



Equilibrium and kinetics studies on As(V) and Sb(V) removal by Fe²⁺-doped Mg–Al layered double hydroxides



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ABSTRACT

Mg–Al layered double hydroxides (Mg–Al LDHs) doped with Fe²⁺ adsorbed As(V) (AsO₄³⁻) and Sb(V) (Sb(OH)₆⁻) from an aqueous solution through anion exchange with Cl⁻ intercalated in the LDH interlayer. Fe²⁺-doped Mg–Al LDH exhibited superior As(V) removal compared with Mg–Al LDH. The oxidation of Fe²⁺ doped in the Mg–Al LDH host layer to Fe³⁺ increased the positive layer charge of the LDH, thus increasing the anion-uptake capacity owing to stronger electrostatic attractive force between the positively charged layer and the anion. However, Fe²⁺-doped Mg–Al LDH was not superior to Mg–Al LDH in terms of Sb(V) removal. This was attributed to the preferential intercalation of OH⁻ over Sb(OH)₆⁻. The As(V) and Sb(V) removal by LDH followed Langmuir-type adsorption, which proceeded via a pseudo-first-order reaction. The equilibrium and kinetics studies confirm that the adsorption of As(V) and Sb(V) by Fe²⁺-doped Mg–Al LDH was the result of chemical adsorption, involving the anion exchange of AsO₄³⁻ and Sb(OH)₆⁻ with the intercalated Cl⁻.

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1. Introduction

Layered double hydroxides (LDHs) are typically given by the formula [M_{1-x}²⁺M_x³⁺(OH)₂](Aⁿ⁻)_{x/n} • mH₂O, where M²⁺ and M³⁺ are divalent and trivalent metal ions, respectively; x denotes the M³⁺/(M²⁺ + M³⁺) molar ratio (0.20 ≤ x ≤ 0.33); and Aⁿ⁻ is the anion, for example, CO₃²⁻ or Cl⁻ (Ingram and Taylor, 1967; Allmann, 1968; Cavani et al., 1991; Mills et al., 2012). They consist of a stack of M³⁺-bearing brucite-like octahedral layers in which the charge of the positive layer is neutralized by interlayer anions. The interlayer space is also occupied by water molecules in the hydration shells of these intercalated anions. LDHs have been investigated for water preservation and purification. For example, LDHs can take up oxometalates such as arsenite, arsenate, chromate, and selenite, or oxyhalides such as bromate, chlorate, and iodate from aqueous solutions (Goh et al., 2008; Mandal et al., 2009; Goh et al., 2010; Goh and Lim, 2010; Yuan et al., 2013; Paikaray and Hendry, 2013; Paikaray et al., 2013; Theiss et al., 2014). Some studies also examined the treatment of antimonate using LDHs (Kameda et al., 2011, 2012a,b).

In a previous study, Mg–Al LDHs doped with Fe²⁺ were

prepared by coprecipitation, and they were then used to remove Cr(VI) from an aqueous solution (Kameda et al., 2014). The doping replaced some of the Mg²⁺ in the LDH host layer with Fe²⁺. The resulting material could remove Cr(VI) from an aqueous solution because of its anion exchange properties. Upon oxidation of Fe²⁺ to Fe³⁺, Cr(VI) was reduced to Cr(III), which then combined with OH⁻ to produce Cr(OH)₃. Chitrakar et al. found that Fe²⁺-doped LDHs can be used to reduce bromate (Chitrakar et al., 2011). In practical usage, co-precipitation techniques employing Fe(III) species have been used to remove As and Sb from wastewater (Inoue and Munemori, 1980; Nishimura and Umetsu, 2000). However, these techniques tend to generate large amounts of sludge, which is contaminated with As and Sb if the reaction conditions are not set appropriately (Umetsu, 2005). Thus, a new method for treating As and Sb in an aqueous solution should be developed. In this work, the application of Fe²⁺-doped Mg–Al LDH has been proposed for removing As(V) and Sb(V). The Fe²⁺-doped Mg–Al LDH is expected to remove As(V) and Sb(V) from an aqueous solution because of its anion exchange properties. Upon oxidation of Fe²⁺ to Fe³⁺, As(V) and Sb(V) are expected to be finally reduced to As₂O₃ and Sb(OH)₃, respectively. That is, the Fe²⁺-doped Mg–Al LDH is expected to act as an anion exchanger, a reductant, and a precipitant, leading to the effective treatment of As(V) and Sb(V). In the present study, equilibrium and kinetics studies were conducted to determine the effect of Fe²⁺-doped Mg–Al LDH on the removal of As(V) and Sb(V)

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from an aqueous solution.

2. Experimental

Fe²⁺-doped Mg–Al LDH was prepared by the dropwise addition of a Mg–Fe–Al chloride solution to aqueous NaOH at constant pH, as was described in our previous paper (Kameda et al., 2014). Fe²⁺-doped Mg–Al LDH contained 16.6 wt% Mg²⁺, 6.3 wt% Al³⁺, 8.7 wt% Fe²⁺, and 4.4 wt% Fe³⁺. Mg–Al LDH was also prepared by dropwise addition of a Mg–Al chloride solution to aqueous NaOH at constant pH, and it contained 25.0 wt% Mg²⁺ and 6.8 wt% Al³⁺. An aqueous As(V) solution containing AsO₄³⁻ ions was prepared by dissolving Na₂HAsO₄ in deionized water. The aqueous Sb(V) solution containing Sb(OH)₆⁻ ions was prepared by shaking Sb₂O₅ in deionized water at 70 °C for 72 h.

Appropriate quantities of Fe²⁺-doped Mg–Al LDH or Mg–Al LDH were added to 500 mL of 1 mM As(V) or Sb(V) solutions so that the Al/As or Al/Sb molar ratios were between 1 and 3. The subsequent suspensions were bubbled with N₂ and stirred at 30 °C for 240 min. The suspension samples were withdrawn at different time intervals and immediately filtered through a 0.45- μ m membrane filter. The filtrates were analyzed for residual As and Sb. For kinetic studies, the Al/As or Al/Sb molar ratios were set at 1 and it was conducted at 10, 30, and 60 °C for 30 min. In order to determine the adsorption isotherm of As(V) or Sb(V) adsorbed by Fe²⁺-doped

Mg–Al LDH, 20 mL of the As(V) or Sb(V) solution and 0.2 g of Fe²⁺-doped Mg–Al LDH were placed in 50-mL screw-top tubes and shaken at 30 °C for 24 h. In the present work, the initial As(V) concentration is 5–25 mmol/L and the initial Sb(V) concentration is 0.3–0.6 mmol/L.

The two types of LDHs before and after the removal of As(V) or Sb(V) were analyzed by X-ray diffraction (XRD) measurements using Cu K α radiation at 40 kV and 20 mA (scanning rate: 2°/min, step size: 0.02). The oxidation states of As and Sb in the LDHs after the removal of As(V) and Sb(V) and As and Sb compounds were investigated using X-ray photoelectron spectroscopy (XPS) with Mo K α radiation. For the adsorption experiments, the residual concentration of As and Sb in the filtrates was determined using inductively coupled plasma-atomic emission spectrometry (ICP-AES) at 189.0 nm for As and 206.8 nm for Sb, with an error of 0.1 mg/L. In addition, pH and oxidation reduction potential (ORP) during the reaction of Fe²⁺-doped Mg–Al LDH were continuously analyzed. ORP analysis was performed using an ORP electrode (Ag/AgCl internal electrode, 3.3 M KCl electrolyte).

3. Results and discussion

Fig. 1 shows the variation in the removal of aqueous As(V) by

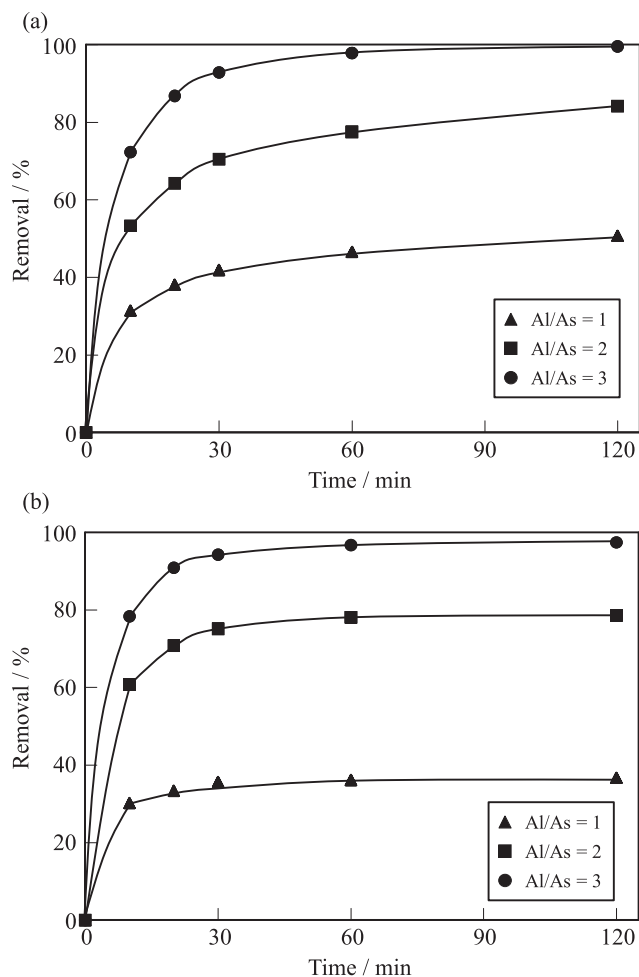


Fig. 1. Removal of As(V) from solution with time for (a) Fe²⁺-doped Mg–Al layered double hydroxide (LDH) and (b) Mg–Al LDH across various molar ratios of Al in the LDH to As(V) in solution at 30 °C.

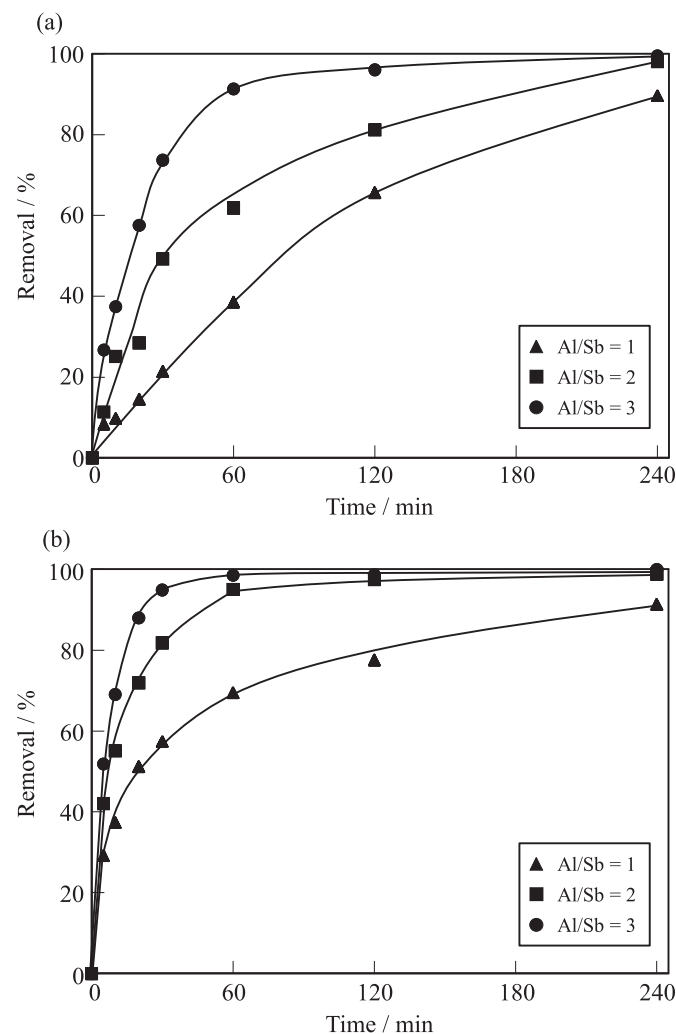


Fig. 2. Removal of Sb(V) from solution with time for (a) Fe²⁺-doped Mg–Al layered double hydroxide (LDH) and (b) Mg–Al LDH across various molar ratios of Al in the LDH to Sb(V) in solution at 30 °C.

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