Functional engineered barriers for geological repositories: A mineralogical approach to improved radionuclide sorption

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The UK multi-barrier design concept for the geological disposal of intermediate level nuclear waste combines reducing conditions from the corrosion of steel canisters with high pH from a cement backfill; the aim is to limit the solubility and mobility of radionuclide species within a geological disposal facility when primary containment fails.

We here investigate the feasibility of developing an extra barrier to the release of radionuclides, by application of cement backfill with the addition of redox-active components containing either blast furnace slag (BFS) or other sulfide minerals. Compared to cement backfill with no addition, we show that these 'Functional Engineered Barriers' are capable of increasing the sorption radionuclides, such as ^{99}Tc, by up to 7 orders of magnitude. Linear combination fitting of μ-XANES data showed that significant reduction of Tc(VII) to insoluble Tc(IV) occurred for BFS- (~40%) and FeS_2-NRVB (~100%) blends, respectively. This is supported by batch sorption data and gamma camera transport experiments that show a significant reduction in diffusive transport. Micro XRF mapping indicated that anionic radionuclide immobilisation occurred at the surfaces of BFS and sulfide mineral particles. Rietveld refinement of high resolution XRD data, collected on the world's first long duration beamline at the Diamond Light Source over a 10 month period, gives insights into the formation of secondary hydration products in these cement blends, which may be responsible for the sorption of anionic radionuclides at the surface of additive particles.