

The effect of high pH and ionic strength on the adsorption of uranium to quartz, sandstone, and volcanic rock

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One possible environment for a nuclear waste repository in the UK is igneous basement rock that is overlain by sedimentary overburden. Intermediate level waste (ILW) will be deposited within steel canisters filled with cement. The steel of the canisters is expected to last at least 10,000 years, after which surrounding groundwaters would have the ability to infiltrate the cemented ILW. Since it is unlikely that a nuclear waste repository would be placed into a fresh water aquifer, it is likely that the fluids surrounding the waste canisters would be briny. The resulting porewaters would be of pH's in excess of pH 10 due to the dissolution of cement and high in ionic strength, therefore, it is important to understand what will happen to U under those conditions.

Speciation calculations suggest that at the high pH values of 8-12 a $\text{Na}_2\text{U}_2\text{O}_7$ crystalline species is predominant but as pH increases over 12 the predominant species is an aqueous $\text{UO}_2(\text{OH})_4^{2-}$ species which may be more mobile. When 2mM NaHCO_3 is introduced into the system a $\text{UO}_2(\text{CO}_3)_3^{4-}$ species becomes prevalent from pH 8-11 with the crystalline species only predominant from 11-12.2 followed by the uranyl hydroxide at pH values higher than 12.2. Therefore the presence of bicarbonate may be able to change under what conditions and how U may adsorb onto rock forming minerals.

In order to test U speciation under those conditions, we examine the effect of high pH and bicarbonate concentration on the adsorption of U to quartz, sandstone, and volcanic rock as a function of time, pH, and U concentration. We will also use spectroscopic techniques to confirm the mechanism of U adsorption under these conditions. Preliminary data suggests that bicarbonate has no effect on the adsorption of 10ppb U to quartz from pH values of 2-7, then hinders adsorption by up to 30% from pH 8-10. However, above pH 12 60% of U adsorbs to the quartz, regardless of bicarbonate concentration. Therefore, bicarbonate could mobilize U if pH values are lower than 12, whereas above 12 the amount of bicarbonate has little effect.