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Experiments to demonstrate chemical containment: Solubility under the cementitious conditions of a repository in the UK

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One option for the long-term management of intermediate level radioactive wastes is deep geological disposal. The waste is grouted with cement in steel canisters for disposal; after closure, the repository may be backfilled with more cement. The cement porewater is expected to be of high pH due to cement dissolution and low Eh due to canister corrosion. This work describes a series of long term experiments to demonstrate precipitation and solubility limitation under these conditions.

The experiments are expected to last 4 years and interim results are presented here. The elements included are Cs, Ni, Eu, U and Th, I as iodide and Se, as SeO32-. Four solutions are being tested, NaOH, saturated Ca(OH)2, NRVB (a limestone-rich backfill) -equilibrated water and cellulose degradation products (CDP) in NRVB. Additional experiments in CDP are being carried out in the presence of reducing agents (metallic Fe and Na2S2O4). Solubility is assessed from both under- and oversaturation directions.

After one year of periodic sampling, steady state has been reached for U and Ni. Ca(OH)2 is a good surrogate for the behaviour of radionuclides in NRVB, showing similar results to the reference backfill. CDP increase the solubility of Ni and U by several orders of magnitude, whereas Th and Eu are less affected. The solubility of Se is clearly conditioned by the availability of Ca in solution, and there is no evidence of a decrease in the concentration of Cs or I by co-precipitation. XRD analyses of precipitated solids show poorly crystalline phases in the oversaturation experiments. Only incipient crystallinity was observed for undersaturation, which could explain the differences observed in measured solubility between the two sets of experiments.

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