

Colloidal particulates in spent nuclear fuel storage: from fundamental properties to effluent treatment

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Colloids have the potential to mobilise radionuclides in radioactive waste decommissioning and disposal scenarios including the legacy ponds and silos complex at the Sellafield nuclear site, UK.[1] Here, we present results from investigations into U colloids under alkaline conditions using a multi-technique approach. Experimental results include both laboratory analysis of the fundamental colloidal and structural properties in U(IV) and U(VI) systems, and characterisation of authentic samples taken from effluent treatment facilities on the Sellafield site.

Silicate was seen to have a significant role in stabilisation of colloidal U(IV), in both core-shell intrinsic U(IV)-silicate particles[2] and colloidal suspension of nano-UO₂ formed via corrosion of metallic uranium[3]. In both cases, silicate enrichment on the particle surface increased colloidal stability via a silicate-rich shell in intrinsic U(IV)-silicate particles, an a silicate coating of nano-UO₂. Experiments probing interactions of U(VI) aqueous species and U(IV) nanoparticles with hydrotalcite (Mg₆Al₂CO₃(OH)₁₆·4H₂O), a key corrosion product of Mg metal cladding of Magnox fuel, illustrated strong interactions between U and hydrotalcite, characterised by EXAFS, TEM and TRLFS, suggesting hydrotalcite and other Mg-based colloids may be a significant vector for U[4].

Finally, authentic samples from a Sellafield effluent treatment facility treating effluents from the legacy ponds and silos were characterised. These samples contained stable colloidal U and Mg phases and radionuclides which were analysed by ultrafiltration, TEM and EXAFS for the first time, with results suggesting U was present as both nano-UO₂ and U(VI)-carbonate sorption complexes. Overall, these results illustrate the close, complex relationship that U and Mg phases have in spent nuclear fuel storage. The laboratory studies closely mirror the authentic samples, and, in particular, illustrate the capability of hydrotalcite colloids to mobilise nanoparticulate U(IV) phases – a new type of ‘pseudo-colloid’. This understanding is also of importance for long-term geological disposal of radioactive wastes where alkaline, reducing conditions are also expected.

References

[1] Z. Maher, *et al.* (2016), *J. Nucl. Mater.* 468, 84 [2] T. Neill, *et al.* (2018), *Environ. Sci. Technol.* 52, 16, 9118–9127 [3] T. Neill, *et al.* (2019), *J. Nucl. Mater.* 526, 151751 [4] C. Foster, *et al.* (2022), *Langmuir* 38, 2576-2589.