



Contribution ID: 182

Type: **Invited**

Study of ^{137}Cs , ^{241}Am and Pu isotopes sorption behavior in the environment: Prague – Vilnius collaboration*

Tuesday, 13 May 2014 11:20 (30 minutes)

Results of long-term collaboration with the Czech Technical University in Prague are reported. Sorption behavior of ^{137}Cs , ^{241}Am and Pu isotopes was studied with the aim of better understanding their migration mechanisms in the Triassic clay selected for engineered barrier of the near surface low and intermediate level radioactive waste repository, as well in soil and bottom sediments from the natural environment.

The Baltic Sea bottom sediments and well characterized clay samples with different amounts and compositions of iron oxides as well as synthetic magnetite, goethite and hematite were used in equilibrium and kinetic sorption experiments. Sorption was studied as a function of pH, a composition of solutions, and a contact time. The character of Cs, Pu and Am bonding was analyzed by sequential extraction (SE). Solvent extraction techniques, ultrafiltration and alpha spectrometry as well as ICP-MS were employed to characterize the oxidation states of the formed plutonium species. Two Chernobyl soil cores were also analyzed on the vertical distribution of radionuclides, their bonding to soil components, as well as on Pu oxidation state distribution. Experimental data obtained from the laboratory and field observations were used in modeling.

Despite their similar sorption kinetics, Pu(IV), Pu(V) and Am(III) showed different bonding to soil, clay and bottom sediment coatings. Pu was predominantly associated with amorphous Fe-oxides and natural organic matter sites, whereas in the case of Am(III), the exchangeable and carbonate sites played the principal role. It has been shown that Pu(V) sorption mechanism includes a very fast Pu(V) reduction (reaction rate $\leq 2.33 \cdot 10^{-3} \text{ s}^{-1}$) to Pu(IV) and partly to Pu(III). Following reduction Pu isotopes were bound to various components of bottom sediments via ion exchange and surface complexation reactions and a slow incorporation into the crystalline structure of Fe minerals. Kinetics experiments showed that the sorption of Cs(I), Pu(V), Pu(IV) and Am(III) to bottom sediments from natural seawater was controlled by the inert layer diffusion process.

The analyses of data obtained from sorption experiments, field observations and speciation of radionuclides have indicated a slightly higher migration potential for Am both in the Chernobyl soil and the natural clay minerals. The exchangeable and carbonate bound Am(III) should display faster migration rate and greater bioavailability in the environment.

*) Dedicated to the memory of Prof. Petr Beneš

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Session Classification: Radionuclides in the Environment, Radioecology 1

Track Classification: Radionuclides in the Environment, Radioecology