Geochemical optimisation of EBS design for HLW

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Prefabricated EBS design

Recent work has shown that prefabricated engineered barrier system (EBS) designs for high-level radioactive waste (HLW) repositories may greatly improve operational practicality and quality management. Prefabrication also allows multi-component EBS designs to be considered which aim to optimise performance (e.g. Fig. 1).

Figure 1: An example of a prefabricated EBS design, the Multi-Component Module (MCM) (McKinley et al., 2000)

Geochemical aspects

Taking the example of disposal of vitrified HLW, the problem of optimisation is discussed from the point of view of geochemistry, in particular balancing the advantages of additional barrier components against the disadvantages of the requirement to determine their long term evolution and, especially, reactions at interfaces.

References


The reductive immobilization of pertechnetate by bioreduced sediments

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Sediments were incubated with Shewanella putrefaciens for varying periods, then washed and pasteurized, and contacted with TcO₄⁻ at a concentration of 20µM. In Hanford sediment (1.3 wt.% Mn) only about 1% of the total Fe (7.8 wt.%) was reduced after 50 d incubation, and extractable Fe(II) appeared after 21 d. In Oak Ridge sediment (0.23 wt.% Mn), 17.8% of the total Fe (4.6 wt.%) was reduced, and Fe(II) appeared in extractions after 10 d. 72% of the Hanford Mn, and all of the Oak Ridge Mn, was bioreduced. Reduction of Tc(VII) was dependent on the incubation time, the extent of Mn(III/IV) reduction, and the amount of Fe(II) formed. Mn(III/IV) oxides buffered the sediment redox potential, and inhibited Tc(VII) reduction. Bulk XANES analysis indicated that Mn(III/IV) oxide minerals were transformed to rhodocrosite (MnCO₃) plus minor Mn oxide. Elemental X-ray microprobe abundance maps for Tc, Mn, and Fe were compared to (SEM) backscattered electron images. Discrete MnCO₃ was absent, and individual micro-XANES spanned the range from Mn(III/IV) to Mn(II) even in the most reduced Ringold sediments. The bioreduction of these oxides was spatially heterogeneous, yielding microscopic regions with variable redox chemistry.

In Ringold sediment, Tc(IV) was associated with Fe-bearing phases, particularly biotites. The biotites were weathered and included abundant interlaminar Fe and Mn oxides. The formation of Tc(IV) solids on Fe-bearing minerals suggested the reduction of Tc(VII) by bioreduced Fe(II), and Mossbauer spectroscopy showed an increase in Fe(II) associated with silicates after bioreduction; relatively small amounts of sorbed Fe(II) were apparently reactive with Tc(VII)O₄⁻. In Oak Ridge sediment, Tc(IV) was disseminated within and associated with one of two morphotypes of weathered shale.