



Research paper

Layered double hydroxides intercalated with sulfur-containing organic solutes for efficient removal of cationic and oxyanionic metal ions

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ARTICLE INFO

Keywords:

Layered double hydroxide
Intercalation
Sulfur-containing organic anion
Potentially toxic metal
Dual-electronic adsorbent

ABSTRACT

Mg/Al layered double hydroxides (LDH) intercalated with sulfur containing-organic anions (L-cysteine, potassium ethyl xanthate, and sulfate sodium dodecyl) were directly synthesized through a facile co-precipitation method at low supersaturation, yielding Cys-LDH, PEX-LDH, and SDS-LDH, respectively. The synthetic LDH samples were characterized by Fourier transform infrared spectroscopy, X-ray photoelectron spectroscopy, scanning electron microscopy, and X-ray diffraction, and were then applied to remove various potentially toxic metal ions from aqueous solutions. The results demonstrated that the Cys and PEX anions were successfully intercalated into the interlayer region of LDH as a horizontal orientation, while the SDS anions as vertical orientation with a tilt angle of 23°. The synthetic LDH samples were a non-porous material with their low specific surface area (2.99–18.2 m²/g). The Langmuir maximum adsorption capacity (Q_{\max}^0) of LDH generally exhibited the following order: SDS-LDH > Cys-LDH > PEX-LDH > LDH. The Q_{\max}^0 of SDS-LDH was 418 mg/g for Cd²⁺, 402 mg/g for Pb²⁺, 206 mg/g for Cu²⁺, 156 mg/g for Ni²⁺, and 97.4 mg/g for MnO₄⁻ adsorption. The feasible adsorption mechanism was also discussed and proposed. LDH intercalated with sulfur containing-organic anions are a promising dual-electronic adsorbent for efficiently removing cationic and oxyanionic metal ions from water environment.

1. Introduction

Potentially toxic metals present in water environment have been regarded as an important issue in the recent years because of obvious toxicity and carcinogenicity. Therefore, the World Health Organization set a maximum acceptable concentration for the potentially toxic metals in drinking water, such as 0.003 mg/L for cadmium, 1.0 mg/L for copper, and 0.0015 mg/L for lead, 0.02 mg/L for nickel, and 5.0 mg/L for zinc (Siegel, 2002). Development of a new method to effectively remove various kinds of metal ions from water or industrial wastewater has been remarkably investigated during the last decade. Among these methods, ion exchange is the most common process in the removal of heavy metal ions. However, the ion exchange is a high cost technique. Thus, the development of a low-cost and high-capacity sorbent for removing heavy-metal ions from water bodies has recently become a crucial issue.

Layered double hydroxides (LDH) have been regarded as an adsorbent to effectively remove anionic contaminants (Goh et al., 2008). The LDH is a hydrotalcite-like mineral, possessing the unique and outstanding characterizations. The structures of LDH regularly

comprise the brucite-like layers with exchangeable anions in the interlayer region. Therefore, LDH often possesses a positive electrokinetic (ζ) potential (i.e., from +56 mV to +7.5 mV within the solution pH range between 3.0 and 12, respectively) and a high isoelectric point (pH_{IEP} often higher than 12.0) values (Tran et al., 2018b). The brucite-like layers have permanent positively charged metal hydroxide because of coexisting trivalent and divalent cations (M²⁺/M³⁺ isomorphous substitution in octahedral sites). The interlayer contained various anionic species (i.e., CO₃²⁻, NO₃⁻, BrO₃⁻, OH⁻, Cl⁻, and SO₄²⁻) and water. The general form of LDH can be written as [M_{1-x}²⁺M_x³⁺(OH)₂]^{x+} (Aⁿ⁻)_{x/n}·mH₂O, where M²⁺ is divalent metal cations, M³⁺ is trivalent metal cations, and Aⁿ⁻ is the interlayer anions of *n* valence. The molar ratio *x*, which is defined as M³⁺/(M²⁺ + M³⁺), normally ranges from 0.20 to 0.4 (De Roy et al., 2001).

Because the interlayer region of LDH contains abundant host inorganic anions (commonly CO₃²⁻ and NO₃⁻), many researchers have successfully applied many corresponding guest organic anions to replace the host anions in the interlayer region of LDH through a two-stage synthesis process (co-precipitation and anionic exchange) or one-stage synthesis process (i.e., direct co-precipitation method). These

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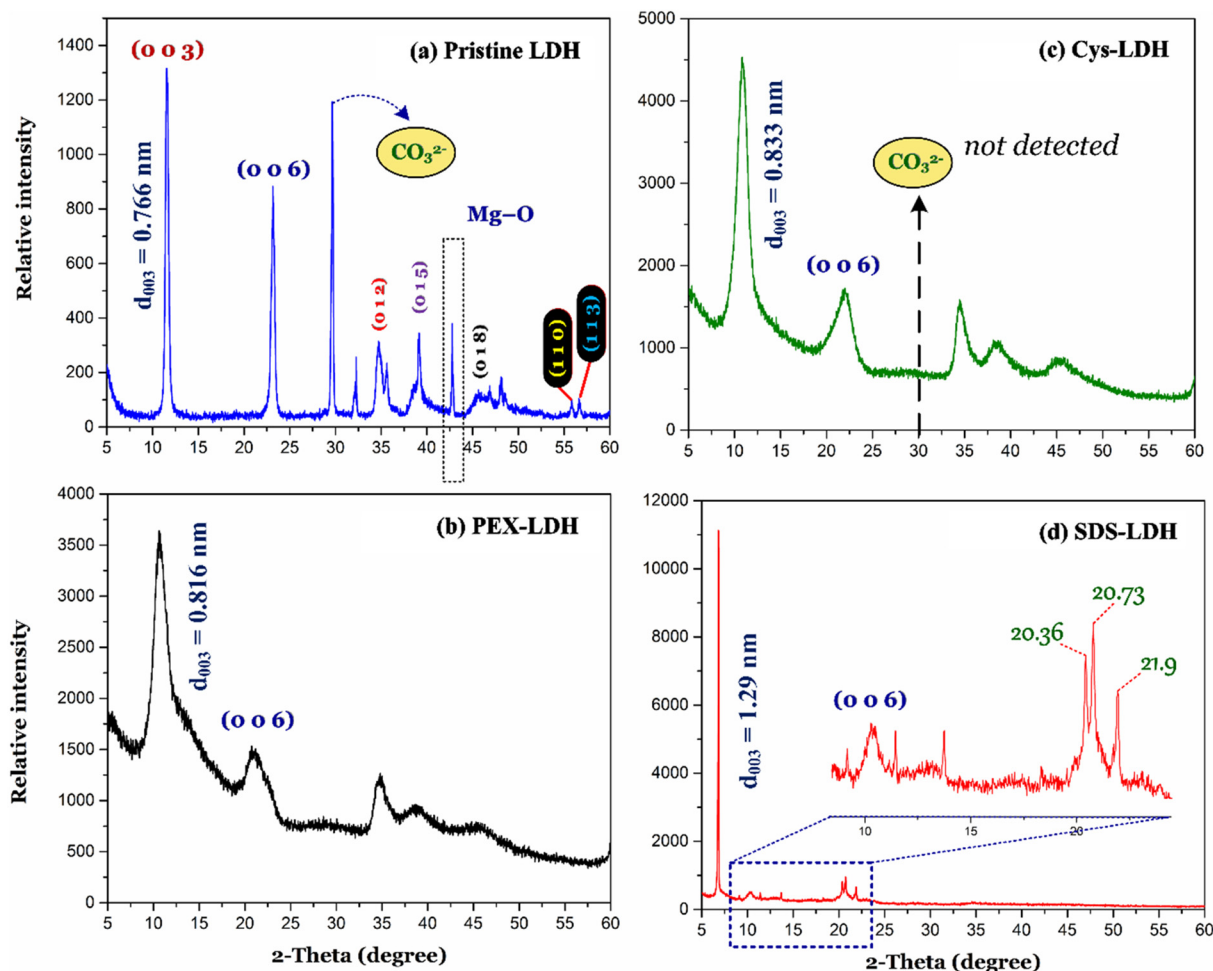


Fig. 1. Representative XRD patterns of (a) pristine LDH, (b) potassium ethyl xanthate-intercalated LDH, (c) L-cysteine-intercalated LDH, and (d) sodium dodecyl sulfate-intercalated LDH.

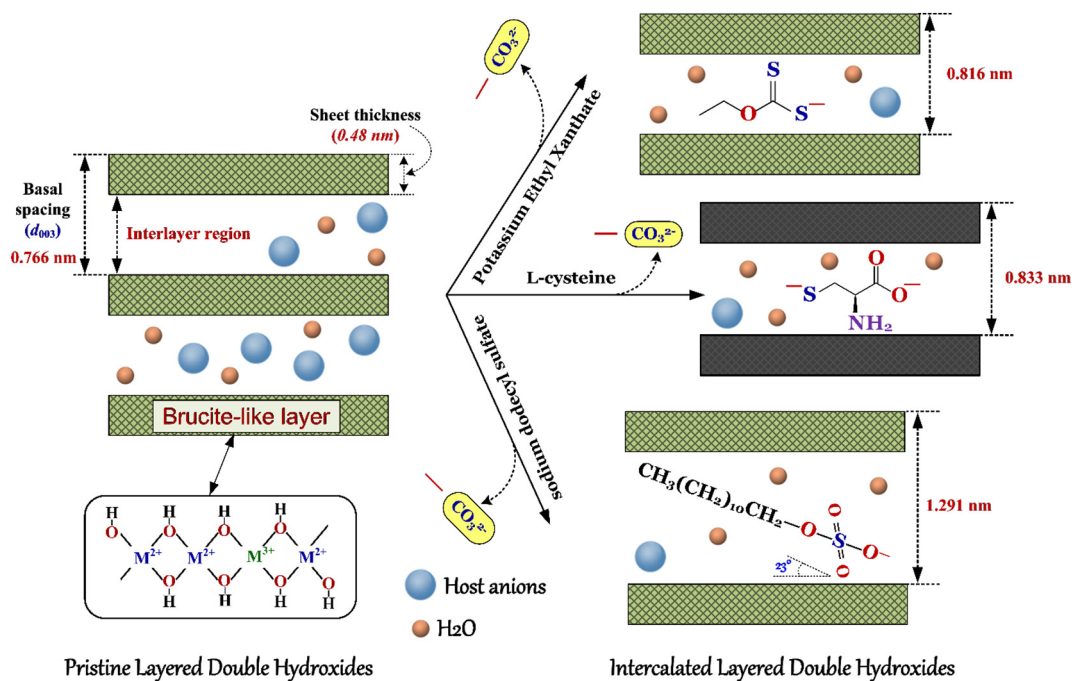


Fig. 2. Schematic representation of spatial orientation of sulfur-containing organic anions in the interlayer region of LDH.

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