Precipitation pathways for ferrihydrite formation in acidic solutions - DTU Orbit (08/11/2017)

Precipitation pathways for ferrihydrite formation in acidic solutions

Iron oxides and oxyhydroxides form via Fe^{3+} hydrolysis and polymerization in many aqueous environments, but the pathway from Fe^{3+} monomers to oligomers and then to solid phase nuclei is unknown. In this work, using combined X-ray, UV-vis, and Mossbauer spectroscopic approaches, we were able to identify and quantify the long-time sought ferric speciation over time during ferric oxyhydroxide formation in partially-neutralized ferric nitrate solutions ($[Fe^{3+}]$ = 0.2 M, 1.8 <pH <3). Results demonstrate that Fe exists mainly as $Fe(H_2O)_6^{3+}$, mu-oxo aquo dimers and ferrihydrite, and that with time, the mu-oxo dimer decreases while the other two species increase in their concentrations. No larger Fe oligomers were detected. Given that the structure of the mu-oxo dimer is incompatible with those of all Fe oxides and oxyhydroxides, our results suggest that reconfiguration of the mu-oxo dimer structure occurs prior to further condensation leading up to the nucleation of ferrihydrite. The structural reconfiguration is likely the rate-limiting step involved in the nucleation process.

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