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学 位 論 文 題 名

Comparison of adsorption and post-adsorption behavior of oxyanions between ferrihydrite and schwertmannite

(フェリハイドライトとシュベルトマナイトの陰イオン吸着および吸着後の挙動の比較)

Water contamination is the serious problem around the world, many toxic trace metals such as copper, lead, zinc, and cadmium occur in ground water as cations. Almost of these metals are insoluble with increasing the pH. At pH of normal natural water, the solubility of most trace metal is limited by precipitation as on oxide, hydroxide. These actions dominantly adsorb to the clay minerals and organic matter. Meanwhile, most oxyanions tend to be less adsorbed as the pH increases to alkaline condition. Under some conditions, these anions can remain in solution with relatively high contents such as arsenate, chromate, and selenate is most common trace contamination in surface and groundwater. To remove the oxyanions is the most difficult task because in the natural water there is the coexisting oxyanion had similar properties with toxic and anions, and there is a high concentration in the natural environment. Therefore, it is important to consider a technology and find out the best material to apply for water contamination treatment.

Chapter1 refers to the background and research objectives of this study. In this chapter described iron oxides and hydroxides minerals, which are excellent scavengers both for hazardous cations and hazardous oxyanions. In iron oxides and hydroxide minerals, there are several mineral species such as goethite, hematite, and ferrihydrite etc. At acid mine drainage, there is mineral precipitation such as schwertmannite, it is a meta-stable iron oxy- and hydroxy sulfate found in acidic iron and sulfate-rich environments such as sulfide metal mines. Schwertmannite and ferrihydrite both well known to play an important role in the removal toxic elements from the acid mine drainage and in natural attenuation processes of hazardous elements in acid mine water.

Chapter 2 schwertmannite and ferrihydrite are metastable iron oxide mineral and it is also well known as excellent adsorbents for oxyanions such as arsenate, but the comparison adsorption behavior of schwertmannite and ferrihydrite with other kinds of oxyanions have not been fully or systematically investigated. With this background, the adsorption properties of oxyanions including arsenate, phosphate, chromate, and selenate of schwertmannite and ferrihydrite were investigated. The result of adsorption capacities between schwertmannite and ferrihydrite, in this conditions shown the schwertmannite's adsorption capacity, is higher than the ferrihydrite's adsorption capacity. But the adsorption selectivity of oxyanion adsorption on both schwertmannite and ferrihydrite was decreased in the following order: arsenate $\geq$ phosphate > chromate $\gg$ selenate.

Chapter 3 schwertmannite and ferrihydrite are metastable minerals and both will transform to stable phase as goethite. The disposal waste from adsorption process was necessary to consider when schwertmannite and ferrihydrite adsorbed oxyanions, the transformation process by dissolution and

precipitation, the toxic element may be released to the environment. The post-adsorption behavior of oxyanion onto schwertmannite and ferrihydrite were investigated. To better understand the stabilization of mineral, the solubility of schwertmannite with different oxyanions was calculated by solid solution theory. The result of the post adsorption compares between schwertmannite and ferrihydrite shown that ferrihydrite more stable than schwertmannite. In the case of comparison of many kinds of oxyanions adsorption on schwertmannite had shown the degree of retardation in the transformation to goethite decreased as arsenate=phosphate>chromate>selenate>sulfate. The solubility increase in the following order: arsenate<phosphate<chromate<selenate≈sulfate. Therefore, oxyanions with a high selectivity can stabilize schwertmannite by lowering the solubility of schwertmannite after adsorption of the oxyanions.

Chapter 4 the natural of adsorption for oxyanions species for a wide range of mineral and environmental conditions is fundamental to the prediction of migration and long-term fate of oxyanions in the natural environments. The application surface complexation modeling as known as the theoretical method which is the tool for prediction of adsorption in the natural system. However, ferric oxide had already established in many of the adsorption conditions, such as ferrihydrite adsorption arsenate and phosphate as inner-sphere complexes, was modeled by extended triple layer modeling (ETLM). Meanwhile, schwertmannite is lacking information about surface complexation modeling. In this study was characterized schwertmannite by comparison with ferrihydrite, as explained in Chapter 2, schwertmannite and ferrihydrite had similar adsorption tendency. Thus, the main propose is to apply surface complexation modeling to schwertmannite by using similar input parameter of ferrihydrite. In this present study, oxyanions speciation reaction equations with the surface of schwertmannite were predicted theoretically following previous work of ferrihydrite. The ETLM result for some oxyanions was shown as a trial for explaining the difference in adsorption behavior of oxyanion on schwertmannite from the oxyanions adsorption on the pure ferrihydrite.

Chapter 5 presents the summary and general conclusion of this study, following from the chapter 2 to chapter 4, concluded that the adsorption behavior between schwertmannite and ferrihydrite were similar anions selectivity. Arsenate and phosphate were inner-sphere complexes with both schwertmannite and ferrihydrite. selenate and sulfate were outer-sphere complexes. Chromate anions are intermediate with an inner and outer sphere with schwertmannite and ferrihydrite. The stabilization of ferrihydrite is higher than schwertmannite due to the solubility of ferrihydrite is lower than schwertmannite. Some of the oxyanions which made inner sphere complexes with mineral can change the stabilization of mineral by lowering solubility.