Coupling Source Term, Mineral Reactivity and Flow in Radionuclide Transport

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Abstract

The focus of this work is to investigate the dissolution of MW25, a non-radioactive simulant of UK high-level nuclear waste borosilicate glass, and to predict its performance in the near field of a geological repository. A single-pass flow-through (SPFT) experimental system was used to measure the forward dissolution rates of MW25. Experiments were conducted in two parts, resulting in a total of 432 experiments. Experiment Part 1 considers the dissolution of the waste glass in deionised water at 40 and 90 ^{o}C and circum-neutral pH. Experiment Part 2 considers the dissolution of the waste glass in simulant groundwaters, with similar compositions to groundwaters of Callovo-Oxfordian clay (lower-strength sedimentary rock (LSSR)) and Borrowdale Volcanic Group rocks (higher-strength rock (HSR)), at 40 ^{o}C and pH 7. Experiments in both parts were conducted at flow rates ranging from ~10 mL d^{-1} to 250 mL d^{-1}. The wide range in flow rates, together with adjustments to the sample surface area, were used to produce a range of ratios of flow rate to surface area (q/S) values.

The forward dissolution rate measured in deionised water was found to be approximately one order of magnitude higher at 90 ^{o}C than at 40 ^{o}C. A similar release was observed for Si, Mg and Al at 40 ^{o}C and 90 ^{o}C, whereas the B, Cs, Na, Li and Mo showed an order of magnitude increase when the temperature was increased from 40 to 90 ^{o}C for low q/S values. The temperature dependence of the dissolution rates was determined using the Arrhenius rate law,

and the resultant activation energy (E_a) shows that the dissolution process is a surface phenomenon. At 90 ^{o}C the net effect of the processes governing MW25 dissolution led to the preferential release of boron and alkali metals relative to the release of Si during the transient dissolution stage, accompanied by an increase in the concentration of silicic acid. This suggests that the solution activity of silicic acid at a higher temperature has a weak influence on the release of the mobile elements.

The forward dissolution rate measured in LSSR simulant groundwater was found to be slightly higher than that measured in HSR simulant groundwater. The dissolution behaviour of MW25 in both groundwaters is consistent with its behaviour in deionised water at 40 ^{o}C, with the dissolution rates of elements increasing as q/S values were increased. However, forward dissolution rates measured in the simulant groundwaters were lower than the forward dissolution rates measured in deionised water under these experimental conditions. This is attributable to the interaction of the components of the simulant groundwaters with the glass, as revealed by post-reaction surface analyses, and a consequential lower alkalinity of the leachates collected in the experiments with simulant groundwater than in deionised water.

Reactive chemical transport simulations of waste glass dissolution and radionuclide release in a hypothetical near field were conducted over a time span of a million years with GoldSim. Model parameterisation was accomplished by coupling direct laboratory measurements and literature data with transport processes. Time and spatial dependence of radionuclide flux on waste glass were evaluated under four different scenarios. The results showed that Scenario 4, which assumes enclosing the waste glass in a steel canister covered by a copper canister and emplacing the waste package in a granite host rock, is optimal for the long-term isolation of the radionuclides. The waste glass was found to play a significant role in the overall performance of the near field.

This study features a new method for estimating the surface area of reacted glass powder more accurately than the geometric surface area estimate, which is the preferred standard method among researchers.