



Synthesis of SrHPO₄/Fe₃O₄ magnetic nanocomposite and its application on Pb²⁺ removal from aqueous solutions



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ABSTRACT

Strontium hydrogen phosphate/Fe₃O₄ magnetic nanocomposite (M-SrHPO₄) was successfully synthesized using a facile method of solution growing and applied to immobilize lead ions (Pb²⁺) from acidic aqueous solutions. M-SrHPO₄ was characterized using scanning electron microscopy (SEM), X-ray diffraction (XRD), Fourier transform infrared (FTIR), Brunauer Emmett Teller (BET) and physical property measurement system (PPMS). The resulting M-SrHPO₄ possessed desirable magnetic property (25.214 emu·g⁻¹), which contributed to the separation of solids from solutions after the immobilization process. The equilibrium removal capacity was 1215.5 mg·g⁻¹ for Pb²⁺ at pH = 4.5 within 150 min. The selective immobilization of Pb²⁺ was slightly affected by some competing cations such as Cr³⁺, Cd²⁺, Co²⁺ and Cu²⁺. It is revealed that the immobilization mechanism of Pb²⁺ was based on dissolution-precipitation theory. All of the experimental data indicated that M-SrHPO₄ was promising for Pb²⁺ removal from acidic aqueous solutions.

1. Introduction

Water contamination has become the major threat to the development of human and society. Removing heavy metal ions is always an urgent problem. Even low concentration of Pb²⁺ can cause harmful damage to human's brain, circulatory and kidney system, which could cause serious diseases such as mental retardation, anemia, cancer, intellectual disability and so on, due to the undegradability of Pb²⁺ [1, 2]. Developing highly efficient materials to remove heavy metal ions has aroused many researchers' attention and passion [3–5]. Materials such as activated carbon [6], resins [7], lignin [8] and clinoptilolite [9] have been widely used in waste water treatment. Those materials with small particle size are difficult to separate from water [10], so they are restricted in the practical application in the aspects of removal efficiency and economic benefit.

Strontium hydrogen phosphate (SrHPO₄), as a kind of significant inorganic material, has been widely investigated by many researchers. Gashti et al. [11] have reported SrHPO₄/gelatin composites synthesized by a biomimetic approach and they found that novel SrHPO₄ crystal morphologies could be produced by gelatin depending on the ion concentrations and the first ion diffused. Roming et al. [12] synthesized SrHPO₄ which was well crystallized, uniform in size and non-agglomerated using a polyol-mediated method. Zheng et al. [13] synthesized anorthic SrHPO₄ nanobelts by a hydrothermal method without using

any surfactant as template. It was reported that SrHPO₄ has been successfully synthesized using a method of electrosynthesis and the growth of adherent micrometer-thick SrHPO₄ coatings could be controlled on the cathode substrate [14]. In addition, solution growing as a simple way to synthesize SrHPO₄ has been widely utilized [15]. SrHPO₄ is increasingly utilized in various fields, for example, luminescent materials, flame proofing and ceramics [16]. Zhuang et al. [17] have first investigated the performance of β-SrHPO₄ synthesized by a hydrothermal method in removing Pb²⁺ from acidic aqueous solution. It exhibited a maximum capacity of 1120 ± 22 mg·g⁻¹ at pH = 3.0. Nevertheless, the significant shortcoming is that it is difficult to achieve solid-liquid separation after the immobilization process, which could cause unknown damage and secondary pollution to our environment.

In the last few years, magnetic separation has been one kind of promising method in waste water treatment on account of producing no additional pollution and generating no secondary waste. Magnetic nanoparticles with small diffusion resistance [18], easy phase separation [19] and possible surface modification [20] have drawn considerable attention and are used in the field of sewage treatment. Many materials have combined with Fe₃O₄ nano-particles, which possess good magnetism property, convenient preparation route and biocompatibility [21], and are widely used to remove heavy metal ions. Verma et al. [22] synthesized glycine-functionalized magnetic nanoparticles entrapped calcium alginate beads which were proved to be effective to

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remove Pb^{2+} . Cui et al. [23] have synthesized magnetic carbonate hydroxyapatite/graphene oxide which was utilized for removing Pb^{2+} and methylene blue. Jiang et al. [24] reported the sorption of lead, zinc, and phenol on to amino and thiol modified magnetic multi-walled carbon nanotubes. Although the separation process is resolved using a magnet by means of the magnetic field, the removal efficiency still needs further improvements.

In this work, strontium hydrogen phosphate/ Fe_3O_4 magnetic nanocomposite (M-SrHPO₄) was synthesized by a facile method of solution growing at 80 °C and utilized for Pb^{2+} removal. The effect of initial pH values and concentrations of Pb^{2+} solution, contact time, temperature and interference of coexisting cations were investigated to determine the performance of M-SrHPO₄ on Pb^{2+} removal. The mechanism of Pb^{2+} removal was also studied according to the experiment results.

2. Materials and methods

2.1. Synthesis of Fe_3O_4

Fe_3O_4 was obtained using a solvothermal method [25]. Ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) (2 g) was dissolved in ethylene glycol (EG) (40 mL). Then sodium acetate (NaAc) (6 g) was put into the above mixed system. After NaAc dissolved, add ethanediamine (ETH) (20 mL) to the above solution. The mixture was stirred at room temperature for 60 min and then was poured into a stainless steel autoclave. The temperature of autoclave was maintained at 170 °C for 7 h. The obtained product was then washed with deionized water and ethanol for two times respectively using a magnet. The final product was dried at 60 °C for 24 h in a vacuum drying oven.

2.2. Synthesis of M-SrHPO₄

Strontium nitrate ($\text{Sr}(\text{NO}_3)_2$) (4.8 g) was dissolved in 100 mL of deionized water and the temperature was then heated to 80 °C. Diammonium phosphate ($(\text{NH}_4)_2\text{HPO}_4$) (1.8 g) was then put into Sr (NO_3)₂ solution after $\text{Sr}(\text{NO}_3)_2$ was completely dissolved and the mixture was stirred for 1 h. Fe_3O_4 (0.3 g) was dispersed into 100 mL of deionized water by means of ultrasonic dispersion for 1 h. Next, the dispersion of Fe_3O_4 was added into the precursor of SrHPO₄ dropwise under mechanical agitation. After the mixed solution was stirred for 24 h at 80 °C, the obtained M-SrHPO₄ was collected with the action of magnetic field, washed for several times using deionized water and ethanol, and dried in a vacuum drying oven at 60 °C for 12 h.

2.3. Characterizations

The powder X-ray diffraction (XRD) patterns were recorded by a D8 ADVANCE diffractometer, with monochromatized CuK_α radiation ($\lambda = 0.154 \text{ nm}$). The Fourier transform infrared (FTIR) spectrum was acquired with a Fourier infrared spectroscopy (FTIR, NICOLET 6700, Thermo, America). The zeta potential was measured with a zeta potential analyzer (Zetasizer Nano ZS, Malvern, Britain). The morphology of β -SrHPO₄ was carried out by a field emission scanning electron microscope (FESEM) (Sirion200, FEI, America). The concentration of Pb^{2+} was analyzed by an inductively coupled plasma-optical emission spectroscopy (ICP-OES) (PE Optima 2100DV, Perkin-Elmer, America). The magnetic properties of samples were measured using a physical property measurement system (PPMS, Quantum Design, USA). BET analysis was carried out by a full-automatic surface area and pore porosity analyzer (ASAP2020HD88, America).

2.4. Immobilization experiments

The effect of pH values (2.0–6.0) on Pb^{2+} immobilization was performed by adding M-SrHPO₄ (0.03 g) into Pb^{2+} solution (30 mL) of

2000 $\text{mg}\cdot\text{L}^{-1}$. The pH were adjusted using dilute nitric acid and dilute ammonia solution. The immobilization kinetics of Pb^{2+} was investigated within 240 min (0.03 g of M-SrHPO₄, 30 mL of Pb^{2+} solution with the concentration of 2000 $\text{mg}\cdot\text{L}^{-1}$, pH = 4.5). Different initial concentrations of Pb^{2+} (500 $\text{mg}\cdot\text{L}^{-1}$ to 2400 $\text{mg}\cdot\text{L}^{-1}$, pH = 4.5) were studied to analyze the immobilization isotherms. M-SrHPO₄ (0.03 g) was put into 30 mL of Pb^{2+} solution (initial concentration of 2000 $\text{mg}\cdot\text{L}^{-1}$) at different temperatures (from 299 to 339 K) to study the effect of temperature values on the immobilization process. To study the effect of different cations (Cr^{3+} , Cd^{2+} , Co^{2+} and Cu^{2+}) on Pb^{2+} removal, corresponding nitrates were dissolved into Pb^{2+} solution (150 $\text{mg}\cdot\text{L}^{-1}$ for all cations and 2000 $\text{mg}\cdot\text{L}^{-1}$ for Pb^{2+}). All the removal processes were carried out using a reciprocal shaker. The removal amount of Pb^{2+} on M-SrHPO₄ was computed using the following formulas:

$$q_e = \frac{(C_0 - C_e)V}{M} \quad (1)$$

$$R_e = \frac{C_0 - C_e}{C_0} \times 100\% \quad (2)$$

where q_e ($\text{mg}\cdot\text{g}^{-1}$) is the removal capacity of Pb^{2+} at equilibrium time. The initial and equilibrium concentrations of Pb^{2+} are represented by C_0 ($\text{mg}\cdot\text{L}^{-1}$) and C_e ($\text{mg}\cdot\text{L}^{-1}$) respectively. V (L) is the volume of lead solution and M (g) is the amount of M-SrHPO₄. R_e is the removal efficiency of M-SrHPO₄ on Pb^{2+} .

3. Results and discussion

3.1. Characterizations of M-SrHPO₄

The XRD patterns of Fe_3O_4 (*1), M-SrHPO₄ (*2) and the standard data of anorthic SrHPO₄ phase (JCPDS No. 70-1215) (*3) are shown in Fig. 1a. It is observed that the phase of M-SrHPO₄ was consisted of the phase of SrHPO₄ and Fe_3O_4 . The diffraction peaks of M-SrHPO₄ (*2) at $2\theta = 25.4^\circ, 25.5^\circ, 25.7^\circ, 29.3^\circ, 29.8^\circ$ and 31.6° represented the (002), ($\bar{2}01$), (200), (120), ($\bar{1}\bar{2}1$) and ($\bar{1}21$) lattice planes of SrHPO₄, respectively. The diffraction peaks at $2\theta = 18.3^\circ, 35.4^\circ, 53.4^\circ$ and 56.9° represented the (111), (311), (422) and (511) lattice planes of Fe_3O_4 and this was in accordance with the standard data of Fe_3O_4 (*1) (JCPDS No. 88-0866). These characteristic diffraction peaks confirmed the component of M-SrHPO₄.

In order to study the characteristics of M-SrHPO₄, the FTIR analysis was performed and the spectra of Fe_3O_4 (*1) and M-SrHPO₄ (*2) are shown in Fig. 1b. The strong peaks at 586.26 cm^{-1} of Fe_3O_4 and 586.41 cm^{-1} of M-SrHPO₄ were attributed to the Fe–O vibration of Fe_3O_4 [26]. The bands at 3446.22 cm^{-1} of Fe_3O_4 and 3444.29 cm^{-1} of M-SrHPO₄ were caused by the stretching vibration of the O–H group. The adsorption peaks at 1641.15 cm^{-1} on Fe_3O_4 and 1646.94 cm^{-1} of M-SrHPO₄ indicated the existence of –NH₂ group [25]. The bands at 2923.60 cm^{-1} and 2418.33 cm^{-1} on M-SrHPO₄ were assigned to hydrogen bridge bonds from HPO_4^{2-} groups [12]. The peaks at 1132.08 cm^{-1} and 1072.25 cm^{-1} on M-SrHPO₄ were associated with the asymmetric stretching vibration of P–O in PO_4^{3-} groups. The peak at 883.25 cm^{-1} on M-SrHPO₄ was caused by the symmetrical stretching vibration of P–O (H). The band at 566.98 cm^{-1} on M-SrHPO₄ was attributed to the bending vibration of PO_4^{3-} groups [20]. It is concluded that Fe_3O_4 was successfully anchored on M-SrHPO₄.

The Barrett–Joyner–Halenda (BJH) method was used to determine the plot of the pore size distribution according to the N₂ adsorption and desorption analysis, and the BET analysis results were shown in Fig. 1c. The specific surface area was $5.05 \text{ m}^2\cdot\text{g}^{-1}$ and the adsorption average pore width was 20.71 nm for M-SrHPO₄.

The method of zeta potential is used to analyze the surface charge properties of β -SrHPO₄ and Fig. 1d shows the measurement results of M-SrHPO₄. The pH_{zpc} , determining the electrophoretic mobility where

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