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Uranium uptake and redistribution during phase transformation and oriented attachment of iron oxide nanoparticles

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At acidic pH, the growth of goethite nanoparticles from a 6-line ferrihydrite precursor proceeds via a two-step mechanism of phase transformation from to goethite (Gt) followed by alignment of Gt particles and growth into Gt rods via oriented attachment (OA). OA is a non-classical crystal growth mechanism in which primary crystals align in a crystallographically oriented manner prior to fusion and ultimate production of a continuously-crystalline secondary crystal. Species adsorbed to the surfaces of the primary crystals must be desorbed or incorporated into the final product. U(VI) is a common oxidation state for U ions under oxic conditions and is readily sorbed by iron oxide (FeOx) nanoparticles. The interaction of U with FeOx has important implications for the biogeochemical cycling of this toxic and radioactive metal, and iron oxides are in fact an attractive system for the immobilization of U and its sequestration from groundwater sources. In this study, we examined the impact of the OA process on U distributions in Gt. High angle annular dark field scanning transmission electron microscopy and electron energy loss spectroscopy were used to collect information on the spatial distribution of U atoms in or on FeOx particles at atomic resolution. X-ray absorption fine structure spectroscopy informed by *ab initio* molecular dynamics was used to determine the local bonding structure and coordination state of U atoms. Finally, inductively coupled plasma optical emission spectroscopy was used to measure differences in the concentration of acid-extractable U as phase transformation and growth progressed. We found that U(VI) sorbs to the surface of 6-line Fhy nanoparticles but has significantly less sorption to the surface of the Gt product. Furthermore, there is little evidence for the incorporation of U(VI) into the goethite structure; although some U may reside at grain boundaries formed during OA.