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Diffusion of Selenium in Crystalline Rock

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In many countries, high-level radioactive waste is planned to be disposed of in deep-lying crystalline rock. The role of the geosphere as a safety barrier is one of the most important issues in repository performance assessment. Repository safety evaluation today requires going from the laboratory and surface-based field work to the underground repository level. The diffusion and sorption of radionuclides have been studied extensively in the laboratory, but only a few long-term in situ experiments have been carried out. In situ experiments are time consuming and cost intensive, and it is commonly accepted that laboratory scale tests are well-established approaches to characterizing the properties of geological media. In order to assess the relevance of laboratory experiments, the Swiss National Cooperative for Disposal of Radioactive Waste (Nagra) have been conducting extensive in situ experiments at the Grimsel Test Site (GTS) in order to study field scale radionuclide transport and retention. Two Long Term Diffusion (LTD) experiments have been performed since summer 2007. Amongst the elements used in these experiments is nonradioactive Selenium. It is used as an analog for the radiotoxic isotope Se-79, which is ubiquitous in nuclear waste.

In the Radiochemistry Laboratory, University of Helsinki, two laboratory scale selenium diffusion experiments have been carried out to support one of the above mentioned long term in situ experiments. The selenium through diffusion experiments are conducted in two rock blocks; Kuru grey granite and Grimsel granodiorite. Changes of the selenium concentration in the inlet and observation holes are followed using ICP-MS technology. Experiments are conducted under oxic conditions. Parallel to the diffusion experiments, selenium sorption onto Grimsel granodiorite and Kuru grey granite was studied with batch experiments and geochemical modelling. Sorption was studied as a function of grain size, pH, Se concentration and pE. Apparent diffusion coefficients (Da) of selenium were determined using a Time-Domain Diffusion (TDD) modelling tool. Da and distribution coefficients (Kd), acquired from batch sorption experiments, were used to calculate the effective diffusion coefficients (De).

Selenium Kd was higher in Kuru grey granite than in Grimsel granodiorite. Kd was also higher under anoxic conditions than under oxic conditions. Diffusion coefficients were higher for Grimsel granodiorite (De = 3×10-12 m2/s) than for Kuru grey granite (De = 2×10-13 m2/s). Selenium probably occurred as selenium (IV) oxide in the Kuru rock matrix judging by the higher Kd and the slower diffusion observed in the Kuru grey granite.

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