



Contribution ID: 184

Type: Poster

## Pu(V) and Pu(IV) oxidation state distribution in natural clay

*Tuesday, May 13, 2014 5:15 PM (1h 30m)*

Environmental behavior of Pu isotopes can be strongly affected by oxidation–reduction reactions which can result in changes of their speciation, solubility and transport. Pu(V) and Pu(IV) sorption to three well characterized samples from the industrial exploitation site Šaltiškiai in North Lithuania was used to study their sorption kinetics and oxidation state distribution in natural clay systems. The composition of clay mineral coatings and their leaching during the sequential extraction procedures were investigated by means of the Mössbauer spectroscopy and X-ray diffraction.

Pu(IV) and Pu(V) were added to achieve their initial concentration of  $3 \cdot 10^{-10}$  mol·L<sup>-1</sup>. Solids were separated by centrifugation at 6000xg after a desired time. Pu activity concentrations were measured by alpha spectrometry. The oxidation state distribution analysis was performed using thenoyltrifluoroacetone (TTA), bis(2-ethylhexyl) hydrogen phosphate (HEDHP) and 1,3-diphenyl-1,3-propanedione (DBM) solvent extraction as well as Pu(III, IV), Pu(V), Pu(VI), Pu(IV) and Pu(IV polymeric) species were separated.

In the natural clay systems the Pu oxidation state distribution analysis in the liquid-phase indicated a comparatively high content (up to 75%) of Pu(III/IV) at pH=5, while polymeric species of Pu(IV) were found to be the dominant species at higher pH. Pu oxidation states in the solid-phase were determined after the desorption step using 3 mol·L<sup>-1</sup> HCl, and Pu(III,IV), Pu(V) and Pu(VI) as well as Pu(IV) at pH~0 were analyzed in parallel. The content of Pu(III) was calculated from mass balance analysis. The obtained results showed that Pu(V) was mainly reduced to Pu(IV), and a small portion of Pu(III) found in the system was explained as a result of reduction by iron-bearing minerals such as montmorillonite and siderite present in the Triassic clay samples.

Experimental data obtained from the laboratory and field observation were used in modeling. The kinetic curves were evaluated by fitting the experimental data with six different types of kinetic models derived for the following six control processes: mass transfer (DM), film diffusion (FD), diffusion in the inert layer (ID), diffusion in the reacted layer (RLD), chemical reaction (CR) and gel diffusion (GD).

Financial support provided by FP7 RECOSY, grant No 212287, the Agency for Science, Innovation and Technology of the Republic of Lithuania (contract No. TAP-36/2010), the Research council of Lithuania (contract No. TAP-54/2010) as well as by the Ministry of Education of the Czech Republic (contract No. MSM 6840770020) is acknowledged.

**Primary author:** Dr LUJANIENE, Galina (SRI Center for Physical Sciences and Technology, Savanorių ave 231, Vilnius, LT-02300, Lithuania)

**Co-authors:** Dr VOPÁLKA, Dušan (CTU in Prague, Department of Nuclear Chemistry, Prague 1, Brehova 7, Czech Republic); Ms KULAKAUSKAITĖ, Ieva (SRI Center for Physical Sciences and Technology, Savanorių ave 231, Vilnius, LT-02300, Lithuania); Dr ŠTAMBERG, Karel (CTU in Prague, Department of Nuclear Chemistry, Prague 1, Brehova 7, Czech Republic); Prof. JUŠKĖNAS, Remigijus (SRI Center for Physical Sciences and Technology, Savanorių ave 231, Vilnius, LT-02300, Lithuania)

**Presenter:** Dr LUJANIENE, Galina (SRI Center for Physical Sciences and Technology, Savanorių ave 231, Vilnius, LT-02300, Lithuania)

**Session Classification:** Poster Session - Radionuclides in the Environment, Radioecology

**Track Classification:** Radionuclides in the Environment, Radioecology