carrying out fault detection of irradiated fuel assemblies at KV-2 facility. Upgrading of KV-2 facility has allowed to identify 85Kr entrance from leaking irradiated fuel assemblies within the range of activities from 2.7•10-10 to 1.6•10-4 Ci/l. As a comparison, the lower volume activity limit of 85Kr nuclide measured by semiconductor gamma spectrometer in laboratory was 1.4•10-8 Ci/l.

Poster SEP / 656

Determination of formation constants of actinide (An = Pu(IV), Th(IV), Am(III) and U(VI) complexes with a hydrophilic SO3-Ph-BTP ligand, using liquid-liquid extraction

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Complex formation between actinide (An = Pu(IV), Th(IV), Am(III) and U(VI) and a hydrophilic anionic form of SO3-Ph-BTP4–ligand, L4–, in water was studied by liquid-liquid extraction experiments performed over a range of the ligand and HNO3 concentrations in the aqueous phase, at a constant concentration of nitrate anions at 25°C. The competition for An ions between the lipophilic TODGA extractant and the hydrophilic L4–ligand leads to the decrease in the An distribution ratios, D, with an increasing L4–concentration.

The model of the solvent extraction process used accounts –apart from An complexation by TODGA and SO3-Ph-BTP4—also for An complexation by nitrates and for the decrease in the concentration of the free L4–ligand in the aqueous phase, due to its protonation, bonding in the An complex and the distribution between the two liquid phases.

The analysis of the results, based on the formal correction of free ligand concentrations, points to the formation of 1:x An –SO3-Ph-BTP complexes in the aqueous phase. The conditional formation constant of
the 1:x complex has been determined.

Poster NAM / 698

Non-destructive determination of elemental composition of samples of various origin using PIXE technique

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One of the crucial advantages of the PIXE (Particle Induced X-ray Emission) technique is its non-destructive approach to the sample treatment during the analytical process. Rare and precious environmental samples can be analysed in order to evaluate the concentration of individual elements presented in the specimen. Composition of various samples and material has been investigated using PIXE technique in the CENTA laboratory. Non-destructive analysis of chondrite and iron meteorites and various mineral samples were carried out using NEC Pelletron. 3 MeV protons were incident in a narrow ion beam (1.5 mm diam.), and emitted X-rays were detected using Canberra BEGe detector. Each sample was measured in 30 positions distributed in a mesh spread over the sample surface. GUPIXWIN software package was used for spectra evaluation. Concentrations of several elements (e.g. Fe, Ni, Cu, Sr, Zn⋯) were determined in analysed samples which were subsequently processed into surface distribution maps of investigated elements. A comparison of results obtained by PIXE, neutron activation analysis and traditional chemistry will be presented

Poster NAM / 843

A Rapid Determination of the Uranium in Environmental Samples

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In this study, a rapid method using single column separation and fusion for determination of the uranium in environmental samples (e.g., soil, sediment, air particulate, etc.) is presented. Various certified reference materials (CRM) of soil and sediment were used to evaluate the accuracy and precision for the developed method. Samples were rapidly digested using an alkali fusion (LiBO2) technique and readily dissolved in dilute nitric acid. Prior to separation using UTEVA resin, concentrated nitric acid has been added until the molar concentration of the sample aliquot becomes greater than 2 M. The prepared solutions were separated from a 20 mL column filled with UTEVA resin. The

purified samples were diluted and measured through inductively coupled mass spectrometry (ICP-MS). The analytical results agreed well with the certified values for all samples. All experiments for five samples including the sample digestion, separation, and determination could be conducted in 1.5 h. Thus, method developed in this study can offer the main advantages over existing methods in terms of simplicity and reliability for routine monitoring and emergency preparedness.

Poster SEP / 651

Solid phase extraction based Lu-177m/Lu-177 radionuclide generator to produce Lutetium-177

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The radionuclide lutetium-177 has become one of the preferred radionuclides for targeted therapy. The low tissue penetration of the emitted β- particles assures an efficient energy deposition on small size tumours (less than 3 mm) and a low radiation dose to the surrounding healthy tissue. This is especially useful when ¹⁷⁷Lu is combined with different targeting molecules, which are internalized within tumour cells, being then possible to treat small primary and metastatic tumours, like prostate, breast, melanoma, lung and pancreatic tumours as well as bone metastasis. Recently we have proposed a ^{177m}Lu/¹⁷⁷Lu radionuclide generator as a new method for the production of lutetium-177 1. The reported separation method, increases the ¹⁷⁷Lu/^{177m}Lu activity ratios from 0.25 (in equilibrium) to values around 250 2. In our current research we are exploiting the convenience of solid-liquid phase extraction for the separation of the two isomers.
In order to achieve this separation, the surface of amino propyl silica has been chemically modified by reacting it with different chelating groups like : 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (DOTA), and its analogous namely, 2,2′,2”-(10-(2,6-dioxotetrahydro-2H-pyran-3-yl)-1,4,7,10-tetraazacyclododecane-1,4,7-triyl)triacetic acid (DOTAGA-anhydride) and Tri-tert-butyl 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetate (DOTA-tris(tert-butyl ester)). The modified silica surfaces are characterized with several techniques including infrared spectroscopy, solid-state 13C-NMR and TGA analysis. The chelator bearing particles are then packed in a column and loaded with ^{177m}Lu. Periodically the produced ¹⁷⁷Lu is eluted an its quality and the efficiency of the process are quantified by gamma spectroscopy.
This system offers advantages like easy of operation and reliability that together with the low kinetics of the chelating moieties make the system a great candidate to be the final design of the ^{177m}Lu/¹⁷⁷Lu generator.

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Poster SEP / 466

Uranium(VI) binding by pine needles prior and after chemical modification

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Adsorption of uranium(IV) by pine needles prior and after carbonization and following surface oxidation has been investigated by batch-type experiments as a function of various parameters (e.g. pH, [U(VI)]o, I, T), and the adsorbents were characterized by FTIR and SEM measurements. The experimental data have been well fitted by the Langmuir adsorption isotherm and the highest adsorption efficiency is observed for the carbonized and surface oxidized material (qmax= 1.15 mol·kg-1 at pH 3), followed by the non-treated pine needles (qmax= 0.28 mol·kg-1 at pH 3) and the carbonized material (qmax= 0.12 mol·kg-1 at pH 3). The highest U(VI) adsorption observed after surface oxidation of the carbonized material is attributed to the presence of carboxylic moieties, which possess increased affinity for the U(VI) cations and form inner-sphere surface complexes.

Poster SEP / 629

Facile Synthesis and Characterisation of NaBiO3 nano-sheets and Selectivly Adsorbtion of Ce3+

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The NaBiO3 nano-sheets were syntheized by a facile method of oxidizing Bi3+ in alkaline solution. The as-syntheized products were used to adsorb Ce3+ in solution and characterized by XRD, SEM, TEM and XPS. The results show that the synthized products have nano-sheets structure with a size of ~50 nm and high selective adsorption performance on Ce3+. After adsorbtion of Ce3+, all Na cations in NaBiO3 nano-sheets have been fully exchanged and replaced by Ce cations and the chemical state of Ce and Bi were Ce4+ and Bi3+ in the generated products. The XPS analysis shows that Bi5+ species were changed into Bi3+ species in the adsorbtion process of Ce3+. So the adsorption process of Ce3+ contains two process of the ion-exchange between Ce3+ and Na+ cations and the oxidation of Ce3+ species, and the selective adsorption process of Ce3+ ion was achieved by an ion-exchange and redox reaction process.

Poster SEP / 610

Fiber materials based on amine-containing copolymer Eudragit® for radioanalitical applications

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The wide use of radioactive materials in human activities leads to the accumulation of a significant amount of radioactive waste (RW). Therefore search for safe methods of handling with RW and analytical determination of their content in environment becomes the most urgent tasks. At present, uranium mining, nuclear fuel production, treatment of various types of RW, civil and military nuclear powered transport, all of these became major sources of technological radioactive nuclides.1. Furthermore territories contaminated by nuclear weapon testes and exposed owing to emergency situations at nuclear fuel cycle facilities represent a significant radiation hazard. A certain contribution to radioactive contamination of ocean and terrain environment have been made by RW generated during drilling at oil wells and offshore platforms In concern with all of these reasons monitoring of radionuclides content in environment especially in radiation-hazardous areas and objects became an important task [2–8].

Current analytical practice and the concept of treatment of low-level activity liquid radioactive waste require the application of highly efficient materials that could strongly bind radionuclides and allow their isolation, separation and concentration. Under technological operations and in laboratory analysis, polymer ion exchangers, inorganic materials and composite sorbents based on them, as well as complex-forming sorbents (CFS) could be used to adsorb radionuclides [9–14].

Due to functional groups fixed on polymeric or mineral matrices CFS could characterize by the ligand binding mechanism. Methods for CFS synthesis include chemical and non-covalent fixation of functional groups on synthetic and natural matrices, impregnation and preparation of composites based on various matrix. In CFS composition could contain some functional groups that involve metal ions in complexation process in media with different pH values. The most effective CFS are containing diphosphoryl, carbamoylmethylphosphinate, aminophosphinate, primary, secondary, tertiary nitrogen atoms, quaternary ammonium groups [15].

However, analytical determination of actinides still stays very important task because of special chemical properties of U, Pu, Am and the large variation in chemical composition of processing solutions. In this connection, the selection of new CFS for effective analytical determination of radionuclides in complex chemical composition solutions, including natural media, nowadays seems to be a very important task.

Depends on the field of use, a certain set of requirements have been presented to the CFS. Only biosorbents, the adsorption materials based on biocompatible polymers, may apply in drinking water treatment especially water for baby food and in biological fluids purification from heavy metal ions and radionuclides. There are a few such biocompatible polymers and the development of new types of them or the expansion the field of application of already existing ones are really urgent tasks [16,17].

In this decade, the Eudragit® substances which are copolymers of acrylic and methacrylic acids derivatives produced by Evonik, Rohm & Haas Gmbh have become widespread in medical practice. These copolymers are produced under a common brand, but they have different composition and different types of functional groups and therefore can be used not only in pharmacology, but also in other areas, in particular for the creation of adsorption materials. Possessing solubility at pH more than 7, Eudragit S provides the direct acid-sensitive drugs delivery into the intestines bypassing stomach acid media [18,19]. Biodegradable materials based on a mixture of Eudragit E [20] or Eudragit RS[21] with chitosan may become an alternative to synthetic resins for the production of sorption materials for environmental application.

In reference [22] shows the possibility of obtaining a chelate compound of Ni with Eudragit C. This complex was used in chromatography for separation the cell wall components from solutions. In addition, the formation of such a strong complex of a divalent ion with a methacrylate copolymer containing a nitrogen atom allows to consider Eudragit E as the starting matter for the synthesis of metal-ligand complexes [23].

The physical-chemical properties of fine fibered materials Eudragit® (amine-containing copolymer of methyl(butyl)methacrylate Eudragit E (Ed-E) (fig 1.) and quaternary salt dimethylaminoethylmethacrylate Eudragit RS (Ed-RS) were studied in radionuclides adsorption processes.

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The ultra-thin fibers of Eudragit\circledR{} have been obtained in electrospun process from copolymer Ed-E and Ed-RS solutions. Dependence of inorganic salt compound at sorption of $^{233,238}\mathrm{U}$, $^{241}\mathrm{Am}$, $^{239}\mathrm{Pu}$, $^{90}\mathrm{Sr}$, ^{90} on Ed-E и Ed-RS has been shown in 5 g/l $\mathrm{NaNO_3}$, $\mathrm{Na_2SO_4}$, $\mathrm{Na_2CO_3}$ solutions. Less influence gave $\mathrm{NO_3^-}$ ions and distribution coefficient ($\mathrm{K_d}$) has stayed more than 10^2 ml/g for $^{233}\mathrm{U}$, $^{241}\mathrm{Am}$, $^{90}\mathrm{Sr}$. Sulfate ion affected strongly on $^{90}\mathrm{Y}$ interaction. $\mathrm{K_d}$ has been reached 6.0·10^2 ml/g in 5 g/l $\mathrm{Na_2SO_4}$ solution. Effective diffusion coefficients have been determined for fibers with 500 nm, 1200 nm diameters and achieved 3.5·10^{-16} and 3.0·$10-15$ $\mathrm{m^2}$/s respectively in $^{233}\mathrm{UO_2^{2+}}$ adsorption process on Ed-E. Biot numbers stay in range 5-8. This shows us that on adsorption influences external diffusion of radionuclides. Moreover, application of Ed-E as radionuclides flocculant in neutral and weakly acid solutions has been demonstrated. According to flocculation experiments Am(III) and Eu(III) could be separate in 200 g/l $\mathrm{NaNO_3}$ solution.

Thus, a group of Eudragit\circledR{} copolymers may become promising for use in the analytical practice of radioactive substances. Therefore, in this work, we have been studied the possibility of the interaction between Ed-E, Ed-RS and radionuclides $^{233,238}\mathrm{U}$, $^{241}\mathrm{Am}$, $^{239}\mathrm{Pu}$, $^{90}\mathrm{Sr}$, $^{90}\mathrm{Y}$ to determine the field of application in radiochemical analysis.

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Poster SEP / 614

Synthesys and characterization of mesoporous manganese oxide sorbents for removal of strontium radionuclides from aqueos solutions

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Currently, sorption technologies are developed very intensively for the decontamination of liquid radioactive waste (LRW) containing strontium radionuclides (the half-life of 90Sr isotope is 28.79 years). The study of novel sorbents for strontium removal is the subject of research around the world. Due to the chemical and thermal stability and radiation resistance inorganic sorbents are the most promising materials for these purposes 1. One of the main required characteristics for sorbents is the high selectivity towards radionuclides due to the complex composition of LRW. Selective removal of strontium ions from solutions with high salinity, including LRW, is a challenging task due to the presence of competing ions in solutions, especially calcium, magnesium and sodium that reduce the uptake of strontium 2.

There is a number of sorbents that demonstrate sorption-selective properties towards Sr2+-ions in saline solutions, e.g. natural and synthetic zeolites and other raw materials, composite magnetic nanoparticles, manganese oxides, titanates and titanosilicates etc. High exchange capacity, stability in alkaline media allow to consider manganese oxides with a layered and channel structure as a promising materials for the removal of radioactive metal ions from aqueous media. Manganese oxides have a structure of octahedral molecular sieves (OMS) with tunnels 2×2 or 3×3, which are formed of octahedra MnO_6. The dimensions of the tunnels of OMS-2 (cryptomelane with ions K^+) and OMS-1 (type todorokite with ions Ca^{2+} or Mg^{2+}) depend on the cations located inside and are about 0.46 and 0.70 nm respectively [3, 4].

At present work mesoporous manganese oxides with developed specific surface area, layered and/or tunnel structure and high ion-exchange capacity were prepared using non-template sol-gel technique by reducing $KMnO_4$ in aqueous media. Physical and chemical properties of manganese oxide sorbents were characterized by means of X-Ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), differential-thermal analysis (DTA), thermogravimetric analysis (TGA), scanning electron microscopy (SEM) and low temperature adsorption-desorption of nitrogen. Sorption of the ^{85}Sr and ^{90}Sr radionuclides from model LRW was studied in batch mode. The influence of inorganic ($MnCl_2$ and H_2O_2) and organic reducing agent (ethanol and polyvinilalchohol), nature of „host"ions (Na^+, K^+ and Ca^{2+}), conditions of sol-gel process (time, temperature) and calcination temperature on the chemical and phase composition, porous structure, surface morphology and sorption properties on sorption properties were studied for prepared manganese oxides.

The results of sorption study for manganese oxides prepared via sol-gel technique by reducing $KMnO_4$ in ethanol/aqueous media is presented in tables 1 and 2. All sorbents are characterized by developed specific surface area (A_{BET}) –up to 293 m2/g. Sample 2 demonstrated the highest sorption (K_d ^{90}Sr=10.6×103 cm3/g) and selective ($D_{Sr/Ca}$=99.8) properties towards ^{90}Sr in the presence of 0.01M $CaCl_2$ background electrolyte. It was established that manganese oxides with tunnel structure (samples 2,3 and 5,6) had higher affinity towards ^{90}Sr radionuclide in comparison with layered modifications (samples 1 and 4). Sorbents in Na^+-form is more effective than in Ca^{2+}-form due to lower charge and higher mobility of Na^+ ions.

Table 1. Synthesis conditions and characteristics of manganese oxide sorbents prepared via sol-gel technique by reducing $KMnO_4$ by ethanol in aqueous media.

Sample	Temperature of sol-gel synthesis, °C	Time of sol-gel synthesis, h	Calcination temperature, °C	Ion-form	ABET, m2/g
1	25	5	150	K+	208
2			350	Na+	203
3				Ca2+	165
4	80	48	150	K+	284
5			350	Na+	293
6				Ca2+	213

Table 2. Sorption properties of manganese oxide sorbents towards ^{90}Sr (0,01 M $CaCl_2$, pH=6.0, V/m=200 cm3/g).

Sample	Ion exchange capacity for Ca2+, mmol/g	Distribution coefficient, Kd (90Sr) ×10-3, cm3/g	Separation coefficient, DSr/Ca
1	0.852	0.75	5.6
2	0.739	10.6	99.8
3	<0.01	5.77	-
4	0.243	1.23	47.7
5	0.44	3.83	66.3
6	-	2.89	67.2

The relationship between the conditions of preparation, physical-chemical and sorption-selective properties of sorbents allow to define general regularities and approaches to the directed synthesis of highly selective materials of strontium radionuclides. The comparison study of developed manganese oxides with others sorbents (zeolites, zirconia and titanosilicates) has showed their higher sorption and selective properties towards strontium radionuclides.

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Poster SEP / 759

Fluorinated carbonates as new diluents for extraction of f-elements

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Fluorinated diluents have been considered as possible alternative for hydrocarbons. Among their advantages one can list particulary their chemical stability and in many cases high polarity. Unfortunately, the best studied polar fluorinated diluents such as *meta*- nitrobenzotrifluoride (F-3, MNBTF) or phenyl trifluoromethyl sulfone (FS-13, PMTS) have density about 1.41 g/cm^3 and higher solubility in acid solutions than the traditional hydrocarbons. Recently, a new class of fluororganic compounds –carbonates of fluorinated alcohols was proposed as potential diluents. Carbonates of 2,2,3,3-tetrafluoropropan-1-ol (BK-1) and 2,2,3,3,4,4,5,5-octafluoropentan-1-ol are available and rather cheap; they have high boiling point, high density (1.578 and 1.722 g/cm^3, respectively) and low solubility in water.

In the presented work, extraction ability of two perspective extracting compounds, CyMe$_4$-BTBP and CyMe$_4$-BTPhen, considered for the SANEX process dissolved in the new fluorinated diluent BK-1 was tested to assess the extractability of trivalent lanthanoids and minor actinoids. The dependences of distribution coefficient D$_{Am}$, D$_{Eu}$, and D$_{Cm}$ and separation factor SF$_{Am/Eu}$ and SF$_{Am/Cm}$ on HNO$_3$ concentration (0.001 –4 mol/L) were evaluated. The SF$_{Am/Eu}$ values higher than 300 were achieved in 2 mol/L and 4 mol/L HNO$_3$ for CyMe$_4$-BTPhen. The highest SF$_{Am/Cm}$ value (about 5.8) was achieved in 4 mol/L HNO$_3$ for CyMe$_4$-BTPhen as well. The extraction of studied Ln(III) and An(III) by the neat diluent (without extraction compound) showed that there is no extraction by diluent itself. Moreover, kinetics of Am(III), Eu(III) and Cm(III) extraction was evaluated and will be presented.

Poster SEP / 645

Modified bio-sorbent wood-decay fungus Fomes fomentarius for pre-concetration of 137Cs in water samples

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The bio-sorbent used in the study was prepared from wood-decay fungus Fomes fomentarius. The chemical modification of bio-sorbent with ferric hexacyanoferrate(II) (Prussian blue) for cesium sorption studies was used. The surface area was calculated by adsorption of argon at 77 K, in accordance to the method of Brunauer et. al, 1938. The surface area of the 0.5 mm grains was 39 m2 g-1. In this study, the effect of pH in the range of 1.0–13.0 on cesium sorption onto modified bio-sorbent was studied. It was found, that the bio-sorbent has a high 137Cs absorbability in the wide range of pH, even in the solutions with high ionic strength. The maximum sorption capacity of the bio-sorbent was 50 mg Cs+ / g sorbent. The effect of competing ions Na+, K+, Ca2+ and Mg2+ was tested. It was found, that the tested cations had no a significant effect on the sorption of 137Cs. It was found, that the most suitable medium for the elution of 137Cs from the bio-sorbent was HNO3 with a concentration higher than 7 mol·dm-3. The developed method for 137Cs determination by using modified bio-sorbent was applied to various types of waters delivered by Water Research Institute in Bratislava. 137Cs was measured on HPGe detector and 137Cs activity was less than the

minimum detectable activity (0.004 Bq L-1) with the exception of water Sokolianky –Tornyosnemeti. The tested modified bio-sorbent is suitable for pre-concentration of 137Cs in water.

Poster SEP / 827

Radiological Characterization of a low and intermediate-level Radioactive Waste Samples from Research Reactor

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The radioactive solid wastes from research reactor and facilities consist of soft wastes, disassembled equipment, laboratory supplies used during research activities, filters and ion exchange resins used in the purification of gas and liquid effluents, and other various type of combustible materials. According to the regulation for low and intermediate-level radioactive wastes in Korea, gross alpha, 3H, 14C, 55Fe, 59Ni, 63Ni, 90Sr, 94Nb, 99Tc, 129I, and gamma emitters (e.g., 58Co, 60Co, 94Nb, 137Cs, and 144Ce) should be quantitatively determined for the disposal treatment.

In this study, analytical procedures were developed to quantitatively determine the radio-nuclides for the various type of radioactive solid waste samples. In the case of radioactive soft wastes, it is difficult to obtain the representative in the whole sample volume due to inhomogeneity for the radionuclide's contamination. Therefore, in order to assure the homogeneity of the sample, the whole samples were cut and mixed repeatedly. The process of sample preparation and measurement is composed of four main processes: direct measurement (gamma emitters), alkali digestion (129I), acid digestion (3H and 14C) and sequential separation and purification using extraction chromatography (e.g., TRU: gross alpha and U isotopes, DIBK resin: 55Fe, Ni resin: 59Ni and 63Ni, SR resin: 90Sr, and TEVA resin: 99Tc). The validated process were applied to radiological characterization for the combustible waste samples from HANARO research reactor and facility of KAERI. The validation results used the standard spiked samples revealed that the methods could be applied for rapidly and satisfactorily recovering the specific target nuclides from samples with a high degree of accuracy and precision.

Poster SEP / 847

Removal of radionuclides from aqueous solutions by composite nanomaterial

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Radioactive waste contains large amounts of hazardous radionuclides. Among them, long lived radionuclides of 137Cs (half-life-time of 30 years) and 90Sr (half-life-time of 28 years) are considered as the most dangerous to human health and environment. The ion exchange technology is one of the most commonly used methods for safe treatment of radionuclide waste.

In the presented work, new composite nanomaterial (TSMW) based on sawdust (wood dust) and titania, has been prepared and tested for sorption of 134Cs and 85Sr from aqueous solutions. The morphology and structure of TSMW were characterized by XRD, BET and SEM techniques.

The batch sorption experiments were used for evaluation of composite nanomaterial retention properties. The sorption of radionuclides onto TSMW sorbent was found to be pH dependent with the uptake increasing with pH. The efficient removal (more than 99%) of Sr(II) was achieved from neutral and base solutions. In case of Cs(I), the uptake was less efficient (70%). The evaluation of equilibrium sorption data for all studied ions indicated that the sorption processes onto TSMW sorbent agreed better with Langmuir isotherm model suggested that sorptions are monolayer.

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Poster NAM / 714

Application of chromatographic techniques in radiocarbon dating

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Chromatographic techniques are applicable in a wide spectrum of scientific fields and are also advantageous in different kinds of interdisciplinary research. By these techniques we are able to analyse compound composition of unknown samples, purity of solvents but they can be applied also for the preparation of analytes with subsequent analysis of stable or radioactive isotopes. Based on obtained results, we can also evaluate the effectiveness of sample preparation routines.

One of the promising application of chromatography is in the field of radiocarbon dating. Using different chromatographic techniques, it is possible to eliminate or remove possible contaminating agents which could distort final results of 14C activity. One of the example is preparative GC (Gas Chromatography) which could isolate e.g. fatty acid from the mixture of lipophilic compounds extracted from bones or archaeological pottery. Further, we can use micro-preparative column chromatography for the removal of humic and fulvic substances originating from soil or for the isolation of steroidal fraction (non-sapofiniable matter) from trans-esterified fat. To examine these techniques we sampled bones from individuals of different age and known date of death. Extracted fats were then separated to neutral (tri-, di- and monoacylgycerols) and steroidal fraction. An aliquot of the neutral fraction was then transesterified obtaining fatty acid methylesters (FAMEs) which were subsequently separated by preparative GC system Agilent-Gerstel to obtain pure C16:0 and C18:0 FAMEs. Isolated FAMEs where then deesterified in acidic environment back to free fatty acids. Steroidal fraction was isolated by two approaches, either by sapofinication of extracted fat and isolation of non-sapofiniable matter or by eluting on micro-preparative silicagel filled glass column (micro-preparative column chromatography) using a suitable elution system. Purity of obtained analytes and solvents was examined by GC-FID (Flame Ionisation Detector) and GC-MS (Mass spectrometry) techniques. Further, all analytes were stripped off solvents quantitatively and processed for the purpose of measuring 14C activity. The analytes were combusted to CO2 which was subsequently graphitized by catalytic reduction using metallic zinc. Samples of graphites were then measured by AMS (Accelerator Mass Spectrometry) using a MICADAS system at Debrecen (Hungary). Preliminary results show, the isolation of fat fractions from bones, including steroidal fraction, could be useful for highly contaminated bone samples –by soil contamination or by plastificators from sample packaging. Nevertheless, the approach of sample processing should be considered for each sample separately.

Poster NAM / 581

Combination of an automated isolation method and gamma spectrometry for determination of Fe-55 in complex samples

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Radioactive Fe-55 is generated by neutron activation of stable iron and, therefore, it can be found in materials from nuclear power plant, which should be decommissioned. It decays by electron capture and emits x-ray of 5.9 keV (24.5% probability). Because of the low energy of emitted ray, the attenuation effect is large and it occurs already in the sample itself, therefore it is the most often measured via liquid scintillation counting (LSC). However, modern HPGe detectors allow the determination of gamma emitters in a wide range of energies (3 keV - 3 MeV). In case of LSC, prior determination, isolation of pure iron from the sample is essential. Most of the samples may contain higher amounts of iron, so chemical and color quenching effects are important since they can significantly decrease detection efficiency. Therefore, within this study, especially for samples with higher amounts of iron, the method for Fe-55 determination by gamma spectrometry is developed. In this case, chemical isolation from the sample is also desirable to decrease attenuation effect and if present, to remove Co-58 and Mn-54 which emit x-rays in the same range (5.4 keV for Mn-54 and 6.4 keV for Co-58). The geometry of the sample is optimized to minimize the attenuation effect. For the chemical isolation of Fe-55 from complex samples an automated method which includes separation of iron from interfering elements on Sr resin or Pb resin column will be described (Sr and Pb resin consist of 4,4'(5')-di-t-butylcyclohexano 18-crown-6 in 1-octanol or isodecanol loaded on an inert chromatographic support). The Sr and/or Pb resin are chosen since they can be used for simultaneous separation of strontium isotopes 89,90Sr which are also present in different kind of materials from nuclear power plant. In almost all published methods, isolation and determination of Sr-89,90 and Fe-55 are separated, while here it will be shown how Sr and/or Pb resin enables simultaneous isolation of 89,90Sr and Fe-55 in one step. This work will also include optimization of the sample preparation method in the sense of achieving a detection limit of Fe-55 as low as possible.

Poster NAM / 612

Study of new luminophores for use in modern scintillation cocktails

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This paper deals with the study of 1-phenyl-3-(2,4,6-trimethyl-phenyl)-2-pyrazoline, 2,2´,7,7´-tetrakis(N,N-diphenyl-amino)-9,9´-spirobifluorene, 2-(4-biphenylyl)-6-phenylbenzoxazole and 9,9´-bifluorenyl as possible replacements of the existing and most widely used luminophore (2.5-diphenyloxazole (PPO)) in contemporary scintillators. The study was focused on the determination of emission wavelength, concentration optimization, the influence of the wavelength shifters POPOP and bis-MSB. In addition, the response to alpha and beta radiation were measured, in particular, the pulse length index (PLI) was determined. Last but not least, the effect of emulsifiers on the shape of the spectrum and the solubility of the aqueous phase was tested. The results obtained were compared with the commercially available Aqualight scintillation cocktail.

Poster NAM / 548

Electronic Structure Studies of Th Systems using X-ray and theoretical Methods

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The content of thorium (Th) on the earth's crust is three to four times higher than the uranium, so the idea of using thorium as the main component of nuclear fuel is currently developing.
In recent years High Energy Resolution Fluorescence Detected (HERFD) and Resonant Inelastic X-ray Scattering (RIXS) were shown to be a highly valuable tool for investigation of the electronic structure of actinides[1,3]. We have studied the electronic structure of several thorium systems by means of HERFD and RIXS and compared the results to X-ray photoemission spectroscopy (XPS) investigations. The research was conducted at Rossendorf Beamline at European Synchrotron Radiation Facility (ESRF), dedicated to actinide science. The recently upgraded ROBL beamline at the ESRF provides now a unique opportunity to study actinide materials by several experimental techniques: Extended X-ray Absorption Fine Structure (EXAFS), X-ray Absorption Near Edge Structure (XANES), HERFD, RIXS and X-ray diffraction techniques2.
The recorded experimental spectral features were characterized using ab initio theoretical calculations (FEFF 9.6). The combined experimental and theoretical data provide a new insight into a fundamental understanding of thorium chemistry significant for topics of high societal relevance.

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Poster SEP / 829

Studies on New Bis-Triazine Ligands in Spent Nuclear Fuel Reprocessing: Tuning the Aliphatic Part and its Effects on Extraction Properties

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The removal of the minor actinides americium(III) and curium(III) from spent nuclear fuel is currently being studied worldwide as part of an ongoing strategy to reduce its long-term heat load and radiotoxicity. This strategy aims to close the back end of the nuclear fuel cycle and ultimately increase the safety and sustainability of civil nuclear energy. Within the framework of several EU research programs, promising solvent extraction processes have been developed that involve the selective extraction of the trivalent minor actinides from the lanthanides from aqueous nitric acid into an organic diluent containing a hydrophobic bis-triazinyl pyridine (BTP), bis-triazinyl bipyridine (BTBP) or bis-triazinyl phenanthroline (BTPhen) ligand. Recent studies have focused on modified ligands containing electron donating or withdrawing substituents attached to the aromatic rings of these ligands, and their effects on minor actinide extraction performance and actinide/lanthanide selectivity.

In contrast, less emphasis has been placed on modifying the aliphatic part of these ligands. Here we present our recent studies on modified bis-triazine ligands (BTP, BTBP and BTPhen) containing 5-membered rings appended to the outer triazine rings, instead of 6-membered rings, and the effects of this modification on ligand solubility, minor actinide extraction performance and actinide/lanthanide selectivity. Using a combination of solvent extraction experiments, NMR titrations and time-resolved laser fluorescence spectroscopy, it has been shown that changing the ring size of the aliphatic part

of these ligands has subtle effects on metal ion speciation, complex stability and minor actinide extraction performance and selectivity.

Poster NAM / 528

A Comparison Study of Simulation for Neutron Induced Prompt Gamma Using MCNPX and PHITS

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PGAA (Prompt Gamma Activation Analysis) is an established nuclear analytical technique for the non-destructive determination of elemental and isotopic compositions. The neutron activation analysis research group in KAERI (Korea Atomic Energy Research Institute) has been developing a fake gold bar detection method using a PGAA. However, a real experiment to verify the detection method was not performed because of the seismic retrofit for HANARO. As an alternative method, a computer simulation based on the Monte Carlo method was used to confirm the detection method. Prompt gamma-rays are the most important element for a PGAA. MCNPX and PHITS, Monte-Carlo radiation transport simulation codes, have succeeded in simulating a prompt gamma-ray spectrum. However, the prompt gamma-ray spectrum generally simulated by MCNPX did not show accurate gamma energy peaks in our previous study. Thus, the simulation was conducted in two separate files to describe the accurate prompt gamma energy peaks. The neutron transport and prompt gamma ray production rate in the sample were first calculated. Photon transport for the detection of prompt gamma rays emitted from the sample was then simulated. The other code, PHITS (Particle and Heavy Ion Transport code System), is a general purpose Monte Carlo particle transport simulation code developed under collaboration between JAEA, RIST, KEK, and several other institutes. According to Ogawa et al., a new theoretical model to simulate gamma de-excitation nuclei was developed based on the Evaluated Nuclear Structure Data File (ENSDF) in FHITS. The model is applicable for neutron capture products and spallation products of 1071 nuclear species from Li to bk. In this study, the neutron induced prompt gamma-ray spectra are calculated after a neutron beam is irradiated onto the target sample using PHITS. The simple geometry of the neutron irradiation system in the PHITS simulation is modeled after the fake gold detection system. The track of neutrons irradiated onto the sample is simulated using a T-track tally, and the prompt gamma-rays emitted from the sample are calculated using a T-cross tally. Finally, a comparison of PHITS and MCNPX code in PGAA is described, and an optimal method for simulation of neutron induced prompt gamma is discussed.

Poster NAM / 622

Initial study on determination of uranium in wiped samples by Total reflection X ray fluorescence spectrometry

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The analysis of wipe samples is of great importance in nuclear safeguards. Total reflection X ray fluorescence spectrometry as a trace nondestructive analysis method can preliminarily analyze the wipe samples and provide references for screening, making it easier to make follow-up analysis plans. The

method of determining uranium in wiped samples by full reflection X ray fluorescence spectrometry was preliminarily discussed in the experiment, and the simulated samples were measured.
The mixed standard uranium solution was prepared with six elements including Pb, Ni, Zr, Fe, Zn, Cr, etc., and Pb, Ni and Zr were obviously interfered in SIMS. Fe, Zn and Cr are common elements in the environment, which represent the three elements of high, medium and low levels in the environment. The internal standard element Ga was added for quantitative analysis. 10 μL the mixed standard solution was taken and added to the sample plate and placed on the heating plate in the 60-70 ℃ for drying. The total reflection X-ray analyzer was used. The results showed that the relative error of the method was less than 10% when the uranium content was less than 50ng. Under 1000 ng for the amount of impurities, the X-ray features of each element peak width is narrow, and the characteristics of X-ray wavelength absorption edge is not very close, so there is no basic interference between each element. The measurement errors of Pb, Ni, Zn, Fe and Cr were all within 10%. The determination of zirconium element is high, which may be due to the similarity between the characteristic X-ray wavelength and molybdenum excitation of zirconium, and th interference to had been when measuring of zirconium. The accurate measurement of Pb and Ni can provide useful information when measuring the uranium particles by SIMS.

Poster NAM / 653

Activation analysis screening of hazardous elements in a dump soil in the Central Bohemia Region

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Most hazardous elements contribute significantly to contamination of agricultural soils. Their content in the soil increases above background level due to anthropogenic influences as industry ballast and transportation and energy production, especially in the surface layer of the humus horizon. Accumulation of hazardous elements in the soil has a significant ecological impact on the speed of the elemental cycling. Due to binding to primary agricultural products they can pass into the food chain.
Soil samples from a dump in the Central Bohemia were collected for determination of selected hazardous elements. Instrumental neutron activation analysis (INAA) and instrumental photon activation analysis (IPAA) were used for the assay of elements such as As, Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb, V, and Zn. Results obtained by the radioanalytical methods were compared with the regulatory values applicable in the Czech Republic.
As expected, the content of hazardous elements in dump soils in the Central Bohemia increases. The amount of these elements is several times higher than the maximum permissible limits. It suggests their anthropogenic origin, probably from the operation of former coal mines and the Poldi Steelworks. Most of the hazardous elements are included in the group of extraneous substances that can significantly contribute to contamination of agricultural soils. A potential long term risk is expected because free access to this area is not disabled.

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Poster NAM / 863

Building of the first AMS laboratory in the Czech Republic –Extension of the CANAM infrastructure

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Building of the first AMS laboratory in the Czech Republic has recently been started in co-operation of the Nuclear Physics Institute of the Czech Academy of Sciences (NPI), the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague and the Institute of Archaeology in Prague of the Czech Academy of Sciences. The laboratory will be located on the premises of NPI in Řež and equipped with an accelerator with maximum terminal voltage 1 MV, a complex of new laboratories, including the class ISO 7 clean laboratory, and offices. The AMS system should be capable of measuring low levels of 14C, cosmogenic radionuclides 10Be and 26Al, actinides, namely isotopes of U and Pu, and selected fission products, e.g., 129I.
The application fields of 14C measurement will involve
• Radiocarbon dating in archaeology, paleoecology and other disciplines
• Bomb peak dating for environmental protection and forensic medicine
• Environmental and geological studies, detection of environmental changes
• 14C in the atmosphere and in the vicinity of nuclear power plants
• Atmospheric 14CO2 and 14C environmental levels (reference background values)
• Green vs. fossil carbon 14C in motor fuels (HVO), pharmaceuticals, foods and chemical industry
• Microdosing of pharmaceuticals labeled with 14C
The applications of 10Be and 26Al measurement will involve
• Geochemistry and paleoclimatic change studies (sediments, rocks, glaciers, …)
• Age determination of meteorites and other extraterrestrial materials
• Determination of cosmogenic nuclides
The measurements of actinides and fission products will be applied for
• Geochronology, environmental control and monitoring
• Natural fission and neutron reactions
• Nuclear forensics and Safeguards, undeclared nuclear activities, nuclear weapon explosions
The new AMS laboratory will complement the existing NPI infrastructure Centre of Accelerators and Nuclear Analytical Methods (CANAM) that comprises at present three laboratories–Laboratory of Cyclotron and Neutron Generators, Laboratory of Tandetron and Neutron Physics Laboratory. The new AMS laboratory will become the fourth laboratory of CANAM and should be operational at the beginning of 2020.

This work is realized within the EU, ESIF, OP RDE, MEYS project Nr. CZ.02.1.01/0.0/0.0/16_019/0000728: "RAMSES - Ultra-trace isotope research in social and environmental studies using accelerator mass spectrometry".

Poster NAM / 664

Precise Determination of U-235 and Ra-226 Photopeak Intensities in Naturally Occurring Radioactive Materials Using Optimization Subroutine Function

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The need for determining the U-235/U-238 isotopic ratio in a naturally occurring radioactive material (NORM) is increasing to warn a leakage of uranium into the environment. During the process of seeking a non-destructive method of analyzing natural matrices for uranium isotopes U-235 and U-238, a major source of uncertainty was found to be the gamma-ray intensities. Ra-226 and U-235 are always found in the presence of each other in soil samples, and each has primary gammas whose energies differ by only 0.496 ± 0.014 keV. The least squares fitting method, using the MIGRAD function optimization subroutine in ROOT framework was implemented to obtain reliable values for the two photopeak intensities of Ra-226 and U-235. This framework was developed to examine the effect of increasing or decreasing the number of degrees of freedom in the fit on the confidence intervals of the fitted intensity parameters (heights of the two Gaussians). An IAEA U1GX natural uranium reference material known to be in secular equilibrium has been counted using an ORTEC HPGe detector with the sample in a Marinelli beaker geometry. It was found that the increased number of degrees of freedom approach (i.e., with a reduced number of free parameters) gives more accurate results for the photopeak intensities, with the determined ratio of U-235 to Ra-226 intensities having discrepancies in the range 0.27-2.8% from the expected ratio for natural uranium that is in secular equilibrium.

Keywords: Uranium, HPGe, NORM, Gamma-ray.

Poster NAM / 745

Preparation of Fluoride Target Matrices for U-236 AMS Measurement

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In recent years, radionuclide ²³⁶U has become a significant analytical tool. Due to it's mostly anthropogenic origin, the main purpose of it's determination resides in tracing of human nuclear activities, though it can be used for studying plenty of natural phenomena like natural fission reactors or deep sea currents as well. Since natural samples usually contain only a trace amount of this nuclide, a highly accurate analytical method like accelerator mass spectrometry (AMS) is required. Target sample preparation involves a pre-concentration of raw water or soil eluate obtained from the natural matrix, followed by conversion of uranium contained to a desired chemical form, commonly a uranium oxide. Apart from the oxides, fluoride target matrices have lately become a significant point of interest, particularly due to use of monoisotopic fluorine that provides a considerable decrease in isobaric interferences occuring in the process of analysis. Several methods of fluoride target preparation have been developed and published recently 1, though none of them provided a pure fluoride matrix without contamination by oxygen. Therefore, a new method of fluoride target sample preparation for ²³⁶U is being developed. The main objective is a substantial reduction of the oxygen content while maintaining the procedure itself simple enough for application in routine analyses. The proposed preparation method involves addition of a suitable carrier for uranium to it's acidic solution, followed by reduction to +IV oxidation state and coprecipitation with the carrier in a form o fluoride. Uranium separation yield for the samples prepared exceeds 99 %. Particular ion currents, ionisation yields and efficiencies are currently being measured at AMS Vera, Vienna.

1 Xianggao Wang, Kejun Dong, Ming He, Shaoyung Wu, Shan Jiang (2013) Nucl. Tech. 182, 235-241

Poster NAM / 818

Application of graphene oxide (GO) and chemically modified GO for radioanalytical separations and preconcentrations.

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Graphene oxide (GO) has two important characteristics: (I) it can be produced by cost-effective chemical methods, and (II) it is highly hydrophilic and can form stable aqueous colloids to facilitate the assembly of macroscopic structures by simple and cheap solution processes. In this work, the possibility of using of GO and GO modified by Mn and Fe have been examined. In order to find potential separation possibilities of prepared sorbents, their extraction behaviour was investigated by batch equilibration technique with use of radioactive tracers or multielemental standards in dependence on pH at room temperature. The sorption of individual elements strongly depend on the pH. In acidic solution, only few elements show the affinity to prepared sorbents: Pa-233, Au, Mo, Ni. Alkali metals are not retained on the resins under all pH conditions. The dependence on the ionic strange was also checked- no influence for the sorption behavior was observed. Obtained results have been verified by column experiments. Due to small dimensions of nanoparticles, there are great difficulties with column preparation.

These sorbents have found application for the separation, preconcentration and determination of Am-241, Co-60, Eu-152, Ba-133 from aqueous solutions.

Also, the analytical procedure for the determination of trace amounts of Pt in tissues and body fluids by RNAA, with the separation of Au-199 on GO-Mn sorbent, was proposed.

Poster SEP / 849

Sorption characteristics of carbon-containing natural materials for removing Tc(VII) from aqueous solution

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Permeable reactive barriers (PRB) enable physical, chemical or biological in situ treatment of contaminated groundwater by bringing it into contact with reactive materials. The reactive material is inserted underground in a natural aquifer and intercepts the pollution plume as it is carried along within the aquifer. Thus the contaminants are treated with neither wholesale soil excavation nor water pumping. This cost-effective clean-up technology has much less impact on the environment than other methods, appearing generally more economical profitable over the long term in comparison with other methods.

In this study sorption characteristics of shungite, lydite and activated carbons as natural material that can be used for creation a part of complex multi-layer PRB were specified. Sorption coefficients (Kd) and kinetic characteristics for shungite, lydite and activated carbons for Tc-99 were obtained. Speciations technetium on these materials were investigated by means of the sequential extraction method. Sorption isotherms for technetium uptake by shungite were obtained.

Was shown that uptake of Tc(VII) by shungite depend both on specific form of carbon occurring in shungite and numerous associate minerals, especially pyrite and pyrrhotite.

Poster NAM / 831

Multielement content of Canadian food samples by instrumental neutron activation analysis

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An instrumental neutron activation analysis (INAA) method was developed for the determination of some major, minor and trace elements in 93 different Canadian food samples. One portion of the sample was irradiated for 1 min at a neutron flux of 4 x 1011 cm-2 s-1 in the Kyoto University Reactor (KUR), Japan. Gamma-ray spectra of the irradiated samples were recorded after 1.5 min decay for 5 min using a Compton suppression Ge detector system. Levels of Br, Ca, Cl, Cu, K, Mg, Mn, Na, and Ti were measured using their short-lived nuclides. Then, another portion of the samples was irradiated for 1 h at a neutron flux of 2 x 1013 cm-2 s-1 in KUR. Gamma-ray spectra of these samples were recorded after one-month decay for 20-30 min using a Ge detector system. Levels of Ag, Ba, Co, Cr, Cs, Fe, Rb, Sb, Sc, Se, and Zn were obtained using their long-lived nuclides. The INAA method used was validated using NIST 1566b Oyster Tissue, NIST 1570a Spinach Leaves, NIST 1575 Pine Needles, NIST 1577b Bovine Liver, and NRCC TORT-1 Lobster Hepatopancreas certified reference materials. Our values agreed well with the certified values. Details of the method and results will be presented.

Poster NAM / 859

Separation and isotope ratio measurements of lanthanides and uranium, using HPIC-SF-ICP-MS, for characterization of spent nuclear fuel

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Elemental and isotopic characterization of spent nuclear fuel is of crucial importance in nuclear reactor facilities for various purposes, including the management of spent nuclear fuel 1. Characterization of spent nuclear fuel samples requires highly sensitive and precise mass spectrometric techniques, such as sector-field inductively coupled plasma-mass spectrometry (**SF-ICP-MS**) and thermal ionization mass spectrometry (TIMS) to accurately measure the isotopic compositions of uranium (**U**), plutonium (**Pu**) and neodymium (**Nd**), especially to determine the burnup 2. A major issue in mass spectrometry, i.e. spectral overlap of isobaric nuclides, can be overcome by the separation of the analyte elements from one another. Unlike TIMS, SF-ICP-MS can be hyphenated with online separation methods [3], e.g., high performance ion chromatography (**HPIC**), thus substantially simplifying the analytical protocol and enhancing the sample throughput.

A Dionex ICS5000+ dual quaternary gradient pump HPIC instrument, equipped with Dionex CS5A mixed bed ion exchange chromatography column, and CG5A as guard column, was connected to both a UV-Vis detector and an Element 2 sector-field ICP-MS instrument, partly installed inside a glove-box. Separation of **lanthanides** (La, Ce, Pr, Nd, Sm, Eu, Gd, Lu) and uranium from the concomitant matrix components and from one another was achieved when 0.1 –0.3 M oxalic acid and 0.5-1.0 M HCl mobile phases were used in gradient and isocratic elution programs respectively. Chromatograms obtained in this way, as well as the analytical challenges involved in optimizing the **HPIC-UV-Vis** [5] and **HPIC-SF-ICP-MS** methods [6] will be presented. In addition, an overview of both data acquisition parameters (incl. the number of nuclides monitored per run, dwell time, number of replicates, integration window, etc.) and analytical method characteristics (incl. calibration and method(s) to correct for mass discrimination while handling transient signals) to arrive at isotope ratio determinations with the best obtainable accuracy and precision will be described.

In conclusion, HPIC hyphenated to a SF-ICP-MS unit in a glove-box, for the separation and isotope ratio measurements of lanthanide elements and uranium, will be presented. Isotope ratio measurement uncertainties, determined at a later stage, will allow the evaluation of the HPIC-SF-ICP-MS method against TIMS. Method validation will be done using environmental samples.

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Poster NAM / 821

Determination of the gross activity of uranium, plutonium, americium and strontium in environmental samples using solid-state scintillation

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Rapid determination of selected gross alpha and beta emitters in environmental matrices by solid-state scintillation technique is discussed. This method is based on sample treatment using microwave reactor and direct measurement of digested products using powder scintillator and alkaline solution as a substitute for traditional liquid scintillation cocktail. The selected group of radionuclides was chosen with respect to their use in nuclear industry, high radiotoxicity, and the possibility of potential misuse. The work aimed at verifying the connection of microwave decomposition using alkaline solution with solid-state scintillation using a powder scintillator YAP:Ce together with an alkaline medium.

Poster NAM / 837

Potentiometric uranyl-selective sensor based on a new supramolecular composition

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A new supramolecular composition containing a clozoborane salt UO2 [B10H9S(C18H39)]2 , polyvinyl chloride (PVC) as a matrix and tris (2-ethylhexyl) phosphate. The membranes of various compositions with different ratios of membrane components were investigated.
The membrane with the best electroanalytical characteristics has the following composition:
Cs B10H9S(C18H37)2 : 1 –3 % (mass),
tris (2-ethylhexyl) phosphate: 65 –72 %
PVC: 27 –32 %
The developed electrode had a linear potential dependence in the range not worse than pUO22+ = 2 –6, The detection limit for uranyl ions was C(UO22+) = 1,6*10 -7 M. The measured selectivity factors are given in the table:
Anion Cs+ Rb+ K+ Na+ Sr2+ Ca2+ Mg2+
Кselect 8•10-5 8,2•10-5 9•10-5 1,1•10-4 3,0•10-5 1,05•10-4 2,2•10-5

As follows from the values of the selectivity coefficients, the developed electrode showed a very high selectivity with respect to uranyl ions in the presence of alkaline and basic alkaline earth cations. From the analysis of selectivity data, it should be noted that the behavior of the proposed supramolecular system does not obey the Hofmeister series. It has been established that even 10 + 4-fold excesses of alkali and alkaline earth ions do not significantly affect the membrane potential. The established series of selectivity series looked as follows:
UO22+ > Li+ > Na > K > Cs > Ca > Mg

Poster NAM / 860

Use of microtron for photon activation analysis

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The Microtron MT 25 is a cyclic electron accelerator with a Kapitza type resonator. The accelerator is located in a laboratory belonging to the Nuclear Physics Institute of the CAS. The Microtron MT25 serves as a source of relativistic electrons (primary electron beam), secondary photon beams (bremsstrahlung) and neutrons from nuclear reactions. Photon beams are frequently used for instrumental photon activation analysis of geological, biological, environmental and others samples. This method allows non-destructive determination of number of elements not determinable by neutron activation analysis or determination some elements with a better sensitivity then INAA. The laboratory is equipped with coaxial HPGe detector and multichannel analyzer. In microtron laboratory was recently installed fully automatic pneumatic post for fast transport of samples between irradiation positions and HPGe detector. This system expands the possibilities of photon activation analysis, because it enables determination of samples with short half-life.

Poster NAM / 866

Performance comparison of experimental liquid scintillation cocktails

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In this article the properties of four new luminophores, from the group of bifluorenes and other substances with multiple aromatic rings, were measured and studied for possible use in liquid scintillation, namely 1-phenyl-3-(2,4,6-trimethyl-phenyl)-2-pyrazoline, 2,2´,7,7´-tetrakis(N,N-diphenylamino)-9,9´-spirobifluorene, 2-(4-biphenylyl)-6-phenylbenzoxazole and 9,9´-bifluorenyl. The measurement was also done for the most commonly used luminophore (2,5-diphenyloxazole (PPO)). To improve counting efficiency the wavelength shifters were added to scintillation cocktail, in particular POPOP and bis-MSB. The main effort was focused on the determination of sample load capacity of the cocktail, especially in the acidic, alkaline and neutral environment. Furthermore, the detection limit and FOM were determined. Quench resistance, an important factor responsible for a reduction in counting efficiency, was determined via the addition of CCl4 and PBS. Lastly, the stability and diffusion of the cocktails were measured. The results were compared with the commercially used Aqualight scintillation cocktail.

Poster SEP / 861

Deep Eutectic Solvents: Promising Media for advanced hydrometallurgical processes for actinides separation

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Deep eutectic solvents (DESs) have emerged as an interesting type of neoteric solvents and have shown their usefulness as environmentally benign sustainable alternative to the conventional organic solvents *1.* DESs are eutectic mixtures of Lewis and Brönsted acids and bases, which can contain a variety of anionic and cationic species *2.* The use of DESs in chemical synthesis and chemical processes stems from their characteristics of green and inexpensive solvents, biodegradable, catalytic activity in some cases. Their simple preparation and the quantitative yields render DESs attractive for academic and industrial research *1.* In particular, DESs have extensive applications in the extraction and separation of a wide range of analytes.
In this communication we reports on the use of some newly synthesized DESs as innovative alternative to the recently proposed imidazolium based ionic liquids (ILs) for processing nuclear waste *3.* To this purpose, new DESs choline acetate-based products, designed in accordance with the CHON rule, were synthesized and fully characterized (HR-MAS NMR, FT-IR and DSC). The new DESs were checked as co-solvents in liquid-liquid extraction tests simulating hydrometallurgical processes for the recovery of minor actinides from the Plutonium and URanium EXtraction (PUREX) raffinate. In particular, Selective ActiNide EXtraction (SANEX) based processes were considered as reference. Therefore, some preliminary liquid-liquid extractions were performed at controlled temperature (25℃) by exploring i) a small library of DESs, ii) different DESs concentrations in the aqueous phase, and iii) different extraction strategies. 241Am(III) and 152Eu(III) were used as representatives of minor actinide and lanthanide families, present in the aqueous feed to be decontaminated. The DES affinity towards actinides and lanthanides and their extraction capability were checked with respect to TODGA, as reference ligand to co-extract Am(III) and Eu(III), and CyMe4-BTBP, as reference ligand to selectively extract Am(III), dissolved in the organic phase.
Results, advantages and drawbacks of the use of DESs in advanced partitioning processes are summarized in the present communication.

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Poster NAM / 540

Monitoring of iodine radionuclides in NPP gas emissions

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Radionuclide 131I is used as a reference nuclide for monitoring of gas-aerosol emissions. Under the normal operating modes, 131I radionuclide release does not exceed admissible values, however, during conduction of some activities in the plant premises it can exceed admissible levels that can be potentially dangerous for personnel because of high toxicity of the iodine. In case of accident, the population can also be exposed to risk of radiation. The situation is complicated by imperfection of equipment used for monitoring of iodine radionuclide content in the air at nuclear plants. The variety of iodine radionuclide forms that include an aerosol component - molecular iodine (I2) and iodine organic compounds (iodide methyl (CH3I), primarly) is also an issue. The sorption extraction of iodine radionuclides from gaseous atmosphere is used both for cleaning and iodine content control and is basically single option nowadays. The activated carbon or coals, impregnated with iodide potassium, elemental iodine, silver nitrate, triethylenediamine, etc. are widely used for sorption of iodine radionuclides. However, the coal flammability does not allow to apply such sorbents at temperatures over 150 ° C. Mineral sorbents for iodine capturing are being developed for more than one decade, however, the creation of an effective, cheap and long-life sorbent is still not solved. Important aspect of this problem is the selectivity of iodine extraction to inert radioactive gases.
NITI has developed the Siloxide sorbent used for capturing the volatile iodine radionuclides in gaseous atmosphere. Siloxide is not flammable and does not emit volatile toxic components within operating temperature range. The coefficient of iodine selectivity to 133Xe is 2·105. The sorbent was tested in laboratory and industrial researches for different types of nuclear power plants. The monitor for determination of volume activity of iodine radionuclides in gaseous atmosphere of NPP has been developed: range of measurements is (10-5 –105) Bq/l with air flow rate of 30 l/min and sorbent volume of 100 cm3. Time of iodine radionuclides release is from 0.1 to 1000 hours. The URAN-10-BGG - monitor for extracting and detecting the iodine radionuclides in the gaseous atmosphere - comprises of Siloxide-based sorption element and scintillation gamma sensor (BDEG type) hooused within uniform lead protection. The monitor has a simple design, sharp selectivity and capability to extract all forms of iodine radionuclide in gaseous atmosphere. The most significant advantage of Siloxide is that it does not occlude inert radioactive gases and is effective even at 100% of humidity. This methodical and hardware solution is applied at NITI reactor research facility, in JSC «SSC RIAR», FSUE "Zavod Medradiopreparat" and on a number of the NPP.

Poster NAM / 760

Radioactivity of chondrites and meteorites from Mars and the Moon: Measurements and Monte Carlo simulations

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Low-level concentrations of primordial and cosmogenic radionuclides were determined in recently fallen chondrites and in meteorites from Mars and the Moon. The radionuclide measurements showed a wide range of concentrations of ^{22}Na, ^{26}Al, ^{46}Sc, ^{54}Mn and ^{57}Co. The absence of the neutron product (^{60}Co) in some of the meteorites indicates that their pre-atmospheric radius was small (on the order of 10 cm). This has also been confirmed by Monte Carlo simulations of the production rates of investigated cosmogenic radionuclides, and their comparison with experimental data. The cosmic-ray exposure ages of the investigated meteorites have been estimated to range from 1 to 5 million years. The agreement between the simulated and observed ^{26}Al activities indicates that the meteorites were mostly irradiated by a long-term average flux of galactic cosmic rays.

NFC 2 / 872

SF and HLW disposal, long term performance and radionuclide migration in UK radioactive waste disposal –an overview

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TAN 2 / 604

Paramagnetic NMR investigations in metal-organic complexes of tetravalent actinides with soft-donor ligands

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Electronic interactions between metal and ligand are the origin of additional NMR chemical shifts observed on nuclei of the ligand in paramagnetic metal-organic complexes. The major two contributors to these paramagnetic chemical shifts are Fermi-contact shifts (FCS) and pseudo-contact shifts (PCS). FCS are due to delocalisation of unpaired electron density in molecular orbitals involving both metal and ligand orbitals and thus report on the bond properties. PCS are originating from distance- and angle-dependent dipolar coupling of electron spins through space and are therefore bearing structural information.

The mathematical separation of paramagnetic contributions in complexes relies on the availability of a suitable diamagnetic reference to subtract non-paramagnetic contributions. For the trivalent actinides no diamagnetic reference in the same series is available in milligram scale. Furthermore, all available theories behind mathematical disentangling of contributions to the paramagnetic chemical shift, even for the lanthanide series, omit the influence of spin-orbit effects that might have a sizeable contribution as well.^[1,2]

Comparing isostructural diamagnetic complexes of both f-element series of tetravalent metal ions (Ce(IV) and Th(IV)) allows for an estimation of additional influences to the chemical shifts and the effect of contributions usually omitted by commonly used mathematical theories.

To assess the chemical bonding situation via the influences on NMR chemical shifts (via FCS) we started to study paramagnetic metal-organic complexes of the tetravalent actinides (An(IV)). With increasing number of unpaired electrons throughout the $5f$-series additional effects to the observed chemical shift are expected. Structural properties of the complexes as derived from PCS contributions can be compared to single crystal X-ray diffraction structures. Herein we report the first results of investigations of N- and N,O-donor ligand complexes of the An(IV) series.

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TAN 2 / 599

Coordination Chemistry of Uranium (U(IV) and -(VI)) with Bidentate N-donor Ligands

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Coordination Chemistry of Uranium (U(IV) and -(VI)) with Bidentate *N*-donor Ligands

Introduction

The bidentate *N*-donor ligands 2,2'-bipyridine (bipy) and 1,10-phenanthroline (phen) have attracted considerable attention in the field of coordination chemistry over the last decades because of their remarkable stability towards a wide variety of transition metals.¹ The coordination chemistry of uranium (U) has been explored with these *N*-donor ligands as well with a primary focus on its hexavalent state (U(VI) as UO₂²⁺). To the contrary, much less attention has been paid for the lower oxidation states, such as a tetravalent state (U(IV)). Here we present a systematic study on the coordination chemistry of U(IV) and -(VI) with bipy and phen under different chemical conditions, such as different solvents and changing the metal / ligand ratio.

Results and Discussion

In this study we succeeded to obtain a series of U(IV) complexes with U:ligand ratios of 1:1 and 1:2, all of which show an eight-fold coordinated uranium centre. In addition to the ligand, chloro and methanolato ligands are coordinating to the metal centre for charge compensation. Interestingly, the complexation between U(IV) and the ligand does occur even in protic solvents, in which the ligand is expected to be protonated. We also obtained another series of U(VI) complexes with both bipy and phen, underlining the versatile coordination chemistry of uranyl (UO₂²⁺). That is, the coordination between uranyl and the ligand depends strongly on the pH of the solvent used. For instance, in media with lower pH mononuclear complexes are formed, showing the uranyl unit in an unusually bent geometry.³ On the other hand, dinuclear uranyl arrangements with hydroxo-bridging are dominated in the media with higher pH.

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TAN 2 / 498

Complexation of americium(III) with citric acid : role of the hydroxo function ?

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Alpha-hydroxycarboxylic acids are important molecules in nuclear field. They can be used as complexing agent or buffer in aqueous phase in actinide selective separation processes. These molecules are also present in the environment as a degradation product of cellulose. Therefore the alpha hydroxyl-carboxylate ligands are likely to influence the speciation in the fuel cycle processes and in the migration mechanisms. In this double context, the behavior and complexation of americium with this type of ligand was studied, taking citric acid as a representative of this family.

Some studies have already been published on actinide(III)-citric acid complexation, but most of them are incomplete because the citric acid is also one of the most complicated compounds of the family due to its three carboxylic acids able to be deprotonated and to coordinate the metallic cation. On one side, the thermodynamic studies provide only the stoichiometry of the different complexes without any information on the coordination spheres (the number of complexing carboxylates, the presence or not of the hydroxo function as a coordinating group and the ligand protonation state). On the other side, the rare structural studies relate to polynuclear or polymeric species unlike the complexes identified in solution in the previous thermodynamic studies.

In this study, thermodynamics and structural data were assessed in solution for the same chemical conditions to finally describe the speciation of the Am(III)-citrate system. A focus was done on the role played by the hydroxyl group: does it coordinate the actinide? Is it protonated? To better understand its role, the Am-citrate system was compared to the Am-tricarballylate system (analogue to citric acid without the hydroxyl group). The two systems were studied at pH = 1 and 3. The complexation constants have been determined and, different complexes were characterized using X-ray absorption, NMR, TRLFS and capillary electrophoresis.

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Microbial processes in a radioactive waste repository

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The nuclear fuel cycle ends by spent fuel reposition in an appropriate facility. The long-term performance of a radioactive waste repository can be significantly influenced by geological, geochemical and also biological phenomena. Microbial processes may negatively affect canister (microbial corrosion) and buffer and backfill material (microbial induced chemical and mechanical changes) as well as the geosphere of the repository (biologically enhance migration of radionuclides). On the contrary, a positive effect of microbes is observed during the open phase of the repository, when microbial processes in the geosphere stop oxygen from reaching the groundwater, and also in the post closure phase, when microbes (denitrifying bacteria, manganese reduction b., iron reducing b., sulphate reducing b., sulphur reducing b., methanogens) reduce the available ions to a lower oxidation state. In both cases, ORP is kept at a low level, which restricts radionuclide migration. Moreover, migration of radionuclides can be restricted by accumulation or biosorption by microbes. Due to the fact that radionuclides can be toxic (or radiotoxic) for the microbes present, the study of radionuclide (or their stable equivalents) toxicity is very important.

In this study, the effect of different concentrations of non-radioactive Cs on the survivability of natural anaerobic bacteria was determined. The impact of Cs on bacteria was examined using molecular-biological methods and transmission electron microscopy techniques. Lower Cs concentrations (0.5 mM) promoted bacteria growth, while higher concentrations (1 mM) limited their growth, and 5 mM was lethal.

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Polydentate phosphinoxide-bearing N-heterocycles as extractants for f-elements

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A new type of heterocyclic "hard-and-soft"phosphinoxide-bearing N-heterocycles for f-elements binding was proposed from the basis of DFT simulation (PBE, B3LYP, cc-pVDZ and cc-pVTZ basis sets). The efficient method for the scalable preparation of the reagents by cross-coupling reactions was developed. As a N-donor scaffold the pyridine, 2,2'-bipyridine and phenanthroline were used and their combination with different dialkyl-, diaryl- or alkylarylphosphinoxides were applied. The structures of the ligands were estimated by X-Ray. All of the synthesized reagents demonstrate selectivity to Am over Eu on extraction from 0.5-5M nitric acid to polar diluents. For the first time the significant affect of P,P-diasteriomers on extraction was observed for phenanthroline-based reagent. Solvate numbers for the extractants strongly depends on the number of donating atoms and less on their nature. The model Eu complexes were prepared and structurally characterized for the extractants based on 2,2'-bipyridine and phenanthroline scaffolds. Phosphinoxides based on 2,6-disubstituted pyridine possess the better among investigated compounds efficiency in Am/Cm separation (SFAm/Cm 2.9-3.5 from 0.1-3M HNO3) which was estimated based on three methods γ-, α- and liquid-scintillation spectrometry. The DTF modeling of the extracting species show that the better fitting of the spatial requirements of the ligand was observed for Am than Cm or Eu . The reagents demonstrate excellent radiation stability: the γ-radiolysis up to 350kGr doesn't decrease the Am, Eu and Cm distribution coefficients. So the new generation of neutral organophosphorous reagents are promising for application in minor actinide separation technology.
The results have been obtained under support of the RSF grant no. 16-13-10451.

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Characterization of different bentonite and its application in the sorption of Cs and Sr from aqueous solution by bentonite-alginate microcapsules

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The bentonite materials are frequently designed as buffer/backfill material for engineered barrier in final disposal for spent nuclear fuels (SNFs) or high-level radioactive waste (HLW) repository. The clay minerals showed a favourable environment for sorption of Cs and Sr, bentonite having high sorption toward Cs and Sr were encapsulated in the biopolymer matrices (sodium alginate, NaALG) in this study. The characterization of bentonite-alginate microcapsules (MCs) enclosing different bentonite (Taiwan, China, U.S) was examined and analysed by SEM, and the sorption of Cs and Sr in different bentonite-alginate MCs was investigated by batch methods. Spherical and elastic granules

were obtained, and the particle size of microcapsules was about 500 -700 μm. A relatively higher distribution coefficient (Kd) of Cs for bentonite-alginate MCs than Sr. The sorption isotherm of Cs and Sr followed a Langmuir-type equation and it indicated that the sorption capacity of different bentonite-alginate MCs in Cs were (233.96 - 266.57mg/g) higher than Sr (19.40 - 63.01mg/g). In fact, the ZH bentonite-alginate MCs has a lowest sorption capacity for Sr due to the ZH bentonite is Ca-type bentonite and the higher Ca2+ component and pyrite (FeS) mineral would may have a soption competetion with Sr2+ in the solution.

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Spectroscopic and batch studies of technetium uptake by siderite

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⁹⁹Tc is a long-lived (t_{1/2} = 2.1 x 10⁵ years) β-emitter formed during the fission of U and is of major concern for radioactive waste disposal. Its environmental mobility is primarily governed by the oxidation states VII and IV, with Tc^{VII} forming the highly mobile TcO₄[−] aquo anion, whereas Tc^{IV} is rather immobile due to the low solubility of its hydrolysis products 1. Redox processes, which are able to convert Tc^{VII} into Tc^{IV}, are hence of paramount importance for the safety of radioactive waste repositories. Fe^{II}-bearing minerals, ubiquitous in nature but also forming as corrosion products of the steel canisters foreseen as a possible first enclosure of radioactive waste, play a vital role in these redox reactions due to their high redox reactivity and high sorption capacity, as has been shown not only for Tc, but also for Se, U, Np and Pu.
We studied the Tc^{VII} uptake by siderite (FeCO₃), a typical Fe^{II} mineral in carbonate-rich environments, in the relevant pH range 7 –12.6 under anoxic conditions by means of batch sorption experiments and by X-ray absorption spectroscopy. Sorption experiments showed that Tc retention by siderite is fast and efficient (log R_d ~5) across the investigated pH range and independent of ionic strength (0.1 –1 M NaCl). Tc K-edge X-ray absorption near-edge structure (XANES) data confirmed that the Tc immobilization is due to the surface-mediated reduction of Tc^{VII} to Tc^{IV}. The local structure of Tc^{IV} in Tc siderite sorption samples and Tc siderite coprecipitates probed by extended X-ray absorption fine-structure (EXAFS) spectroscopy revealed three different species: In the pH range 7.8 to 11.8, TcO₂-dimers form inner-sphere sorption complexes at the surface of the initial siderite phase as well as on secondary magnetite or green rust formed during the redox reaction. Between pH 11.9 and 12.6, a mixed Fe/Tc hydroxocarbonate precipitate (chukanovite-like) is formed.
The results showed that siderite contributes effectively to the retention of Tc under repository conditions through formation of strong sorption complexes and precipitation of hydroxocarbonate phases with low solubility.
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TAN 2 / 493

Thermodynamics of the complexation of neptunium (V) with halides in aqueous solution at elevated temperatures

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Introduction

Deep geological salt rock formations are in discussion to serve as host rock for a repository of high-level nuclear waste. This approach requires a well-founded knowledge of the geochemical behaviour of the radionuclides in diluted to concentrated salt solutions to model their potential migration and retention in the near- and far-field of the repository over long time scales. In the past a broad variety of complexation reactions of actinides in their most important oxidation states were studied. The thermodynamic functions (log β_j^0, $\Delta_r H_m^0$, $\Delta_r S_m^0$, $\Delta_r C_{p,m}^0$) and modelling parameters ($\epsilon_T(i,k)$) are summarized in the NEA thermodynamic database.[1] However, most data are only available for 25 ℃ or fixed ionic strength.[2a,b] Temperatures up to 200 ℃ are expected in the near-field of a repository for high-level nuclear waste, which will significantly alter the thermodynamics of the geochemical reactions of the radionuclides in solution. Thus, detailed thermodynamic data at 25℃ as well as at increased temperatures are mandatory for a comprehensive long-term safety assessment.

Results and Discussion

In this work the complexation of $NpO_2{}^+$ with F^- and Cl^- was studied as a function of the temperature (20 –85 ℃) and the ionic strength ($I_m(NaClO_4)$ = 0.5 –4.0 mol kg^{-1}) using near infrared (NIR) absorption spectroscopy. Systematic evaluation of the spectroscopic results clearly confirms the formation of two distinct complex species $[NpO_2(L)_2]^{1-n}$ (n = 1, 2) for L = F^- and Cl^-. Applying the specific ion interaction theory (SIT) and the integrated Van´t Hoff equation the thermodynamic functions (log β_j^0, $\Delta_r H_m^0$, $\Delta_r S_m^0$) and the binary ion-ion-interaction parameters ($\epsilon_T(i,k)$) for the different complexation reactions are obtained. With increasing temperature the chemical equilibrium shifts towards the complexed Np(V) species for both ligands. The thermodynamic stability constants increase by 0.5 - 1 orders of magnitude with increasing temperature. Furthermore, all complexation reactions show positive values of $\Delta_r H_m^0$ and $\Delta_r S_m^0$. Thus, the reactions are endothermic and driven by the high gain of entropy.

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TAN 2 / 497

Actinide samples production at Mainz University for applications in chemistry and physics research

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Rare actinide isotopes are essential in many basic science research projects, yet working with them always features unique aspects, especially due to their limited availability and often high radioactivity. In our contribution, we will focus on actinide targets produced via molecular plating (MP) 1, a technique refined over decades and well-confirmed to be ideal in many cases. Recent highlight results that were obtained with actinide samples from our institute in accelerator experiments include studies on the synthesis, nuclear, and chemical properties of the superheavy elements at GSI

Darmstadt [2-6] using Pu-244, Am-243, Cm-248, Bk-249, and Cf-249 targets; nuclear reaction studies aiding in the identification of the optimum route towards elements beyond Z=118 performed at the ANU Canberra, Australia [7]; the determination of the first ionization potential of the last actinide element, lawrencium at JAEA Tokai, Japan [8]; fission cross section studies on Pu-242 performed at CERN n_TOF [9] and the HZDR nELBE facility [10]. The MP technique is also suitable for other applications, providing sources of decay-daughters. Recently, a direct signal from the long-sought exotic low-energy nuclear isomeric state in Th-229 at an excitation energy below 10 eV was observed at LMU Munich [11], using alpha-decaying U-233, which feeds the isomer in a 2% branch.
An overview over our actinide target production and characterization capabilities will be given, and the recently developed "Drop-on-Demand"ink-jet printing technique [12] will be introduced, which overcomes some of the limitations of MP, like the need for an electrically conducting backing or the restricted sample geometry.

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NFC 2 / 588

Effect of background electrolyte composition on the sorption behavior of Th(IV) and Zr(IV) on the muscovite (001) basal plane

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Reliable long-term predictions about the safety of a potential nuclear waste repository must be based on a sound, molecular-level comprehension of the geochemical behavior of the radionuclides. Especially, their reactivity at the water/mineral interface will control their mobility and thus hazard potential.1 A recent study has found a surprising dependency of the uptake of Th(IV) on the muscovite (001) basal plane on the composition of the background electrolyte.2
Two effects were observed a sorption reducing effect of perchlorate relative to chloride and a sorption increasing effect of Li relative to Na. Thus, a simple change from NaClO4 medium to LiClO4 led to an increase in surface occupancy by more than two orders of magnitude, which subsequently leads to the formation of Th(IV)-(hydr)oxo-nanoparticles. A mechanistic interpretation is hitherto not available, so it remains unknown whether cation and anion effects occur independently and whether the background electrolyte's cation affects the formation of nanoparticles in solution or increases sorption at the water/mineral interface.
To probe whether anion and cation effects occur independently, Th(IV) sorption was studied in the

presence of LiCl and KCl ([Th] = 0.1 mM, pH = 3.3, I = 0.1 M) using the surface X-ray diffraction techniques crystal truncation rod (CTR) diffraction and resonant anomalous X-ray reflectivity (RAXR). The finding show strong uptake at the muscovite basal plane in both cases, exceeding the surface occupancy previously described in NaCl media,[3] thus confirming that the cation effect is indeed independent of the background electrolyte's anion.
To elucidate whether the observed differences occur, when oligomers are present before introduction of the mineral surface, we studied the uptake behavior of Zr(IV). Zr(IV) has a much more pronounced hydrolysis, and similar subsequent formation of oligomers and nanoparticles compared to Th(IV). The interfacial structure of muscovite was characterized in contact with Zr(IV) in solutions of various background electrolytes MCl (M = Li –Cs, [Zr] = 0.1 mM; pH 2.5, I = 0.1 M). In parallel, we performed AFM to characterize the morphology of any particles found on the mineral surface. The results clearly show that only small differences are induced by the electrolyte composition, which are generally well explained by the alkali cations sorption affinity and speciation at the muscovite (001) basal plane. Apparently, the background electrolyte effect is suppressed (or not effective at all) when the initial speciation of the metal is as small oligomers, indicating that the effects observed for Th(IV) occur at the water/mineral interface, and not in solution.

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SEP 2 / 521

Interaction of human serum albumin with Cm(III) using time-resolved laser fluorescence spectroscopy (TRLFS) and NMR

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Radionuclides, in particular actinides, can cause a serious health risk upon incorporation. Regarding the development of potential decontamination therapies, a detailed understanding of the different biochemical reactions of incorporated actinides is highly required. In the human body, actinides preferentially interact with blood serum components. One representative of utmost importance is human serum albumin (HSA), the most abundant protein in the human blood plasma. HSA binds and transports a range of predominantly water-insoluble molecules such as fatty acids, hormones and drugs as well as metal ions. Since albumin possesses four partially selective metal binding sites it can potentially also bind and thus transport incorporated actinides.
The complexation of Cm(III) with HSA is investigated in the pH range from 3.5 to 11.5 using time-resolved laser fluorescence spectroscopy (TRLFS). The Cm(III) HSA species displays an emission band at λ_{max} = 602.5 nm and is the dominating species between pH 7.0 and 9.3. The fluorescence lifetime of 152 µs correlates with three to four water molecules and five to six ligands (amino acid residues from the protein, OH^-, $CO_3{}^{2-}$ etc.) in the first coordination sphere of the metal ion. For the complexation of Cm(III) with HSA at pH 8.0 a conditional stability constant of logK = 6.2 ± 0.5 is determined.
The investigations regarding the identification of the Cm(III) HSA binding site were focused on the two binding sites with the highest affinities to metal ions. These are the N-terminal Cu and Ni binding

site (NTS) and the multi metal binding site (MBS), which binds predominantly Cd and Zn. Competitive titrations of Cm(III) HSA with Cd(II), Zn(II), Cu(II) and Ni(II) point to complexation of Cm(III) at the NTS site. This is confirmed by NMR measurements with Eu(III) and Am(III). With increasing Eu(III) and Am(III) concentration the signal of proton H8 which is located on the His residue at the NTS decreases. Further TRLFS and NMR experiments were performed with recombinant human serum albumin (rHSA), an albumin variant with single deletion of Asp from the N-terminus. The results indicate an involvement of this Asp residue in Cm(III) binding confirming the NTS as the preferential Cm(III) binding site.
The results presented in this study focus on the identification, speciation and structural investigation of Cm(III) HSA. For the first time thermodynamic data were derived which allow a quantitative description of the complexation reaction. Under physiological conditions (pH 7.4, 37.5℃) about 90% of the Cm(III) is coordinated to albumin which indicates that complexation with HSA is a relevant biochemical reaction incorporated actinides might undergo enabling the transport of actinides in the human blood.

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RER 1 / 695

Contamination of the Baltic sea with radionuclides and potential risk to aquatic biota

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Contamination of the Baltic sea with radionuclides and potential risk to aquatic biota

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Data on long-lived radionuclide concentrations in water and sediments obtained during various projects performed by SRI CPST (Institute of Physics) as well as from HELCOM-MORS database have been analysed in order of better understanding transport, redistribution and accumulation of radionuclides during 1986-2015. In addition, the assessment of biota exposure was performed for the Baltic Sea (Lithuanian economic zone). Average activity concentrations of 137Cs, 40K, 90Sr and 239, 240Pu in surface water, sediments and biota for period of 2011-2015 from EPA and SRI CPST database were used for modelling. The species considered for this part of the sea included seaweed (*Cladophora glomerata*, *Flucelloria lumricalis* and *Fucus vesiculosus*) and fish species such as plaice (*Pleuronectes platessa*), Baltic herring (*Clupea harengus membras*), and bream (*Abramis brama*). ERICA Assessment Tool was used for evaluation of dose rates for biota. The screening value 10 microGy/h proposed in ERICA tool was used for evaluation of the risk. The total (including internal and external) estimated dose rates were compared directly to the selected screening dose rate to enable the assessment of the risk to biota.
137Cs activity concentrations were measured with HPGe detectors (GEM40P4-76, efficiency 40%, resolution 1.85 keV (FWHM) at 1.33 MeV and GX4018, resolution 1.8 keV/1.33 Mev and efficiency 42 %). All dried samples were ashed, and Pu isotopes were separated from the matrix as well as from interfering radionuclides using the extraction chromatography method. 242Pu was used as a tracer for the chemical yield determination. Plutonium was extracted with TOPO/cyclohexane solution

and TEVA and TRU resins were applied for the final cleaning (Lujaniene, 2013). Then plutonium isotopes were electrodeposited on the stainless steel discs. 239,240Pu activity activities were measured by means of alpha spectrometry.

Wide variations of 137Cs and 239,240Pu activity concentrations in bottom sediments of the Lithuanian waters (Sothern Baltic Proper) of the Baltic Sea were observed in 2011-2015. Average 137Cs activity concentrations varied from 7.6±0.5Bq/kg to 200±10Bq/kg dry weight (d. wt.), while in the Curonian Lagoon activities ranged from 1.24±0.07Bq/kg to 10.4±0.5Bq/kg d. wt. The highest 137Cs activity concentrations in the Baltic Sea in bottom sediments were reported for the Gulf of Finland and the Bothnian Sea (HELCOM-MORS database).

239,240Pu activity concentrations in seaweed ranged from 0.48±0.04 mBq/kg to 17.6±1.5 mBq/kg, d. wt. The lowest activities were found in the *Cladophora glomerata* collected in the Curonian Lagoon, while the highest ones in the *Fucus vesiculosus* from the Baltic Sea.

Activity concentrations of 239,240Pu in the fish species varied from 0.3 mBq/kg to 1.3 mBq/kg, wet weight (w. wt.). Low plutonium activity concentrations were measured in the *Abramis brama* from the Curonian Lagoon, whereas higher activities were characteristic of the *Clupea harengus membras* and *Pleuronectes platessa* species collected in the Baltic Sea.

Dose rate evaluation has shown that main contribution to dose rate in the Sothern Baltic Proper was from naturally occurring 40K, similar results were obtained for the Gulf of Finland and the Bothnian Sea where the doses to biota were mainly dominated by the natural radionuclides 210Po and 40K (Outola et al., 2011).

137Cs was the main contributor to the dose rate from all studied artificial radionuclides. The lowest dose rates resulted to pelagic fish and the highest to seaweed. Higher activities of 137Cs in bottom sediments is the reason for this difference, as the lowest dose rates resulted to fish and the highest to sediment-associated organisms. The contribution from the other artificial radionuclides evaluated (90Sr, 239, 240Pu) to the total dose were negligible.

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SEP 2 / **737**

Uranyl –Carbonate –Ca(2+)/Mg(2+) aqueous system spectroscopic experimental and theoretical study - Molecular modelling meets environmental protection

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Aside to pure Physical Chemistry interest, determining uranium speciation is important for environmental monitoring or within experimental studies for possible remediation protocols (and possible application for prospection and extraction is of interest as well). Natural water samples dominated by system $UO_2{}^{2+}$ - CO_3^{2-} - Ca^{2+}/Mg^{2+} - H_2O have been chosen in this study.

Cryogenic Time-Resolved Laser-Induced Fluorescence Spectroscopy (TRLFS) and in-Capillary UV-VIS spectrophotometry have been used for a direct speciation in low concentration ranges (10^{-8} M –10^{-3} M, 10^{-6} M –10^{-2} M).

However, subsequent spectral series data analysis face two major problems –

1. Even robust Multilinear methods (ranging from Parallel Factor Analysis to PCA respecting matrix dependence of luminescence life-times of each component) suffer from ill-conditioned spectral data (as individual components are nearly indistinguishable in both spectral and temporal domains).
2. Data analysis provides us with a set of individual component spectra and concentration profiles. But how to assign components to a particular chemical species? In particular, when independent information on chemical composition of studied natural sample is limited - individual component assignment from, often incomplete, literature data could be insufficient.

We propose first-principles-based Computational Chemistry protocol to tackle both problems. Aside to the spin-orbit splitting including quasi-relativistic and relativistic spectral parameter estimation extending the preliminary study 1, a protocol connecting Classical Molecular Dynamics sampling, ground and excited state geometry optimization, normal mode computation and multimode Franck-Condon Factor computation has been proposed for individual components spectra computer simulation (Fig. 1). The preliminary versions of protocol were applied to aquo complex $[UO_2(H_2O)_5]^{2+}$ 2 and tris(sulfate) complex, $[UO_2(SO_4)_3]^{4-}$.

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Literature

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Time evolution of Fukushima-derived radiocesium in the western subtropical gyre of the North Pacific Ocean by 2017

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An accident of Fukushima Dai-ichi Nuclear Power Plant (FNPP1) in March 2011 resulted in a large amount release of radiocesium (134Cs and 137Cs) into the North Pacific Ocean. Radiocesium deposited on and discharged directly into coastal area of Japan in the north of the Kuroshio Front had been transported eastward in surface layer and reached to the North American Continent by 2015 and early 2016 (Smith et al., 2017). On the other hand, radiocesium deposited on the south of the front, namely the western subtropical gyre, had been transported southward to 15°N through subsurface layer due to subduction of the subtropical mode water (STMW) by the end of 2014 (Kumamoto et al., 2017). However, time evolution of Fukushima-derived radiocesium in the subtropical gyre is still unclear because observational data of radiocesium in subsurface layer are less than those at surface. Here we show our new radiocesium data obtained between 2015 and 2017 in the western subtropical gyre. Seawater samples were collected from surface to 800 m depth during cruises of KS-15-14 (October 2015), KH-16-3 (June 2016), KM16-08 (September 2016), KS16-09 (Nov. 2016), and KM17-01 (Jan. 2017). The seawater sample was acidified using nitric acid and then radiocesium in the seawater was concentrated onto ammonium phosphomolybdate (AMP). Radiocesium in the AMP was measured using gamma-ray detectors. Uncertainty of the radiocesium measurement was estimated to be about 8 %. A meridional transection of 134Cs in 2015/2016/2017 along 142-145°E between 25°N and 35°N was compared to those observed in Jan./Feb. 2012 (Kumamoto et al., 2014), Oct. 2012 (Kaeriyama et al., 2016), and Mar.-Jun. 2014 (Kumamoto et al., 2017) along 142-149°E. In Jan./Feb. 2012, a high 134Cs plume was lying in subsurface layer around 300 m depth between 25°N and 35°N and the highest activity concentration of 134Cs (more than 25 Bq/m3) were observed at 32°N. In Oct. 2012, the peak of 134Cs subsurface maximum (about 10 Bq/m3) moved southward to the south of 30°N. In Mar.-Jun. 2014, there was still the subsurface maximum layer around 300 m depth although the peak concentration decreased to 5 Bq/m3, which was observed at 34°N. These temporal changes indicate that 134Cs subducted to the subsurface layer just after the FNPP1 accident had remained within the western subtropical gyre by the end of 2014. The southward and then northward movements of the 134Cs peak between 2012 and 2014 imply anticyclonic re-circulation of STMW within the gyre. In 2015/2016/2017, the subsurface maximum layer (~3 Bq/m3) was also observed although the peak layer deepened from 300 m to 400 m depth approximately. This deepening is probably derived from vertical diffusion toward deep layer and erosion by newly-subducted STMW in shallow layer. Subsurface maxima of 134Cs (~2 Bq/m3) at the western edge of the gyre (26°N/128°E and 24°N/131°E) in 2017 suggest that Fukushima-derived radiocesium have been spread in the whole western subtropical gyre. This work was partially supported by Grant-in-Aid for Scientific Research on Innovative Areas, the Ministry of Education, Culture, Sports, Science and Technology Japan (KAKENHI), No. #24110004.

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236U and 233U around Greenland coast

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We report for the first time an environmental uranium-236 (236U) and uranium-233 (233U) dataset, on samples in the Greenland marine environment during the 5-year period (2012-2016). Results are discussed in terms of time evolution and spatial distribution characters of radionuclides (236U) concentrations and isotopic ratios of 236U/238U and 233U/236U. Our results indicate that 236U concentrations and 236U/238U atomic ratios along Greenland coast are distributed within a relatively narrow range of (3.6-8.2) × 107 atoms/L and 1× 10-9, respectively, but still up to 4 times the expected value from direct global fallout (236U=0.8 × 107 atoms/L and 236U/238U=1× 10-9).

Slightly higher 236U/238U ratios as well as high 236U concentrations were observed on the west coast of Greenland compared to the eastern side for 2012, 2015 and 2016, which is in contrast to the expectation that the major source term of 236U near the Greenland coast are the two European reprocessing plants. This feature might be interpreted as a hint for another 236U input in the western Greenland Sea, but may also be caused by the complexity of water currents or by the slow turnover rate. A first preliminary source term assessment was performed using the measured 233U/236U ratios indicating considerable differences in the contributions from the various source terms for 236U between the east and west coast of Greenland.

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Speciation and luminescence properties of Eu(III) and U(VI) in solutions using complexing agents

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Time-resolved laser-induced fluorescence spectroscopy is fast, easy and accurate optical method for low concentration determination and spectral characteristics of rare earth elements (e.g. Eu) and actinides (mainly U, Cm, Am) based on their luminescence properties.

Having Nd:YAG crystal as a laser source (emission wavelength 1064 nm) with the repetition frequency 10 Hz and Optical Parametric Oscillator (OPO) emitting laser beam ranging from 210-2400 nm it is possible to use UV and VIS light to excite the lanthanide and actinide ions in solutions.
For determining spectral characteristic of Eu(III) and U(VI) and their complexes, a 394 nm and 418 nm absorption wavelengths are used respectively. For Eu solutions, a complexing agent TODGA (N,N,N,N-tetraoctyl diglycolamide) as well as EDTA (especially its disodium salt), succinic, glycolic and phthalic acid with different metal-to-ligand ratio are used in an environment of pH ranging from 2 to 10. Solutions containing Uranium are acidified to pH 1 with commercially available Uraplex or phosphoric acid as complexing agents. Additionally, different matrices are used for concentration determinations and fluorescence lifetime measurements. For assessment of species of Europium and Uranium in different solutions a geochemical model PHREEQC is used.

Measurements are performed using quartz cuvettes with a 10 mm optical path at temperature 20℃ in a thermostatted cuvette holder. The fluorescence spectra are recorded by grating monochromator in addition to ICCD camera.

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Structure and thermodynamics of the complexes of neptunium (V) with dicarboxylates in aqueous solution

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Introduction
The complexation of actinides with different organic ligands is an important topic in terms of nuclear waste disposal. Depending on the host rock these molecules are either naturally occurring organic compounds as in pore waters of clay rock formations (e.g. acetate, lactate) or model compounds (e.g. oxalate, malonate, succinate) of organic macromolecules (polycarboxylate based cement additives, humic substances) and their degradation products. These compounds may serve as complexing agents towards actinides and thus influence their migration behaviour. A detailed thermodynamic description of these processes is needed for a comprehensive long-term safety assessment of a nuclear waste repository. However, most of the so-far available data is valid only at 25 ℃ and/or fixed ionic strength.[1] The thermodynamic functions (log β_j^0(T), Δ_rH$_m^0$, Δ_rS$_m^0$, Δ_rC$_{p,m}^0$) and SIT ion-ion interaction parameters (ϵ_T(i,k)) are often missing. Due to the radioactive decay, elevated temperatures are expected in the near-field of a repository. This will considerably alter the (geo)chemical reactions of the actinides and thermodynamic data of them at increased temperatures are mandatory for a comprehensive thermodynamic model.

Results and Discussion
In this work the complexation of Np(V) with oxalate (Ox^{2-}), malonate (Mal^{2-}), and succinate (Succ^{2-}) is studied as a function of the temperature (20 –85 ℃) and ionic strength (I$_m$(NaCl or NaClO4) = 0.5 –4.0 mol kg^{-1}) using near infrared absorption spectroscopy. The structure of the formed complexes is investigated by extended x-ray absorption fine structure spectroscopy (EXAFS). The formation of the complex species [NpO$_2$(L)$_n$]$^{1-2n}$ (n = 1, 2) is confirmed for all ligands. Application of the specific ion interaction theory (SIT) and the integrated Van´t Hoff equation yields the thermodynamic functions (log β_j^0(T), Δ_rH$_m^0$, Δ_rS$_m^0$) and the SIT-specific binary ion-ion-interaction parameters (ϵ_T(i,k))

for the different complexation reactions. The results show, that with increasing length of the alkyl chain of the ligand the reaction enthalpy of the complexation reaction shifts from exothermic (Ox^{2-}) to endothermic (Mal^{2-}, $Succ^{2-}$) and the stability constants decrease by 2.5 –6 orders of magnitude. The EXAFS data reveal that the change of the thermodynamic properties results from different coordination modes and ring sizes of the formed chelate complexes. Ox^{2-} and Mal^{2-} form 5- and 6-membered chelate rings with Np(V) by coordinating in a side-on mode to Np(V) whereas $Succ^{2-}$ is not capable of forming a chelate ring and coordinates in an end-on fashion.

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RER 1 / 675

I-129 and I-127 speciation in sea water by IC-ICP-MS and AMX

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Abstract

Anthropogenic ^{129}I release increased the $^{129}I/^{127}I$ ratio by several orders of magnitude. In the 1960s, detonations of atmospheric nuclear weapons contributed to the ^{129}I release; today the main sources are the reprocessing plants in La Hague and Sellafield. 90 % of ^{129}I is released into the ocean, gaseous emissions account for only 10 %. Today, this continuous release has increased the $^{129}I/^{127}I$ isotopic ratios from some 10^{-12} in the prenuclear age to 10^{-7} or higher in wide parts of Europe.

Numerous measurements of water samples from the English Channel to the North Sea were published. Total activity concentrations of ^{129}I and isotopic ratios in sea water are well known. However, findings by Hou et. al.[1, 2] indicate disequilibria of the chemical species containing ^{129}I compared to those containing ^{127}I. The present work aims at understanding chemical speciation of the released ^{129}I. Chemical reactions and kinetic aspects of iodine chemistry in sea water are modelled.

Water samples were taken in the vicinity of La Hague. In addition to total ^{129}I and ^{127}I, chemical speciation was investigated at trace concentrations, differentiating iodide, iodate and organo-iodine species.

The results show higher contents of ^{129}I in sea water around La Hague compared to measurements from the North Sea. In addition, the ^{129}I content was higher at the southwest coast of La Hague as compared to the northeast coast. $^{129}I/^{127}I$ ratios of 10^{-6} up to 10^{-5} were found for total iodine and iodate, while $^{129}I/^{127}I$ ratios of 10^{-7} to 10^{-6} were measured for the iodide fraction. Iodate/iodide ratios show lager variation for ^{127}I with values between 2.6 to 7.4 compared to ^{129}I with a range from 1.6 to 4.5. In addition, iodate/iodide ratio of ^{129}I decreased with the distance to La Hague, while the iodate/iodide ratio of ^{127}I did not show any correlation with the distance. Further indication for organic bound iodine co-eluting with iodide was found.

In addition, laboratory batch experiments were performed to simulate the behavior of radioactive ^{129}I. Different iodine chemistry in sea water compared to deionized water was studied with ^{125}I as iodide in a first experiment. A second experiment was performed with iodate and iodide in synthetic sea water to compare the behavior of the two species. In a third step, organic molecules were added as possible reaction partners for the iodine species.

An equilibrium between iodate and iodide was obtained. Iodate spiked solutions contained 10 to 20 % of iodide. Similarly, iodide spiked solutions contained 10 to 20 % iodate as measured by ^{125}I gamma ray spectrometry.

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Complexation of protactinium(V) with nitrilotriacetic acid

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Protactinium, as a ^{235}U decay product, is naturally present in the environment as ^{231}Pa isotope (alpha emitter with a half-life of 32,400 years). Over the years, this isotope is accumulated in uranium tailings and stocks of yellow cake. Modelling the behavior of this element in the geosphere requires thermodynamic and structural data relevant to environmental conditions. The present work concerns a study of the complexation of Pa(V) with nitrilotriacetic acid (NTA, $N(CH_2COOH)_3$), a chelating agent that can be regarded as a model for polyaminocarboxylic acids.

Because of the strong tendency of Pa(V) towards hydrolysis and polymerization, speciation studies have been conducted with the element at ultra-trace scale ($C_{Pa} < 10^{-10}M$) [1,2]. A systematic study using liquid-liquid extraction with a β-diketone as extractant has been performed at constant ionic strength and temperature. Under these experimental conditions, the variations of the distribution coefficient of $^{233}Pa(V)$ as a function of NTA and proton concentrations provides information on the stoichiometry of the complexes Pa-NTA in aqueous phase and also on their mean charge (slope method). Results indicate the formation of two successive complexes that are likely to be $PaO(NTA)$ and $PaO(NTA)_2^{3-}$). The formation of complexes (1:1) and (1:2) are observed with actinides at oxidation states +3 and +4. In contrast, only (1:1) complexes are formed with actinides +5 and +6. In addition, capillary electrophoresis inductively coupled plasma mass spectrometry experiments have been performed with several actinides ($^{239}Pu(IV)$, $^{243}Am(III)$, $^{231}Pa(V)$) at tracer scale. $Pu(IV)$ and $Am(III)$ namely are known to form complexes of charge -2 ($Pu^{IV}(NTA)_2^{2-}$) [3] and -3 ($Am^{III}(NTA)_2^{3-}$) 4. Comparison of electrophoretic mobility for the complexes $Am(III)$, $Pu(IV)$ and $Pa(V)$ with NTA, confirms the charge -3 for the maximum stoichiometry complex.

The formation constants of $PaO(NTA)$ and $PaO(NTA)_2^{3-}$ have been deduced from solvent extraction experiments. The value obtained for the (1:1) complex is similar to those relative to actinides + 6 whereas the formation constant of the (1:2) complex is close to the ones observed for the actinides at the oxidation state +3. Thus, these results emphasize the distinctive feature of protactinium chemistry as compared to the other actinides.

To complete the results already obtained, a structural study by X-ray absorption spectroscopy (XAS) and density functional theory (DFT) calculations is currently being processed to determine the coordination geometry and the interatomic distances of the formed complexes. Preliminary results on the structural study performed with the maximum stoichiometry complex will be also presented.

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RER 1 / 658

Sedimentary record of plutonium in the North Yellow Sea and response to catchment environment of inflow rivers

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239Pu (T1/2=24110 yr) and 240Pu (T1/2=6563 yr), because of their high chemical toxicity, long half-lives and high particle affinity, are not only good indicators for radioactive pollution but also useful geochemical tracers for better understanding a variety of marine processes. Pu isotopes have been intensively studied in the East China Sea to investigate their source terms, transport, scavenging and deposition processes in the past years. However, studies regarding Pu isotopes in the Yellow Sea especially the North Yellow Sea are very limited. The North Yellow Sea is adjacent to the Bohai Bay and surrounded by the Liaodong Peninsula, Shandong Peninsula and Korean Peninsula. The coastal areas of the NYS are densely populated areas, where are very close to North Korea. Nuclear activities including NWTs in North Korea have caused a great concern on the radiation exposure to the public in the past years. The Hongyanhe nuclear power plant (NPP) with 4 units located in Dalian (39.792°N, 121.475°E) on the Liaodong Peninsula has already being in operation from 20th September 2016. In addition, there are plans to build 4 more NPPs in China's three northeast provinces. Potential releases of radioactive materials including Pu from NPPs to the surrounding environment and the consequences are also a major concern of the local inhabitants and the authorities.
This work, for the first time, reports sedimentary records of 239Pu and 240Pu in the northern North Yellow Sea in order to elucidate their source terms and deposition process as well as the response to human activities in watersheds of inflow rivers. The results of 240Pu /239Pu atom ratios in all sediment samples indicates that plutonium in the northern North Yellow Sea was mainly originated from the global atmospheric fallout, without any significant contributions from nuclear weapons testing in the North Korea in the past few years, the Fukushima accident in 2011, as well as the Pacific Proving Ground (PPG) sourced Pu. The observed large variation in the spatial distribution of Pu isotopes in the study area was mainly attributed to the re-suspension and transportation of fine sediments influenced by the coastal currents. From the depth profiles of 239+240Pu activities with easily observed onset fallout levels (1952) and fallout peaks (1963) in the two sediment cores, 239+240Pu was found to be a useful time marker for studying the modern sedimentary process in the northern North Yellow Sea. The riverine Pu was quantified to contribute 15-27% to the total global fallout Pu inventory in the study area, much lower than that in the Yangtze River estuary (77-80%), indicating a better soil conservation in the northeast China compared to the Yangtze River's drainage basin. Riverine input Pu increased after 1980s in the sampling sites reflected the increased soil erosion degree caused by the land use and cover change due to the increased human activities in the northeast China.

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Alpha Radiolysis of DOTA ligand in aqueous solution

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In nuclear fuel reprocessing, after separation of uranium and plutonium using the PUREX process, minor actinides (americium and curium) are the main contributors to the long term radiotoxicity and heat load of the spent fuel. Therefore, new processes have been developed to separate americium and curium using polyaminocarboxylic acids as selective complexant. Due to the presence of radioactive elements, these organic ligands in solution are subjected to radiolysis, thus leading to a possible impact on the process efficiency. Understanding the behavior of polyaminocarboxylic acids when exposed to ionizing radiation is important for further development of minor actinides separation processes. The aim of this work is to study the radiolytic stability of polyaminocarboxylic acids and determine if their stability is different when the ligand is free in solution or involved in a complex with a metallic cation.
In this study, the cyclic polyaminocarboxylic acid, DOTA (1,4,7,10-tetraazacyclo-dodecantetraacetic acid) ligand was chosen as a reference molecule, due to its interesting properties regarding the complexation of actinides (An) +III and +IV. DOTA¬An complexes with a 1:1 stoichiometry are very stable.1 The preliminary experiments consist in external radiolysis on a cyclotron facility or a gamma irradiator of DOTA free in solution or DOTA involved in a complex with a lanthanide (non-radioactive metal). Then the nature and the energy of the ionizing radiation, the composition of the aqueous solutions (pure water or nitric acid) and the effect of the complexation will be examined on all these systems.
More precisely, the degradation of free DOTA and DOTA involved in a complex with neodymium (DOTA-Nd) in aqueous solutions was investigated by external radiolysis on a cyclotron facility. The DOTA solutions were irradiated by alpha particles (4He2+) with energies about 9.4 MeV and 60.7 MeV at a maximum deposed dose of 18kGy. Yield formation of gases like H2 and CO2 as well as yield formation of products in solution (H2O2 and/or HNO2) were determined for different concentrations of DOTA. Moreover, nuclear magnetic resonance (NMR) and electrospray ionization mass spectrometry (ESI-MS) analysis on irradiated DOTA solutions enabled the identification of some DOTA's degradation products which are mainly due to the decarboxylation of the molecule or the opening of the cycle.

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NFC 3 / 748

Hot test of advanced Purex process and its results

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The hot test of advanced Purex processing (Fig.1) based on organic reducing agent was carried out by using experimental reactor UO2 spent fuel with low burn-up in CRARL(China reprocessing and radiochemistry laboratory).

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Fig. 1: The flow-sheet of advanced Purex process.

Several used fuel rods were chopped, dissolved and then separation processing was conducted. Various determination methods were used including K-edge and L-edge densimeter for U or Pu and U/Pu solution, time resolved laser fluorescence trace of U,and X-ray fluorescence or X-ray prediffraction of graphite crystal for low concentrate Pu,and so on. The results such as the recovery of U and Pu, separation factor of U and Pu, decontamination of U and Pu from fission products are satisfactory. During and after the experiment, radioactive discharges into the environment and internal exposure of the staff were monitored.

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Coumarins and their reaction ability in free-radical reactions

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Coumarins reaction ability with free radicals was studied. Antioxidant properties for coumarin, esculetin, esculin in reaction with carbon-centered hydroxyethyl radical, superoxide anion radical are shown. The influence of zinc ions on the change of coumarins reaction ability is investigated.

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Soft Donor Ligands for Actinide/Lanthanide Separations in Used Nuclear Fuel and for Hg(II) Coordination and Extraction in Alkaline Tank Waste Processing

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Minor actinides in used nuclear fuel are responsible for much of its radiotoxicity and heat generation. Actinide(An)/Lanthanide(Ln) separation processes via selective complexation and solvent extraction by designed soft S- and N-donor ligands can take advantage of the slight differences in relative hardness of An(III) vs. Ln(III), leading to potentially selective separations. Furthermore, the presence of organic mercury in the alkaline high-level waste at the Savannah River Site has sparked interest in synthetic soft-donor ligands for Hg(II) and organic mercury separation, coordination, and extraction.

As part of our efforts in selective Hg(II) and Ln/An(III) coordination, soft-donor ligands containing thioamide and/or pyridine sites that are derived from o phenylenediamine or from dipicolinic acid, were synthesized and studied for complexation of Hg(II) and f-elements by structural and spectroscopic methods. These frameworks allowed for comparative studies between ligands that contain the C=O vs. the C=S group: For example, while the O-donor dipicolinamide was shown to bind Ln(III) by UV-Vis and NMR in acetonitrile, the S-donor dithiopicolinamide ligand did not show any binding towards Ln(III). Yet the same dithiopicolinamide was shown to extract Am(III) vs. Eu(III) selectively from acidic solutions. Hg(II) complexation studies by UV-Vis titrations showed binding in solution via the formation of a new UV-Vis absorbance at 279 nm, and selectivity in the presence of Ca(II). The X-ray structure for the dimeric Hg(II) complex shows each Hg(II) center bonded to two S donors from two distinct thioamide ligands and one pyridine N atom, thereby resulting in two tri-coordinated Hg centers.

Pyrazole ligands that contain varying substitution patterns showed remarkable differences in Ln(III) complexation and fluorescence sensing capabilities, which show some variation along the Ln(III) series. The role of ion-pairing and solvation in explaining these differences will be discussed.

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Study on the gamma radiolysis of TODGA and CyMe4BTPhen in the ionic liquid [N1888][NO3] using solvent extraction and UHPLC-ESI-MS analysis

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Separation of minor actinides (MA, i.e. americium, curium and neptunium) from the highly radioactive raffinate originating from spent nuclear fuel reprocessing, followed by an efficient transmutation of Am could contribute to a significant decrease of the long-term radiotoxicity and heat load of vitrified highly radioactive waste (1).
The effect of 60Co gamma radiation on two different solvents was investigated; (i) 0.05 M N,N,N',N'-tetraoctyl diglycolamide (TODGA) in the ionic liquid methyl-trioctylammonium nitrate ([N1888][NO3]) and (ii) 0.01 M 2,9-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-1,2,4-benzotriazin-3-yl)-1,10-phenanthroline (CyMe4BTPhen) in [N1888][NO3].
In the first system, TODGA showed similar degradation pathways (cleavage of bonds within the DGA core of the molecule) as identified in former studies using aliphatic hydrocarbon diluents (2). The main degradation products N,N-dioctylamide, dioctylacetamide, dioctylglycolamide and dioctylformamide were identified by ultra-high performance liquid chromatography coupled to an electrospray-ionization accurate mass-mass spectrometer (UHPLC-ESI-am-MS). The residual TODGA concentration of the solvent after 1 MGy absorbed dose was found to be ca. 45 %, suggesting a high stability of the ligand when diluted in the ionic liquid [N1888][NO3].
The second system, composed of CyMe4BTPhen in [N1888][NO3] was irradiated up to an absorbed dose of 400 kGy. UHPLC-ESI-am-MS analyses showed the presence of a single ligand degradation product, an adduct formed between CyMe4BTPhen and the [N1888]+ cation. In our study, the residual concentration of CyMe4BTPhen was higher than 80 % after 100 kGy of absorbed gamma dose. Nitric acid showed a sensitization effect on the ligand in [N1888][NO3]. Former irradiation studies in 1-octanol showed a protective effect of HNO3 on the ligand (3).
The results of this study show that the sensitivity of these well-known ligands towards radiation-induced degradation can be mitigated by a proper choice of the diluent.

RCH 1 / 682

Photolytic removal of arsenic and mercury(II) from aqueous solutions

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Due to the pollution of industrial effluents with toxic metals, often in ionic forms, and their spreading to the environment, much work is currently underway to develop the most efficient methods of separation of heavy metals (e.g. mercury, cadmium, chromium or arsenic) from aqueous solutions.

Arsenic is known for its very high toxicity. Its removal by conventional methods, aimed largely at ions removal, is complicated by the predominant non-ionic nature of its more dangerous and mobile trivalent form. This difficulty can be minimized using photochemical techniques. The study focuses especially on the impact various solid modificators (e.g. aluminum oxide, zinc peroxide) coagulated during UV irradiation on the arsenic removal (1 mmol/L AsO2-). The UV irradiation of the systems with aluminum salts, formic anions (HCOONH4) as scavengers of •OH radicals and hydrogen peroxide proved to be very effective due to the high rate of arsenic removal after 15 minutes of irradiation: 93% using Al(NO3)3 and 99 % with AlCl3. In the case of AlCl3, the aluminum oxide precursor, the arsenic was removed also by adsorption on previously photolytically prepared precipitate. After several days, 98 % of As was dissolved out of the aqueous solution. A highly efficient system was a solution containing zinc nitrate, formate, and hydrogen peroxide from which zinc peroxide was precipitated due to UV irradiation. More than 98% of arsenic was then removed. A 92% degree of arsenic removal during photolysis was achieved in the system with nickel, formate and hydrogen peroxide.

Optimal conditions were sought also for the photochemical reduction of mercury(II) at a concentration 1 mmol/L. In the systems containing dissolved mercury acetate, perchlorate or chloride in the presence of scavengers of •OH radicals (methanol, ethanol, isopropanol activated by acetone, and ammonium formate) the reduction to monovalent or to metallic mercury in the form of a precipitate occurs. In the system containing mercury acetone and isopropanol activated by acetone mercury was reduced to metallic mercury, in the system with mercury chloride with isopropanol and acetone a mixture of metallic mercury and calomel was formed, while other studied scavengers did not have such strong reduction properties and reduced Hg2+ only to monovalent mercury. The time course of UV radiation along with changes in pH was also monitored. In most irradiated systems the pH value decreased. The highest degree of mercury removal was achieved in systems containing mercury (II) acetate or chloride, and isopropyl alcohol with acetone, where more than 98% of mercury was removed in the insoluble form.

In UV irradiated (medium-pressure mercury lamp with a maximum operating power of 400 W) solutions containing studied metals their concentration was determined by the AAS method.

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Partitioning of minor An(III) and Ln(III) by the 1,2,4-triazine extracting compounds

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Partitioning of trivalent minor actinoids from lanthanoids and mutual separation of minor actinoids is a current issue studied within reprocessing of irradiated nuclear fuel and nuclear reactors of the IVth generation, i.e. closing of a nuclear power cycle. The objective of this contribution is to summarize results obtained for various liquid-liquid or solid-liquid extraction systems, and/or hydrophilic masking agents based on 1,2,4-triazine extracting compounds (BTP, BTBP, BTPhen) proposed for separation of the above mentioned groups of elements. Properties of new monohalogenated CyMe$_4$-BTBP were compared with the reference molecule of r-SANEX, CyMe$_4$-BTBP. The monohalogenated BTBP were much better soluble in octan-1-ol and cyclohexanone. Obtained values of Am/Eu separation factor (SF$_{Am/Eu}$ > 110) were similar to the reference molecule but without the need to use a phase transfer agent. The best results for mutual separation of minor actinoids were obtained in extraction system with CyMe$_4$-BTPhen where a kinetic separation effect SF$_{Am/Eu}$ = 7.9 was observed in the mixture of octan-1-ol with toluene, while in cyclohexanone SF$_{Am/Eu}$ was equal to 5.5–6.7. This effect was explained by computational modelling. Hydrophilic sulfonated ligands were tested for their ability to complex Am(III) and prevent its extraction by TODGA within the i-SANEX process. Four various tetrasulfonated BTBP or BTPhen ligands were able to selectively complex Am(III) in 0.28–0.77 mol/L HNO$_3$; SF$_{Am/Eu}$ = 138–934 was achieved by extraction to TODGA. The described system provides a possible alternative to the American TALSPEAK process as it works at higher HNO_3 concentrations and without the use of any buffer. The highest value of $SF_{Cm/Am}$ = 4.6 for mutual minor actinoids separation was obtained with tetrasulfonated BTPhen in aqueous phase. TODGA-PAN composite sorbent performed well for chromatographic Cm(III) and Am(III) partitioning with $SF_{Cm/Am}$ = 4 when tetrasulfonated BTPhen was used as an eluting agent. The best system for solid-liquid extraction was the BTPhen–SiO_2–Fe_2O_3 MNP. It can be used for selective separation of Am(III) at higher acidities with values of $SF_{Am/Eu} \approx 1670$, and as a non selective extraction system for Am(III), Eu(III), and Cm(III) at lower acidities with values of mass distribution coefficient $D_w > 700$.

RCH 1 / 739

Hydroxyl radical formation in dilute aqueous solutions through photolysis by sub-5-eV photons

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We have observed the formation of OH radicals in aqueous solutions under the effect of 253.7 nm (4.89 eV) UV radiation delivered from a low-pressure Hg-lamp 1. By combination of chemical actinometry (iodide-iodate system) with the tools conventionally used for the characterization of radical yields during radiolysis of aqueous solutions (a fluorescence dosimeter - coumarin-3-carboxylic acid - and the traditional ferrous sulfate dosimeter) we determined the part of radiant energy efficiently absorbed to contribute to the formation of OH radicals via the photolysis of water. The effective quantum yield of the OH radical formation was determined to be approximately 0.08. A combination of at least two 4.89 eV photons would be needed to deliver enough energy for direct H-OH dissociation (that needs between at least 5.0 and 5.4 eV to take place). Since direct two-photon effects can be considered as less probable in the case of the Hg-lamp, we suggest a mechanism based on the interaction of two long-living excited states of water molecules. Such an interaction could provide the energy required for the dissociation of one water molecule into OH and H radicals.
1 K. Tomanová et al.: At the crossroad of photochemistry and radiation chemistry: formation of hydroxyl radicals in diluted aqueous solutions exposed to ultraviolet radiation, Phys. Chem. Chem. Phys. 19, 29402 (2017).

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Gas-phase nitration of metallic uranium, zirconium, and aluminum

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Oxidative embrittlement (voloxidation, volume oxidation) of both the fuel itself and of Zircaloy fuel claddings shows promise as modification of head-end operations of the PUREX process. The gas-phase conversion of oxide fuel in various atmospheres (O2, air, O3, H2O vapor, air–CO2 mixture) at 300–600°C ensures almost complete removal of volatile components (3H, 14C, 129I, radioactive noble gases) already before starting the radiochemical reprocessing of the fuel. DEOX process (DEclad via OXidation) has been developed for breaking down strong zirconium claddings. In this process, the fuel claddings are broken down and transform into ZrO2 powder as a result of the reaction of zirconium with oxygen at 1100°C. On the other hand, along with the oxide fuel, other kinds of fuel (e.g., metallic fuel) also require radiochemical reprocessing. This fuel contains not only U, but also other metals (Zr, Al, Mg, etc.). A hydrometallurgical scheme involving the steps of SNF dissolution both in an acid and in an alkali is suggested for reprocessing of this type of SNF.
Our previous studies dealt with the gas-phase conversion of U3O8, MoO3, SrO, ZrO2, and their mechanical mixtures into water-soluble compounds in the NOx–H2O (vapor)–air and HNO3 (vapor)–air atmospheres in the temperature interval from 25 to 150℃ show that in the course of gas-phase conversion U3O8 and SrO transform into water-soluble compounds (nitrates, hydroxynitrates), whereas MoO3 and ZrO2 undergo no changes. The principal possibility of separating U from Mo and Zr by gas-phase conversion of the oxides in the NOx–H2O (vapor)–air and HNO3 (vapor)–air atmosphere was established. Therefore, it was interesting to examine the possibility of the gas-phase conversion of metallic U, Zr, and Al in the NOx–H2O (vapor)–air or HNO3 (vapor)–air atmosphere (hereinafter, nitrating atmosphere) into water-soluble compounds. That was the subject of this study.
Gas-phase conversion of metallic U, Zr, and Al into water-soluble compounds in the NOx–H2O (vapor)–air and HNO3 (vapor)–air atmospheres was studied. Monolithic Umet and powdered Zrmet undergo gas-phase conversion to form water-soluble compounds (nitrates, hydroxynitrates), whereas monolithic Zrmet remains unchanged. The degree of conversion of Almet into water-soluble compounds in the examined nitrating media at 25–150°C does not exceed 10%. The principal possibility of separating U from Al and Zr by gas-phase conversion of monolithic samples in a nitrating atmosphere was demonstrated.

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Thermodynamic analysis of fluorine regeneration from uranium hexafluoride with a flame of hydrogen-containing fuel and oxygen-containing oxidant using

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The uranium hexafluoride depleted in the U-235 isotope (DUHF) is a by-product of isotope uranium enrichment in the nuclear fuel cycle (NFC). The accumulated DUHF amount in the world reaches 2 million tons, including in Russia up to 1 million tons.
Uranium hexafluoride is a very toxic substance (maximum allowable concentration in the air is 0.015 mg•m-3), volatile (p = 0.1 MPa at 56 ℃) and prone to hydrolysis. Therefore, DUHF large quantities storing is a risk to the environment.
At the same time in the composition of DUHF there is a fluorine, the important chemical substance that is advisable to regenerate.
The only industrial technology of DUHF processing developed by the "AREVA":
UF6 gas + 2H2Ogas ⟶ UO2F2 sol + 4HFgas, T = 250-400 oC; (1)
3UO2F2 sol + 3H2Ogas + H2 gas ⟶ U3O8 sol + 6HFgas + H2Ogas, T = 450-600 oC. (2)
At this temperature the most stable uranium compound is UF4. In order to avoid UF4 formation and to regenerate fluorine completely, process (2) is carried out with an excess of water. Therefore, the product of this process is an azeotropic aqueous solution of HF, which does not allow the NFC closing for fluorine, because the uranium fluorides production based on anhydrous hydrogen fluoride.
The authors performed a thermodynamic analysis of the possibility of HF obtaining from the UF6 in

the flame of the hydrogen-containing fuel and oxygen containing oxidant:
UF6 gas + CmHnNk gas + O2 gas ⟶ UOx sol + mCOy gas + 6HFгаз + 0.5kN2 gas –Q, (3)
where CmHnNk –hydrogen-containing substance, m≥0, n>0, k≥0, x=2÷3, y=1 or 2.

For elements U-H-O-F-N the thermodynamically equilibrium composition of substances were calculated depending on temperature and the ratio of atoms. The calculation was developed based on the method of minimization of Gibbs energy on the molecular composition. It was shown that at temperature above 1100 K at a specific ratio of atoms hydrogen fluoride is practically the only fluorine-containing substance, and uranium dioxide is virtually the only uranium-containing substance.

This temperature in the reaction zone can be obtained using the process of recovery of UF6 in the combustion regime (3). For hydrogen, methane, ammonia as a fuels and oxygen and air as an oxidizers were calculated adiabatic temperature of the products and temperature taking into account thermal radiation of a heterogeneous flame. The values of these temperatures for the stoichiometric ratio of the HF obtaining in all cases exceeded 1100 K.

Also, we calculated the cooling rate of the products of the process necessary to minimize the rate of reverse reaction of uranium dioxide fluorination with hydrogen fluoride.

Thus, the recovery of UF6 in the flame of the hydrogen-containing fuel and oxygen-containing oxidizers at temperatures above 1100 K, and fast cooling products can become the basis for a method of hydrogen fluoride obtaining from DUHF.

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Albumin-binding PSMA Ligands: Next Step Towards Improved Radionuclide Therapy of Prostate Cancer

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The prostate-specific membrane antigen (PSMA) has emerged as an attractive target for imaging and therapy of prostate cancer. Despite many advances in the past years, the treatment of metastatic castration-resistant prostate cancer (mCRPC) still remains challenging. The aim of this study was to optimize radionuclide therapy of mCRPC through the development of albumin-binding PSMA ligands with increased uptake of radioactivity in the tumor compared to the clinically employed Lu-177-PSMA-617.(1)

A variation of albumin-binding PSMA ligands was prepared using a solid phase synthetic approach and radiolabeled with lutetium-177 (Lu-177; T1/2 = 6.65 d) within 10 min at 95 °C. The Lu-177-labeled compounds were tested for stability, lipophilicity and potency of binding to human serum albumin (HSA). Cell uptake experiments were performed in PSMA-positive PC-3 PIP and PSMA-negative PC-3 flu cells. In vivo time-dependent biodistribution up to 8 days post injection (p.i.) and SPECT/CT imaging studies were performed in BALB/c nude mice bearing PC-3 PIP/flu tumor xenografts.

Lu-177-labeled PSMA ligands (50 MBq/nmol, radiochemical purity ≥98%) were stable in the presence of L-ascorbic acid over 24 h (≥95%). In vitro investigations revealed significant binding of the novel radioligands to HSA (≥95%). PSMA-specific uptake in PC-3 PIP tumor cells was high (60–63%, 4 h incubation) and comparable to Lu-177-PSMA-617. Biodistribution studies showed enhanced blood circulation of all radioligands and, as a consequence, an increased tumor uptake over time. The most promising radioligand showed a more than 2-fold increased area under the curve (AUC) for the tumor uptake as compared to Lu-177-PSMA-617. The tumor-to-kidney (~5.9) and tumor-to-blood ratios of AUCs (~46) were generally high, however, somewhat lower than ratios obtained for Lu-177-PSMA-617 (~37 and ~71, respectively). Finally, SPECT/CT imaging confirmed the high accumulation and retention of activity in PSMA-positive tumors as well as increasing tumor-to-blood and tumor-to-kidney ratios over time.

Taken all of the characteristics into account, the herein applied concept of albumin-binding PSMA ligands may potentially enable more potent and convenient radionuclide therapy of mCRPC using lower quantities of activity and/or its less frequent applications.

(1) Benesova et al., (2015) J Nucl Med. 2015; 56(6): 914–920.

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Radiochemcial analysis of long-lived radionuclides using mass spectrometry: New progress in the methodology

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Human nuclear activity has produced a large amount of radionuclides, some of them was released to the environment, while most of them are remained in various radioactive waste to be deposited. Among these radionuclides, long-lived radionuclides such as 14C, 36Cl, 79Se, 99Tc, 126Sn, 236U, 237Np, and isotopes plutonium, neptunium, americium and curium are most important in view of waste depository, decommissioning, environmental radioactivity and tracer studies of environmental processes. In addition, natural process also produce many radionuclides, such as 14C, 10Be, 26Al, 36Cl, 129I, 210Pb, 226Ra, these radionuclides are also important for radiation risk, especially in geological dating and environmental process investigation.
Radiometric methods based on counting of radioactive decay is conventional methods for measurement of radionuclides, especially for short-lived radionuclides, but the sensitivity is not sufficient for measurement of long-lived radionuclides (> 10 ky) in low-level. Mass spectrometry typically used for measurement of stable isotopes by counting the number of atoms of nuclides is suitable for measurement of long-lived radionuclides. However, the interferences from the samples matrix, isobars, polyatomic ions, tailing of high-level stable isotopes with a m/z close to the analyte radionuclide are the major challenge in its application for the determination of long-lived radionuclides of low-level samples. In the past years, mass spectrometry, especially accelerator mass spectrometry (AMS) and ICP-MS analytical techniques has been developed very rapidly, which significantly benefit to the determination of long-lived radionuclides, and stimulated its application in many research fields.
In our laboratory, we have developed serious analytical methods for the determination of long-lived radionuclides in environmental and waste samples by using radiochemical separation for removal interferences and AMS and ICP-MS for their sensitive measurement. An overview of these methods will be presented, with a focus on the following methods. (1) A novel ICP-MS method for determination of plutonium isotopes (239Pu, 240Pu, 241Pu) by using sequential mass spectrometers and reaction/collision cell. A removal efficiency of uranium of up to 1E8 has been achieved by employing a mixed reaction gasses to sufficiently removal of uranium and significantly improve the sensitivity of plutonium. (2) A sensitive AMS measurement method for determination of ultra-low level of 129I and 236U with microgram carrier isotopes (127I and 238U, respectively). New radiochemical separation and target preparation techniques were developed enable to prepare suitable and effective targets. (3) A novel AMS measurement method for rapid directly determination of 14C and 129I in environmental samples without chemical separation.

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Sulfonated calix-baskets for complexation of Barium and Radium

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Understanding the coordination chemistry of the heavy group 2 metal chemistry, especially of barium as surrogate for radium, is mandatory not only for adiopharmaceutical applications of radium. This is from high importance since Radium-223 is the only approved therapeutic alpha-emitter (by EMA and FDA). Unfortunately, the applications are limited. To date, radium-223 is only in use as RaCl₂ for the treatment of bone cancer metastases. To overcome this limitation, which is also true for other group 2 metals, special cage-like compounds have to be developed as ligands like sulfonated calix4crowns to stably bind the Ba²⁺ and Ra²⁺ to avoid a release in vivo. This will be the basis for a future application of heavy group 2 metals and not only of radium to treat other cancer entities than bone metastases. Ra²⁺ can then be included in radiopharmaceuticals which contain a chelator and a biologically active molecule part to find the

tumor cell.
For this purpose, a series of modified calix4crown-6 derivatives was synthesized to chelate barium, which serves as non-radioactive surrogate for radium-223/-224. These calixcrowns were functionalized with sulfonate moieties including deprotonable groups. Furthermore, the corresponding barium complexes were synthesized. Stability constants of these complexes were measured using UV/Vis titration experiments to determine logK values. Further radiolabeling and extraction studies were performed with [¹³³Ba]Ba²⁺ and [¹²⁴Ra]Ra²⁺ to further characterize the binding affinity of calixcrowns.

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Synthesis and radiofluorination of a novel monocarboxylate transporter 1 inhibitor for tumor imaging by PET

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Monocarboxylate transporter 1 (MCT1) is an integral plasma membrane protein that bi-directionally transports lactate and ketone bodies and is highly expressed in non-hypoxic regions of human colon, brain, breast, lung and other tumors. Accordingly, MCT1 inhibitors are regarded to be of potential clinical use. In the current study we developed a new 18F-labeled radioligand for in vivo imaging of MCT1-overexpressing brain tumors by PET.
A new fluorinated analogue of α-cyano-4-hydroxycinnamic acid (RM231) was synthesized from m-anisidine via alkylation, ortho formylation and Knoevenagel condensation in 50% overall yield. Its MCT1 inhibition activity was evaluated via [14C]lactate uptake assay on rat brain endothelial 4 cells. The mesylated precursor was similarly prepared in 52% overall yield. Radiosynthesis of [18F]RM231 was achieved by a two-step reaction, starting with the radiofluorination using [18F]-K2CO3-K222 complex followed by protective group removal via hydrolysis under optimized reaction conditions. RM231 showed relatively high MCT1 inhibition activity (IC50 = 12 nM). The radiolabeled intermediate was obtained by an optimized procedure (acetonitrile, 5.5 mg of K222, 0.7 mg of K2CO3, 12-15 GBq of K18F, 100°C, 8 min) with 44-50% yield determined by radio-HPLC analysis (N=3, non-isolated). The final product was obtained by hydrolysis with TFA in dry dichloromethane at room temperature for 10 minutes with 29% yield (radio-HPLC, non-isolated).[18F]RM231 could be obtained after separation using semi-preparative HPLC (RP C18 column; 30% ACN, 20 mM NH4CO2H). Currently, attempts are made to stabilize and formulate the final product appropriately for biological investigation. The newly developed MCT1 radioligand is anticipated to be a useful agent for imaging of the tumors with PET. Accordingly, animal studies on the new radiotracer are currently under investigation.
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Determination of 129I in aerosols using pyrolysis and AgI-AgCl coprecipitation separation and AMS measurement

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Airborne radioactive iodine is a key concern for transport and dispersion of radioactive contamination and radiation exposure evaluation during nuclear accidents and nuclear emergency preparedness. Long-lived 129I in aerosols is vital for reconstruction of level and distribution of short-lived and highly toxic 131I, as well as understanding the knowledge of atmospheric circulation process of iodine. However, aerosol 129I concentration is hard to measure due to its low concentration in the areas remote from nuclear pollution sources. In this study, a novel method for 129I in aerosols collected on glass fiber filter was developed using separation of high-temperature pyrolysis and AgI-AgCl coprecipitation coupled to highly sensitive AMS measurement. The chemical yield is 85±5% for iodine. The detection limit for 129I in aerosol is 1.8e6 atoms, which is 4 times lower than that using ashing-solvent extraction method in previous study. For aerosol samples collected in Asia with 129I/127I ratio of (1-10) e-9, only 500 m3 of air is sufficient for determination of 129I. Using this newly developed methods, the aerosol samples collected in an inland Chinese city were measured for 129I concentrations ranging within (0.31-7.3) e5 atoms/m3, and 129I/127I ratios of (0.26-7.4) e-8, which is is comparable to those in aerosols collected in Japan and Brazil, while 1-2 orders of magnitude lower than those in Europe.

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Alpha Radionuclide Therapy Using Polymeric Nanocarriers

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Alpha radionuclide therapy (ART) is a very powerful tool for the treatment of small tumour metastases. Due to their short range and high LET, alpha particles are much more efficient at killing cells than the commonly used beta radiation. Furthermore, their short range stops them from destroying neighbouring healthy cells. One of the main problems which still needs to be solved before ART can successfully be implemented in the clinic is the recoil problem: upon alpha-decay the daughter nuclide receives a recoil energy decoupling it from any targeting agent, allowing it to diffuse throughout the body to irradiate healthy tissue.

We have developed polymeric nanocarriers capable of retaining the recoiling daughters of the alpha-emitting radionuclide 225Ac and thus limit healthy tissue toxicity. Using a Monte Carlo-based simulation tool, a number of different polymersome designs have been simulated to optimize the recoil retention. Subsequently, polymersomes have been prepared with 225Ac co-precipitated with an InPO4 nanoparticle inside the vesicle, as the use of high-Z materials results in a much-reduced recoil range as compared to an aqueous environment. Using this new formulation, recoil retentions have improved significantly as compared to earlier published results by Wang et al., where 225Ac was encapsulated using a hydrophilic chelate 1.

Excellent results have been obtained in vitro, where the potential of 225Ac-loaded polymersomes has been evaluated in U87 glioblastoma multicellular spheroids. We have found that polymersomes distribute themselves throughout the spheroid after 4 days which, considering the long half-life of 225Ac (9.9 d), allows for irradiation of the entire spheroid. Our studies indicated that even at low radionuclide activity the 225Ac polymersomes deliver a very high dose, with spheroid growth

inhibition was already observed at just 0.1 kBq of 225Ac added. The therapeutic efficacy upon intratumoural administration of 225Ac-polymersomes has been tested in vivo in BALB/c mice bearing an MDA-MB-231 tumour. The retention of the vesicles upon intratumoural administration has been shown to be very high (46.0 ± 21.5% and 37.0 ± 23.9% at 1 day and 7 day p.i. respectively), whereas the tumours which have been injected with 225Ac-DOTA retained less than 1% (1 day p.i.), demonstrating the advantage of using the vesicles intratumourally. The accumulation of recoiled 213Bi in the kidneys was limited, with a kidney to tumour ratio of only 1:30. Immunohistochemical analysis of the tumours has shown an increase in double-stranded breaks in the group treated with 225Ac-polymersomes, indicating their suitability for tumour irradiation.

Large strides have thus been made towards the clinical implementation of polymersomes in ART. The incorporation of nanoparticles in polymersomes has allowed for high retention of the 225Ac mother and daughter nuclides, and both in vitro as well as in vivo their potential in destroying tumours has been demonstrated.

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Isolation of lanthanides from spent nuclear fuel by means of high performance ion chromatography (HPIC) prior to mass spectrometric analysis

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An important characteristic of spent nuclear fuel is its burnup, which is a measure of fissionable material consumption prior to fuel replacement. Isolation of lanthanides, most importantly neodymium, from spent nuclear fuel is necessary to determine the burnup experimentally. As an introduction, various ion chromatographic strategies for lanthanide isolation described in literature will be discussed briefly. In one approach, lanthanides are bound to a mixed-bed ion-exchange column and are eluted, with oxalic acid, as negatively charged oxalate complexes (e.g.1). The lanthanides elute sequentially in order of decreasing ionic radius, from La to Lu. In a second approach, a cation-exchange column containing sulfonic acid functional groups as active sites on the stationary phase, and a hydroxy carboxylic acid as mobile phase can achieve lanthanide separation (e.g.2). Lanthanides then elute in the opposite order to the first approach. Finally, reverse phase monolithic C-18 columns are a third type of chromatographic columns that can be used to separate lanthanides, provided these columns are first modified to dynamic ion-exchangers, by using e.g. camphor-10-sulfonic acid (e.g.[3]).
Secondly, an analytical method to isolate pure and complete fractions of neodymium, samarium, europium, gadolinium and dysprosium by means of high performance ion chromatography (HPIC) using a cation-exchange column and gradient elution with alpha-hydroxyisobutyric acid solutions of different concentrations and pH will be presented. Evaluation of intermediate precision and robustness against changes in pH of the eluent, lanthanide concentration and uranium matrix concentration indicated that the method proposed was reproducible. Eluent pH was identified as the most important parameter affecting the elution of the lanthanides. In addition, investigation of the elution behaviour of the most important fission and activation products and actinides indicated that in order to prevent the accumulation of cesium on the cation-exchange column, isocratic elution with a 1.0 M α-HIBA solution after elution of the lanthanides was required. Characteristics of the method presented will be discussed in relation to other studies dealing with the subject. Finally, sample preparation methods for removal of organic carbon prior to mass spectrometric analysis optimized by using acid digestion followed by UV photo-oxidation will be discussed. This presentation summarizes a recently published article 4.

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RPH 1 / 685

134Ce/134La for Positron Emission Tomography of Targeted Alpha Therapeutics

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The radionuclides employed in targeted alpha therapy are often actinides (e.g. 225Ac, 227Th) with multiple radioactive daughters. Successful treatment relies on the radiopharmaceutical's ability to "hold-on"to 225Ac (or 227Th) while it is delivered to the target cancer cell. Imaging surrogates of the radiopharmaceutical permit non-invasive pharmacokinetic assays and enable rapid screening of prospective compounds in living subjects. 134La emits positrons, enabling its use in positron emission tomography (PET) imaging in the form of an in vivo generator with its parent, 134Ce. Due to similar ionic radius and coordination geometries, the 134Ce/134La pair can be used as a surrogate for 225Ac or 227Th (non-PET isotopes) in the investigation of novel alpha-emitting radiotherapeutics.
This talk will focus on using the unique LANL capability at the Isotope Production Facility (IPF) at Los Alamos Neutron Science Center (LANSCE) to develop a production method for 134Ce (t1/2 = 75.9 h, daughter 134La t1/2 = 6.67 m, 2.7MeV β+). In addition, the approach to scale-up for bulk 134Ce production will be discussed. This process involves separation of microscopic (μg) amounts of 134Ce from macroscopic quantities (g) of lanthanum target material. Particular attention is focused on the Ce-La separation development that relies on "in-house"production of a 139Ce tracer (t1/2 = 137.6 d, 165.9 keV γ) via the 141Pr(n,3n)139Pr⟶139Ce reaction using the secondary neutron flux near a stack of IPF production targets.1

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NAM 2 / 477

In-Housing characterization of a working standard americium solution

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For many reasons the production and supply of nuclear certified reference materials is becoming increasingly difficult and expensive, some of these non-routine materials being utterly not commercially available at all. This lack of CRM may be critical for the capability of labs to support analysis. For these reasons, many labs, especially in the field of nuclear community, are involved in this issue and have to develop and implement within the lab working standard to meet the requirements of a certified reference material, our lab developed recently a methodology to overcome this problem to limit the risk of significant systematic errors in measurement systems.
This presentation deals with the entire process to prepare and maintain an "in-house"or working standard from a legacy material that consists of a batch of americium oxide powder, used for the determination of Americium assay by ICP-OES.
That includes procedures to handle and weigh, methods to dissolve and dilute but also to assess the americium concentration value and its uncertainty by different analytical methodologies ; a focus is made on the determination using the Isotopic Dilution - Thermal ionization mass spectrometry (ID-TIMS) method.
All the results are processed with an ANOVA approach to ensure accurately the concentration of the prepared mother solution.
To extend this procedure to others samples that may contain plutonium, a specific chemical pretreatment must be carried out to separate americium from other actinides to avoid isobaric interferences. This partition procedure includes a multi stage column-based extraction chromatography system and will be described.

RPH 1 / 593

119Sb: promising radionuclide for Targeted Auger Therapy - Production radiochemical separation and chelation aspects.

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Introduction: 119Sb is one of the most potent radionuclides for Targeted Auger Therapy due to convenient energy and numbers of Auger electrons as well absence of any other accompanying emissions [1-3]. Antimony-119 has a half-life of 38.5 hours which is well suited for radiotherapeutic application. In the present work: production, radiochemical separation and chelation preference of antimony radioisotopes were studied.
Production of antimony isotopes was performed by proton irradiation of a natural tin target with energies of 13 and 16 MeV as well as by neutron bombardment of natural tin and antimony. Radiotracers were used for establishing radiochemical procedures for isolating of bulk mass of tin from produced no-carrier-added antimony isotopes
Radiochemical separation: Dissolution procedure for irradiated tin target was established by using hydrochloric acid and hydrogen peroxide. Further liquid-liquid extraction with ethers or ketones was applied for separation of antimony from bulk tin mass. Methods based on solid phase extraction chromatography and ion exchange chromatography in various media were tested as well.
Chelation: Several common chelation systems along with some of the novel candidates from -pa family were tested to identify chelation preference of antimony radioisotopes.
Acknowledgments: We would like to thank Dr. Jan Kozempel from Charles University in Prague, Czech Republic and Dr. Steffen Happel from Triskem International, Bruz, France for synthesizing

and providing samples of custom resins for Sn/Sb separation. TRIUMF receives federal funding via a contribution agreement with the National Research Council of Canada.

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NAM 2 / 519

ICP-MS measurement versus radiometric detection for Radium-226 analysis in environmental liquid matrices

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Among the natural isotopes of radium, Radium 226 is one of the most radiotoxic agent. It is part of the decay chain of Uranium 238 and, with a half-life of 1600 years, it is the most common isotope of Radium present on Earth. Due to its chemical properties, the metabolic pathway of Radium is similar to that of Calcium and it can concentrate mainly in the bones and thus increase the internal dose and the risk of occurrence of certain diseases. For this reason, Radium 226 is of paramount importance in the field of radiation protection.

In order to optimize its environmental monitoring and to better understand its biogeochemical dynamics and to quantify its transfer, the analysis of Radium 226 at the lowest possible levels is necessary. Our aim was to implement an analytical procedure complying with the requirements of environmental monitoring and radioecological studies all over French territory, but also to assess a robust protocol for accidental or post-accidental situations, where higher activity levels are expected. One way for lowering detection limits is to measure this radioactive isotope by inductively coupled plasma mass spectrometry (ICP-MS).

In this context, we choose to develop Radium-226 analysis by ICP-MS in environmental liquid matrices. The selection of the internal standard, the influence of the interfering species on the measurement and the ICP-MS measurement parameters will be discussed. The protocol has been tested with standard solutions and proficiency test samples, and showed very good accuracy.

A comparison of the analytical performances of mass spectrometry versus radiometric methods (limit of quantification, time of analysis…) demonstrates the strengths of radium-226 analysis by ICP-MS.

Poster TAN / 486

Self Diffusion of Bk3+ in aqueous solutions at neutral pH and pH 2.5. Comparison with the trivalent f-elemnts (Eu3+ , Gd3+ , Tb3+ , Tm3+)

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Key words : Self-diffusion (coefficients)/ Solvation / Actinide/ Lanthanide / Trivalent (aqueous ions)/ Hydolysis.

Abstract:

In this paper, we present a realistic model for estimation of trivalent actinides and lanthanides ions self-diffusion coefficients. We take account hydrolysis phenomena. We use Fuoss theory and Bjerrum approach to calculate Kh the thermodynamic hydrolysis constant. We use Marcus summarized data for different radius. We suppose that ion structure skills the same in solid state (salt) or in aqueous solution, regarding the water molecules surrounding central ions (M ≡ La3+ , Ac3+).
Taking account hydrolysis, experimental self-diffusion which allowed us to deduce the structure of the entity hydrated particularly for Berkelium ion.
All my gratitude to Pr R.G Haire my supervisor at Oak Ridge National Lab USA.

Poster RER / 428

Sorption of 137Cs from aqueous solutions onto layered double hydroxides containing the Fe(CN)64–ion in the interlayer space

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Early we studied the sorption of microamounts of 60Co2+, 85,90Sr2+, 90Y3+, 131I–, 131IO3–, 137Cs+, and 233,238U(IV) onto layered double hydroxides (LDHs) of Mg, Cu, Al, and Nd, containing various anions in the interlayer space, and also onto layered double oxide (LDO) of Mg and Al, prepared by thermal decomposition of the corresponding LDH. It was found that LDH-Mg-Al-Anion (Anion = CO32–, NO3–) solid phases did not take up 131I in the form of 131I–and 131IO3–from aqueous solutions, whereas the efficiency of the 131I–and 131IO3–removal from aqueous solution with LDO-Mg-Al exceeded 99%. Synthetic analogs of hydrotalcite of the composition LDH-Mg-Al-Nd-CO3 and LDH-Mg-Nd-CO3 allowed 90Sr2+ and 90Y3+ to be removed from aqueous solutions with more than 99% efficiency. The compounds LDH-Mg-M3+-CO3 (M3+ = Al, Nd) remove 233,238U(VI) from aqueous solutions of complex chemical composition with more than 99% efficiency. 60Co is efficiently sorbed from 10–3–10–5 mol/l aqueous Co(NO3)2 solutions onto LDH-Mg-Al and LDH-Mg-Nd with CO32–ions in the interlayer space. At the contact time of the solid and liquid phases (τ) of 15 min and V/m = 50 ml/g, Kd exceeds 2·104 ml/g for LDH-Mg-Nd and does not exceed 5·103 ml/g for LDH-Mg-Al. On the other hand, 137Cs+ was not noticeably taken up from aqueous solutions by any of the previously studied LDHs and LDOs. The highest values of Kd, equal to ~25 ml/g, were obtained for LDH-Mg-Nd-CO3 at τ = 2 h and V/m = 50 ml/g. In the other cases, Kd did not exceed ~5 ml/g.
Inorganic sorbents containing ferrocyanide ions are widely used for removing cesium radionuclides from aqueous solutions. Therefore, we prepared in this study samples of LDH-M2+-M3+-Fe(CN)6 (M2+ = Mg, Cu, Ni, Zn; M3+ = Al, Fe) and determined their physicochemical properties, including the performance in the 137Cs+ sorption.
Sorption of 137Cs from aqueous solution onto LDHs of the composition LDH-M2+-M3+-Fe(CN)6 (M2+ = Mg, Cu, Ni, Zn; M3+ = Al, Fe) was studied. The LDH-Mg-M3+-Fe(CN)6 (M3+ = Al, Fe) take up 137Cs from 10–5 mol/l aqueous CsNO3 solutions extremely weakly, whereas the LDH-M2+-M3+-Fe(CN)6 (M2+ = Cu, Ni, Zn; M3+ = Al, Fe) solid phases efficiently take up 137Cs. After 15-min contact of the solid and liquid phases, the apparent distribution coefficient Kd* of 137Cs ranges from ~3·102 to ~104 ml/g for M2+ = Cu, from ~4·103 to ~2·104 ml/g for M2+ = Ni, and from ~5·103 to ~3·104 ml/g for M2+ = Zn at V/m = 50 ml/g. As the NaOH concentration in the solution is increased, the sorption performance of the examined LDHs, except LDH-Ni-Al-Fe(CN)6, drastically decreases, which is due to the formation of LDH-M2+-M3+-OH (M2+ = Ni, Zn; M3+ = Al, Fe) having low ability to take up 137Cs.
Thus, synthetic analogs of hydrotalcite of the composition LDH-M2+-M3+-Fe(CN)6 (M2+ = Cu, Ni, Zn; M3+ = Al, Fe) allow 137Cs+ removal from aqueous solutions with more than 99% efficiency. Apparently, these compounds can find use for removing cesium radionuclides from neutral aqueous solutions in various flowsheets.

Poster RCH / 437

Comparison of two types of LuCl3 pharmaceutical substance (NCA and CA) for production of radiopharmaceuticals for the Peptide Receptor Radionuclide Therapy.

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Over the past decade, the scientific community involved in the research and development of therapeutic radiopharmaceuticals (RPhs), has been taking growing interest in the isotope of the rare-earth element, Lu. 177Lu radionuclide (T1/2 = 6.65 days) exerts the properties of a "soft"β—emitter (Eβmax = 498 keV), which makes it convenient for treatment of small malignant formations (the maximum range in soft tissues Lmax = 2 mm) of different nature and localization, and the gamma component (Eγ = 208 keV) allows to visualize the agent biodistribution by means of scintigraphic imaging.

Lutetium is the smallest representative of lanthanides with a characteristic degree of oxidation (+3) and abilities to form complexes with various ligands, in particular with electron-donor bifunctional chelating agents (DTPA, DOTA, EDTA) conjugated to a molecule specific to receptors on the surface of a malignant cell, this principle underlies the Peptide Receptor Radionuclide Therapy (PRRT). Thus, selecting the optimal peptide transporter, it becomes possible to control the biodistribution of 177Lu in a patient's body, and the use of a chelator with a high stability constant of the metal complex (lg KML) results in minimizing the diffusion of free 177Lu ions into healthy organs and tissues.

177LuCl3 salt in a 0.05 M solution of HCl is used as an active pharmaceutical substance when producing RPh for PRRT. However, the possibility of using different target materials to obtain 177Lu in the reactor according to the following reactions:

176Yb (n,γ) 177Yb ⊠ 177Lu + β- + ῡe (1)
176Lu (n,γ) 177Lu + 177mLu (2)

leads to formation of two types of raw material: without a carrier (NCA) and with a carrier (CA), distinguished by the amount of lutetium isotopic impurities, which decrease the specific activity of the finished RPh. Meanwhile the presence of the long-lived nuclear isomer, 177mLu, (T1/2 = 160 days) in the raw material leads to an increase in unreasonable radiation loads on the patient's body.

The works on labeling of the peptide, the octreotide analogue with a conjugated chelator: DOTA-Tyr3-octreotate (DOTA-TATE) by means of 177LuCl3 NCA raw material (SA = 102.41 Ci/mg at the moment of first labeling) and CA (SA = 30.2 Ci/mg at the moment of first labeling) were carried out. In both cases, the peptide sample weight, the reaction mixture volume and the synthesis conditions remained unchanged, but 177LuCl3 volumetric and specific activity was changed.

The aim of the work done was to identify the optimal molar ratio of peptide to lutetium (νpept : νLu), and of peptide to lutetium-177 isotope (νpept : ν177Lu) for both types of raw material. The effect of a decrease in the specific activity of the raw material during storage on the yield of the labeling reaction was considered. The control indicator of the finished product quality was the incorporation yield of 177Lu+3 ions into the composition of the transporter molecule of not less than 95 %, defined by ITLC and HPTLC methods in the system ammonium acetate - methanol.

Poster RCH / 496

Interfacial formations in extraction systems

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Liquid extraction is widely used in radiochemistry for the concentration and separation of elements. When the organic phase containing the extragent contacts with the aqueous phase in which contains

element the reactions occur in the interfacial layer. As a result of the reactions, several compounds are formed, among which there are often those that do not directly pass into either the organic or the aqueous phase. Their molecules accumulate in the interfacial layer, aggregate and coagulate forming cruds. At a certain ratio of the quantities of the extracted element and the extragent, a precipitate is formed that can sediment to the bottom of the vessel or settle in pipelines connecting the extractor with other processing equipment. Interfacial formations have a negative effect on the extraction of the element.

This report presents the results of studies on the formation of interfacial formations in the extraction of rare-earth elements (REE) by solutions of di- (2-ethylhexyl)phosphoric acid (D2EHPA).

The resulting di-(2-ethylhexyl)phosphate lanthanide (has some surface activity) is able to accumulate in the dynamic interfacial layer (DIL) of the extraction system. Over time, the accumulation of REE in the DIL increases. The processes of aggregation, coagulation and polymerization are proceeding, as indicated by the absorption at 1180 cm-1 and 1090 cm-1 in IR spectra of interfacial formation. These indicate the presence of bridge alkylphosphate groups in linear polymers. The formation of structure in the DIL leads to the appearance of interfacial formations, the mechanical strength of which is not great for both the elements of the yttrium and the cerium subgroups. However, in the case of elements of the cerium subgroup, the interfacial formations deforms reversibly, and in the case of elements of the yttrium subgroup it is partially destructed (for example, when heptane is used as a diluent of D2EHPA).

Kinetic curves of accumulation of REE in the DIL are characterized by the presence of a region when the accumulation of a REE remains constant. The length of this region and its position on the kinetic curve of accumulation depends on many factors (the concentration of REE and extraction reagent, acidity of the medium, the nature of REE and diluent of the extragent). The amount of the REE accumulating in the DIL depends on these same factors. In the heptane system used as the diluent of the extragent, when Er(III) is extracted from the chloride solution, the accumulation is higher (23% at the time of 60 min) than in the system with toluene being the more polar diluent (12% to this same time). A region with a constant value of accumulation of a REE in the DIL is short, often weakly expressed and observed almost at the very beginning of the experiment (⊠5-7 min). In systems with toluene, the region with a constant value of accumulation is well expressed, longed (15⊠40 min) and at a lower degree it depends on the concentration of the extragent. An increase in the concentration of D2EHPA and a decrease in the concentration of the REE, all other things being equal, lead to a decrease in the accumulation of the REE in the DIL. The obtained dependences can be explained by the change in the intensity of interfacial phenomena, of which spontaneous surface convection is important.

Thus, the formation of interfacial formations in the DIL is due to chemical reactions, adsorption, formed molecules, their aggregation and polymerization, partial emulsification, in view of the appearance and development of spontaneous surface convection, the phase separation and the formation of structure.

Poster RER / 429

Sorption of U(VI) onto layered double hydroxides and oxides of Mg and Al, prepared using microwave radiation

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Layered double hydroxides (LDHs) belonging to two-dimensional supramolecular systems are compounds of the composition [(M2+)1−x(M3+)x(OH)2]•[(An−)x/n•mH2O], where M2+ and M3+ are cations in oxidation states 2+ and 3+, respectively; An−is practically any anion or anionic complex. An advantage of LDHs over other layered compounds is the fact that various anions can be intercalated in their interlayer space, with the substitution of anions in the interlayer space occurring without break of the layered structure. The presence of M2+ and M3+ ions in the LDH structure suggests the possibility of ion exchange with cations of radioactive elements in aqueous solution.

Previously we studied the sorption of U(VI) onto LDH-M2+-M3+ (M2+ = Mg, Zn; M3+ = Al, Nd)

and LDO-Mg-Al from aqueous solutions of various compositions. In all the studies, the synthesis of LDH-Mg, Zn, Al, and Nd containing various anions in the interlayer space took no less than 24 h. This is caused by the need for long recrystallization of the precipitate in the course of its formation and heating at 60–80°C. On the other hand, it is known that the use of microwave radiation (MWR) allows considerable acceleration of various physicochemical processes. In this connection, it was interesting to determine whether short microwave heating of a freshly formed LDH precipitate in the mother liquor can replace its prolonged heating at 80°C. In so doing, it was necessary to determine how the change in the synthesis procedure will influence the ability of the synthesized LDH to take up U(VI). The above problems became the subject of this study.
LDHs of Mg and Al, containing CO32–ions in the interlayer space (LDH-Mg-Al-CO3), and layered double oxides of Mg and Al (LDO-Mg-Al) were prepared using MWR. The use of MWR allows not only acceleration of the synthesis of both LDH and LDO, but also preparation of compounds with high kinetic characteristics of the U(VI) sorption. The degree of U(VI) sorption (α) from 10–2 mol/l aqueous U(VI) solutions at a sorption time of 4 h and V/m = 50 ml/g exceeds 99.0%. In sorption from more concentrated (10–1 mol/l) aqueous U(VI) solutions under similar conditions, α on all the samples does not exceed 37.5%.
It was found that the use of MWR in the synthesis of LDHMg-Al-CO3 and LDO-Mg-Al allows not only acceleration of the synthesis of both LDH and LDO, but also preparation of compounds with high kinetic characteristics of the U(VI) sorption.

Poster TAN / 532

Structural diversity of uranyl complexes with aliphatic monocarboxylate ligands

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In most of the cases interaction between uranyl ions UO22+ and anions of aliphatic monocarboxylic acids L leads to formation of mononuclear anionic complex units [UO2L3]–. Crystal structures of corresponding compounds are usually three-dimensional due to electrostatic interactions or hydrogen bonding among the complex ions [UO2L3]–and different counter ions. Such compounds have long been studied, especially with acetate ions. However, over the past several years we were able to substantially enlarge the number of uranyl compounds with higher homologues of acetate ion, such as propionate, butyrate and valerate. The results of our synthetic efforts are somewhat surprising. Here we highlight several peculiarities of this family of compounds.
First, there are rare examples of compounds of uranyl ions with anions of aliphatic monocarboxylic acids L constructed of larger heteronuclear clusters. For example, the crystal structure of n-butyrate containing compound {Sr(H2O)4[UO2L3]2}·2H2O is constructed of trinuclear neutral clusters and the unprecedented neutral octanuclear clusters K[UO2L3]{R(H2O)4[UO2L3]2}2 were observed in crystal structures of two similar K and Sr or Ba propionates of uranyl ion.
Formation of heteronuclear clusters in the mentioned systems is possible mostly due to the bridging function of anions of aliphatic monocarboxylic acids, which is considered unusual for these ligands. Such coordination mode allows the typical [UO2L3]–complex units to bind in larger architectures. According to statistical analysis of the CSD, the bridging function is in fact the most common for butyrate and valerate ions. Thus, even such featureless at the first view monotopic ligands as the ions of aliphatic monocarboxylic acids may facilitate formation of heteronuclear clusters with uranyl ions if other metal atoms except for U are available. This is another curious fact about this family of compounds. Besides, information on the possible and preferred coordination modes of ligands derived in the course of our studies is extremely important for crystal structure prediction and crystal engineering.
The current versions of CSD and ICSD contain respectively about 0.2 and 3.7% of crystal structures featuring cubic crystal system and excluding the inversion center. Curiously and unexpectedly, many uranyl monocarboxylate complexes are noncentrosymmetric and crystallize in cubic crystal system. Noncentrosymmetric crystals may show nonlinear optical (NLO) activity. This third curious fact enhances the value of aliphatic monocarboxylate complexes of uranyl ions and motivated us for

the search of new crystals with high NLO activity.

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Poster RCH / 500

Influence of gamma radiation dose rate and some other parameters on the radiation protection of microbial cells by OH radical scavenging

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The effects of dose, dose rate, hypothermia (1 hour at 0 ℃ before and after irradiation) and simultaneous action of two different scavengers on the radiation protection of microorganisms due to the scavenging of OH radicals were investigated. The quantitative evaluation of this protection was carried out by means of the slope k = dσ / dQ where σ = ln S0 / ln S, Q is the scavenging efficiency of the scavenger and S0 and S are the surviving fractions of cells after irradiation without and with scavenger, respectively.

Methanol and ethanol were used as scavengers. The radiation sensitivity of both prokaryotic (bacteria Escherichia coli) and eukaryotic (yeast Saccharomyces cerevisiae) cells was studied. Irradiation was carried out by gamma radiation of radionuclide 60Co in Gammacell 220. The values of dose rates ranged from 18.7 to 41.7 Gy h-1.

The σ-values were found to be higher than 1 and they increase linearly with the scavenging efficiency Q for both microorganisms in intervals from 0 to 1.86 x 109 s-1 for methanol and from 0 to 4 x 109 s-1 for ethanol. These results lead to the conclusion that both scavengers (methanol and ethanol) protect living cells against ionizing radiation. The scavenging of radicals is probably one of the main mechanisms of protection against irradiation. The specific protection k of both yeast and bacteria does not depend on the dose of gamma radiation and linearly increases with dose rates in above mentioned interval for both alcohols. Both protections σ and k are higher for bacteria than for the yeast. It may be related to the dissipation of DNA molecules in cytoplasm of bacteria. No effect was found when the microbial culture undergoes the hypothermia before and after irradiation. No synergic effect was determined when both scavengers were used simultaneously.

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Poster TAN / 586

Synthesis, structure and yhe state of [MIII(UO2)6O4.5(OH)6]•6H2O (MIII –Nd, Sm, Eu, Gd, Dy) in aqueous solutions

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Method of synthesis of the REE hexauranates is proposed in this work. The composition and structure of these compounds were defined, processes of dehydration and thermal decomposition were studied, the state in aqueous solutions was investigated by X-ray fluorescence spectrometry, chemical analysis, IR spectroscopy, high-temperature X-ray diffraction and thermography. The choice of

MIII (MIII –Nd, Sm, Eu, Gd, Dy) in present study was limited by those rare earth elements which form the compounds with same chemical composition. All synthesized compounds are formular analogues and their chemical composition corresponds to general formulae MIIIU6O19.5*10H2O. The REE hexauranates have not only same chemical formulae but they are also crystallographic analogues. The presence of intense reflection in the region of small angle 2θ, that is characteristic of uranyl compounds, indicates the formation of layered structure type of the compounds under study. For evaluation of functional composition of MIIIU6O19.5*10H2O, we have performed the IR spectroscopic researches. The IR-spectrums contains four groups of vibrations associated with uranyl moieties UO2δ+, bounds U–Oeq in the equatorial plane of uranium polyhedron, H2O molecules and uranium-hydroxide groups. To assess the state and role of H2O molecules in the formation of the MIIIU6O19.5*10H2O structure it was held thermographic analysis in combination with X-ray powder diffraction and IR-spectroscopy. The dehydration process of studied compounds occurs in two stages. First endoeffect on the DTA curve is observed at 195-203°C and, according to thermogravimetric study, it results to the removal of seven out of ten of the water molecules and the formation of MIIIU6O19.5*3H2O phase. The ν(H2O) and δ(H2O) bands in the range 3600-3400 cm-1 and 1613-1620 cm-1 disappear, but ν(U-OH) and δ(UOH) at 3320 cm-1 and 962 cm-1 remain in the IR spectrum of the dehydration product. This transformation of IR-spectrum can take place if the dehydration product contains the equivalent number of hydroxyl groups OH instead of three H2O molecules. In this case the resulting compound would have the composition MIII[(UO2)6O4.5(OH)6]. The dehydration process ends at 302-332°C that is accompanied by a complete destruction of the crystal lattice.
The state of the REE hexauranates in aqueous solutions obeys the general laws, that is in good agreement with their similar structure. Among the variety of factors, the most significant influence over state of MIII[(UO2)6O4.5(OH)6]*7H2O compounds in heterogeneous systems provides the acidity of an aqueous phase. All MIII[(UO2)6O4.5(OH)6]*7H2O compounds are chemical stable and preserve their structure in heterogeneous aqueous-salts system in wide acid-base interval from pH 0 to 11.5-12.0. The structure of hexauranates is completely destroyed in strongly alkaline medium. Na2U2O7 and MIII(OH)3 are formed under these conditions. The REE hexauranates solubility varies by several orders of magnitude from 10-7 M in neutral solutions to 10-4-10-1 M in acid and strong alkaline conditions. The nature of interlayer REE atom does not influence significantly over solubility of studied compounds.

Poster RER / 440

Marine organism concentration factors and sediment distribution coefficients

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The concentrations factor of 14 heavy metals in marine organisms at the neighbouring sea of Korea were measured and investigated, respectively. The 359 seawater samples, 332 fish samples, 20 Cephalopods and 22 Crustaceans, 15 Molluscs, 38 Macroalgae were measured the concentrations of heavy metals. The concentration factors in 314, 22 sampling locations were analyzed in concentration of seawater and marine organisms, and in that of seawater and sediment, respectively.
The concentrations in seawater were Sr(7,030 μg•kg-1), Rb(106 μg•kg-1), Fe(13.6 μg•kg-1), and Cs(2.08 μg•kg-1), respectively. The concentrations of muscle of marine organism were Sr(56.1 mg•kg-1), Fe(23.0 mg•kg-1), Zn(9.12 mg•kg-1), Cs(5.64 mg•kg-1), and Mn(1.88 mg•kg-1), respectively.
The concentration factors in all of organism were highest in P, Mn, Cs, Fe and lowest in Na, Sr, Rb, Mo. The concentration factors of muscle in fishes showed strikingly high P(383,000), Mn(42,700), Cs(25,000) and Sr(3.06), respectively. Highest concentration factors of P(286,000), Cu(169,000), Mn(111,000), and Sr(10.1) were measured in Molluscs, respectively. The concentration factors of macroalgae showed P(127,000), Mn (64,800), Cs(25,500), and Sr(15.1). The results indicated that the concentration factors of 14 heavy metals in the Echinoderms, Molluscs were higher than that in the fish and cephalopods except for Mo, Ba.
The concentrations of sea sediment were Fe(32,700 mg•kg-1), Mn(705 mg•kg-1), Sr(467 mg•kg-1), and Cs(0.889 mg•kg-1), respectively. The sediment distribution coefficients showed Fe (88,600,000), Mn(8,020,000), Cs(4,560), and Sr(71.2) , respectively.

The mean concentration factors and the sediment distribution coefficients of the heavy metal were similar to the recommended value from IAEA (2004) and IAEA (2010).

Poster RCH / 598

Modification of eukaryotic cells’radiation sensitivity by various hydroxyl radiacal scavengers

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The influence of ionizing radiation on living cells’radiation sensitivity can be affected by many parameters like dose rate, temperature, level of oxygen in irradiated system or presence of some chemicals (radioprotectors or radiosensitizers). Radioprotectors can reduce radiation sensitivity of living cells to ionizing radiation due to their high reactivity with hydroxyl radicals produced during radiolysis of water.
The aim of this work was to study the influence of various scavengers on radiation sensitivity of yeast (*Saccharomyces cerevisiae*, haploid strain, type *a*, DBM 272) under gamma irradiation of Co-60 at the dose rate and the dose 40 Gy/h and 150 Gy, respectively. *Saccharomyces cerevisiae* cells were cultivated on Sabouard chloramphenicol nutrient agar layer M 1067, HIMEDIA for 5 days at 30 ℃. The results were gained by the counting of colonies grown up from surviving irradiated or nonirradiated cells in presence of various concentrations of the scavengers (methanol, ethanol, t-butanol, iso-butanol, n-amyl alcohol, dimethyl sulfoxide, acetone, potassium formate and DL-alanine).
The radiation protection σ was defined as a ratio of natural logarithm of surviving fraction of the cells without ($s0$) and with scavenger (s). The specific radiation protection k, i.e. radiation protection based on hydroxyl radical scavenging, was defined as a slope of the dependence of radiation protection σ on a scavenging efficiency Q. The scavenging efficiency Q is defined as a product of scavenger’s concentration and the reaction rate constant kOH of the reaction of hydroxyl radical with the scavenger.
It was found that the radiation protection σ of all scavengers linearly increases with increasing scavenging efficiency Q. It means that all scavengers under study act as the radioprotectors. The specific radiation protection k hyperbolically decreases with increasing rate constant kOH and it linearly rises with increasing concentration c of scavengers at the constant scavenging efficiency Q. Therefore, the radioprotection seems to be a complex process.

Poster TAN / 591

Synthesis and study of cesium uranate Cs2U6O19•10H2O

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There is only one stable cesium isotope with a mass number of 133 in nature. However, an artificial radioactive cesium isotope - cesium-137 is the most radiationally hazardous and practically important. Its main source is the fission reaction of uranium nuclei. The efficacy of separation of ¹³⁷Cs from nuclear waste depends on the chemical and structural compatibility of uranium (VI) and cesium especially in aqueous media.
Cesium uranate Cs₂U₆O₁₉•10H₂O was synthesised for the first time via a reaction between synthetic schoepite UO₃•2.25H₂O and aqueous solution of CsOH. The composition and structure of the compound was studied using

X-ray fluorescence spectrometry, X-ray powder diffraction, IR-spectrometry, differential thermal analysis and thermogravimetry.
Elemental composition of the cesium uranate was established via XRF after dissolving the compound in aqueous solution of nitric acid. Following spectral lines were used in the analysis: CsLα and ULα. The amount of water in the formula unit was determined by heating the sample to 600°C. The compound was established to be a hydrate with a formula: Cs2U6O19•10H2O.
X-ray powder diffraction shows that the compound is an individual crystalline phase. Reflections at small 2θ angles and pinacoidal peaks indicate layered structure of the studied compound.
Four types of absorption bands are found on the IR-spectrum. They correspond to the molecular water, the uranyl moiety UO₂^{δ+}, UOH bonds, and U-O_{eq} bonds in the coordination polyhedron of U. Characteristic band for δ(H₂O) is shifted from 1595 cm⁻¹, as observed for gaseous H2O molecules, to 1624 cm⁻¹ due to the forming of hydrogen bonds. Due to the same reason both symmetrical and asymmetrical stretching ν_{as}(H₂O) and ν_s(H₂O) are not resolved into the constituents and give a wide integral band at 3508 cm⁻¹. A band at 925 cm⁻¹ is assigned to asymmetrical stretching ν₂(UO₂^{δ+}). Absence of symmetrical stretching band indicates that the uranyl moiety has a linear configuration and equal bond lengths. A band at 996 cm⁻¹ and a shoulder at 3315 cm⁻¹ are assigned to bending δ(UOH) and stretching vibrations ν(UOH) of uranium hydroxide fragment respectively. Bands at 400 to 600 cm⁻¹ can be assigned to planar vibrations of U-O_{eq}.
The hexauranate is not thermally stable. Six molecules of H₂O leave the Cs2U6O19•10H2O by 160°C. Upon further heating to 310°C the remaining 4 molecules of H₂O evaporate and the compound becomes amorphous. New crystalline structure forms at 600°C and remains virtually the same in the temperature interval from 700°C to 1000°C.
Conducted experiments allow us to predict the structure of the synthesised cesium hexauranate. Oxygenous coordination polyhedra of U form the negatively charged layers, while Cs⁺ cations and water molecules are situated in interlayer space. They along with hydroxide groups bind the layers together creating a three dimensional lattice.
All experiments were conducted using analytical instruments made by Shimadzu Corp.

Poster RER / 455

Uranium isotopes (234U and 238U) in calcium and magnesium supplements

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The objectives of this research were to investigate the naturally occurring 234U and 238U in calcium and magnesium supplements, find the correlations between 234U and 238U concentration in medicament and its chemical form, as well as calculate the effective radiation dose connected to analyzed supplement consumption. The analyzed Ca and Mg pharmaceutics contained their organic or inorganic compounds; some from natural sources as shells, fish extracts, or sedimentary rocks.
Uranium is widely spread in nature, occurs in over 160 minerals, locally at high concentrations. Isotopes 234U and 238U occur naturally in uranium decay chain; both of them are alpha emitters of low radioactivity and radiotoxicity. The harmful effect of uranium results from its high chemical toxicity which is comparable to lead. The occurrence of uranium in environment can also be result of human activity –nuclear industry, combustion of fossil fuels, production and use of phosphorous fertilizers use of depleted uranium for military purposes. Next calcium and magnesium are one of the most essential elements in living organisms and their deficiencies are common so their supplements have become extremely popular.
The highest 234U and 238U activity concentrations, in both Ca and Mg supplements, were found in all samples of natural origin –marine shells or sedimentary rocks. Among chemically processed supplements (during production), there was one exception, namely chelate compound, which contained high uranium concentrations as well. Similarly to 210Po and 210Pb analysis in Ca and Mg supplements, the results showed, inorganic forms of analyzed supplements were richer in 234U and 238U than organic.

On the basis of 234U and 238U content calculated in analyzed calcium and magnesium supplements, the annual effective radiation doses were estimated and obtained data showed there is no radiological risk connected to uranium ingestion with calcium and/or magnesium supplements.

Poster TAN / 648

Development of an atomic beam apparatus for chemistry of the heaviest elements

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Chemical studies of the heaviest elements provide crucial and challenging opportunities not only to advance our understanding of properties of matter at the limits of existence but also those to elucidate the influence of relativistic effects on atomic electrons and to architect the periodic table of the elements at the farthest reach [1,2]. The influence of relativistic effects on electronic orbitals has, so far, indirectly inferred through a comparison of chemical properties of the heaviest elements with those of their lighter homologues and those predicted by theoretical calculations. The first ionization energy (IP_1) is one of the most sensitive atomic properties which reflect the outermost electron configuration. Thus, accurate IP_1 values of the heaviest elements allows us to give significant information on valence electronic configuration affected by relativistic effects.
The ground-state electronic configuration of element 103, lawrencium (Lr), is predicted to be $[Rn]5f^{14}7s^27p_{1/2}$ which is different from that of the lanthanide homologue Lu, $[Xe]4f^{14}6s^25d$, because the $7p_{1/2}$ orbital is expected to be stabilized below the 6d orbital by strong relativistic effects [3]. Lr is expected to be the first element where relativistic effects would directly change the electronic ground-state configuration with respect to the normal prediction of the periodic table.
In the previous paper 4, we reported the determination of IP_1 of Lr by using a novel technique based on a surface ionization process coupled to an on-line mass separation technique. Our experimental IP_1 is in excellent agreement with theoretical calculations. This good agreement with predictions obtained using relativistic calculations, which favour a $7s^27p_{1/2}$ configuration in the Lr atom, supports this ground-state configuration.
In the next stage, we plan to directly determine the ground-state configuration of Lr by applying the Stern-Gerlach technique of magnetic deflection of atomic beams. Here, a well-collimated atomic beam passes through an inhomogeneous magnetic field that splits the beam into $(2J + 1)$ components; J is the total electronic angular momentum of the atom. From the number of beam split components, the ground-state configuration of the Lr atom can be determined unambiguously. We have just started the development of an apparatus for effective extraction of the atomic Lr beam by exploiting a small cavity type atomic beam source that can be heated by electron bombardment. In the contribution, we present the status and achievement, and future prospect of this program.

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Poster RER / 456

Extraction efficiency of 210Po in Polish herbal teas

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Air and food are the main sources of many chemical elements, also natural and artificial radionuclides transferred to human organisms. The intensity of radioisotopes intake depends on the place of residence, local radiation quantity, diet habits and food origin. So far, during annual radiation doses evaluations in Poland, the most often consumed food products were taken into account. Among naturally occurring radionuclides, their potential ingestion and internal expose, the most important seems to be 210Po and its parent nuclide 210Pb.

Presented are results of a study on 210Po extraction efficiency in Polish herbal teas and risk to human consumer due to exposure from highly radiotoxic decay particles emitted by 210Po. 12 most popular commercially available Polish herbal teas, and their infusions in tap water and filtered water were analyzed and 210Po activity concentrations were calculated.

The results in dried plants were between 2.11±0.09 for milk thistle and 33.70±0.42 Bq•kg-1 dry wt. for cistus. The extraction efficiencies into tap water ranged from 4.93±0.39 % for lime to 27.40±1.43 % for elder, while for activated carbon filtered water were between 7.55±0.47 % for lime and 20.32±1.09 % for elder and there were no statistically significant differences between both extractions. There was no correlation found between 210Po extraction efficiency into infusions and 210Po activity concentrations in dried herbs. Herbal teas consumption should not contribute significantly to the annual effective radiation dose in Poland.

Poster RCH / 606

XPS study of ion irradiated and unirradiated CeO2 samples

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Cerium dioxide, CeO2, is a fluorite structure ceramic widely used as an inactive structural surrogate to UO2 and PuO2 to avoid difficulties associated when working with radioactive materials. This material is suggested to be used as an inert matrix for perspective nuclear fuels and highly radioactive waste disposal. Irradiation studies, where CeO2 is exposed to ions with different mass and energy, are extensively taking place in the recent time. The attempt of these studies is to replicate the effect of radiation damage by fission fragments that is taking place in UO2 based fuels. X-ray photoelectron spectroscopy (XPS) proved to be an effective tool for determination of the cerium ionic composition (Ce3+ and Ce4+).

XPS determination of the cerium oxidation state in compounds faces difficulties due to the complex structure in the valence- and core- electron spectra. Therefore, this work employed an original technique for cerium oxidation state determination on the basis of the core- and valence electron fine spectral structure parameters.

This work considers the effect of fission-energy ion irradiation on the electronic structure at the surface of bulk and thin film samples of CeO2 as a simulant for UO2 nuclear fuel. For this purpose, thin films of CeO2 on Si substrates were produced and irradiated by 92 MeV 129Xe23+ ions to a fluence of 4.8 × 1015 ions/cm2 to simulate fission damage that occurs within nuclear fuels along with

bulk CeO_2 samples. The irradiated and unirradiated samples were characterised by X-ray photoelectron spectroscopy. The as-produced samples were found to contain mostly the Ce4+ ions with a small fraction of Ce3+ ions formed on the surface in the air or under X-rays. The core-electron XPS structure of CeO_2 was associated with the complex final state with vacancies (holes) resulted from the photoemission of an inner electron. A technique of the quantitative evaluation of cerium ionic composition on the surface of the samples has been successfully applied to the obtained XPS spectra. This technique is based on the intensity of only one of the reliably identifiable high-energy peak at 916.6 eV in the Ce 3d XPS spectra. This method yielded that the surface of unirradiated thin film sample AP1 contained Ce3+ ions (AP1: 97% Ce4+ and 3% Ce3+). A 129Xe23+ (92 MeV and 4.8 × 1015 ions/cm2 fluence) irradiation was found to increase the Ce3+ content in thin film sample AP2g (87% Ce4+ and 13% Ce3+) and bulk samples AP4g (92% Ce4+ and 8% Ce3+) and AP5g (93% Ce4+ and 7% Ce3+). Concentration of Ce3+ ions was shown to grow significantly as the film thickness decreased and the film fragmented (AP3g: 29% Ce4+ and 71% Ce3+).
The work was supported by the RFBR grant № 17-03-00277a.

Poster RER / 848

Investigation of a wetland contaminated by uranium mine tailings in Central France

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The release of uranium from mine tailings may present a hazard to the environment, which is the reason for the monitoring of the relevant storage sites in many countries. Studying the behavior of released radionuclides at these sites serves to better estimate the local risk and can help to improve the understanding of the geochemistry of the involved contaminants, e.g. for the application in transport modelling.

The storage site Roffin, located in the Region of Auvergne, France, contains approximately 30 000 t of mill tailings from the adjacent processing plant of the same name, which operated from 1947 to 1956. After the shutdown of the plant, the responsible operator has remodeled the site several times over the decades, in order to meet updated environmental standards 1.

Recent gamma-ray surveys have shown elevated radiation levels alongside a creek downstream of the storage site, especially in a wetland area in some two hundred meters distance of the site. Drill cores taken in this area show uranium concentrations up to 2000 ppm in the upper 30 cm, with peak concentrations in a whitish, clayey layer with a thickness of about 5 cm at a depth of 20 cm. Besides this anomalous layer, the soil is of the histosol type, with very high contents of organic matter and mostly saturated with water. The goal of our study is to identify the involved uranium species in the solid and aqueous phases, in order to understand the influence of discharge history and geochemistry on the risk presented by this contamination.

Sequential extractions performed on the different layers of the soil following the protocol of Tessier et al. 2 indicate a majority of the uranium to be bound to soil organic matter. Yet scanning electron microscopy analysis (SEM) of the white layer shows the presence of particles containing high uranium concentrations with sizes around 10 µm. Energy dispersive X-ray spectra (EDS) of some of these particles give compositions corresponding to a specific mineral processed in the plant, which is Parsonsite [$Pb_2(UO_2)(PO_4)_2$]. Dating the soil with the C-14 of the soil organic matter and the depth profile of Cs-137 from nuclear fallout further suggests that the origin of the white layer is connected to the active period of the site. X-ray absorption spectroscopy performed on the soil shows a

variable distribution of U(IV) and U(VI) in the different layers. Porewater obtained by centrifugation contains uranium concentrations up to 1000 ppb.

Further studies aim to quantify the distribution of uranium between the different solid phases of the soil, as well as the identification of the main species in the porewater.

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Poster RER / 763

Adsorption of Uranium from Saline Lake Brine by Amidoximated Mesoporous Silica

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Amidoximated mesoporous silica was prepared by grafting and co-condensation methods in this work, respectively. The materials were characterized by FTIR, SEM, XPS and physisorption apparatus. The effects of pH, adsorption time, initial uranium concentration, adsorbent amount and coexisting ions on uranium adsorption by amidoximated mesoporous silica were investigated. The experiment results showed that the uranium adsorption ratio and capacity were improved after modification by amidoxime group. It can be deduced that uranium adsorption could be attributed to the coordination of N atoms in oximido- and amido- in amidoximated mesoporous silica. The uranium adsorption from saline lake brine on amidoximated mesoporous silica were studied. The results showed that the uranium adsorption capacity could reach to 3.5 mg/g with amidoximated mesoporous silica, which was prepared by co-condensation method. Amidoximated mesoporous silica can be a potential adsorbent for uranium extraction from saline lake brine.

Poster RER / 772

On the radioecological issues of natural radionuclides in water and sediment of a highly contaminated lagoon from Mexico

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Some aquatic ecosystems from Mexico, such as rivers and lagoons, are affected by wastes enriched with potentially toxic elements generated by precious metal mining activities. Contamination of water, sediment and affected biota has produced several environmental issues, including biological effects in some species of flora. On the other hand, mineral processing activities may also generate effluents containing high activity concentration levels of natural radionuclides and, in this case, the wastes may be considered as technologically enhanced naturally occurring radioactive materials (TENORM). The release of TENORM in the aquatic environment may represent a radiological and ecological contamination issue. Therefore, the need for monitoring and assessing the radioecological

impact of mining activities has been increased. In this study, measurements of the activity concentration levels of natural radionuclides in water and sediment from a continental lagoon in Central Mexico are performed. Methods include the use of alpha-particle spectrometry with passivated implanted planar silicon detectors for the analysis of uranium and polonium in water. For isolation of U and Po of water samples, a radiochemical process based in co-precipitation with iron hydroxide and liquid-liquid solvent extraction using tributyl phosphate was employed. High-resolution gamma-ray spectrometry with hyper-pure germanium (HPGe) detectors was used to analyse gamma emitter radionuclides in sediment samples. Preliminary results show that levels of uranium isotopes in surface water are 1-5 orders of magnitude higher than reported levels for drinking water from North America. Because water from the suited lagoon is used for irrigation of nearby crops, a discussion of the radioecological implications of the obtained results is presented.

Poster RER / 474

Radionuclides and stable metals in drainage from old uranium mines

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An investigation was carried out on the acid drainage (pH 3.96) of an old uranium mine (Quinta do Bispo mine, Portugal), containing activity concentrations of 61000±7300 mBq/L of 238U, 886±60 mBq/L of 226Ra and 504±27 mBq/L of 210Po, and relatively high mass concentrations of Ni, Al, Fe, Mn, and Zn. This mine water is treated with addition of BaCl2 and pH increase with addition of hydroxide, in order to co-precipitate radionuclides with barium while the overlying water is released into surface streams. This overlaying water still contained 8740±747 mBq/L of 238U, 250±22 mBq/L of 226Ra and 30±1 mBq/L of 210Po, with radionuclide activities remaining in the treated water ranging from 6% to 26% of initial concentrations. Stable elements present in mine water partly remained in treated water at 3% to 76% of their initial concentrations. The sludge from water treatment contained thus most of radionuclides and metals removed from mine water by co precipitation. This sludge, after sun drying and curing for months at the disposal pond, was leached with water and large fractions of contaminants could be re dissolved. Barium, used in the water treatment, was the only element in concentrations higher in treated water and sludge elutriates than in the original mine water. Results indicate that current treatment of uranium mine drainage with barium is reasonably effective in reducing concentrations of radioelements and stable metals, but treated mine water still contains significant amounts of contaminants. Disposal of sludge in uncovered landfills may also originate leachates and surface runoff toxic to aquatic fauna. Therefore, improved treatment of mine drainage is needed to remove contaminants to much lower levels and abate environmental contamination.

Keywords: uranium mines, acid mine drainage, radionuclides, mine water treatment

Poster RER / 839

Determination of Presence of Artificial Nuclides in Air using NuRMS EGS Air Sampler and WIMP 120 Gross Alpha/Beta Counter

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The determination of the presence of artificial radionuclides in the air is mainly accomplished by using gamma spectrometry analysis of filter samples. This work suggests very fast, reliable and efficient technique for tentative quantitative determination of the activity of artificial nuclides such as transuranic elements, strontium, yttrium, etc. in the air based only on the gross alpha and beta volume activity. The main aspect of this technique is to provide information about approximate results for the gross alpha and beta activity of air filters, thus it is convenient as triage technique. Unlike gamma spectrometry, this method is not qualitative but allows preliminary estimation of the presence of non-natural radionuclides. The glass microfiber filters of porosity of 2.7 μm and diameter of 110 mm were used for sampling the air. The influence of increment of the dust layer on self-absorption of alpha/beta particles was also studied. This method is particularly suitable for screening situations where radionuclide identification is not necessary.

Poster RCH / 607

The XPS structure and the peculiarities of the chemical bond nature in CeO2

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Cerium dioxide is known as a non-radioactive structural substitute of actinide oxides (UO2 and PuO2). CeO2-based ceramics is suggested as an inert 239Pu or 235U bearing matrix for nuclear fuel, as well as a matrix for high-level waste disposal. CeO2 is also used an exhaust gas afterburning catalysts and in electronics.
X-ray photoelectron spectral structure of CeO2 valence electrons in the binding energy range 0–~50 eV was analyzed. The core-electron spectral structure parameters and relativistic discrete-variational calculation results for the CeO812- (D4h) cluster reflecting cerium close environment were taken into account. Comparison of the valence and the core-electron spectral structures showed that formation of the inner (IVMO) and the outer (OVMO) valence molecular orbitals contributes to the spectral structure more than the many-body processes. The Ce 4f electrons were established to participate directly in the chemical bond formation in CeO2 loosing partially their f nature. They were found to be localized mostly within the outer valence band. The Ce 5p atomic orbitals were shown to participate in the formation of both the inner and the outer valence molecular orbitals. A most of part in the IVMO formation is taken by the filled Ce 5p1/2,3/2 and O 2s atomic shells, while the Ce 5s electrons participate weakly in the chemical bond formation. The composition and the sequent order of the molecular orbitals in the binding energy range 0–~50 eV was established. A quantitative scheme of the molecular orbitals for CeO2 was built. This scheme is fundamental also for the interpretation of other X-ray spectra of CeO2. Evaluations yielded that the IVMO electrons weaken by 37 % the chemical bond formed by the OVMO electrons.
The work was supported by the RFBR grant № 17-03-00277a.

Poster RCH / 643

Electron Beam Irradiated Carboxymethyl Cellulose Hydrogel Containing Silver Nanoparticle/Activated Carbon Composites for Fast Wound Healing

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In the present study, the carboxymethyl cellulose (CMC) hydrogel containing activated carbon-silver nanoparticles (Ag NPs) composites (Acc-Ag composites) was fabricated for the highly functional wound-dressings by electron beam irradiation. For this, the Acc-Ag composites were first prepared using a thermal treatment under an inert argon (Ar) atmosphere. During the thermal treatment, the Ag ions were successfully changed to metallic Ag NPs, and NO3 anion with a potential cytotoxicity was simultaneously eliminated. Thereafter, the Acc-Ag composites were mixed with an aqueous CMC solution, and then the hydrogel was prepared by electron beam irradiation. The optimum manufacturing condition of the CMC hydrogel containing the Acc-Ag composites was examined with a variation of composite/hydrogel ratio and irradiation dose. The various physical/chemical properties of CMC composite hydrogel were characterized. The resulting CMC composite hydrogel was examined via wound regeneration test. Therefore, the CMC hydrogel containing the Acc-Ag composites was found to have a great potential for functional wound dressings.

Poster RER / 592

129I records of nuclear activities in the East China Sea inner shelf

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Abstract:
Anthropogenic 129I on surface environment mainly originates from three sources, including nuclear weapon tests, nuclear fuel reprocessing plants, nuclear accidents, with different pathway. This leads to 129I deposition history recorded in sediment is not the same in different regions. Due to its long half-life and high conservative feature in the ocean, 129I has been used as an effective environmental and oceanographic tracer. Due to the same production ways and properties with the short-lived and high radiation hazard 131I, 129I also can be applied to evaluate the radioactive influence of early nuclear activities [1,2,3].
With the rapidly increased numbers of nuclear power plants constructed along the China coast, the environmental radioactive impact in East coastal area of China has raised high concern. The impact of other nuclear activities such as the nuclear weapons tests Lop Nor, Fukushima accident and nuclear reprocessing plants are also concerned. To elucidate these issues, we analyzed two sediment cores collected in East China Sea, covering the periods of 1959-2011 and 1960-2011, respectively, to obtain a temporal fallout of 129I in this region.
The results show that 127I concentrations range 5.0-42.5 μg/g, 129I concentrations 0.05-1.2×107 atoms/g, and the 129I/127I ratios were 15.0-66.0×10-12. The 129I level are similar to that obtained in the similar latitudes elsewhere. Significantly enhanced 129I level than pre-nuclear (1.5×10-12 for

129I/127I ratio and 0.043×107 atoms/L for 129I concentration) was observed in these two sediment cores, with similar peak values in the layer corresponding to 1965-1967, 1970-1973, 1975-1977, 1980-1983. According to the peak time coincidence with Lop Nor NWT, the source of 129I before 1980s was regarded from Lop Nor NWT, but less related to Semipalatinsk NWT. The transportation mechanism of radioactive substance from Lop Nor was simulated on basis of the local general circulation of atmosphere and the transmission characteristics of radioactive materials from NWT [4,5,6]. It shows that the radioactive material could reach ECS (East China Sea) through the direct atmospheric dispersion. Meanwhile, the regional deposition of Lop Nor turned into soil in the Jinsha River Valley area that is one of the most important branches of Yangtze River. And the 129I can be continuously leached out by rain and entered to YDW (Yangtze River Diluted Water) in ECS within Yangtze River input [7]. Finally, 129I was further transferred to sediment by the biochemical processes [8]. The 129I signal in the sediment core after 1980s was mainly from European nuclear fuel reprocessing plants at La Hague and Sellafield by the direct atmospheric release and the re-emission of marine discharges in the highly contaminated seas [9]. In comparison, the Fukushima nuclear accident had no detectable effect on the ECS until September 2011.

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Poster RER / 618

Natural and anthropogenic radionuclides in Barra de Valizas – Aguas Dulces area, 290 RAMSAR site, Uruguay

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Barra de Valizas –Aguas Dulces area is placed in the strip coast of the 290 Ramsar site, located in the eastern region of Uruguay, on its Atlantic coast. The studied area included two coastal towns and a freshwater lagoon.
The activity concentration of 238-U, 226-Ra, 210-Pb, 232-Th, 40-K and 137-Cs radionuclides in sand, soil and *Baccharis articulata* were evaluated. For the quantification of those radionuclides, 2 kg sand and soil of 20 cm from the top surface layer, and the hole plant of Baccharis articulata placed in the same soil were collected. Each sample was dried at 60°C, vegetation sample was milled and all samples were sealed into 100 mL flasks. After approximately four weeks in order to reach secular equilibrium of the 238-U and 232-Th series, samples were measured by gamma-spectrometry with a High Pure Germanium Detector GMX35P4-76-RB, 35 % efficiency and 1,75 % energy resolution for the 60-Co photopeak. IAEA reference materials were used for efficiency calibration, and self-absorption correction was applied for 238-U and 210-Pb determinations. 238-U was studied by the

234-Th photopeak (63.3 keV), 226-Ra by the 214-Bi photopeak (609.3 keV) and 232-Th by the 228-Ac photopeak (911.1 keV). 40-K (1460.0 keV) and 210-Pb (46.5 keV) were evaluated by their own photopeaks, and 137-Cs radionuclide by the 137m-Ba (661.7 keV) photopeak.

The activity concentration for 238-U, 226-Ra and 210-Pb in soil were found to be 14.3 –19.5, 8.69 – 13.21, 16.60 - 32.3 Bq/kg, respectively, whereas the activity concentration for 238-U, 226-Ra and 210-Pb in sand were 10.9 –35.8, 7.53 –26.19, 11.1 –23.9 32.3 Bq/kg, respectively. Sand and soil samples show high concentration of 238-U than 226-Ra, the 226-Ra/238-U activity ratio was in the range 0.51 –0.83, therefore there is a low radioactive equilibrium disturbance, because of selective leaching of radium. The 210-Pb/226-Ra activity ratio was found to be in the range 0.91 –3.70, the equilibrium disturbance is higher in soils than in sands samples, showing an amount of unsupported 210-Pb, which increased with organic matter.

The activity concentrations for 232-Th, 40-K and 137-Cs in soil were 21.4 –45.6, 186.6 –307.6, 0.12 –1.00 Bq/kg, respectively. The activity concentration for 232Th in sand is higher than in soil, 39.8-749 Bq/kg, whereas the activity concentration of 40-K is lower: 120.0- 160.3 Bq/kg. The activity concentration of 137-Cs in sand samples is below the minimum detectable activity (MDA).

The mean activity concentrations for 210-Pb, 40-K and 137-Cs in *Baccharis articulata* were 58.7 + 6.1, 238 + 11, 1.82 + 0.65 Bq/kg, respectively. 238-U, 226-Ra, and 232-Th were below the MDA.

The transfer factors for 210-Pb, 40-K and 137-Cs were, 2.33 + 0.47, 1.209 + 0.086 and 1.57 + 0.73 respectiely.

The area shows higher 232-Th activity concentration than worldwide mean for both soil and sand, whereas 226-Ra activity concentration is only higher than worldwide mean for Aguas Dulces´ sands.

The 238U series shows disequilibrium for both sand and soil, with high concentration of 238-U and 210-Pb.

210-Pb, 40-K and 137-Cs transfer from soil to vegetation, although more determinations must be made in order to know the 210-Pb fallout intake.

Poster RER / 626

Neptunium speciation and accumulation by soil components and biota

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Artificial radionuclides and, first of all, the most dangerous isotopes of transuranium elements(TUE) became constant and irreversible components of the biosphere because of the experimental nuclear explosions performed, the wrong concept of the disposal of radioactive wastes to open water reservoirs, and also process upsets and emergency situations at NFC enterprises .

It is known that many radionuclides can accumulate by biogeocenosis components and incorporate into food chains. However, only the behavior of gamma-emitting short-lived isotopes has been studied comprehensively at present. Bioaccumulation of alpha-emitting and the most toxic isotopes of actinide elements have been studied less due to methodological difficulties of radiochemical analysis. However, laboratory researches show a high degree of actinide sorption, primarily of neptunium, by microorganisms and plants.

The main problem of determining the coefficients of actinide accumulation by biota is the impossibility of direct detection of their content and the significant influence of the organic substance on the yield of the analyte during alpha-spectrometric determination. A method for determining actinides according to the luminescence of crystallophosphors was developed and tested in the Vernadsky Institute of Geochemistry and Analytic Chemistry of the Russian Academy of Sciences. A photometer and membrane modules for the separation of actinides in physicochemical forms, as well as for their concentration, were created. The method is particularly useful when analyzing natural objects, because it doesn't require prior separation of actinide elements, as well as cleaning the sample from alkali and alkali-earth elements. Actinide luminescence is induced by the ultraviolet radiation of the crystallophosphor located at room temperature. Detection limit of the photometer for determining the neptunium contents in simulated groundwater is 3 pg/ml(0.3 pg of Np in a quarts crucible).

The present research studies the processes of bioaccumulation and biotransformation (changing of

speciation) of the most toxic, alpha-emitting actinides on real biological objects, selected in the areas of Krasnoyarsk Mining and Chemical Combine (KMCC), and in the impact zone of the Kraton-3 underground nuclear explosion (Yakutia).
It has been established that the relative content of water-soluble and exchange forms of neptunium is 5-10 times higher than the relevant values for plutonium and americium. Among long-lived radionuclides only 90Sr has similar solubility. It was found that, for all of the studied soil types, the concentration of radionuclides in water-soluble and exchange forms,which are most mobile and determine in the series 237Np(39.1-75.4%) >241Am(3.9-21.7%) > 239Pu(4.1-
20.1%). In poorly soluble forms (acid-soluble and residues) irrespectively of the soil type, the Concentration of radionuclides changed in the inverse order : 239Pu(68.4-85.7%)> 241Am(19.6-
-36.2%) >237 Np (9.7-31.8%)
In the soil organic matter the main content of neptunium was found in the fulvic acids fraction and low molecular weight acids (below 43%), and moreover, up to 25% of neptunium is directly bonded to the low molecular weight substances of non-specific nature. This, apparently, provides its high bioavailability.
Neptunium is found in the aquatic vegetation of the Yenisei River. The water plant Fontinalis antipyretica (water moss), in the dry biomass of which the maximum specific activity of neptunium amounts up to 10 Bq/kg, accumulates it most actively.
The content of neptunium in the vegetation in the impact zone of the Kraton-3 underground nuclear explosion increases in the line: lingonberry (Vaccinium vitis-idaea) < mountain ash (Sorbus aucuparia) < sedge (Carex riparia) < lichen (Hypogymnia physodes) < larch needles (Larix sibirica) < moss (Cladonia rangiferina).

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Polonium 210Po and radiolead 210Pb in forest mushrooms of family Boletaceae from Poland and China and its contribution to the effective radiation dose

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This study aimed to assess potential radiotoxicity to human consumers from 210Po and 210Pb accumulated in several species of mushrooms from the family Boletaceae that are traditionally collected in Poland and China. Fruitbodies of B. edulis were from the Yunnan province of China and from the northern part of Poland, and individuals of pine bolete Boletus pinophilus, summer cep Boletus reticulatus, iodine bolete Boletus impolitus and lurid bolete Suillelus luridus were from Poland.
Mushrooms typically grow in forests and fields, but almost all ecosystems will favor their growth in the correct substrate medium. They are considered as organisms that well bio-concentrate in fruitbodies certain mineral constituents absorbed by mycelium e.g. heavy metals or radiocaesium. Hence, to some degree mushrooms can be useful as indicators aiming to evaluate degree of soil pollution, while fruitbodies of edible species when loaded with heavy metals may pose a risk for consumer.
Boletus edulis collected near Ninger in the Puer county in Yunnan exhibited greatest activity concentration of 210Po, i.e. 53±1 Bq kg-1 dry biomass, while B. reticulatus of 210Pb, i.e. 15±1 Bq·kg-1 dry biomass.
The data showed, Chinese mushrooms contained significantly more 210Po and 210Pb when compared to Polish king bolete (B. edulis). Also the results indicated, in both cases of Chinese and Polish Boletaceae, the values of 210Pb activity concentrations were lower than 210Po.
In order to identify the potential radiotoxicity, on the basis of previously calculated 210Po and 210Pb content in dried and unprocessed culinary fruiting bodies of analyzed mushrooms, the effective radiation doses were calculated, using the effective dose conversion coefficients from analyzed radioisotopes ingestion for adult members of the public recommended by ICRP. The results showed

the consumption of 1 kg of whole dried mushrooms would give the effective radiation doses at 1.04-63.4 microSv from 210Po and 0.70-10.02 microSv from 210Pb, but higher values of the effective dose would be received from Chinese mushrooms ingestion. However, the results mean if consumers would eat the analyzed mushrooms, they should not increase significantly the total effective radiation dose from 210Po and 210Pb when compared to other sources of 210Po and 210Pb from typical diet.

Poster RER / 487

Po-210 in marine flora and fauna from an area affected by NORM effluents and atmospheric emissions

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Polonium is a radioactive element present in many components of the marine and terrestrial environment. ^{210}Po, the most abundant naturally occurring radioisotope of polonium, is one of the descendants of the uranium (^{238}U) decay series. Rocks, soil, water, sediment, and biota, contain traces of ^{210}Po in variable amounts. ^{210}Po may be radiotoxic to humans and non-human biota if high levels of this radionuclide are detected in the food chain. Polonium may be bio-accumulated by marine organisms, depending on the biogeochemical characteristics of the environment but also due to the contribution derived from human activities. Certain industrial activities generate naturally occurring radioactive materials (NORM); industrial residues and by-products containing significant amounts of polonium and other natural radionuclides are considered NORM. For example, some effluents and atmospheric emissions associated with the production and processing of oil and gas, introduce variable, but sometimes, elevated levels of natural radionuclides to the near environment. Monitoring and evaluation of the activity concentration levels of polonium in ecosystems affected by the discharge of NORM residues are needed. In this contribution, a survey of the levels of ^{210}Po measured in different species of flora and fauna around an oil and gas facility in the southern Gulf of Mexico is presented. Alpha-particle spectrometry with passivated implanted planar silicon (PIPS) detectors was used for radiochemical analysis of polonium. The obtained results show that ^{210}Po is been bio-accumulated by flora (algae, seagrass, and mangrove) and fauna (molluscs, crustacean, and fish) from the studied region. An assessment of the ^{210}Po levels in the different flora and fauna analysed samples is performed.

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Poster RER / 495

Is human hair a proper 210Po and 210Pb monitor of their increased natural activity in human body?

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Polonium 210Po and radiolead 210Pb belong to the uranium series decay chain with half-lives of 138.376 days and 22.2 years respectively. They are relatively toxic to humans, especially α-emitting 210Po, both chemically and radiologically, and are accumulated to a considerable extent. The main sources of chemical elements intake are air and food but their intensities depend on living place, level of local contamination, also radiochemical, diet habits and food origin. Also cigarettes are significant source of 210Po and 210Pb.

This study aimed to determine the activity concentration of polonium 210Po and radiolead 210Pb in human hair as well as investigate the utility of human hair as a non-invasive indicator of metal exposure for radiotoxic, naturally occurring 210Po and 210Pb, and find differences in their concentrations in hair considering the age, gender, hair color or diet of people who donated the samples. The results ranged from 0.10±0.01 to 12.8±0.80 Bq⌧210Po ⌧kg-1 dw and from 0.44±0.04 to 52.9±4.45 Bq⌧210Pb⌧kg-1 dw. Statistical analysis showed significant differences within age, hair color and cigarette smoking groups. The data show hair could be used as an indicator of 210Po and 210Pb exposure in occupational and environmental surveys for various populations, although more detailed knowledge on analyzed objects should be taken.

Poster RER / 515

Advancements in the measurement of 137Cs, 134Cs and 90Sr in seawater

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On March 11, 2011, the Tohoku earthquake and the subsequent series of tsunamis severely damaged the Fukushima Dai-ichi nuclear power plants (FDNPPs). Failure of cooling systems led to explosions releasing radioactive gas and debris to the atmosphere along with cooling water that was directly discharged to the sea after being in contact with the nuclear fuel. The resulting release of 90Sr (T1/2 = 28.9 years), 137Cs (T1/2 = 30.17 years), and 134Cs (T1/2 = 2.07 years) was the largest ever uncontrolled input of artificial radionuclides into the ocean. This release increased pre-Fukushima concentrations in surface waters off the coast of the FDNPP that were on average 1 Bq·m−3 for 90Sr and 1−2Bq·m−3 for 137Cs by factors 1000 or more in 2011. Presented are methods for 90Sr and 137Cs, where 90Sr is analyzed in 20 liter seawater samples that have passed through a Cs absorber. Samples were spiked with stable 133Cs and processed through 5 ml of a wetted composite inorganic absorber of potassium-nickel hexacyanoferrate(II) (KNiFC) in a binding matrix of modified polyacrylonitrile (PAN). Gamma spectrometric measurements of 134Cs and 137Cs were performed using a Canberra high purity germanium well detector (GCW4030S) integrated with a cosmic guard and processed through a Lynx 32K channel integrated multi-channel analyzer. Calculated MDA's for 134Cs and 137Cs are 0.4 mBq/kg and 0.2 mBq/kg respectively. Stable 133Cs in initial and final aliquots was analyzed on an Element 2 ICP-MS Thermo Fisher Scientific. Average recoveries were 98.62% and corrections were applied to measured Cs activities. 90Sr activity was determined via separation chemistry of its 90Y daughter. Samples spiked with stable Yttrium underwent iron hydroxide co-precipitation and purification using a DGA resin. 90Y beta decay was measured using a GM-25-5 Beta Multicounter by Risø National Laboratory. Average recovery determined by ICP-MS analysis of stable Yttrium was 81.5% and a MDA for 90Sr = 0.4 mBq. The application of Fukushima isotope ratios of 137Cs and 90Sr can be applied as longer term Fukushima water mass tracers, which include tracing North Pacific circulation and mixing.

Poster RER / 544

Method development for the determination of uranium isotope ratios by MC-ICP-MS

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Accurately determined uranium isotope ratios are essential for geochronological dating and tracing of different environmental processes. In addition, they are of paramount importance for nuclear safeguards and nuclear forensics.
Multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) has become an important tool for determination of uranium isotope ratios in both liquid and solid samples. Its features are better accuracy and precision compared to alpha spectrometry, which is comparable to thermal ionization mass spectrometry while maintaining higher sample throughput. This makes it an ideal tool for many applications.
The objective of this study was to develop, validate and verify accurate analytical method for the determination of uranium isotope ratios (235U/238U and 234U/238U) with correctly estimated measurement uncertainty. Measurements were carried out with Nu plasma II, (Nu instruments Ltd, UK) MC-ICP-MS with the high-efficiency sample introduction system Aridus IITM (Cetac Technologies, NE, USA). At first, routine optimization and calibration of the MC-ICP-MS was performed. Then the appropriate aliquot concentration of purified uranium fractions was selected from several different concentrations of a uranium standard of known isotopic composition. Instrument mass bias was corrected with external standard –sample –standard bracketing technique.
The developed method was applied for analysis of uranium isotope ratios in uranium fractionations obtained from water samples of karstic aquifer (Ljubljanica catchment). After pre-concentration, uranium precipitate was further purified on UTEVA column and then the organic composition of resin was destroyed by using concentrate HNO3 and H2O2. Before measuring the proper aliquot of uranium isotope ratio on MC-ICP-MS, the exact concentration of uranium in sample was determined by two different methods, by radiochemical neutron activation analysis (RNAA) and by inductively coupled plasma mass spectrometry (ICP MS). The comparison of this two different methods was also performed.

Poster RER / 551

Detection of 2017 ruthenium-106 fallout in grass in Northern Czechia

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Traces of radioactive isotopes of ruthenium in the atmosphere were reported in the beginning of October 2017 by several European stations monitoring the airborne concentrations of gamma emitters (e.g., IRSN, 2017). As of February 2018, the source in Eastern Europe has not been publically identified. ^{106}Ru is a fission product with a half-life of 371.5 days. It is used as a medical isotope and despite its relatively short half-life, also its use as radionuclide thermoelectric generator was suggested (IRSN, 2017). ^{106}Ru values, up to dozens of mBq•m^{-3} have been detected in Czech stations (SURO, 2017), where also the isotope ^{103}Ru was detected in levels 3–4 orders of magnitude lower.

In October 2017 we collected grass samples from four stations in Liberec region in northern Czechia. The samples were taken in order to test, if radioisotopes related to wet deposition during the period of positive ^{106}Ru atmospheric detection, can be measured in detectable amounts and whether a deposition ratio could be derived. Grass is an environmental medium, in which radionuclide fallout would be concentrated after emission followed by wet deposition. Grass is at the same time an

important part of the terrestrial food chain. For both reasons, grass is one of the first environmental media to be investigated in emergency plans. Our grass samples contained ^{106}Ru detectable by standard gamma spectrometric procedures in the range of hundreds of mBq•m^{-2}. The observed activities of ruthenium and natural radionuclides were evaluated using a simple deposition model in context of the reported airborne activities and meteorological conditions.

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Poster RER / 661

210Po distribution in different compartments of the Briozzo Lagoon

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210Po is a natural radionuclide, which is present in the environment as a result of the decay of 238U and is characterized by high radiotoxicity and bioaccumulative behavior in certain tissues of living beings.
The main source of 210Po explaining its presence in different compartments of the environment is the exhalation of 222Rn from the ground and its subsequent decay in the atmosphere, resulting in 210Po deposition on earth surface through fall out and rain out.

In shallow coastal lagoons, although the main contribution of 210Po to water and sediments is directly from the atmosphere, rivers and runoff can be additional sources to the water column. In the lagoons, aquatic organisms are capable of concentrating within their tissues various toxic elements including radionuclides, although the concentration of most of these elements or radionuclides in the medium occur generally at trace levels.

The Briozzo lagoon, is a natural freshwater shallow lake, located on the east coast of Uruguay, without anthropic influences and an average depth of 3 meters. In that area of Uruguay, several black sand deposits are present, which have radioactive activity due to their content of thorium and uranium. In order to study the distribution of 210Po in the different compartments of this ecosystem, samples of water, sediment, clams (Diplodon sp), freshwaters snails (Pomacea caniculata) and different species of aquatic plants (Luciola peruviana, Salvinia sp., Schoenoplectus californicus), were collected.

The activity concentrations of 210Po in all the samples analyzed have been determined by applying the high-resolution alpha-particle spectrometric technique. In particular, an alpha-particle spectrometric system, Alpha-Analyst from Canberra Co., formed by a total of eight independent chambers working in parallel, each one equipped with a PIPS type silicon detector (450 mm2 active area), has been employed, being reached typical minimum detectable activities in the order of 10−1 mBq. The application of this technique implies the previous isolation and deposition in thin layers of the radioelement of interest in order to avoid interferences in the measurements.

In addition to the activity concentrations of 210Po determined in the different environmental matrices, the corresponding transfer or concentration ratios were calculated. In particular, high bioaccumulation of 210Po in the analysed aquatic organisms was inferred.

Poster RER / 663

A study of quantitative analysis of Korean granite using ICP-AES/MS/XRF

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Geological reference material is essential for analytical research and has been widely used for a chemical analysis to obtain accurate geochemical data. Also, it is useful to evaluate the accuracy and precision of the analysis, to develop and improve analytical techniques. Although there is a trend that demand for these geological reference materials is increasing but development of Korean geological reference materials has been ceased since the 1990s. In this study, therefore, as a preliminary part of research for development of Korean geological reference materials, we selected granites that can represent Korean rocks and determined the concentration of major/minor/trace elements of granite sample using various analytical instruments. The major elements of the granites were obtained by XRF analysis, and concentration of minor/trace elements were measured using ICP-MS/AES. For the selection of a candidate of Korean reference materials, following options should be satisfied; represent the Korean rocks, chemical compositions are homogeneous, not include structure like fissure or fold, fresh rock is available in a large volume. Based on these options, we selected two Jurassic granites and one Cretaceous granite which are representing Korean rocks. XRF (Bruker, S8 Tiger) was performed at the Center for Research Facilities of Gyeongsang National University. X-ray fluorescence (XRF) analysis has the problem of volatilization of volatile elements like alkali metal, Pb, Cs and Zn during melting at high temperature when glass bead produced. In order to minimize these problems, glass beads were prepared at 1020℃ for 10min. In addition, ICP analysis was carried out at the Korea Basic Science Institute in Ochang. Inductively coupled plasma mass spectroscopy/atomic emission spectroscopy (ICP-MS/AES) are most widely used analytical techniques to determine the concentration of major and trace elements in the rock samples. The complete dissolution of rock samples is, however, difficult when they contain refractory minerals such as Zr, Hf, and REEs. Generally, in the case of a granite sample, there are methods for complete dissolve including acid digestion, glass bead digestion, microwave assisted procedure, high-pressure bomb procedure. In this study, we digested three granites using mixed acid (HNO3:HF:HClO4=4:4:1), aqua resin, 1% v/v HNO3 for complete dissolution of granite samples. Precision and accuracy of the analytical techniques are evaluated by using reference materials supplied by USGS and GSJ and then we calculated concentration of chemical composition of granite rocks. Based on these results, the possibility of development as a Korean reference material will be examined by comparing the results of analysis carried out by other analytical institutions and determined the reference value. We expected that these reference materials are to be utilized in various field such as research institutes like a universities and laboratories, as well as in analytical institute supporting analysis study.

Poster RER / 666

Retrospective measurement of U-236 in human lungs

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Human lung ashes were collected in Vienna, Austria, during the early sixties of the last century in order to determine short-lived fission products from the nuclear test explosions by gamma-spectrometry (1). Later on in part of these samples also plutonium was determined by alpha-spectrometry (2).
Last year we started to investigate some of the remaining samples with regard to 236U (half- life 2.3·107 years) which is produced via 238U(n,3n) by fast neutrons and by thermal neutron capture on 235U; the cross section for the latter reaction is about 1/6 of the 235U fission cross section.
The ash-samples were dissolved in half-concentrated nitric acid, a certain amount of 233U was added as a spike and uranium was then separated by anion exchange (UTEVA) and co-precipitated with Fe(OH)3. After calcination the samples were measured by accelerator mass spectrometry (AMS) at the VERA laboratory. The derived 236U/238U isotopic values were in the range of 10-8, as expected for global-fallout samples, corresponding to 107 atoms 236U per g of lung-ash. Although the values for individual samples from the same year scatter rather strongly, the calculated mean values clearly display the well-known bomb-peak. The new results will be discussed in connection with the old published data as well as with unpublished 137Cs data.

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Poster RER / 715

The DESTRUCT software for calculation of barriers evolution parameters

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Forecast of engineering barriers evolution is an important problem of radioecology.
The goal of the following study is the development of complex methodology of modeling different processes, occurring during evolution of multibarrier system, and creating a uniform approach to safety analysis.
Methods of the description of the major factors, important for modeling multibarrier system (thermal emission, illitization of bentonite buffer, degradation of concrete and glass matrices, corrosion of a metalwork, processes on barrier/barrier border) are developed.
Algorithms of sodium phosphate glass matrix leaching modeling, corrosion of steel, degradation of concrete, bentonite evolution at a chloritization and an illitization are developed. The described algorithms are implemented in the form of templates for calculation of chemical processes at specific conditions of barriers evolution.
The basic structure of the main modules of DESTRUCT software is created. It consists of blocks of data processing, modeling of evolution processes of safety barriers, modeling of radionuclides migration in a near field, postprocessings of calculation results, interactions between the blocks providing prognosis of dynamics of parameters changes of materials.
The developed models are tested on the example of calculation of processes of barriers degradation and radionuclides transport for a a prospective Russian radioactive waste disposal site.

Results of leaching calculation from various glass matrices, calculations of corrosion rates in anaerobic conditions in a bentonite environment depending on temperature and acidity of the environment, changes of of mineral phases array in concrete are presented in this study
Application of DESTRUCT has allowed to estimate rate of steel corrosion in. The illitization is proved to be neglectible at predicted temperature fields.
As a result of test of DESTRUCT complex a number of problems of various barriers evolution assessment and radionuclide transport are solved. Influence of physical and chemical processes on evolution of parameters of steel corrosion and evolution of concrete barriers, bentonite swelling, evolution of glass matrices and also radionuclide transport through the multibarrier system for a prospective radioactive waste disposal is estimated.
Tests have shown efficiency of the applied approaches to assessment of evolution of multibarrier system. At the same time need for development and updating of chemical models of processes is confirmed.

Poster RCH / 820

European Tools and Methodologies for an Efficient Ageing Management of NPP Cables

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TeaM Cables is a European collaborative Research and Innovation project focusing on nuclear cable ageing.It has received 4.2 M€ of funding from the Horizon 2020 Euratom Research and Training Programme of the European Union. TeaM Cables started in September 2017 for a duration of 4.5 years.
TeaM Cables aims at providing NPP operators with a novel methodology for efficient and reliable NPP cable ageing management by:
1. developing cable ageing models and algorithms, which are based on multi-scale studies and can be tailored to cover variations in fillers, additives and degrees of crosslinking,
2. developing methodologies for non-destructive testing techniques and their associated criteria identified from multiscale relations,
3. developing a novel "open access"tool, the "TeaM Cables tool", integrating all the models developed and providing the residual lifetime of cables by crossing non-destructive measurements with predictive models and knowledge of cable exposure conditions (wiring network in the NPP).
TeaM Cables methods and tools will allow NPP operators to safely extend the plant life duration of generation II and III reactors and thus contribute to the production of sustainable energy responding to future energy needs.

Poster RCH / 841

Radiolysis of ammonium succinate in aqueous solution: Relevance in chemical evolution.

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Chemical evolution encompasses the formation of biologically relevant compounds started from simple inorganic molecules and promoted by an energy input. Among the possible energy sources for these processes is radioactivity. This research is done in this framework, and it is aimed to study the effect of ionizing radiation on oxygen-free, aqueous solutions of ammonium succinate. Particularity is concerned with the stability this compound in an aqueous medium and the presence of high radiation fields and comparing it with the behavior of succinic acid. Succinic acid and its ammonium salt are molecules formed readily in prebiotic experiments and are essential in chemical evolution processes, as participants in metabolic pathways, and as starting material for more complex molecules. Several products were detected, including some containing-N compounds and the formation can be explained via the attack of the radiolytic products of water. The abstraction and dimerization reactions were the more important. The decomposition increased with the irradiation dose given rise to biologically relevant compounds.
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Poster RER / 875

Natural radionuclides in acid mining drainage waters after a not controlled discharge from a pit lake in the south of Spain

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Poster RER / 876

210Po distribution in different compartments of the Lagoon of Briozzo

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Poster TAN / 484

Adsorption of the nihonium species on gold surface: a relativistic density functional theory study

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The discovery of long-lived isotopes of superheavy elements (SHEs) in nuclear fusion reactions of ^{48}Ca with actinide targets signifies a close approach to the island of particularly shell-stabilized nuclei. While the emergence of the new elements is a fantastic discovery itself, the longevity of certain isotopes is thought-provoking for chemical investigations, both experimental and theoretical. Successful chemical identification of copernicium (element 112, Cn) and flerovium (element 114, Fl) has been recently followed by the recognition of the discovery of nihonium (element 113, Nh) confirmed at the fourth IUPAC/IUPAP Joint Working Party. Although preliminary experimental results have been recently obtained in FLNR JINR (Dubna), the chemical properties of nihonium remain of top interest. From the theoretical standpoint, investigations of the SHE chemistry are especially challenging as they require an understanding of the electronic structure in the presence of strong fields of heavy nuclei and thus governed by relativistic effects.

Gas thermochromatography on gold is a unique method of chemical detection of SHEs. The corresponding experiments, however, are extremely sophisticated and expensive and produce very scarce data on the chemical properties of SHEs. Moreover, the correct and detailed interpretation of the available experimental data cannot be performed without preliminary theoretical modeling. The main effort of our research is focused on the description of the SHEs atoms/molecules adsorption on a gold surface from first principles –a key component for quantitative theoretical predictions. The desorption energies of SHEs on a gold surface are estimated using the cluster model. To simulate the electronic structure of such systems, we combine accurate shape-consistent relativistic pseudopotentials and non-collinear two-component relativistic density functional theory. As in important result of such modeling, we recommend estimates of the Nh desorption energies on gold –110 kJ/mol.
Strong relativistic effects suggest dramatic dissimilarities in the chemical behavior of SHEs and their formal lighter homologues. The calculated adsorption energy for single atoms of nihonium on a gold surface differs substantially from the experimentally measured adsorption energy on gold of its nearest homolog, thallium. This casts doubt on the usefulness of the experiments with Nh formal homologues for understanding its chemistry. Despite manifest deviations of the chemical properties of the SHEs from the trends observed in their lighter formal homologues in the respective groups of the periodic table, finding chemical pseudo-homologues appears a practically meaningful issue.
It has been shown experimentally that the desorption temperatures and energies of Cn and Fl atoms from a gold surface are fairy close to each other and overall lower than those of their immediate homologs Hg and Pb, observed on the same surface. This confirms theoretical predictions concerning the electronic structure of Cn and Fl atoms: the strong relativistic stabilization of the s and p1/2 shells in both Cn $(6d^{10}7s^2)$ and Fl $(6d^{10}7s^27p^2_{1/2})$ results in a closed-shell character of the ground states of these atoms. Due to this unique feature of the 7^{th} row of the Periodic Table, the electronic structure of a Nh atom can be interpreted as a Fl atom with a hole in its closed $7p_{1/2}$ subshell. This observation seems to render astatine a closer chemical "relative"of Nh in comparison to the formal homologue Tl. Thus, At might be a plausible chemical species for model experiments aiming at finding the optimum experimental conditions for further explorations of the Nh chemistry. The predicted adsorption energies for At and AtOH on gold are 130±10 kJ/mol and 90±10 kJ/mol, respectively. This confirm the experimental observation on the formation of AtOH molecules in presence of trace amounts of water and oxygen in the carrier gas. Due to the similarities in the chemical properties of AtOH and NhOH molecules, one may expect, that the formation of NhOH is indeed possible under experimental conditions similar to those in the experiments on At.
The work was partially supported by the by the Russian Science Foundation grant No. 14-31-00022.

Poster TAN / 546

Sputtering and heating as factors affecting the durability of actinide targets and metallic foils in experiments on synthesis of superheavy nuclei

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Sputtering of actinide targets, Ti target backing and window foils irradiated by intense heavy ion (HI) beams in long-term experiments has been considered on the grounds of available models and experimental data. Experiments on synthesis of superheavy nuclei (SHN), which are carried out with the Dubna Gas-Filled Recoil Separator (DGFRS), are the examples of such kind of experiments. High fluxes of HIs and heat generation, which are realized within a relatively small area and thickness of these elements of DGFRS, are inherent in such experiments. At present, the {48}^Ca beam with the intensity of about 1 pμA allows obtaining several atoms of SHN per month at their production cross section of several pb and an efficiency provided by DGFRS. The detailed study of properties of SHN produced in the experiments with the use of complete fusion-evaporation reactions induced by the {48}^Ca projectile on actinide target nuclei, which lead to nuclei with 112≤Z≤118, implies the use of beam intensities significantly higher than those used in the discovery experiments 1. Moreover, synthesis of SHN with Z>118 implies the use of the beam heavier than {48}^Ca beam particles ({50}^Ti, {54}^Cr etc.). One may expect the production cross sections less than 0.05 pb for SHN formed in the

fusion-evaporation reactions with these heavier projectiles. It means that for the observation of two decay events of SHN produced with this cross section one should collect the beam dose of 10^{20} particles if the 0.4 mg/cm^{2} actinide target and total DGFRS efficiency of 40% are implied. This dose of particles passed through a stationary target may cause total disappearance of radioactive target material at the end of the experiment if the sputtering yield of the material is estimated as 10^{-2} atom/ion 2. In the case of the rotating target one can essentially reduce the yield of sputtered atoms due to the gain in the irradiation area. The question arises of whether these estimates are reliable to be taken into account in future experiments.
Heating the target and target backing foil as a single whole caused by an intense HI beam can be estimated with the use of some approximations. The temperature of the target and target backing is calculated as a function of time in the conditions of pulse heating followed by subsequent cooling with radiation emitted from their surfaces. Such pulsing mode corresponds to the rotating target irradiated by a continuous HI beam in the experiments. Estimates show that radiative cooling in the conditions of pulse heating can be the most effective way of heat transfer to the surroundings at the temperature of several hundred degrees. Such temperatures can be reached on the surfaces of the target and foils irradiated by HI beams at the intensities exceeding 10^{13} particles/s 3. Yu.Ts. Oganessian, V.K. Utyonkov, Nucl. Phys. A 944 (2015) 62.
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Poster TAN / 701

Np(VI) and Pu(VI) complex formation with malonate ions in low acid solutions

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Carboxylic acids of different composition are the most frequently encountered components of a variety of natural media. They are active complexing agents for actinide ions in higher oxidation states and sometimes, depending on actinide ion pare redox potential, can play reductant role. Taking into account that actinide elements in higher oxidation states have an increased solubility in the solutions of pH range (what is often closed to the environmental media), it is became clear the importance of the understanding of their behaviour under such conditions. In the present work we have studied interaction of Np(VI) and Pu(VI) with anion of malonic acid (CH2(COO)22- = L2-).
Synthesis of Pu(VI) solid compounds from aqueous solutions and their characterization also require special data on the behavior of Pu(VI) and Pu(V) in the presence of complexing agents. There is very limiting information on this topic for both Pu and Np. Some data were reported on complexation of the neptunyl ion with aromatic polycarboxylates, particularly, phthalate. Earlier we published new data on Np(VI) and Pu(VI) complexation with phthalate in neutral aqueous solutions. In this study we have gained additional information on the complexation and reduction of Pu(VI) and also on the disproportionation of Pu(V) in malonate solutions. For comparison, the behavior of Np(VI) was also examined.
Complexation of PuO22+ in solutions containing malonate anions was studied by spectrophotometry. Mono- and bimalonate complexes are formed. The monomalonate complex was isolated as PuO2L*3H2O. It was found to be isostructural with UO2L*3H2O. Isolated plutonium solid complex forms rhombic crystals with the unit cell parameters a= 9.078(2),b= 7.526(2), and c= 6.2005(15)Å, space group Pmn21. The electronic absorption spectrum of the monomalonate complex is characterized by a strong band at 843nm. In malonate solutions, Pu(VI) is slowly reduced to the pentavalent state even in the cold.
In contrast to Pu(VI), Np(VI) is noticeably reduced by the malonate ions even at room temperature, making it possible to examine the kinetic features of the reaction An(VI) An(V). In this work we have performed a series of experiments to examine the effect of the temperature, Np concentration, and solution composition on the reaction kinetics.Kinetic curves illustrating the general picture of the temperature dependence of the process of neptunium reduction are measured. Calculations have been performed showing that they (kinetic curves) are not described by first- or second-order reaction equations. A mechanism of neptunium(VI) reduction is proposed, the rate-determining stage of

which is the intramolecular reduction of neptunium (VI) with the formation of the monomalonate complex Np (V) and the radical ion. The proposed mechanism is confirmed experimentally. The activation energy of the process is estimated. It is concluded that the synthesis of NpO2L*3H2O, a crystalline analogue of the previously described uranyl and the plutonium compound prepared in the course of this work, seems impossible in view of the high rate of reduction of Np (VI) by malonate ions even in the cold. The results obtained will be useful in determining the optimum conditions for the synthesis of crystalline compounds of penta- and hexavalent plutonium and neptunium.

Poster RER / 775

Radioiodine's behaviour in wastewater treatment plants samples

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Nuclear medicine involves the administration of I-131 to patients in order to treat or diagnose thyroid lesions. Commonly, it is administered orally in a liquid or capsule form. Once this radioisotope enters into the patient's body, it is mainly excreted via urine and in this way it can reach wastewater treatment plants (WWTP) 1. Consequently, radioiodine can be present in the different stages involved in this kind of facilities. Thus, WWTP workers can be exposed to this isotope via external irradiation or via inhalation due to the possible resuspension of dewatered sludge particles in air 2. Regarding to this topic, several studies can be found in the literature dealing with the determination of such radionuclide in samples from WWTP [2–4]. In particular, in a previous study carried out by our research group I-131 was found, among other radioisotopes commonly used for medical purposes, in sludge samples taken from a WWTP which collects wastewater from a hospital located in its area of influence. Besides the presence of I-131 in sludge samples, in recent years, attention has been also paid to speciation of I-131 in sludge samples in order to investigate the behaviour of this radioisotope in the process carried out for the sludge treatment [5,6]. However, there is limited literature related to this topic. With this in mind, one of our aims is to evaluate the radioiodine speciation into different sludge samples collected from a WWTP located in Spain by using gamma spectrometry and in this sense try to increase the existing data in this field. Moreover, as some works suggest [7,8], radioiodine can be potentially mobilized to air during the anaerobic digestion process performed in the WWTP. Taking this into account, air sampling was carried out from the digester and dewatered sludge areas of this WWTP to be able to determine if the workers can be potentially exposed under these circumstances.

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Poster TAN / 705

Volatile compounds for high-intensity beams to reach superheavy elements

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Research in the field of superheavy elements (SHE) has gained quite a significant progress with introduction of doubly magic ⁴⁸Ca beams into the experimental campaigns aimed to reach the shores of the island of stability 1. As known cross-sections of reactions, leading to SHE is limited to pico-barns thus requiring high beam intensities (now available within an order of magnitude less than 1 pμA) and experimental sessions lasting for months.
Admitting high importance of ⁴⁸Ca beams in SHE research one should take into consideration that modern and future experiments in that field require production of beams of other elements 2, i.e. ⁵⁰Ti, ⁴⁶Ti. Beams of such nuclides as ⁵⁴Cr, ⁵⁸Fe, ⁶⁴Ni and some others are also attracting attention. As stated higher, experiments make necessary long-term stability and high concentration of substance (vapor pressure), containing the nuclide, in the ion source. Some approaches to production and regeneration of such compounds as well as collation of them was performed.

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Poster TAN / 758

On the carbonyl complexes of Group VI-VIII transition metals: from molecular orbitals to structures

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The first synthesis of a superheavy carbonyl, seaborgium hexacarbonyl, has witnessed the step of the studies on superheavy element chemistry into a new era. The carbonyl complexes of element 107 (bohrium) and 108 (hassium) are the next targets for the investigation. In "few-atom chemistry"condition, the structures, properties and stabilities of bohrium carbonyls are influenced by both the relativistic effect and the John-Teller effect. Previous isothermal chromatography (IC) and thermal chromatography (TC) have proved the existence of the mononuclear carbonyls [M(CO)_x, M=Mo,W/Tc,Re/Ru,Os] with the short-lived lighter homologues in Group VI-VIII. However, some of these carbonyls are not stable (in the form of radicals); some are radioactive (technetium carbonyls). This brings much difficulty in the structural characterization.
In our recent investigations, the laser ablation and ionization time-of-flight mass spectrometer (LAI-TOF-MS) technique and the theoretical approach, including the molecular orbital theory (MOT), the ligand field theory (LFT) and the density functional theory (DFT), are applied to give a systematical explanation of their structural information and the transition mechanism for these mononuclear carbonyl complexes.
In LAI-TOF-MS, it is found that the 17e cations, [M(CO)₆]⁺ (M=Mo, W) and [M(CO)₅]⁺ (M=Ru, Os) for Group VI and VIII, and 18e [Re(CO)₆]⁺

for Group VII can be formed spontaneously in gas phase. This is confirmed by the theoretical calculations on complexes formation energy. Also, by MOT, LFT predictions and DFT calculations, it can be determined that in ground-state the geometries of Group VII hexacarbonyl cations and Group VI neutral hexacarbonyls are in the same point group (O_h), while those of Group VII pentacarbonyl anions and Group VIII neutral pentacarbonyls are in the same point group (D_{3h}). By energy calculation, the coordination number of the most stable mononuclear carbonyls are determined to be 6 for Group VI molecules/cations and Group VII cations, 5 for Group VII radicals/anions, Group VIII molecules/cations and Group VI anions, and 4 for Group VIII anions. These results also accord well with the LAI-TOF-MS experiments.

Poster RCH / 832

Radiation-induced preparation of ZnO:Ga-based scintillators with band gap modulation

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Radiation- and photo-induced precipitation was used for the synthesis of nanopowder scintillators based on ZnO:Ga. It is very efficient method published rarely outside of our research group. In principle, an aqueous solution of precursors (in the case of ZnO usually zinc nitrate or formate, a scavenger of OH radicals and/or hydrogen peroxide) is irradiated by non-ionizing (UV) or ionizing (γ, or accelerated electrons) radiation, causing the interaction with water radio- or photolysis products and leading to the precipitation of the solid nanocrystalline product - ZnO precursor (zinc carbonate hydrate, peroxide or directly zinc oxide). The following thermal decomposition leads to the formation of nanocrystals with particle size in the range of 20-100 nm, depending on the irradiation and heat treatment conditions.

The principle of band gap modulation consists in shifting the conduction band edge by affecting the energy levels of 2p orbital of Zn2+ by foreign ions and their energy levels. These foreign ions may occupy either the lattice position of Zn2+ ions or interstitial positions. If the concentration of foreign ions is sufficient, the bottom edge of the conduction band shifts. Doping by appropriate ions, such as Cd, Mg, Ca or Sr leads to the narrowing or broadening of the band gap. In summary, the changes in the band gap structure will be directly affected by the electron configuration of doping ion in the crystal lattice and with increasing concentration the shift of BG increases (Makino et al., 2001; Lange et al., 2012). The effect of doping ion can be observed as "red"or "blue"shift in emission spectra to lower or higher energies.

Cadmium and magnesium ions were chosen for the investigation of the ZnO band gap modulation, based on the previous promising results (Procházková 2013). It was shown that the concentrations up to 1mol% of Cd in ZnO structure have no effect on the intensity and position of exciton-related emission in UV range, but the increasing intensity of defect-related emission in visible range was observed. At higher Cd concentrations, modulation of the band gap and appropriate shift of emission maxima were observed. Significant blue or red shift of the UV emission maxima was obtained with the maximal admixture of 13 mol % of Cd or Mg ions, without any emission in the visible spectral range. It was manifested that the UV exciton-related emission can be shifted in the range of 376-425 nm at the cost of the decrease in total luminescence intensity.

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Poster RER / 568

Characterization of radioactive particles generated in low-yield atmospheric weapon tests

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It is a well-known fact that a series of different nuclear sources associated with the nuclear weapon and fuel cycles have contributed to the release of radioactive particles to the environment. In particular, at local scale, and following nuclear weapon tests, it has been observed that a major fraction of released refractory radionuclides such as uranium (U) and plutonium (Pu) are present as entities ranging from sub microns to fragments.
These radioactive particles can be found for example in the zone were the Trinity test (USA) was performed, and in the Ground Zero area at Semipalatinsk (Kazakhstan) were the former Soviet Union carried out atmospheric weapon tests in the 1950s. In both commented areas, low-yield atmospheric/surface weapon tests generated radioactive debris that was partially fused with soils at very high temperatures, producing glassy materials (particles) containing fission and activation products and transuranic radionuclides.
The physico-chemical characterization of these particles is essential in order to integrate them into environmental impact assessments, including the linking of particle characteristics to weathering rates and ecosystem transfer. With this end, several radioactive particles from the Trinity and Semipalatinsk Test Sites, identified initially by autoradiography and/or gamma-ray spectrometry, have been characterized (morphology, elemental, density) by the application of a series of non-destructive microanalytical techniques: SEM-EDX (scanning electron microscopy), micro-PIXE (proton induced X-ray emission), micro-CT (computed tomography) and micro-XRF (X-ray fluorescence). The main results and conclusions obtained in these characterizations, form the core of this paper.
After characterization, some of the isolated particles were dissolved by applying an alkali fusion procedure and the Pu-isotopic content determined. In particular, the 238Pu/239+240Pu activity ratios by alpha-particle spectrometry (AS) and the 240Pu/239Pu atomic ratios by accelerator mass spectrometry (AMS) were determined. Quite low 240Pu/239Pu atom ratios were obtained in the particles from both sites, fact that can be explained because the low yield of the explosion did no alter appreciably the 240Pu/239Pu atom ratio in the debris in comparison with the originally existing in the nuclear fuel (formed mainly by 239Pu).

Poster TAN / 826

On the formation and stability of metal carbonyl complexes from superheavy elements and their homologues

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The chemical investigation of molecular transactinide species is still a challenging task mainly due to the low production cross sections and short half-lives of all superheavy (SHE) isotopes available

so far. Despite those challenges, there is currently a lot of effort going on in this field as it promises a wealth of insights into relativistic effects, into the structure of the periodic table and into the properties of SHE atoms and molecules in general. A new approach to investigate the chemistry of transactinides is by synthesizing their corresponding metal-carbonyl compounds, which provides a number of advantages over alternative strategies. In theory, this strategy would allow the further investigation of Sg, Bh, and Hs, the very first chemical investigation of Mt as well as direct measurements of thermodynamic properties of their carbonyl compounds. A first experiment in 2014 lead to the successful synthesis of $Sg(CO)_6$.[1] Further experiments targeted at investigating the thermodynamic stability of this new compound were strongly hindered by unexpectedly low chemical yields for $Sg(CO)_6$ and its homologues $Mo(CO)_6$ and $W(CO)_6$, as well as the formation of yet unidentified side products.

In order to overcome these limitations, this study aims at gaining additional understanding on the formation and survival of metal-carbonyl complexes under single-atom-chemistry-conditions. Next to determining optimized reaction conditions for future experiments with transactinides, this project also aims at gaining a general understanding of the mechanisms leading to the formation and stability of metal carbonyls. For this purpose, a ^{252}Cf-source is used to produce short lived Mo-, Tc-, Ru- and Rh-isotopes as representative chemical homologues for Sg, Bh, Hs and Mt respectively. By connecting this source to a gas-loop that is filled with a gas mixture containing carbon monoxide, volatile carbonyl-complexes can be selectively and reproducibly synthesized. Those complexes are transported to a charcoal-trap by a gas-stream, where they are adsorbed until their decay can be registered by γ-spectrometry, which allows their quantification. Since this strategy is completely independent from accelerator beam-time, it is possible to execute a wide variety of experiments in a short time in order to probe the properties and responses of the carbonyl-formation-reaction to multiple physical and chemical parameters.

Here, we present results from studies of the influence of trace amounts of common impurities, of kinetic energy of the recoiling fission products, of pressure variations and of overall gas composition. Further studies concerning the formation and identification of side products, kinetic effects, alternative strategies for overall gas purification as well as theoretical simulations to support the experimental data are one the way.

1 J. Even et al, Science 345, 1491 (2014)

Poster TAN / 865

Extraction of Indium and Thallium Isotopes as the Homologues of Nihonium Into the Ionic Liquids

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This work represents a part of the research dealing with the study of properties of homologues of SHE. The aim of the experiments was to test the possibility of the separation of thallium from indium as the homologues of nihonium using the ionic liquids as the organic phase. Short-lived thallium and indium isotopes were generated by irradiation of Au and Ag target foil on U120M cyclotron at Nuclear Physics Institute of Czech Academy of Science in Rez near Prague. The irradiation was performed with beam of 3He ions accelerated to average energy of 47 MeV.
Two ionic liquids (1-hexyl-3-methylimidazolium bis (trifluoromethanesulfonyl) imide - [Hmim][NTf2] and Tributylmethylammonium bis (trifluoromethanesulfonyl) imide - [Tbma][NTf2]) were used as the organic phase. The extraction was carried out from the medium of hydrochloric acid in the concentration range from 0.2 to 5M. The influence of the oxidizing agent (NaClO2) was tested, whereas

Tl1+ is extracted successfully only in the higher concentrations of HCl, Tl3+ is extracted efficiently in the whole range of HCl concentrations.
Dependences of the distribution ratios on various extraction parameters will be presented. Based on the results, it can be concluded that extraction of indium and thallium into the tested ionic liquids is possible and will be used in the following detailed experiments.

Poster RER / 488

Determination of Pu-238,239+240, Am-241 and Sr-90 in air filters affected by Ruthenium isotopes and collected in autumn 2017

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At the very end of September and in October 2017 artificial radioactive isotope Ruthenium-106 has been detected in air over Europe. It was primarily found in Northern Italy and Central Europe. ^{106}Ru was detected by means of aerosol sampling stations used in continuous monitoring of presence of radioactive isotopes in ground-level air. In Krakow, ^{106}Ru was firstly detected on filter which had been exposed in period 25.09 - 02.10.2017 (standard week exposition) but in order to increase time resolution, next filters were changed every day. Early measurements involved assessment of activity concentration of ^{106}Ru in collected samples. Soon traces of ^{103}Ru were also found in the most active sample with ^{106}Ru/^{103}Ru activity ratio close to 3500. Moreover, due to unknown reason and source of release, further study also focused on determination of ^{241}Am, ^{90}Sr, ^{238}Pu and $^{239+240}$Pu in three samples with the highest activity concentration of ^{106}Ru, collected in time periods: 25.09-02.10, 02-03.10 and 03-04.10.
Air filter samples were collected at the Institute of Nuclear Physics PAN in Krakow using aerosol sampler which works at nominal flow rate 500 m^3/h and utilizes Petryanov filter FPP-15-1.5 (polyvinyl chloride) with high efficiency for collecting aerosols. After exposition, filters were compressed into pellets (5 cm diameter, about 4 mm height) and measured using low background gamma-ray spectrometer with HPGe detector (^{106}Ru (half-life 373 days) is a pure beta-emitter but within few minutes achieves secular equilibrium with its daughter isotope - ^{106}Rh (half-life 30 s) which decay is followed by gamma-ray emission). Subsequently, samples underwent radiochemical procedures in order to separate isotopes of Pu, Am (using Dowex-1x8 and TEVA) and Sr (using Sr-Resin). Determination of activity concentrations of actinides were carried out using alpha spectrometry and ^{90}Sr was determined by means of LSC technique.
Investigations showed that the highest concentration of ^{106}Ru in ground level air in Krakow was present in period 2-3 October 2017 and reached 16.7 ± 1.2 mBq/m^3. During following days its activity concentration was constantly declining and reached limit of detection on 5.10.2017. However, ^{106}Ru was still detectable in following weeks at levels three order of magnitude lower.
Preliminary results of conducted radiochemical analyses demonstrated that the highest activity concentrations of $^{239+240}$Pu and ^{241}Am were 2.42 ± 0.93 nBq/m^3 and 7.15 ± 5.45 nBq/m^3, respectively. The ^{238}Pu concentrations were below detection limit (estimated at 0.77 nBq/m^3). Results of ^{90}Sr concentrations, will be presented at the conference.

Poster RER / 501

Accumulation of technogenic radionuclides by plants of meadow natural communities

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Contamination of the aboveground parts of plants caused by root uptake and dust particles sedimentation on a surface of plant organs above the ground. The overall aim of the work is evaluate of foliar uptake of radionuclides in plants groves in areas with a high contamination density of soil by the radionuclides.
The studies were performed at the territory of Polessky State Radiation and Ecological Reserve (PSRER). We select Smooth brome grass (Bromus inermis Leyss.) and Wormwood (Artemisia campestris Leyss.) as on objects of study. Resuspension of radionuclides was estimated by deposition on the plates with Petriyanov tissue. Radionuclides washings from the surface of plant were used to estimate a contamination density. Absorbed dose rate of γ- radiation on the soil out the sampling site was 4.3-5.6 µGy•h-1; flux density of α- and β-particles, were: 62-82 and 91-125 pulse•s•cm2 respectively. Density of soil contamination was for Cs-137 –3.5 MBq•m-2, for Pu-238, Pu-239,240 and Am-241 –13.4; 29.1 and 82.9 kBq•m-2 respectively.
Resuspension values on sampling sites for Cs-137, Pu-238, Pu-239,240 and Am-241 were 1850, 0.54, 1.91 and 37.2 mBq•m-2 in day respectively.
Ratio of daily fallout activity on plates to soil contamination density was 0.53•10-6, 0.3•10-8, 0.4•10-8 and 0.42•10-6 respectively.
The radionuclides of the Chernobyl outburst of bioavailability for the plants was: Cs-137 > Am-241 > Pu-238 > Pu-239, 240. The specific activity of Cs-137, Pu-238, Pu-239,240 and Am-241 in the aboveground parts B. inermis was 626, 0.15, 0.12 and 0.54 Bq•kg-1. The specific activity of Cs-137, Pu-238, Pu-239,240 and Am-241 in the aboveground parts of A. campestris was: 1002, 0.35, 0.44 and 2.76 Bq•kg-1.
Am-241 and Cs-137 has higher capacity for air transfer than isotopes of plutonium so it causes a significant contribution to the total contamination of plants. The contribution of radionuclides adsorbed on the surface of the aboveground parts of plants in total pollution was 7-13% for Pu isotopes, 11-25% for Am-241 and 1.5-9.2% for Cs-137. Biological features of plants affect the accumulation of radionuclides to plants, and the morphology of plants to the level of surface contamination.

Poster RER / 517

Sorption of Sr and Cs onto Czech natural bentonite –Experiments and modelling

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Within the DGR project in the Czech Republic, the local Mg/Ca bentonite named BaM (Bentonite and Montmorillonite) has been tested with the aim to assess its sorption qualities with regard to cations necessary for the performance assessment of the repository.Sorption batch experiments present a tool to estimation of sorption coefficients which provide useful information on the transport properties of barrier materials. Depending on the requirements of the performance assessment and of the preferred transport models, various sorption models might be applied on the experimental data, which enable us to draw conclusions on different levels of understanding.
Hereby, data from batch experiments with Sr and Cs on Czech natural Mg/Ca bentonite which were performed in two background electrolytes, CaCl2 and NaCl respectively, of the same ionic strength (Is = 0.1 mol·L-1) are presented. Several values of solid-to-liquid ratios (m/V, [g·mL-1]) and a range of tracer total concentrations (C0, [mol·L-1]) were tested. The total concentration was controlled by non-active carrier (SrCl2·6H2O or CsCl) and the distribution between solid and liquid phase was assessed with the use of radioactive tracer (85Sr or 137Cs), concentration of which was negligible. On the basis of previously performed kinetic experiments, the equilibrium time was set to be 3 days for Sr and 7 days for Cs. The experimental vials were placed on the horizontal shaker for the desired contact time after which samples were centrifuged (966 g for 10 minutes) and 2 mL of supernatant were measured in a well-type NaI(Tl) scintillation detector. The average pH values at the end of experiments were 7.5 ± 0.1 in case of CaCl2 background electrolyte and 8.0 ± 0.1 in case of NaCl background electrolyte.
The sorption of Sr showed linear sorption isotherm in the whole range of tested concentrations.

However, there appeared to be a dependence of distribution coefficient (Kd, [L·kg-1]) on the value of m/V, especially pronounced in NaCl background electrolyte. This dependence might be described by power function. After such recalculation, sorption data are applicable for the use in the modelling of Sr transport in the compacted bentonite. Sr Kd values measured in NaCl background electrolyte were generally higher than those in CaCl2 background electrolyte for the given experimental conditions.

The sorption of Cs was non-linear due to Cs dual sorption mechanism on the clay material. The major mechanism is ion exchange and the minor mechanism is specific sorption on the frayed edge sites (FES). The sorption on the FES is dominant in the lower Cs concentration range whereas it is superimposed by ion exchange in higher concentration range. This dual sorption mechanism results in the large dependence of Kd values measured for each Cs initial concentration, which ranged from tens to thousands L·kg-1. The Kd values were higher in lower concentration range and the non-linearity was more pronounced when CaCl2 was used as a background electrolyte. No significant influence of m/V ratio was observed, even though certain trends might be identified. The sorption isotherm of Cs was described by two-site Langmuir isotherm.

Because the main mechanism of Sr sorption onto clay material is ion exchange, which is supported by observed fast sorption kinetics and the linearity of sorption isotherm, the geochemical sorption model used was one-site ion exchange model realized in the PhreeqC3 programming environment. The model was described by five ion exchange reactions. The ion exchange sites initial concentration and composition was based on the real composition of bentonite BaM. Equilibrium constants for exchange reactions of four major exchangeable cations were taken from literature (Bradbury and Baeyens, 2002) and equilibrium constants for tracer cations were fitted.

For the case of Cs, two-site ion exchange/surface complexation model was applied. The fitted equilibrium constant for ion exchange reaction was found to be significantly higher than those for other cations. Surface complexation was described by single reaction. Because of such simplified approach, the fitted constants of surface complexation reaction differed for each background electrolyte used with that for CaCl2 background electrolyte being higher which is in agreement with the result of empirical sorption model.

Poster RER / 525

Study of oil shale processing parameters on their sorption properties for liquid radioactive waste treatment

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One of the most important tasks of the nuclear industry is to develop effective liquid radioactive waste treatment technologies with the aim to minimize their amount and immobilizy for future safe storage. Various sorption materials are used to extract radionuclides from aqueous solutions. The products of oil shale processing can be the promising sorbents, as their mineral components are principally clay-marly composition and the fine structure of kerogen distribution in the amount up to 30 % by weight.

Natural oil shale samples of Belarusian origin were processed under various conditions: thermal decomposition, steam treatment, γ-irradiation. Processing conditions for oil shale are presented in the Table 1.

Experimental study was carried out to determine the sorption properties of obtained material towards the model solutions containing radionuclides ^{137}Cs, ^{60}Co and ^{85}Sr (neutral aqueous solution, 0,1 M NaCl and 0,05 M $CaCl_2$ solution). Distribution coefficient (K_d, ml / g) and sorption degree (S, %) are used for practical evaluation of sorption properties of the samples. The phase composition of the sorbents was studied by X-ray analysis. XRD patterns were collected on a DRON-3 powder diffractometer (CuKα radiation, 2θ = 10 –70 grad). The sorption and texture properties of the sorbents were evaluated from isotherms of low temperature (−196°C) physical adsorption–desorption of nitrogen, measured by the volumetric method on an ASAP 2020 MP surface area and porosity analyser (Micromeritics, USA).The resulting values of distribution factor in aqueous solution are presented in the Figure 1.

The best sorption properties towards radionuclide ^{137}Cs have shale samples 7, 8, 14. No influence

of solution composition is shown on ^{137}Cs sorption: high distribution factors were obtained as for solutions without electrolyte background as for solutions with competing stable cations Na^{+}, Ca^{2+}. The effect of competing cations is significant towards radionuclides ^{60}Co and ^{85}Sr. The best sorption properties in the neutral aqueous solution (pH = 6.5) have: shale sample № 2 –towards ^{60}Co; shale sample № 9 - towards ^{85}Sr.
The specific surface area of sorbents varies between 12 –29 m^{2}/g, the pore volume is 0.033 –0.097 cm^{3}/g, the pore size is 11 –17 nm.
Values of distribution coefficient (K$_{d}$) for samples of oil shale are 10^{3} –10^{4}. These values are rather high. Finally, based on these data, we can talk about high efficiency of natural sorbents on the basis of oil shale to extract radionuclides from solutions simulating liquid radioactive waste.

Poster RER / 526

Retention of Tc(VII) by pyrite nanoparticles

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One of the major pollutants in nuclear waste is ⁹⁹Tc, a redox-sensitive fission product from ²³⁵U and ²³⁹Pu with a long half-life of 2.14 X 10 ⁵ years. Under environmental oxidizing conditions, ⁹⁹Tc mainly exists in aqueous solution as pertechnetate, TcO₄⁻, which is a highly water-soluble ion that does not sorb significantly on minerals or sediments 1, so that, it is considered as inert with high risk to migrate through the environment. Under reducing conditions, Tc mobility is hampered by formation of Tc(IV) with significantly lower solubility 2. Pyrite, on one hand, is a good sorbent for Tc(VII) due to the presence of the functional groups FeOH and FeSH, and on the other hand, Tc(VII) is reduced to Tc(IV) by the interaction with structural Fe²⁺ and possibly with sulfur atoms. Moreover, Bruggeman et al. [3] found considerably increased Tc sorption when adding pyrite to boom clay in the presence of humic substances, which also favor the Tc retention [4,5].

In this work synthetized pyrite nanoparticles [6] were used to investigate Tc sorption in classical batch sorption experiments.

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Poster RER / 562

The adsorptive behaviour of heavy metals on magnetic nano-sorbents

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THE ADSORPTIVE BEHAVIOUR OF HEAVY METALS ON MAGNETIC NANO-SORBENTS
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Currently, water is one of the most vital human resources and is of the economic, social, political and environmental importance throughout the world. With the rapid development of the economy and industry, large amounts of wastewater containing different kinds of heavy metal ions and organic pollutants are generated. Unlike organic contaminants, heavy metals are not biodegradable and tend to accumulate in living organisms and many heavy metal ions are known to be toxic or carcinogenic. Nano-composites possessing magnetic properties can be used for the efficient removal of various toxic elements from contaminated solutions. Magnetic nano-materials, apart from having the high surface area to volume ratio, possess so many unique properties which make the remediation process economical, efficient and environmentally friendly. In this work we synthesized magnetic nano-sorbents (magnetite (M), magnetic graphene oxide (MGO), magnetic Prussian blue (MPB), magnetic Prussian blue with graphene oxide (MPBGO)) and applied them to the Cu(II), Co(II), Ni(II) and Pb(II) removal.
The batch technique was used to study the adsorption (concentration, pH dependences, dosage, reuse and etc.) of Cu(II), Co(II), Ni(II) and Pb(II) and three sets were conducted for each experiment. The kinetics of the adsorption process were investigated to determine the effect of the initial concentration of metal ions on the Qe (Equilibrium concentration) with respect to time and the time required to achieve the equilibrium adsorption. Two kinetic models, the pseudo-first-order and the pseudo-second-order, were used to analyze the rate of sorption. To analyze the rate controlling steps such as mass transport and chemical reaction processes, the intraparticle diffusion model (IDM) was applied to kinetics results.
Sorption dependences on the metal ion initial concentrations showed that, the maximum sorption capacities were characteristic for M, MGO, MPB and MPBGO in experiments with Ni (II) (150.86 mg/g), Co (II) –(145.9 mg/g), Pb (II) - (778.9 mg/g) and Cu (II) - (138.5 mg/g). Magnetic nano-sorbents are characterized by fast kinetics –the equilibrium is reached in 60 minutes. Results with M and MGO indicated that one step occurred in the sorption process (ion-exchange reactions on the surface). MPB and MPBGO showed multiple characterizations, indicating that two steps occurred in the sorption process.

Poster RER / 587

Soil-plant transfer ratios of tellurium and the estimation of committed effective dose from ingestion of radio-tellurium released by the Fukushima Daiichi Nuclear Power Plant accident

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Radio-tellurium isotopes such as Te-127m and Te-129m were released into the environment as a result of the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident. A large amount of Te-127m and Te-129m was released, and these isotopes have relatively long half-lives (109 and 33.6 days, respectively). As such, ingesting food harvested from the contaminated area shortly after the accident could have contributed to residents'internal radiation levels. However, our understanding of the environmental behavior and human radiation dose of radio-tellurium is limited. We previously reported soil-plant transfer factors (TF) and calculated the effective dose in the public.(1,2) The data obtained showed very large variations in the TF, and thus in the committed effective doses calculated with these TFs. In this study, we reanalyze the previous data, explain the mechanisms relating to the variations, and discuss the factors that may affect the estimated radiation dose values.
Experimentally-obtained soil-plant TFs were 1.2×10−2 to 3.0×10−2 for radish roots (Raphanus sativus var. sativus) and 1.4×10−2 to 5.1×10−2 for Komatsuna leaves (Brassica rapa var. perviridis). These values are much lower than those provided in the International Atomic Energy Agency technical report. The committed effective doses of radio-tellurium (Te-127 and Te-129m) were estimated using these TFs. Assuming that leafy vegetables such as Komatsuna were ingested one year post-harvest (30 April 2011), the committed effective doses were calculated to be 3.2% that of radio-cesium (Cs-134 and Cs-137) for an adult living in the southern area of Fukushima Prefecture. Dose coefficient, TF, and market dilution (the ratio of contaminated vegetables in the market) are likely the factors that significantly affect the estimation of committed effective dose.

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Poster RER / 647

Dissolved State of Radon Atom with Cluster Molecules of Solvent

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Radon is a rare gas element which has only radioactive isotopes. It is supposed to be an important and useful element because we could find anywhere on the earth and we might utilize for medical use in cancer therapy.
There are several measurements available such as an ionization chamber measurement and a track counter method. We can also measure the radon concentration in solution by using liquid scintillation counting which needs an extraction process into organic solvent while the element has a notable water solubility compared to the other rare gas elements.
However, dissolved state of radon atom in water is not well known thus far. In order to clarify the state, the interaction between radon atom and water molecules needs to be investigated.
In this study, we assume the interaction of a rare gas atom trapped inside a cluster of molecules in liquid, that were theoretically proposed in 20011 to estimate the enthalpy change due to their association based on a theoretical calculation. Besides, we performed measurement of distribution ratios of radon between mixed solutions of ethanol in water and dodecane to investigate the radon dissolution on cluster formation to be related to the concentration of alcohol in water.
For Ne, Ar, Kr, Xe, and Rn atoms into a cluster consisting of 20 H2O molecules, complexation energies were estimated by the DFT method (wB97XD/Aug-cc-pVDZ) with the BSSE correction. Since the calculated complexation energies and the solution enthalpies[2,3] showed a strong correlation, we have found that rare gas and water clustering is an important factor in the dissolution.
The performed experiments in this study also demonstrated that suppressed is the transfer of radon into the mixed solution in the concentration where the interaction of H2O and ethanol makes a layer

structure[4,5] rather than an cluster structure.
In conclusion, both of theoretical and experimental approaches to solution of rare gas atoms in water suggest a possible association with a cluster consisting of H2O molecules. The results will give one important information on the cluster formation of molecules.

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Poster TAN / 662

Calculation of francium hyperfine anomaly

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In recent years, the precision achieved in resonant ionization spectroscopy experiments coupled with advances in atomic theory has enabled new atomic physics based tests of nuclear models. The measurements of hyperfine constants and isotope shifts are highly sensitive to the changes of the nuclear charge and magnetic radii because they depend on the behavior of the electron wave function near the nucleus. The hyperfine structure splitting measurements can serve as a very useful tool for the understanding of changes in the nuclei shape in isotopic series of heavy elements. Magnetic hyperfine constants are usually assumed to be proportional to the nuclear magnetic moments. However, this is true only for the point-like nucleus. For the finite nucleus, we need to take into account both distribution of the magnetization inside the nucleus and dependence of the electron wave function on the nuclear charge radius. Together, these corrections are known as the hyperfine anomaly. In this report, we discuss how to calculate the hyperfine anomaly for many-electron atoms with the available atomic packages. All francium isotopes are unstable, it making difficult to carry out measurements of their nuclear g-factors by conventional techniques. Calculation of the hyperfine anomalies in this case can substantially improve the accuracy for nuclear g factors of francium isotopes, obtained from the ratios of the magnetic hyperfine constants.
In this work the magnetic hyperfine constants for low-lying states of neutral Fr atom are calculated within the configuration interaction (CI) method, the CI plus many-body perturbation theory (CI+MBPT) method, and the CI plus all-order (CI+AO) method. The effect of the Breit corrections and spin-polarization of the core are considered. Contribution of spin-polarization of the core significantly change the hyperfine anomaly for the $7p_{3/2}$ state.
We conclude, that for the precision measurements of g factors it is preferable to use the hyperfine constants for s and $p_{1/2}$ states, while the $p_{3/2}$ states are least useful.
The work was supported by the by the Russian Foundation for Basic Research grant No. 17-02-00216 A.

Poster RER / 688

Rare elements and radionuclides in aerosols of the Novosibirsk vicinity (Russia, West Siberia)

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The aerosol particles accumulated during winter in the snow cover of the Novosibirsk vicinity are studied by the means of the gamma-ray spectrometry, ICP-MS, X-ray fluorescent analytical methods. These studies allowed revealing the contribution of separate industrial enterprises into the general technogenic pollution of the megapolis with uranium and thorium.

The work was supported by the Russian foundation for basic research (RFBR) under project No. 09-05-00839 "Mineral and geochemical features of technogenic aerosol in Siberia"and No.14-05-00289 "Element and mineral-phase composition of technogenious aerosol as method basis for an assessment of environmental pollution of the urbanized and mining territories of Siberia". The part of the work relating to the measurement of spectra was done by analyst Yu.P. Kolmogorov using the infrastructure of the Shared-Use Center "Siberian Synchrotron and Terahertz Radiation Center (SSTRC) based on VEPP-3 of the Budker Institute of Nuclear Physics SB RAS.

Poster RER / 699

Application of radioactive and stable isotopes to trace organic matter in the Baltic Sea

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Application of radioactive and stable isotopes to trace organic matter in the Baltic Sea
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Keywords: 137Cs, 239,240Pu, 14C, δ13C , tracer studies

Pu isotopes, Δ14C and δ13C of total organic carbon, lipid and phospholipids (PL) fractions as well as humic asids of the sediments were applied to study the sources of organic substances including chemical warfare agents (CWA). The compound-specific δ13C analysis, PL-derived fatty acid biomarkers and an end-member mixing model were used to estimate a relative contribution of the marine, terrestrial and fossil, petroleum hydrocarbon sources to organic carbon in the sediments, to assess a possible effect of petroleum hydrocarbon contamination on radiocarbon signatures and to elucidate a possible leakage of CWA at the Gotland Deep dumpsite.
Liquid extraction, chromatography (GC-FID, Restek Rxi-1ms column and GC/MS - QP2010, Rxi-5Sil MS 0.22 mm x 25 m column), mass spectrometry and stable isotope compound specific analyses (Finnigan Trace GC ultra gas chromato¬graph, combined with the Thermo Finnigan Delta plus Advantage stable isotope spectrometer) as well as AMS (1.0 MV HVE Tandetron AMS) were applied. Measurements of Δ14C and δ13C in sediments and in different classes of organic substances as well as concentrations of n-alkanes in bottom sediments collected during 2011-2017 in the open Baltic

Sea waters were carried out to clarify their role in the formation of complicated organic carbon mixtures.
In addition phosphofipid derived biomarkers were measured in samples of sediments and suspended matter in order to obtain information about microbial communities and their response to the contamination. Radionuclide studies of sediments sampled during 2011-2014 in the open Baltic Sea and in Curonian Lagoon showed wide variations with low 137Cs and 239,240Pu activity concentrations in the Lagoon and higher levels in the open Baltic Sea, including the Gotland Deep. Similarly, more depleted δ13CTOC values in sediments were found in the Curonian Lagoon when compared with the open Baltic Sea, in agreement with the expected transport of natural organic matter of the terrestrial origin to the Baltic Sea. It has been shown that contrary to the variation of δ13CTOC, the most depleted Δ14CTOC values were found in the Gotland Deep, which could indicate a leakage from dumped chemical warfare agents, rich in fossil carbon (petroleum, natural gas, coal).

Poster RER / 703

Radionuclides distribution in roach and pike organs

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Fish is part of the food chain and actively involved in radionuclides migration processes. The investigations of real samples from radioactively contaminated water reservoirs were studied. The reservoir R-4 of the "Mayak"Production Association using as a low-level waste (LLW) storage and it is unique in the number and radionuclides composition. The study purpose was to establish the radionuclide distribution parameters within the artificial reservoir R-4 fishes.
The reservoir R-4 characteristics: the water area is 1.3 km2, the water volume is 3.8 mln.m3, the average depth is 3.1 m. The total activity is 555·106 Bq. As a result of field work, the following samples were selected: water, bottom sediments and fish (pike, roach). Investigated fish are the main species of the reservoir ichthyofauna. Samples of fish were divided into organs: scales, muscles, liver, gills and frozen.
Samples preparation for alpha (U, Pu, Am) and gamma (Cs) measurements included the following steps: ashing in a muffle furnace (8 hours, 450°C), acidic autopsy (HNO3 conc., 4 hours) and co-precipitation ((NH4)2HPO4 + NH4OH). All samples were analysed using gamma-spectrometry (Canberra GR 3818). Alpha spectrometry was used for alpha-emitted nuclides determination (CANBERRA Model 7401) after separation using extraction chromatography (UTEVA, TRU (TrisKem International).
It was investigated three sample of each fish species. The mass of the pike was varied from 2 to 3,5 kg; mass of roach –100-180 g. The summary activity of dose-forming 137Cs is higher in pike (1930 Bq/g), than in roach (1590 Bq/g), that is typical for the accumulation of radionuclides along the food chain. Distribution of activity through the organs is equal for pike and roach, accumulation occurs by chain: muscles ⟶ gills ⟶ scales ⟶ bones ⟶ blood. Activity in muscles is equal, the greatest difference in values related to accumulation of radionuclides by external organs.
The alpha-emitted nuclides total activity in external organs is to 2-3 orders higher than in internal organs. The highest americium activity in all studied roach's organs indicates its bioavailability. The greatest content of plutonium (141,3 Bq/g) and americium (5,8 Bq/g) observed in roach gills; uranium –in scales (3,4 Bq/g). For pike also highest activity was found in gills (10 Bq/g) and scales (2,9 Bq/g).
The radionuclides concentration ratio (CR) was calculated. 137Cs CR (total activity in fish/activity in reservoir water) 835 for pike and 1100 for roach are approximately the same, that indicates ion form sorption from water by external organs and accumulation in muscles. Calculate uranium coefficient is impossible because of its absence in water, that indicate other proceed ways to organs. For other alpha-emitted radionuclides CR is one order different (for pike is less, than for roach). To explain this distribution additional calculations and analysis of accumulation Pu and Am by each organ are needed.

Poster RER / 709

Inverse estimation of upscaling of dispersion for cesium transport in crushed granite media

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Crystalline rock is suitable the safety disposal of high-level radioactive waste (HLW). Fracture networks connections can dominate flow behaviors at the crystalline rock. In this study, advection-dispersion column experiments of cesium (Cs) through crushed granite (0.297 - 0.840 mm) were constructed where synthetic groundwater (GW) and seawater (SW) were employed as the liquid phase. This study conducts multi-length column tests (length: 2, 4, and 8 cm). The other parameters of columns are diameter of 5 cm, bulk density of 1.45 g/cm3 and porosity of 0.45.

This study presents the HYDURS-1D model simulation to estimate the parameters of transport in 1D crushed granite media. The dispersivity for multi-length crushed granite is estimated by using Marquardt-Levenberg optimization algorithm in the HYDRUS-1D model. The estimated dispersivity can provide a valuable reference for radionuclide transport of waste repositories.

Poster RER / 742

Concentration of Be-7 in Saudi Arabia

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Exposure to alpha, beta or gamma radiations from natural sources, which is varies by location and altitude, affect the living organism. The main sources are earth crust (83%) and cosmic (17), with annual exposure 2.0 mSv and 0.4 mSv, respectively. The objective of this study is to identify the radionuclide concentration in certain cities (Riyadh , Hagl and Khafji), of Saudi Arabia. The radionuclide were identified using gamma spectroscopy equipped with a high-purity germanium (HPGe) detector during twelve months. The results illustrate the observation of Be-7 (T1/2= 53.3 days), K-40 (T1/2= 1.251×109 years) , Cs-137 (T1/2= 30.17 years) and Ra-226 (T1/2= 1600 years) with average activity concentration in surface air samples (mBq/m3) of 11.8±0.7, 1. 9±0.1, 0.01±0.0 and 0.24 ±0.03 at the same order in Riyadh city which is located in the central of Saudi Arabia. In addition, comparable concentrations were measured in Hagl and Khafji cities which are located in both north corners of Saudi Arabia. Various concentration of Be-7 were noticed during certain time in a year, depending on the variation of the environmental conditions (climate) and the geographical location. This can be attributed to fact that B-7 is produced in upper atmosphere and transported to troposphere.

Poster RER / 768

Aggregation of Bentonite Colloids：Influence of Background Electrolyte, pH and Humic Substance

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Colloids existing in soils and groundwater aquifers can facilitate the transport of environmental contaminants including radionuclides as well as hydrophobic organic compounds. The mobility of colloids depends strongly on dispersion and aggregation of the particles. Therefore, the environmental fate and stability of colloidal bentonite particles under various conditions are crucial for assessing the safety of radioactive waste repository. In China, bentonite from Gaomiaozi region (Inner Mongolia, China) has drawn widely concerns as a buffer material in high-level radioactive waste repository. Erosion of the back-filled bentonite blocks by flowing in-situ groundwater is expected to produce bentonite colloids easily.

In present work, aggregation of bentonite colloids were investigated by the photon correlation spectroscopy (PCS), effects of colloids concentration, pH, background electrolytes (including Eu3+, Ba2+, Sr2+, Ca2+, Mg2+ and Na+) and humic substance on colloidal aggregation were discussed in detail, and the DLVO model were employed to demonstrate the aggregation process. Results showed that the stability of GMZ Na-bentonite colloids was dependent significantly on cations concentrations and pH values, while slightly on colloids concentrations and anions concentrations, aggregation was getting obvious and kinetically fast when increasing either salinity or acidity. The presence of humic acid enhanced colloidal stability and thus acted as an unstable role in repository terrains. The results in present work can provide a fundamental understanding on environmental behavior of bentonite colloids as well as a vital reference for safety assessment of high-level radioactive waste repository.

Poster RER / 769

240Pu/239Pu atom ratios in the Bay of Bengal water column

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The plutonium isotopes, 239Pu and 240Pu, have been added to the surface oceans mainly as a consequence of global fallout from atmospheric nuclear weapons testing. There were two major sources of Pu isotope in seawater in the western North Pacific Ocean: global (stratospheric) fallout which occurred mainly in the late 1950s and early 1960s, and local fallout from the Pacific Proving Grounds (PPG) in the Marshall Islands which occurred mainly in the early 1950s. Since the 240Pu/239Pu atom ratio is characteristic for the Pu emission source, information on Pu isotopic signature is very useful to better understand the transport process in the oceans and to identify the sources of Pu. The mean atom ratio of 240Pu/239Pu from the global stratospheric fallout is 0.180 ± 0.014, whereas that from close-in tropospheric fallout from the PPG is 0.33 –0.36 . The 239Pu and 240Pu inventories and 240Pu/239Pu atom ratios in seawater will provide important and useful data for understanding the process controlling Pu transport. The objectives of this study were to measure the 239Pu and 240Pu concentrations and 240Pu/239Pu atom ratios in seawater from the Bay of Bengal and to discuss the transport processes of plutonium in the ocean.

Seawater samples were collected from the surface to the bottom in the the Bay of Bengal with acoustically triggered quadruple PVC sampling bottles during the R/V Hakuho-Maru cruise. The Bay of Bengal station is located on the eastern end of the broad Bengal Fan. The 239Pu and 240Pu concentrations and 240Pu/239Pu atom ratios were measured with a sector field inductively coupled plasma mass spectrometer (SF-ICP-MS), which was equipped with a guard electrode to eliminate secondary

discharge in the plasma and to enhance overall sensitivity.
The 239Pu, 240Pu and 239+240Pu concentrations in the surface water were 0.71, 0.56 and 1.27 mBq/m3, respectively, and they increased with depth; a sharp subsurface maximum over the depth interval 200 - 400 m was identified, and they decreased with depth. The 239Pu, 240Pu and 239+240Pu inventories in the entire water column were 6.9, 5.8 and 12.7 Bq/m2, respectively. The atom ratios of 240Pu/239Pu ranged from 0.22 to 0.24. The atom ratios in water column from the Bay of Bengal were significantly higher than the mean global fallout ratio of 0.180 ± 0.014. These high atom ratios proved the existence of close-in fallout plutonium originating from the Pacific Proving Grounds.

Poster RER / 853

Frequency distributions of radiocesium in edible wild mushrooms

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After the Fukushima Daiichi Nuclear Power Plant (FNPP) accident in March 2011, a huge amount of radioactive cesium was released over a widespread area in eastern Japan. People were seriously concerned about food safety. Therefore, the Ministry of Health, Labor and Welfare established a provisional regulatory value of 500 Bq/kg for radiocesium in cereals, vegetables, meat, and fishery products. In April 2012, a new maximum limit of 100 Bq/kg was established as a new standard of radiocesium in general food. Since 2012 April, the results of monitoring has indicated that the violation rates were extremely low, much less than 1%, for agricultural products including cereals, vegetables and fruits, meat and milk, fish and fishery products, except for wild mushrooms. Even in 2018, several cities in Miyagi prefecture which locates 50-160 km north from the FNPP still prohibit the shipment of wild mushrooms. However, the decisions by the government do not always reflect the practical pollution of the wild mushrooms because of poor sampling data. We collected many edible wild mushrooms at several forests in Miyagi prefecture and investigated the frequency distribution of the radioactivity of the samples. We discuss annual, air dose rate and species dependences of the radioactivity concentration.

Poster RER / 825

Plutonium, thorium and 137Cs in the bottom sediments of the deep parts of some Mazurian Lakes (Northeastern Poland)

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Results for, Cs-137, Pu and Th isotopes activity concentration found in the upper part of bottom sediments taken from a deep part of 29 lakes on Mazurian Lakes area (North-Eastern Poland). Samples were collected by diving. Results for 137Cs were obtained using low background gamma ray spectrometry, Pu-238, Pu-239+240, Th-230 and Th-232 were obtained by means of alpha spectrometry and those for 241Pu were obtained using liquid scintillation spectrometry. Activities for Pu-239+240 range from 81(13) mBq/kg to 3820(265) mBq/kg, whereas for Pu-238 they range from 9(5) mBq/kg

up to 549(122) mBq/kg. Arithmetic mean activity for Pu-239+240 was 1285 mBq/kg with the standard deviation equal to 1012 mBq/kg. Analyses of Pu isotopic ratios allowed for the discussion of Pu origin. Maximum percentage of Pu-239+240 activity from Chernobyl fallout was 24%. It was significantly lower then found in previous measurements in forests at the same region of Poland. It suggest a difference in Chernobyl hot particles fallout between forests and lakes. Surface deposition of Pu-239+240 was calculated. No clear relationship was found between the plutonium and main chemical matrix components of sample nor with the trophic status of the lake. Pu activities were not well correlated with measured Cs-137 activities. Pu activities were not correlated with Th activities. Th-230 and Th-232 were strongly correlated.

Poster RER / 833

Study of Po-210 content in the urine of people living in the homes with high radon concentrations

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The objective of the presented research is to find a possible effect of living and/or working in places where the radon concentration exceeds the Czech guideline (reference) level of 300 Bq/m3. The first step was to determine 210Po concentration in urine samples from such people. Twenty-nine non-smokers volunteered to provide a urine sample excreted within 24 hours. They were asked to not consume fish, seafood, liver and kidney one week before and during the urine collection. These factors influence the intake of 210Po, and therefore its excretion in urine, as it was shown in the review paper by Hölgye and Straková (2009).

The values of 210Po concentration found in urine samples were in the range of 3.1 - 28.8 mBq/day. The arithmetic mean (AM) was calculated at 7.64 ± 5.78 mBq/day and a geometric mean (GM) was 6.4 mBq. On the other hand, people who have not been exposed to high radon concentrations had a range of 1.4 - 10.8 mBq/day of daily excretion of 210Po, AM 4.1 ± 2.0 mBq/day and GM 3.7 mBq/day, as was found by Hölgye (2013) in a group of 40 Prague citizens. Based on these results, it can be suggested that the individuals staying in an environment with increased radon activity concentration excrete more 210Po in the urine.

Knowledge of 222Rn activity concentrations in homes of the volunteers alone is not adequate to determine the possible impact of radon content in houses on 210Po excretion. To evaluate this effect, additional data about the environment in which the people under investigation are living and/or working and the habits of these people (ventilation, outdoor stay, etc.) will be obtained and used in this study.

Poster RER / 569

Natural radionuclides in acid mining drainage waters after a not controlled discharge from a pit lake in the south of Spain

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The Iberian Pyrite Belt (South of Spain) is plenty of acid pit lakes as a result of former mining. These pit lakes are leaching some limited amounts of acid water into their close environment: acid mining

drainages (AMD). But in 2017, an accident occurred in an abandoned mine called "La Zarza"and from the associated pit lake approximately 250,000 m3 of acid water were discharged into a small tributary of the Odiel River. Policy makers decided the immediate application of countermeasures to decrease the impact of the release, being the main one to increase the pH of water affected by AMD . With this end, the water discharged from the pit lake was halted and amounts of calcareous rocks were fallen into the AMD in some zones.
In this work, Uranium and Thorium concentration were determined in waters collected immediately after the accident in the small tributay affected by the discharge and fin the Odiel River, and the obtained results are presented and discussed . In the case of the Odiel River, the results corresponding to the samples collected after the accident are compared to previous levels observed under normal conditions.
Although the Uranium and Thorium concentrations are clearly elevated in the AMD, an irregular distribution, related to the presence of calcareous rocks in some zones (discharged as main countermeasure against the low pH of water) were observed in the small tributary. On the other hand, no differences of levels in the Odiel River, comparing previous levels and levels at the moment of accident are observed. Consequently, a positive effect of countermeasures (stopping the discharges and the use of calcareous rocks) can be inferred.

Keywords: Pyrite mines, radioactivity, environmental impact.

Poster RER / 842

Geochemical modeling of strontium transport in nitrate solution. focus on dispersion edge processes

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The practice of liquid radioactive waste (LRW) injection in deep stagnant aquifers in Russia dates back to 1963 year. The safety assessment and safe operation of existing LRW repositories require complex modeling of basic geochemical processes. Sorption onto mineral phases tends to be the main process affecting the dynamics of migration of radioactive and other contaminants in geological media. Meanwhile, the behavior of macrocomponents in contaminated subsurface aquifers depends on a number of parameters, including microbial processes.
The purpose of this paper is to create a multiparametric model of strontium migration in subsurface aquifers, with such a model to consider the contribution of nitrate ions as the main macrocomponents of radioactive waste, highlighting the role of microbiological denitrification, the effect of concentration blurring on the dispersion surface.
The ion exchange adsorption and transport models were complemented by nitrate biodegradation process due to bacterial denitrification. Calculations of migration were carried out in the PhreeqC 2.18 software.
Sorption experiments were performed with a sandy–clay rock taken from the geological stratum into which the liquid radioactive waste was injected. The cation exchange capacity (CEC) of native rock is 4.40 mg-eq. per 100 g. The model draws on the assumption that ion sorption occurs at Sr Selective (8.12% of total quantity) and Sr Nonselective ion exchange sites.
Calculations for denitrification parameters were made during laboratory experiments with microbial communities isolated from samples collected from Russian deep LRW repository in Krasnoyarsk region. Laboratory modeling of microbiological processes was used to establish that the denitrifying microorganisms inhabiting the Severny MCC repository are capabale of living in solutions with up to 8 g/l of nitrate ions, i.e. on the waste dispersion surface and in the low-active radioactive waste repository area, while the denitrification rate will be suppressed as nitrate ion concentration increases over 3 g/l.
It was revealed that biodegradation of a nitrate-ion can strongly influence strontium transport. It follows from the outputs provided in this work that biodegradation of the nitrate ion is observed due

to the dispersion front dilution, while the strontium complex with nitrate ion does not participate in sorption, and the strontium front is advancing faster.

Poster RER / 835

Biogeochemical impact on Uranium migration in aquifers near to sludge depository

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The work presents data on the aerobic and anaerobic microbiological processes impact on Uranium migration in upper aquifers polluted with sulfate and nitrate ions near to sludge depository of Novosibirsk chemical concentrate plant. Sludge depository was built in 1960s and still works. Uranium concentration in subsurface water reaches in several zones 1-2 mg/L and its migration depends on some geochemical aspects of water horizon: mineral sorption capacity, TOC, Eh, oxidizers concentration (O2, NO3, Fe+3, etc.) and biogeochemical processes.
We have studied chemical and microbiological characteristic of water samples of contaminated and uncontaminated areas and have obtained main data for biogeochemical modelling of Uranium migration capacity.
It was indentified, that different microbial communities consist of aerobic organotrophic, denitrifying, sulphatereducing and fe-reducing bacteria, and there where strains capable of forming biofilms on sandy minerals in aquifer.
Biogeochemical modelling in PHREEQC software shows, that Uranium immobilization is possible in local areas with high content of organic substances, and the most important microbiological process is the redox potential reduction due to aerobic respiration. After that in anaerobic conditions redox-dependent Uranium reduction can be expected. Moreover, in the presence of sulfate ions, further anaerobic processes of microbial sulfate reduction and iron reduction lead to the formation of iron sulphide, which plays the significant role as an antioxidant buffer in the case of oxygen migration.
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Poster TAN / 702

Np(V)-phenanthroline-dicarboxylic acid compounds: Synthesis, structure, spectral studies, complexation in solutions

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The interest in interaction of actinide "yl"cations with anions of heterocyclic carboxylic acids appreciably increased recently, which is caused by a number of factors. In particular, it became clear that the coordination behavior in the series of hexavalent (U, Np, Pu) and pentavalent (Np, Pu, Am) actinides in some systems significantly varies. For more detailed study of the coordination interaction of f elements with various kinds of ligands, it is necessary to expand the range

of the experimentally studied complexes. In this aspect 1,10-phenanthroline-2,9-dicarboxylic acid, C12H6N2(COOH)2 (H2PDA), is of much interest. Its specific feature is the rigid structure determined by the phenanthroline core, with fairly strictly fixed distances between atoms of functional groups. These properties of the PDA2–ion allow us to expect increased selectivity of the complexation with metal ions depending on their ionic radius. Therefore, this ion is considered as promising ligand for separation of elements with similar properties. Among complexes of actinyl ions, before this work only one uranyl compound, UO2(PDA), has been isolated in the crystalline form and structurally characterized. Here three novel complexes of Np(V) with 1,10-phenanthroline-2,9-dicarboxylic acid, of the compositions [(NpO2)2(PDA)(H2O)3]•H2O (I), (NH4)2[NpO2(PDA)]2•3H2O (II), and [C(NH2)3]2[NpO2•(PDA)]2•4H2O (III) were synthesized and structurally characterized by a single crystal diffractometry. All the compounds were synthesized by the hydrothermal procedure. The Np atoms in the crystal lattices of all the compounds have the pentagonal bipyramidal coordination surrounding, with the [C12H6N2(COO)2]2–anions acting as chelate-bridging N,O-donor ligands. In the structure of I, two crystallographically independent NpO2+ dioxocations participate in the cation–cation interaction leading to the formation of tetrameric cation–cation complexes. The nonequivalence of the Np atoms is manifested in splitting of the main absorption band of Np(V) in the electronic spectrum of solid compound I. The structures of II and III are based on dimeric anionic complexes [NpO2(C12H6N2(COO)2)]22–. Only one kind of complexes, NpO2(PDA)–, was detected in the solution, and high value of the concentration stability constant β ~1012 L mol–1, is due to tetradentate coordination of the ligand.

Poster RCH / 767

A Study on Post-effects of Proton Beam Irradiation of Tantalum Metal Using Probe Emission Mössbauer Spectroscopy

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For the irradiations, 40 mμ thick tantalum foils were used that have been annealed after being rolled and labelled with 57Co. Diffusion impingement of evaporated radioactive atoms onto the surface of tantalum substrates was carried out for 140 minutes at 1270-1275 K (E-7 Torr) that was followed by cooling both the foils and the furnace down to the ambient temperature.
Irradiation conditions for the samples in the MGC-20 isochronous cyclotron are given in the Table below:

Conditions of Irradiation Foil 1 Foil 2
Fluence, particles/m2 2.6 E21 2.6E21
Integral beam current, μA 11.45 11.45
Proton energy losses, MeV 6.95/5.24 5.08/2.86
Heat released, Watts 19.6 25.4
Temperature of targets, K (310+/-3) (310+/-3)

The Mössbauer Spectra measured before and after irradiation are a superposition of the two single lines that correspond to different positions of the 57Co atoms in BCC Tantalum lattice: Singlet 1 corresponds to the 57Co atoms at the boundaries of crystallite grains; Singlet 2, to the 57Co atoms in the lattice sites (the isomer shift that is characteristic for interstitial atoms).
The nuclear prehistory has no practical influence on the proportion of spectral lines: the low temperature of targets in the process of irradiation excludes the possibility for the impurity atoms to migrate within bulk samples; the fraction of Singlet 1 is (37+/-2) %, for Singlet 2, it is (63+/-2)%.
The presence of a significant number of own radiation-induced defects (that are formed resulting from irradiation) in the vicinity of the Mössbauer atom causes the experimental lines to be broadened. As the proton energy decreases (i.e., as the cross section of their interaction with atoms of the target increases), this effect is enhanced and is especially strongly revealed for the interstitial 57Co atoms.

Poster RCH / 855

ELI Beamlines - Research Program 4: Applications in Molecular, Biomedical and Material Science

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At the ELI Beamlines facility the first endstations dedicated to user experiments that are starting just now are those of the team fulfilling the Research Program 4: Applications in Molecular, Biomedical and Material Science. The endstations will utilize optical, extreme UV and plasma X-ray beams to perform pump-probe experiments on diverse sets of samples (free atoms, molecules and their clusters / materials / biochemical compounds) with temporal resolution down to tens of femtoseconds. As one of the possible options, it will be possible to perform research in ultrafast radiation chemistry induced by either hard photons or multi-photon ionization processes. The poster provides an overview of the current instrumental capabilities and outlook for their future developments and upgrades.

Poster RCH / 633

Fabrication of Antibacterial Foam Dressing Including Radiation Assisted Silver-Carbon Composites

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Dressings for human wounds have been aimed at protection, removal of exudate, inhibition of exogenous microorganism invasion, and improved appearance. Protection was accomplished by covering the wound with dressing materials. Wound areas that are kept just damp may heal faster, but accumulation of exudates under the dressing can cause infection. Silver is important in the treatment of wounds due to its anti-microbial properties, and commonly used in the treatment of major burn injuries where bacterial infection is common. Also, activated carbons are the most widely used industrial adsorbent for removing contaminants from gaseous, aqueous, and non-aqueous streams. We fabricated activated carbon powder by using radiation technology. We purified lignin from black liquor and then electron beam irradiated to stabilize lignin for carbonization.

Finally, we fabricated and characterized the functional polyurethane (PU) foams containing silver nanoparticles and activated carbon composites. Various PU foams containing Ag and AC composites were prepared and their cytotoxicity, anti-microbial activity, and in vivo wound healing ability were evaluated. Ag and AC composite particles were homogeneously dispersed in PU foams. In particular, PU foam had sufficient anti-microbial activity against two pathogenic bacteria and had good biocompatibility. PU foams showed effective wound healing compared to controls. It will be a useful approach as a functional wound dressing material.

Poster RCH / 634

Electron Beam Treatment and Characterization of Lignin Obtained from Black Liquor by A Mini-Pilot Scale Extraction System

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Black liquor is biomass-based industrial waste obtained from the chemical pulping process of wood. The lignocellulosic materials such as lignin, hemicellulose and other extractives are dissolved into black liquor. Lignin, which can also be extracted from black liquor, exhibits good thermal stability and high carbon yield. In the present study, compared to the lignin obtained by a lab-scale experimental set-up, a relatively large amount of lignin was extracted from black liquor by a mini-pilot scale extraction system. Lignin extraction was performed by adjusting the pH value by adding sulfuric acid and, then neutralizing by filtering and washing it with distilled water. The extracted lignin was treated by irradiation process at different electron beam absorption doses. Electron beam irradiation process was performed at ambient temperature in air by using industrial-level radiation facility. Prior to electron beam processing, a certain amount of the extracted lignin was placed in a polyethylene bag. The electron beam energy used was 1.14 MeV, the current was 7.2 mA, and the moving conveying rate was 10 m/min. The irradiation was done in the electron beam channel, at which the samples to be irradiated were placed in the moving cart. The electron beam absorption dose was controlled by the number of sample irradiations exposed in the channel. The absorption doses ranging from 100 to 1000 kGy were irradiated to the extracted lignin. The extracted lignin and the irradiated lignin were characterized through various analytical methods such as attenuated reflectance- Fourier transform infrared spectroscopy (ATR-FTIR), proton-nuclear magnetic resonance spectroscopy (1H-NMR), thermogravimetry analysis (TGA), and scanning electron microscopy (SEM). The result shows that the lignin obtained from black liquor by a mini-pilot scale extraction system indicates chemical characteristics of typical lignin. The present study suggests that the lignin extracted by a scaled-up process and also the irradiated lignin may be used for preparing lignin-based activated carbon through carbonization and activation processes.

Poster RER / 857

Sorption properties of microbial treated aquifer grounds in according to technetium, uranium, neptunium, plutonium and americium

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Ground water pollution by long living radionuclides can cause their introduction into subsurface environment and drinking water sources. Such actinides as uranium, neptunium, plutonium, americium and technetium as fission product are ones of the most important radionuclides in radioecological management because of their toxicity and long living. Their migration ability in environment depends on variety of factors namely redox conditions, presence of dissolved oxygen, ground water content and etc. Radionuclide contamination often is coupled with high concentrations of nitrates, sulphates and dissolved organic compounds, which in their turn can cause the development of indigenous microflora which uses such compounds in its biochemical processes. Iron containing minerals are considered to be important components for radionuclide sorption and reduction. Biochemical

cycle of iron is known to be the one of the most common and accessible in subsurface environment. Bacteria are able to consume avaible iron from iron-containing minerals with changing their structure, surface area, oxidation state of iron and therefore their sorption properties.

The aim of this work is to estimate the role of microbial treatment (including dissolution of initial minerals and precipitating biogenic iron-containing phases) on sorption properties of aquifer grounds in according to technetium, uranium, neptunium, plutonium and americium.

Pyrite, siderite, gematite and biotite were treated by natural microbial community collected from boreholes located on SCC site (Tomsk region) near deep underground LRW repository of injection type aquifer horizonts from the depth 300-400 meters contaminated by nitrates, sulphates and uranium. Biogenic depend changes of minerals were studied. Changes in sorption properties of biogenic treated samples were estimated as well.

Modeling was performed using PHREEQC code and llnl.dat data base.

The work was supported by the Russian Science Foundation, project No. 17-17-01212.

Poster RCH / 730

Radiolysis of adenine in aqueous solutions and clay suspensions

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The aim is this work is to establish if the presence of a solid surface, like a clay mineral, during the irradiation of adenine (purine bases) alter the stability, formation, and distribution of radiolytic products in comparison with samples of those molecules without the mineral. The results showed that these bases could receive higher irradiation doses with less decomposition to the samples without clay (recovery of 96-98.%). The irradiation was carried out in a 60-Cobalt-Gamma source with irradiation doses up 91 kGy. This results can be applied to chemical evolution studies in which the preservation of organic compounds to energy sources may be important for the stability and formation of more complex prebiotic organic compounds.

The analysis of the irradiated samples was carried out by infrared spectroscopy, high-pressure liquid chromatography (HPLC) and HPLC-mass spectrometry.

This work was supported by CONACyT (grant No. C001-CONACyT-ANR-188689) and PAPIIT (grant No. IN226817). J.C. received supported from a CONACyT fellowship and from the Posgrado en Ciencias Químicas. We thank Chem. Claudia Camargo, M.Sc. Benjamin Leal, and Phys. Francisco Flores for their technical assistance.

RER 2 / 727

Environmental radioactivity studies in Kabul and northern Afghanistan

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Enhanced environmental radioactivity was reported for some spots of the Kabul suburbs during the Soviet era. As residential houses have been built in these places, 51 soil and rock samples as well as 51 all-purpose water samples were collected in three phases and measured in order to quantify the primordial isotopes and daughter nuclides of the respective decay chains and possible contamination by manmade radionuclides. For the rocks and soil samples gamma spectroscopy was used as main technique, while ICP-MS and ICP-OES were used as main technique for water analysis. Furthermore, alpha spectroscopy, μ-XRF, PXRD, TOF-SIMS and LSC were used to verify the gamma spectroscopy and ICP-MS results. Activity concentrations in soil and rocks ranged between 160 to 28600 Bq/kg, 73 to 383000Bq/kg, and 270 to 24600 Bq/kg for uranium, thorium, and potassium, respectively, all measurement methods did not indicate any anomalies and identified the samples as samples high in natural radioactivity exhibiting a thorium containing cheralite mineral structure. Uranium and thorium concentrations in waters ranged from 0.01ppb to 26 ppb and from 2 ppb to 150 ppb, respectively, no considerable amounts of lead and radium were detected. Most of the investigated waters are safe to drink, some exhibit element concentrations exceeding the national and international recommended values 1.

To assess the level of hazardousness imposed by the enhanced activity in the living spaces further, radon gas concentrations in eight basements, four living rooms and four caves from different locations in Kabul and Panjsher, Afghanistan, were measured using eight active radon exposure meters recently developed by the Helmholtz Center in Munich, Germany 2. The two-phase measurements lasted from a week to a year. In the first phase of measurements which lasted one week, the mean activity concentrations ranged from 6 to 120Bq/m3 and 25 to 139Bq/m3 for the basements and caves, respectively. In the second phase of measurements which lasted one year, the mean activity concentrations ranged from 33 to 2000 Bq/m3 and the corresponding effective annual doses calculated for the inhabitants were in the range between 0.6 and 33.4 mSv. As some of the values are rather high and exceed the recommended recommendations by IAEA and ICRP, based on the local conditions a number of simple recommendations has been proposed for the possible reduction of effective annual dose caused by radon in the measurement locations.

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PAR 1 / **467**

Radionuclides for theranostic applications

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Theranostic medicine is a new integrated therapeutic system which can diagnose, deliver targeted therapy and monitor the response to therapy. Many of the "neutron rich"radionuclides that emit both imaging photons as well as therapeutic beta particles and which would be potentially excellent choices for metabolic radiotherapy and theranostic applications are produced by nuclear reactor with a very low specific activity - AS. In selected cases they can be produced by bombardment of targets by charged particle beams, in No Carrier Added Form –NCA - with very high AS. In particular if the irradiations are made with deuteron beams some more indisputable advantages are obtained: higher cross sections; more radionuclide of interest production with a lower amount of the bombarding target; lower price of the target itself; advantage in the radiochemical separation because the dissolution of the target, the separation of the radionuclides are simpler, the amount of waste is lower and the radioprotection implications as for the personnel as for the discharge are contained.
At the Radiochemistry Laboratory of LASA, a wide range of high specific activity accelerator-produced radionuclides have been produced since the 70-ties in NCA form. In particular the results about the production of Re-186g and Lu-177g, Pd-103 and Zr-89 by deuteron irradiations will be presented. These radionuclides can be used for the new emerging branch of Nuclear Medicine related to theranostic nanomedicine: liposomes, dendrimers, quantum dots, iron oxide, nanomicelles, perflurocarbon and carbon nanotubes can be used as carriers for the development of radionanomedicines.

PAR 1 / 543

Flexible irradiation facility for the production of radionuclides

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Reactor production of radionuclides is well established. However, more and more new, unconventional production routes are being requested as consequence of new applications in nuclear medicine. In order to be able to fulfil new irradiation requests it is important to design flexible irradiation facilities. Here, we present new irradiation facility with built in flexibility allowing tuning the radiation field according to the desired application. The flexibility is built in using three shielding blocks that can be composed of different materials and can be exchanged if necessary. We will illustrate the idea of this facility using lead as a shielding block, which has been used to reduce gamma flux while keeping the neutron flux unchanged. The lead variant of the facility has been used to increase the specific activity of Ho-166 polymeric microspheres used in liver cancer treatment. These microspheres can currently be irradiated for a limited time since they get damaged by the present in the reactor pool gamma photons, limiting the amount of radioactivity produced. Utilizing the new facility we show at least 4-fold increase of the 166Ho specific activity without damaging the polymeric material.

RER 2 / 681

Biogeochemical factors of Uranium and Technetium migration in subsurface water near to RW repository

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Biogeochemical factors of Uranium and Technetium migration in subsurface water near to RW repository
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The report presents our data on the impact of microbiological factors on the behavior of uranium and technetium in the upper aquifers (10-15 m) near to the radioactive waste surface repository (Russia, Tomsk region). Pollution of groundwater continued for 50 years, and now, in addition to radionuclides, the concentration of nitrate ions reached 5000 mg/L. Nitrate ions and some dissolved oxygen lead to high values of redox potential, forming conditions for high mobility of uranium and technetium in higher oxidation states.
In the samples of ground water an active microbial community capable of using nitrate, uranyl, pertechnetate ions for cell respiration was found. Treatment of microflora with organic substrates

in the laboratory modeling tests has led to the microbial processes intensification followed with consumption of nitrate ions to molecular nitrogen, reduction of pertechnetate and uranyl ions to less soluble species. The results of geochemical modeling have shown that the immobilization of technetium in the environmental occurs mainly in the form of a tetravalent oxide and in a mixture with a biogenic sulfide. Uranium was immobilized as uraninite. Biogenic sulfide ions can play the role of an antioxidant buffer when new portions of the oxidant enter the system.
The process of formation of polysaccharide microbial biofilms on the host sand horizons was studied. It was found that in some cases microbial biofilms supported on the surface of minerals and rocks can increase their sorption capacity for uranium and technetium.

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PAR 1 / 480

Technetium isotopes for electron tracking Compton camera imaging

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In this study, we produced Tc isotopes with high energy gamma emitting for developing of Compton camera imaging instead of SPECT measurement with 99mTc. Technetium-99m is used in radioactive diagnostic tests, for example as a radioactive tracer that a nuclear medical gamma camera can detect in human body. It is well suited to the role because it emits readily detectable 141 keV gamma rays, and its half-life is 6.01 h. There are at least 31 commonly used radiopharmaceuticals based on 99mTc for imaging and functional studies of the brain, myocardium, thyroid, lungs, liver, gallbladder, kidneys, skeleton, blood, and tumors. In medical diagnostic test, ▯-rays emit from 99mTc-labelled compound in human body are measured with Single-photon emission computed tomography (SPECT). The SPECT can achieve high position resolution using pine-hole collimator. In the SPECT measurement, the use of collimator deduces detection efficiency of gamma-rays from 99mTc remarkably. In recent years, the Compton camera which is originally developed for the astrophysical studies was applied for medical diagnostic usage. Compton camera imaging can realize high position resolution without collimator. Because of no collimator using, the Compton camera makes higher gamma-ray detection efficiency. Compared with SPECT with 99mTc, the Compton camera imaging technique can be expected that radiation exposure deduce to 10-20%. For the Compton camera imaging require technetium isotopes emitting higher energy gamma-rays. Two technetium isotopes, 95mTc and 96Tc are candidates for Compton camera imaging. The feasibility study of Compton camera imaging using gamma rays emitted from 95mTc and 96Tc was carried out. Using the ETCC (Electron Tracking Compton Camera) at Kyoto University, we obtained imaging pictures of the gamma rays emitted from 95mTc. Also recycling method of enriched Molybdenum oxide target developed in this study and the possibility of these Tc isotopes, 95m, 96Tc, of substitution for 99mTc in diagnostic will be discussed in the conference.

RER 2 / 438

Artificial Groundwater Recharge Process Study By Natural Isotopes Tracers

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The interaction between groundwater and surface water was important to know groundwater recharge process of the artificial recharge system. To understand recharge process, natural isotopes such as 222Rn and 87Sr/86Sr ratio variation were used. And also these tracers used to identify groundwater mixing phenomena for the artificial groundwater recharge of the water curtain greenhouse system. The groundwater was sampled from three monitoring well for every month from December 2013 to March 2016. And elements, 222Rn and 87Sr/86Sr ratio variation were analyzed. The concentrations of Fe, Mn, Si, F were varied during the water curtain cultivation period due to the surface water intrusion. And the concentration of 222Rn was decreased when water curtain cultivation and artificial groundwater recharge were started and slowly increased after water curtain cultivation ended. The concentration of 222Rn was changed from 400 pCi/L to 2500 pCi/L according to the well. The 87Sr/86Sr ratio variation of the three monitoring well showed different appearance. Among them OB-12 and OB-14 showed similar variation during artificial groundwater recharge period but OB-13 showed different ratio variation. This means groundwater-surface water mixing was occurred by different groundwater route.

PAR 1 / 550

^{99}Mo production from α-induced reaction on ^{96}Zr

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Diagnostic imaging using ^{99m}Tc ($T_{1/2}$ = 6.0 h), which is produced from the decay of ^{99}Mo ($T_{1/2}$ = 66 h), is relying on the production of ^{99}Mo from nuclear reactors to be shutdown. Alternative routes to produce the ^{99}Mo and ^{99m}Tc isotopes are being investigated. One of the reactions to create ^{99}Mo is the ^{96}Zr(α,n)^{99}Mo reaction. There are several experimental cross section datasets measured earlier, which are different from the TENDL data in the peak region of the excitation function of the reaction. To evaluate the production yield of the ^{99}Mo isotope it is very important to provide reliable and consistent cross section data. Therefore, we performed two experiments to measure the cross sections the ^{96}Zr(α,n)^{99}Mo reaction.

Two stacked targets were created from thin Zr foils with natural isotopic composition (10 μm and 20 μm) and Ti foils (5 μm) for monitoring the beam parameters. Both stacked targets were irradiated by 50 MeV alpha beam with intensity of about 200 pnA for 2 hours at the RIKEN AVF cyclotron. No chemical separation was applied. Cross sections of the ^{96}Zr(α,n)^{99}Mo reaction were derived by measuring the γ rays from decay of ^{99}Mo using a high resolution, low background γ-spectrometer based on a HPGe detector.

The measured activation cross sections for formation of ^{99}Mo are different from the available experimental data measured earlier in different studies. According to the newly measured excitation function the peak is located at around 14 MeV, which is lower than those for the literature data. On the contrary, the amplitude of the excitation function measured in this work is higher than those of the earlier studies.

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Production of cosmogenic radionuclides in Ge spectrometers placed in deep underground: a comparison of measurements with Monte-Carlo simulations

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Environmental and rare nuclear physics experiments require operation of Ge spectrometers in deep underground laboratories. Cosmic-ray background of Ge gamma-spectrometers is decreased to such low-levels that observation of cosmogenic radionuclides produced e.g. in germanium (^{68}Ge, ^{60}Co, ^{65}Zn, ^{54}Mn) and copper (^{54}Mn, ^{57}Co, ^{60}Co, ^{58}Co) is possible. There are several ways of activation of construction materials by cosmic rays:

1. during production of Ge crystals and cryostats in the factory and their transport to an underground laboratory all the construction materials are irradiated by secondary cosmic rays (nucleons, muons)
2. after installation of the Ge spectrometer in a deep underground laboratory the cosmogenic radionuclides produced at sea level (mainly the short-lived ones) are decaying
3. new generation of cosmogenic radionuclides may be produced even at deep underground by penetrating muons and their secondaries, as well as by neutrons originating from radioactive contamination of Ge spectrometer construction materials, detector shielding, and surrounding environment.

In thiswork wepresent experimental background gamma-ray spectra of the large-volume OBELIX Ge spectrometer operating in the deepest underground laboratory in Europe (LaboratoireSouterrain de Modane, 4,800 m w.e) just after its installation and several years later, and compare them with Monte Carlo simulations. The background simulation model is based on GEANT4 software package.

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Concentrating of fission 99Mo from very low enriched uranium by extraction with higher hydroxamic acids solutions in alcohols C8-C10

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99mTc is very important for medical practice, and efforts to develop the best procedure for the production of its parent 99Mo for the subsequent use in 99mTc generators are being made for more than four decades. For this purpose, a composite ceramic target containing HEU dioxide and a filler metallic powder (Al, Mg, etc.) has been irradiated in a high-flux reactor to obtain concentrate solu-tion for fission Mo recovery by sorption, extraction or precipitation. However, this method has not been recommended by IAEA because of nuclear nonproliferation. Another option is to use irradia-tion of LEU dioxide without filler in commercial-level fluxes and to rise 99Mo concentrating capabil-ity as an alternative method to 98Mo or 100Mo irradiation.

Extraction of Mo, U and certain FP from HNO3 solutions with 0.2% solutions of higher hy-droxamic acids (HA) in alcohols poorly soluble in water has been studied for 99Mo concentrate pro-duction from solutions of different enriched U targets, including those of 3% 235UO2.

The process chemistry of Mo has been supposed for both steps of Mo interphase transfer. Mo extraction by the solvent containing caprinohydroxamic acid (CHA) or laurylhydroxamic acid (LHA), is a slow process limited by first-order reaction in aqueous phase.

The DMo decreases with increase of HNO3 concentration which is typical of cation-exchange processes. The log–log plot of DMo vs. total CHA or LHA concentration is slightly S-shaped, but it is linear with the slope of 2.0 for DMo after deduction of Mo extraction by alcohol vs. free HA concentration. DMo in the case of CHA is near indifferent for alcohol content, but it has the slope of 1.0 for LHA.

Mo backwashing has been performed by interphase autocatalytic thermo-chemical HA oxidiz-ing destruction with 5 mol/L HNO3 at 95 oC in combination with Mo complexing in aqueous phase by a salt-free reagents. Alkaline scrubbing has been chosen for final regeneration of the alcohol.

The rig trials included dissolution of U-Al model or real targets in 8 mol/L HNO3 containing 0.2 g/L Hg and 0.2 g/L HF at 95 oC, allowing further I2 and Ru compounds air stripping. The fur-ther concentrating process was tested in counter-current and batch variants and the latter was found to be rather effective.

Mo extraction recovery was performed using 27 mmol/L CHA in 20% n-decanol with Isopar-M in 3 steps: extraction, scrubbing and backwashing in the vessels of decreasing volume. The simulate feed contained, mol/L: HNO3 -1.2; Al –1.2; Fe - 5·10-3; U –0.11; Hg - 1·10-3, 239Pu –1.4·10-4, Mo – 3.2·10-5, as well as 15 MBq/L 99Mo, 5.2 MBq/L 125I and 4.1 MBq/L 239Np.

The achieved total concentrating factor was 180 at process duration 2 h. Mo decontamination factors were for: U ~ 1.5·106, 125I ~ 850, 239Pu > 105, 239Np > 106, Al > 106, Fe - 4.6·104, Hg ~ 2·104.

The feasibility study has indicated that the compact extraction flowsheet and simple batch equipment are suitable for profitable 99Mo recovery from standard 3-5% 235UO2 targets. Final Mo decontamination for Tc generator production can be performed by sorption and/or by sublimation.

Patents RU:
1. 2 522 544 (2014);
2. 2 575 028 (2016);
3. 2 624 920 (2017)

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Monte Carlo simulation of environmental background sources of a HPGe detector operating in Modane underground laboratory

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Low-level gamma-spectrometers have been used in wide range of measurements from environmental radioactivity monitoring to fundamental nuclear physics. HPGe gamma-spectrometers play important role in radioactivity monitoring and material screening for experiments searching for rare nuclear events, especially in underground experiments. In general, three components of background can be observed: a radioactive contamination of detectors construction parts, cosmic-ray component and environmental radioactivity. While in surface laboratories the cosmic ray component plays the main part of the background, in underground laboratories natural radioactivity of construction materials of HPGe spectrometers (e.g. cryostat material, detector holder, cold finger) has been identified as the dominant components of the detector background. In order to understand origin of induced radioactive background, or to evaluate the background before the detector construction, a simulated background spectrum can be obtained with Monte Carlo simulation. In investigation of different radionuclide contaminants, decay events are simulated for different parts of the detector separately. Under investigation are gamma-rays of different origin: radioactivity of laboratory and surrounding rocks that mostly includes the natural radioactive series (^{238}U and ^{232}Th) and ^{40}K, and neutron capture and muon bremsstrahlung in rocks. The paper will discuss in detail sources of environmental background of a HPGe spectrometer operating in Modane underground laboratory and compare them with Monte Carlo simulations carried out using GEANT4 package.

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Production cross section measurement of alpha induced reaction on natYb to produce medical RI 177Lu

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Theranostics, which combines therapy and diagnosis, is one of hot topics in nuclear medicine. Theranostic RI to provide such new medical treatment have been investigated. We focus on ^{177}Lu ($T_{1/2}$ = 6.6 d), which is one of candidates of theranostic RI since it emits both beta particles and gamma-rays, which are suitable for therapy and diagnosis, respectively. However, the production route of ^{177}Lu is not established yet based on nuclear reaction data. It is important to measure and evaluate nuclear reaction data of ^{177}Lu. In some existing production routes, we performed an experiment on the alpha induced reaction on ^{nat}Yb. There is one data set for this reaction up to 40 MeV earlier, but a peak position is not clear. Therefore, we performed an experiment up to 50 MeV to evaluate and verify earlier data.
Our experiment was performed at the AVF cyclotron of the RIKEN RI Beam Factory. The metallic foils of ^{nat}Yb (purity: 99%) and ^{nat}Ti (purity: 99.6%) were stacked as a target. The target foils were cut into 8x8 mm2 foils from three ^{nat}Yb foils and one ^{nat}Ti foil. Their average thicknesses were found to be 16.43, 16.15, 16.93 and 2.40 mg/cm^2 by measuring their surface and weight. The stacked target was irradiated by a 50 MeV alpha particle beam with an intensity of 207 particle nA for 2 hours. After 3 days cooling time to decay out ^{177}Yb, the parent of ^{177}Lu, each target foil was measured by gamma-ray spectrometry with a HPGe detector. A decay gamma-ray (208.keV (10.36%)) from ^{177}Lu was used to derive its cross sections.
Our result is different from both previous data and TENDL-2015 data.
To observe the peak position of this reaction, we need higher incident energy.

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Neutrons and gamma-rays as environmental sources of HPGe detectors background operating underground

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Backgrounds induced by neutrons and gamma-rays are important background components for all experiments looking for rare events, such as dark matter interactions or neutrinoless ββ decay. Neutrons and gamma-rays can be produced by natural radioactivity, via spontaneous fission or (α, n) reactions and by interactions of cosmic rays. Predicting all background components correctly is crucial for designing efficient shielding and applying appropriate event-rejection strategies.
In deep underground laboratories only the hard component of cosmic rays contribute to background. Merely muons and neutrinos are able to penetrate so deeply. Next significant background component is radioactive contamination of detector construction materials and of the laboratory.
In order to investigate neutron induced-background, interactions of neutrons with a Ge detector were studied experimentally as the first step. Monte Carlo simulations using the GEANT4 simulation tool were carried out next for surface and deep underground laboratory.
The principal energy deposition mechanisms of neutrons with energies up to 11 MeV in the Ge detector are elastic and inelastic scattering. The dominant process for slow and thermal neutrons is the neutron capture, for fast neutrons the dominant processes are elastic and inelastic scattering. At neutron energies from 3.5 to 4.5 MeV, the inelastic scattering is the most probable interaction of neutrons with all naturally occurring germanium isotopes. This process is of interest for the background induction by fast neutrons, as indicated by cross sections of these reactions.
Detailed analysis of both, experimental and simulated spectra was carried out. Elastic and inelastic scattering of fast neutrons were observed, as well as their capture. A typical feature of neutron interactions with a Ge crystal of the detector are triangular gamma-ray peaks. Such peaks were clearly observed in experimental spectrum obtained from a surface laboratory as well as in the simulation. Whereas, inelastic scattering will always contribute to the background of Ge detectors in the energy region of interest, Ge peaks are observed also in the background spectrum measured in deep underground laboratory and in the simulation, too. Copper and lead as well as aluminium and tin influence the spectrum strongly. Interactions of neutrons with these materials produce many gamma-lines visible in the spectrum and become a background source, which can hide or imitate the signal. In underground laboratories gamma-rays from radioactive contamination of material surrounding the detector as well as from contamination of construction parts of the detector itself contribute to background.
The experimental results were compared with GEANT4 simulation. Precise geometry of the setup was coded including individual material impurities. The simulated spectrum reproduces the main features of the measured spectrum fairly well considering the complexity of the interactions. Integral count rates were compared and the agreement of 98% was achieved.
The results of this work showed that the GEANT4 simulation toolkit is appropriate tool for estimation of the neutron and gamma-ray background components.

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MEET-CINCH A Modular European Education and Training Concept In Nuclear and radioCHemistry

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In order to maintain European nuclear operations, expertise in nuclear and radiochemistry (NRC) is of strategic relevance. NRC contains key knowledge and techniques needed by a modern society and is certainly needed in addressing many societal challenges.1 The MEET-CINCH project is the third CINCH-based project aiming on cooperation in education in nuclear chemistry and radiochemistry. In the first two projects, CINCH and CINCH-II, status quo in NRC education at European universities was assessed, minimum requirements for bachelor, master and postgraduate programs to achieve approved NRC curricula were defined, and a number of theoretical and practical courses were developed using hands-on and e-learning approaches and platforms. The third consecutive project is designed to address the end-users in a more focused way offering platforms for immediate practical value. Building on the results of the previous projects, MEET-CINCH will counteract the massive lack of NRC expertise by three actions. A teaching package for high schools and a MOOC on NRC for the general public are built in order to attract young persons to the NRC field and convey them its fascination and relevance. Two additional actions focus on vocational training and (university) education. MEET-CINCH will develop new education and training approaches based on remote teaching and the flipped classroom concept further developing material generated in the previous projects, such as the NucWik platform (https://nucwik.wikispaces.com/) and the remote controlled RoboLab experiments (https://nucwik.wikispaces.com/RoboLab+Exercises). MEET-CINCH will provide ECVET course modules in an e-Shop adapted to the needs of end-users. After the end of MEET-CINCH the e-shop will be continuously operated by The European Network on Nuclear and Radiochemistry Education and Training (NRC-network, http://nrc-network.org/) as part of a sustainable European Fission Training Scheme (EFTS).

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The consortium includes 13 partners from ten European member states; both academia and nuclear laboratories are represented. Networking on national and European level will be an important part of the project, facilitated by having ENEN (European Nuclear Education Network) as one of the partners and by having structural links with other European associations such as the EuCheMS Nuclear and Radiochemistry Division and the NRC-Network.

To meet the objectives of the project, the proposed activities have been organised into three technical and two managerial work-packages that closely copy the project pillars listed above. Each of the work-packages is further subdivided into several Tasks. The technical WPs are:

- WP1: Nuclear Awareness and Dissemination
- WP2: Sustainability and Evolutionary Development of VET tools
- WP3: Novel Education and Training Approaches (Flipped Classroom)

These activities are supported by Euratom under a Horizon 2020 project No. 754972 (Call NFRP 2016-2017-1).

1. C. WALTHER, Angew. Chem. Int. Ed., 55, 9102 (2016).

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Analysis of environmental radionuclides at ultra-low levels: Limitations for underground experiments on rare nuclear processes

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Accelerator mass spectrometry, inductively coupled plasma mass spectrometry, neutron activation analysis and underground gamma-ray spectrometry have been investigated for ultra-sensitive analysis of long-lived radionuclides (^{238}U and ^{232}Th) in isotope sources and in construction materials designed for underground studies of rare nuclear processes and decays. Specific features of each method are compared with the aim to optimize analytical conditions for different types of samples. Detection limits below 1 nBq/g are feasible, depending on the analytical method and sample matrix. Analytical results obtained with various methods for different samples are presented, and limitations of presently available technologies and plans for future developments are discussed.

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Future Perspectives of Nuclear and Radiochemistry Education in the New Era of the Silk Road According to Chinese Nuclear Energy Development

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The training of qualified workers through the peaceful development along the New Silk Road is one of our tasks, since our Lab. is located in the capital of Gansu province, Lanzhou, which is now as then located at the Silk Road and closely related to it's development. But, what does nuclear mean? This question can be answered in different ways. In the past, nuclear meant armament, threats and similar things. But in the future, nuclear will stand only for peaceful and sustainable energy. Therefore, I will look about this issue from the perspective of the development of the Silk Road. In Chinese writing, there are two characters which sound alike, but mean different things. But the fact, that they sound alike, demonstrates that they are somehow related, anyway. So, for me, in Chinese language, nuclear energy means peace through development. And that has been, after all, the fundamental idea of the Silk Road in the past, and that is what it will be in the future.

I. CHINA'S FUTURE PERSPECTIVE

There are presently 35 nuclear power plants at 11 locations. Another 28 Gigawatts are under construction, and until 2020, 58 Gigawatts will be online. And, they shall not be built along the eastern coast of China, but more and more in the interior parts of China. In this endeavour, all work is aimed at an peaceful and sustainable development of nuclear power in China. Thus, the percentage of nuclear power in the production of electricity will be expanded to 4% by 2020, to 10% by 2035 and, together will renewable energy, to 30%, by 2050. To reach this aim, China needs to build and develop more nuclear power plants.

But, first of all, I want to show why we did choose nuclear energy:
- because we have to re-evaluate, for a number of reasons, coal as an energy source;
- because we do not want to create a conflict, between economic growth and the environment;
- because we will be more able to even out energy prices;
- because we want to overcome the limitations of other energy sources in regards to the availability of resources and their compatibility with the environment;
–because the development of nuclear energy serves to boost productive powers, especially in the area of nuclear power technology.

The challenges we are confronted with, in this endeavour, are
- the safety of the installations;
- the protection of the installations against terrorism as well as the emission of dangerous substances;
- the handling of radiation products;
- economic competitivity;
–as well as sustainability.

But, aside from these general challenges we are confronted with, there are some challenges that are specific for China. Among these, there are:
- the availability of nuclear materials and resources in this area;
- competitivity in the construction and installation of nuclear power plants;
- our own capacities for innovation in nuclear technologies;
- the construction of nuclear power plants in the interior parts of China;
–and the training of skilled workers in the area of nuclear technology and nuclear chemistry.

This last challenge for China, concerning the training of skilled workers in the area of nuclear and nuclear and radiochemistry, is a task of our university of the city of Lanzhou, which is located at the Silk Road. How important this area is for China, today, is demonstrated by a look at the past, the present and the future of nuclear and radiochemistry in China.
1895-1942 - The first steps: founding of radiochemistry education and research in China;
1942-1986: Golden Era: construction and use of nuclear technicians;
1986-2000: Decay;
2000-2011 Renaissance: of nuclear technicians as well as nuclear and radiochemistry;
2011-?: Fukushima After the incident in Fukushima, it was, at first, not clear how China would proceed, and if China would stick to its plans. Therefore, there were intensive discussions among specialists. They arrived at the conclusion that China will continue to develop its nuclear energy.

II. CURRENT TRAINING IN NUCLEAR AND RADIOCHEMISTRY

While in the last century, our universities hesitated and in some cases stopped some nuclear programs, we have now restarted programs in the areas of radiochemistry, nuclear technology, nuclear physics, nuclear medicine etc. Similar developments occur at a number of institutions outside of universities. We have to recognize, that the number of nuclear workers needed depends on how many nuclear power plants are in use.

Therefore, it is clear that a faster development of nuclear sciences and radiochemistry will enlarges the labour market for skilled workers, there will be a greater demand for skilled workers, and this creates a challenge, weather we will be able provide for this demand. The areas of skills that are affected by this are planning, construction and running of nuclear power plants and the nuclear fuel cycle. If China is to provide for this demand, this would require that the number of skilled nuclear workers rises to 25.000 until 2020.

But not only in nuclear technology and nuclear physics, chemistry, we will need skilled workers, because of these development plans, but also in areas like finances, law and protection of the environment.

Thus, the challenge is:
- to provide for the needed skilled workers;
- the lack of teachers;
- laboratories are too old and badly equipped;
–there is not enough done to attract students.

Political decisions can liberate all subordinated institutions. We will need decisions by the People's Congress, to provide for a long-term financing for these programs.

Progress that has already been made:
- The government has decided in favour of a sustainable financing for training programs in the areas of nuclear technology and nuclear physics;
- companies get involved and share their facilities with universities;

–universities work to renew their departments and equipments.

III. CONCLUSIONS

Concluding, I can say that the peaceful use of nuclear energy along the Silk Road will enhance the training of skilled workers in the areas of nuclear and radiochemistry. Chinese people love and hope to set up a peaceful world. We should ensure peaceful use of nuclear energy. We hope everyone in the world enjoy the peaceful development.

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CORONA Academy - regional training center for VVER competences

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The main objective of the CORONA Academy is to enhance the safety of nuclear installations through further improvement of the training capabilities aimed at building up the necessary personnel competencies. CORONA II project is an international project funded by the EC which has all its support to achieve a real and effective implementation. Specific objective of the proposed CORONA II project is to proceed with the development of state-of-the-art regional training center for VVER competence being viable solution for supporting transnational mobility and lifelong learning amongst VVER operating countries. The proposed CORONA Academy will maintain the nuclear expertise by gathering the existing and generating new knowledge in the VVER area. The selected form of the CORONA Academy, together with the online availability of the training opportunities will allow trainees from different locations to access the needed knowledge on demand. The selected courses will cover the whole range of training of VVER specialists from the university until reaching high professional skills and competences in the area. The CORONA Academy will meet the social goals of EURATOM program by providing training and source of knowledge also for non-nuclear specialists including school teachers, journalists and government officials.

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Determination of ultra-trace levels of uranium and thorium in electrolytic copper by radiochemical neutron activation analysis

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Significant improvements in construction of Ge detectors for ultra-sensitive gamma-ray spectrometry systems located underground have been made in recent years. A special attention in the construction of the spectrometer was paid on requirement to achieve as low as possible background rates at the lines of progenies in the decay chains of U-238 and Th-232. Radiopurity of construction materials used in underground experiments has been crucial for background. Underground installations require detectors made of radioactivity free materials with minimum contamination by natural radionuclides.

The aim of the present study was determination of impurities of uranium and thorium at ultra-low levels in electrolytic cooper by using radiochemical neutron activation analysis (RNAA) via their induced nuclides 239U/239Np and 233Pa, respectively. After irradiation of cooper together with uranium and thorium standards, various separation techniques were used for their separation from the matrix. For isolation of short-lived U-239 solvent extraction in combination of TBP and toluene was used, while for separation of Np-239 and Pa-233 extraction chromatography by using TEVA and TK-400 resins were applied. A special attention was paid on the estimation of radiochemical recovery, which was determined in each sample aliquot by using U-235, Np-238 and Pa-231. For quantification of induced nuclides and tracers used in, the Ge gamma-ray spectrometry was applied. Results obtained showed that electrolytic copper contains impurities of uranium and thorium in ultra-trace levels; up to 45pg/g (550 nBq/g) for uranium and up to 80 pg/g (330 nBq/g) for thorium. The electrolytic copper contains also impurities of Ag, Au, Sb, Se and Zn as observed using the k0 INAA technique.

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INAA of elemental admixtures in carbon-based nanomaterials for battery electrodes

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Carbon-based nanomaterials (nanocarbons) exhibit exceptional mechanical, electrical, thermal, and optical properties. These materials are attractive in various fields of science and technology, including energy storage. Despite the interest in nanocarbons as electrodes for supercapacitors and batteries, additional elements present at variable quantities could influence their electrochemical response. These admixtures often originate from the production process of the nanocarbons and details on concentration is often unavailable. Absence of such information is especially bothersome in the case of expensive commercial materials. Due to the high chemical resistance of nanocarbons, elemental characterization is a challenging task, particularly if total sample decomposition and solubilization is required. Moreover, while routine digestion methods are being developed worldwide, results validation is complicated due to a lack of well-characterized reference materials for many of the known nanocarbons. Instrumental neutron activation analysis (INAA) is an excellent method to establish reference values of total mass fractions in many types of materials. Recognized as a primary method of chemical analysis, INAA has high potential for accuracy, matrix independence, and very low limits of detection for many elements. Moreover, INAA is a non-destructive method and does not require sample dissolution.

We examined six different nanocarbon samples by INAA, i.e., carbon black, expandable graphite (purified and non-purified), reduced graphene oxide, and two specimens of single walled carbon nanotubes. The test portions, each with a mass of about 30 mg, were irradiated in the LVR-15 experimental reactor (Research Centre Řež, Ltd.) at a thermal neutron fluence rate of $3\cdot10^{13}$ cm^{-2} s^{-1}. The irradiation time was 1 min or 2 hrs to be able to determine elements forming radionuclides with half-live from 2.5 min to several years. Quantification was performed by co-irradiation of a set of synthetic multielement comparators. Quality control of the results was demonstrated by analysis of the SWCNT-1 Certified Reference Material (NRC Canada) and US NIST Standard Reference Materials SRM1633b, SRM2711, and SRM1547. Comparing the results determined for these six nanocarbons highlights how different their composition truly is, even when sold as (or presumed to be) “pure” carbon materials. Mass fraction of some elements span from sub-μg g^{-1} to units of wt %. The INAA results can now be used to validate the results from more routine analytical tools. A comparison with the values determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) at KAUST for some nanocarbons is presented.

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A new blended-learning hands-on training course in nuclear chemistry

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INTRODUCTION
The lack of trained nuclear chemical specialists has been identified in all branches of nuclear industry and other areas where skills in NRC are required, such as radiopharmacy, nuclear medicine, radiation protection and radioecology, and many others. Retraining general chemistry graduates is one of the options for mitigating this problem. Hands-on Training in Nuclear Chemistry (HOT in NRC) aims at efficient delivering the basics of nuclear and radiochemistry to trainees with chemical background at Master level, who need to extend their skills and knowledge to the field of nuclear and radiochemistry. The course was developed by Department of Nuclear Chemistry, CTU in Prague in collaboration with CINCH consortium during the CINCH I and CINCH II projects (www.cinch-project.eu).
EXPERIMENTAL
The course consisted of 40 hours covering the fundamentals of nuclear chemistry followed by 40 hours of practical laboratory exercises. The course was designed flexible to cover needs of end-users with reasonable schedule and relevant materials available. Prior to attending the course, the participants had to pass distant-learning courses at CINCH Moodle related to a) general radiation protection and b) radiation protection rules and restrictions of the laboratory controlled area.
Participants had to successfully finish theoretical and radiation protection parts before entering the on-site course as well as an interview on the priniciples and procedures of each of the tasks before starting the practical/laboratory work.
RESULTS
Based on the experience and feedback obtained during the pilot and second run of the course, all the course materials were updated and finalized. All the lectures delivered during the theoretical part of the course were recorded, post-produced using the SlidesLive™ technology. All the e-learning materials and the course are available at CINCH Moodle, additional study materials at NucWik. The contents of the theoretical part includes:
Structure and properties of atomic nuclei. Classification of radionuclides. Kinetics of radioactive decay. Radioactive equilibria. Binuclear reactions. Yield of nuclear reactions. Natural radioactivity. Radioactive decay chains. Nuclear fission, fission products. Hot atoms chemistry. Szilard-Chalmers system. Radiation chemistry. Actinides and transactinides.Interaction of IR with matter (α, β, γ, neutrons). Detection of ionizing radiation (detector types, principles). Dosimetry of ionizing radiation.
The practical exercises included have been:
1. Handling of radioactive materials - work behind shielding and in glove box.
2. Preparation of solutions with required activity.
3. Contamination survey, decontamination, wipe smear samples.
4. Radionuclide generator preparation and milking. Radioactive equilibria.
5. Sample activation via neutron irradiation.
6. Decay curve measurement and deconvolution, half-life determination.
7. Gamma-ray spectrometry –calibration, efficiency, measurement.
8. Liquid-liquid extraction of uranium. Uranium specific activity, estimation of its isotopic abundance.
9. Liquid scintillation counting.

The next run of the Hands-on Training in Nuclear Chemistry is planned in June 2018 under MEET-CINCH project and will take place at Department of Nuclear Chemistry, CTU in Prague.
ACKNOWLEDGEMENT
1. CINCH-II: "Cooperation in education and training In Nuclear Chemistry"has been supported by the European Atomic Energy Community's 7th FP (EURATOM FP7 2007-2011), grant No. 605173
2. MEET-CINCH: "A Modular European Education and Training Concept In Nuclear and radioChemistry"supported by HORIZON 2020 Framework Programme for Research and Innovation, grant No. 754972.

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Sequential anion-exchange separation of ultra-trace actinides and lanthanides with an automatic system

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Plutonium and americium belong to anthropogenic elements, and these ultra-trace elements are detected from environmental samples. Origin of these elements comes from the nuclear bomb tests and the accidental release from nuclear power plants. Isotope ratio and elemental abundance of these actinides provide information on the source term and the elapsed time 1. Uranium and thorium in environmental samples also play a key role to investigate origin of the sample, dating of mineral formation, and history on mineralization. In most environmental samples, trace actinides are contained with major elements including Na, K, and Fe. The ultra-trace elements of interest have to be separated from these major elements to measure accurate isotope ratio and abundance. It is essential to separate ultra-trace Am from large amounts of lanthanides in a sample to accurately measure the Am isotope ratio. An automatic system for sequential separation of Pu, U, Th, Pb, and lanthanides for ultra-trace analysis in environmental samples was developed in our previous works 3. In this work, automatic and sequential separation of actinides including Am, Th, U, Pu, and lanthanides from other elements was achieved by using a small anion-exchange column. Americium was completely separated from lanthanides by eluting the Am with a mixture of highly pure acids including acetic acid, hydrochloric acid, and nitric acid without any special chelate reagents. This simple technique has excellent ability to separate actinides from the matrix elements including lanthanides. The detailed experimental results and optimized separation conditions will be shown in this presentation.

This work was supported by financial aids (JSPS KAKENHI) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan (Scientific Research (C): Grand No. 22550143, 25340078, 16K00592).
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EDU 1 / 671

Nuclear and Radiochemistry 2nd editon –a textbook published by Elsevier

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This book is the second edition of Nuclear and Radiochemistry first published in 2012 and aims to provide the reader with a detailed description of the basic principles and applications of nuclear and radiochemistry. Its content is based on the authors'more than 50 and 25 years of experience, respectively, as professors of nuclear and radiochemistry at both the B.Sc. and M.Sc. levels in the Imre Lajos Isotope Laboratory of the Department of Physical Chemistry at the University of Debrecen, Hungary.The second edition is improved and complemented, especially by some novel aspects of nuclear chemistry applications, mainly in industry and nuclear medicine.
Although the book contains all modern aspects of nuclear and radiochemistry, it still has a characteristic local flavor. Special attention is paid to the thermodynamics of radioisotope tracer methods and to the very diluted systems (carrier-free radioactive isotopes), to the principles of chemical processes with unsealed radioactive sources, and to the physical and mathematical aspects of radiochemistry. This approach originates from the first professor of the Isotope Laboratory, Lajos Imre, who himself was Otto Hahn's disciple and coworker.
The 480 pages of the book is divided into 14 chapters. Chapters 1–6 discuss the basic concepts of nuclear and radiochemistry and Chapters 7–14 deal with the applications of radioactivity and nuclear processes. There are separate chapters dedicated to the main branches of modern radiochemistry: nuclear medicine and nuclear power plants, including the problems of the disposal of nuclear wastes. One chapter (Chapter 10) deals with nuclear analysis (both bulk and surface analyses), including the analytical methods based on the interactions of radiation with matter.

Acknowledgement: The 2nd edition was supported by the EU and co-financed by the European Regional Development Fund under the project GINOP-2.3.2-15-2016-00008 and by the Hungarian National Research, Development, and Innovation Office (NKFIH K 120265).

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Application of HPLC-NAA for the speciation analysis of As, Fe and Se in infant formulas.

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Elemental speciation of As, Se and Fe in infant formulas is of prime importance in order to identify the sources of the specific chemical forms of trace elements. It enables to get better information about their metabolic pathways. In the case of As, it was found, that dietary exposure to inorganic As for children under 3 years old (including rice based food) is estimated to be about 2 to 3 fold that of adults. The objective of this study was to elaborate the HPLC- NAA method for speciation analysis of arsenic, selenium and iron in in infant formulas and food. In dependence on an element, appropriate extraction procedures have been used (waterbath or enzymatic). Anion-exchange (for As and Se) and size exclusion chromatography (for Fe) has been used for the separation of the individual species. Separated species, elemental standards and blank were irradiated for 1 h at thermal neutron flux of 1014 cm-2 s -1 (nuclear reactor Maria, Świerk, Poland). After appropriate cooling time (1-15 days), samples were measured by ⍰-spectrometry; the time of measurements varied between 3600 and 20000 s. Obtained results demonstrate, that using chromatographic separations coupled with NAA detection, enables to determine selected chemical forms of As, Se and Fe at low concentration level with good accuracy. It is especially important, because these elements are problematic for ICP-MS 1.

Acknowledgments: The studies were supported by the National Science Centre, in the frame of the scientific project: New analytical procedures for the determination of chosen Se, As and Fe chemical forms in infant formulas based on neutron activation analysis (No. 2014/15/B/ST4/05047).

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The European Network on Nuclear and Radiochemistry Education and Training

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The European Network on Nuclear and Radiochemistry (NRC) Education and Training 1 is a non-profit cooperative platform of mainly, but not exclusively, European universities, research organizations and national research centers providing NRC education and training. Also industrial partners and end-users either conducting training themselves or having need for respective training courses for their personnel are eligible for membership. The aims of the NRC network include:

- cooperating in NRC education and training in Europe
- promoting development of NRC education and training in Europe
- representing NRC education and training community towards other organizations and society
- promoting and organizing student and teacher exchange between partners
- organizing common courses in NRC

The NRC network was founded as outcome of the CINCH (FP7-CA-249690) and CINCH II (EC-GA No. 605 173) EU projects and is closely linked to the MEET-CINCH project (EC GA No. 754 97) which started in June 2017. From this link, open source teaching material [3,4], remote controlled experiments and (to a limited extend) travel funds can be obtained.

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Additional members to the Network are accepted by application and by approval of the general assembly. To become a member of the Network the candidate organization needs to be a relevant provider or end-user of NRC education and/or training. For the application process, evaluation criteria and an application form are available [5].

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NFC 4 / 585

Sorption and solidification of iodate

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After the Fukushima nuclear power plant accident, radionuclides containing fission products were released from the fuel debris into the cooling water. For the treatment of the contaminated cooling water, the Advanced Liquid Processing System (ALPS) facility has been operating for 7 years. Most of the cationic species of fission products are well eliminated in ALPS by sorption and/or coprecipitation. On the contrary, anionic species of the fission products are still unknown to eliminate or not. Especially iodine-129 having a high toxicity and an extremely long half-life (16M years), is present as different chemical species of iodide (I-), iodate (IO3-), and/or organo-iodine (org-I). Thus, sorption of I-129 should be studied by controlling chemical speciation. Here we report the sorption behaviors of IO3- species on the several sorbents involving activated carbon, CeO2-modified activated carbon, Ag-attached zeolite, titanate attached activated carbon, and MgO. In addition, the solidification of IO3- with alkaline-activated metakaolin (AAMK), so-called geopolymer, were studied. We measured distribution coefficients by the sorbents, leaching behavior from the I doped AAMK into a distilled water and into a sea water. We analyzed chemical species of IO3- in sorbents and AAMK by XANES using synchrotron X-ray, and position of I and chemical components present with I in the sorbents and AAMK were analyzed by SEM.

Sorption of IO3- showed that high distribution coefficients could be obtained in acid solution for activated carbon, CeO2-modified activated carbon, Ag-attached zeolite, and titanic acids attached activated carbon, and in alkaline solution for MgO. XANES analysis showed that the sorbed I was present as IO3-. SEM analysis dis not detect I within the sorbents. These results indicate that IO3- is associated by sorbents with outer sphere complexes. Almost all IO3- was solidified with AAMK. The leaching test showed that less than 1% of I in AAMK were released into distilled water and into sea water. The XANES spectrum of I in AAMK was nearly the same as that of the KIO3 standard, indicating that I is present as IO3- ion in AAMK. These results indicate that I as the chemical species of IO3- is well recovered by the sobents in acid solution, and is also well solidified in AAMK.

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Radionuclides’uptake from contaminated soil into vegetables and consequent dose assessment

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Radioactive wastes with elevated natural radionuclides content, like wastes from uranium ore processing, can be deposited relatively close to agricultural areas. It is therefore important to understand the behavior and transport of those radionuclides in the environment. In support to this knowledge, a plant pot experiment was carried out, involving three different types of vegetables (radish, savoy and rocket) grown in substrates, contaminated with different amounts of added uranium mill tailings (UMT). The experiment involved plants grown in non-contaminated substrate and four soils with various amounts of added UMT content, representing various contamination scenarios. After five months of growing period the investigated plants were harvested, and activity concentrations of U-238, Th-230, Ra-226 and Pb-210 were determined in the substrates and all plant parts, such as shoot and root systems.

Based on measurement results, soil-to-plant transfer factors, expressed in terms of concentration ratios, were calculated. The results obtained indicated significant linear correlation between the content of radionuclides in soil and plants as well as between the content of the radionuclides in plants and soil parameters.

Finally, annual effective dose due to the radionuclides ingested by consuming the studied vegetables were calculated. Different scenarios with respect to consumption and activity concentrations were taken into account. The total annual effective ingestion dose and UMT content in soils showed highly significant linear correlation. However, the assessed dose was far below the world ingestion average dose, even if assuming that the activity concentrations of Po-210 and Pb-210 in the investigated samples were in equilibrium. Nevertheless, given the fact that the dose was correlated with the UMT content, caution should be taken when consuming vegetables grown in areas close to uranium ore processing sites.

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Disposal options for molten salt reactor waste

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Experimental research on active materials often goes along with the generation of compositionally complex waste streams for which a suitable route towards safe (interim) storage is lacking. This has worldwide resulted in the accumulation of significant volumes of highly diverse types of waste. The complexity of the streams invokes the need for tailored solutions for the individual components. At different nuclear laboratories across Europe (NRG, SCK.CEN, CEA, Halden reactor), a research irradiation programme on advanced nuclear fuels, including fuel concepts as Accident Tolerant Fuels, Fast Reactor Fuels and Molten Salt Fuels, is running. Within this programme various nuclear fuels have been/are being /will be irradiated at different conditions. Research on possible reuse but mainly final disposal of the spent fuel is an important part of the new nuclear fuel concepts. A prerequisite for any route is that the waste form can be accepted by the national organizations for waste disposal.
At NRG, research on molten salt reactor (MSR) fuels is ongoing. A part of the research is dedicated to waste handling following irradiation experiments in the HFR Petten. The created spent fuel waste will be finally disposed by the national organization for waste disposal (COVRA). To get this new waste accepted, it must be first fully characterized and it is foreseen that the fluoride waste must be transformed to chemically stable and acceptable waste streams (e.g. oxides and cementable waste).
It is foreseen that if new forms of waste are offered for disposal, additional tests related to final disposal are required; the chemical stability of the immobilized waste forms, most notably cemented waste, must be tested by specific leaching experiments to meet the waste acceptance criteria.
A comprehensive literature survey has been done to summarize the possible ways of handling the MSR waste including molten salt specific challenges (such as radiolysis leading to fluorine gas formation). Based on the review, different routes have been identified for experimental cold tests. Simulated fuels will be prepared to perform "semi"hot tests to validate the procedures. The proposed and tested route for waste handling will be finally applied at NRG to irradiated and fully characterized MSR fuels.

RER 3 / 603

Effect of wet and dry processes on the gastrointestinal absorption of radiocesium adsorbed to soil particles in rats

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Radiocesium is a major radionuclide discharged into the environment as a result of the Fukushima Daiichi Nuclear Power Plant accident. Because radiocesium isotopes have relatively long physical half-lives (134Cs: 2.065 years; 137Cs: 30.04 years), they are the most significant long-term radioactive contaminants in the environment. When radiocesium is deposited on solid ground, it remains in the soil surface layer for a long time owing to its strong adsorption on soil particles. Soil aging enhances the adsorption of radiocesium on soil particles and limits its phytoavailability 1. In this study, the effects of soil aging on bioavailability were investigated by estimating the absorption of radiocesium adsorbed on soil particles in rats after wet and dry soil weathering processes.
Soil samples were dried at 45 ℃, grinded in a mortar, and passed through a 250-μm mesh. 134CsCl was added to the soil samples and dried at 40 ℃. Subsequently, wet and dry weathering processes (watering and drying at 40 ℃) were repeated up to 20 times, and each soil sample was encapsulated. Soil samples were administered to five female Wistar rats aged 8—10 weeks in capsules by intragastric cannulation. After administration, the whole-body activity of 134Cs was measured over time by a whole-body counter. 134Cs absorption was estimated using the least-squares method with a biphasic curve with fast and slow compartments.
After adding 134Cs to the first soil sample, the absorption rate of 134Cs in rats was 24 ± 6.8%. After 10 and 20 repetitions of wet and dry processes, the absorption rates decreased to 12 ± 3.1% and 1.0 ± 0.9%, respectively. These absorption rates are significantly lower than that of ionic forms such as CsCl 2, and the International Commission on Radiological Protection's recommended absorption value (absorption rate = 1.0) 3. It was also found that wet and dry processes made the adsorption of 134Cs on soil particles stronger and limited the isotope's bioavailability. Therefore, it can be inferred that when humans or animals ingest soil particles contaminated with radiocesium, the amount of time between deposition and ingestion influences the absorption rate and internal exposure dose.

This work was supported by JSPS KAKENHI Grant Number 16J10094, and by the Research on Health Effects of Radiation organized by Ministry of the Environment, Japan.

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NFC 4 / 468

Determination of mineral-specific distribution coefficients of 133Ba from thin sections of granitic rock using filmless electronic autoradiography

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The final disposal of spent nuclear fuel will be performed in Finland in a geological repository in crystalline granitic rock at a depth of 400 meters 1. The processes affecting the transport of radionuclides from the spent nuclear fuel to the biosphere need to be carefully taken into account when considering the overall long-term safety of the repository 2. Consequently, it is necessary to study the sorption and diffusion properties of safety relevant radionuclides from laboratory scale all the way up to in-situ scale in the different barriers of the repository, for instance, the bedrock. In this study the mineral-specific sorption of 133Ba was studied in thin sections of veined gneiss and pegmatitic granite obtained from the in-situ site in Olkiluoto using filmless electronic autoradiography (i.e. the BeaverTM). The thin section samples were equilibrated with 1 mL of groundwater simulant made to resemble the fracture groundwater in the Olkiluoto site after which it was replaced with 1 mL of the same groundwater simulant with added 133Ba 3. After a week, the groundwater simulant was pipetted from the thin section samples and it was measured with gamma spectrometry.

The spatial distribution of barium activity in thin sections was measured with two autoradiography methods; digital autoradiography using IP plate technique (Fuji 5100) and the BeaverTM, which is based on a gas detector incorporated by micromesh Parallel Ionization Multiplier (PIM) 4. The BeaverTM allows real-time counting of charged particle emission from the analyzed surface with high sensitivity and linearity. Methylmethacrylate standards of 133Ba were made in order to determine the counting efficiency to derive activities of the measured count rates.

The distribution coefficients of 133Ba for main minerals were determined from the thin sections with the BeaverTM measurements and from the activity decrease in solution with gamma spectrometry. The mineral-specific distribution coefficients of 133Ba were found to decrease in the order: biotite > plagioclase > potassium feldspar > quartz, which was in good agreement with previous results [5]. The BeaverTM has proved to be an efficient method for quantifying heterogeneous sorption of sorbing radionuclides on rock materials. In addition, sorption experiments on thin sections provide a way to upscale distribution coefficients from crushed to intact rock. The sorption data obtained in this study will be utilized in the heterogeneous diffusion modelling as well as in the interpretation of results from the in-situ through diffusion experiments.

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A study on distribution characteristics of 37Cs, 90Sr and Pu isotopes in the mushroom and lichen in South Korea.

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Various mushroom samples, lichens and pteridophytes are collected nationwide in 2017 and their contents of artificial radionuclides (137Cs, 238Pu, 239,240Pu and 90Sr) are measured to address the growing concerns regarding the radioactive contamination of food products in Korea.
The analysis shows that the activity concentrations of 137Cs range from minimum detectable activity (MDA) to 77 Bq/kg of fresh weight and the highest concentration was found in the Sarcodon imbricatus. The activity concentrations of 239,240Pu in the mushrooms varied from MDA to 0.13 Bq/kg of fresh weight. Among the samples, Umbilicaria escu¬lenta (lichen) showed the highest concentration. On the other hand, the activity concentration of 90Sr in the mushrooms showed the MDA to 19.6 Bq/kg of fresh weight, the highest concentration of 90Sr was found in the Phellinus linteus.
The activity ratio ratios of 238Pu/239,240Pu vary from 0.023 to 0.038, with a mean of 0.028. This ratio is consistent to the global fallout ratio (0.020-0.030) within 2σ. In addition to the activity ratio of 238Pu/239,240Pu, the atom ratio of 240Pu/239Pu will be reported later as a supplement to identify the sources.

NFC 4 / 584

Profile analysis of Aare granite samples after radionuclide migration

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Several laboratory through-diffusion experiments with HTO, ³⁶Cl, selenate, ²²Na, ⁸⁵Sr, ¹³⁴Cs, and ¹³³Ba were performed on samples from experimental and monitoring borehole of the Long Term Diffusion Phase III experiment in Grimsel Test Site. Salinity dependent cation excess and anion exclusion effects were observed. Anion effective diffusivities were almost half of water effective diffusivity. Na and Sr effective diffusivities were 1.5 and 5.5 times higher than water effective diffusivity, respectively. Heterogeneous character of granite diffusion path was observed experimenting with Cs and Ba.
Using numerical modeling, unknown parameters of porosity, geometrical factor, and distribution coefficient can be evaluated from the break-through and depletion curves. The associated uncertainty of diffusion parameters might be thus significant. It might be reduced by obtaining tracer concentration profile in the rock sample. We applied two experimental procedures for profile analysis. The first consisted in sawing the sample on thin slices using Struers Secotom Hot Cell saw and diamond cut-off wheel. The second procedure was an abrasive technique commonly used for cementitious materials and argillaceous rocks. Instead of grinding paper, artificial corundum (Al₂O₃:B80 and Al₂O₃:B150) mixed with cooling water was used. Benefits and drawbacks of both methodologies will be discussed. The profit of having the concentration profile, as the third dataset from the through-diffusion experiments, will be demonstrated, presenting Na, Sr, and Cs experiments.

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Vertical Migration of Radionuclides in Soil Samples from Pripyat

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The accident of Chernobyl led to significant contamination in large regions of Europe, particularly in Ukraine, Belarus and Russia. In the Chernobyl exclusion zone (CEZ), which includes a 2200 km^2 large area around the Chernobyl nuclear power plant, the highest amount of radioactivity was distributed. The decrease of surface activity concentrations is governed not only by physical decay but also by vertical migration. In this study the vertical distribution of ^{137}Cs, ^{90}Sr, ^{241}Am and plutonium in upper soil layers from the exclusion zone was investigated to get information on the retention mechanisms of these nuclides. Therefore, several drill cores of 30 cm length were collected in the city of Pripyat, which is about 4 km away from the nuclear power plant. Each drill core was cut into 10 layers of different thicknesses. Due to their half-lifes ^{137}Cs ($T_{1/2}$=30,17 a) and ^{90}Sr ($T_{1/2}$=28,78 a) still dominate the contamination in this region. Because of their relevance to long-term considerations the different isotopes of plutonium (^{238}Pu, ^{239}Pu, ^{240}Pu) as well as ^{241}Am being the daughter of ^{241}Pu were also investigated.

Methods and Measurements

In the first step, each sample was analyzed by γ-spectrometry to determine the amount of ^{137}Cs and ^{241}Am by their characteristic γ-lines (^{137}Cs: E=661,7 keV; ^{241}Am: E=59,54 keV). In the next steps, the soil samples have been ashed and plutonium and strontium were chemically separated. For the detection of plutonium α-spectrometry was used. The ^{90}Sr measurements were performed by liquid scintillation counting (LSC).

Results

Figure 1 shows the specific activity of ^{241}Am, ^{137}Cs, ^{90}Sr and the three plutonium isotopes in different depths of one drill core. While the absolute specific activities in layers of equal depth differ considerably, the decrease follows similar slopes in all cores and all nuclide below the 15 cm. Each nuclide is detected down to 30 cm depth.

Figure 1: Specific activities in soil layers.

Important information on the different migration behavior is obtained from the specific activity ratios of different nuclides (Figure 2).

Figure 2: Ratio of ^{241}Am and $^{239+240}$Pu activities for two cores.

From the decrease in the ratio of ^{241}Am and $^{239+240}$Pu activities migration rates of plutonium and americium are deduced by a simple model.

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In-situ migration experiment with radionuclides in granitic rock (Josef gallery, Czech Republic)

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Crystalline rocks are being considered as potential host rocks for the construction of deep geological repositories (DGR) for radioactive waste in a number of European countries including the Czech Republic. The aim of the project (PAMIRE) was to determine the degree of reproducibility the laboratory determination of tracer transport processes of the tracers compared to the results of the the larger scale experiments (dm scale); subsequently to implement it into the in-situ granitic real conditions of the and in the models evaluating the radionuclide migration into the rock.
The work consisted of laboratory experiments on cm and dm samples (diffusion 3H, 36Cl, 125I, electromigration experiments 131I, sorption, visualization of pore space etc), in-situ experiments and modeling. Laboratory experiments at various scales have confirmed that the diffusion migration in the rock of a given type (tonalite) uses only part of the pore space, being is available for migrating radionuclides. The presence of cracks, although visually completely healed, leads to the formation

of preferential paths, which then lead to an increase in diffusion coefficients of migrating radionuclides. On the basis of the evaluated data, a concept of pore space of samples of granitic material was proposed.
In-situ experiments were carried out in the Josef underground laboratory (CZ). Due to the significantly damaged rock environment in the Josef underground laboratory, the project focused on a rock environment with the presence of fissures (filled/opened) and a description of its behavior with regard to the potential migration of radionuclides. Advection is considered as a main processes within such a type of test.
A system consisting of a flow field intersecting a water bearing fracture, was first identified at the Josef Underground Laboratory. Here two boreholes were drilled and fitted with instrumentation so as to determine and characterise an appropriate location for the potential injection of a radioactive tracer into the fractured rock massif. A number of methods were employed in order to describe the rock system in niche JP-57, fissure system and the hydraulic flow field in detail prior to tracer injection. Detailed system description included thorough core descriptions, fracture determination, Optical Borehole Imaging (OPTV), Acoustic Borehole Imaging (HRTV), tracer dilution tests, hydrochemistry monitoring and hydraulic tests. The pressure levels were monitored in the multipacker system. The pressure responses of the system were tracked during tracer tests and the flow from the selected intervals were measured.

Based on the findings and results, the tracer experiment instrumentation was developed, focusing namely on the maximum accuracy of the tests, the measurement parameters and the setting of the tests. In 2017, 21 tracking tests were performed and 33 penetration curves were evaluated to be used conservative tracers (NaCl, KI). The aim of the tests was to thoroughly test the rock environment, tracer behaviour, instrumentation and optimal setup of the test system for active experiments. Moreover, discussion with the SÚJB, the regulator, about the ensuring necessary legislative requirements for radiation safety and the implementation of active tracer tests proceeded. The GoldSim program was used to demonstrate the safety of the experiment under consideration and the Modflow-MT3D program was used to determine the hydraulic conditions in the area concerned.
After the regulator approval, two tests, using 3H (2 MBq.l-1), were performed on 22 September and 6 October 2017 as the very first pilot tracer test of such a kind in Czech Republic. Subsequently, evaluation of tracer tests and penetration curves was performed using Qtracer2. The transport model was set up in MT3DMS on the basis of a verified hydraulic model implemented in MODFLOW2000. The results of both tests were consistent and well monitored. The experimental balance (HTO) were around 95% during tracer tests, with return back rate 99%.

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RER 3 / 679

The role of nuclear forensics in determining contamination sources

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The Waste Isolation Pilot Plant (WIPP) is a radioactive waste repository operated by the U.S. Department of Energy (DOE) for the permanent disposal of defense transuranic (TRU) and TRU-mixed wastes (wastes that also have hazardous chemical components). Located near Carlsbad, New Mexico, the WIPP facility is the world's first deep geological disposal facility licensed to accept TRU waste having alpha activity > 37000 Bq/g for radioactive isotopes with atomic numbers higher than uranium and half-life greater than 20 years. Since the facility opened for operations in March 1999, WIPP has disposed of more than 91,000 m3 of TRU waste in more than 170,000 containers, allowing the remediation of 22 generator sites nationwide.

Four years ago, the WIPP experienced its first minor accident involving a radiological release. On February 14, 2014, a waste container in the repository underwent a chemical reaction that caused the container to overheat and breach, releasing some of its contents into the underground. A small amount of radioactivity escaped through the ventilation system and was detected above ground. It was the first unambiguous release from the WIPP repository. Soon after the radiation release event, the Department of Energy-Office of Environmental Management appointed an Accident Investigation Board (AIB) to determine the cause of the radiation release. The AIB used both chemical and radiological forensic analyses of the materials ejected from the drum to pinpoint the exact location and cause of the drum breach. To complement the AIB investigation, the DOE also chartered a Technical Assessment Team (TAT) of specialists to perform a comprehensive, independent scientific review of the mechanisms and chemical reactions that may have resulted in the release of radioactivity. The Carlsbad Environmental Monitoring and Research Center (CEMRC), an independent radiological monitoring organization, also performed extensive monitoring of the above ground environmental samples. The CEMRC's environmental samples and the TAT samples from repository were examined to determine isotope ratios for comparison with what was known of the potential source materials in the drum believed to be the source of contamination. The TAT, the AIB, and CEMRC interpreted the isotope ratios found as providing forensic evidence of the contamination source. This paper discusses the difficulties and uncertainties inherent in making definitive forensic claims based on the variability in the key ratios of the breached drum and the sample media collected from the WIPP underground as well as those collected outside the repository. The TAT and AIB also relied on a photographic examination of the waste in the room where the release occurred. CEMRC's analytical results independently support the TAT and AIB conclusions. Following a lengthy recovery process the facility recently resumed waste disposal operations.

SEP 3 / 574

Silica-based extractants for metal uptake and their direct conversion to a waste form

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Silica-based materials offer the opportunity for "selective decoration"where a large variety of adduct, ion exchange and chelate forming organic ligands can be attached directly to the surface. With the correct choice of ligand, in combination with high surface functionalisation, it is possible to offer high sorption capacity, fast exchange kinetics and selectivity for a variety of metal ions. The functionalised silicate backbone is also more compatible with the final waste forms utilised in the nuclear industry. For example, the metal-loaded silica could be directly converted to a glass-based waste form, currently the preferred option for high level waste disposal in the UK.

Here we report on our ongoing experiments to remove uranium, thorium, lanthanum and strontium from aqueous solutions using various silica-based resins, including bistriazinyl bipyridine silica (BTBP-Si), bistriazine silica (BT-Si), neocuproine-Si and the commercially available ethyl/butyl phosphonate silica (Phosphonics POH1). We have investigated a range of conditions, including pH, ionic strength, metal loading capacity and kinetics, which have been fitted using simple mathematical models, as well as performing column studies. We will also report on our ongoing efforts to convert the loaded silica resins into borosilicate-type glasses.

NAM 4 / 696

Rapid Method for the Determination of Actinides and 93Zr in Radioactive Waste, Biological and Environmental Samples

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A novel method based on the retention and sequential elution of all actinides on a single small DGA resin® column has been developed and published recently1. DGA resin® is an actinide selective extraction chromatographic material which contains N,N,N',N'-tetraoctyldiglycol-amide sorbed onto Amberchrom CG-71 inert support (developed by Horwitz et al.2 , available from Triskem International). According to the original procedure, nuclear power plant waste samples were wet ashed, actinides were pre-concentrated by co-precipitation, and the chromatographic procedure was optimized by changing the nature, the acidity of the eluents, the oxidation states of the actinides, the concentration of complexing agent, and the temperature. Alpha sources were prepared by micro-coprecipitation. 241Am, 239,240Pu, 238Pu, 238U, 235U, 234U were determined by alpha spectrometry, 237Np was determined by ICP-MS.
Making advantage of the extremely high distribution ratios (Dw) of all tri- and tetravalent actinides on DGA resin, the pre-concentration step was omitted and sample solutions originating from 100 mL waste concentrates were directly loaded and processed, therefore recoveries were increased, actinides were successfully determined.
Due to the high retention (Dw) of Zr on the resin, the method was extended for determination of 93Zr in radioactive wastes using ICP-MS for detection.
The method was also adopted for the analysis of environmental samples (soil, sediment). The rapid fusion technique using NaOH was applied to 5g of sample according to Maxwell et al.3 Sample solution was loaded without pre-concentration on the resin followed by the optimized sequential elution process.
The method was also adopted for the determination of actinides in 100-200 mL urine samples where not only the pre-concentration but the urine destruction was omitted.
High recoveries and decontamination factors were obtained in each alpha and ICP-MS source. The methods became much simpler and faster, typically the complete analyses of actinides and 93Zr could be performed in one day.

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SEP 3 / 577

Remediation of Radioiodine from aqueous used nuclear fuel (UNF) waste streams by Lewatit A365 and Purolite S985

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Some nuclear accidents and nuclear fuel treatment (such as reprocessing) can release radioactive iodine isotopes, principally 129I and 131I, into gaseous streams and aqueous solutions. Iodine-131

raises concerns in nuclear accidents due to its high activity and the potential for uptake into the human body but does not pose a long-term disposal risk due to its short half-life (t1/2 = 8.04 d). Iodine-129 raises concerns due to its extremely long half-life (t1/2 = 1.57 × 107 y) and high mobility in most geological environments. During aqueous reprocessing of used nuclear fuel (UNF), iodine partitions between the various gas streams and the various aqueous phases, and iodine can be released to varying degrees in almost every process within the facility. Subsequently, a more efficient method of capturing and immobilizing this iodine from various waste streams before disposal or discharge is required. Current research is looking at aqueous scrubbing of radioiodine from acidic conditions due to compatibility with aqueous based back end processes of UNF reprocessing. This research looks at the effectiveness of two commercially available anion exchange resins for aqueous iodide uptake under a variety of conditions that may be present in reprocessing. Lewatit A365 and Purolite S985 have previously never been considered for iodide uptake, therefore our study aims to determine the suitability of these resins for their industrial implementation.
pH-dependence, isotherm, kinetic and column simulation data has been obtained, allowing for a detailed comparison of performance between the two resins, using iodide ISE analysis. Purolite S985 was found to display greater uptake capacity than Lewatit A365 under all experimental conditions tested, with Langmuir model-fitting demonstrating a maximum uptake capacity of 548.9 mg g-1 in comparison to Lewatit's 435.9 mg g-1. Uptake capacities when compared to commercial applications such as silver mordenite and silver-nitrate impregnated silica and alumina are considerably greater, suggesting these ion-exchange resins could be a superior alternative. Furthermore, our studies have found that addition of iodine enhances iodide uptake by each resin which could dramatically improve uptake where both species are present such as in UNF streams.

NAM 4 / 553

Rapid Determinations of Sr-90, Am-241 and Plutoniumisotopes in Emergency Exercises

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Introduction
An exercise for emergency situations was held in the Austrian Agency for Health and Food Safety in cooperation with the federal state Carinthia. The Exercise was funded by the Federal Ministry of Sustainability and Tourism. Therefore 21 soil samples, 19 ground vegetation samples and 6 sediment samples were collected. During former emergency exercises samples were only measured by gamma spectrometry. This time 10 samples were selected after the gamma measurements to perform an exercise with radiochemical sample preparation for Sr-90, Am-241 and Plutoniumisotopes. These samples were analysed using a rapid method and afterwards by using a method for monitoring measurements.
The goals of these exercises are first to establish sample preparation methods and analyses which can be performed at once after an emergency situation with all the equipment and software needed at hand. Second to train the people which have to perform those analyses so that they can perform them "blindfolded"and third to determine the activity concentrations now present in these samples with the monitoring measurements.

Material and Methods
Internal standards were added to 4 of the 10 samples before the sample preparation. The results of the monitoring measurements were used to evaluate the results of the rapid determination. Therefore ashing, leaching and adding of the internal standards to the sample was conducted and afterwards two subsamples were created –one for the monitoring and one for the rapid determination.
The separation of the analytes for the rapid determination was conducted with Sr-Spec Resin and DGA Resin. Sr-90 was measured with Liquid Scintillation Counting and the chemical recovery was measured by Sr-86 and Sr-88 with ICP-MS. Am-241 and the Plutonium isotopes were measured with

alpha spectrometry and Pu-239 was measured with ICP-MS.
During the analyte separation for monitoring, an ion exchange chromatography and in soil samples TEWA resin was used in addition to the other resins.

Results and Discussion
12 samples can be analysed within 48 hours even by one person. If two people are conducting the separations 12 samples can be analysed within 24 hours (ashing and digestion not included). The results of the samples with internal standard addition were in consistence with the calculated value. Furthermore the results of the rapid methods were in agreement with the monitoring determination. Within two soil samples Pu-239/240 and Am-241 could be determined with the rapid method although no internal standard was added. The activity concentration of sample 7 was 1.78 ± 0.12 Bq/kg for Pu-239/240, 0.049 ± 0.012 Bq/kg for Pu-238 and 0.744 ± 0.074 Bq/kg for Am-241. The activity concentration of sample 8 was 1.54 ± 0.09 Bq/kg for Pu-230/240, 0.0364 ± 0.0062 Bq/kg for Pu-238 and 0.573 ± 0.046 for Am-241. Thus the rapid determination can be successfully used to determine samples with elevated activity concentrations compared to the current values.

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Synergy of sequential injection analysis and molecular recognition technology for low-level Sr-90 activity determination

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The routine monitoring, as a part of radiation protection from radioactive materials released into the environment, is established in many countries worldwide. A radionuclide of particular interest is Sr-90, which is, due to its long half-life, one of the most hazardous radionuclide. As it is pure beta emitter, its isolation prior determination is necessary. In recent years, materials based on solid phase extraction are used as a replacement for purification steps of conventional methods, which are mostly time consuming and labor intensive. The introduction of such materials provide partial automatization and miniaturization of separation procedures. The products based on molecular recognition technology, AnaLigSr-01 and SuperLig 620, provided by IBC Technologies, more known as materials encased in Empore™ Strontium RAD Disks, are promising materials, since it is possible in short time to separate Sr2+ from many interfering cations, which are present in much higher concentrations, such as Na+, K+, Mg2+, Ca2+ etc. The drawback of these materials is that they, besides Sr2+, sorb Pb2+ and Ba2+. These cations may interfere during measurements on available instrumentation. Therefore, within this research an extensive study is done to effectively separate strontium from these elements. The goal is to develop rapid and easy to handle method for precise determination of strontium, with detection limits below 50 mBq/L. The final results show, that by changing pH or molarity of complexing agents, pure fractions of Pb2+, Sr2+ or Ba2+ can be obtained.

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Determination of Mo-93 (and Nb-94) in nuclear decommissioning waste from a nuclear reactor

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Molybdenum is an alloying component of structural materials used in nuclear reactors. The activation product of stable Mo-92 (14.65% abundance) is Mo-93, which has a long half-life ((4.0±0.8)×10^3 years); so it is an important contributor of nuclear power plant wastes (especially dismantling wastes). As it decays by electron capture; its proper detection is very difficult. X-ray spectrometry, LSC and mass spectrometry (ICP-MS) can be used, but chemical separation is needed to separate Mo-93 from the matrix and interferences before detection. Although some methods have been reported for its determination, a more thorough isolation is required in case of nuclear decommissioning waste, where activities of Co-60, Sb-125, Zn-65 and many other radionuclides are 3-8 orders of magnitude higher, than that of Mo-93. So a newly developed method for this purpose will be presented.

Several metallic samples from a nuclear power plant were analyzed. Each sample was dissolved using aqua regia and HF, and prepared in diluted HF acid for separation.

A combined chromatographic separation procedure was applied to separate Mo from the matrix and interfering radionuclides. The majority of activity was removed using cation exchange chromatography, as the activation products of most metallic components (Mn-54, Fe-55, Co-60, Ni59, Ni-63, Zn-65) were retained on a cation exchange resin as cations. However, Mo as anion (e. g. [MoF7]-, [MoOF5]-, [MoO2F3]- or [MoO2F4]2-) passes through the cation exchange column and remains in effluent; just like anion complexes of Zr (Zr-93) and Nb (Nb-93m, Nb-94) ([ZrF6]2-, [NbF6]-, [NbOF5]2-), as well as Tc-99 ([TcO4]-).

Due to the removal of the main gamma emitters at this stage, Nb-94 in the effluent can directly be measured using gamma spectrometry.

Before measurement of Mo-93 using LSC, further separation of Mo is required from all other radionuclides, especially from the anions of Zr and Nb. The different affinities of these anions to TEVA resin were utilized to perform a nearly clean isolation of Mo from Zr and Nb; and by this procedure most of interferences can be removed.

For further purification, the separated Mo-93 was prepared in a diluted HNO3 solution, and loaded to an alumina (Al2O3) column. After rinsing with diluted HNO3, the adsorbed Mo-93 was finally eluted with ammonia solution. This solution was concentrated and prepared in water for LSC measurement after adding scintillation cocktail.

The recovery of Mo in the whole procedure was determined by measuring stable Mo in the separated solution and the initial solution using ICP-OES, and found to be more than 70 %. The decontamination factors for the key interfering radionuclides are higher than 10^5.

An internal solution of Mo-93 was prepared from a proton irradiated niobium target, and standardized using X-ray spectrometry using a HPGe detector (to determine the activity of Mo-93 due to its X-ray radiation). This solution was used to prepare quench curve of Mo-93 for the calibration of the LSC instrument, and quantitative measurement of Mo-93 in the separated sample solutions.

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The binding ability of amidoxime ligands with different substituents to uranyl

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The sequestering of uranium from the ocean is an attractive choice for clean energy due to its vast inventory (about 4.5 billion tons). In this regard, the extractant with high binding affinity to uranyl (UO22+) is desired. In the past few decades, amidoxime (AO) has been screened as the most promising candidate, and various absorption materials bearing this ligand or its derivatives have been developed and applied in both mimic seawater and field experiments. To date, sequestering uranium from ocean at the industrial level is still limited. Enhancing the binding capability of the extractant towards uranyl has always been an issue of top-priority.

In this report, we have developed a novel "one-pot"reaction for the synthesis of amidoxime compounds, which can be used for studying the substituent effects systemically. The results from electrospray ionization mass spectrometry (ESI-MS), fluorescence quenching experiments, and theoretical calculations provide consistent conclusions on the electronic effect of the substituent on AO–UO2 bonding.[1-4]

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NAM 4 / 520

The AUTORAD platform for the automated radiochemical analysis

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Innovative analytical methods for the determination of radionuclides are essential to optimize and reduce costly and time consuming sampling, and simultaneously to reduce potential radiation exposure of the operating personal. The AUTORAD platform is an automated and portable system for the separation, pre-concentration and determination of key radionuclides using flow scintillation counting. AUTORAD uses a Lab-on-Valve device coupled to a multi-syringe module, allowing detection at ultra-trace levels depending on the system configuration and counting time. It has been applied and qualified for the determination of 90Sr, an important fission product generated during irradiation of nuclear fuel, in view of radiation protection, environmental monitoring, radioecology, facility decommissioning and radioactive waste management.

SEP 3 / 567

Pre-concentration and selective separation of Ra2+ in the presence of a wide range of concurrent cations using a new developed resin

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Radium is the daughter nuclide of Naturally Occurring Radioactive Materials (NORM), uranium and thorium, which exists in several isotopes. Accordingly it incites great interest in various fields of

application such as: environmental monitoring, analytical and medical uses(Gott et al. 2016). Due to its extensive use (in food products, in heating pads and suppositories, in cosmetics, as a fertilizer …) during the early 20th century, many sites across Europe and North America have been contaminated(Harvie 1999). Therefore, various regulatory authorities have shown an interest in monitoring and enforcing limits for radium levels in drinking water and industry(Morvan et al. 2001). Radium's high mobility in the environment and its chemical similarity to calcium both explain its classification as a highly radiotoxic radioelement.
For all these reasons, the necessity to concentrate/separate radium has gained importance in scientific research academic or industrial fields. MnO2 resin or disks are the most common materials used to extract radium from ground and surface water of low Ra-activities. But these systems show some limitation, especially in salty or mineral waters, rich in calcium and magnesium. So there is a need for the development of more selective supports for radium based on Molecular Recognition Technology (MRT).
This question is at the center of this work, led by TRISKEM, which aims to develop a specific resin for a selective extraction of Ra in environmental applications. The developed resin consists of a metal-selective ligand, either chemically bonded to a silica support or impregnated onto another organic polymer support. This study characterizes the adsorption and desorption of Ra, on the newly developed Ra-resin, in the presence of high concentrations of interfering cations, especially calcium. Several French mineral waters and environmental samples have been tested to check the efficiency of the new Ra-resin compared to MnO2 disks and chelex resin. Resins were tested for selectivity and efficiency as a function of different key parameters (pH, time, Ca/Mg concentrations and Ba concentration). The results are very promising for environmental applications.

Key words: Radium, selective extraction, specific resin, concurrent cations, environment.

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TRLIF and TRLIC Laser Spectroscopy and Detection of Actinides/Lanthanides in Solutions

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This work is devoted to applications of the time-resolved laser-induced luminescence (TRLIF) spectroscopy and time-resolved laser-induced chemiluminescence (TRLIC) spectroscopy for detection of lanthanides and actinides. Pu, Np, and some U compounds do not produce direct luminescence in solutions, but when excited by laser radiation, they can induce chemiluminescence [1-4] of chemiluminogen (luminol in our experiments). It is shown that multi-photon scheme of chemiluminescence excitation makes chemiluminescence (TRLIC) not only a highly sensitive but also a highly selective tool for the detection of lanthanides/actinides.
Results of the experiments on Eu, Sm, U, Pu, and Np detection in different solutions are presented.

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NFC 5 / 436

New applications of the hot compressed water method for the chemistry of the nuclear fuel cycle

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The hot compressed water (HCW) is a remarkable reaction medium. In the nuclear field, the HCW method has been successfully applied in chemical synthesis and separations, nuclear waste treatment, studies of accelerated corrosion of cladding materials, etc. Moreover, the supercritical water (SCW) reactor is a concept of GenIV reactors under consideration.
We have employed the HCW (here pressured water above 120 oC) method in chemical reactions relevant to the nuclear fuels production. Thus, we have proved for the first time that the hydrothermal attack of the An(IV)-oxalates take place under HCW, with formation of AnO2 (An= Th, U, Np, Pu) nanocrystals (NC's). This new synthetic method is uncomplicated, fast, versatile, close to quantitative, and reproducible. With respect of other hydrothermally methods employing organic solvents, this one presents the advantage that the decomposition path does not lead to residual carbon impurities which can be absorbed to the NC's surface. But the main achievement consists in the destructuring of the plates-like agglomerates specific to the oxalate-to-oxide thermal conversion, with direct effect on the sintering behavior of the NC's. Even extremely effective in the case of the actinides, the hydrothermal decomposition of cerium oxalate does not lead to the desired reaction product.
It was shown that HCW supports water elimination from molecules such us carbohydrates and alcohols. Very intriguing, on have found that also Ce(IV), Th(IV) and U(IV) hydroxides dehydrates under HCW, forming even smaller correspondent NC's than compared with the hydrothermal decomposition of the oxalates. Thus, we have proposed the hydrothermal decomposition of Ce(IV)-hydroxide at temperatures above 120 oC as an effective method to manufacture CeO2 NC's. Moreover, the method proved to be successful for the production of (U,Ce)O2 noncrystalline solid solution.
For the synthesis of the uranium and thorium carbides, we are currently developing a method of reacting U(VI), U(IV) and Th(IV) with different dicarboxylic acids, and subsequent decomposition of the formed complexes under inert/ reducing atmosphere.
Safety assessment for a lead-bismuth eutectic cooled reactor requires detailed knowledge of potential Pb,Bi/ MOX interaction products that could form in the event of a pin breach. Investigations on the phase relation in the Bi-U-O ternary system have shown that the formation of Bi2UO6 is likely under mild thermal conditions. Beside the classical reaction in solid state, we have also synthesized this phase by reacting different components under HCW.
In the field of radioactive and nuclear wastes, remediation of uranium contaminated waters by using magnetite and zero-valent iron particles is a well-established treatment method. We have shown that magnetite NC's may form by the decomposition of a number of inorganic compounds under SCW conditions. Presently we are studying the reductive incorporation of uranium in the magnetite structure concurrently with the decomposition reaction under HCW.
Mineral-like hydrated phosphates (relevant for the dissolution of the phosphate based ceramic waste forms) could be also obtained by using this method.

TAN 3 / 655

Electrochemical Behavior of Plutonium in Nitric Acid and n-Tributyl Phosphate

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The redox behavior of actinides is particularly complex because these elements have both many degrees of oxidation and many disproportionation reactions. In the case of plutonium, the most common oxidation states in the aqueous phase are +3 to +6. For the +3 and +4 oxidation states the

solution ions are in the aquo hydrate form whereas the +5 and +6 oxidation states are in the "yle" form, ie in the trans-dioxo form. These differences in chemical forms of the ions in solution is the source of slow kinetics between the aquo and "yles" ions, whereas in the same family the kinetics of the redox reactions is fast. Moreover, it is now well known that under certain conditions Pu(IV) and Pu(V) is disproportionate. The coupling of these two phenomena leads to complex chemical reactions with mechanisms that are not yet perfectly established.
Since the treatment / recycling of irradiated fuels is carried out industrially in aqueous nitric acid (HNO3) and organic phase n-tributyl phosphate (TBP) for extraction cycles, it is useful to better understand and control the redox equilibria of plutonium in these processes. Electrochemical techniques therefore seem perfectly suitable for studying the redox behavior of plutonium.
This presentation concerns the electrochemical behavior of plutonium in HNO3 medium as well as in TBP. The two redox couples Pu(IV)/Pu(III) and Pu(VI)/Pu(V) will be described more from a thermodynamic point of view. The redox mechanism of the Pu(IV)/Pu(III) pair in a nitric medium will also be discussed following the use of the coupling of electrochemical and spectroscopic techniques.

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New Insights into the Chemistry of the An-Se-oxosystem

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Complexity of An chemistry in aqueous oxo-salt systems is dominated by their redox behavior. It's became even more complex in the systems where oxo-salts are also redox sensitive, for example in Se and I based systems. In order to better understand the chemistry of An in Se based oxo-systems we performed a series of systematic research on reactivity and phase formation of An bearing selenates and selenites in Th, U and Np systems. Th is good modeling elements for AnIV because it is not redox sensitive and crystal chemically very similar to the UIV, NpIV, and PuIV. Uranium is quite stable in UVI form under ambient atmospheric conditions and can mimic AnVI. Np shows a great diversity in oxidation states and its chemistry is perfectly illustrates complexity of An based systems.

In our study we found that for Th system pH is one of the most important factors for phase formation and their structures. At nearly neutral pH only SeIV phases have been obtained. By changing of pH to the lower range we found very strong hydrolysis with formation of Th based oxo-clusters interlinked by SeVI.1
In U based system we found only UVI solid precipitates with different content of SeVI and SeIV which is strongly depends on the initial oxidation of Se.
The Np system is way more complex in comparison to the both previous. We started from an NpV stock solution and surprisingly no NpV solid phases were observed in the precipitate. Two novel one-dimensional Np(IV) selenites were found from reactions of NpV with SeIV: A2[Np(SeO3)2(HSeO3)2]•(H2O) (A=K, Rb). Hereby NpV most likely dispro-portionated into NpIV and NpVI and the lower solubility of NpIV selenites led to a further shift within the equilibrium. Simultaneously novel zero-dimensional NpVI nitrates also formed. The experiments of NpV interacting with SeVI reveal the first NpVI selenates: Cs2[(NpO2)2(SeO4)3], K7[(NpO2)9(SeO4)13.5(H2O)6]•(H2O)20, K2[(NpO2)2(SeO4)3(H2O)2]•(H2O)3.5, and Rb2[(NpO2)2(SeO4)3(H2O)4]•(H2O)3. All of which show two-dimensional layered structures. Such difference in phase formation demonstrates a strong dependence of precipitates chemistry from initial Se valence.2

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TAN 3 / 533

Sublattices of actinide atoms in crystal structures

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As known, in classical crystal chemistry special attention is paid to closest sphere packings, which satisfy the Laves'maximum space filling (MSF) principle. The most important to crystal chemistry closest sphere packings are the face-centered cubic (fcc) and hexagonal close-packed (hcp) ones. In terms of such packings, atoms in crystal structures are treated as hard spheres of constant radius and realize coordination number (CN) equal to 12.

Extensive calculation of single-element sublattices in all crystal structures contained in ICSD and CSD in 2006–2008 allowed one to rethink the value of Belov's 12-neighbor rule. That project involved calculation of characteristics of Voronoi–Dirichlet (VD) polyhedra of more than 12 million crystallographically independent atoms. It revealed that the most common type of VD polyhedra for more than eighty chemical elements had 14 faces, while VD polyhedra with 12 faces were predominant for sublattices of only seven chemical elements: argon and six actinides (Ac, Pa, Am, Cm, Bk or Cf).

Increased prevalence of VD polyhedra with 14 faces was explained within the stereoatomic model of crystal structure, in which atoms are regarded as "soft" deformable spheres of constant volume. The total deformation of soft spheres forming the thinnest covering corresponding to VD tessellation is minimal in the case when the centers of the spheres form a body-centered cubic (bcc) lattice. As opposed to fcc and hcp lattices, the bcc lattice features CN equal to 14. As the number of structures of actinide containing compounds has risen up by a factor of 2-4 in the last ten years, it was interesting to check if the occurrence of VD polyhedra with 12 and 14 faces has changed, and to study the reasons of different CN's in sublattices of different elements.

In the course of this study, more than 18 thousand crystallographically independent atoms of 10 actinide elements from Th to Es were analyzed. It was shown, that VD polyhedra with 14 and 12 faces still remain the most common in actinide sublattices. Curiously, U-sublattices with 20 or more independent crystallographic sorts of U atoms feature VD polyhedra with 15 faces as the most common. Similar VD polyhedra with 15 faces are characteristic for the model system 'ideal gas', which lacks both short-range and long-range order. This similarity allows us to assume that sublattices of crystals containing 20 or more crystallographically nonequivalent A atoms have no short-range order but conserve long-range order. Thus, crystals with such sublattices can be treated as antipodes of quasicrystals, which feature short-range order, but lack long-range order.

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NFC 5 / 527

Heat treatments of Cl– and water-contaminated PuO2 and its analogues

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Magnox PuO2 packaged before 1975 has become contaminated with Cl-, from thermally degraded PVC bags it was stored in, and water, from exposure to the atmosphere. Heat treatment is the preferred method for decontaminating the PuO2 for storage before either subsequent reuse as Mixed

Oxide (MOX) fuel or safe disposal in a Geological Disposal Facility (GDF). Ion chromatography experiments carried out on the contaminated PuO2 show that leachable chloride on the solid decreases and volatilised chloride increases with increasing heat treatment temperature. Powder XRD shows no change in fcc $Fm\bar{3}m$ crystal phase with heat treatment, although high heat treatment temperatures (> 600℃) sinter the crystals, increasing the crystal size, and anneal the lattice damage caused by self-irradiation, decreasing the lattice parameter. When PuO2 is heated to 225℃ and cooled in a sealed (Baskerville) vessel, a non-condensable mixture of gases (including hydrogen, helium, NO and CO), ideal above ~113℃, is produced, and the monolayers of water on the surface of the PuO2 decrease. Repeating this experiment with 0.05 mL water in a side chamber increases the number of water monolayers on the PuO2 surface.
PuO2 artificially chloride-contaminated, by exposure to dry HCl vapour, and humidified in a sealed pot with H2SO4 (conc.) shows similar results to the Magnox PuO2 when heat treated at various temperatures. CeO2 analogue nanocrystals have also shown an increase in size when exposed to HCl vapour and following heat treatment at temperatures higher than the original calcination temperature. Studies on CeO2 analogues also showed that the sorption mechanisms of chloride and water are linked. Upon heat treatment of the contaminated CeO2 at 900 ℃, the nanocrystal morphology resembles that of PuO2 in TEM images, and the chloride, which was homogeneously spread on and within the CeO2 particles, is no longer detectable by EDX.
XPS and UPS studies on CeO2, CexO2-x, ThO2, UO2 and U metal thin films, involving in situ sputter deposition synthesis, HCl/Cl2 contamination and Thermal Programmed Desorption-Mass Spectrometry (TPD-MS), showed that adsorbed Cl- is not volatilised up to 800℃, but diffuses beneath the surfaces of the films. No change in oxidation state is observed for the metals in the metal oxides, but U metal oxidises upon contamination and TPD, with Cl- the only Cl species detected. XPS studies on spin coated CeO2 thin films contaminated with HCl vapour ex situ show a change in Ce oxidation state at high heat treatment temperatures from +4 to +3 for the thin films originally calcined at low temperatures, but Cl- remains the only Cl species present and also decreases on the CeO2 surface, following heat treatment at high temperatures.

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Thermal decomposition and structural changes of lanthanide-doped uranium dioxide particles prepared by internal gelation

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In any advanced nuclear fuel cycle, partitioning and transmutation (P&T) is a key strategy to reduce spent nuclear fuel's radiotoxicity and heat generation: Long-lived minor actinides (MA) are partitioned from spent nuclear fuel and subsequently converted to fuel pins or targets. These are used as nuclear fuel in fast reactor systems where the actinides are fissioned to short-lived radionuclides.
An essential link between the partitioning and the transmutation is the conversion of the separated minor actinides into solid precursors to fabricate fresh fuel suitable for minor actinide recycling. Among transmutation fuels, particle fuel offers certain advantages compared to pellet fuel. Particles can be prepared without any formation of dust via an aqueous synthesis pathway. Moreover, the fuel-manufacturing process offers advantages in terms of automation up to and including the rod filling step. Strategies to fulfil this task are sol-gel processes, where a stable solution containing the desired metals, the sol, is converted into a solid gel.
Within this study, the feasibility of the sol-gel route via internal gelation was investigated for the fabrication of neodymium- and cerium-doped UO_2 microspheres. Nd^{3+} was used as simulant for the minor actinide Am^{3+}. Cerium was used in the tetravalent and trivalent state to simulate plutonium and to investigate the influence of the metal's oxidation state on the synthesis process and final product. Depending on the chosen preparation conditions, stable microspheres with lanthanide contents ranging from 5 % to 40 % could be prepared. Additionally, non-doped reference particles

were fabricated using the same synthesis parameter and conditions.

A method to convert the gelled microspheres into a dioxide form was investigated in detail. The thermal decomposition pathway under oxidizing conditions was first investigated by simultaneous thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC), coupled with evolved gas analysis via mass spectrometry (MS). The calcined particles were then sintered for 10 h at 1600 ℃ under reducing conditions. The study also focussed on assessing the crystal structure evolution in the intermediate products via powder X-ray diffraction (XRD).

Single phase solid solutions according to the formula $U_{1-y}Ln_yO_{2\pm x}$ were found in case of the sintered particles. A cubic fluorite structure with a lattice parameter linearly decreasing with increasing lanthanide content according to Vegard's law was observed via XRD. In case of the intermediate products after calcination a more complex, mixed-phase behaviour was found. The non-doped particles contained a single orthorhombic phase, whereas the doped particles contained the orthorhombic phase and an additional fluorite phase. Interestingly, the relative amounts of fluorite phase depended on the amount of dopant and maximum temperature applied during the calcination. For compositions with *Ln* contents of 30 % calcined at 1300 ℃ only the fluorite phase and no orthorhombic phase was found. The implications of these results on the conversion strategy will be further discussed.

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Astatine: Halogen or Metal?

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The halogen family is generally assumed to stop at iodine. Does astatine, element 85, continue the vertical trend of the halogens, does it behave like a metal? Unfortunately, little is known of the basic chemistry of astatine: it is a rare (only short-lived isotopes can be produced) and "invisible" (the amount of produced 211At requires working at ultra-trace concentrations) element. In the past decade, a program on astatine chemistry has been developed in Nantes in perspective to use 211At for targeted alpha therapy 2. The purpose of this presentation is to give an overview of the results obtained thanks to an original methodology combining experimental [3-5] and theoretical tools [6-8]

Astatine behaves as halogens. At- was evidenced by ion-exchange chromatography 4 and electromobility [5] measurements. Most of the current labelling protocols are developed based on iodine chemistry and lead to the formation of of astato(hetero)aryl labelled compounds by electrophilic demetallation. An alternative approach with nucleophilic astatine based on aryliodonium salts precursors was recently developed. A much higher reactivity of astatide than predicted was observed compared to iodide. Such discrepancy was rationalised by DFT calculations predicting differences in reaction pathways [9]. More recently, experiments have shown a first indirect indication of halogen bonding between AtI and Lewis bases, further supported by relativistic quantum mechanical calculations. AtI was found to be a stronger halogen-bond donor than I2 [10].

Astatine behaves as metals. The range of existence of At- is limited [11] with the presence of two At species at the oxidation states +I (At+) and +III (AtO+) [12] formed in more oxidizing conditions. Astatine cationic species can interact with both inorganic and organic ligands [11, 13-14] to form complexes. Interestingly, At+ forms with I- and Br- the heaviest ternary trihalogen species, IAtBr- in aqueous solution [15].

To conclude, astatine can be considered as a chameleon, as was highlighted by Visser in 1989 [16]. An important question may arise from this work: can we use the metal character of astatine to develop alternative labelling strategies in light of our recent results rationalizing the limit of the classical labelling protocol [17-18]?

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NFC 5 / 719

MCNP modeling of impurity activation in graphite moderator and reflector of a L-54M-type nuclear research reactor

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The CeSNEF L-54M was a homogeneous nuclear research reactor fueled by a uranyl sulfate aqueous solution enriched by 19.94% 235U. It was commissioned by Politecnico di Milano to Atomics International in the late ‘50s. After 20 years of operation, in 1979 the reactor was definitely shut down. Since then, Politecnico di Milano decided to manage the entire structure according to in-situ safe storage strategy, so as to take advantage of short-lived radionuclides decay by reducing the total amount of radioactive material to be removed and allowing safer dismantling operations. Recently, Politecnico di Milano decided to restore the unrestricted re-use status, thus L-54M decommissioning has been launched. During the last years, preliminary environmental radiological characterization of the surrounding topsoil has been performed 1. At the same time, analysis of few samples of irradiated graphite and concrete evidenced the presence of gamma emitting radionuclides (above all 60Co and 152Eu), 3H and 14C, even though a more extended radiological characterization would be required for implementing L-54M decommissioning.
In this work, a Monte Carlo N Particle (MCNP) 3D model of the reactor has been developed and validated in order to accurately assess the activation of the reflector graphite stack. This study is aimed at better addressing the future radiological characterization efforts and reducing the associated analytical costs. In order to build an accurate MCNP model, rigorous geometrical and materials information about the main reactor components were required and collected from blueprints, security and safety reports, as well as from experimental and academic works published in the 1960s. The L-54M reactor pile has been modeled in detail up to the outer boundary of the concrete shielding. In order to verify the model and ascertain its accuracy, several simulated data have been compared with available experimental ones with satisfactory agreement. In particular, neutron flux profiles in the experimental irradiation channels, criticality condition, control rods inventory and calibration have been considered. Afterwards, a superimposed fine mesh has been used to obtain the neutron fluence in the graphite reflector. For each radionuclide of interest, the main production reactions have been considered and the activation precursors have been assessed thanks to a detailed elemental characterization of virgin graphite by ICP-MS analyses following a suitably developed oxidation

and acid digestion procedure. The MCNP data have been properly elaborated and converted to activity concentration values, taking into account the exponential decay up to the measurement date. A satisfactory accordance between MCNP simulations and preliminary experimental data has been highlighted. Following this promising study, further samples have already been collected at different positions within the graphite stack in order to better corroborate the model outcomes.

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TAN 3 / 547

Impact of charge transfer to ligands on magnetism of Uranium compounds

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Uranium metal as well as other light actinides are weak Pauli paramagnets, i.e. do not exhibit magnetic moments. The reason is a large overlap of the 5f wave functions centered on nearest U neighbours. The 5f states form consequently a broad 5f band. Increasing the U-U spacing in compounds makes the 5f band narrower and eventually U moments are formed and order (ferro- or antiferromagnetic ordering) unless the hybridization with ligand states, which can similarly broaden the band, is too strong. The critical U-U distance, d(U-U) necessary (but not sufficient) to form U magnetic moments is 340-360 pm. From handful of exceptions with lower d(U-U), the most conspicuous is uranium hydride, UH3. Despite low d(U-U) = 331 pm in the common beta-UH3 form, ferromagnetism with moments exceeding 1 magneton Bohr appears below relatively very high Curie temperature Tc = 165 K. We found that hydrogenating U alloys with early d-metals (Mo, Ti, Zr) Tc can even increase, exceeding 200 K. A hint why it is like that was provided by ab-initio calculations indicating a charge transfer from the U-7s and 6d states towards the H-1s states. The charge transfer surprisingly does not involve the 5f states, which remain populated to 2.7-2.8 electrons per U atom. Such situation reduces the 5f-6d hybridization as one of the delocalizing mechanisms, and a narrow 5f band remains at the Fermi level. Magnetism is much more pronounced than in case the U metal is hypothetically blown up to the volume of UH3. A thorough study of UH3 by XPS/UPS indicates indeed fingerprints for the charge transfer. This work opens new suggestions where to seek for more magnetic uranium compounds. Similar charge transfer has to take place e.g. in binary and ternary pnictides. If the transfer is too large, as e.g in oxides, the product becomes non-metallic, and ordering temperatures have to drop down. The highest Tc = 216 occurs in UCu2P2. Much larger d(U-U) in this case is perhaps contra-productive, the exchange interaction at a long distance is getting weaker. Therefore we studied a lattice compression in external hydrostatic pressure. Indeed, a dramatic increase of Tc has been traced so far to the pressure of 4 GPa, yielding Tc = 255 K. As the pressure derivative is still high, there is a good chance that a room-temperature 5f ferromagnet, with all its specific features originating from the strong spin-orbit interaction, as giant magnetic anisotropy, will soon become reality.

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Homogeneous precipitation of thorium by thermal decomposition products of urea

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Although thorium dioxide is considered as a replacement for UO_2 as a matrix for nuclear fuel, there are practical problems which must be resolved before deployment. These include the poor sinterability and the cost of manufacture. ThO_2 is the most stable binary oxide and thus requires higher temperatures than UO_2 to sinter to the required densities of ≥95%. The sintering temperatures can exceed the 1750 ℃ commonly achievable by commercial furnaces. In addition, the decay products of ^{232}Th are shorter lived compared to ^{238}U decay products and the activity of stocks can increase significantly during storage. Should these decay products such as the longer lived ^{228}Ra remain in the liquid phase during powder production, waste disposal costs will rise significantly. If radium can be incorporated into the solid phase, these costs can be avoided.

Homogeneous precipitation using urea is a promising method for preparing ThO_2 for fuel applications, as it produces polycrystalline ThO_2 agglomerates, which readily form necks even at low temperatures 1. This makes such ThO_2 an excellent candidate to counteract the poor sinterability inherent in ThO_2. Nonetheless, very little is known about the process. To correct this, the precipitation of Th from nitric acid solutions was followed over time as a function of the urea decomposition temperature and amount. 2

Urea decomposes to form ammonia and carbonates, both of which are useful in the precipitation. Ammonia readily precipitates thorium, but leaves radium as highly soluble $Ra(OH)_2$. The carbonate produced by the urea process can serve to correct this, as $RaCO_3$ is very insoluble. The ammonia indeed caused >90% Th to pass into the solid phase at a pH above 4. These efficiencies increased over time to 99%. Despite supersaturation with CO_2 already occuring at low pH, the binding of Ra into the solid phase only begins to happen after Th precipitation is complete, above pH 7. Even after 24 h of heating, reaching pH 8.5, this process remains incomplete and only 80% of the radium is precipitated.

The produced precipitate is composed of nanometric, crystalline ThO_2, with adsorbed H_2O and CO_2. Prolonged heating causes the individual ThO_2 crystals to grow gradually. Unfortunately, the precipitate is very soft and readily forms a gel. This poorly affects the flowability of the powder and thus its potential industrial application.

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TAN 3 / 638

Thermodynamic and Structural study of uranyl phosphonate-based ligands as potential uranium decorporation agents

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ABSTRACT: Because of the widely use of uranium in fuel cycle, especially in the reprocessing process, the environmental and occupational exposure opportunities have increased. Although uranium has very limited radioactive dose impact, its chemical toxicity still need to consider, e.g. uranyl ion (UO22+, U(VI) could cause renal injury. The only way to remove or prevent the internally deposited uranium is by using decorporation agents to accelerate excretion. It is known that phosphonate group has a really strong affinity to complex U(VI). Two phosphonate-based ligands were synthesized and tested for its uranium-binding properties.

Fig.1 structure of bisphosphonate-based ligands
The two ligands could provide 5 coordination at equatorial planar, 3N & 2O, for the uranyl. L3 was designed only for uranyl with relatively high lipophilic property due to the 3 aromatic group, meanwhile L4 could donate 7 coordination totally, 3N & 4O, which should also has high affinity to chelate minor actinide. 1
Due to the hydrolysis of uranium, low pH is required to have a major free uranyl species in solution, thus pH 3 was chosen. Then for physicochemical study for blood serum, pH 7.4 was suggested.
Then ligand uranyl complex was preliminary studied with TRLFS. Due to formation of precipitation at pH 3 with NaClO4 or NaNO3 as ionic strength, although Cl- will quench the fluorescence of uranyl, NaCl was been used. For L3 at pH 3, the uranyl ligand complex has a strong fluorescence with a red shift about 7 nm, the time constant is 0.48 um. For L3 at pH 7.3, the uranyl ligand complex has no fluorescence no matter with NaCl or NaNO3 which suggests there is a configuration change. The same study was done with L4. At pH 3, the uranyl l4 complex fluorescence spectra have a red shift about 5 nm, the time constant is 0.83 um. At pH 7.3, the uranyl l4 complex fluorescence spectra have a red shift about 9 nm, the time constant is 25 um. Thus, the coordination mode for two pH shouldn't be the same.
Then ATR-FTIP was been used to study the uranyl ligand complex at two pH. At pH 3, each phosphonic acid group has deprotonate one proton. At pH 7.4, there is no proton for the phosphonic acid group.

Fig.2 FT-IR spectra in absorption mode of free L4 and L4-U, pH = 3 &7.4. Normalization was performed on the band at 1600 cm−1 (not shown) and spectra were shifted in ordinates
Three type of phosphonate-based ligands /uranyl complexes mode under pH 3 and 7.4 had calculated with DFT. Then EXAFS was implement.

Fig.3 Minimum energy conformation obtained from DFT calculations (B3LYP) in aqueous solution for [UO2(H4L4)] at pH 3(left) & [UO2(H4L4)] at pH 7.4(right) system

Fig.4 U LIII edge k3 -weighted EXAFS spectra (left) and the corresponding Fourier transforms(right) of the ligand–U complexes formed. The experimental spectra are given in line and the fits are given in dash line.

Table 1. Structural parameters of the ligand–U complex formed at different conditions

samples	path	Ndeg	sigma^2(× 10−3 Å2)	RFIT
UO2-L3-PH3	U-Oyl	2	3.67	1.79508
	U-Op	2	2.92	2.30471
	U-Npyr	1	2.08	2.43804
	U-Namine	2	9.93	2.95359
	U-P	2	14.95	3.69177
UO2-L3-PH7.4	U-Oyl	2	3.03	1.79963
	U-Op	2	7.42	2.32334
	U-OOH	1	7.42	2.32334
	U-Npyr	1	0.72	2.47119
	U-Namine	2	14.81	2.97252
	U-P	2	10.23	3.76969
UO2-L4-PH3	U-Oyl	2	3.87	1.80601
	U-Op	2	4.04	2.31739
	U-Npyr	1	2.92	2.49759

U-Namine 2 27.17 2.8262
U-P 2 19.48 3.68678
UO2-L4-PH7.4 U-Oyl 2 4.02 1.78448
U-Op 2 8.2 2.28005
U-Npyr 1 8.2 2.31105
U-OOH 1 4.85 2.93286
U-Namine 2 10.46 2.23852
U-P 2 9.14 3.56492

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Electrochemical behavior of Co(II) in 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide

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Because of their characteristic properties, room temperature ionic liquids (RTILs) are considered to be suitable alternatives to presently used systems in the field of spent fuel reprocessing and decontamination/recycling of materials contaminated with radionuclides. Electrochemical separations from RTILs are highlighted processes in the field and Co is one of the most common contaminants during recycling of material coming from many industrial processes. *1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide* ([EMI][NTf2]) was chosen as a carrier system in the recycling scheme for Co-contaminated material because of its hydrophobicity, satisfactory solubility of the most Co compounds and sufficient electrochemical window.

Both chemistry and electrochemistry of Co in [EMI][NTf2] was studied. Chemical forms of Co in the solution are discussed as well as the mechanism of electrochemical reduction of Co(II) ions and possibilities of the Co deposition on the solid electrode. It was shown that even high lattice energy Co compounds can be dissolved in [EMI][NTf2]. However, if Cl- ions are present in the system, complexes with mixed ligands appear and certain concentration is needed to form complex reducible to the pure metal. Metal Co deposits were subsequently obtained and its characterization is presented as well.

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A long-term monitoring of 14C in liquid discharges from the NPPs of the Czech Republic

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During a normal operational regime of the nuclear power plants (NPPs) equipped with pressurized light water reactors (PWR), 3H and 14C are two biggest contributors to the effective dose of population. While 3H is responsible for the highest dose from all nuclides discharged in liquid effluents, 14C causes the highest dose from the nuclides liberated in airborne discharges. Monitoring of the 14C in the liquid effluents is not part of the standard monitoring procedure of the most NPPs with PWR.

There are two operating NPPs with PWRs in the Czech Republic: Temelín (ETE) and Dukovany (EDU). Monitoring of 14C in the liquid effluents has not been routinely performed. Therefore there is a lack of comprehensive information about contribution of liquid effluents to the total amount of discharged 14C. Generally accepted report by Kunz (1985) states that more than 95% of 14C from NPPs with PWR is discharged via gaseous effluents. The 14C liquid discharges from Swedish NPPs with PWRs were reported by Magnusson et al. (2008) to be about 0,3% of 14C total release, while results of the long-term monitoring of 14C releases from the French NPPs with PWR reveal relatively high annual values of 7,5% of total 14C release in liquid effluents (CEPN 2005).

On the basis of ambiguous results reported in literature, the regulatory body State Office for Nuclear Safety (SUJB) requested introduction of a long-term monitoring of 14C in liquid effluents from both EDU and ETE.

A method for determination of chemical forms of 14C in liquid discharges from the nuclear power plants was developed in collaboration between the National Radiation Protection Institute (NRPI) and the Nuclear Physics Institute (NPI CAS) (Svetlik et al. 2017). During the year 2016, the Laboratories of Environmental Control of the both Czech NPPs were provided with this method and till the end of the year 2017 each one-month liquid release batch was sampled and the sample was analyzed. Activities of 14C in both organic and inorganic forms were determined.

Comprehensive results of this unique two years lasting monitoring will be presented. Percentage of 14C in form of liquid effluents in total 14C releases was calculated for the period of observation, calculated values are in conformity with values reported by Kunz (1985). Contribution of 14C in form of liquid effluents to the effective dose of a representative person was calculated and compared with contribution of 3H.

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Development of procedures for separation of uranium and thorium from selenium matrices for their determination by mass spectrometry techniques

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Large-scale experiments which have been or are being established in order to answer some fundamental questions of nuclear physics, such as the SuperNEMO experiment, have been recently arising the need for radiopurity measurements on the nBq/g or even sub-nBq/g levels. One of the biggest challenges of these experiments is to reduce the background radiation to minimal values. The isotope source deposited on a foil to be used in the SuperNEMO experiment, which shall be exclusively devoted to the search for the neutrinoless double beta-decay, is made of enriched 82Se powder. Despite the fact the 82Se powder material have been purified before being further processed, it still contains trace amounts of natural uranium and thorium. 238U and 232Th decay to short-lived beta emitters (214Bi and 208Tl, respectively), which are very dangerous because the energy of their beta-electrons is higher than the double beta-decay energy of 82Se. Therefore, precise determination of 238U and 232Th in the source is crucial for the evaluation of the contribution of their progenies to the background signal. Several methods have been exploited for low level analysis of uranium and thorium in various construction materials, though it is assumed that the proposed detection limits could be reached only with the use of the state-of-art mass spectrometric techniques, i.e. inductively coupled plasma mass spectrometry (ICP-MS) and accelerator mass spectrometry (AMS). However, both ICP-MS and AMS generally require pre-concentration of the radionuclide of the interest and its almost complete separation from the original sample matrix. Selenium, uranium and thorium can be separated from each other by e.g. solid phase extraction (SPE) or anion exchange chromatography. Here we shall present results of testing of straightforward procedures for separation of 238U and 232Th from selenium using UTEVA resin (TrisKem) and Anion Exchange resin (Cl-) form (TrisKem). Modelled samples were prepared from commercially available high-purity elemental selenium powder (99.999%; Alfa Aesar), whose quality was checked by low-energy mass scanning, and spiked with 232U and 229Th. Overall radiochemical yield of the procedures was calculated from alpha spectrometry determinations of 232U and 229Th, as well as from gravimetric measurements of selenium.

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Time and temperature depend changes of radionuclides sorption behavior of clays in Yeniseysky site environment

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Sorption behavior is one of the most important characteristics of material considered to be used in engineered barrier on the sites for radioactive wastes repositories. One such site in Russia is located near Krasnoyarsk on the Yeniseyskiy site, in the Nizhnekansky rock massive. It is designed for intermediate and high level radioactive wastes, including self-heating radioactive wastes containing actinides 1. Consequentially, during the loading period and the initial timeframe of operation local areas of elevated temperature could exist 2.

We studied sorption behavior of clays of different mineralogical content from sites located in the vicinity of the repository site both at room temperature and at 90°C to simulate the processes that could be important in the near repository zone. The sorption behavior of the clays was estimated within the following scheme: sorption kinetics study, distribution coefficients (Kd) calculation, and speciation of sorbed radionuclides according to the Tessiers technique.

All sorption experiments were done with dried, crushed and sieved clay samples of different content (natural bentonites, Na-bentonite, kaolin clay) and with model groundwater containing actinides and some fission products. It was shown that increasing the temperature of contact caused increasing Kd and sorption rates. A higher temperature of contact shifts the speciation of sorbed radionuclides to more strongly bounded species. Increasing the contact time also causes stronger bonding of radionuclides to clay.

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Ca-Mg phosphates as an effective sorbents of 60Co and 85Sr radionuclides

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High risk of radiation for human health makes continuous interest in finding effective methods for radionuclides immobilization. Depending on the level of radioactivity, sources, and amount of pollution different methods are used for immobilization of radionuclides, details of which are summarized in some review papers: precipitation, evaporation, solvent extraction, membrane technique, filtration, sorption and ion-exchange. Each method has its advantages and limitations in use, so studies are underway in all these areas [1, 2].

The aim of this work was to study the efficiency of Ca-Mg phosphate sorbents based on dolomite for 60Co and 85Sr radionuclides removal from aqueous solutions depending on the electrolyte content in the wide range of pH. It was important to determine the influence of competing ions in the solution on the stability of Ca-Mg phosphate sorbents and the effect of the surface changes of sorbents on their selectivity towards 60Co and 85Sr removal from solutions with high salinity.

Dolomite from the Ruba (Belarus, Vitebsk region) deposit with the following chemical composition (wt. %): SiO_2 1.1, Fe_2O_3 0.4, Al_2O_3 0.5, CaO 30.3, MgO 20.0, SO_3 0.4, K_2O 0.2, Na_2O 0.1 and calcination loss 47.0 was used for the preparation of Ca-Mg phosphates. Sorbents with different chemical and phase composition (Fig. 1) were obtained in various dolomite phosphating conditions: PD-1 – mixture of Ca-Mg hydrogen phosphates, PD-2 –mixture of amorphous Ca-Mg tertiary phosphates with admixture of $MgNH_4PO_4$ and PD-3 –mixture of MgO, $MgHPO_4$, $CaCO_3$ with admixture of amorphous $Ca_3(PO_4)_2$ [3, 4].

Fig.1 XRD patterns of Ca-Mg phosphate sorbent PD-1 (a), PD-2 (b) and PD-3 (c)

The process of radionuclides 60Co and 85Sr uptake from model seawater solutions with salt content 0 35 g/L by phosphate sorbents produced from natural dolomite were studied. It was shown that the efficiency of phosphate sorbents depends on the dolomite phosphating conditions which determine the chemical and phase composition of sorbents, acid-base properties and consequently different selective-sorption properties towards 60Co and 85Sr radionuclides in aqueous and model seawater solutions.

Fig. 2 Solution pH after contact with sorbent (pHf) vs pH of model solution (pHo) with salinity 35.0 g L-1

The influence of pH on the removal efficiency of 60Co and 85Sr is determined by the value of pHpzc,

which depends on the composition of sorbents and processes of protonation-deprotonation on their surface. For the studied sorbents values pHpzc increased in the row of PD-1 < PD-3 < PD-2, with the values of 7.2, 7.3 and 8.4, respectively (Fig. 2). Only for PD-2 sorbent the value of pHpzc exceeded pH in model solution, which ensures the independence of the degree of sorption and distribution coefficient on solution pH in the pH range 3.0 –8.5 4.
It was found that for the studied phosphate sorbents the efficiency of 60Co and 85Sr sorption from model solutions decreased with salinity increase, due to the increase in ionic strength of the solutions and the competing effect of metal cations in the background electrolyte. The most effective sorbent for 60Co and 85Sr radionuclides showed sample consisting of tertiary phosphates of calcium, magnesium and magnesium ammonium phosphate (PD-2): Kd 60Co from 2.44×104 to 3.32×103 cm3/g and Kd 85Sr from 2.66×103 to 0.14 × 103 cm3/g in the salinity range 0.0-50.0 g/L, respectively.

Fig.3 Effect of salinity on the lg Kd of 60Co (a) and 85Sr (b) radionuclides on Ca-Mg phosphate sorbents (V/m=500 cm3/g)

Thus, high effective Ca-Mg phosphates sorbents towards 60Co and 85Sr were synthesized. Effect of sorbents phase and chemical composition, salinity and pH of solution were studied. The highest sorption efficiency and selectivity was obtained for PD-2 sorbent. The simplicity of Ca-Mg phosphates preparation and use of widely occurring natural raw material dolomite for their production give the scientific basis for industrial implementation of novel sorbents for treatment facilities. The produced materials are promising for application in seawater and liquid radioactive waste treatment from 60Co and 85Sr radionuclides.

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Modern geoecological situation of ”Crystal”peaceful underground nuclear explosion vicinity (Siberian Plate)

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Modern geoecological situation of “Crystal”peaceful underground nuclear explosion (PUNE) area basing on the geochemical and the geophysical sounding data is considered. New data of the contents of radionuclides in surface and underground waters are obtained.
Geophysical transient electromagnetic sounding, in other words - time-domain electromagnetic sounding method (TDEM) along two profiles above the epicentre of PUNE was applied. The revealed geoelectric layers conform to the geological structure of the area. There is a local geotechnogenic anomaly under the epicentre of the explosion: it is a sharp rise of the aquifer levels by 260-300 m up to the cavity of PUNE.
The destabilization of geotechnogenic system «”Crystall”cavity –geological environment –surface ecosystems» is caused by the mining of the kimberlite pipe “Udachnyi” at a distance of 3.5 km to

southwest from the "Crystal". Deepening of the open-cast mine and its drying promoted the formation of a great cone of depression around the mine. Radioactive underground brines from the zone of the explosion moved to this depression, mixing with the brines of underlying horizons in the cone of depression.

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Keywords: peaceful underground nuclear explosion (PUNE), Yakutia, geotechnogenic system, permafrost, underground waters, brines, radionuclides, tritium, time-domain electromagnetic sounding method (TDEM), ecological risk

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Radioactive medicinal springs: Searching for activating geomaterials

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Radioactive mineral waters (rich in dissolved 222Rn) are highly valued for their medical use. Therefore they have been intensively explored in last years (2005–2017) in the Lusatian and Saxothutingian units of the Bohemian Massif. Springs of radioactive medicinal water are bound up with the Cambrian/Ordovician orthogneisses and Tanvald granite of the Variscan age in the Lusatian unit and Smrčiny/Fichtelgebirge Variscan granite in the Saxothuringian unit respectively.

More than 130 water sources with activity above 1500 Bq/L (i.e. "radioactive mineral waters") were found in the whole territory. The highest radon activity reaches 13.2 kBq/L in case of the Břetislav spring in Plesná near Cheb. Waters are cold (5–9.5 °C) and low-mineralized (TDS < 200 mg/L), activities of 228U, 226Ra is generally low; 222Rn is the only important component.

The occurrence of radioactive sources is completely mysterious. In the overwhelming majority of cases, only radioactive water is present, but no solid geomaterials with increased radioactivity. The secret of activation takes place somewhere deep in the Earth. Only in two cases the active materials were found directly on the surface near the spring outflow.

The most significant 16 sources were technically captured for scientific observation and their balneological use. During the works, more than 50 samples of solid geomaterials and other samples of surrounding rocks were collected for comparison. The samples taken are mainly from the outlet channels of the spring where they were in long-term contact with running water.

Samples were examined for radionuclide content (gamma and alpha spectroscopy and alpha autoradiography) as well as mineralogical methods (optical microscopy, XRD, EM-EDA).

Although geomaterials have long-term contact with water, most of them have only low activity comparable to that of surrounding rocks. Only in rare cases have samples with elevated levels of radionuclides been found.

In these cases, a close relationship between mineralogical composition and affinity for individual radionuclides was found. While radium (226Ra) concentrates in hydrated Fe oxides (amorphous Fe oxides and goethite of varying degrees of crystallinity) and Mn hydrated oxides (birnessite and asbolane), uranium (238U) is preferentially bound to amorphous hydrated oxides of Al and Ti (so call "uranium leucoxene").

The question was how to distinguish geomaterials from water activating and water activated. In this case, the ratio of 210Po/226Ra was used as the most appropriate indicator. Activated materials have a ratio of 210Po/226Ra greater than 1 due to radon inflows in the system. On the other hand, the water-activating materials we are looking for have a ratio of 210Po/226Ra less than 1 due to radon long-term escape. Only four samples meeting this criterion were found in the whole sample set. But only one of them was actually in contact with water in a natural situation. Nature is still guarding its secrets.

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Studies on graphene-based materials for the removal of radionuclides from aqueous solution.

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Graphene-based materials (GBMs), such as graphene oxide (GO) have previously been shown to demonstrate exceptional surface sensitivity 1, sorption capacity 2 and selectivity 3 for radionuclides. In addition, it has been found that the selectivity of GO towards certain radionuclides can be further improved by covalent attachment of chelating ligands. Such modifications have been shown to lead to enhanced targeted sorption and removal of long-lived actinides, in particular, U(VI), from aqueous solution [4, 5]. Therefore, ligand-modified GO is believed to be an ideal sorbent material for use in aqueous nuclear waste treatment. In such applications, selective removal of radionuclides could potentially lead to a reduction in the total volume of nuclear waste designated for long-term storage in reprocessing facilities [6].

This work aims to present the recent developments made to modify the surface of GO, which was designed with a high affinity for U(VI). The sorption behaviour of the modified-GO materials was investigated in the form of batch experiments, as a function of pH, sorbent to solution ratio and contact time, with aliquots analysed by inductively coupled plasma mass spectrometry (ICP-MS). Finally, a combination of surface characterisation techniques was performed to assess the suitability of the modified-GO material for waste processing.

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Sorption properties of selected oxidic nanoparticles for the treatment of spent decontamination solutions based on citric acid

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Nanoparticles of metal oxides with high specific surface area show high adsorption towards selected radionuclides with fast kinetics. Metallic nanooxides were tested for separation of selected radionuclides (Am-241, Co-60, Cs-137, Sr-90) from simulated spent decontamination solutions containing citric acid. Following materials, prepared by photoinduced synthesis, were tested: NiO, NiO/TiO2 and ZnO:Cu. Buffer solutions with concentration of citric anions 0.01 mol/L were tested. Sorption properties (sorption kinetics and weight distribution ratios) were investigated in the course of the experiments performed in pH range 2–12. Additionally, the influence of the presence of surfactants on the sorption was studied. The results obtained show that metal oxides in the form of nanoparticles adsorb radionuclides rather in the basic pH range. Outcomes will be used for the development of method for in situ treatment of spent decontamination solutions containing citric acid produced during radiation accident.

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Molecular Insights into Uranium Adsorption to Granite Gained from TRLFS, EXAFS and EPMA Study

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Hexavalent uranium is a prominent radioactive contaminant in both sediments and aquifers around nuclear activity sites, and poses a potential health and environmental risk to the biosphere. Understanding on the detailed interaction at the solid-liquid interface between uranium and granite is crucial for the safety assessment of high level radioactive waste geological repository in granitic terrain, as well as prediction of uranium's geochemical fate in the environment. However, the interaction between uranium and granite is complicated by the ubiquitous ligands in natural media such as phosphate, humic substances et al, thus the detailed insight into U-granite interface is critical in governing the subsurface mobility of uranium in disposal environment. Full understand on complicated interaction mechanism between uranium and ligands at the granite-water interface requires full identification of surface species with the aid of sensitive spectroscopic techniques.

In this work, the adsorption of uranium on granite in both absence and presence of phosphate was investigated by a combination of batch measurements and spectroscopic techniques, including cryogenic time-resolved laser induced fluorescence spectroscopy (TRLFS) and extended x-ray absorption fine structure (EXAFS). Results showed that phosphate is beneficial for uranium immobilization, the spectroscopic confirmation revealed that multiple surface species including inner-sphere complexes and surface precipitates were formed on granite surface with their abundances varying as a function of acidity. The EPMA results showed that uranium mainly located on mica mineral, thus the detailed interaction between uranium and mica was further investigated. The results showed that uranium sorption on phlogopite mica was strongly dependent on pH while minimally affected by the ionic strength, multiple inner-sphere surface species (including ≡SOUO2+, ≡SO(UO2)2(OH)2CO3- and ≡SOUO2(CO3)x1-2x) were formed with their abundance varying as a function of pH, and a portion of uranium precipitated as uranyl oxyhydroxides at pH > 9. The presence of HA made significant difference on uranium sorption behavior as well as surface species. The finding in this work is helpful for understanding on the geochemical fate of uranium in granitic environment as well as setting a reliable reference for surface complex models.

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Comprehensive studies on third phase behavior during solvent extraction of U(VI)/Th(IV) mixtures

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The third phase formation during liquid/liquid extraction has been studied for decades by several authors[1−3] starting in the 50's. Those studies were focused on the determination of the maximum organic loading that can be reached for a given metal in order to avoid organic phase splitting during the industrial applications such as the PUREX process. More recent studies have been conducted using a physical approach[4−5] to propose models accounting for phase segregation phenomena. A new interest on third phase studies appeared in the early 2000's with the investigations related to the Th-cycle[6−8]. Most of the models and results rely on SANS/SAXS or FTIR data acquired on metal-free or single-metal systems, and concentrations are often determined through indirect analysis.

Despite the huge amount of data concerning LOC (Limiting Organic Concentration) of actinides (IV) for different phosphate based solvents, a lack of data remains in the literature for $U^{(VI)}$ alone and in the presence of $Th^{(IV)}$ or $Pu^{(IV)}$ for TBP/dodecane solvent. Besides, there are few values concerning the composition of both heavy and light organic phases after splitting, and when available, the data covers only a narrow range of the 3^{rd} phase existence.

We have determined $U^{(VI)}$LOC values for previously described system at different HNO_3 and TBP concentrations which have not been yet reported in the literature. The behavior of $U^{(VI)}$ and $Th^{(IV)}$ after phase segregation and the composition of each phase was characterized using X ray fluorescence spectroscopy (XFS), allowing metal and TBP direct titration in organic medium for the first time ever according to our knowledge. Nitric acid and water contents were also estimated using acid/base and Karl Fischer titration respectively. Trends were established for $U^{(VI)}$ and $Th^{(IV)}$ alone in the third phase, as well as for bimetallic $U^{(VI)}$/$Th^{(IV)}$ systems.
A parallel has also been drawn by studying transition metals behavior in HCl/TBP/dodecane systems (analyzed by XFS and ICP-OES). We showed that LOC values and metal distribution can vary considerably from a metal to another even when working in the same conditions of extraction. We are currently investigating molecular species, as some other authors[9−10], that may be involved in phase splitting in order to determine contributions of metal and acid to phase segregation phenomena.

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RER 4 / 483

Contamination of forest fuel materials by long-lived radionuclides and estimation of its threat to human health during the wildfires in Belorussian part of Chernobyl exclusion zone

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After 30 years of accident on the Chernobyl NPP radioactive contamination in 30-km Chernobyl zone is determined by long-lived radionuclides of Cs-137, Sr-90 (with the half-life time 30.1 and 29.1 years, respectively) and alpha-emitting isotopes of Pu-238,239,240 and Am-241, also known as transuranium elements (TUE) (Sokolik 2004; Kashparov et al. 2003).
The overall aim of this article is to determine the volume activity of long-lived radionuclides in the surface air by experimental simulation of forest fire and estimate the potential health threat through calculation of effective doses for firefighters.
Materials and methods. Experimental simulation of forest fire, radiochemical analysis, spectrometry measurements, statistical analysis.
Results and conclusions. The data on airborne activity concentration was obtained by experimental simulation of forest fire in the smoke chamber. The common trend is the increasing of airborne activity of radionuclides with increasing of its activity concentration in combustible materials. Concentration of TUE in smoke aerosols was three order of magnitude lower than for cesium. Activity of Am-241 in aerosols was 1.5-2 times higher than activity of total plutonium. Pu and Am released in small amounts from the source of fire and can be transported only by ash and water particles because of high values of boiling points in comparison with cesium
During forest fire suppression in radioactively contaminated zones firefighters are at risk of additional exposure. The total effective dose can be presented as summary of external and internal irradiation. Dose of external exposure for adults is formed by contaminated forest site: from radioactive soil and vegetation. Due to forest fires, radioactive smoke plume can be an additional source of irradiation.
Calculated data shows that contribution of TUE to the total effective dose can reach 75 %. At the same time, the duration of inhalation intake is lower than the duration of personel staying in contaminated area. Thus, in the longer term, external irradiation from radioactive soil and vegetation will give a more significant contribution to the total dose. The risk of exceeding of annual effective dose limits is low during small fires, such data are of great importance to inform the population and reducing of social and psychological stress in the society, caused by the influence of mass media.

RER 4 / 628

Americium and cesium distribution among sequential extraction fractions in bottom sediments and reed from artificial reservoir

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Artificial reservoir V-17 (Old Swamp) was formed as a result of the construction of dams in 1952 and 1954 (MAYAK Production Association), located in a natural depression relief. Intermediate level liquid radioactive wastes (ILLW) have been discharged to this reservoir since 1949. The water surface area of the lake is 0.13 km2 with the volume of 0.36 Mm3. The maximum depth is 6.5 m with the average value of 2.8 m. Among c.a. 74 PBq deposited to the reservoir, the major portion is concentrated in the bottom sediments. Production Association "Mayak", Chelyabinsk region, Russia, - one of the largest nuclear facility in the Russian Federation.

Bottom sediments are playing the "geochemical barrier" role in the radionuclides migration processes. The radionuclides mobility/availability in bottom sediments depend on the bond strength and absorption mechanisms, which is determined by the radionuclides chemical properties, the state and forms of their presence in radioactive fallouts, the bottom sediments physicochemical features in absorbing complex particular mineralogical and chemical composition, etc. The aim of this study is to identify the 137Cs and 241Am chemical being forms in bottom sediments and reed of the reservoir V-17.

The following samples were collected: reservoir water, bottom sediments, reed (Phragmites australis). Plant samples have been washed out of the soil residues and sediments, air dried and divided into roots, stems and leaves. All samples were analyzed using gamma-spectrometry (Canberra GR 3818). The activity of 137Cs and 241Am in water, sediments and of reservoir-17 in "Mayak" territory were determined. For the sequential extraction, the following procedure was used: 1) Reservoir water imitated salts mixture (watersoluble) ⟶ 2) MgCl2 (exchangeable) ⟶ 3) NH4Ac (carbonates) ⟶ 4) NH2OH·HC (Fe-Mn oxides) ⟶ 5) 30%H2O2 (organic) ⟶ 6) HNO3 conc. (residue).

Results shows the total activity of 137Cs: (1.81 ± 0.06)·101 Bq/ml in water, (1.50 ± 0.01)·105 Bq/g in bottom sediments, (3.20 ± 0.04)·103 Bq/g in roots, (2.3 ± 0.1)·103 Bq/g in leaves, (7.9 ± 0.1)·102 Bq/g in stems. 241Am: (2.1 ± 0.1)·10-2 Bq/ml in water, (5.27 ± 0.05)·103 Bq/g in bottom sediments, (2.4 ± 0.3)·101 Bq/g in roots, (2.7 ± 0.7)·10-1 Bq/g in leaves, (5.0 ± 0.1)·10-2 Bq/g in stems. Maximum activity consider to bottom sediments and reed roots. Minimum activity was determined in water and reed leaves.

Poster RPH / 433

Human serum albumin microspheres labelled with rhenium-188 as the basis for new therapeutic radiopharmaceuticals

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Over the past several decades nuclear medicine turned into an integral part of clinical practice in developed countries. The therapeutic radiopharmaceuticals (RPh), developed and currently used in Russia, are applied for treatment of oncological diseases, but due to their versatility, RPh can be useful in other fields of medicine.

For this purpose formulation development of a medication on the basis of β–emitting radionuclide 188Re as an active substance and human serum albumin microspheres (HSA) as a carrier was carried out.

The benefits of 188Re include the possibility to obtain it in medical institutions in the form of perrhenate ions (ReO4)–by means of the 188W/188Re generator, or in specialized institutions using the extraction technique. In both cases, the obtained substance allows to prepare RPh without a carrier, which is an undeniable advantage and leads to an increase of the medication specific potency.

HSA were chosen due to their biodegradability, affinity to the human body, high stability in vivo and in vitro, and also tight binding of 188Re to a sulfur atom of cysteine in the polypeptide. Various sizes of microspheres in RPh make it possible to create medications with different pharmacodynamic and pharmacokinetic attributes.

We investigated the influence of auxiliary substances on the labelling degree. Since 188Re (VII) is incapable to form bonds with HSA, a reducing agent was additionally used to facilitate 188Re (IV) production. An antioxidant in RPh contributes to reducing the possible negative effects of radiolysis. A transchelator promotes the better incorporation of 188Re into the polypeptide molecule composition by forming a complex in a low stability constant, followed by a re-ligand effect on the polypeptide molecule. An emulsifier improves HSA hydrophilic properties.

In view of the foregoing, we carried out works to study the effect and search for optimum concentration values of auxiliary substances necessary to achieve a labelling yield of at least 95%.
The quality analysis of the finished RPh was carried out taking into account two parameters: pH and the value of radiochemical impurities (the ratio of activity of 188Re ions unbound to HSA to the total activity of the medication). The timing stability of the prepared RPh was also studied; the study was conducted within 72 hours from the time of preparation in physiological solution. It was found out that during this period of time 188Re is tightly bound to HSA and is not released into the external environment.
Thus, this pharmaceutical composition is an excellent preformulation for topical treatment of joint diseases by means of intra-articular administration of the medication.

Poster NFC / 431

Application of microwave radiation for decomposition of uranyl nitrate in the silica gel matrix

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Decomposition of aqueous solutions of uranyl nitrate in a matrix of granulated silica gel (KSKG grade) under the action of microwave radiation (MWR) was studied. Microwave irradiation leads not only to formation of solid decomposition products UO3, UO2(OH)NO3, and their hydrates in pores of KSKG granules, but also to accumulation of gaseous NOx and H2O. The presence of NOx in KSKG pores leads to HNO3 formation in the course of washing of sorbent granules with water. This prevents hydrolysis of uranyl nitrate and formation of UO2(OH)2•H2O in KSKG pores. Washout of uranium with water and HClO4 solutions from the KSKG fraction containing products of decomposition of 2 and 10 g of the initial UO2(NO3)2•6H2O under the action of MWR (hereinafter denoted as KSKG-P-I) was studied. Upon about 7-day contact of the solid and liquid phases at the total ratio S : L = 1 : 20, from 5 to 14% of U passes into the aqueous phase from KSKG-P-I samples obtained in experiments with 10 and 2 g of UO2(NO3)2•6H2O, respectively. In the course of repeated treatments of KSKG-P-I with water, pH of the wash water increased from 3 to 6, owing to removal of NOx from KSKG pores. Then an insoluble phase of uranyl hydroxide UO2(OH)2•H2O, which can also be presented as hydroxylated uranium trioxide UO3•2H2O, is gradually formed from the solution obtained by treatment of KSKG-P-I with water. On treatment of KSKG-P-I with HClO4 solutions (pH 1–2), virtually all uranium species formed by MWR treatment of aqueous uranyl nitrate solutions in KSKG matrix dissolve (at a contact time of the solid and liquid phases of about 21 days, the amount of U that passed into HClO4 solutions is about 90%). The amount of the U form that is not extracted with HClO4 solutions and remains in KSKG granules is about 12% of its initial amount. X-ray phase analysis suggests that the uranium species remaining in KSKG are silicate compounds formed by sorbent saturation with a uranyl nitrate solution and subsequent MWR treatment.

The work on the study was financially supported by the Presidium of the Russian Academy of Sciences (program for fundamental research № 34P “Actual problems of surface physicochemistry and creation of new composite materials” /Academician A.Yu. Tsivaddze/).

Poster RPH / 505

Fully automated GMP [68Ga]GaDOTA-TOC production using TRACERlab® MX module

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The application of 68Ga –labeled peptide for tumor diagnosis is growing very fast in the recent years. One of the traced peptide for tumor diagnosis in nuclear medicine is [68Ga]GaDOTA-TOC. The aim of the work was GMP preparation of [68Ga]GaDOTA-TOC for application in nuclear medicine for a routine diagnosis using automated production with TRACERlab® MX module and quality control according to the standards of European Pharmacopoeia. 68Ga for peptide tracing was obtained from 68Ge/68Ga generator iThemba Labs with initial activity 1850 MBq. 5 mL of 0.6 M HCl elution solution from the generator was collected to the vial and for synthesis of 68Ga labeled peptide a commercial cassette and chemical kit from ABX was used. 68Ga was collected on PS-H+ cartridge, washed and transferred to reaction vials with 5 M NaCl in which 50 µg of DOTA-TOC dissolved in 1.5 M HEPES was prepared. The temperature in the reactor vessels was kept 6 min at 125oC. The solution with reaction mixture was past trough a pre-conditioned Light C18 column and washed with water. Labeled [68Ga]GaDOTA-TOC was eluted with a mixture 1:1 ethanol/water to the vial and diluted with PBS buffer. Prepared solution was transferred to a dispense chamber trough a sterile filter. Decay correcting yield was about 89%. Parameters for quality control (QC) were appearance, pH, radionuclide purity, radionuclide identity, radiochemical purity, HEPES concentration, microbiological quality, bacterial endotoxins and ethanol concentration. All validation parameters of [68Ga]GaDOTA-TOC QC analysis prepared were fulfilled. Radiochemical purity was better than 98.8%. 68Ga impurity in colloidal form was less than 1% and free 68Ga3+ was less than 0.1%. Prepared [68Ga]GaDOTA-TOC was used in patients with neuroendocrine tumours (NET) with better sensitivity and spatial resolution in comparison to standard imaging procedure.

Poster PAR / 444

Production of 51Cr by proton irradiation of natV and purification by precipitation and ion exchange chromatography

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European demand for chromium has grown dramatically, leading to the need for a detailed understanding of recycling of steel sludges and separation methods. To simulate these processes, we will use the radiotracer technique. 51Cr (T1/2 = 27.7 d) was choosen as a radionuclide. The isotope can be produced by the nuclear reaction natV(p,n)51Cr at a cyclotron. We used our recently installed cyclotron Cyclone® 18/9 (IBA) for the irradiation of natV (99.75% 51V). The vanadium foil was put in an aluminium holder with a diameter of 10 mm and a depth of 100 µm. The target was covered by a 100 µm thick aluminium foil. The irradiation was done at a beam of 16 MeV protons and a current of 10 µA for 4 hours. For the separation of 51Cr we established a multistage treatment. After cooling for 20 hours, the vanadium foil was dissolved with 2 ml conc. nitric acid. After addition of 20 mg iron(III) chloride, the hydroxide was subjected to a threefold cycle of precipitation with ammonia and dissolution with nitric acid. Vanadium(V) is soluble under these conditions. The separation of the radionuclide 51Cr and iron(III) was performed by ion exchange chromatography with AG 1-X8 (BIORAD) in conc. HCl. The 51Cr solution was eluted on the resin and the resin was washed six times with 2 ml conc. hydrochloric acid to remove the iron(III). The combined 51Cr solutions were evaporated to dryness and the residue was dissolved in 0.01 M sulfuric acid. The detection of 51Cr was done by gamma-counting (320 keV; 9.91%). The radiochemical yield was 66% at a production rate of 0.575 MBq/µAh.

Poster NFC / 457

The Micro-Optical Ring Electrode: A Sensor for Multiple Actinide Ions Monitoring

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We present on the development of the Micro-Optical Ring Electrode (MORE) for the analysis of transuranium elements in aqueous mixtures. The MORE is a photo-electrochemical device based on a ring microelectrode that uses the insulator interior to the ring as a light guide. This single device exploits the unique photophysical and electrochemical properties of multiple analytes present in mixtures to quantify them. Our study aims to develop a protocol for the analysis of ions of uranium, neptunium and plutonium, which are most relevant to the nuclear industry, especially in the areas of decommissioning and fuel-reprocessing, where speedy and safe identification of radioactive contaminants is essential.

The technique offers a number of advantages over traditional methods in the area of radiochemical analysis such as real time monitoring in-situ, reduced radiation dose impact to the analyst as a result of greatly reduced sample handling and preparation, improved analysis times allowing quick decision making and a much reduced cost (e.g. compared to ICP-MS and Scintillation counting).

Here we present results of our study on successfully generating photocurrents from uranium containing solutions, and describe the response of the sensor as a function of wavelength of illumination, concentration of sensitizer and scavenger species, and include a discussion of limits of detection and interference from additional analytes. The effect of the MORE's surface finish on photocurrents will also be discussed.

Poster RPH / 708

Determining nuclear data of the isomer 195mPt production through neutron irradiation of enriched 193Ir at the reactor IBR-2

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^{195m}Pt is isomeric radionuclide with the great prospect for use in nuclear medicine, due to high yield of Auger electrons, good for small malignancies treatment, mild gamma-radiation for medication distribution control without almost any damage to healthy tissues. These properties perfectly suit personalized approach for treatment of cancer which is developed into theranostics in nuclear medicine, i.e. simultaneous therapy and diagnostics using one particular drug. However no method leading to ^{195m}Pt production with high specific activity in sufficient amounts for reasonable price is known.
In this work production method of double neutron capture of enriched ¹⁹³Ir has been

discussed 1. Only 2 neutron capture cross sections are known out of 6 crucial values. It is impossible to deduce yield of ^{195m}Pt with this set of input data 2.
For calculation of the unknown cross sections the full scale model experiment at the reactor IBR-2 was carried out in Dubna. For this purpose 2 targets of ¹⁹³Ir powder (98.5 enrichment, m, 20 mg each) were wrapped in 2 aluminum foils and put in 2 aluminum containers. Then irradiation with the neutron flux of 2.3x10¹² cm⁻²s⁻¹ during 18 days occurred. Direct measurement of ^{195m}Pt appeared to be difficult because of the enormous radiation background. Therefore dissolution and separation of Ir and Pt isotopes were undertaken. According to the requirements the most feasible option for dissolution was found to be electrochemical technique and extraction chromatography with TEVA resin and HCl eluent for separation.
With the help of the results of ^{195m}Pt radioactivity determination on HPGe gamma-detector Canberra retrieving of previously unknown cross sections was carried out.

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Poster NFC / 509

Volatilisation of 134Cs, 131I and 210Po from lead bismuth liquid metal solutions studied by the transpiration method

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One of the prospective directions in the development of the generation IV reactors is the use of liquid metals as a coolant. At the same time, to ensure compliance with all the requirements, it is necessary to solve many practical problems and challenges. The subject of this work are radiochemical investigations required for licensing of the MYRRHA reactor conducted within the MYRTE-project as part of the EU-Programm for Research and Innovation "Horizon 2020"1. The previous studies on the volatilization of radionuclides from liquid lead-bismuth eutectic (LBE) relevant for the safety of future accelerator driven systems will be extended to measuring the volatilization behaviour of fission products such as Cs, I and also the activation product Po from dilute solutions in LBE. Here, we describe the development and optimization of the samples preparation and experimental equipment for the transpiration method used for volatilization studies, through which it became possible to obtain reliable experimental data. Various series of experiments have been performed using the optimized setup, using 134Cs, 131I and 210Po tracers. In this report, we summarize the methodological development and show results in comparison with existing literature data determined by other authors.

Poster RPH / 819

Recycling of O-18 enriched water used in F-18 cyclotron production.

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Different methods are available for the purification of recovered enriched water from FDG synthesis units. Procedures for organic solvents elimination as: irradiation of the water with UV light, oxidation with KMnO4/NaOH (Rötering et al., 2015), electrolysis (Huang et al., 2003; Weber et al., 2003), ozonolysis (Asti et al., 2007) or He gas purging are proposed. All these methods can remove organic impurities, but they cannot eliminate metallic impurities coming from the [18F]F-targets. Some of the inorganic and metallic impurities can be removed by solid-phase extraction (SPE) methods through ion–ion exchange reactions, IC–Chelate cartridges (Razpotnik et al., 2003), or slow distillation under vacuum (Rötering et al., 2015). All authors report very good results for purification and negligible decreasing of [18F]FDG yields using recycled [18O]H2O but without clear view about generated radioactive waste and radiation safety of involved personal.

A simple, effective, easy-handling and reliable method for recycling of [18O]H2O, used for 18F production, is developed based on co-precipitation, co-crystallization and distillation. Preliminary experiments with normal H2O were used to determine the optimal purification conditions. The obtained recycled [18O]H2O had comparable quality to commercially distributed 18O enriched water. The loss of enriched water was about only 7 % and losses of 18O enrichment about 1.3% ± 0.2%. The method ensures elimination of radioisotopes (99.7% ± 0.1%), organic compounds (99.5% ± 0.1%) and trace metals (99.8% ± 0.1%) with special attention on radiation safety and generated radioactive waste.

Key words: H218O; Purification; Recycling

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Poster PAR / 660

Development of a Switchable Radioisotope Generator

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Alpha induced reactions are being implemented in the imaging and industrial fields. The alpha induced reactions will be implemented in those fields by the patent pending Switchable Radioisotope Generator (SRG). The SRG allows control over the alpha induced reactions by manipulation of the alpha producing material and the target isotope. To prove the effectiveness and practicality of the SRG, analytical calculations, simulations, and experimental measurements were conducted.

Activity estimations were conducted for 9Be and 10B, along with eight other materials, to find the

highest reaction rate. This was to insure large error propagation would not occur when taking background radiation into consideration. 9Be was selected for alpha irradiation due to the high alpha capture cross section and low cost compared to other materials. Neutron Spectroscopy was used to determine the spectrum of neutron energy produced between 241Am and 9Be, with a preliminary measurement yielding a positive spectrum.
Simulations done in MCNP include the experimental setup to measure angular and energy dependence of neutrons. Cross section libraries were obtained through ENDF, then processed through NJOY in ACER format, such that they may be read into MCNP. MCNP6.1.1 Beta was used for initial simulation studies, but was found to process input files too slow. MCNP6.1.1 Beta was rebuilt and compiled through MCNP6 source code to enable Message Passing Interface (MPI), which allows for variance reduction by using multiple processors and faster processing times. The alpha source and target material geometry was defined in the MCNP input card, along with material compositions, data directories for the XSDIR file, and tallies for the energy and angular dependence of neutrons. The input cards were fed into MCNP6.2 using MPI. Preliminary results show a strong bias in forward scattering with energies ranging from 50-150 keV. Results from the final simulations were compared to the experimental data for conclusions and validation of the SRG.
The analytical calculations, experimental results, and simulation studies show how the SRG will function, and provide data validation for analytical calculations done in databases such as ENDF. The combination of the work performed will determine if the SRG is feasible in the imaging and industrial fields. The final results obtained from this project will determine if applications of the SRG will change.

Poster NFC / 513

Stability investigations of actinide doped calcium silicate hydrate phases in highly saline electrolytes

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To evaluate the retention potential of the concrete barrier in a nuclear waste repository towards actinides in the presence of high saline water, leaching experiments with actinide-doped calcium silicate hydrate (CSH) phases were performed in highly saline electrolytes. Therefore, U(VI)- and Cm(III)-doped CSH phases with different C/S ratios (1.0-2.0) were synthesized directly in presence of either U(VI) or Cm(III) and characterized by time-resolved laser-induced fluorescence spectroscopy (TRLFS), infrared (IR) spectroscopy, powder X-ray diffraction (XRD), thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), and scanning electron microscopy (SEM). The time-dependent release of Ca, Si, U, and Cm from CSH phases into brines containing either 2.5 M NaCl, 2.5 M NaCl/0.02 M Na₂SO₄, 2.5 M NaCl/0.02 M NaHCO₃ or 0.02 M NaHCO₃ for U(VI)-doped CSH phases or 2.5 M NaCl/0.02 M NaHCO₃ or 0.02 M NaHCO₃ for Cm(III)-doped CSH phases was monitored in batch leaching experiments for 30 or 60 days, respectively. Subsequently, leaching induced changes of the CSH structure and of the U(VI) or Cm(III) coordination environment were investigated with TRLFS, IR spectroscopy and XRD.

Site-selective TRLFS studies of the Cm(III)/CSH binding at 8 K revealed a luminescence line-narrowing effect, which could be ascribed to the presence of a continuum of similar curium species on two different sorption sites in the CSH interlayer. The leached CSH phases showed pronounced differences in terms of decomposition behavior and actinide release depending on their C/S ratio, leaching electrolyte, and incorporated actinide. U(VI)-doped CSH phases leached in 2.5 M NaCl showed an increased release of Ca and a U(VI) precipitation as uranophane while the leaching in 0.02 M NaHCO₃ led to a mobilization of U(VI) as an aqueous Ca₂UO₂(CO₃)<sub> species.

In contrast, Cm(III) was not mobilized by NaHCO₃ but incorporated into newly formed secondary phases like calcite and vaterite as detected by site-selective TRLFS. The comparison between leaching experiments performed in 0.02 M NaHCO₃ and 2.5 M NaCl/0.02 M NaHCO₃ revealed that the presence of 2.5 M NaCl increases the carbonate-induced

U(VI) mobilization from CSH phases with higher C/S ratios while no influence on the Cm(III) retention was detectable.

Poster NFC / 862

Oxidative dissolution of compounds present in corrosion layers in primary circuit stainless steels

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Dissolution of corrosion layers is strongly connected with the problem of decontamination and decommissioning of nuclear facilities, which has become of the current issues in many countries. The key process to decontaminate metals, is to dissolve surface corrosion layer and remove radionuclides. From corrosion studies on the surface of stainless steel under primary circuit conditions of pressurized water reactors, can be concluded, that the dissolution of the corrosion layer is associated with the oxidation of insoluble built-in Cr3 + to soluble Cr6 +. Because of this fact, the experiments of dissolving corrosion layers started with dissolution of chromium (III) oxide.
The oxidation and dissolution of common corrosion layer compounds was investigated in acidic environment of HBF4 and H2SO4 at various concentrations and temperatures. As tested corrosion layer compounds, chromium (III) oxide, magnetite and several spinel minerals synthetized to substitute spinels created in primary circuit of nuclear power plant during the operation were used; the dissolved metals were measured by Atomic Absorption Spectroscopy and ICP-MS. The results show that it is possible to corrupt and dissolve highly stable corrosion layer compounds with elevated content of chromium.

Poster NFC / 856

Prospective of syntetic K-Struvite as solid radioactive waste matrix

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Recent years some alternative techniques are being developed for solid state matrixes producing for immobilisation of liquid and mixed state radioactive wastes. Magnesium-potasium-phosphorus ceramics (MPP ceramics) matrix are considered to be a promising solid state matrix for immobilisation of liquid and mixed state radio-active wastes as alternative to cement and glass matrix. The major advantage of this matrix is low leach rates since it is a structure analogue of natural mineral K-struvite (MgKPO4•6H2O). Another technological advantage is low temperature synthesis technique in comparison with the glass form production.
There are some difficulties in MPP ceramics syntesis. First of all it is high reac-tion rate and heat emission during reaction. High amount of heat produced during synthesis could cause water boiling and water loss while it is necessary for dissolving sodium hydrophosphate. It could be problematic to stir fast large amounts of reaction mixture, especially in case of heterogenic radioactive wastes. In this regard method of additions could be useful. In this case small portions of chemicals are constantly added to the reaction volume until the required ceramics volume is met.
Species of MgO play important role in this process. MgO should be low soluble and provide low rare leach of cation to the liquid phase during formation process. Heating MgO at 1200-oC provides

formation of crystalline phase of periclase which is stable and promoting for ceramic synthesis. Usage of amorphous MgO is possible only as dry powder course it hydrolises while being put in water with forming low binder component. The lack of water in the reaction mixture causes formation of K2Mg3(P2O7)2(H2O)10 phase in addition to MgKPO4•6H2O while exceed of water cause formation of Mg2KH(PO4)2(H2O)15 phase.
Powder Roentgen diffraction of MPP ceramic samples revealed that oxide com-ponent added in system in stechiometric ratio does not react in total and some amount is left in the system as inert filler. In this case it is promising to replace part of MgO with mineral additions to increase durability of material.
Keeping MPP ceramic samples for one year decreases leach rate to 10-6-10-5 g/sm2*day that is comparable with phosphate glass and is one order of magnitude less than leakage rate of cement matrix. It is shown that durability of MPP ceramics increases with time.
During development of MPP ceramic production major criteria such as content and component's addition scheme, technic and equipment requirements for produc-tion were determined. As a result of applying of test facility 300m3 of low level radi-oactive wastes was produced and encapsulated in containers.

Poster NFC / 539

Immobilization of high level waste in magnesium-potassium-phosphate compound

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Mineral-like magnesium-potassium-phosphate (MPP) compound samples were synthesized during the immobilization of a high level waste (HLW) simulator. The phase composition and structure of compounds were studied and their physico-chemical properties, including mechanical and thermal stability, were determined. The high hydrolytic stability of the MPP compound was established: the differential leaching rates at the 90th day of contact with water under 23 0C were for 137Cs - 4.6·10-7, 90Sr –9.7·10-7, 239Pu - 3.7·10-10 and 241Am - 9.6·10-10 g / (cm2 / day). The leaching of radionuclides during the first 2 weeks was caused by the dissolution or washing out from the surface of the compound, when the experiments were being continued for 90 days the leaching was done by the depletion of the surface layer. The permanence of the phase composition, structure, and hydrolytic stability of MPP compound irradiated by an electron beam (absorbed dose -1 MGy) was established, it means that the MPP compound is stable to radiation. The quality profile of MPP compound corresponds to current regulatory requirements, which make possible to consider the compound as perspective material for industrial solidification of HLW.

Poster RPH / 868

Surface modification of 99mTc-HAp-NPs

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Hydroxyapatite (HAp) is one the main mineral bones and teeth components and biocompatible material used in orthopaedic implants. It might be used as a drug carrier in diagnostics and treatment of cancer. The sufficient in vivo stability without fast aggregation of the Hap-NPs is required for theranostic application. Various phosphonic acids were studied as potential HAp stabilisers and

technetium-99m chelator.
Hydroxyapatite nanoparticles were prepared by precipitation of Ca(NO₃)₂ with (NH₄)₂HPO₄ at pH=11. The precipitate was washed, lyophilized and crushed. Stabilized samples were prepared from already-made HAp-NPs by ultrasound dispergation in corresponding phosphonic acid solution in water (1 mg/ml).
The hydrodynamic size distributions of studied stabilized particles were determined using dynamic light scattering (Zetasizer, Malvern, UK). Samples were labelled with technetium-99m eluted from ⁹⁹Mo/^{99m}Tc generator (DRYTEC, GE Healthcare). The labelling yield ranged to about 90 %. Subsequent in vitro stability studies were carried out in bovine serum, bovine plasma, saline and 5% albumin solution. Measurements of released activity revealed that samples exhibit the highest stability in saline (released activity of about 10 %). The lowest stability was shown to be in blood plasma (released activity of about 20 %).

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Poster NFC / 869

The effect of dissolved hydrogen on the oxidative dissolution of highly Pu-doped MOX and externally irradiated UO2

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The effect of dissolved hydrogen on the oxidative dissolution of highly Pu-doped MOX and externally irradiated UO2

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The release of radiotoxic species from future final geological nuclear waste repositories is governed by the oxidative dissolution of the UO2-matrix in the case of a canister failure. At the depths of a final geological repository, the conditions are reducing with very low oxygen concentrations 1. In repositories built in granitic bedrock, as in the designs planned in Sweden, Canada and Finland, anoxic conditions are further ensured by the redox chemistry of the copper canister as well as with the surrounding biotite, magnetite and organic matter 2. As the UO2 matrix is highly insoluble in the U(IV) state 3, the main mechanism of dissolution is through the formation of locally oxidizing conditions near the fuel surface through radiolysis. Even though radiolysis produces both reducing and oxidizing species, as both radicals (H·, e-aq, HO2·, O2·, ·OH) as well as molecular products (H2, H2O2) 4, the net redox conditions due to radiolysis are oxidizing at the immediate surface due to the higher reactivity of O2 compared to H2 [5]. Dissolution of the UO2 matrix leads to the release of radiotoxic fission products, which are to a large extent (>95%) contained in the matrix [6]. H2, which is formed in substantial amounts in the final repository through anoxic corrosion of the massive iron

inserts, has in several studies been shown to protect the UO2-surface from radiolytic oxidation. The hydrogen effect is propagated through activation on the radioactive UO2 surface on active sites, or on metallic ε-particles (Mo, Pd, Ru, Rh and Tc).
It is not yet known if the radioactivity must be internal for the active surface sites to be present on the UO2-surface. This has been investigated using 241Am-sources separated from low-enriched UO2-pellets by 30µm using glass-threads in a 10mM NaHCO3-solution to externally irradiate the water volume close to the surface. The radiolytic oxidation of the UO2-matrix is studied under Ar- and H2-atmospheres. The dissolution of highly Pu-doped MOX has also been investigated, under both Ar- and D2-atmospheres and the extent of which the hydrogen effect protects this surface will be presented.

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Poster NFC / 600

Modeling of technological processes for their optimization and designing of industrial facilities in SNF reprocessing.

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Nowadays computer models of fuel fabrication/refabrication, SNF reprocessing and waste management processes are under development as part of technologies front-end and back-end of closed fuel cycle. One of them is software package "VIZART"[1,2]. Code "VIZART"is used for material flow calculations of technological schemes and dynamic calculations of equipment work, taking into account capacity, transfer time between apparatuses and volumes of storage vessels. Results of such calculations are needed for development of initial data for equipment and facilities design.
Developed mathematical models are used to calculate regular transition and emergency modes, mass-dimension characteristics of equipment items, working hours and resources. The codes of individual processes in SNF reprocessing are included in "VIZART"as stand-alone modules and can be used both for calculation of technological modes and clarifying of coefficient and parameters of operation balance and dynamic calculations.
For example, model of linear crystallizer 3 was used to choose its optimal working mode for the crystallization refining of uranyl nitrate hexahydrate (UNH) melts in 2017. Temperature of crystallization, composition of initial solution of UNH melt, composition of washing solution, flow rates can be varied in this mathematical model. The yield of main component into crystals and its flow distribution also can be calculated. Optimal composition of initial solution, the temperature of crystallization process in purpose getting maximum yield of UNH crystals in experiments were chosen on the base of pre-test calculations. The estimation of filling time for crystal receiving vessel can be calculated by this mathematical model, as well. Knowing the filling time helped to create a working timeline for crystallizer. Further analysis of obtained experimental data and values calculated with mathematical model showed discrepancy of values on an average no more than 5 %.

Obtained results allow to clarify the parameters of dynamic calculations in «VIZART» of U-Pu-Np mixture crystallization refining. Working time and volumes of solutions in storage vessels were determined, as well.
Nowadays there are more than 30 individual models of main operations in hydrometallurgical and pyrochemical SNF reprocessing.

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Poster NFC / 852

Approaches to inner safety barriers development

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Currently proposed concepts for safe isolation of radioactive wastes imply development of a multi-barrier system which consists of both natural and engineered barriers. Hosting rock of depository serves as a natural barrier. It appears for already-existing depositories and disposal facilities. It is naturally chosen on the stage of disposal development. Vertical and horizontal curtains of polymeric materials, clay back filling, slurry-type walls appear for engineered barrier. It serves as impervious barriers for filtration prevention. Besides there is another type of engineered barrier for prevention of contaminants migration it consists of both organic and inorganic sorbents, biosorbents etc.
Technology of inner barriers creation is determined by both geometric parameters of repository and species of solid or solidified radioactive waste will have been placed therein. Nowdays Russian Federation has the experience of constructing an inner safety barrier system namely when industrial uranium-graphite reactor (PUGR) decommissioning. As barrier material one used a mixture of three components: kaolinite (Kampanovskoye deposit), bentonite (Khakass deposit), vermiculite (Tatar deposit). It was a composition design what reveals high sorption activity being cost effective. Besides chosen fractional composition possessed quite high index of powder flowability. The total volume of involved barrier material amounted more than 40 thousand m^3.
Another type of inner barrier was developed for reactor storage of solid waste. This type of storage presumes a hodgepodge of solid radioactive wastes of irregular shapes made of graphite, metal, organic compound and so on. In this case clay suspension in gel (thick slurry) was offered to play a role of barrier materials, which was injected into storage room. Such hind of barriers has high penetrability and prevents migration of radionuclides. Clays from the local depository near Tomsk (Kornilovo) were chosen as a component of barrier material along with the mixture of clays which was either used as back filling for reactor. Optimal solid-liquid ratio was 2:1. Large-scale experiments confirmed the possibility of storage room full filling without any cavities left.
The developed approaches to inner safety barriers construction allow researches to choose the appropriate technology of their creation depending on the type of the object and its technical features.

Poster NFC / 649

The study of the behavior of coatings based on silica-organic com-

position OS 51-03 of the containment during severe accidents at NPP

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The work describes an experimental study of the determination of chemical stability of protective coatings under conditions of severe accidents at nuclear power plants. Another task of the work was to evaluate the thermal degradation of the coatings and the analysis of the resulting solids (debris), which can cause overload of cooling systems, as well as be a source of release of combustible gases.
To ensure reliable corrosion protection of the inner containment shell was used the protective coating including plating the steel surface of the shell is aluminum, with the subsequent drawing on it organic silicate composition OS-51-03. This coating has been proven as stable in chemically aggressive environments and in streams of radiation at temperatures up to 200 ℃.
In conditions of a severe accident environment inside the containment shell becomes highly aggressive because of high temperatures, pressure and high radiation levels. When conducting an experimental study of coating parameters for such a corrosive environment was simulated dynamically.
In the course of the work have been obtained and investigated technology solutions, determined the by their viscosity and density taking into account the dynamics in the course of a severe accident. Was determined the particle density of the chemical debris and conducted their sedimentation analysis to determine fractional size. Also was evaluated the corrosion and hydrogen security of operation of a covering veneer protective shell.
The data obtained allow to justify the choice of type of coverage and its compliance with requirements of regulatory authorities. The behavior of the organic silicate composition OS-51-03, was applied on a the aluminum surface of the protective shell of a nuclear power plant, in conditions of a severe accident can be described using the obtained data. Composition OS-51-03 has shown itself resistant to high temperatures, pressures, high radiation levels, aggressive liquid and gaseous media, so it is recommended for use as a protective coating for the design, construction and operation of NPP.

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Poster NFC / 834

Uranium bioremoval from liquid radioactive waste in alcaline conditions using different types of microorganisms

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Liquid radioactive waste (LRW) represent serious environmental problem due to their multicomponent composition: presence of long-lived radionuclides with variable oxidation states as well as nitrate ion in high concentrations. One of the most problematic types of LRW are alkaline waste generated in the PUREX process of spent nuclear fuel recovery. Annually are generated about 6490 m3 of alkaline LRW with pH>9, which contain uranium in concentration up to 3 g l-1 and sodium carbonate in concentration range 80–150 g l-1. The aim of the present work was to use microorganisms: bacteria Halomonas mono and cyanobacterium Arthrospira (Spirulina) platensis, isolated from extremely alkaline habitat for uranium removal from synthetic LRW. Elemental composition of microbial biomass was determined using neutron activation analysis at the pulsed fast reactor IBR-2 (FLNP JINR, Dubna). Biosorption experiments showed that bacteria Halomonas mono possess higher maximum biosorption capacity for uranium in comparison with cyanobacterium Arthrospira (Spirulina) platensis. The kinetics of uranium sorption onto studied microorganisms follows a pseudo-first order rate equation. The strong involvement of carboxyl, hydroxyl, carboxyl and amide groups in studied metals binding was ascertained by FTIR analysis and potentiometric titration. Bioreduction studies carried out with Halomonas mono cells showed highness of uranium reduction in alkaline conditions, resulting in the bioreduction of 64% of uranyl ions. Results of geochemical modeling showed formation of uraninite by Halomonas mono. It can be concluded that bacteria Halomonas mono can be efficiently applied for uranium removal from liquid alkaline radioactive waste by means of biosorption and bioreduction processes.

Poster NFC / 693

Selective separation of neutron poisons from spent nuclear fuel using a carboxyl-functionalized ionic liquid

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Reprocessing of spent nuclear fuel became the key issues for the environmentally friendly and sustainable development of nuclear energy. For the future development of nuclear energy requirements, the concept of an accelerator-driven advanced nuclear energy system (ADANES) has been proposed in China. ADANES consists of a burner system and a fuel recycle system. The waste transmutation, breeding, and power production are implemented in the burner driven by the neutron source outside the accelerator. It is not necessary to carry out the fine partitioning for minor actinides due to the higher-power neutron source. Hence, in this fuel reprocessing, only part of the fission products containing neutron poisons from spent nuclear fuels will be separated and disposed. The rest spent nuclear fuel, uranium dioxide along with transuraniums including Pu, Np and Am will be regenerated as new nuclear fuels and burned subsequently.

Neutron poison is a substance with a large neutron absorption cross-section in nuclear reactors, such as xenon-135 (σ = 2,000,000 b (barns) and samarium-149 (σ = 74,500 b). Actually, some lanthanides or rare earth elements, which accounts or about 1/3 of the fission products, have large neutron absorption cross sections and are neutron poisons. So these lanthanides should be separated from spent nuclear fuel before the next transmutation step.

In this work, water-saturated [Hbet][Tf₂N] was used to selectively dissolve rare earth oxides from the simulated spent nuclear fuel, leaving most of the valuable uranium oxides as solid in fuel matrix. The selective dissolution of rare earth elements is driven by the reactivity of the carboxylic acid group located on the cation of the ionic liquid under the appropriate experimental conditions. The lanthanide oxides dissolves easily at 40 ℃ with complete dissolution in a short time while UO₂ have very low solubility at this temperature with negligible amounts being dissolved. The dissolved rare earth metals by [Hbet][Tf₂N] will be in solution phase and the rest undissolved solid particles are uranium dioxides. After centrifugal separation, the valuable and reused uranium oxides or most spent nuclear fuel will be obtained. Rare earth metals dissolved in [Hbet][Tf₂N] can be stripped with an acidic solution. Such a selective dissolution system that automatically separates the rare earth metals including some other fission products is innovative and applicable to spent nuclear fuel reprocessing.

Poster RPH / 850

Hydroxyapatite nanoparticles labelled with 18F

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Hydroxyapatite based materials including nanoparticles (HAp-NPs) are widely used in medicine because of their stability in various biological media 1. The HAp-NPs also belong to promising drug carrier systems for medicinal radionuclides such as F-18 as the part of multipurpose theranostic system with other therapeutic and diagnostic radionuclides 2. HAp-NPs can show uptake by cancer tissue due to EPR effect, therefore diagnostic and therapy of the diseases are expected to be very accurate and beneficial.
Fluorine-18 was obtained in a form of irradiated water for medicinal purposes and was used without further purification. The labelling was performed with ready-made HAp-NPs (5 mg) in 0.9% NaCl solution. Labelling was carried out with pristine NPs and NPs decorated with selected ligands: NPMIDA and IDMP (0.05 mg). Experiments with pristine HAp-NPs have shown F-18 uptake with maximal yield of about 93% (15 min incubation), while NPMIDA-HAp-NPs and IDMP-HAp-NPs have shown yields 99% and 98% respectively. Further experiments have shown that uptake kinetics is quite fast (over 90% yield within 5 minutes). *In vitro* stability experiments have shown good stability of 18F-HAp-NPs nanoconstruct in a bovine blood serum and plasma (more than 90% of activity remained bound to the NPs in 7,5 hours), and slightly worth results in 5% albumin solution and physiological saline (under 90%). NPMIDA and IDMP decorated nanoparticles have shown good stability in a physiological saline (over 95% of bound activity in 6 hours), between 65 –80% in serum and 40 –50% in 5% albumin solution.
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Poster NFC / 720

The influence of accelerated electrons on low flammability limits for extraction mixtures based on light hydrocarbon diluent – isopar M

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During reprocessing of spent nuclear fuel by PUREX process uses an extractant that consist of a mixtures of TBP in a light hydrocarbon diluent. In the Russian industrial practice mainly used saturated aliphatic hydrocarbons, but currently discusses the possibility of using branched-chain hydrocarbons, which have advantages properties over the first. Because in the technology process extractant is exposed by radiation in them proceed radiation-chemical processes that lead to the formation of degradation products and changes of the extractant properties. One of the important parameter from the point of view of explosion safety of technological process –the lower flammability limit (LFL) which defines admissible temperature for use of combustible liquid. The goal of this study was to identify the effect of ionizing radiation on LFL of the extraction mixture with a light hydrocarbon diluent - Isopar M. For this purpose the extraction mixture "30% TBP in the diluent Isopar M" was investigated and LFL were determined for "fresh" (75 C) and irradiated mixtures. The method of determining LFL was based on the standard [ISO 4589-84]. Ionizing radiation was simulated by using an electron accelerator with electron energy about 8 MeV and the irradiation was carried out up to absorbed doses of 2 MGy. In this study were tested the mixtures that were pre-saturated by nitric acid before irradiation and mixtures without saturation. For the purpose of regeneration of the extraction mixture, soda flushing, air bubbling and steam distillation were tested.
In this study were tested the mixtures that were pre-saturated with nitric acid before irradiation and mixtures without saturation. For the purpose of regeneration of the extraction mixture, soda flushing, air bubbling and steam distillation were tested.

Poster NFC / 757

Features of radionuclides sorption by bentonite clays

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Bentonote clays are widely used as engineering barrier materials for radioactive waste disposal into geological formations due to their low permeability, considerable sorption capacity in relation to radionuclides, good swelling ability, heat resistance, thermal conductivity. Nowdays several bentonites deposits in Russia and
CIS countries are currently being developed that are Zyryanskoe (Russia), 10th Khutor (Russia, Khakassia republic), Dashkovskoe (Russia), Taganskoe (CIS, Kazakhstan).
Possible transformations of bentonite barrier properties, which can happen when isolating liquid radioactive waste, were modeled by the treatment with concentrated nitric acid at 90 ° C during 5 hours.
Sorption of microquantities of 90-Sr, 137-Cs, 233-U, 241-Am was held from aqueous dispersions (liquid to solid ratio 1:100) and pH magnitude at 7.2-8. Results of sorption experiments are shown in Table 1.

Table 1. The results of radionuclides sorption on bentonite samples from distilled water
Bentonite sample Condition of samples Radionuclides adsorption (S,%) Specific surface area, m2 / g
90 Sr 137Cs 233U 241Am
Taganskoe natural 95 83 95 99 110,2
acid modified 99 98 98 99 280,1
Dashkovskoe natural 94 95 81 97 63,9
acid modified 99 98 99 99 135,4
10th Khutor natural 89 89 74 99 13,33
acid modified 94 89 95 99 95

Zyryanskoe natural 89 89 85 99 44,8
acid modified 99 96 96 96 192,98

Adsorption values of thermo-chemically modified bentonite samples appeared to exceed the values of adsorption for natural samples. Therefore the adsorption degree for 137Cs on natural Taganskoe bentonite is 83%, whereas on modified one it is 98%.
The increase in radionuclides sorption on modified bentonite clays samples is likely due to the transformation of surface properties. Acid treatment of bentonites resulted it washing the octahedral cations out as well as dissolvation of presumable organic films on the surface of the montmorillonite particles. That leads to an increase in the specific surface area and the appearance of relatively large pores. (Zakusin et al., 2015, Krupskaya et al., 2017).
Speciation of radionuclides were investigated by sequential extraction method.

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Poster PAR / 756

Production of positron emitter radionuclide 68Ga via generator 68Ge → 68Ga

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68Ga (T1/2 = 68 min) is used for molecular imaging in positron emission tomography. 68Ga decays via 1.92 MeV positron emission (89%) and electron capture (11%). Due to suitable half-life for radiolabeling and easy availability from radionuclide generator, interest in 68Ga labelled compound has increased.
The most convenient method of 68Ga production is usage of 68Ge → 68Ga radionuclide generator. Currently available commercial generators have number of disadvantages and need to be improved. The shortcomings are the usage of hydrochloric acid, large volumes of eluate and the presence of low-charge metal cations impurities, which prevent the formation of gallium complexes during labeling.
In this study the sorption behavior of Ge on ion-exchange resins in various solutions of oxalic and hydrochloric acids are carried out. The optimal scheme for efficient separation of germanium and gallium was also developed. The scheme is based on anion exchange resin and mixture of the acids C2H2O4 and HCl. Three generators 68Ge → 68Ga and two elution methods (direct and reverse) were tested. Further use of the tandem column will significantly improve the quality of the resulting product and will enable the selection of conditions for direct labeling.

Poster NFC / 761

Characteristics of Decontamination Foams Containing Various Chemical Components

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It is required to secure decontamination technology that is easy to apply and minimizes the generation of secondary wastes for large-scale equipment and large-area decontamination, which are difficult to be applied structurally when massive repair or decommissioning of nuclear facilities. In this respect, decontamination foam has a potentially wide application in the removal of contaminants from metallic walls, overhead surfaces, and elements of complex components and facilities. The decontamination efficiency can be enhanced by improving the contact time between the chemical decontamination agents and the contaminated surface through the introduction of stabilizers into the decontamination foam.
In this study, the stability of a decontamination foam containing mesoporous silica nanoparticles as a stabilizer and the decontamination performance of radioactive isotope contaminated specimens were evaluated. A process for waste liquid treatment after foam decontamination was also proposed. Decontamination performance tests using RI contaminated specimens were carried out in a decontamination foam prepared only with a solution containing only 1 wt% EM100 surfactant and in a decontamination foam prepared with a solution containing 1 wt% EM100 surfactant and 1 wt% various silica nanoparticles. The stability of the decontamination foam was increased due to the addition of various types of silica nanoparticles. In particular, the liquid film thickness of the decontamination foam containing mesoporous silica nanoparticles was the largest. As a result, the decontamination coefficient for RI contaminated specimens increased as the thickness of the foam liquid film increased due to the longer contact time between RI contaminated specimen and the chemical decontamination agent as the thickness of the foam liquid film increased.

Poster NFC / 523

Diffusion and sorption of radionuclides in veined gneiss from Olkiluoto, Finland

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Diffusion and sorption of radionuclides in veined gneiss from Olkiluoto, Finland

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A project "rock matrix REtention PROperties"(REPRO) at ONKALO, the underground rock characterization facility in Olkiluoto, Finland, consists of extensive series of in situ sorption and diffusion experiments that are supplemented by laboratory work 1. A supporting laboratory Through Diffusion Experiment (TDElab) described here aims to study transport of 3H, 36Cl, 133Ba and 134Cs in Olkiluoto veined gneiss with strong foliation. The non-sorbing elements 3H and 36Cl provide data on diffusion coefficient of the rock while the sorbing nuclides, 133Ba and 134Cs, provide also data on distribution coefficient. Solution with known activities of 3H, 36Cl, 133Ba and 134Cs was injected into injection hole in the rock sample (length 29 cm; diameter 29 cm) and their diffusion to four observation holes were monitored weekly by collecting water samples from the observation and injection holes. The water samples were analyzed using beta and gamma measurements. The observation holes located 1 cm and 4 cm away from the injection hole so that main transport directions were perpendicular and parallel to foliation.

After 39 weeks, 46 % of initial 3H and 27 % of initial 36Cl were intruded into the rock while the intrusion of initial 133Ba and 134Cs were 82 % and 95 %, respectively. Diffusion of all radionuclides was fast along foliation that indicates plausible fractures along the foliation. A breakthrough of all four

radionuclides was observed at the distance of 1 cm from the injection hole, along the foliation. No diffusion of sorbing radionuclides was observed at a distance of 4 cm in observation hole perpendicular to foliation. Diffusion of non-sorbing 3H and 36Cl was clearly slower perpendicular to foliation. The results of TDElab will be interpreted by modelling with Time Domain Random Walk (TDRW) method 2. The effects arising from diffusion, sorption, structural and mineralogical heterogeneity and foliation can be taken into account with the TDRW method.

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Poster NFC / 536

Formation and transformation of physical and chemical iodine forms by foreign substances in the primary coolant

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Iodine radionuclides largely define radiation hazard at nuclear power plants under both normal operation and emergency. The factors that determine the high level of potential hazard of iodine are its volatility and a possibility to remain in various physical and chemical forms such as iodide (I-), molecular (I2), iodine organic, for example, methyl iodide (CH3I), iodate (IO3-) or periodate (IO4-). The problem of accidental release confinement requires considering the efficiency of capturing various physical and chemical forms of iodine using the existing methods. The most hardly perceptible are iodine organic compounds, in particular, iodide methyl that accounts for the main proportion of radioiodine organic fraction.

The life-time tests of naval marine nuclear prototype investigated the content of volatile forms of the iodine radionuclides in gas phase that are formed during the the primary coolant degassing while sampling. It was considered that partitition coefficient of volatile organic forms of iodine (primarly, iodide methyl) in water/gas system was from 3 to 5; partitition coefficient of volatile inorganic forms (primarly, molecular iodine) was from 20 to 200. The volatile physical and chemical forms passing from the coolant into a gas phase during sample degassing are defined only for the most long-lived radionuclides of iodine 131I and 133I. The fraction of volatile organic and inorganic forms of the iodine radionuclides passing into a gas phase does not exceed 3,3·10-2% and 2,5·10-4%, respectively. Also, the life-time tests defined the fraction of iodate ions in the primary coolant. It was found that the fraction of iodate ion did not exceed 3,2% under steady-state operation of prototype facility, and after filtration process using ion-exchange filters the fraction considerably increased which can be indicative of various coefficients of iodide and iodate purification. The main form of iodine nuclides in the primary coolant is iodide form. The observed deviation of coefficient was related with reactor operating modes and disabling of filters. Content of the iodine radionuclides (as iodate ions) in the primary coolant depends not only on the reactor power operation, but also on concentration of ammonia. The regularities of iodate ions formation for 131I differ from regularities of iodate ion formation for other iodine nuclides (132I, 133I, 134I, 135I). It can be related with large life-time of 131I nuclide in the coolant. A certain dependence on ammonia concentration, and, accordingly, on pH is observed for 132I, 133I, 134I, 135I iodate ions. The content of 132I-135I iodate ions did not exceed 2-3% and did not depend on pH oscillations within admissible values of actual water chemistry.

The research of iodine radionuclide transformation in the coolant has been conducted at reactor power cutback 60% → 10% → 0%. The content of iodates in the coolant upon the first power transient did not change. The volume of iodates has considerably increased at reactor power reduction at the second transition to minimum controlled power level. After the reactor shutdown, the content of iodates of 131I and 133I has increased almost to 25%. This growth can be explained by the change of oxidation-reduction potential and decrease in coolant temperature when the reactor is shutdown.

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Enhancing the technology for chemical washing of reverse osmosis modules used in treatment and evaporation of liquid radioactive waste

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Over the last decades, membrane methods (especially reverse osmosis) have been increasingly used for treatment of liquid radioactive waste (LRW). The reverse osmosis methods are most efficiently used in combination with microfilters and ion exchange filters. Based on experience of using the NITI-designed modular sorption membrane system (MSMS) in LRW treatment and concentration process, an automated version of MSMS, MSMS-A, has been developed, which includes an automated reverse osmosis module. A drawback of the membrane technology in water treatment applications is that reverse osmosis elements are often clogged with low soluble calcium and manganese carbonates, iron hydroxides, and other residues. This reduces the efficiency of the reverse osmosis module and the quality of filtrate. 95–97% of quality reduction is determined by fouling of the membrane surface and only 3-5 % by densification of the membrane capillary porous structure. Moreover, contamination of membranes leads to an increase in gamma exposure rate from the reverse osmosis equipment, thus causing troubles to maintenance. Therefore, it is of current importance to improve the process of regeneration washing of reverse osmosis elements.
The authors investigated the efficiency of regeneration solutions prepared with phosphoric acid, ethylene diamine tetra-acetic acid (EDTA), citric acid, and oxalic acid. The phosphoric acid was shown to be poorly efficient in washing the reverse osmosis elements, as compared to organic acids. The choice among organic acids was based on the consideration that radioactive wastes generated from washing would be solidified by cementing. EDTA deteriorates the quality of cement compounds. The oxalic and citric acids form low-solubility compounds with calcium contained in cement clinker. In addition, they are commonly used as agents in preparation of decontamination solutions. Based on results of testing the citric acid solution as the most efficient and least toxic substance was chosen for chemical washing.
Testing of the spent washing solution has shown that iron compounds and other corrosion products make up to 85% of washed-off residues and the remaining are magnesium, calcium, and aluminum compounds. An automated module for washing the reverse osmosis elements has been developed with the purpose to ensure radiation safety of the process. Control of operation of the automated reverse osmosis module and module for washing reverse osmosis elements is provided from operator workstation. Spent washing solutions are drained to LRW tank. The portion of acid that has not reacted during washing process serves as a sludge inhibitor, mitigating fouling of reverse osmosis elements in next cycles of LRW treatment. This prevents sludging in the tank itself.

Poster NFC / 823

Competition of U(VI) hydroxide and carbonate complexation in alkaline solutions – implications for U(VI) retention by Ca-bentonite

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Thorough understanding of the aqueous complexation of U(VI) with ubiquitous inorganic ligands, such as hydroxide and carbonate, is crucial for predicting U(VI) mobility in natural and engineered systems, since retardation processes largely depend on the metal speciation. U(VI) hydrolysis 1 and complexation with carbonate in weakly alkaline media 2 have been extensively studied. This work, however, systematically elucidates the U(VI) speciation in (hyper)alkaline solutions, where OH- and CO32- can occur in approximately equimolar quantities and compete for the complexation with U(VI). Such (hyper)alkaline conditions can evolve within deep geological repositories for radioactive waste by corrosion of concrete. The effect of pH-dependent changes in speciation on U(VI) sorption affinity was investigated by batch sorption experiments with Ca-bentonite, which is considered as buffer and backfill material within such repositories. Spectroscopic measurements provide information on the underlying retention mechanisms on the molecular level.
Time-resolved laser-induced fluorescence spectroscopy (TRLFS) proofs the formation of (calcium) uranyl carbonate complexes in aqueous solution in the presence of carbonate. However, these complexes only form up to a certain pH. A sudden change of speciation to uranyl hydroxides was detected above pH 10 at low carbonate concentrations (0.5 mM) and above pH 11 at high carbonate concentrations (100 mM).
Batch sorption experiments reveal that this ligand replacement of carbonates by hydroxides goes in hand with an increase in U(VI) retention by Ca-bentonite. The study shows that an almost complete sorption of U(VI) can be obtained in (hyper)alkaline repository environments, even though carbonate is present in substantial amounts.
In order to clarify the mechanisms responsible for the very strong U(VI) retention, uranyl complexes on the bentonite surface were examined directly, using x-ray absorption spectroscopy (ROBL-beamline, ESRF, Grenoble). EXAFS spectra (extended X-ray absorption fine structure) did not show any indication of precipitates, implying that adsorption is the dominant retention process. In all samples with high U(VI) retention, the derived uranium coordination is identical irrespective of the amount of contained carbonate. According to atomic distances and coordination numbers for U-Oeq, U(VI) surface complexes shift from a 5-fold to a 4-fold coordination in the equatorial plane with increasing pH. Attachment might be facilitated by charge balancing cations (i.e. Ca2+) that mediate between the negatively charged clay surface and the anionic aqueous U(VI) hydroxide complexes.
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Poster NFC / 570

Application of the method of electrolytic-plasma treatment for decontamination of removable equipment of nuclear power plants

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Operation of the nuclear power plant is accompanied by the radioactive contamination of internal and external surfaces of equipment. Usually, the main part of surface contamination of the equipment's nuclear power plant is due to activated corrosion products. Decontamination of removable equipment can be done in various ways, depending on its size and design features. Electrochemical decontamination method is used to remove firmly fixed radioactive contamination, when the application of other methods is inefficient or technically difficult. Recently, there is considerable interest in electrolyte-plasma treatment (EPT) 1, as a kind of electrochemical method. Electrolyte-plasma method is an environmentally safe way of high-quality surface treatment of products, because solutions of neutral salts are used as electrolyte. The application of the electrolytic-plasma treatment method for the decontamination of removable structural equipment was not considered in the world practice before.
Objective: To investigate the possibility of using the electrolytic-plasma treatment method for decontamination of the removable equipment of nuclear power plants.
In the study, parts from steel AISI 321 were subjected to electrolytic-plasma treatment. Preparation and radioactive contamination of parts was carried out in accordance with GOST 50773. 6% aqueous solution of ammonium sulfate (grade of p.a.) was used as an electrolyte. The processing time was 4 minutes. The cycle "contamination-decontamination" is carried out 3 times. Measurement of radioactive contamination were performed using γ- and β-analyzer MKS-AT-1315.

From the analysis of the data obtained it can be concluded that the use of EPT is effective for the decontamination of parts and equipment as simple geometric shapes and complex. Decontamination is carried out in a short period of time.

Poster PAR / 770

Measurements of proton-induced production cross sections for Sc radioisotopes from natural Ca

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Keyword –proton induced nuclear reaction, experimental cross sections

The Coordinated Research Project (The International Atomic Energy Agency), entitled "Therapeutic Radiopharmaceuticals Labelled with New Emerging Radionuclides (⁶⁷Cu, ¹⁸⁶Re, ⁴⁷Sc)"is focused on production methods of the ⁶⁷Cu, ¹⁸⁶Re and ⁴⁷Sc radioisotopes as Theranostic Radionuclides for possible radiopharmaceutical development as well as research purposes.
Little information on ^{nat.}Ca(p,x) ^{46, 47, 48}Sc reaction in the energy range below 60 MeV could be found in the literature1. First our results of the yield of ^{46, 47, 48}Sc in reaction ^{nat.}Ca(p,x) were published in the last year2.
For the proton bombardments CaCO₃ targets of natural isotopic compositions and different thickness were prepared. The CaCO₃ targets were made into 12 and 10 mm diameter pellets of different thickness 0.13 - 0.35 g/cm² thickness. The target pellet was placed between Al, Cu or Cu, Ti monitors. These monitors were used to determine a proton beam flux. Several alternating pellets and foils were placed as stacks inside an Al target holder. The stacked targets were bombarded by 60 MeV protons for about 5 hours. The average beam current was 20 nA. This irradiation was performed at the AIC-144 cyclotron at the Institute for Nuclear Physics Polish Academy of Sciences in Cracow. The incident energy for each target pellet was calculated by the proton energy loss in passing through other targets and monitors.
Production cross sections for ^{42, 43}K, ⁴⁷Ca, ^{46, 47, 48}Sc have been measured for the ^{nat.}Ca(p,x) reactions up to 60 MeV. Measurements of the experimental cross section for the reaction ^{nat.}Ca(p,x) are presented as a function of the mean proton energy in the thick target. Cross section reaching a peak of 0.9 mb at around 33 MeV for ⁴⁶Sc and 0.8 mb at around 12 MeV for ⁴⁸Sc. Obtained values of energy for above-mentioned Sc radioisotopes are in good agreement with results reported by Michel et al. 1. Our cross section results are higher by about 20% than cross section data in Michel et al. work 1. However, direct comparison of obtained experimental cross section values is difficult because irradiated Ca targets had different chemical forms. The optimum energy range for the ^{nat.}Ca(p, 2n)⁴⁷Sc reaction is 24 - 17 MeV with the peak at about 20 MeV for CaCO₃ target. The cross section value is about 2 mb.

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Poster NFC / 559

A comparative study of radium and strontium uptake by cementitious materials

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Cementitious materials will be very important structural elements of a deep geological repository, but they can be used also as a part of engineering barriers. A comparative study of radium and strontium uptake by different cementitious materials was therefore performed. Three cementitious materials were studied: a hardened cement paste (HCP CEM II) and two types of concrete (Concrete CEM I and Concrete CEM III, both actually utilized in the storages of institutional radioactive waste in Czech Republic); all crushed and sieved on fraction ≤ 0.4 mm. The cementitious materials were analysed by X-ray diffraction and thermogravimetric analysis and used to the comparative study of equilibrium sorption of the ^{223}Ra (used as an analogue of ^{226}Ra) and Sr with radiotracer ^{85}Sr under various conditions. On same materials, the diffusion experiments with both isotopes were also done. Sorption experiments with ^{223}Ra were carried out in a carrier-free arrangement, concentration of ^{223}Ra was approximately $1\cdot10^{-12}$ mol·L^{-1}. Sorption experiments with Sr were carried out mainly with concentration of Sr $c = 3.5\cdot10^{-4}$ mol·L^{-1} traced with ^{85}Sr, but some of the experiments were also done as carrier free. However, all the cementitious materials used contain Sr, which may affect the experiment. Varying conditions included the temperature, the liquid-to-solid ratio *V/m*, and the composition of the starting solution which was either Portlandite water (saturated $Ca(OH)_2$) or synthetic water CW based on granitic water equilibrated with $Ca(OH)_2$, which is a representative of cement water.

Comparing obtained K_d values corresponding to Sr and Ra uptake, it was determined that Sr reaches distribution coefficients in range of 10 – 150 L·kg^{-1} and Ra in 70 – 900 L·kg^{-1}. The influence of rising temperature (over 50 °C) on the sorption of both elements was observed –rising temperature increases K_d values, especially for lower liquid-to-solid ratio. In the case of Ra, higher K_d when the liquid phase was changed from Portlandite water to CW were reached, but there is no significant influence of the liquid phase change for Sr. Liquid-to-solid ratio also affects differently results of the experiments. In the range of *V/m* from 10 to 600 L·kg^{-1}, K_d values are mostly comparable for Sr, the K_d for Ra are rising in general. The steep increase of K_d for *V/m* = 1000 L·kg^{-1} for both elements was observed, accompanied by unclear influence of rising temperature for this phase ratio too.

Poster NFC / 774

Study of Europium Speciation Using Time-resolved Laser-induced Luminiscence Spectroscopy

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Radioactive wastes, especially high-level waste (HLW) and intermediate-level waste (ILW), generated predominantly in nuclear fuel cycles contain a wide variety of radionuclides including long-term radionuclides and the wastes contain also various chemicals, often complexation agents used for decontamination purposes. Due to the presence of large quantities of long-term radionuclides, HLW and ILW have to be stored in deep geological repositories. Design of such repositories has to ensure long-term isolation of radioactive waste from the biosphere. In order to satisfy this condition, multibarrier approach is applied. However, such an approach is not perfect, the fail is possible and therefore radionuclides might enter environment. The mobility of radionuclides in the environment is affected especially by their speciation. Knowledge of speciation is therefore crucial in terms of deep geological repository design and protection of the environment. Therefore, this work is focused on the study of complexation of europium commonly used as a chemical analogue of trivalent actinides. Disodium salt of ethylenediaminetetraacetic acid (EDTA), glycolic acid, succinic acid and phthalic acid were used as complexation agents and the study was performed by means of time-resolved laser-induced fluorescence spectroscopy method (TRLFS). Europium complexation with complexation agents mentioned above was studied with varying pH and in the case of succinic and phthalic acid also with varying ligand-to-metal ratio.

These experiments served as a basis for europium distribution diagrams acquistion, for determination of luminiscence spectra as a function of pH and ligand-to-metal ratio and the mean lifetime measurements were used for determination of number of water molecules surrounding europium, the mean lifetimes of resulting complexes and indentification of europium complexes. Finally, time-resolved luminiscence spectral characteristics of indentified complexes were determined and deconvolution of luminiscence spectra of the identified complexes was performed.

Poster NFC / 516

The assessment of the composition of the gas phase in the processes of stabilizing plutonim valent states during hydrometallic processing of spent nuclear fuels

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In the technological system of high-burnt irradiated nuclear fuel reprocessing that is being developed, before entering the affination phase, all the reductant from the previous phase of reducing reextraction must be deconstructed and the Pu(III) must be oxidized to Pu(IV). For this process a reagentless oxidation method using an activated carbon catalyst (VNIITU-1) was proposed. As a result of this catalytic oxidation, gas evolution occurs on a large scale, which in turn points to an organic reductant oxidation.

The tests to determine the composition of the gas phase were conducted in 2 to 4 mol\L [HNO3] at 40 ^{0}C. The plutonium reductants used were carbohydrazide (0.05-1 mol\L) and diformylhydrazine (0.1-1 mol\L). The examination of the composition of the solution, flowing out of the catalytic column, in search of non-decomposable catalytic discharge was conducted. Data regarding the quantity and composition of the gas phase in a measure of reductant was obtained. The specters of this gas phase were recorded and analyzed by use of a Fourier-transform spectrometer.

Poster NFC / 668

Development of tools for studying radionuclide transport in fractured rock environment

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The main goal of the project Development of tools for Studying Contaminant Transport in fractured rock environment (TH02030543; 2lsConTra), funded by Czech Technology agency, is to develop tools for evaluation of migration and retention of potential contaminants in the rock environment as the key input values the safety assessment of anthropogenic activities. Those may release contaminants (radionuclides, heavy metals, nanoparticles) into the environment, hence groundwater. It is necessary to describe the processes which undergo in different environments on the path from the contaminant source to the environment, concerning that the main transport medium is groundwater and that one of the main routes of man exposure is drinking water.

The project aims is to build a laboratory physical models that include both artificial and natural fracture, to instrument them and to perform migration experiments that would study migration processes of the species in the rock environment, including dispersion, diffusion and sorption. The experiments will be modelled using both conventional software (MODFLOW) and in-house code (FLOW123).

As the fate of radionuclides in the environment are nowadays of high importance, those physical models will be presented as a potential tool for identification and description of processes that could influence their retention on the way toward the biosphere.

Poster NFC / 530

Investigation on the sorption behavior of Se(IV) and Se(VI) onto modification bentonite

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Selenium-79 (79Se) is one of the important long-lived fission products exists in radioactive waste and has toxic anionic species in an aqueous environment. Consequently, 79Se is considered as a key radionuclide that should be taken into consideration in safety assessments in radioactive waste repository. Commenly, the usage of bentonite as a buffer material is suggested in radioactive waste repositories. Although bentonite expresses high sorption abilities to cations, the situation seems different to anions due to the negative surface charge of bentonite. Recently, the preparation of modified bentonite was studied to investigate their sorption properties in aqueous.

The present work focuses on the sorption behavior of Se(IV) and Se(VI) on inorganic and organic modification bentonites, which were produced by modification of sodium bentonite with iron hydroxy oxide (Fe-OOH) and hexadecyl trimethyl ammonium bromide (HDTMA). Batch techniques were used to investigate the sorption ability of modification bentonites to Se(IV) and Se(VI) under different environmental conditions. The results show that the bentonite sample was successfully modified and the host structure is significant changed, leading to obvious increase Se(IV) and Se(VI) sorption. Compared to organic modification, the inorganic modification with iron hydroxy oxide shows more powerful sorption abilities towards Se(IV) and Se(VI).

Poster NFC / 672

Decontamination of anthropogenic polluted soils from Ra-226 decay family radionuclides and mercury using hydroclassification technique

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Vast territories with soils contaminated by radionuclides (RN) and/or mercury have appeared as a result of anthropogenic accidents, long-term activity of mining and processing plants, heat power plants, nuclear fuel cycle industry, as well as a consequence of decommissioning of such objects. These contaminants easily get involved to migration processes in the environment and frequently cause sustainable negative effects on ecosystems.

Modern technologies of decontamination of soils polluted by RN and/or heavy metals are based on the techniques, which either require chemicals in the processing or involve merely physical steps, as well as their combination. But the problem is that the high effective and relative cheap purifying technology has not created yet.

In this report the results of systematic researches devoted to decontamination of soils polluted by RN and/or mercury using hydroclassification technique involving pulse column equipment are discussed. This technique provides fractionating of contaminated soils and concentrating of RN in a fine fraction. As a rule, the resulting mass content of this fraction does not exceed 20% from initial mass.

The objects of the study were soils contaminated by radionuclides (RNS) or mercury (MCS). The soils have been sampled in one of the industrial plants in Russia. The samples under the study contain a significant part of construction waste along with natural soil components.

Specific activity (Q) of the initial RNS due to Ra-226 approached to the value of 12 Bq/g. Mass content of the fraction -0.01 mm ~35%. It has been determined that Ra-226 generally occurred in a form firmly coupled with organic matter and clay (~90% of total Ra-226). Mercury content in the initial MCS was extremely high and approached to the value of 350 mg/kg. Mass content of the fraction -0.01 mm ~30%. In the initial MCS, besides mercury metal, the other chemical forms like organic and inorganic mercury compounds have been detected.

Decontamination of the soils has been carried out using enlarged laboratory setup. The main apparatus of the setup was pulse hydroclassification column equipped with KRIMZ filling that ensured productivity of up to 500 kg/h. The column has been already successfully tested on soils contaminated by Cs-137. As a result of the hydroclassification of soils, up to 90% of total Cs-137 was concentrated into clay (≤15% of total mass of soils).

Hydroclassification of RNS in the optimized operation conditions provides concentrating of up to 66% of the total Ra-226 in clay fraction and returning of up to 85% mass of the initial soils for recovery $Q \leq 4$ Bq/g.

As a result of hydroclassification of MCS ~63% of total mass content of the metal was concentrated in clay fraction (28% mass). Considering that mercury content of the most part of decontaminated soils had not exceeded 15 mg/kg, such soils can be used for filling pits with overlapping of a layer of clean soil with a height of at least 0.5 m.

The results obtained allowed a development of the flow sheet and the process flow diagram of stationary and mobile setups including pulse column apparatus for decontamination of soils from Ra-226 decay family RN and/or mercury.

Poster NFC / 654

Degradation of diluent in TODGA –F-3 extraction system under α-, β-, γ- irradiation

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Future nuclear fuel cycles based on fast reactors are being developed in many countries. New nuclear fuel cycles include the reduction of long term radiotoxicity of nuclear waste by partitioning and transmutation of long lived transplutonium elements. So the selective recovery of transuranium elements, especially actinides (III) –americium and curium, from the high lever radioactive waste (HLW) created by the reprocessing of spent nuclear fuel is rather actual problem.

Liquid-liquid extraction is one of the known and proven methods of separation of the actinides (III) from PUREX rafinates. Some isotopes of americium and curium are strong α-emitters and the solvents, used for the partitioning of HLW are exposed to α-particles emitted by the isotopes along with β- and γ-radiation emitted by fission products.

We have developed a process, using a solvent based on well-known ligand N,N,N',N'-tetraoctyldiglycolamide (TODGA) 1 in polar diluent meta-nitrobenzotriflourite (F-3) 2. Such diluent significantly increases the capacity of solvent in comparison with traditional hydrocarbons and prevents the third phase formation 3.

But this diluent contains flourine atoms so the fluoride ions can be produced as a result of degradation of the F-3 under irradiation. The fluoride ions can significantly increase the corrosion rate of steel. In case of evaporation of the solutions created by reprocessing of HLW, the high concentrations of fluoride ions can lead to very serious problems.

In this work the effect of α-, β-, γ- irradiation of the solvent on yield of fluoride ions was studied. The yield of fluoride ions was not very significant. G-value for yield of fluoride ions –about 2 ions / 100 eV under α- irradiation, about 0,7 ions / 100 eV under γ- irradiation and about 0,67 ions / 100 eV under β- irradiation.

The system shown good resistant extractions properties and slight G-value of fluoride ions.

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Poster NFC / 674

The use of a cement compound for treatment radioactive waste from the reprocessing of SNF fast reactors

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Cementing methods is mainly used for low and intermediate level radioactive waste. Currently, cement stone is almost never considered as a matrix for immobilizing high level waste due to probable low durability of as well as to radiolysis cement hydrates and pore-water under high radiation dose. The objective of this report is the assessment of cement compounds affected by high radiation dose typical of high level activity waste. Cement compound samples of different water-cement ratio were affected by up to 108 Gy radiation dose during the research.

Irradiated cement samples were observed to examine changes in regulated properties. Gas emission from cement samples and crystal phase structural changes were also assessed during the research.

The research of structural changes of cement stone due to ionized radiation were done by the following methods: X-ray phase, Petrographic, Electron micrograph, Electron- microscopic analyses.

Comparative assessment of the samples before and after irradiation shows that cement compounds meet regulated requirements of final radioactive product.

Compressive strength was considered as a criterion. The samples subjected to hardening for 28 days have from 4 to 8 times more compressive strength than the required 5 MPa. The research proves that irradiated compounds have the same durability after frost resistance and emersion tests.

As a result of the experiment, the concentration of hydrogen emitted from cement compounds was also measured. The quantity of emitted hydrogen from higher water-cement ratio is a bit higher due to higher water level. The research verifies that the highest level of hydrogen is not higher than 10-3 mol/ (per sample).

The studies of structural and morphological characteristics of cement samples by the above mentioned methods show that structural and phase changes in the samples are not revealed.

The results of the research show that examined cement compounds withstand radiation dose up to 108 Gy.

Poster NFC / 740

Stability of chain and cyclic monoamide resins for selective separation of U(VI) against γ-ray irradiation in HNO3

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In order to develop resins with selectivity to U(VI) in HNO3 media, we have synthesized several polymer beads with the structure of a monoamide as the functional group. For wider applications of these resins including treatment of highly-radioactive solutions, it is necessary to investigate their stability under irradiation conditions. In this study, resins with a chain-type monoamide and a cyclic-type one as the functional group were irradiated by γ-ray in HNO3 and adsorptivities to FP ions were examined. The structure change in the resins by irradiation was also studied.

N-methyl-N-vinylacetamide (VMAA) 1 was used the chain-type monoamide resin. For the cyclic type resin, that with a 6-membered ring of 1-(4-vinylbenzyl)piperidin-2-one (VBPP) was newly synthesized. The resins and HNO3 of up to 6 M were mixed in a Pyrex sample tube at a ratio of 5 cm3/g, respectively, and used as the samples for γ-ray irradiation. Irradiation by the Co-60 source was performed up to 0.95 MGy at room temperature under ambient atmosphere. The irradiated resins were separated from the supernatant and washed using distilled water to avoid further degradation of the resins by HNO3. Adsorptivities of the irradiated resins to FP ions were examined in HNO3 by the batch method similarly to the previous studies[1-3]. The resulting resins and the supernatants were also analyzed by IR and NMR, respectively.

For irradiated VMAA, the Kd values for Pd(II) and Re(VII) were found to become lower than those of the neat ones. This indicate that the degradation properties are similar to those of chain-type monoamide extractants4. On the other hand, the Kd values of the irradiated VBPP for Pd(II) and Zr(IV) were found to be increased at lower concentration of HNO3 with increasing dose. Polyvinylpolypyrrolidone (PVPP), which is a cyclic monoamide resin and has a 5-membered pyrrolidone ring, changes the chemical structure by γ-ray irradiation in HNO3, where the degradation starts with the ring opening and the final products have been found to be a primary-amine-type anion exchange resin and oxalic acid[5]. This study suggests that the major route of change in the chemical structure of VBPP is basically identical to that of PVPP and that the resulting resin structures differ according to the number of carbon atoms in the cyclic monoamides.

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Poster RPH / 824

Conjugation, radiolabelling and in vitro studies of VAP-P1, a vascular adhesion protein-1 targeting peptide

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Background and Aims:
Vascular adhesion protein-1 (VAP-1) is an endothelial glycoprotein present on the endothelial surface of tissues affected by infection/inflammation that recruit lymphocytes from blood. VAP-P1 is a VAP-1 selective peptide (sequence GGGGKGGGG) which, according to previous studies, conjugated with 1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid (DOTA) and labelled with Ga-68, could be used as imaging agent capable to differentiate between normal bone healing and bone infection.

The aim of this paper is to study the potential of 1,4,7-triazacyclononane-triacetic acid (NOTA)-conjugated and Cu-64 labelled VAP-P1 peptide for possible clinical applications.

Methods:
Synthesized peptide VAP-P1 was conjugated with NOTA-SCN, purified using semi-prep HPLC and lyophilized. The conjugate was then labelled with Cu-64. In vitro plasma protein binding and stability in human serum in time were determined using transfected Chinese Hamster Ovary cells (CHO) that express human VAP-1 and HT-29 (negative control). Each experiment was performed in octaplicate, and the data were reported as percentage of total added activity.

Results:
The peptide was conjugated with NOTA chelator and labelled with 68Ga and 64Cu with radiochemical purity ≥ 95 %. Radiolabelled analogue [Cu-64]NOTA-VAP-P1 presented here was rapidly internalized into CHO cells versus negative control HT-29. The internalization of the surface bound peptide was time-dependent. It sharply increased during the first 30 min reaching internalization levels of 80–90 %.

Conclusions:
In vitro studies showed that NOTA-VAP-P1 labelled with Cu-64 may provide a faster and a simpler method of diagnostics of inflammation/infection compared to the method of labelled leucocytes. The concept deserves to be tested in vivo.

Key words: Cu-64, NOTA-VAP-P1, VAP-P1, VAP-1, labelling, conjugation, PET, CHO cells, plasma protein binding, human serum stability

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Poster NFC / 828

Performance of Decontamination Foams Containing Various Chemical Components

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Foam decontamination process has a potentially wide application in the removal of contaminants from large components with complex shapes or large area or large volumes. This process can be applied to any direction regardless of walls, floors and ceilings, and basically it has the advantage of generating less secondary wastes. However, there is a disadvantage that the decontamination factor (DF) is relatively low only by one batch application. In order to improve the DF of the foam decontamination process, many attempts have been made to introduce various chemical decontamination agents into the foam and enhance the contact time of the foam between the chemical decontamination agent and the contaminated surface by introducing a viscosifier to improve the stability of the foam. In this study, we investigated the effects of 'push-blowing' cyclic foam filling process in which newly generated foam is periodically brought into contact with the surface of the decontamination object, rather than the 'one batch application' process which applies the decontamination foam only once. In addition, the decontamination performance of the foams containing various chemical components such as chemical reagents and silica nanoparticles was evaluated according to the characteristics of surface contamination of the decontamination object. Decontamination performance tests were carried out by applying various decontamination foams to the simulated specimens contaminated with non-fixed particles and oil, and the specimens with fixed corrosion oxide films. The decontamination performance was improved by keeping the physicochemical properties of the decontamination foam substantially constant by applying 'push-blowing' cyclic foam filling process. In the removal of contaminants from the simulated specimens by the decontamination foam, non-fixed contamination consisting of a mixture of Eu2O3 particles, TBP and fluorescent material was completely removed within 4 hours by neutral decontamination foam containing 1 wt% of M-5 silica nanoparticles and 1 wt% of EM100 nonionic surfactant. For the removal of the fixed contaminants, the NiFe2O4 film coated on the stainless steel specimen having a thickness of 500 nm was completely removed within 2 hours by decontamination foam composed of 0.5M HF, 0.5M HNO3 and 1 wt% EM100. On the other hand, only about 3% of the NiFe2O4 was removed during the same time period by the decontamination foam consisting of 2 M HNO3, 1 wt% EM100 and 1 wt% M-5. FeCr2O4 film with a thickness of 10 ㎛ produced in the autoclave was completely removed within 2 hours by the oxidizing decontamination foam containing 0.1 to 0.5 M Ce(IV), 2 M HNO3, 1 wt% TBS as an anionic surfactant and 1 wt% M-5 nanoparticles, but almost not by the decontamination foam consisting of 0.5 M HF, 0.5 M HNO3 and 1 wt% EM100. It was found that the corrosion oxide film containing chromium can be removed by the oxidizing decontamination foam while it was difficult to remove by the reducing decontamination foam.

Poster RPH / 838

Pharmaceutical development of the theurapetic radiopharmaceutical based on β-emitting samarium-153 in heat-sensitive carrier for brachytherapy of tumors of various location

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The present development is aimed at creation of thermo-collapsing radiopharmaceuticals for brachytherapy based on thermo-sensitive copolymers with chelated beta-emitting radionuclide 153Sm. The preparation is a polymer solution and is made of two components, each loads individual function.
The task set in this development is solved with component-wise division of the radiopharmaceutical's key functions. Primarily, the first functional subsystem is prepared: this is a thermo-sensitive copolymer of n-isopropylacrylamide and allylamine which is exposed to etherification on aminogen groups by the chelating agent of diethylenetriaminepentaacetic acid dianhydride, and to the following labeling of 153Sm3+ due to formation of chelate complexes. Separation of the labeled copolymer

component from low-molecular active and inactive compounds in the reaction mixture is performed by elution with acetate buffer on a chromatographic column filled with swollen sephadex. Column effluent containing active copolymer fractions actually is the first functional system which serves as a hybrid solvent to prepare a semidelute solution of the polymer –gelifier. Gelifier's role is to build a fluctuation quasi network which is formed by contacts of links from different chains.
Auxiliary materials such as copolymer-carrier and polymer-gelifier are responsible for formation of the required conditions in the preparation to hold the active substance tightly by creation a firm compact gel, while acetate buffer and salt component provide the required pH and isotonicity of the solution.
Action mechanism is the following –radiopharmaceutical solution after intratumoral injection at the injection site quickly achieves the body temperature and due to self-collapsing of the heat-sensitive polymer-gelifier turns into a firm polymeric clot. This phenomenon provides fast loss of translational mobility of immobilized radionuclides chelated with linear chains of the copolymer-carrier. As a result, there are formed sources of local therapeutic irradiation which affect cancer cells and destroy them almost without any impact of healthy tissues. When therapeutic action is completed the polymer coils are naturally removed from the body.
There were performed the pre-clinical trials of the radiopharmaceutical for brachytherapy of a number of prostate malignancies and other solid tumors. According to the pre-clinical trials results the preparation is recommended for clinical trials for local therapy (brachytherapy) of tumors of various location. First phase of clinical trials is reasonable to be performed with patients of the one clinical entity of the disease –prostate cancer.
The preparation injection should be performed intratumorally and to the several tumor points (at the rate of 1 injection / 1 cm3 of tumor tissue), 0.1 ml in volume. Recommended dosage of the injection for patients with average body weight (70 kg) is 1.6 GBq.

Poster RPH / 836

Synthesis and structure of adeninium pertechnetate.

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Pertechnetate (TcO4-–) is among the anions that are considered the most environmentally mobile. This is due to its relatively large size and low negative charge density distributed at the four oxygen atoms [1-4]. The negatively charged anions repel from the negatively charged surface of most minerals. Conversely, they are attracted by positively charged residues of nitrogenous bases like adeninium in acidic environments. The model characteristics for this behavior might be understood by analyses of the corresponding structures in the adeninium compounds. We first synthesized the compond of adeninium pertechnetate (C4H6N5)TcO4 (I) by interaction of 0.1 M aqueous solutions of (C4H6N5)Cl with 0.1 M NaTcO4 followed by slow evaporation of the resulting solution till the single crystals started being formed. Compound (I) is described by the formula (C4H6N5)TcO4, it contains in its composition doubly charged adenine cations, two perrhenate anions and crystallizes as colorless crystals in the monoclinic syngony, Z = 8, pr gr. _____. The parameters of the unit cell are: a = 9.2585(3), b = 9.3131(3), c = 20.7305(5) Å, The system of hydrogen bonds in (I) is represented by classical N-H ⋯ O bonds.
The structure contains 4 crystallographically independent formula units. There is pseudo-translation b/2. The anions associated with pseudotranslation have different orientation, the orientation of the cations is preserved.
The X-ray diffraction experiment was performed at the Center for Collective Use of Physical Methods of Investigation the IPCE RAS on an automatic four-circle diffractometer with a two-dimensional Bruker KAPPA APEX II detector (MoKα radiation, λ = 0.71073 Å).

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Poster RPH / 846

LABELLED SUPERPARAMAGNETIC IRON OXIDE NANOPARTICLES

Authors: Zuzana Sobkuliaková[1]; Veronika Valová[1]; Ekaterina Kukleva[1]; Michal Sakmár[1]; Olga Mokhodoeva[2]; Martin Vlk[1]; Ján Kozempel[1]

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Superparamagnetic iron oxide nanoparticles (SPIONs) are being studied as contrast agents and drug delivery vehicles in medicine, mainly for easy preparation, low toxicity, biocompatibility and their magnetic properties. SPIONs have become a promising tool in the magnetic resonance imaging, magnetic drug targeting, hyperthermia anti-cancer strategy, and enzyme immobilization. The scope of our work was the synthesis and characterization of SPIONs and their labelling with Tc-99m useful for SPECT diagnostics and also with Ra-223 for targeted alpha particle therapy [1, 2].

SPIONs were prepared by co-precipitation method and then stabilised with 0.1 M sodium citrate, the pH range during the synthesis of iron oxide SPIONs was between 8-14 with maintaining molar ratio of Fe(3+)/Fe(2+) 2:1 under inert condition. The structure and composition of the synthetised nanoparticles were confirmed by FTIR and XRPD. The size of the nanoparticles was determined by dynamic light scattering (DLS). The stability of the nanoparticle dispersion was determined by the measurement of Z-potential.

Labelled SPIONs were synthetised contacting suspension of 1-5 mg SPIONs with commercial generator eluate containing Na^{99m}TcO₄ (200-250 MBq) in physiological saline, respectively ²³³Ra(NO₃)₂ 50-100 kBq in PBS buffer. Stock Ra-223 solution was eluted from generator 1. Commercial MDP and HDP kits were labelled with 200 –250 MBq Na^{99m}TcO₄and were subsequently contacted with SPIONs in 30 min. Labelled Tc(99m)-phosphonates coordinate on SPIONs surface and their stability was studied. SPIONs labelled with Tc-99m (T1/2 = 6 h) and Ra-223 (T1/2 = 11,43 d) both with excellent yields (> 90%) were also studied for a stability and period of five half-lives in physiological solution, plasma, serum and albumin.

The multimodality of SPIONs has led the biomedicine research in a theranostic research, which is the integration of diagnostic imaging and therapeutic function into a single platform, and has potential for in vivo experiments on physiological mice model.

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Poster NFC / 851

Evolution of mineralogy and radionuclide diffusion on Portland-type -cementitious materials due geochemical and thermal effects

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Experimental work within the CEBAMA project focuses on alteration and interaction studies on cementitious materials (based on Portland type CEM II, w:c = 0.45, OPC) in contact with groundwater (GW) from Underground Research Facility Josef and with bentonite suspension (Czech B75 bentonite) in pressure vessels during three sampling campaigns (9/18/27 months) under in-situ (10℃) and high temperature (95℃) load. The first two sampling campaigns after 9 and 18 months were performed, and evolved solutions and solid samples were analysed. The main goal was to understand how the interaction processes affect the transport properties of cementitious materials. The HTO through-diffusion experiments on these evolved cylindrical samples (thickness of 8 mm, diameter of 50 mm) provided three types of datasets: depletion and breakthrough curves obtained during the experiments and concentration profiles in samples, which were obtained by a newly developed abrasive technique after the experiment´s termination.

A precipitate of carbonates, mainly calcite, was found on the surface of some samples, that was the most significant on samples after interaction of samples in bentonite suspension under 95℃ load; the oversaturation of calcite can be linked to large carbonates pool in bentonite and to calcium originating from the cement paste. Based on chemical analyzes of evolved solution of groundwater in vessels and XRD after the interaction, the high temperature speeded up the dissolution / transformation of ettringite (C₄AF), which resulted in increase of sulfate concentration. Further analyzes of evolved solution revealed an increase of potassium and calcium. The heated samples contained also hydrothermal mineral katoite (C₃ASH₄) and miss the phases with stability under lower temperatures like CO₂/Cl-hydrocalumites.

The first results of HTO through-diffusion experiments on samples from the first sampling campaign showed that the high temperature raised values of effective diffusion coefficients (*D*_e), approx. in order of magnitude, which could be related to dissolution of material under higher temperature but also to the decrease of elasticity (microcracks). The mean values of HTO effective diffusion coefficients (for evaluation using GoldSim software) from three parallel experiments are the following:

- *D*_e = 2.8•10⁻¹¹ m²•s⁻¹ in the series: OPC + B75; 95℃; 9 months;
- *D*_e = 0.3•10⁻¹¹ m²•s⁻¹ in the series: OPC + B75; 10℃; 9 months;
- *D*_e = 2.0•10⁻¹¹ m²•s⁻¹ in the series: OPC + GW; 95℃; 9 months;
- *D*_e = 0.4•10⁻¹¹ m²•s⁻¹ in the series: OPC + GW; 10℃; 9 months.

The preliminary results (the concentration profile dataset were not considered) from through-diffusion experiments on samples from the second sampling campaign which are terminated between March and April 2018 showed no significant changes between two sampling periods.

Acknowledgement

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Poster PAR / 731

Metal-Organic Frameworks as Scaffolds for Isotope Separation

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Metal Organic frameworks (MOFs) are self-assemblies of metal ions and organic ligands, known for their high crystallinity and porosity. The wide range of high and accessible porosity allows them to be explored as alternatives for traditional porous materials and for many new applications. Some of them exhibit an extraordinarily huge surface area together with a high chemical stability.
One of the most studied and applied MOFs, MIL-101, was selected as the starting material for the production of chromium-51 and its enrichment based on the Szilard-Chalmers effect. After finding the most optimal synthesis conditions, several samples of MIL-101 were irradiated in the HOR reactor at Delft under different neutron fluxes and irradiation times. Upon neutron capture, the formed ⁵¹Cr atoms will get into a short-lived excited state that relaxes through the emission of prompt gamma photons. The recoil imparted on the emission of these photons is energetic enough to break chemical bonds and allow for the separation of the atom involved. Once that the different samples were irradiated the optimal conditions for the collection of the freed ⁵¹Cr were investigated. The key point for getting the best results resulted in avoiding the dissolution of chromium from the framework by selecting the right pH and extracting agents for the elution.
The use of new materials, like MOFs, with high stability and surface area may open new possibilities for isotope separation based on Szilard-Chalmers effect thanks to their widely accessible surface area, their high metal content and their high chemical (and radiation) stability.

Poster PAR / 741

Assessment of Staff Radiogenic Risk from radionuclide exposure at Nuclear Medicine Department

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Staff and patient are exposed to un avoidable radiation exposure to a wide range of radionuclides, as 99mTc, 67 Ga and 131I during nuclear medicine procedures. Although the frequency of the nuclear medicine is increasing, few data are available regarding ambient and patient exposure worldwide. The The International Commission on Radiological Protection (ICRP) has lowered the annual dose limit for the eye lens from 150 mSv to 20 mSv (i.e., by a factor of 7.5) for occupational exposures, therefore it is crucial to evaluate its impact in the existing programs on radiation protection and safety. Therefore, measurement of staff doses and ambient doses are important.
The objectives of this study are to measure the ambient radiation and patient dose during whole body bone scintigraphy, thyroid and renal scan procedures. The study was carried out at Alnilain Daignostic Center, Khartoum, Sudan. Staff radiation exposure personnel were calculated as a function of administered dose distance from the patient and at different times after the administration and workload. A calibrated survey meter and thermoluminescent dosimeters (TLDs- GR200A) were used to measure the ambient dose and staff dose, respectively. Prior to measurements, all TLD were calibrated in terms of air kerma free-in-air under reproducible reference condition using 99m Tc

with activity 10 mCi (370 MBq). Quality control performed before administration of the radiopharmaceutical and doses are carefully calculated. All scan procedures were performed using MiE single head gamma camera (Orbiter 37 Gamma camera) after administration of 20 mCi, 4 mCi and 5 mCi of 99mTc.
The average ambient dose equivalent rate equal to about 12, 25 and 10 µSv/h was obtained at distance of 1 m, at 1.3 m from patient during bone, renal and thyroid scan respectively. Injection room and hot lab has ambient dose equivalent rates of 1.0 and 30 µSv/h at the same order. The maximum dose were recorded at the reception area equal to 180 µSv/h. Staff may exposed to a dose range from 8.0 to 12.5 mSv annually. Knowledge of ambient dose values is crucial in order to determine exposure personnel who may limit the time spent at high dose areas. The dose values are within the safety limit in the light of the current practice. Although, the ambient dose is high compared to previous studies, the staff exposure was below the annual dose limits in the light of the current workload. Appropriate isolation of the patients, training of staff and a strict compliance with the established radiation safety standards are crucial in order to avoid unnecessary radiation exposure.

Poster NFC / 549

The rate and mechanism of uranium and lanthanum leaching from magnesium-potassium-phosphate compound

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Long-term controlled storage or disposal is one of the major stages of liquid radioactive waste (LRW) management. The preparation of LRW for this stage includes the conversion of waste into a stable solidified form. In recent years, researches devoted to the development of new methods of LRW solidification are carried out using a magnesium-potassium-phosphate (MPP) compound based on the MgKPO4·6H2O matrix. The matrix is formed in an aqueous solution at room temperature as a result of the acid-base reaction between magnesium oxide and potassium dihydrogen phosphate and it is an analog of the natural mineral K-struvite. It was shown that the LRW immobilization method in the MPP compound combines versatility, simplicity of implementation and relative cheapness similar to cementing, as well as high physicochemical stability of the compound, which is not inferior to glass.
The samples of the MPP compound were synthesized during the immobilization of nitric acid solutions of uranium and lanthanum in this research work. It has been established that uranium in the MPP compound is in the form of potassium uranyl orthophosphate K(UO2)PO4·3H2O (meta-ankoleite), and lanthanum - in the form of phosphates LaPO4·0.5H2O (rhabdophane-La) and Mg0.6K0.68La0.36PO4·6H2O. The differential rate of leaching of uranium and lanthanum at the 28th day of contact of the compound with water under 23 ℃ is, g / (cm2·day): 1.7·10-6 and 6.4·10-6, respectively. The degree of leaching of uranium and lanthanum is 0.02 and 0.05%, respectively.
The mechanism of uranium and lanthanum leaching of MPP compound was studied according to the de Groot and van der Sloot model. It was shown that the mechanism of uranium and lanthanum leaching varies during the contact of the compound with water. During the first 7 days uranium leaching occurs due to dissolution of the surface layer of the compound, where single particles of hydrated uranyl nitrate were probably localized, however then the uranium leaching is uniquely determined by the diffusion mechanism from the inner layers of the compound. The behavior of lanthanum in leaching was almost the same. During the first 14 days leaching of lanthanum occurs due to dissolution of the surface layer of the compound, then during the subsequent 14 days - due to its depletion, however then leaching of lanthanum similar to leaching of uranium is uniquely determined by the diffusion mechanism from the inner layers of the compound.

Poster NFC / 579

TPE and RE extraction during long-lived radionucide partitioning in combination with PUREX-process using single TBP-bazed

solvent

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One of the ways for diminishing risks during storage and burial of Purex-process HLW, which are arising from high heat evolution by -emitting 90Sr & 137Cs, is recovery of long-lived TPE radionuclides, especially 241+243Am, which could be recycled in NFC by inclusion in the fuel for fast reac-tors. Commercial TPE partitioning in Russia has been carried out and developed using fluorinated diluents, such as metanitrobenzotrifluoride (F-3) 1. They are recognized now as corrosion activa-tors at HLW evaporation due to their destruction products generated by radiolysis and hydrolysis. So, the progress in TPE partitioning is connected with the use of non-halogenated diluents.

The goal of this work was TPE and RE partitioning by solvent extraction with dropping of Cs & Sr to raffinate in the frames of the joint extraction cycle of Modified Purex-process using the sin-gle TBP solution in paraffine diluent as a solvent. The influence of component concentration on the effectiveness of 241Am & 154Eu extraction by 30% and by 50% TBP in hydrocarbon fraction C13 from nitric acid solutions in the presence of salting-out reagents (ammonium, aluminum and iron ni-trates with nitrate-ion concentration up to 6 mol/L) has been investigated in the course of laboratory scale experiments.

The parameters of the above combined process have been optimized computer simulation us-ing the model developed in KRI. It has been shown that 50% TBP in hydrocarbon diluent is the limiting composition of the single solvent for the Modified Purex-process. It is the maximum TBP concentration in the 1st extraction cycle which does not lead to the phase inversion and to increase of U backwash specific volume. At the same time, it is the minimum one for efficient TPE recovery from evaporated HLW raffinate of the 1st cycle in the presence of salting-out reagent, which could further form a matrix for HLW immobilization.

Rig trials on TPE and RE recovery from real bottoms solution after evaporation of HL raffi-nate generated in WWER-1000 fuel reprocessing have been carried out in the hot cells using mixer-settler units. The flowsheet has included TPE&RE extraction, solvent product scrubbing, backwash-ing and solvent regeneration. The HLW feed has been adjusted by metallic iron dissolving in the feed solution up to salt concentration 1 mol/L for both decrease of acidity and forming of the salt-ing-out agent. TPE and RE were recovered by 99.98% and further backwashed by weak nitric acid solution with decontamination factor from 137Cs up to 7500. The patent application has been filed.

The further investigations would be devoted to TPE and RE partitioning, their concentration during backwashing and to testing of the single-solvent hybrid extraction cycle.

Poster NFC / 721

The effect of preliminary irradiation on the dynamics of gas evolution under thermal oxidation of the diamides based extractant in the diluent F-3

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Investigation of causes of the accidents, leading to explosions and fires at nuclear facilities, is an important component in ensuring nuclear and radiation safety because of the significant impact of such accidents on the personnel, population and environment. Among a large num-ber of nuclear facilities, the most dangerous are SNF reprocessing plants, which are currently using processes such as PUREX, UNEX, which consist in the extraction/back-extraction of target components by organic solutions from nitric acid media. This combination of organic reductant and nitric oxidant poses a potential danger of uncontrolled exothermic chemical re-actions, that more than once led to explosions at the radiochemical enterprises both in Russia and abroad.

The aim of this work was to study the explosiveness and radiation stability of extraction mix-tures: di(N-ethyl-4-fluoroanilide)-2,6-pyridinedicarboxylic acid (Et(pFPh)DPA), di(N-ethyl-4-ethylanilide)-2,2'-bipyridine-6,6'-dicarboxylic acid (DYP-9) and di(N-ethyl-4-hexylanilide)-2,2'-bipyridine-6,6'-dicarboxylic acid (DYP-7) in the diluent F3. Ionizing radiation of radio-nuclides was simulated by irradiating samples on a linear electron accelerator UELV-10-10 S70 with a vertical scanning electron beam with an energy of 8 MeV. The average dose of 4.68 kGy was absorbed by the sample, passing through an electron beam with a fixed velocity of 1.65 cm/s at a current of 500 µA. The number of passes provided the required total ab-sorbed dose: 0.1, 0.5 and 1 MGy. The thermolysis of the irradiated samples in contact with aqueous nitric acid solutions was carried out in an apparatus for studying the parameters of a thermal explosion at an elevated pressure. This apparatus contains a thermostat where auto-clave with volume of 300 cm3 was placed with a sample. The volume of the samples was 30 ml. The error in the pressure measurements was less than 0.5%. The thermolysis duration in all the experiments was 5 hours. The temperature of the autoclave during these experiments was 170 and 200 ℃.

Following results have been obtained when the irradiated samples were heated in the closed autoclave containing extraction mixtures irradiated to doses from 0 to 1 MGy and 14 mol/L nitric acid solution:

- pressure values were from 18 to 23 atm.;
- the highest pressure values were close for all the studied extraction systems with the diluent F-3;
- the rate of pressure increase, as in the case of the diluent FS-13, varied almost in the same interval: from 0.014 to 0.019 atm./s;
- the rate of pressure increase was almost independent on the absorbed dose;
- insignificant exothermic effects were detected;
- the maximum increase in temperature (self-heating) of the samples as a result of exothermic processes was from 4 to 10 ℃.

Poster NFC / 722

Salts of organic bases application for in-cycle regeneration of the turnaround extractant

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The extraction system that supposed to be used for extraction of radionuclides must have suitable extraction and hydrodynamic characteristics, as well as to be explosive- and fire-safe in operation. The main reason of the worsening the circulating extractant quality is the formation and accumulation of various organic compounds under irradiation. The main products of radiolytic degradation in the extraction system based on tributyl phosphate in a hydrocarbon diluent are: monobutylphosphoric, dibutylphosphoric and phosphoric acids, butanol, carbonyl compounds, nitrocompounds and nitrates, carboxylic acids and esters, various saturated and unsaturated hydrocarbons. The in-cycle regeneration stage is used to remove impurities from the system. At this stage the extractant is washed with solutions of basics or alkaline metal carbonates. However, if a sufficient amount of higher carboxylic acids is accumulated in the system, then extractant washing leads to the formation of their salts, which are strong emulsifiers.

The objective of this study is to evaluate the possibility of application of salts of organic bases (guanidin carbonate and methylamine carbonate) and sodium bicarbonate at the stage of in-cycle regeneration of the degraded extractant. The 30% TBP solutions in Isopar-M isoparaffin diluent (fraction of branched hydrocarbons up to 14 carbon atoms) were used as extraction systems. Irradiation of the organic solutions preliminarily saturated with 3 mol/L nitric acid was performed up to doses of 250, 500 and 750 kGy using a linear accelerator UELV-10-10-S70 with 8 MeV energy. The regeneration step was carried out in a separatory funnel with a glass stirrer by two contacts (each step for 10 minutes) at equal volumes of the organic and aqueous phases. Aqueous solutions of 0.5 mol/L NaHCO3, 1 mol/L guanidine carbonate or 1 mol/L methylamine carbonate were used as regenerating solutions. The radiolysis products were analyzed by Fourier transform IR spectroscopy.

Poster NFC / 573

Simulation of solute transport from deep repository to near surface

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In The Czech Republic a selection of locality for deep repository of spent nuclear fuel is currently in progress. For the safety evaluation it is necessary to verify that the radionuclides penetrating the engineered barriers and a geosphere are not reaching a biosphere in high concentrations. Flow123D and GoldSim SW are used for the solute transport simulations in a geosphere. The advantage of Flow123D is that it allows simulating the transport on a 3D computational mesh. A simulation result is a time development of concentration/activity of the radionuclides. Information from the surface elements is used for effective dose computation.

The issue is that the computed concentration/activity is actually several meters below the surface (on a saturated zone boundary) and hence doesn't include dilution a sorption in unsaturated zone. This contribution tries to tackle this issue by comparison of results of two different models: Computation of effective dose from transport simulation with 1) only the saturated zone included 2) both saturated and unsaturated zone included.

Ad 1) Flow123D is used for the simulations. Results are underground water fluxes across element sides and concentration developments (in time) on mesh elements. From those it is possible to compute mass flux of individual isotopes on each element. We assume a conservative model of men using the contaminated water for drinking and for cultivation and breeding of products for direct consumption. The consumer basket based on the Czech statistical office data is common for both models. The model doesn't account for mixing with rain water in the unsaturated zone.

Ad 2) Flow123D is used for simulations in the saturated zone. Its outputs serve as inputs for ResRad SW used for simulations in the unsaturated zone. ResRad is primarily designed for simulations of different tasks hence it was necessary to modify it.

ResRad is predominantly used for simulation of solute transport from sources located near the surface. It assumes that the initial activity of isotopes is known and that no activity is added during the simulation period (zero mass source). Transport is simulated from the source through the unsaturated zone and it continues to the food chain. The result is an effective dose (time dependent) for a man living in the area.

From Flow123D, the concentration/activity value on a surface element [kg/m3] along with water flux through it [m3/y] are used to compute a mass flux [kg/y] entering the unsaturated zone which represents the intensity of released amount of substance. From this intensity it is possible to determine an effective dose interval for a man.

The result is a comparison of both models. In the unsaturated zone model, the following parameters are variated: unsaturated zone layers thickness, isotope sorption coefficients and the rain water infiltration amount.

Poster NFC / 746

Application of molecular modelling for studying the stability of organic ligands in nuclear fuel cycle

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The crucial agents in partitioning of high level liquid nuclear waste are organic ligands capable of selective complexation of the actinides and lanthanides. To evaluate their applicability, it is necessary to study the stability against undesirable reactions of these ligands, causing their structural changes. One of the main cause for degration is represented by reactions with highly concentrated free radicals.

In this work, we present the summary of theoretical studies for examining the stability of DGA-ligands (TODGA and its derivatives) and CyMe$_4$-BTBP from the last years. The techniques include the analysis of electronic and orbital structure, kinetic reaction properties, or determination of degradation products. One of the recent studies reveals the mechanism and reasons for selectivity of reactions with α-hyroxyoctyl radicals at various positions on the pyridine ring in CyMe$_4$-BTBP, which is surprisingly different from the expected situation described on pure pyridine.

Poster NFC / 749

Sorbents for treatment of highly radioactive liquid waste, originated from VVER NPP severe accident

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The post-accident measure demand has raised after Fukushima Daichi accident. One of the main challenges was related to treatment of large volumes of the highly radioactive waste waters, being generated during the severe accident mitigation. Therefore UJV Řež in the cooperation with CEZ, a.s. and Slovenské elektrárne, a.s. has started to review the concept and potential design of radioactive contaminated water treatment device, dedicated to the VVER 440 and 1000 NPP severe accident, followed by experimental programme, studying sorbing properties of selected sorbents for critical radionuclides.

The potential apparatus concept has been developed as an integral part of the implementation of Fukushima Daichi post-accident measures in VVER units. The volume of treated water is expected to be 10 000 m3, presuming that the treatment of the contaminated water would start 6 months after the successful stabilization of the accident progression based on ERO response according to plant-specific SAMGs. The most important contaminants in the coolant solution would be Cs and Sr radioisotopes, altogether with actinides.

The first important step was to determine of the radionuclide source term and the composition of the solution. The apparatus itself might consist of several columns with various sorbent materials, dedicated to the radioisotope to be eliminated. Potential sorbent were identified, including commercial and laboratory sorbents and nanomaterials. Finally a set of 48 materials was selected.

Nowadays, the research has been focused on material sorptive properties in order pre-select the candidates for further research, studying effect of radiation and coolant solution. Sorbing materials include commercial materials, experimental materials (being developed for specific purposes) and nanomaterials.
Sorption experiments with 137Cs and 85Sr were performed in the first round in order to select the most efficient sorbents of the set that would be submitted to further research (influence of post-accident solution, influence of irradiation due to nuclides in the solution etc.)

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Poster NFC / 830

The use of columns of the zeolite clinoptilolite in the remediation of aqueous nuclear waste streams

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Samples of the zeolite clinoptilolite, mined from Mud Hills in California, have been tested in column experiments to determine their ability to remove radioactive Cs+ and Sr2+ ions. The clinoptilolite was effective in removing both Cs+ and Sr2+ from aqueous solution. Increasing concentrations of Ca2+, Mg2+, Na+ and K+ competed with the Cs+ and Sr2+, and increased elution of Cs+ and Sr2+. Ca2+, Mg2+ and K+ were more effective competitors than Na+, with Na+ concentrations 1 –2 orders of magnitude higher required to produce the same effect. For Na+, it was found that if the concentration was reduced, then the column performance recovered rapidly.
The Mud Hills clinoptilolite has been used in an effluent treatment plant (SIXEP) at the Sellafield nuclear reprocessing site in Cumbria (United Kingdom). There was a large variation in performance for both Cs+ and Sr2+ across the mine site. However, a subsection of the site was identified that provided material within the required specification. This material has been used to remove 134/137Cs and 90Sr successfully from effluents for 3 decades.
A coupled chemical transport model has been developed to predict the performance of the clinoptilolite. The removal of Sr2+ from the effluent was better than expected, considering the zeolite Si/Al framework composition. The formation of Sr-bicarbonate complex ions may be responsible.

Poster NFC / 845

Radionuclides behavior during liquid deactivation of irradiated graphite bushings

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In the current time the task of management the accumulated volumes of irradiated graphite obtained by uranium-graphite reactors decommission (over 250,000 tons in the world) is acute in Russia along

with other countries. It is known during the reactors operation the carbon and chlorine present in the nuclear graphite become activated. Besides various incidents during reactor exploitation occur resulting in contamination of both block and bushing graphite with fission products and actinides. Conditions of the reactor operating determine both properties of irradiated graphite and mechanisms of its pollution. Therefore, a comparative study of the decontamination effectiveness of different species of irradiated graphite is of considerable interest.
Thus the purpose of this work is to study the radionuclides distribution in the volume of decommissioned reactor graphite bushes and to assess the effectiveness of irradiated graphite decontamination by using liquid reagent treatment methods as well. The authors used the samples of irradiated graphite sampled from bushings after two years of operation in the reactors.
Involved graphite samples belonged to the graphite masonry, some zones of which were undergone the incidents. For deactivation experiments the sampled graphite were milled and averaged. Solutions containing acids, alkali, oxidizing reagent, mixtures of acids with fluoride ion were used. Experiments lasted 30 days.
At first stage solutions of 2.5 M H2O2, 5 M NaOH, 1-2 M H3PO4, HCl, HNO3, mixtures of acids with fluoride ion and etc. were used at a temperature of about 22℃. Afterwards were used more aggressive solutions. The efficiency of the deactivation process of bushing graphite samples is minimal in solutions of hydrogen peroxide and alkali, slightly higher in acid solutions, but generally does not exceed 10%, including for Cl-36 and C-14. Increase in the concentration of hydrochloric and nitric acids among with fluoride ions addition gives rise to increasing of Cs-137, Co-60, and Am-241 deactivation degree by almost an order of magnitude. Cl-36 and C-14 extraction degree increases as well. However for Pu-239, U-238 and Sr-90 this effect was not observed. As the temperature rises the recovery of Pu-239 and U-238 increases, while the process time decreases.
It should be noted that an increase in the decontamination time at elevated temperature practically does not affect on the efficiency of Cl-36 and C-14 extraction process.
In the long term taking into account the peculiarity of irradiated graphite properties, it is proposed to evaluate the possibility of using liquid deactivation to reduce the activity of both non-removable and removable graphite elements.

Poster NFC / 854

Can be barium and strontium used as analogues to radium in clay migration?

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Most of the migration data for radium in the performance assessment of deep geological repository, which are used in modelling the long-term safety, were obtained from experiments performed with its chemical analogues - barium and strontium. To our knowledge, there are few data available on radium migration in the engineered barrier of compacted bentonite, possibly due to difficult work with Ra-226. In this study, we applied short-term Ra-223 on planar source diffusion experiments and sorption experiments with the Czech commercial bentonite (named as BaM). Sorption and diffusion behaviour of Ra-223 has been compared with those of Sr-85 and Ba-133.
The apparent diffusion coefficients were as follows Sr>Ba>Ra that is in line with results from batch experiments. Radium apparent diffusion coefficient was one order of magnitude lower than for strontium. Radium distribution coefficient from batch experiments was 4.8 times higher than for strontium and 2.4 higher than for barium.
Based on the results, a difference between radium and its chemical analogues appear to be more significant than expected from reference diffusivities. Therefore, we have focused on the chemistry of these elements under certain conditions in order to answer the title question.

Poster NFC / 858

Development of barrier system in hosting rocks

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Inner and outer safety barriers are constructed when decommissioning nuclear fuel cycle facilities takes place. Combination of different barriers prevents leakage of radionuclides and other pollutants into environment.
The outer barriers formation in geological media is carried out by injection method that allows one to minimize significance of hosting rocks contribution for depositories development. Injected polymer Si-Al compounds which serve as barrier material are to meet special requirements namely low viscosity in wide temperature range, high permeability, low sensitivity to dilution, low viscosity during sol-gel process, adjustable time of gel-formation. Proposed gel-forming composition consists of liquid glass, polymer current agent and modifier. Liquid glass being a main constitution component provides strong binding to hosting rocks. In its turn polymer current agent provides gel structure strengthening and improvement of adhesion to rocks. Modifier determines sorption properties. Developed composition possesses gel-formation time of 17 hours, distribution coefficient for actinides reaches 3·10^4 cm^3/g. Mixture of grouted sand with this composition has filtration magnitude 10^(-3) –10^(-4) m per day. Such barrier allows to prevent spread of radionuclides and nitrate-ions, that is confirmed by the results of calculations and five-year monitoring.
Permeable reactive barriers (PRB) appear for outer safety barriers as well. PRB allows in situ treatment of groundwater using a reactive material that adsorbs, decomposes and removes contaminants. Different materials can be used for creation of complex multi-layer PRB. In this study sorption characteristics of apatite, vermiculite, lightweight expanded clay aggregate (LECA), wood sawdust, perlite, natural zeolite (TREYD) and shungite were specified. Distribution coefficient (Kd) for Cs-137, Sr-90, U-238, Pu-239, Am-241, Tc-99 and stable Cr(VI) were obtained. Speciations of these radionuclides on materials were investigated using the method of sequential extraction.

PAR 2 / 623

On line gas phase separation of At and other halogens under ambient conditions

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Recently, first gas-phase chemical experiments aimed to investigate the adsorption behavior of the heavy elements Cn and Fl on selenium surfaces were conducted. During this experimental campaign, an unexpected formation and transport of a volatile At species was observed. Monte-Carlo simulations of the deposition pattern of this species on quartz, selenium, and gold surfaces were performed, and the corresponding adsorption enthalpies ΔHadsSiO2(AtOxHy), ΔHadsSe(AtOxHy), and ΔHadsAu(AtOxHy) were estimated. The formation of a volatile AtH species in an *in-situ* reaction with atomic hydrogen - being present in trace amounts in the carrier gas - is suggested.

RER 5 / 435

The development of a digital gamma-gamma coincidence/anticoincidence spectrometer and its applications to monitor low-level atmospheric 22Na/7Be activity ratios in Resolute, Canada

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7Be (half-life, 53.2 d) and 22Na (half-life, 2.6 a) are naturally occurring radionuclides of cosmogenic origin. Intrusions of stratospheric air masses into the troposphere followed by dry or wet deposition are the main processes transferring 7Be and 22Na to the earth's surface. These two atmospheric cosmogenic nuclides are simultaneously generated in the upper atmosphere and have similar behaviors after production. They are interesting because their activities and activity ratio can be used as a tracer and radiochronometer for the stratosphere-to-troposphere downward air mass exchange if the current 22Na measurement limitations are overcome.

The analysis of 22Na and 7Be in air-filter samples collected on a daily basis has typically been based on a measurement done with a single HPGe detector using their respective 1274.5 keV and 477.6 keV gamma-rays. There are several practical problems associated with 22Na measurement by this method. In addition to very low 22Na activity concentration (about four orders of magnitude lower than that of 7Be), other important problems include the low measurement efficiency, the losses due to cascade summing and the background from Compton scatter from 208Tl and 40K. This decreases sensitivity and degrades the detection limit of the 1274.5 keV peak, restricting measurement of 22Na in particular.

To improve 22Na detection limit, a digital gamma-gamma coincidence/anticoincidence spectrometer was developed and examined for low-level cosmogenic 22Na and 7Be in air-filter sample monitoring. The spectrometer consists of two bismuth germinate scintillators (BGO) and an XIA LLC Digital Gamma Finder (DGF)/Pixie-4 software and card package. The spectrometer design allows a more selective measurement of 22Na with a significant background reduction by gamma-gamma coincidence events processing. It has been demonstrated that this improved spectrometer provides a more sensitive and effective way to quantify trace amounts of 22Na and 7Be with a critical limit of 3 mBq and 5 Bq respectively for a 20 h counting. The use of a list-mode data acquisition technique enabled simultaneous determination of 22Na and 7Be activity concentrations using a single measurement by coincidence and anticoincidence mode respectively.

Using the newly developed spectrometer, the aerosol samples collected at Resolute, NU, Canada (74.71°N, 94.97°W) airborne particulate monitoring station from 2016 to 2017 were counted. The activity concentrations of 22Na and 7Be were analysed. Based on the results from the Resolute station, the study confirms that the seasonal distribution of 22Na to 7Be activity concentration ratios has a significant peak in spring and winter, and it's relatively low in other seasons. It may indicate that the stratosphere-to-troposphere downward exchange events affect Northern Canada more frequently during spring and winter.

RER 5 / 877

Challenges for environmental restoration of the first uranium mining and milling in Brazil

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The Osamu Utsumi Mine located at the Poços de Caldas plateau, Minas Gerais state, Brazil, was the first uranium mine exploited in the country, in the Poços de Caldas Mining-Industrial Complex

(CIPC) of the Nuclear Industries of Brazil (INB). The operational phase for extracting and processing uranium lasted from 1982 to 1995, when activities were interrupted initiating the decommissioning process. INB had the liability for presenting a mine closure plan following requirements of IBAMA (Brazilian Institute of Environment and Renewable Natural Resources) and CNEN (National Nuclear Energy Commission). Several studies have been conducted to provide useful information to support the establishment of the closure plan. The low grade uranium deposit occurring mainly in the form of pitchblend was associated with a primary mineralization of Zr, REEs, U,Th, Mo. The pyrite present in the rock played an important role in the generation of acid drainage. It was evidenced that the acid drainage from the open pit, waste rock piles and tailings dam is of utmost relevance due to environmental impact and neutralization costs. Two major water courses, the Antas River flowing to Poços de Caldas city and the Soberbo River flowing to Caldas city, have been potentially contaminated by receiving releases of the mining and milling operation, thereby impairing hydric system quality. Huge amounts of waste rocks corresponding to 45 x 106 m3 were dumped into several piles around the mine pit. The largest waste rock pile (WRP-4) has a surface area of 569 x 103 m2 and a deposited volume of 12.4 x 106 m3 in the valley of Consulta stream of the Rio Verde basin. A Plan for Environmental Remediation (PRAD) was developed guided by a Reference Term written by the Environmental and Nuclear regulators to provide solutions for site decommissioning encompassing the mine pit, waste rock piles, tailings dam and industrial area. The present study is based on a comprehensive sampling of the WRP-4 and the tailings dam performed for chemical and radiological characterization and will be thorougly discussed.

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Production of Np isotopes in nuclear reactions for a standard material in accelerator mass spectrometry

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The techniques of highly sensitive mass spectrometry is achieving rapid development recently. Several elements or nuclides that were not supposed to be applicable in that technique are now quantitatively analyzed. Especially, for long-lived actinides elements, the techniques are becoming more and more important as a promising alternative of radioactivity measurements. The tracer nuclide for chemical recovery determination, which has to be non-existent in nature and not contained in target samples, is absolutely necessary for that purpose. Tracers for several elements are available now, but the tracer for neptunium has not been developed, yet. We seek for an efficient method for production of Np-236 in the ground state of 1.54×10^5 y in half life as a candidate of the tracer nuclide.

In present study, we try the Np tracer production in the reactions of Th-232 + Li-7 and U-238 + p to aim to perform an application for Np contamination in environmental samples as the goal.

For irradiation of beam, two types of target stacks were prepared. One includes several targets of electrodeposited Th or U of ca. 1 mg/cm^2 on Al foils. The other includes a thick target disc of 100 mg/cm^2 Th or U. The former stack was utilized to check the production rate and interfering products depending on the projectile energy dumped in the stacking targets, and the latter was for the total properties. Foil of silver or copper of natural abundance was also used to monitor beam intensity calculated from produced radioactivity of In-111 or Zn-65 during the irradiation as well as current monitoring with a Faraday cup in the beam course.

We have performed irradiation with Li-7 ions of 42 MeV at the AVF cyclotron of RIKEN and proton

of 39 MeV at the AVF cyclotron of RCNP of Osaka University. In order to isolate Np atoms from the irradiated targets, the chemical procedures were performed. As an example for irradiated Th samples, the target material was dissolved in 3M HNO_3 with Np-237 tracer for checking chemical recoveries and dried up in heating. Then, procedure of dissolution of the residue in conc. HNO_3 and drying up was repeated three times. Finally, the sample was dissolved again in conc. HCl, and adjusted to the 10 M HCl solution of 4 mL. The solution was subjected to the separation procedure using a TEVA resin column and treated with 10 M HCl, followed by 3 M HNO_3, for purification, and finally 0.1 M HCl for elution of Np.The purified samples of Np were subjected to alpha spectrometry and gamma spectrometry to check the by-products emitting radiation.
As a preliminary result, the measured gamma spectra show that the final solution from the sample was still contaminated by protactinium products. The intensities of uranium X-rays and the gamma rays following decay of Np-236m of 22.5 h in half-life suggest that the production rates of Np-236g were around 5×10^9 atoms/g-Th/h/p micro A and 5×10^10 atoms/g-U/h/p micro A, assuming the isomeric ratio of products, m/g, of ca. 5. The ratio was estimated from production data of the same nuclides in proton induced reaction of 238U1). Spiking amount of Np-236 per sample is estimated to be around 10^8-10^9 atoms.
Analysis of the results is still in progress and additional experiments are under planning at present.

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RER 5 / 694

Impact of nuclear power plants on radiocarbon in the atmosphere and biosphere of Slovakia

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There have been two nuclear power plants (NPPs) in operation in Slovakia - Jaslovské Bohunice and Mochovce. Radiocarbon as one of the most important radionuclides for delivering of long-term radiation doses to the public has been regularly monitored in the NPP surrounding areas as well as in the city of Bratislava. In the vicinity of both NPPs atmospheric carbon dioxide has been sampled on a monthly basis and its radiocarbon activity has been measured by gas proportional counting. The atmospheric $^{14}CO_2$ data from these two sampling stations will be presented and compared with results from other sampling sites from both urban and rural parts of Slovakia.
Carbon dioxide is assimilated by plants during photosynthesis and therefore connects radiocarbon in the atmosphere with the surrounding biosphere. Therefore stinging nettle samples were taken from the sampling area for comparison with the atmospheric data. These biota samples provide information about ^{14}C activity accumulated during given plants'growth. Annual tree rings have been used as an archive of past radiocarbon levels in the biosphere as well. Tree ring samples from the Jaslovské Bohunice area covering a period of 20 years, measured by accelerator mass spectrometry, were compared with atmospheric radiocarbon data collected at the Jaslovské Bohunice and Bratislava (background) stations.

RER 5 / 710

Speciation of Uranium: Validation of a database by modelling techniques and comparative experiments

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Uranium mining legacies and enhanced radioactivity in usable groundwater and surface water resources cause environmental hazards. To predict radionuclide migration, for installing effective water purification technology and as decision basis for remediation strategies of the contaminated sites a profound knowledge of uranium speciation in these waters is essential. Predictive modelling of uranium speciation in natural waters uses equilibrium thermodynamic data and adequate speciation software. However, available thermodynamic data sets differ considerably with respect to number of uranium compounds and with regard to completeness and consistency. Hence, model results may vary widely depending on the used data set[1]. In addition, data for radionuclides published in recent years have only been partially integrated into the databases.

The validity of the thermodynamic data for the environmental media affected by uranium mining legacies is of utmost importance. Therefore, a comprehensive and consistent database was established according to current knowledge. The uranium data included in the database is based on the NEA TDB[2] and is modified or supplemented as necessary e.g. for calcium and magnesium uranyl carbonates. The specific ion interaction theory[3], which is sufficient for the considered low ionic strengths, is used to estimate activity coefficients. The success of this approach was validated by comparative experimental investigations and model calculations (PHREEQC[4]) for several model systems. The waters differ in pH (2.7 –9.8), uranium concentration (10^{-9} - 10^{-4} mol/L) and ionic strength (0.002 - 0.2 mol/L). We used chemical extraction experiments, ESI-Orbitrap-MS and time-resolved laser-induced fluorescence spectroscopy (TRLFS) to measure the uranium speciation. The latter method is nonintrusive and therefore does not change the chemical composition of the investigated waters. This is very important, because any change of the system under study may also change the speciation.

For all investigated systems at least the two species with the greatest expected relative abundance were detected. Moreover, all species with expected concentrations > $1{\cdot}10^{-7}$ mol/L were observed. The present database is consistent and can be considered validated for the model systems and comparable waters, whereas for waters of very different composition an additional validation is necessary. The validity range of the database generally extends over a wide range of compositions, pH (2-12) and also to higher ionic strengths (up to 3 mol/L).

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PAR 2 / 512

A Novel Method to Prepare Reference Sources for Radioactivity Metrology Using Ion Exchange Membrane

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For now, most radioactive reference sources are manufactured with electro-deposition. However, the electro-deposition procedure takes quite a long period of time and the operations are conducted using flammable solvents under high temperature. In addition, the products of electro-deposition may be oxidized and exfoliate after years of usage which can cause extra uncertainty of quantitative metrology. Thus, a more convenient method to prepare thin, homogeneous and chemical and mechanical stable references sources is required.

HP Membrane (DuPont Nafion PFSA Membrane) was selected to fix the radioactive nuclides (Am-241, Sr-90, etc.) and the aluminium plates were used as the supporter. The adsorption was conducted

in the quartzose reactor. Some primary parameters of the reference sources, such as activity, surface emission rate, self-absorption, uncertainty and uniformity were tested to evaluate whether the quality meet the requirements of class one reference sources.
In the further investigation, we will focus on manufacturing novel membranes which are more suitable for preparing reference sources. Now available commercial membranes, as DuPont Nafion membranes, do not specially satisfy the need because of their space network determined by the cross-linkage. The radioactive nuclides adsorbed inside the membranes may reduce the emission rate and the uniformity. Therefore, new membranes especially the thin ones with functional groups distributing uniformly only on the surface are needed. On the other hand, we will check the interaction between the functional groups and radioactive particles emitted from the nuclides adsorbed and furthermore evaluate the radiation damage to ensure the stability of the reference sources.

RPH 2 / 689

CRAB2RABBIT solid target solution

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The CRAB2RABBIT system represents a newly developed solution for processing of cyclotron solid targets. It cosnsists of two parts - the RABBIT, which is an automatic loading and transportation system, and the CRAB - a radiochemical module enabling dissolution of the solid target and its radiochemical processing.
As designed, the CRAB2RABBIT system covers and automatically controls all the steps of production of a labelled compound, beginning with transport of the prepared target to the cyclotron, followed by loading to a target holder, unloading after the irradiation, transport to the selected hot cell, dissolution, purification and labelling. The systems enables processing of main radionuclides produced by solid target irradiation, such as Cu-64, Ga-68, Zr-89 and others.

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An experimental approach to reactive transport in geomaterials: GeoPET

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Detailed understanding of reactive transport in geomaterials of chemical species, including radionuclides, is required for the utilization of the subsoil, e.g. for designing ore production by in-situ leaching, or for radioactive waste disposal. To complement the well-established conventional approach, i.e. computer model simulations based upon bulk material parameters and geochemical data bases, we apply process tomography with positron emission tomography (GeoPET) for direct observation and parameterization of the reactive transport processes. This enables to consider heterogeneity as pervasive feature of processes in complex media. One example is localized flow meandering along fractures, where preferential flow may jeopardize leaching efficiency. On the other hand, fissure networks through otherwise tight material could provide fast transport pathways through geological

barriers.
Our workflow consist of
1. production of appropriate PET-nuclides and labelling,
2. transport experiment on samples of drill core size with the labelled species,
3. recording of PET-data (list-mode-files) during the course of tracer propagation,
4. computation of PET-frames with appropriate frame rate and correction for material effects,
5. parameterization of the spatiotemporal data set with the target parameters effective volume distribution and velocity distribution.
The choice of PET-nuclides is broader than in common biomedical PET applications, because longevity and toxicity of the tracers are inconsiderable, but spatial resolution and efficient corrections for attenuation and scatter require attention. The development of the GeoPET method during the past decade is described in Kulenkampff et al. (2016).
As illustration, we present an example from ore leaching, where the leaching solution is flown through an artificial fracture. During leaching we experimentally determined the macroscopic flow field with GeoPET. With these hydrodynamic data we are able to establish a realistic and light-weight reactive transport model which can directly serve for efficient design of leaching.
The procedure is one good example for the benefit of radiotracers for unravelling complex processes by non-destructive molecular imaging. We strongly suggest utilizing this distinguished tool, in particular for .parameterization and upscaling of heterogeneous reactive transport models.

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RPH 2 / 822

Radioactive Gold Nanoparticles with Beta Energy and Auger Electron Cascades In Nanomedicine: Green Nanotechnology and Radiochemical Approaches

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Theranostic Tumor-Specific Gold-198 Nanoparticles Through Green Nanotechnology—Implications In Nanomedicine For Concurrent Molecular Imaging and Tumor Therapy

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Auger Electrons were once neglected for oncological applications because of their low energy and consequent short range. However, our recent discovery of the production of well-defined and clinically optimal Au-198 nanoparticles, through green nanotechnology, has changed the landscape of Au-198-derived Auger electron cascades, in oncology. Au-198 provides highly useful biologic toxicity to tumor cells and tumor masses for therapeutic applications in oncology. Internally deposited radionuclides, within tumor cells, result in radiation-induced ionizations, excitations, nuclear recoil, chemical transmutations, and local charge effects. γ-Photons, x-ray photons, and energetic negatrons and positrons which have a range of activity equivalent to many cell diameters are characterized by low linear energy transfer and oxygen-dependent biologic effects. In this context, 198Au provides a desirable beta and gamma energy emissions that can be used either to destroy tumor cells/tumor tissue (βmax = 0.96 MeV; half-life of 2.7 days) or to image the neoplasms in real time (gamma-ray energy of = 0.407 MeV). Its penetration range (up to 11 mm in tissue or up to 1100 cell diameters) is sufficiently long to provide cross-fire effects to destroy tumor cells, but short enough to minimize radiation exposure to tissues near the capsule periphery. Au-198 and its nanoparticles comprise of over 85% of the non-radioactive gold. This composition is 'Tailor-Made'for Auger, Coster-Kronig, and super-Coster-Kronig transitions with subcellular ranges (nanometers) as Au-198 decays by electron capture and/or internal conversion to extremely low-energy electrons through Auger effects. Our studies have already generated extensive therapeutic efficacy data (Katti et.al: PNAS 2012) from the treatment of tumors in mice and tumor bearing dogs (where the disease mimics human cancers). These findings have presented compelling prospects for the clinical translation of Auger electron-emitting radionuclides (such as Au-198) in treating human cancers. We envisioned that radioactive gold nanoparticles, which are inherently theranostic, will present realistic prospects in achieving optimal therapeutic payloads in prostate and other solid tumors because of their (i) size; (ii) inherent affinity toward tumor vasculature and (iii) most importantly through their favorable radiochemical properties. We have considered a scientifically sound and futuristic approach, involving green nanotechnology, for the synthesis, stabilization and incorporation of tumor specific features into radioactive theranostic gold nanoparticles. We strongly believe that it is imperative to minimize/eliminate the utility of toxic chemicals in the overall generation of nanoparticles for various medical and allied applications. Our approach to achieving tumor specificity and optimal retention of therapeutic payloads of radioactive gold nanoparticles at prostate tumor sites (and within various disease sites) has involved surface conjugation of radioactive nanoparticles with epigallocatechin-gallate (EGCg) and using a host of tumor specific biomolecules from various herbs and plants. We hypothesized that the redox properties of EGCg (comprised of polyphenolic constitution) could be effectively utilized by using it as a reducing agent to convert radioactive gold precursor to the corresponding radioactive gold nanoparticles—a 100% green nanotechnological process without the intervention of any toxic chemical. An additional advantage of using EGCg is its ability to target 67kDa Laminin receptor (Lam 67R) which is over expressed on human prostate cancer cells and also in various other human tumors. EGCg has been known to bind to Lam 67R with excellent specificity and selectivity. Therefore, the fabrication of EGCg functionalized radioactive gold nanoparticles (198AuNP-EGCg) in our laboratory may be considered as a genesis of 'Green Nanotechnology in Medicine'. In this lecture, I will present experimental results that validate our hypotheses and also present full details encompassing: (1) synthesis and complete characterization of EGCg and various phytochemical functionalized tumor specific radioactive gold nanoparticles; (ii) evidence of endocytosis due to Laminin receptors on prostate tumor cells and quantitative estimation of AuNP-EGCg within PC-3 cells using neutron activation analysis (NAA) (ii) experimental results on the theranostic applications of Au-198 prostate tumor-specific AuNP-EGCg through in vivo studies in mice and tumor bearing dogs; and (iv) therapeutic efficacy studies of 198AuNP-EGCg in prostate tumor bearing mice and dogs, demonstrating excellent tumor retention resulting in over 85% inhibition of tumor growth through singular intratumoral injection. The overall oncological implications of this and a range of new nano theranostic agents in treating human prostate and various other solid tumors will also be discussed.

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SEP 5 / **478**

Deep learning approach for An/Ln separation problem

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A problem of separation of trivalent f-elements is one of key problems of spent fuel reprocessing as well as processing of lanthanide-containing mineral resources. Chemical similarity leads to development of long cascade separation equipment with low efficiency on each step. Development of new selective ligands may allow us to simplify separation scheme and increase process efficiency.
"Deep learning" is one of popular trends in science and technology nowadays. It has already proved its chemical usability in drug design area and continues spreading over other areas of chemistry.
Here we present results of development of neural net models that were trained to predict complex stability constants for a number of trivalent f-elements. While requesting a lot of time and computational resources for training, such models are easy in use and quite fast for prediction purposes. We reached determination coefficient (R^2) values up to 90~95% that allow us to determine perspective in terms of separation ligands at a design stage.

NAM 5 / 615

Determination of iron concentration in brain tissues using PIXE method

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Particle Induced X-rays Emission (PIXE) analysis is non-destructive method used in many fields of interest. With this method, an absolute concentration of investigated elements can be determined at ppm levels. Collaboration between the Medical Faculty and the CENTA laboratory was set up to use PIXE analysis for determination of iron concentrations in rabbit and human brain tissues, which offers 2 or more orders better resolution than Energy-Dispersive X-ray Spectroscopy (EDX). First PIXE measurements and determination of iron concentrations were done for rabbit brain tissues which were placed on thick silicon wafers. Secondly, instead of the thick silicon wafer we focused on the development of a thin sample holder dedicated for a human brain tissue. A new sample holder consists of the square-shaped plastic frame and a mylar foil which is attached to the frame using a double-sided tape and promises significant background reduction in comparison with the thick silicon wafer. With this configuration, a thin brain tissue (~ 5 μm) can be placed on this sample holder and can be measured using PIXE method as a thin sample as more than 99.99 % of proton beam passes through the sample. This holder can be also used for EDX and other measurements. Preliminary results of iron concentration measured in a healthy human brain tissue (substantia nigra) exhibit an inhomogeneous iron distribution with concentrations up to 2 μg/cm^2. A comparison of the gender, age or health status dependency on iron concentration distributed in human brain tissue measured by PIXE and those carried out by EDX method will be presented, as well as a comparison of simulations of proton (PIXE) and electron (EDX) beam interactions with the brain tissue samples.

SEP 5 / 554

Crystallization purification full scale test of U-Pu mixture

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Nowadays the concept of closed nuclear fuel cycle (CNFC) for fast neutron reactors (FR) is developed in Russia. CNFC can be implemented for FR with sodium coolant using mixed uranium-plutonium oxide (MOX) fuel and for reactors with lead coolant using mixed U-Pu-(Np-Am) nitride (MINT) fuel. By “PRORYV”Project the combined “PH-technology”was proposed as the technology for reprocessing of MOX and MINT fuel 1. This technology combines pyrochemical and hydrometallurgical methods of SNF reprocessing.

To work on the hydrometallurgical part of “PH-technology”the full-scale crystallization purification test bench was made at JSC “Siberian Group of Chemical Enterprises”2. This test bench consists of such steps as SNF extraction, reductive striping of Pu, U stripping, concentration of striped Pu-Np mixture and crystallization refining of U-Pu-Np mixture.

At present time according to current industrial SNF technologies Pu is separated, transferred into oxide form and then at the stage of fuel fabrication added to uranium. However the crystallization process due to the similarity of U(VI) and Pu(VI) properties allows to recover them simultaneously as uranyl and plutonyl nitrate hexahydrates that is much in line with concept of non-proliferation of nuclear materials.[3,4]

The crystallization experiments of pure uranyl nitrate hexahydrate and mixture of uranyl and plutonyl nitrate hexahydrates were performed with the use of the full-scale crystallization purification test bench. The crystallization purification experiments of uranyl nitrate showed that such elements as Ce, Nd and La don't co-crystallize with U and mostly get into the mother-washed solution that allows increasing the level of purification by the factor of 1-2. During the experiments of uranyl and plutonyl nitrate hexahydrates co-crystallization the co-crystallization coefficient range from 0,7 to 0,8 according to the process conditions.

Achieved results have confirmed not only earlier successfully proven possibility of combined separation of U(VI) and Pu(VI) from nitric acid solution as uranyl and plutonyl nitrate hexahydrates, but also showed the ability of using crystallization process for SNF reprocessing in industrial scale.

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NAM 5 / 578

Alpha-spectrometry in aqueous solution

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Contemporary alpha ray spectrometry suffers from number of shortcomings: e.g. high time consumption, requirement of expensive chemicals and electrodeposition equipment as well as changing electrodeposition yields. Nevertheless, these and other unfavorable aspects have to be accepted to obtain decay informations about the alpha particle emitting radionuclides.

The aim of our current project is to overcome most of these unfavorable aspects. One way to reach that goal is to couple the chemical sample preparation with the detection process itself. This could be realized by modifying the detector surface with an extraction agent. Based on the type of grafted extraction agent, the well-known radiochemical separation techniques [KRA13] could be performed directly on the surface of an alpha detector. In order to test the feasibility of such an approach the well-established simulation programs SRIM (the Stopping and Range of Ions in Matter) [ZIE10] and AASI (Advanced Alpha Spectrometry Simulations) [SII05] were used to predict the fundamental properties of alpha spectra obtained by grafting a specifically bonding layer to a detector surface.

Figure 1 shows an example of such a simulation where 4.2 MeV alpha particles were emitted from a reasonably thick water layer (30 µm) towards a detector with a thin grafted layer (red line). As one would have expected, the spectrum shows a wide range of alpha-particle energies with no structure up to the emission energy of 4.15 MeV. If one simulates, on the other hand side, the decay from the grafted layer a spectrum is obtained with high resolution as indicated by the low full width at half maximum (blue line). In a real system one would expect a superposition of both contributions.

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Based on these promising results a practical approach was attempted. As a proof of principal, alpha detectors were modified with sulfonic acid functional groups, a renowned class of strong cation exchangers [INT86]. It was possible to attach [238U, 234U] uranyl cations onto the detector surface immersed in a solution containing these ions while acquiring the spectrum. In fact, the obtained spectra closely resembled the results of the simulation. One observes clearly distinguished peaks over a continuum. Moreover, by applying a hydrochloric acid solution, the uranyl cations could be redissolved. The resulting spectra taken from demineralized water showed neither the continuum nor the typical alpha peaks at 4.18 and 4.75 MeV, respectively.

The ability to graft molecules by covalent bonding to detector surfaces enables the selective extraction and detection of radionuclides in aqueous solution. This innovation could be beneficial to all applications of alpha ray spectrometry, from environmental measurements to the investigation of the actinide and transactinide elements.

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SEP 5 / 557

Study on chemical separation method for the analysis of isotope ratio of 135Cs to 137Cs in soil

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Study on chemical separation method for the analysis of isotope ratio of 135Cs to 137Cs in soil
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In order to confirm the completion of the decommissioning of a facility, the radioactivity concentration of soil at the site has to comply with criteria for site release. However, extensive area in the eastern Japan was contaminated by radiocesium as a result of the accident at the Fukushima Daiichi Nuclear Power Station. It is essential the contamination by radiocesium during operation and decommissioning of the facility is discriminated from that by the fallout due to the accident for the decision of site release.
The isotope ratio of 135Cs to 137Cs could be used to determine the origin of 137Cs. Zheng et al. reported an analytical method to estimate the isotope ratio of 135Cs/137Cs in soil contaminated by the accident, which involved the ammonium molybdophosphate (AMP)-selective adsorption of Cs and subsequent two-stage ion-exchange chromatographic separation, followed by the detection of 135Cs and 137Cs via triple-quadrupole inductively coupled plasma-mass spectrometry (ICP-MS/MS) 1. Nuclides that could possibly interfere during the measurement of 135Cs and 137Cs using ICP-MS/MS are 135Ba, 137Ba, 95Mo40Ar, 97Mo40Ar, 119Sn16O, and 121Sb16O. Zheng et al. minimized the influence of Mo from AMP by using a collision cell with N2O gas. However, N2O gas has been designated as a banned substance in Japan since February 28, 2016. Thus, we developed a scheme for the chemical separation of Cs that does not use AMP.
Environmental soil contains approximately 628 ppm of Ba, 1.1 ppm of Mo, 2.1 ppm of Sn, and 0.4 ppm of Sb 2; our sample solution contained similar amounts of Ba, Mo, Sn, and Sb in a 1 M HCl solution. The major elements and Ba were crudely separated by co-precipitation with iron hydroxide and carbonate by adding 10 mg of Fe as FeCl3, 0.2 g of NaHCO3, and NH3 solution and subsequent boiling to grow the precipitation, followed by suction filtration. In this instance, anionic Mo, Sn, and Sb were also roughly removed by an Anion-SR disk set under a filter paper. The transfer rates of Ba, Sn, and Sb into the filtrate were less than 3%, 1%, and 18%, respectively. There was a considerable variation in that of Mo. The filtrate containing Cs was evaporated to dryness, the residue dissolved in a 3 M HNO3-H2O2 solution, and then, refluxed for 5 h to remove the ammonium salt. After evaporation and dissolution in 3 M HNO3, Cs was purified by solvent extraction using calix4arene-bis(t-octylbenzo-crown-6) in 1-octanol.
This research project has been conducted as the regulatory supporting research funded by Secretariat of Nuclear Regulation Authority (S/NRA/R), Japan.
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Comparison of 3 radio-imagers (Cyclone™/Beaver™/µ-imager™ DFINE) to characterize radioactive emissions of 3H, 14C and natural Uranium samples.

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Radio-imagers, mainly dedicated to biological and medicine applications, can be used to reconstruct 2D mapping of radioactivity on a surface. The resulting image, called Digital Autoradiography (DA), gives a non destructive analysis of radioactivity.
Recently, a new scope for the radio-imagers has emerged: nuclear facilities decommissioning. Indeed, in such context, surface contamination need to be accurately localized and characterized in

terms of activity and radionuclide identification. DA method appears to be a suitable candidate for this issue.

Currently, various radio-imagers are commercialized, mainly for research purposes, but up to now, there is no device able to investigate surface contamination at the scale of a facility. The MAUD project (Measurement by Digital Autoradiography) aims at designing a new in situ device able to assist in the dismantling process of nuclear facilities. For this purpose, three DA technologies are currently investigated: 1) Cyclone™, a laser used to read phosphor screen and developed by Perkin Elmer, 2) Beaver™, the Gas Detector designed by AI4R company and 3) μ-imager™ DFINE, the Solid Scintillation Detector developed by Biospace Lab.

Autoradiographic measurements of a rock sample containing natural Uranium, and of samples artificially labeled with 3H and 14C, have been acquired using the three radio-imagers quoted above, with an exposure time of 1 hour. The present study proposes a comparison of the resulting images, in terms of qualitative and quantitative results, in order to identify the strength and the weakness of each device.

A preliminary study has shown that a linear correlation should be expected between the counting of the Beaver™ and the DFINE one. It will also be possible to estimate the efficiency of each detector for the three kinds of radionuclide tested.

This work has been performed within the Investments for the future program of the French Government and operated by the French National Radioactive Waste Management Agency (Andra). These researches are also funded by CEA.

SEP 5 / 620

A magnetic nanocomposite based on natural hydrocolloids for efficient removal of liquid radioactive waste

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Efficient remediation of liquid radioactive waste, produced as in research and medicine, is crucial for increasing safety during the necessary storage of this material. A novel bio-magnetic nanocomposite sorbent based on a natural product (Karaya gum) and magnetite nanoparticles was tested for the efficient removal of radioactive phosphorus 32P from liquid radioactive waste. This non-toxic nano-composite is well-suited to be used as a nano-hydrogel for storage of radioisotopes on a smaller space and without the risk of spills inherent to the initial liquid material. The maximum adsorption capacity of the nanocomposite synthesized in this study was found to be over 15 GBq/g. We present a thorough morphological characterization of the novel synthesized bio-magnetic nano-composite, as well as discuss the possible phosphorus adsorption mechanisms.

NAM 5 / 680

Methodology for the preparation and validation of plutonium age dating materials

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The present work describes a method for the preparation and validation of plutonium age dating reference materials 1. The prepared reference samples can be used to validate experimental protocols

for determining the production date of plutonium via the 234U/238Pu, 235U/239Pu, 236U/240Pu and 241Am/241Pu chronometers. The starting material was prepared using reactor-grade plutonium, which was purified using a dedicated method to guarantee high Pu recovery, while maximizing U and Am separation efficiencies. The U and Am separation factors were determined by the addition of high-amounts of 233U and 243Am spikes and their re-measurement in the final product. Th and Np were measured from all aliquots, but it was not aimed for Np/Am or Th/U chronometers.
The age values obtained for the test samples using the different parent/daughter pairs (chronometers) are in excellent agreement and agree also with the known production date. The date of the last chemical separation is known to within one hour, and not biased by any residual daughter nuclides. The different forms (liquid and solid) and concentrations allow the use of various techniques, e.g. alpha spectrometry, ICP-MS, TIMS or SIMS. The method was demonstrated to be directly applicable for the preparation of a certified reference material (similar to IRMM-1000).
The prepared material is intended for quality control and assessment of method performance in nuclear forensics and safeguards.

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NAM 5 / 625

Characterization concept for the disposal of radioactively contaminated mercury wastes from the decommissioning of nuclear facilities

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The disposal of mercury from nuclear applications that contains radionuclides due to contamination or neutron activation is very challenging, not only due to its radiotoxicity but also due to its high chemotoxicity and mobility in the environment. Elemental mercury has been used in several nuclear facilities, for example as coolant and shielding material in early fast reactors, as sealant in hot cells, as target material in spallation targets and as catalyst in isotope separation research. In Jülich, about 600 kg of radioactively contaminated mercury originate from the decommissioning of hot cell facilities in which mercury was used as a sealant material. A disposal concept for such radioactive mercury wastes is currently developed in the framework of the German national research project PROMETEUS (PROcess of radioactive MErcury Treatment under EU Safety-standards), funded by the German Federal Ministry of Education and Research under grant numbers 15S9266A–B. This disposal concept includes a (radiological) characterization, decontamination and possibly a clearance of the purified mercury as well as a safe disposal, i.e. conversion and solidification / immobilization of the radioactive residues.

A comprehensive radiological characterization concept for mercury was developed and is the focus of this work. The main challenges of the radiological characterization originate from the inhomogeneity of the waste material as well as the inhomogeneous activity distribution and the high density of mercury which leads to a high intrinsic radiation shielding. The characterization starts with the non-destructive gamma-spectrometric measurement of the samples, for which a special gamma detector arrangement was set up consisting of two semi-planar HPGe detectors facing each other. This setup allows the simultaneous measurement of thin mercury samples from opposite sides, thus increasing the efficiency of the measurements and also allowing the detection of inhomogeneities in the samples. The gamma detector setup is not only interesting for waste characterization, but furthermore it can be used for clearance measurements after a decontamination procedure of mercury wastes. This decontamination procedure including vacuum distillation and washing techniques aims at a separation of (radioactive) residues from mercury to enable a subsequent clearance of the mercury.

Following the non-destructive gamma-spectrometric characterization, aliquots of the material are dissolved to perform further characterization methods, i.e. LSC measurements and alpha spectrometry. A chemical separation of mercury and the different radionuclides is performed to quantify alpha-emitting nuclides. This is achieved by dissolution of the material and a subsequent separation by extraction chromatography. Mercury is thereby separated from the alpha-emitting nuclides as it significantly reduces the efficiency and resolution of alpha-spectrometric measurements and in a subsequent step different alpha-emitters are separated from each other to be measured accurately. This paper presents recent results from the chemical separation and characterization of radioactively contaminated mercury wastes.

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Interaction of U(VI) with α-isosaccharinic acid under acidic conditions: characterization of the formed complexes

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Cellulose is a very common organic polymer and also present in considerable amounts in low and intermediate level wastes (LILW). Since it is considered to stabilize LILW with cementitious materials and to use cement-based materials as construction and backfilling material, the alkaline degradation of cellulosic material has to be taken into account. This process will lead to the formation of water-soluble carboxylates and hydroxycarboxylates, with isosaccharinic acid (isa) being the main degradation product. It was shown that the α-form of the polyhydroxy-carboxylic acid is a stronger complexant compared to the β-form and that the interaction with radionuclides affects the solubility and the sorption behavior adversely 1. Only a limited number of studies focused on the interaction of isa with UO₂²⁺. Hence, there are gaps in thermodynamic databases for this system. The basis for reliable thermodynamic data is a detailed structural knowledge about the formed species in the system of interest.
This issue prompted us to characterize the formed complexes in the UO₂²⁺-isa system on a molecular level. Our approach to elucidate the mechanisms in aqueous solution combines different spectroscopic techniques (UV-vis, ATR-FTIR, EXAFS, and luminescence) with DFT-calculations. The mutual influence of UO₂²⁺ and isa on their speciation as well as the three detected complexes will be discussed. The outcomes are on the one hand the basis for thermodynamic investigations as well as for studies under neutral and alkaline conditions. They provide on the other hand also important information concerning the behavior of α-isa as ligand, including dominant binding motifs.

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SEP 5 / 631

Crystal structure and solubility of radium sulfate and carbonate

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Radium is the heaviest alkaline-earth metal. It has no stable isotopes and the most long-lived and abundant isotope is ^{226}Ra with a half-life of 1600 years. Radium and its short-lived daughters are among the most radiotoxic elements. Enhanced radium concentrations can be found in different waste streams: uranium and other metal mining, fossil fuel combustion, oil and gas production among others. Moreover, radium ingrowths occur in spent nuclear fuel, which makes it a critical element for long-term safety assessment of final nuclear waste repositories 1. One of the main factors controlling radium migration in the environment is its co-precipitation with other alkaline-metal sulfate phases, for example barite ($BaSO_4$). In some natural sulfate free waters, radium mobility can be controlled by its co-precipitation with carbonate minerals, for example, witherite ($BaCO_3$) 2. The mechanism and degree of radium co-precipitation with barite and witherite depends mainly on crystal structure and the solubilities of pure barite, witherite, $RaSO_4$ and $RaCO_3$. The crystal structure and solubility of barite, witherite and other non-radioactive alkaline-earth metal sulfate and carbonate phases are well established, but little is known about the properties of $RaSO_4$ and $RaCO_3$. The objective of this work was to study the crystal structures and solubilities of radium-barium sulfate and carbonate and obtain more insight into radium co-precipitation mechanisms and the migration of radium in the environment.

For this purpose, an old radium source was disassembled to obtain radium-barium sulfate ($Ra_{0.76}Ba_{0.24}SO_4$). Radium-barium carbonate ($Ra_{0.76}Ba_{0.24}CO_3$) was synthesized from the $Ra_{0.76}Ba_{0.24}SO_4$ obtained by three cycles of $Ra_{0.76}Ba_{0.24}SO_4$ heating in 1.5 mol·L^{-1} Na_2CO_3 up to 85 ℃, cooling and subsequent removal of supernatant 3. The $Ra_{0.76}Ba_{0.24}SO_4$ obtained was characterized by an X-ray Diffraction (XRD) technique. Rietveld refinement showed that $Ra_{0.76}Ba_{0.24}SO_4$ crystallizes in the *Pnma* (62) space group and is isostructural with pure barite ($BaSO_4$). Density Functional Theory (DFT) and Vegard's law were used to obtain atomic coordinates and unit cell parameters of pure $RaSO_4$ (a=9.129(8), b=5.538(3), c=7.313(5)) 4. The synthesized $Ra_{0.76}Ba_{0.24}CO_3$ was characterized by XRD and Extended X-Ray Absorption Fine Structure (EXAFS) techniques. EXAFS refinement indicated that radium is nine-coordinate in solid $Ra_{0.76}Ba_{0.24}CO_3$ with a mean Ra-O bond distance of 2.885(5) Å. Comparison of the $Ra_{0.76}Ba_{0.24}CO_3$ and witherite ($BaCO_3$) XRD patterns showed these solids are not isostructural.

The solubilities of $Ra_{0.76}Ba_{0.24}SO_4$ and $Ra_{0.76}Ba_{0.24}CO_3$ were measured from over- and undersaturation as a function of ionic strength (NaCl media). The concentration of radium was measured by gamma spectrometry after separation of the aqueous phase from colloids using ultracentrifugation to calculate its concentration and the apparent solubility product. The extended specific ion interaction theory (ESIT) was used to extrapolate the apparent solubility product of $Ra_{0.76}Ba_{0.24}SO_4$ and $Ra_{0.76}Ba_{0.24}CO_3$ to zero ionic strength and obtain the Ra^{2+} - Cl^- ion interaction coefficients. It was shown that the apparent solubility products of $Ra_{0.76}Ba_{0.24}SO_4$ and $BaSO_4$ as well as those of $Ra_{0.76}Ba_{0.24}CO_3$ and $BaCO_3$ increase almost identically from zero ionic strength with an increase in the ionic strength of NaCl which indicates that Ra^{2+} and Ba^{2+} undergo similar short-range interactions with Cl^- in aqueous media as might be expected given the similar ionic radii of Ba^{2+} and Ra^{2+}. The solubility of $Ra_{0.76}Ba_{0.24}SO_4$ at zero ionic strength is a little lower (0.2 $\log_{10}$ units) than the solubility of pure barite ($BaSO_4$). The results reveal that both the solubility and crystal structure of $Ra_{0.76}Ba_{0.24}SO_4$ is very similar to the solubility and crystal structure of barite and that radium and barium form an ideal solid solution in the sulfate phase. The solubility of $Ra_{0.76}Ba_{0.24}CO_3$ at zero ionic strength is significantly higher (1 $\log_{10}$ unit) than the solubility of pure witherite ($BaCO_3$). This confirms that $Ra_{0.76}Ba_{0.24}CO_3$ and pure $RaCO_3$ are not isostructural with witherite ($BaCO_3$).

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Study of phosphate species in soils by radioisotopic labeling

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The nutrient cycle of soils is mainly determined by the concentration of macro and micro nutrients in the soil solution directed by the interactions between the solid phase of soil and the soil solution. These interactions are essential in case of nutrients, such as phosphate anion, strongly binding some soil components. Phosphate anion can precipitate with different cations (e.g. iron, calcium, aluminum) as well as react with the soil organic components. Phosphate ions can be sorbed on soil by different mechanisms, namely weakly, tightly sorbed and precipitated phosphate species were mentioned. The quantity of phosphate uptaken by plants as well as the optimal fertilization depends on the ratio of these phosphate species. The dissolved in water and weakly sorbed phosphate is considered as utilized by plants. The ratio of tightly sorbed phosphate can be significant, but in slow processes this tightly sorbed phosphate can transform to weakly and soluble species.
The ratio of water soluble/weakly sorbed phosphate can be studied by heterogeneous isotope exchange using P-32 labeled phosphate ions; the quantity of water soluble/weakly sorbed phosphate as well as the transport rate of phosphate in a steady state between the soil and soil solution can be determined. In our previous work1, a correct thermodynamic and mathematical model for the evaluation of the heterogeneous isotope exchange of the radioactive P-32 isotope was constructed and tested.
In this work, the sorption and heterogeneous isotope exchange of phosphate of Hungarian soils is studied and using the correct mathematical interpretation of the processes is applied for the determination of the ratio of water soluble/exchangeable to mineralized/organic phosphate after incubation of the soils with different phosphate quantities and time. In plantation experiments, the uptake of phosphorous as well as the change of the water-soluble/exchangeable phosphate is studied under the effect of plantation.

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NFC 6 / 479

Radiochemical characterisation for a decommissioning purpose at NRG: The case of ion exchange resins

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The High Flux Reactor in Petten, Netherlands, is used for the production of medical isotopes as well as a material research reactor; during its exploitation several types of radioactive wastes are produced and accumulated.

That is the case in particular for the Ion Exchange Resins (IER) in use to treat the water of the primary water circuit of the reactor and the demineralizing basin. Several treatments are in favor to treat spent resins such as steamed reforming (THOR process), cementation, or incineration and cementation of the ashes.

The Nuclear Research and consultancy Group (NRG) is pursuing a politic of footprint reduction and waste volume minimization to achieve enhanced waste management and cost reduction. The treatment of the used resins has therefore been changed and the disposal route switched from cementation towards incineration.

The storage capacity on the Petten nuclear site being limited, the Consultancy & Services team developed in the timeframe of a year an extensive range of characterization techniques, both destructive and non-destructive to meet criteria for transportation and incineration of the resins and the final storage of the ashes.

Treatment and characterization of IER has already been practiced by several operating reactors, however the approach taken by NRG offers a combination of characterization techniques and the integration of some of the complex issues which have to be encountered in the waste management business:

- Representative sampling/Homogeneity
- Nuclides analysis selection
- Homogeneity guarantee
- Analysis method
- Reproducibility

Despite its apparent homogeneity, the presence of hot spots inside an IER container is always a risk; in order to control it recommendations mentioned in IAEA-tecdoc 1537 are applied: *"[⋯]a simple and stable waste stream could be declared homogeneous if NDA measurements of Cs-137 and/or Co-60 made at different locations are within a 30% relative interval."*
3 Sampling campaigns have been carried on batches of used IER from various years. (N.B: IER are changed on average every two years and are stored by years and types, anionic or cationic). Based on the low dose rate of anion exchange resins the sampling could be achieved technically easily. Validation of the homogeneity is carried out by gamma spectrometry using the relative spread of the Co-60 and/or Cs 137 content measured.

The required characterization is foreseen to be provided by the following methodology:

1. Non-Destructive:
2. Gamma spectrometry (Specifically for Co-60 and Cs-137)
3. Alpha spectrometry (under advanced developement)
4. Total alpha/beta via Liquid Scintillation Counter
5. Destructive:
   -Specific beta-emitting nuclide analysis
6. H-3
7. C-14
8. Cl-36 (under advanced development)
9. Fe-55

10. Ni-63
11. Sr-90
12. Tc-99 (under advanced development)

Destructive analysis, elaborated by the Consultancy and Services group, consists as a first step on a complete digestion of the resin to maximize the recovery of radionuclides. The desired radionuclides are chemically isolated by selective separations and extractions and later measured using a Liquid Scintillation Counter.

The key step of the selective separation has been developed at NRG with good recovery rates (in the order of magnitude of 75% for most of the radionuclides) achieving limited losses of activity and better accuracy in the measurement. This selectivity has been checked and validation following ISO -17025 standards is on-going for many radionuclides presented.

Due to the controlled radiochemical process in combination with the very low counting activity of the LSC in use at NRG low sample activities are possible. These developed radiochemical methods are foreseen to be extensively applied to other decommissioned or to be decommissioned components of Nuclear Power Plants as well as for various complex waste streams.

Results (to be updated with latest results at the time of the presentation) show an excellent recovery level for the most important nuclides. More importantly, homogeneity can be assumed within a batch of ion exchange resins.

The results provided so far are fitting the profile of required characterization for both transport and incineration. It also constitutes a "radionuclide identification card", and, as such, can participate to the standardization of the treatment and characterization of such waste streams.

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U(VI) toxicity onto canola cells: Correlation of microcalorimetric data with cell viability and U(VI) speciation

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The transfer of radionuclides into the food chain is of central concern for the safety assessment of both nuclear waste repositories and radioactive contaminated areas, such as legacies of the former uranium mining. The interaction of radionuclides with plants is mostly described by transfer factors without knowing the underlying processes. However, previous studies showed, for instance, a speciation-dependent influence of radionuclide uptake and translocation in plants 1. Heavy metal stress induces the synthesis of metal-binding metabolites, storage of metal chelates in vacuoles or the secretion into the rhizosphere 2, which changes the plant cell metabolism.
We studied the interaction of U(VI) with canola cells (*Brassica napus*) as model system for plants focusing on the concentration-dependent impact of U(VI) on the cell metabolism. The metabolic heat flow of the cells was monitored by isothermal microcalorimetry, a highly sensitive real-time monitor that allows the detection of actinide toxicity in environmentally relevant concentrations. The calorimetric data were compared to the enzymatically determined cell viability. The U(VI) speciation in the cell culture medium was studied by time-resolved laser-induced fluorescence spectroscopy (TRLFS) and thermodynamic modeling to correlate the impact of U(VI) on the cell activity with its speciation 3.
Brassica napus cells showed a temporal decrease in metabolic thermal power and a general reduction of heat production with increasing U(VI) concentration. So far, metabolic calorimetry suffered from the lack of models describing metabolic decline. To overcome this, the model-independent descriptor "metabolic capacity"that allows the evaluation of calorimetric data of declining metabolic phases

was introduced in our work. The obtained normalized "metabolic capacities"and the normalized enzymatically determined cell viabilities showed an almost ideal correlation and were, to a very good approximation, linearly related at U(VI) concentrations up to 200 µM U(VI). The combination with TRLFS and thermodynamic modeling indicated that the cell metabolism was affected predominantly by U(VI) hydroxo species 3.

This presentation will demonstrate the potential of life cell microcalorimetry for radioecological studies, including the discrimination between chemotoxic and radiotoxic effects of uranium at the low dose regime.

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NFC 6 / 652

Carbon-14 release from irradiated stainless steel

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Carbon-14 (half-life 5,730 years) is a key radionuclide in the assessment of the safety of a geological disposal facility (GDF) for radioactive waste. In particular, the radiological impact of gaseous carbon-14 bearing species has been recognised as a potential issue. Carbon 14 is expected to be released from a GDF over a timescale of several thousand years. A number of radioactive gases will be generated from waste materials within a GDF, with carbon 14 bearing methane (14CH4) likely to dominate any carbon 14 transported in a gas phase, potentially reaching the biosphere at low activity concentrations. Sources of carbon-14 include irradiated graphite, irradiated steels and Zircaloys, irradiated reactive metals, spent ion–exchange resins and spent fuel.

The objective of this work is to measure the rate and speciation of carbon-14 release from irradiated stainless steel on leaching under high-pH anaerobic conditions, representative of a cement based near field for intermediate- and some low-level wastes (ILW/LLW). In particular, this includes measurements of releases to the gas phase as well as to solution. The gas phase carbon-14 collection method allows for the discrimination of carbon-14 released as 14CO2, 14CO (and volatile oxidized species) or 14C-hydrocarbons. The carbon-14 solution analysis method used to date has measured the inorganic carbon-14 release only. Work is in progress to measure the total carbon-14 release to solution that includes any dissolved organic carbon-14 species. Three experiments have been in progress in NRG's Hot Cell Laboratory at Petten for a period of 20 months: two contain irradiated stainless steel with similar total inventories of carbon-14 (and also cobalt-60); the third is a control experiment with unirradiated stainless steel from the same batch. The steel samples are being leached in 0.1 mol dm 3 NaOH solution. The three experiments have been sampled six times to date and analytical data are now available for the carbon-14 releases to the gas phase, inorganic carbon-14 to solution and cobalt-60 releases to solution up to 13 months leaching. The experiments are still running and further sampling is planned after 2 years.

The presentation will cover: initial characterisation of irradiated steel samples to assess their suitability for use in the experiments; the approach selected and the conceptual design of the experiments for measuring carbon-14 releases from irradiated steel samples in a hot cell (i.e. a shielded cell); the preparation, installation and commissioning of the equipment in a hot cell. The main part will focus on the presentation and discussion of the results up to 13 months leaching.

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Strontium-90 in Polish cow milk

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Strontium-90 is released to the environment during accidents in nuclear facilities and nuclear explosions. It can penetrate human organisms with food, including milk. Once absorbed into the body, it is accumulated mainly in bones. Studies were conducted in order to evaluate the current levels of ⁹⁰Sr in cow milk after more than 30 years from the Chernobyl catastrophe and calculate the annual effective dose (E) from ⁹⁰Sr. For determination of ⁹⁰Sr, an indirect method based on measurements of ⁹⁰Y (⁹⁰Sr decay product) was used. All measured ⁹⁰Sr activity concentration values were very low, far below the permitted limits in European Union and Eurasian Customs Union. They ranged from 0.016 to 0.048 Bq/L of milk. There were no statistically significant differences between ⁹⁰Sr activity concentrations in milk noted in various parts of Poland. An average annual effective dose due to consumption of cow milk (37 L/year) was calculated to be 0.035 µSv/year. It was only about 0.01‰ of the dose received from natural radiation sources (2.4 mSv/year). In conclusion, domestic cow milk was safe in terms of ⁹⁰Sr contamination.

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Role of animal teeth for estimation of environmental pollution of Sr-90 and Cs-137 just after the Fukushima Daiichi Power Plant Accident

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The Fukushima-Daiichi Nuclear Power Plant (FNPP) accident released a substantial amount of radioactive nuclides into the atmosphere and caused extensive contamination of the environment. Cs-137 and Sr-90 are typical fission nuclides that have a great influence on the environment for several ten years after the accident because of its half-life (30.1 y and 28.9 y). After the deposition onto the environment, the nuclides diffused into soil, plant, and water. Specific activities (ratio of radioactivity to the mass of the element in the sample) are associated with incorporation of the radionuclide into animals and plants. However, the specific activities should have drastically changed in the first a few years. Since the FNPP accident, several years have passed and the landscape has changed. The pollution has been decreased around the FNPP by both decontamination and weathering. Thus, it has become very hard to estimate the initial deposition and the internal and external exposure in the early stage.

Formation mechanisms of teeth are quite unique because replacement of component hardly occurs after the formation stage of the teeth. The present authors reported that the concentration of Sr-90 in teeth formed after the FNPP accident reflects Sr-90 pollution in the environment [1, 2].

In this paper, we propose the alternative way to estimate the initial deposition and past incorporation of the radionuclides into animals. We will discuss past pollution of Sr-90 and Cs-137 estimated from animal teeth.

Cattle were collected near the FNPP (5 km west from the FNPP (10-30 μSv/h) and 16 km southwest from the FNPP (1 μSv/h)). We collected young cattle (teeth were developing at the accident). Radioactivity of Sr-90 was determined with a gas-flow GM detector after chemical separations. Radioactivity of Cs-137 was determined with a Ge detector.

Specific activities of Sr-90 in the teeth were compared with those in soluble fractions of soil. The results suggest that Sr-90 in the soluble fractions of the soil might migrate into teeth and contributed to the amount of Sr-90 in the teeth. Thus, the specific activities of Sr-90 in teeth reflect the extent of Sr-90 pollution in the environment and incorporation in the cattle during the formation stage.

Specific activities of Cs-137 in the teeth were compared with those in internal organs. The results indicate that Cs-137 in the teeth recorded high incorporation until the collection time. Cs-137 in the teeth indicated past incorporation from polluted environment because component in the teeth hardly replace after the formation stage.

It is concluded that Sr-90 and Cs-137 in teeth could be indicators of past pollution in the environment and incorporation into the cattle.

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Assessment of the release behavior of 14C from irradiated nuclear graphite from a German Research Reactor

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Irradiated graphite (i-graphite) is a typical nuclear waste arising from the decommissioning of some nuclear power plants and research reactors. According to the current nuclear waste management strategy in Germany, i-graphite discharged from commercial and research reactors is to be disposed of in a deep geological repository for low and intermediate level radioactive wastes, which requires reliable information on radionuclide inventory and their release behavior under repository relevant conditions. Typically, i-graphite contains a number on radionuclides (activation products), like 3H, 14C or 36Cl, which may potentially be released during storage and disposal. Among them 14C, a weak beta-emitter with a half-life of 5,700 years, is of major concern due to radiation hazards by potential incorporation into the human body. Despite years of research activities on i-graphite, the binding forms of 14C as well as the mechanisms of 14C release from i-graphite and its speciation remain unclear; therefore the focus of the present work is on the understanding of 14C release behavior from i-graphite, during the operational phase of a repository, as well as under the long-term conditions relevant to final disposal.

Outcomes of earlier studies demonstrated that 14C can be present in i-graphite in different chemical forms, like elemental C, CO2 absorbed or trapped in the closed pores or as organic C species. These 14C-species have different mobility in i graphite, which can be affected by a number of factors, e.g.

graphite microstructure, temperature, composition and pH of infiltrating groundwater, promoting the 14C release into the gaseous phase. The behavior of 14C in i-graphite was investigated by means of model leaching tests using i-graphite acquired from the Rossendorf Research Reactor (RFR). The effects of various storage conditions, such as temperature and composition of leaching solution, were investigated. For the quantitative determination of volatile 14C species at relatively low concentrations a development of a customized analytical method was required, which will be discussed in details.

The results of the study demonstrated that the main volatile 14C fraction (> 83%) is in form of CO2; the remaining - 17 %, are comprised of 14CO or/and organic 14C species. Model experiments with i-graphite also revealed almost negligible 14C release into the gaseous phase, when i-graphite is stored under cementitious conditions (i.e. in a basic media with pH>12.9). In this case a dominant fraction of 14C remains in the liquid phase in form of carbonate species, which can be immobilized due to reaction with the hydrated cement phases. Consistent with earlier studies on AVR-graphite, a small fraction of formic acid was detected in the leaching solution. The annual release rate of volatile 14C for RFR-graphite into the gas phase was determined to be <1%/year, which allows RFR-graphite disposal with a maximum permitted container loading in the German repository Konrad. In addition, pros and cons of a thermal pre-treatment of RFR-graphite for a selective separation 14C were evaluated.

The work leading to these results has received funding from the European Union's European Atomic Energy Community's (Euratom) Seventh Framework Programme FP7/2007-2013 under grant agreement no. 604779, the CAST project.

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Study of 210Po in Indian tobacco using Liquid Scintillation Spectrometry

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A fast radioanalytical method has been developed to measure 210Po in tobacco samples. After microwave digestion Polonium is extracted quantitatively from a 1 M hydrochloric acid solution into a solution of tri-octylamine in toluene. The extract was measured by Liquid Scintillation Counting in alpha mode. Caused by the high counting efficiency the counting time per sample can be reduced significantly compared to conventional alpha.spectrometry allowing for high sample throughput. The method was applied to 11 brands of cigarettes, beedis or chewing tobaccos from India. Based on the resulting activity concentrations and typical consumption habits annual and lifetime equivalent doses were calculated. They were found to be ~100-700 µSv/a or ~5-35 mSv, respectively.

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Radiochemical analysis of biological shielding of VVER-440 reactor for deep geological repository

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The decommissioning of nuclear facilities is connected with a huge amount of radioactive waste. The biological shielding of reactor vessel is an example of very heavy and voluminous part of NPP

decommissioning waste. It is mainly composed of concrete which is long term exposed to gamma and neutron radiation.
ÚJV Řež a. s. owns one concrete block of biological shielding from NPP Greifswald's fifth reactor. This reactor was the same type VVER-440 as it is in NPP Dukovany. There is taken the advantage of experimental gamma analysis and destructive analysis of hard to detect nuclides (14C, 36Cl and 41Ca) in this work. The aim of this work is to compare experimental values with calculated ones.
The samples from the concrete block were taken with a special care to prevent release of volatile nuclides. Sampling was performed in 7 distances from 50 mm to 750 mm from the lining edge closer to the reactor. These 7 samples were analysed by Central analytical laboratory in ÚJV Řež, a. s. First of all, the high-resolution gamma spectrometry measurements were done using HPGe detector. Nuclides 60Co, 152Eu and 154Eu were identified in the samples. After gamma measurement, samples were decomposed by alkaline leaching. Content of 14C, 36Cl and 41Ca was analysed from the leachate.
14C was separated by precipitation from the leachate with solution of BaCl2. The precipitate was mixed with scintillation cocktail and measured with LSC Quantulus 1220. 36Cl was separated by extraction chromatography using Cl-resin produced by Eichrome. Small part of the eluate was used to calculate the yield of separation on ICP-MS and the rest was mixed with scintillation cocktail and measured with LSC. Evaluation of 41Ca was quite problematic. There was high content of calcium in the concrete so it was not possible to prepare thin layer sample for X-ray measurement, as a standard detection procedure. It was the reason to modify 41Ca detection from X-ray spectroscopy into LSC measurement. However the standard separation procedure used for X ray gamma spectroscopy was not good enough for LSC measurement even after some modifications. Then another separation procedure had to be used, where the procedure published by Xiaolin Hu (2005) was chosen and fit perfectly for this purpose. Finally, after separation the sample was mixed with scintillation cocktail and measured with LSC.
Obtained results were compared with theoretical values computed by Monte Carlo simulations. The comparison showed quite big differences between experimental and theoretical values. One of the possible explanations is that the differences come from approximations made in theoretical MCNP model based on available inadequate chemical and elemental/isotopic composition of analysed concrete, where missing elements had to be taken from the literature for similar (not the same) concrete. Especially the content of elements Eu and Cl is crucial to know likewise the content of hydrogen which moderates the neutron flux.

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”Dry”decontamination by films and pastes

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This study was aimed to development and application of polymer composite formulations for "dry" decontamination of the spent nuclear fuel storage of mobile NPP "PAMIR"with gas-cooled nuclear reactor using heat carrier N_2O_4.
Polymer composite formulations for "dry"decontamination and their operation characteristics (decontamination efficiency, consumption rate), and at evaluating the chemical resistance and radionuclide fixation strength in the "dry"decontamination waste obtained using the new decontaminating pastes were developed. Polymer composite formulations for decontamination were developed on the basis of a polyvinyl alcohol solution as a binder with active additives (HNO_3, HBF_4, 1-hydroxyethane-1,1-diphosphonic acid and its triammonium salt, and a synthetic detergent) and fillers (natural rottenstone; rottenstone modified with nickel ferrocyanides; clinoptilolite modified with iron(III) and calcium chlorides, sodium phosphate, and potassium ferrocyanide; hydrolytic lignin). The developed polymer composite materials (pastes) exhibit high decontaminating ability ($DF=10^2–10^3$) and low adhesion to the tested surfaces of stainless and carbon steels (including painted surfaces), plastic compound, self-leveling floors, and Teflon surface. The ^{137}Cs leach rates from dry decontamination wastes were determined by long-term leaching test in accordance with GOST (State Standard) 29114–91. Results show that rottenstone modified with nickel ferrocyanide fixes strongly (up to 99 %) cesium radionuclides in the solid waste from decontamination with pastes.
The current work directions deal with implementation of decontaminating gels on decommissioning

of the spent nuclear fuel storage pools, spacing grids of storage pools and further release of decommissioning material and waste from regulatory control.
Total square of decontaminated equipment was 62,4 m^2, decontaminating gel consumption for 2-lauer coating –1,2 kg/m^2. DF = 10^1–6•10^2, formed solid waste was 0,3 kg/m^2.
The developed formulations ensure reliable, efficient, and safe storage of radioactive waste from “dry”decontamination of the external surfaces of the equipment and premises with the new formulations.

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Use of Digital Autoradiography to characterize the effect of acidic leaching on mobilisation of 238U series radionuclides in ISR context.

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This work is based on a comparative analysis of 2 sand samples from the ISR (in situ recovery) experimental site of the Dulaan Uul uranium prospect (Mongolia). Both sample were polished thin-sections. The first sample (raw ore) come from the untreated ore body and the second (leached sand) is from a part of the ore body which was leached during 6 months. Previous chemical analysis 1 indicate U-concentration and U-Ra ratio of 400 ppm and near 1 (secular equilibrium) respectively in the untreated ore sample. U-concentration of 6 ppm and U/Ra ratio of 29 were measured in the sample of leached ore.
Three techniques were used in this work: i) alpha digital autoradiography was used as a mapping tool to locate radioactive regions on the thin-section at micrometric scale, ii) QuemScan chemical mapping was used to obtain a large chemical/mineralogical map of the samples at micrometric scale and iii) a scanning electron microscope coupled with focused ion beam (SEM-FIB) was used to produce transversal nanometric cross sections directly from the thin sections. Alpha autoradiographs (i) were coupled with the elementary chemical maps (ii) to precisely locate radioactive regions of interest. The MEB-FIB (iii) was used to study the mineralogy, petrography and chemistry.
Alpha maps of both samples display high active spots, diffuse radioactive and inactive zones. This heterogeneity illustrates that, for both samples, the alpha activity location is related with the mineralogy and the petrology of the sample even after acidic leaching. Using the autoradiograph/QuemScan maps coupled we observed on the raw ore sample an association between U-rich phases and Fe-Ti minerals, mainly Ilmenite (FeTiO3). Two types of Fe-Ti minerals were identified: the type 1 is composed by strongly altered minerals with dissolution holes sometimes filled by clay minerals. Nanocrystals of uraninite (UO2) coat the dissolution surfaces of Fe-Ti minerals. The type 2 show little trace of alteration and is not associated with clays or uraninite. The same approach was applied on the leached sample. The remaining U was associated in zircon crystals. Hot spots of alpha activity were located in zones containing Fe-Ti minerals and more rarely barite. The barite shows two specific morphologies. The first is dendritic and often associated with clay minerals. The second one shows overgrowth on a needle-shaped core.
These results bring new information about the fate of intermediate radionuclide in ISR conditions. The relationship between the alpha activity and the mineralogy of the leached samples indicate that daughters’elements appear to be immobile during the leaching process. Intermediate elements seem to be still associated with Fe-Ti minerals. Mineralogical observation may also indicate others process like adsorption on clay phases or co-precipitation with barium sulphates. This work uses digital autoradiography as a rapid and powerful tool to localize and to have an overall understanding of the mobility of the alpha radio-isotopes. Coupling with the uranium content is fundamental to locally deduce the radioactive equilibrium state in the thin-sections.

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Tc immobilization on gamma alumina: a study of the reductant presence and absence

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Technetium isotope ⁹⁹Tc is a fission product of environmental concern, due to its high mobility and its elevated lifetime (2.13×10⁵ years). Among their possible oxidation states, Tc(VII) and Tc(IV) are the most stable ones. On one hand, Tc(VII) occurs under oxidizing redox conditions, being pertechnetate (TcO₄⁻) the main species, which is considered an inert and mobile anion that hardly interacts with minerals. On the other hand, Tc(IV) is present under reducing conditions and it is mainly found as solid, TcO₂, since it has a very low solubility product. Studies dealing with Tc immobilization in groundwater normally consider the use of reductants or mineral containing reductant moieties to favor the transformation of Tc(VII) to Tc(IV) with the aim of decreasing its migration in water fluxes.
Nano particular gamma alumina is a well suited sorbent for anions because of the high values of surface to volume ratio, specific surface area and point of zero charge. Indeed, we have already observed the efficient sorption capacity of gamma alumina against Se(IV) anions 1. Thus, per se, alumina is a promising candidate to sorb Tc(VII). Nevertheless, for Tc(VII) low sorption is expected. However, previous studies have reported the higher Tc retention when the mineral is in contact with a reductant [2,3]. Thus, we will study the reduction of Tc(VII) to Tc(IV) on nano particular gamma alumina in presence of Fe²⁺.
This work has been developed in the frame of VESPA II project, supported by the German Ministry of Economy and Energy (BMWi).
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Environmental transfer of radionuclides from phosphogypsum at an uncovered waste disposal site in Portugal

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Phosphogypsum from past production of phosphoric acid was dumped into self-contained ponds on the south bank of Tejo River, at the industrial area of Barreiro near Lisbon, for decades. Production and disposal of phosphogypsum was discontinued in the early 90's due to changes in industry, and a contamination assessment was performed at that time. Since then large amounts of phosphogypsum have remained exposed to weathering while a decision on the management of this material was pending. The waste piles were re visited in 2016 and a survey of radioactivity distribution in the phosphogypsum and in the area was carried out. Determinations of radionuclide concentrations (238U, 235U, 234U, 230Th,226Ra, 210Pb, 210Po, 232Th) were performed by alpha spectrometry in samples of phosphogypsum, plants spontaneously grown on the waste piles and in surroundings, groundwater, and in atmospheric aerosols. Similar analyses were performed also in samples from a comparison site further away and in a similar environment. In general, radionuclide transfer from phosphogypsum to plants was minimal, although radium (226Ra) dissolution from phosphogypsum piles and its percolation into the groundwater was noticeable. Ambient radiation doses and radionuclide concentrations in the terrestrial environment around the piles and at the comparison site are compared, and showed reduced dispersal of radioactive elements in the terrestrial environment. Options for reuse of phosphogypsum and phosphogypsum confinement on site are discussed.

Key words: phosphate industry, radioactivity, waste management, environmental contamination.