

## XAS Investigation of Lanthanide Ion Sorption on Iron Oxide and Cr-Substituted Iron Oxide Surfaces

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Iron and steel structures have been widely used in nuclear industrial facilities. Over a period of time, the surfaces of these structures develop passivity layers that contaminants can reside in<sup>1</sup>. These layers are comprised of a mixture of oxides that may include hematite, magnetite, maghemite, and foreign-ion-substituted iron oxide (typically Cr-substituted iron oxides), depending on the environmental conditions. The incorporation between contaminants and the passivity layers may become an essential problem for some industries, especially for nuclear facilities where decontamination processes are inevitable. To decontaminate these surfaces, it is necessary to dissolve the surface oxide layers to release the contaminants. The stability of the surface species, however, is directly related to their local co-ordination environments, with development of multiple bonds between the sorbents and sorbates leading to greater chemical resistance. Hence, the extent of the dissolution of the oxide layers is also dependent on the structure of the surface species residing on these layers. As a result, it is important to identify the local structure and co-ordination chemistry of sorbed species at the mineral-water interface in order to develop molecular scale predictions of the efficacy for the removal of contaminant metals. This direct structural information can only be obtained from the results by microscopic methods such as X-ray Absorption Spectroscopy (XAS) analysis<sup>2</sup>.

In this study, several synthesized oxides<sup>3,4</sup>, including hematite, maghemite, and magnetite, with different extent of Cr substitution that resemble the oxide layers on iron and steel surfaces were used as the sorbents. Two lanthanides, europium (Eu) and neodymium (Nd) were selected as the sorbates, which also served as analogs of the trivalent actinides (e.g., Am, Cm). Both macroscopic sorption studies and XAS analysis had been

conducted to characterize the surface species structure at the oxide/water interfaces. The total metal ion concentrations were 10<sup>-4</sup> M with 0.01 M NaNO<sub>3</sub> as the background electrolyte at a pH range between 4 to 9. Although the solid characteristics were quite different from each other (e.g., PZCs, surface areas, colors), the results from macroscopic sorption experiments showed that there were little differences in metal ion sorption isotherms obtained on different oxides with an all adsorption edges falling between pH 6 to 7. XAS results from both EXAFS and XANES analysis, on the other hand, indicated that while lanthanides were specifically sorbed on the oxide surfaces, the structural co-ordination of sorbed metals varied upon different sorbents. This result suggested that while the extent of metal ion sorption on the oxides under investigation were similar; the dissolution processes of these contaminant-oxides may be different due to their different binding structures. The results from this study provide valuable information of the chemical structures of the contaminants at the oxide-water interfaces and thus be a significant step toward successful decontamination.

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