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Transformation of REE-coprecipitated ferrihydrites and associated REE partitioning and redistribution

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Rare earth elements (REE) are powerful geochemical tracers and are widely used in modern technologies. Their interactions with Fe (oxyhydr)oxides, such as ferrihydrite (Fhy), hematite (Hem), and goethite (Gth), are closely linked to resource formation (*e.g.,* regolith-hosted REE deposits, REE-enriched marine sediments). Previous studies have explored the transformation of Fhy, a typical precursor to crystalline Fe (oxyhydr)oxides, in the presence of REE and their impact on phase transformation. However, the mechanisms controlling REE partitioning and redistribution on Fe (oxyhydr)oxides during Fhy transformation remain unclear. Here, we monitored the transformation of REE-coprecipitated Fhy under oxic conditions, choosing Ce (a redox-sensitive light REE) and Lu (a non-redox-sensitive heavy REE) as representatives. Synthetic pure and REE-bearing Fhy [Ce/Fe (w/w): ~0.007; Lu/Fe (w/w): ~0.004] were incubated at 40 °C and pH = 6.5 for up to 140 days. Powder X-ray diffraction (XRD) data showed that Fhy transformed to Hem and Gth in all systems, with much higher Hem/Gth ratios in the Lu system than in the pure and Ce systems. Scanning transmission electron microscopy coupled with energy dispersive X-ray spectroscopy (STEM-EDX) results revealed a homogeneous distribution of Ce and Lu on the secondary Fe phases. Sequential chemical extraction combined with inductively coupled plasma mass spectrometry analysis (ICP-MS) indicated that Ce and Lu were primarily retained in the solid phases throughout the experiments, with >60% present as strongly-bound forms and <40% as weakly-bound forms. The proportions of different REE species varied during phase transformation, showing a clear pattern: strongly-bound species gradually decreased with a concurrent increase in weakly-bound species at the early stage, followed by a reverse trend at the later stage. In the presence of Ce, the first stage lasted nearly twice as long as in the Lu system. We hypothesize that these results may be related to the distinct transformation pathway of Fhy to Hem and Gth, as well as their mechanisms for sequestering different REE. Overall, these findings will improve our understanding of how Fe (oxyhydr)oxides regulate the geochemical behavior of REE and provide insights into the availability and extraction strategies of Fe (oxyhydr)oxides-associated REE resources.