



Efficient removal of phosphate by facile prepared magnetic diatomite and illite clay from aqueous solution

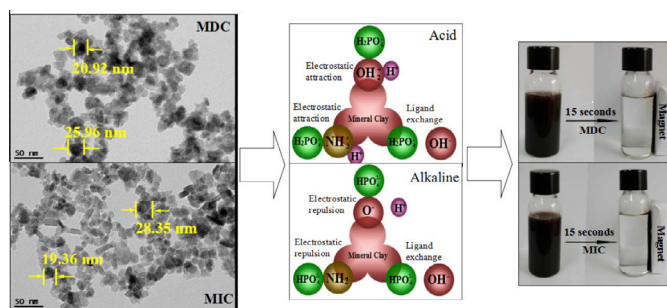
Jian Chen, Liang-guo Yan^{*}, Hai-qin Yu, Shuang Li, Li-lu Qin, Guo-qiang Liu, Yun-fei Li, Bin Du

School of Resources and Environment, University of Jinan, Shandong Provincial Engineering Technology Research Center for Groundwater Numerical Simulation and Contamination Control, Jinan 250022, PR China

HIGHLIGHTS

- The magnetic clays were synthesized via a simple one-pot solvothermal method.
- The adsorbents had ordered structure, rich functional groups and strong magnetism.
- Phosphate adsorption by the magnetic clays was fast and efficient.
- The magnetic clays can be quickly and easily separated using a magnet.

GRAPHICAL ABSTRACT



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ABSTRACT

We present magnetic diatomite and illite clay nanocomposites by loading Fe_3O_4 nanoparticles onto the raw diatomite or illite clay surfaces via a simple one-pot solvothermal method. The magnetic clays have ordered structure, rich functional groups and strong magnetism as verified by X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning and transmission electron microscope (SEM, TEM), and vibrating sample magnetometer (VSM). Subsequently, the magnetic diatomite (MDC) and illite clay (MIC) were used as adsorbents for fast and efficient removal of phosphate from aqueous solution by batch equilibrium experiments. The adsorption kinetic, isothermal and mechanistic properties were studied. The results demonstrated that the phosphate adsorption onto MDC and MIC was pH-dependent, completed within 30 min and possessed higher capacities than the raw clays. The adsorption kinetic and isotherm data followed the pseudo-second-order equation and the Langmuir model, respectively. The adsorption mechanisms of phosphate were ascribed to the electrostatic attraction and ligand exchange. Furthermore, the MDC and MIC endowed a superior selectivity, regeneration, reusability and can be quickly and easily separated using a magnet before and after the adsorption process.

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1. Introduction

Phosphate is regarded as one of important nutrient element to support the growth of biological organisms in aquatic environment [1]. Nevertheless, the presence of extensive phosphate in water

body resulting from anthropogenic activities such as industrial development, agricultural production and population explosion leads to the eutrophication of receiving waters, which causes the overgrowth of algae, depletion of dissolved oxygen, deterioration of water quality, and depopulation of aquatic animals [2–4]. Therefore, there is an urgent demand for a highly effective, reliable, and economical technique for removal of phosphate.

^{*} Corresponding author. Tel.: +86 531 82767617.

E-mail addresses: yanyu-33@163.com, chm_yanlg@ujn.edu.cn (L.-g. Yan).

Up to date, numerous approaches have been developed to reduce and control the concentration of phosphate in wastewater. Compared to other methods, adsorption is relatively useful and cost effective for phosphate removal due to its simplicity and flexibility of design, cost-effectiveness, greater selectivity, faster regeneration kinetics and ease of operation [5,6]. Moreover, various types of adsorbents for phosphate removal have been tested. Among them, iron oxides/hydroxides have been greatly studied due to their outstanding properties like high adsorption capacity, larger specific surface area and hydrodynamic properties [7]. Yoon et al. used magnetite nanoparticles to adsorb phosphate from aqueous solution and examined the adsorption kinetics and isotherms [8]. Zelmanov et al. investigated the phosphate adsorption properties of iron oxide/hydroxide nanoparticles-based agglomerates [9]. Wang et al. studied the differences between ferrihydrite complexes and ferrihydrite–humic acid for phosphate adsorption through kinetic and isotherm experiments [10]. We can easily see that iron oxides/hydroxides show great removal efficiency for phosphate. Nevertheless, sophistication in operation, high-cost and particles agglomeration may limit the practical application and lead to the loss of the material properties.

Among different kinds of clay minerals, the diatomite and illite clay show great potential as adsorbents. Diatomite is a natural material formed from the remains of diatoms, which grew and were historically deposited in sediments of seas or lakes [11]; and possess a 3-dimensional porous exoskeleton comprised of silica and hollow structure with a specific pattern of nanopores build by amorphous silica [12]. Illite is a common soil component in temperate to cold region. It is a non-expanding, typical 2:1 cationic layered silicate including two silicon oxygen tetrahedron and an alumina oxygen octahedral with potassium as the interlayer cation. They are plentiful and natural available minerals in the world and possess excellent performance such as porosity, small particle size, good bonding ability, great electrical insulating property and thermal stability [13]. Furthermore, there was little information about combining magnetic nanoparticles with diatomite and illite. Toster et al. synthesized the diatom frustules coated with magnetite nanoparticles [14], the resulting material exhibited superparamagnetic behavior and high adsorption capacity for phosphate. A new composite material was prepared based on the deposition of nanosized zero-valent iron particles and cyanocobalamin on a diatomite matrix and applied for the catalytic transformation of persistent contaminants [13].

Additionally, the mixture containing phosphate solution and adsorbent powder was hard and time consuming to separate after adsorption. The addition of magnetic component could greatly improve the separation efficiency. A three-component composite, $\text{Fe}_3\text{O}_4@\text{C}/\text{Mg}/\text{Zn}/\text{Ni}-\text{Al}$ LDH was prepared and applied as magnetic adsorbent for removal of phosphate in aqueous solutions [15]. Compared to centrifugation and gravity sedimentation, magnetic separation can achieve fast separation under the effect of magnetic field. Magnetic Fe_3O_4 is commonly used as a magnetic addition due to its unique magnetic response and large surface area [16].

Therefore, the aim of this study was to load magnetic Fe_3O_4 onto the raw diatomite (RDC) and illite clay (RIC) via a facile one-pot solvothermal route and evaluate the adsorption performance of the magnetic diatomite clay (MDC) and magnetic illite clay (MIC) for phosphate removal in aqueous solution. The adsorbents were characterized by XRD, FTIR, SEM, TEM, EDS, VSM and zeta potential. Batch experiments were investigated for the effects of various functions on the phosphate adsorption, such as adsorbent dosage, contact time, phosphate concentration, initial solution pH and coexistent anions. The adsorption kinetic, isotherm and mechanisms of phosphate on MDC and MIC were also discussed in detail.

2. Materials and methods

2.1. Materials and chemicals

The clay used in this study, RDC was obtained as powder through Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China) without any treatment. RIC was supplied by Lingshou County Xinnuo Mineral Co. (Hebei, China) and milled into a fine powder. The chemicals, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{CH}_3\text{COONa} \cdot 3\text{H}_2\text{O}$, NaOH , HNO_3 , KH_2PO_4 , NaCl , NaNO_3 , $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$ were analytical reagent grade and received from Kermel Chemical Reagent Co., Ltd. (Tianjin, China), ethylene glycol, ethylenediamine were also analytical reagent grade and received from Tianjin Fuyu Fine Chemical Co., Ltd. (Tianjin, China).

2.2. Preparations of MDC and MIC

The magnetic nanoparticles (Fe_3O_4) can be synthesized by solvothermal process in our previous study [15,17]. In this work, we further prepared the composites MDC and MIC similar to this method. Firstly, $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (2.0 g) was dissolved in ethylene glycol (40 mL) under vigorously stirring to form a clear brown solution. Then, $\text{CH}_3\text{COONa} \cdot 3\text{H}_2\text{O}$ (6.0 g) and ethylenediamine (20 mL) were added into the brown solution. Afterward, RDC (0.8 g) and RIC (0.8 g) were dispersed into the above solution, respectively. The mixture was stirred for 30 min and sealed in a stainless steel with polytetrafluoroethylene liner, followed by being heated for 8 h at 200 °C. After cooling down, the resultant solid was washed several times with deionized water and dried at 60 °C overnight. The collected solids were lightly ground to pass through a 100 mesh sieve and stored in a desiccator for further phosphate adsorption studies.

2.3. Batch adsorption experiments

A series of batch equilibrium tests were conducted to investigate the phosphate adsorption performances of MDC and MIC. The adsorption experiments were conducted as follows: a certain amount of MDC or MIC composites were added to 20 mL of phosphate solution. This mixture was shaken on a constant temperature oscillator at 200 r/min. Then, the suspension was separated by magnetic separation. The supernatant was filtered through a 0.45 μm membrane filter, and the residual phosphate concentration was determined by the molybdate blue spectrophotometric method, at a wavelength of 700 nm using a UV–Vis spectrophotometer (UV-2450, Shimadzu). Each of the experimental results was conducted three times under identical conditions and the average value was used. The phosphate adsorption capacity q_e (mg/g) (Eq. (1)) or removal ratio R (%) (Eq. (2)) was calculated by the following expression:

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

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

where C_0 (mg/L) and C_e (mg/L) are the initial and equilibrium concentration of phosphate in solution, m (g) is the adsorbent dry weight, and V (L) is the suspension volume.

To be specific, the adsorption of phosphate (25 mg/L) from aqueous solution by different adsorbent dosages (0.02–0.5 g/20 mL) was carried out at 25 °C and original pH of the solution for 60 min shaking. For the effect of contact time, the samples (25 mg/L phosphate concentration, 0.15 g adsorbent and original pH) were withdrawn from the constant temperature oscillator with shaking at predetermined time (0–90 min). In the

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