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DISSERTATION ABSTRACT

博士の専攻分野の名称 博士（工学） 氏名 Paul Clarence Magdael Francisco

学 位 論 文 題 名

Title of dissertation submitted for the degree

Transformation of iron oxide and hydroxide phases and its effects on contaminant mobility

(鉄酸化物および水酸化物の相変換と汚染物質の移行に対する影響)

Iron oxides and hydroxides [Fe-(hydro)oxides] are one of the most common phases in the earth's crust. Due to their ubiquity in the natural environment, they have long been recognized for their ability to regulate the concentrations of hazardous elements in nature. Furthermore, the relative ease with which they can be synthesized under controlled conditions has made them attractive materials for industrial applications like water-treatment and contaminant remediation. The feasibility of Fe-(hydro)oxides as long-term sinks for contaminants both in nature and in engineered environments depend on their behavior during phase transformation. In this work, the effects of foreign ligands on the transformation behavior of Fe-(hydro)oxides as well as on the behavior of contaminants were investigated.

Chapter 1 reviews the literature on the general properties of Fe-(hydro)oxides, their distributions, recognized applications as well as current knowledge on transformation mechanisms. In this chapter, the factors governing Fe-(hydro)oxide transformation and contaminant transport are identified.

Chapter 2 focuses on the effects of Si, one of the most common ligands present in the natural environment on the transformation kinetics of ferrihydrite. In this work, it was observed that even a small amount of Si significantly retards the crystallization of both goethite and hematite. This work provides new insights on the impacts of Si on ferrihydrite solubility as well as on the morphology of goethite and hematite. New quantitative data on transformation rates as well as activation energies are also presented in this chapter, which may be useful in predicting the crystallization behavior of goethite and hematite in alkaline environments (e.g. high-level waste disposal repositories).

Chapter 3 focuses on the behavior of selenite taken up by ferrihydrite during the crystallization of goethite and hematite, a contaminant originating from weathering of sedimentary rocks as well as from high-level wastes. In this work, a fraction of the selenite co-precipitated with ferrihydrite was released during transformation, with another fraction becoming immobilized within the crystalline phases. A combination of chemical extraction experiments as well as diffraction and spectroscopic studies show that the retention mechanism involves the incorporation of selenite within the crystalline phases. The findings presented in this chapter provides insights that can be used to predict the behavior of selenite or other contaminants during the evolution of Fe-(hydro)oxide sorption substrates.

Drawing from the insights presented in Chapters 2 and 3, Chapter 4 focuses on the effect of Si on the behavior of selenite during ferrihydrite transformation. The retardation of transformation by Si results in the retardation of selenite release back into solution. Furthermore, the presence of Si results

in the release of more selenite back into solution. These observations are attributed to the competitive effect of Si for uptake sites on Fe-(hydro)oxide surfaces.

This work presents important insights on the stability of Fe-(hydro)oxides and their capability to retain contaminants in the presence of elements. These findings may be useful in a broad range of contexts, such as on the behavior of Fe-(hydro)oxides in nature and in engineered environments.