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Exploring the impact of Fe-atom exchange on the fate of U(VI)-incorporated goethite

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Over 60 years of nuclear activities have resulted in a global legacy of radioactive waste and contaminated land. Higher activity wastes are destined for disposal in a deep underground geological facility (GDF), with iron (oxyhydr)oxide phases expected to be ubiquitous in and around the repository. Additionally, uranium will be a significant radionuclide in many of these wastes. Given that a range of uranium-incorporated iron (oxyhydr)oxides have been reported in the literature, these mineral phases may be considered a secondary barrier to the migration of uranium in the environment. However, the long-term stability of these phases under fluctuating geochemical conditions is unknown. Stable iron oxyhydroxides (e.g. goethite) have undergone extensive recrystallisation (>90%) during Fe-atom exchange, with incorporated species released and/or reduced during the recrystallisation process. Here, the stability and fate of uranium-incorporated goethite during Fe-atom exchange was investigated. A U(VI)-goethite species was hydrothermally synthesized and reacted with aqueous Fe(II). The system was monitored using geochemical analysis and X-ray absorption spectroscopy (XAS), with an aqueous ^{57}Fe(II) tracer used to track the extent of Fe-atom exchange. This revealed that only ~2% of structural Fe(III) was exchanged with aqueous Fe(II), with the retention of incorporated U confirmed by acid digestions. Despite this, M_{IV}-edge HR XANES and L_{III}-edge EXAFS revealed an ingress of near-surface U(V) as the Fe-atom exchange reaction progressed. Overall, these results suggest that Fe-atom exchange of the outer-most layers of goethite occurred, exposing near-surface U(VI) to the ambient solution chemistry. This provides further insight into the long-term fate of U contaminants in the environment, and the efficacy of iron (oxyhydr)oxides as an immobilization pathway.

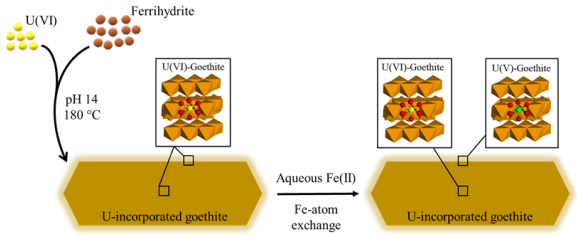

Figure 1. Fe-atom exchange of U(VI)-incorporated goethite.