

Uranium sorption to multi-mineral systems associated with radioactive waste disposal

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The establishment of a radioactive waste facility in the deep subsurface involves proving that natural barriers can prevent the escape of actinides to the surface via fractured rocks. Since much of the waste will be stored encased in cement, the dissolution of the cement will leave the near-field pore waters at pH values above 10.5 for several hundred thousand years. Much has been done to understand the mobility of uranium in neutral to low pH environments, but little has been done to understand the complexity of sorption to multi mineral systems in the high pH environments associated with radioactive waste disposal.

To this end we investigated U sorption on the Borrowdale Volcanic Group (BVG) and St. Bees Sandstone (SS) in order to mimic the crystalline basement rock with sedimentary overburden model for waste disposal, which is a model put forth as an option for waste disposal in the United Kingdom. In addition, adsorption experiments using a single mineral, quartz - a common rock forming and fracture sealing mineral, were used to calibrate our experimental set up.

Batch adsorption experiments were conducted as a function of pH and uranium concentration, with and without the addition of sodium bicarbonate. Various uranyl aqueous species are formed at different U concentrations. To understand whether these different species can influence the adsorption of U we varied the concentration of U in our adsorption experiments from 10 ppb up to 10ppm. We found that the percentage of U sorbed to the geomaterials did not depend on the concentration of U added. This indicates that the sorption of U to the geomaterials goes against the electrostatic gradient.

Bicarbonate was added to the system as it is a common component of groundwaters. While the U species in solution changes significantly as a function of U concentration when no bicarbonate is added, the addition of bicarbonate produces different U species that do not change as a function of U concentration. The effect of bicarbonate on the adsorption of U was to lower the percent of sorbed U onto all single and multi-mineral systems when pH is greater than 7. The predominant U species in that pH range is $\text{UO}_2(\text{CO}_3)_3^{4-}$. While the sorption of U in bicarbonate free systems was able to overcome the electrostatic gradient, the electrostatics governed the adsorption in the bicarbonate system.