

Speciation change of Pu(IV) in granite underwater with high salinity

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Granite is chosen as a candidate back rock for disposal of highly radioactive waste in China, and physicochemical behaviour of radioactive nuclides in granite underwater is a key factor for the safety assessment of the depository. ^{239}Pu is one of the most important long-lived radioactive nuclides in the waste with high chemical and radiological toxicity.

The chemistry of Pu in aqueous solution is very complicated because it may occur in different speciation, including colloids and polymers besides soluble Pu ions in different valence. Many works have reported the enhanced migration of Pu colloid in field survey as well as in laboratory experiments, but the speciation change of Pu after entering the granite underwater is not yet clear. This work tries to understand the speciation distribution evolution when Pu(IV) is introduced into a granite underwater with high salinity collected from a granite area in northwest China. It is shown that about 90% of added Pu transformed into colloids (either as Pu polymer or as pseudo-colloids by adsorbed on particles in water) after one year and most of them precipitated or adsorbed by container walls. For soluble Pu, the valence distribution is investigated by sequential solvent extraction to be Pu(IV) (71%) > Pu(V) (24%) > Pu(VI) (5%). For colloidal Pu, the a modified Tessier Sequential Extraction procedure was employed to give a Pu speciation in particle as carbonate (57.3%) > Organic (21.7%) > Reducible (11.2%) > Residual (5.1%) > exchangeable (4.7%). Also the study demonstrates that shaking and air introduction have great impact on Pu speciation and thus on Pu migration in underwater. Other observations about Pu colloid in our laboratory will be reviewed.