Incorporation of ^{90}Sr into alkaline altered sediments

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The use of cementitious materials is ubiquitous at nuclear facilities and in radioactive waste packaging. Water in contact with fresh cement produces a highly alkaline solution dominated by K and Na hydroxides causing localised areas of high pH groundwater at the concrete/soil interface. Silicate minerals within sediments and clay barrier materials are known to alter to zeolite and feldspathoid phases under these conditions. The effect of alkaline pore fluid induced weathering reactions on the mobility of radionuclides is not well understood, especially with respect to potentially highly soluble fission products such as ^{90}Sr (as Sr^{2+}).

Here we used 20 g.L^{-1} batch experiments containing sediments representative of the UK Sellafield nuclear site in a pH 13.5 high ionic strength cement-based leachate to investigate Sr^{2+} sorption as a function of time. Experiments initially contained 20 ppm Sr^{2+} (spiked with 30 Bq.mL^{-1} ^{90}Sr tracer) and were sampled from 2 days up to one year (one experiment was aged for one year at 70°C). Change in Sr speciation within sediments was determined using X-ray absorption spectroscopy and sequential extraction techniques.

At 2 days 72.6±8.6% ^{90}Sr was removed from solution, which rose to 93.8±3.3% at 10 days but then decreased to 81.8% after a year. In the 70°C aged sample sorption increased to 98.0±2.5%. This suggests that initial alteration enhances Sr sorption but further recrystallisation releases some sorbed Sr. Sequential extractions show that the majority (65-75%) of ^{90}Sr remains in the $MgCl_2$ exchangeable fraction even after one year. In the 70°C aged sample, 25.2±5.8% ^{90}Sr was found to be residual. EXAFS analysis revealed two Sr-O-Si(Al) bond distances at 3.69 and 3.84 in a 10 day sample and at 3.57 and 3.83 Å in a one year sample, consistent with weak Sr sorption to aluminosilicate phases. EXAFS spectra from a 70°C aged sample contain evidence for a single Sr-O-Si(Al) bond distance at 3.45 Å consistent with Sr incorporation in a neoformed feldspathoid phase such as cancrinite. These results indicate that alkaline altered sediments could be a sink for ^{90}Sr in the environment, however even after alteration ^{90}Sr may remain exchangeable with other ions in solution.

Reactive transport modelling to quantify arsenic mobilization and capture during aquifer storage and recovery of potable water

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Aquifer storage and recovery (ASR) is an artificial recharge technique which is increasingly used as a water management tool to augment depleted groundwater resources. ASR is a critical component of the long-term water supply plan in various regions, including Florida and Australia. However, under particular, site-specific conditions the viability of ASR as a safe and cost-effective water resource may be impacted by elevated arsenic concentrations that are detected during recovery of the injectant. This study describes a conceptual and process-based reactive transport model of the coupled physical and geochemical mechanisms controlling the fate of arsenic during ASR. The conceptual/numerical model assumes that (i) arsenic is initially released following pyrite oxidation triggered by the injection of oxygenated water (ii) then largely complexed to neo-formed hydrous ferric oxides before (iii) being released again during recovery as a result of both dissolution of hydrous ferric oxides and displacement from sorption sites by competing anions. Multi-cycle hydrochemical data from an affected site where oxic, potable water was injected into a reducing pyrite-containing storage zone were used to evaluate the model. For this site a detailed assessment of the partitioning of arsenic among mineral phases, surface complexes and aqueous phases during injection, storage and recovery is given, together with an evaluation of temporal and areal extent of arsenic mobilization and capture.