Iron oxyhydroxide formation during base hydrolysis of iron(III) nitrate solution: Application to radioactive effluent treatment

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The formation of iron (oxyhydr)oxide (FeOx) nanoparticles is a key process in many industrial effluent treatment processes and natural systems. The high adsorptive capacity of FeOx, and their ability to co-precipitate aqueous ions makes them effective industrial decontaminants of radionuclides. The Enhanced Actinide Removal Plant (EARP) at Sellafield, UK is one such facility where radionuclides are removed from radioactive effluent streams by inducing FeOx precipitation. In order to better understand and inform this crucial radioactive effluent treatment process, this study aims to elucidate the fundamental process of FeOx nucleation growth and aggregation during base hydrolyis of an acidic iron (III) nitrate solution.

FeOx particle formation during the EARP treatment process was replicated in the lab using an automated small volume reactor. Here, the pH of a 1 M nitric acid ferric nitrate solution (400 ppm Fe) was raised to pH 9 by the addition of NaOH over approximately 45 mins. Experiments were further performed with sulfate, phosphate and borate present to investigate the effect of these effluent constituents on particle formation. To characterise these processes, *in situ* and time resolved Small Angle X-ray Scattering (SAXS) measurements were performed at the Diamond Light Source, in conjunction with ATR-FTIR and electron microscopy using TEM techniques (*in situ* / cryo TEM).

Results show that 2-line ferrihydrite was produced in all systems with SAXS data and TEM images indicating ~ 1 nm diameter colloidal particles/clusters in solution at pH 0.3 when the solution is undersaturated with respect to ferrihydrite. These nanoparticles/clusters are persistent below ~ pH 1.2 after which aggregation becomes significant. Between pH 1.2 and 3.5, aggregation and precipitation dominate. The results of ATR-FTIR analysis provide insight into the mechanisms of phosphate, borate and sulfate interactions during the hydrolysis process, with both inner and outer sphere complexation observed for different systems. These results highlight the complexity of particle formation in this system and underpin management of the EARP process effluents for the future.