Sensitive Plutonium Determination for Migration Studies in a Granitic Matrix

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It is expected that plutonium introduced into the environment exists in the 4^+-state and thus is immobile due to its low solubility in groundwater and strong sorption onto rocks. But mobile colloids - suspended particles in the sub-micrometer size range onto which plutonium is sorbed - can enhance the transport of plutonium in groundwater. This has been observed at the Nevada test site, USA, where quite a number of underground nuclear tests were carried out. The $^{240}Pu/^{239}Pu$ isotope ratio found in ground water samples 1.3 km north of the underground nuclear test site indicates that the plutonium in the water stems from the test site. The authors of [1] argue that colloidal groundwater migration must have played an important role in transporting the plutonium.

Bentonite, which is used to seal nuclear repositories, can produce colloids in contact with water. If fractures would occur in the bentonite shielding, plutonium sorbed on bentonite colloids could enter the far field, i.e., granite, and furthermore the groundwater system. In collaboration with the Forschungszentrum Karlsruhe (FZK) migration studies of plutonium in a granitic matrix (Grimsel field laboratory) with and without bentonite colloids have been performed.

The experiments at the Grimsel field laboratory have been done by H. Geckeis et al., FZK. Plutonium-242 tracer (without bentonite colloids) or plutonium-244 tracer (with 20 mg/l bentonite colloids) have been introduced into the water flow going through the fractures of granitic rock, simulating the behaviour of a granitic far field. After migration through the rock, samples have been collected at different retention times. The plutonium concentration has been measured by ICP-MS at FZK and - for lower concentrations - with RIMS [2] in Mainz, down to the detection limit of 10^7 atoms.

Figure 1 shows the tracer breakthrough curves for Pu-242 and Pu-244. The normalised concentration of plutonium in the samples is plotted versus the retention time.

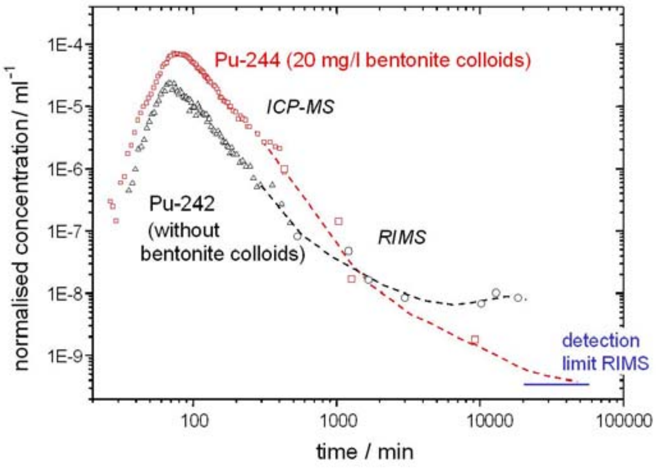

Figure 1: Tracer breakthrough curves of plutonium in a granitic matrix without (black points) and with (red points) bentonite colloids.

The maximum of the tracer breakthrough curve with bentonite colloids (red points) is higher than the one without colloids (black points), indicating that more plutonium is transported in a granitic matrix with colloids than without such colloids. Without colloids, a plateau at later retention times is observed with a slight increase of the concentration, indicating the release of retarded plutonium previously absorbed on the surface of the rock whereas with bentonite colloids, a permanent decrease of the normalised plutonium concentration down to the detection limit of RIMS has been found.

Literature:

[1] Kersting, A. B. et al. (1999): Migration of plutonium in ground water at the Nevada Test Site. Nature. 397. S. 56 - 59.

[2] Grüning, C. (2001): Spektroskopie und Ultraspurenanalyse von Plutonium mittels Resonanzionisations-Massenspektrometrie. Doktorarbeit. Universität Mainz.