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Modelling Iron-Bentonite Interactions

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The presence of both iron canisters and bentonitic clay in some engineered barrier system (EBS) designs for the geological disposal of high-level radioactive wastes creates the potential for chemical interactions which may impact upon the long-term performance of the clay as a barrier to radionuclide migration. Flooding of potential radionuclide sorption sites on the clay by ferrous ions and conversion of clay to non-swelling sheet silicates (e.g. berthierine) are two possible outcomes deleterious to long-term performance.

Laboratory experimental studies of the corrosion of iron in clay show that corrosion product layers are generally thin ($< 1\ \mu$m) with magnetite, siderite, or ‘green rust’ occurring depending upon temperature and ambient partial pressure of carbon dioxide. In theory, incorporation of iron into clay alteration products could act as a ‘pump’ to accelerate corrosion. However, the results of laboratory experiments to characterise the products of iron-bentonite interaction are less than unequivocal. The type and amounts of solid products appear to be strong functions of time, temperature, water/clay ratio, and clay and pore fluid compositions. For example, the products of high temperature experiments (> 250 °C) are dominated by chlorite, whereas lower temperatures produce berthierine, odinite, cronstedtite, or Fe-rich smectite. Unfortunately, the inevitable short-term nature of laboratory experimental studies introduces issues of metastability and kinetics. The sequential formation in time of minerals in natural systems often produces the formation of phases not predicted by equilibrium thermodynamics.

Evidence from analogous natural systems suggests that the sequence of alteration of clay by Fe-rich fluids will proceed via an Ostwald step sequence. The computer code, QPAC, has been modified to incorporate processes of nucleation, growth, precursor cannibalisation, and Ostwald ripening to address the issues of the slow growth of bentonite alteration products. This, together with inclusion of processes of iron corrosion and diffusion, has enabled investigation of a representative model of the alteration of bentonite in a typical EBS environment. Simulations with fixed mineral surface areas show that berthierine dominates the solid product assemblage, with siderite replacing it at simulation times greater than 10 000 years. Simulations with time-dependent mineral surface areas show a sequence of solid alteration products, described by: magnetite -> cronstedtite -> berthierine -> chlorite. Using plausible estimates of mineral-fluid interfacial free energies, chlorite growth is not achieved until 5 000 years of simulation time. The results of this modelling work suggest that greater emphasis should be placed upon methods to up-scale the results of laboratory experiments to timescales of relevance to performance assessment.