



Phosphate adsorption on lanthanum loaded biochar



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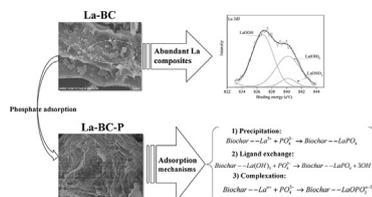
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HIGHLIGHTS

- Various lanthanum composites were doped on the surface of biochar for absorbing phosphate.
- The adsorption mechanism was investigated by means of SEM-EDS, XRD, XPS and FT-IR spectroscopy.
- Precipitation, ligand exchange and complexation were the dominant adsorption mechanism.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 22 August 2015

Received in revised form

21 January 2016

Accepted 2 February 2016

Available online xxx

Handling Editor: X. Cao

Keywords:

Biochar

Phosphate

Adsorption

Lanthanum

ABSTRACT

To attain a low-cost and high-efficient phosphate adsorbent, lanthanum (La) loaded biochar (La-BC) prepared by a chemical precipitation method was developed. La-BC and its pristine biochar (CK-BC) were comparatively characterized using zeta potential, BET surface area, scanning electron microscopy/energy dispersive spectrometer (SEM-EDS), X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and Fourier transform infrared spectroscopy (FT-IR). The adsorption ability and the mechanisms during adsorption process for the La-BC samples were also investigated. La loaded on the surface of biochar can be termed as La-composites (such as LaOOH, LaONO₃ and La(OH)₃), leading to the decrease of negative charge and surface area of biochar. La-BC exhibited the high adsorption capacity to phosphate compared to CK-BC. Adsorption isotherm and adsorption kinetic studies showed that the Langmuir isotherm and second order model could well describe the adsorption process of La-BC, indicating that the adsorption was dominated by a homogeneous and chemical process. The calculated maximum adsorption capacity was as high as 46.37 mg g⁻¹ (computed in P). Thermodynamic analysis revealed that the adsorption was spontaneous and endothermic. SEM, XRD, XPS and FT-IR analysis suggested that the multi-adsorption mechanisms including precipitation, ligand exchange and complexation interactions can be evidenced during the phosphate adsorption process by La-composites in La-BC.

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1. Introduction

Excess discharge of phosphate from agricultural and industrial

productions into runoff is one of the major factors for water eutrophication. The minimum margin concentration of phosphate to induce eutrophication is 0.02 mg L⁻¹ (Yao et al., 2013). Eutrophication could facilitate the fast growth of blue and green algae that leads to serious environmental and aesthetic problems (Kilpima et al., 2014). An emergent demand is to develop some effective technologies for removing phosphate from natural water system.

Among most of the removal technologies for phosphate including chemical, physical and biological methods, adsorption is

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a promising alternative (Jung et al., 2015; Park et al., 2015). Various adsorption materials, such as activated carbon, carbon nanotubes and zeolite, have been successfully employed for the removal of phosphate (Wang et al., 2014; Yang et al., 2014). However, most of these adsorbents were costly or had undesirable adsorption capabilities. Some cost-effective and high-efficient adsorbents should be developed and applied.

Biochar, a carbon-rich by-product derived by pyrolyzing biomass (e.g., agricultural waste, wood chips, algae, manure, sewage sludge, etc) under oxygen-limited conditions, has been widely used as a low-cost adsorbents to adsorb heavy metals, nutrients (ammonium, nitrate and phosphate), dyes and organic contaminants from aqueous solutions (Wang et al., 2015; Zhou et al., 2014; Cao et al., 2011). However, due to negatively charged surface and contained little anion exchange capacity, biochar generally has no capability to adsorb anions, such as arsenic and phosphate (Hollister et al., 2013; Yao et al., 2012). In order to enhance biochar adsorption ability of anions, several strategies have been developed to improve its physicochemical properties (Chen et al., 2011; Zhang et al., 2013). A novel MgO-biochar nanocomposites originated from pyrolyzing the mixture of agricultural wastes and MgCl₂ solution had an excellent adsorption ability to phosphate and nitrate (Zhang et al., 2012a). The maximum adsorption capacity based on Langmuir isotherm model were reached about 100 mg g⁻¹ (computed in P) and 20 mg g⁻¹, which were much higher than that of most commercial adsorbents. Zhou et al. (2014) demonstrated that biochar impregnated by some chitosan and zerovalent iron could efficiently remove arsenic and phosphate compared to pristine biochar.

Lanthanum (La) has attracted significantly attentions in wastewater treatment technology due to its high phosphate affinity and its interaction in adsorbents for enhancing anions adsorption (Liu et al., 2011; Zhang et al., 2010; Ou et al., 2007; Tian et al., 2009; Wu et al., 2007). It was confirmed that the impregnation of La on activated carbon fiber or mesoporous silicate materials can facilitate the La homogeneous distribution and enhance the adsorption capability to phosphate (Zhang et al., 2010; Liu et al., 2013). During the synthesis process of La-composite, activated carbon fiber and mesoporous silicate materials were regarded as support materials to disperse La due to their large surface area and abundant pores. The popular support materials at present were rather cost. La related adsorbent would be widely applied if the cheap support materials are developed.

Based on the cost-effective and large surface area of biochar and the high phosphate affinity of La, an adsorbent (La-BC) would be prepared from La and biochar by a set of treatments in order to achieving high removal efficiency of phosphate. The characteristics of the adsorbent (La-BC) such as the ash content, ultimate analysis, zeta potential, BET surface area, SEM-EDS, XPS, FT-IR were identified for further understanding of the inherent adsorption mechanism between biochars and phosphate. The influences of phosphate initial concentration, contact time, solution pH, and temperature on the adsorption process were examined, together with the adsorption isotherms, kinetics and thermodynamics.

2. Materials and methods

2.1. Materials

Oak biochar originated from the pyrolysis of oak chips under 500 °C was used as the support materials for La, its detailed preparation method was consistent with our previous study (Wang et al., 2015). Lanthanum nitrate (La(NO₃)₃), sodium hydrate (NaOH), nitric acid (HNO₃), potassium dihydrogen phosphate (K₂HPO₄), ascorbic acid and molybdenum acid ammonium with analytical

grade were purchased from KeLong Co. Ltd.

2.2. Preparation of adsorbent

NaOH with a concentration of 1 M was added into 1000 mL La contained solution (0.1 M) with magnetic stirring for adjusting pH to 10 to get La precipitation. After ultrasonic dispersing 2 min, 30 g of oak biochar was immersed and mixed in La precipitation under 10 min ultrasonic time and 120 W ultrasonic power. The biochar composites was obtained after vacuum-filtered, washed with deionized water, dried at 105 °C for 6 h and calcined by tube furnace (OTL 1200, Nanjing NanDa Instrument Co., Ltd., China) at 400 °C which was labeled as La-BC while pristine biochar was labeled as CK-BC.

2.3. Characterization of the adsorbent

The pH of biochar was performed by a pH meter (PHS-3C, LeiCi Instruments Co., Ltd., China) with a ratio of 1:20 (m/v) for biochar to deionized water. Ash content was determined according to the standard methods in ASTM D 1762-84. Ultimate analysis including C, H, and N was carried out at an Elemental Analyzer (EA112, Thermo Finnigan, USA). Specific surface area (SA) was measured using N₂ sorption at 77 K on a NOVA-2000E analyzer (Quantachrome Instruments, USA) and the calculation was based on the Brunauer–Emmentt–Teller (BET) method. Surface morphologies and basic elements were observed with a scanning electron microscopy (SEM) equipped with an energy-dispersive X-ray spectroscopy (EDS) (SU1510, Hitachi, Japan). Surface elemental composition was also carried out on an X-ray photoelectron spectroscopy (XPS) (Amicus, Shimadzu, Japan). Functional groups on the surface of biochar were analyzed by a Fourier Transform Infrared Spectrometer (FT-IR) (Nicolet 6700, Thermo Fisher Scientific, USA). The vibrations corresponding to the wavenumbers range from 400 to 4000 cm⁻¹ were collected with the resolution of 4 cm⁻¹. The zeta potential (ZP) was measured by a laser particle size analyzer (Zetasizer NANO ZS90, Malvin, UK). The crystalline materials and their crystallographic structures of biochar were checked by X-ray diffraction (XRD) using a computer-controlled X-ray diffractometer equipped with a stepping motor and graphite crystal monochromator (I-2, Nicolet, Madison, WI, USA).

2.4. Adsorption experiments

A phosphate stock solution with a concentration of 1000 mg L⁻¹ (computed in P) was prepared by dissolving a calculated quantity of K₂HPO₄ in deionized water. The required initial concentration of phosphate solution was prepared by diluting the stock solution. The pH of the solution was adjusted using 0.1 M NaOH or 0.1 M HNO₃. 0.1 g biochar was added into 150 mL screw-flasks filled with 50 mL phosphate solution then sealed and shaken at 120 r min⁻¹ for 24 h. At the end of adsorption process, the solutions were filtered through 0.22 μm membrane syringe filters and determined the concentration of the phosphate using the ascorbic acid method (Zhang et al., 2012b). All experiments were carried out for three times and more runs would be operated if the experimental data was unreasonably scattered.

The equilibrium experiments were carried out as aforementioned procedures in various initial concentrations from 1 mg L⁻¹ to 400 mg L⁻¹. As for the contact time experiments, 1 mL solution was taken out at given time intervals by 10 min–48 h. The impact of pH on the phosphate adsorption was conducted with the pH range from 3.0 to 9.0. The temperature experiments were studied at various temperature corresponding to 15, 25, 35, and 45 °C.

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