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Adsorption mechanisms and redox behaviors of cerium on iron oxides: insight from mineral morphology and interfacial structures

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Rare earth elements (REEs) are considered critical metals and have been fueling the development of green technology. Considering the limited natural reserves around the world, weathered products enriched in REEs have demonstrated potential in future mining exploration. Nevertheless, geochemical processes that control the fate and transport of REEs on secondary minerals, especially iron (Fe) oxides, are yet not well studied. Cerium (Ce) distinguishes itself from other REEs by its multiple oxidation states and its potential to be a new tracer in the reconstruction of paleo-redox conditions. Besides surface adsorption to weathered products like clay minerals, Ce(III) can be abiotically oxidized to less soluble Ce(IV) by air or through oxidative scavenging at the mineral-water interface, resulting in unique concentration signals compared to its REE neighbors, known as Ce anomaly. While oxidative adsorption of Ce on manganese oxides has been broadly accepted, research into Ce redox behaviors during adsorption by iron oxides remains inadequate and even shows inconsistency. In this study, a series of macroscopic experiments and synchrotron-based X-ray spectroscopic analyses were carried out to illuminate the impacts of iron oxide mineralogy on Ce adsorption and redox behaviors. Ce adsorption isotherms of synthetic ferrihydrite, goethite, and hematite with different crystallography were examined to analyze the influence of iron oxide morphology, mineral structure, and crystallinity on Ce adsorption. X-ray adsorption fine structure (XAFS) spectroscopy was utilized to determine the oxidation state of adsorbed Ce on iron oxides. This study provides new insight into fundamental adsorption mechanisms of Ce on iron oxides, which will also promote our understanding of Ce enrichment in weathering settings.