

Effect of Organics on Radionuclide Partitioning in Nuclear Fuel Storage Ponds

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Abstract : Sellafield has a number of fuel storage ponds, some of which have been open to the air for a number of decades. This has caused corrosion of the fuel resulting in a release of some activity into solution, reduced water clarity, and accumulation of sludge at the bottom of the pond consisting of brucite ($Mg(OH)_2$) and other uranium corrosion products. Both of these phases are also present as colloidal material. ^{90}Sr and ^{137}Cs are known to constitute a small volume of the radionuclides present in the pond, but a large fraction of the activity, thus they are most at risk of challenging effluent discharge limits. Organic molecules are known to be present also, due to the ponds being open to the air, with occasional algal blooms restricting visibility further. The contents of the pond need to be retrieved and safely stored, but dealing with such a complex, undefined inventory poses a unique challenge. This work aims to determine and understand the sorption-desorption interactions of ^{90}Sr and ^{137}Cs to brucite and uranium phases, with and without the presence of organic molecules from chemical degradation and bio-organisms. The influence of organics on these interactions has not been widely studied. Partitioning of these radionuclides and organic molecules has been determined through LSC, ICP-AES/MS, and UV-vis spectrophotometry coupled with ultrafiltration in both binary and ternary systems. Further detailed analysis into the surface and bonding environment of these components is being investigated through XAS techniques and PHREEQC modelling. Experiments were conducted in CO_2 -free or N_2 atmosphere across a high pH range in order to best simulate conditions in the pond. Humic acid used in brucite systems demonstrated strong competition against ^{90}Sr for the brucite surface regardless of the order of addition of components. Variance of pH did have a small effect, however this range (10.5-11.5) is close to the pH_{pzc} of brucite, causing the surface to buffer the solution pH towards that value over the course of the experiment. Sorption of ^{90}Sr to UO_2 obeyed Ho's rate equation and demonstrated a slow second-order reaction with respect to the sharing of valence electrons from the strontium atom, with the initial rate clearly dependent on pH, with the equilibrium concentration calculated at close to 100% sorption. There was no influence of humic acid seen when introduced to these systems. Sorption of ^{137}Cs to UO_3 was significant, with more than 95% sorbed in just over 24 hours. Again, humic acid showed no influence when introduced into this system. Both brucite and uranium based systems will be studied with the incorporation of cyanobacterial cultures harvested at different stages of growth. Investigation of these systems provides insight into, and understanding of, the effect of organics on radionuclide partitioning to brucite and uranium phases at high pH. The majority of sorption-desorption work for radionuclides has been conducted at neutral to acidic pH values, and mostly without organics. These studies are particularly important for the characterisation of legacy wastes at Sellafield, with a view to their safe retrieval and storage.

Keywords : caesium, legacy wastes, organics, sorption-desorption, strontium, uranium

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