



Magnetite/Lanthanum hydroxide for phosphate sequestration and recovery from lake and the attenuation effects of sediment particles

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ABSTRACT

An effective approach for eutrophication control and phosphate recovery remains a longstanding challenge. Herein, we present a new technique for phosphate sequestration in lake and phosphate recovery using novel magnetically recoverable magnetite/lanthanum hydroxide [M-La(OH)₃] hybrids that can be prepared using a simple one-pot synthesis method. Batch studies show that M-La(OH)₃ exhibits a strong sorption towards phosphate with sorption capacities of up to 52.7 mg-P/g at pH 7.0 in water. A simple model indicates that the efficiency of M-La(OH)₃ for phosphate sequestration in lake is significantly attenuated by 34–45% compared to that in water, due to interference from sediment particles. However, our results demonstrate that sediments suspensions mixed with a M-La(OH)₃ content of 1–3% exhibit a capability of up to 1.2 mg-P/g for sequestering external phosphate compared with that of 0.2 mg-P/g for pristine sediment at pH 7.3. M-La(OH)₃-mixed sediment suspensions appear to effectively sequester phosphate over an environmentally relevant pH range from 4 to 8.5. Phosphorus (P) fractionation experiments indicate that the enhanced phosphate sorption by M-La(OH)₃-mixed sediment suspensions is mainly due to the increased fractions of NaOH-P and inorganic P. This work indicates that the M-La(OH)₃ has the potential for phosphate sequestration and recovery from lake.

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1. Introduction

About 30–50% of global water sources are facing the problem of eutrophication as a consequence of excessive dissolved phosphorus (P) in freshwaters (Funes et al., 2016; Liu et al., 2016). To counteract the occurrence of eutrophication, the successful reduction of dissolved P mainly as the form of phosphate in freshwater highly depends on both of controlling internal P release from sediments and limiting introduction of external P inputs (Ye et al., 2017). Despite the great progress in limiting P inputs from point contaminant sources of P particularly wastewater discharges (Fang et al., 2017a,b), the control of non-point P sources such as

agricultural runoffs is still problematic. As a result, the external P inputs into freshwaters from these sources still occur. In addition, the capacity of lake sediments to reduce the continuously introduced P is limited and can be affected by redox conditions (Sondergaard et al., 2003).

Several techniques such as sediment dredging, sediment oxygenation, sediment capping with metal salts [e.g., aluminum (Al) and calcium (Ca)] have been adopted to sequester/immobilize phosphate in sediment, of which the control of P release using chemical salts or materials receives increasing attention due to its high efficiency and energy effectiveness (Funes et al., 2016; Zhang et al., 2012). The addition of Al or Fe has been widely applied for P sequestration in lake restoration through effectively precipitating phosphate in sediment (Deppe and Benndorf, 2002; Funes et al., 2016; Yamada et al., 2012). Although these materials can effectively reduce dissolved P from freshwater, the capped P in lake sediment may be re-released into overlying water phase due to the aging of materials and variations of environmental conditions such as pH and redox potential (Deppe and Benndorf, 2002; Egemose

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et al., 2009).

Owing to the strong affinity of lanthanum (La) with phosphate, La-based bentonite (i.e. Phoslock) was first invented for phosphate removal (Copetti et al., 2016; Douglas et al., 1999, 2016; Haghseresht et al., 2009; Robb et al., 2003). And later, a number of La-based materials such as La modified alumina and zeolite/lanthanum hydroxide have been developed for effectively sequestering P in sediments with a promising P sorption capacity of up to 108 mg/g (Lesnichaya et al., 2009; Xie et al. 2014, 2015; Zhang et al., 2014, Zhang et al., 2012; Wang et al., 2017). It shows that La can be stably present in sediments with a negligible effect on aquatic organisms (Copetti et al., 2016; Balusamy et al., 2015). Despite the advantage of La for phosphate removal, the extensive application of no recyclable La-based materials leads to the high chemical consumption of non-renewable and expensive La resource.

A potentially feasible solution is to magnetize the nanoparticles through hybridizing with biocompatible Fe_3O_4 nanoparticles so that they can be separated, recovered and reused; and this technique has recently been extensively applied for developing magnetic sorbents and catalysts (Funes et al., 2016, 2017; Fang et al., 2017a,b; Lai et al., 2016; Wang et al., 2016). Motivated by this concept, P recovery from lake sediment and overlying water using a La-based magnetic material appears an alternative solution for tackling P resource depletion, restoring eutrophic lake and avoiding high consumption of La resource. However, there is limited knowledge in this regard that can be found in the literature. A very recent work reported a core/shell $\text{Fe}_3\text{O}_4@\text{SiO}_2@\text{La}(\text{OH})_3$ magnetic sorbents for phosphate removal from wastewater (Lai et al., 2016). However, their method is relatively tedious and complicated. Thus a more facile synthesis method that can produce more effective La-based magnetic sorbents is needed. Moreover, most studies have been focusing on P removal or recovery from wastewater, the application of such technique for P sequestration and recovery from lake has been rarely studied. In particular, the effects of sediment particles on the performance of the magnetic La-sorbents for P removal from lake during resuspension/sedimentation process at the water-sediment interface remain unclear.

Thus this work is devoted to developing a facile one-step hydrothermal method to synthesize magnetite/lanthanum hydroxide magnetic hybrids (M-La(OH)₃) for P recovery from lake. The M-La(OH)₃ magnetic sorbents were prepared and systematically characterized by X-Ray diffraction (XRD), transmission electron microscopy (TEM) and vibrating sample magnetometer (VSM). The sorption performance of M-La(OH)₃ for phosphate sorption from aqueous solution was first investigated in terms of sorption kinetics and capacity and the effects of pH. Hereafter, the efficiency of M-La(OH)₃ for phosphate recovery from sediment suspension and the effects of sediment particles on the performance of the magnetic sorbents was systematically assessed. Finally, the P fraction in pristine and M-La(OH)₃-amended sediment before and after loading external phosphate, as well as after M-La(OH)₃ extraction were determined.

2. Materials and methods

2.1. Materials and reagents

All chemicals used in this study were analytical reagents of high purity. Iron (III) chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$; 98%), iron (II) chloride ($\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$; 99%) and KH_2PO_4 (99%) were purchased from Aladdin (China). deionized water (DI) with a conductivity of 18 M Ω /cm was used to prepare all solutions. A 1000 mg-P/L stock solution of phosphate was prepared by dissolving 1.109 g of KH_2PO_4 in 250 mL DI water.

2.2. Synthesis and characterization of M-La(OH)₃

A simple one-pot precipitation and hydrothermal method was adopted for the preparation of M-La(OH)₃. Based on the nominal mass ratio of La(OH)₃, the synthesized sorbents were designated as M-5%La(OH)₃, M-10%La(OH)₃, M-20%La(OH)₃, M-30%La(OH)₃, M-50%La(OH)₃ and M-60%La(OH)₃. The actual contents of La in the M-La(OH)₃ were determined using Inductively Coupled Plasma/mass spectrometer (ICP-MS); Optima 5300 DV, Perkin-Elmer. Detailed description of synthesis method and determination of La contents are given in Text S1 of Supplementary Material.

The crystalline structure of the synthesized sorbents was characterized using a Bruker D8-Focus X-ray diffractometer by applying monochromated $\text{CuK}\alpha$ radiation ($\lambda = 0.15418$ nm) in the range of 20–80 °2 θ . The morphology, particle size and elemental composition of the sorbents were analyzed by a JEOL 2100F TEM equipped Energy-Dispersive X-ray spectroscopy (EDS) operating at 200 kV. The magnetic property of the samples was measured by a vibrating sample magnetometer (VSM; PPMS-9T, Quantum Design, USA). The Brunauer-Emmett-Teller (BET) surface areas of the synthesized M-La(OH)₃ hybrids were determined through N₂ adsorption/desorption over a Micromeritics ASAP 2010 system at 77 K.

2.3. Phosphate sorption in synthetic water

Both sorption kinetic and isotherm experiments were first performed in 50 mL polypropylene (PP) plastic tubes to study and compare the removal efficiency of phosphate by the synthesized M-La(OH)₃ with different La(OH)₃ ratios at pH 7.0 \pm 0.1. A preliminary experiment showed that the adsorption of phosphate to the plastic tube is negligible. The sorption kinetics study was carried out by spiking an appropriate amount of phosphate stock solution into each 40 mL M-La(OH)₃ suspension (0.1 g/L) to achieve an initial phosphate concentration of 5 mg-P/L. The pH of the suspensions was maintained at pH 7.0 \pm 0.1, and the ionic strength was controlled with 0.01 M NaCl. At each reaction time, sample was filtered through a 0.22 μm mixed cellulose membrane filter and the filtrate was stored for further analysis. The sorption isotherm experiments were conducted with the same ionic strength of 0.01 M NaCl as above at pH 7.0 \pm 0.1. Appropriate amounts of phosphate stock solution were spiked into 0.1 g/L of M-La(OH)₃ stock solution to obtain varying initial phosphate concentrations of 1–10 mg-P/L. The sample tubes were then shaken in a mechanical shaker at 25 \pm 1 °C at 300 rpm for desired time depending on different experiments (i.e. up to 3 h for kinetic experiments, and 2 h for sorption isotherm experiments). The effect of pH on the phosphate sorption by M-La(OH)₃ sorbents was also investigated with a M-La(OH)₃ dosage of 0.1 g/L and an initial P concentration of 5 mg-P/L at 25 \pm 1 °C in a pH range of 3–11 by adjusting pH with NaOH or HCl. Thermodynamics of phosphate sorption by the M-La(OH)₃ was investigated with a M-La(OH)₃ dosage of 0.1 g/L and an initial P concentration of 5 mg-P/L at pH 7.0 \pm 0.1 at 25, 35 and 45 °C, respectively. The effect of redox state on the phosphate sorption capacity of the M-La(OH)₃ was further investigated with a M-La(OH)₃ dosage of 0.1 g/L and an initial P concentration of 5.9 mg-P/L at pH 7.0 \pm 0.1 by comparing the phosphate sorption without controlling O₂ with that under anoxic conditions in a Lab2000 anaerobic glovebox (O₂ < 1 ppm; Etelux instrument). After reaction, all samples were collected after filtering suspensions through 0.22 μm cellulose membrane filters, and then analyzed using the method given in Section 2.5. In addition, the loss of La from the M-50%La(OH)₃ as a function of time and pH was investigated with a M-50%La(OH)₃ dosage of 0.1 g/L with and without the addition of P (5 mg-P/L). After reaction certain time, the samples were filtered through 0.22 μm cellulose membrane filters, and the filtrates were

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