



Simultaneous removal of lead and phenol contamination from water by nitrogen-functionalized magnetic ordered mesoporous carbon



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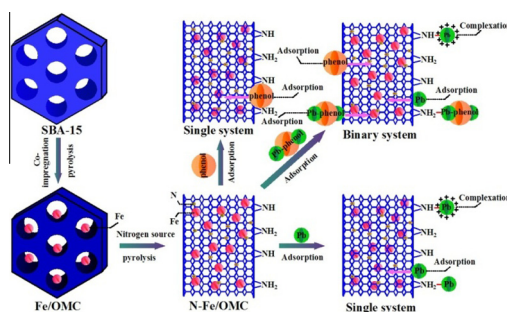
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HIGHLIGHTS

- N-Fe/OMC was applied as a novel adsorbent to remove Pb(II) and phenol.
- The high capacities and fast rates for Pb(II) and phenol adsorption were observed.
- Partial complexation and competition between Pb(II) and phenol were verified.
- Excellent magnetic separation performance and effective reuse.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 4 July 2014

Received in revised form 19 August 2014

Accepted 20 August 2014

Available online 27 August 2014

Keywords:

Lead

Phenol

Adsorption

Nitrogen-functionalized

Magnetic ordered mesoporous carbon

ABSTRACT

In this study, a novel nitrogen-functionalized magnetic ordered mesoporous carbon (N-Fe/OMC) with uniform pore size (3.8 nm) and excellent magnetic property (8.46 emu/g) was fabricated through simple impregnation then polymerization and calcination. The resultant adsorbent exhibited more preferential sorption toward Pb(II) and phenol than simple equivalent mixture of magnetic ordered mesoporous carbon (Fe/OMC) and pristine ordered mesoporous carbon (OMC). Binary adsorption showed that the coexistence of Pb(II) and phenol at low concentrations would slightly accelerate their jointly adsorption because of partial complexation between Pb(II) and phenol. While at high concentrations, Pb(II) adsorption would be inhibited in relation to the competition from phenol, but phenol adsorption was scarcely affected due to the directly phenol molecular adsorption pattern. Pb(II) adsorption was more suitable in alkaline solution and affected by ionic strength on account of electrostatic interaction, whereas phenol adsorption was better in neutral pH and hardly interfered by ionic strength as the adsorption was a physical combination process. Thermodynamics indicated that the uptakes of Pb(II) and phenol were endothermic and exothermic processes, respectively. Moreover, N-Fe/OMC could be regenerated effectively and recycled by using dilute NaOH and acetone solutions. These superior properties demonstrate that N-Fe/OMC is attractive for practical applications in treatment of water contamination by Pb(II) and phenol.

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1. Introduction

Heavy metals and phenolic compounds are common pollutants in surface water and ground water, and pose a potential threat to the flora and fauna because of their toxicity, carcinogenicity and accumulation in the food chains [1,2]. For example, lead ions (Pb(II)) may cause stomachache, dysphoria, even cancer in human beings, and phenol also may contribute to nausea and skin corrosion. Additionally, heavy metals and phenolic compounds may occur together in contaminated water and soil, giving rise to more serious damage to the environment with their combined toxicity and relative mobility [3,4]. Such phenomenon commonly exists in printing, agricultural irrigation and plastic manufacturing. Consequently, finding effective ways to treat these pollutants, especially for their simultaneous removal has aroused worldwide interest recently. Among all possible techniques, adsorption is one of the most attractive approaches for heavy metals and phenolic compounds removal due to its high efficiency, simple and safe treating processes, versatility for different water systems and low cost [5]. Many adsorbents, including active carbons [6], chitosans [7], resins [8] and carbon nanotubes [4] have been applied for removal of heavy metals or/and phenolic compounds. However, there are some defects, such as low efficiency, bad stability and difficult separation, limiting their practical application.

Compared to traditional adsorbents, ordered mesoporous carbons (OMCs) with large surface area and pore volume, unique pore size, and excellent physicochemical and thermal stability are promising candidates in pollutant removal [9–11]. Whereas the adsorption behaviors of OMCs for specific pollutants, such as heavy metals and low molecular weight organic matters are often hindered due to the overlarge pore sizes and ordinary hydrophilicities. Previous studies demonstrated that transition elements, for example, metal nanoparticles of Co, Fe and Pt [12–14] or nonmetal atoms of N and P [15,16] can be utilized as catalysts to accomplish primary amorphous OMCs graphitization, strengthening their excellent acid-base stability, catalytic and hydrophilic properties. Besides, these transition elements introduction sometimes brings about some magical results. For example, introduction of magnetic nanoparticle of Fe can greatly improve their practical application in separation and reutilization [17], and introduction of nonmetal heteroatom of N element will equip OMCs with $-NH_2$ and $-NH$ groups [18], boosting the chelation with heavy metals or organics.

According to the above considerations, it is supposed to triply improve the features of OMCs by incorporating with magnetic nanoparticles, and subsequently introducing nonmetal atoms to enhance and extend their physicochemical properties. Recently, similar functional carbon materials have been reported to be applied in the aspects of catalysis and electrochemistry [13,19]. But their outstanding abilities in adsorption and separation scarcely have been mentioned.

In this paper, a novel functional adsorbent, nitrogen-functionalized magnetic ordered mesoporous carbon (N-Fe/OMC) was prepared by adopting two-step method that primarily synthesizes magnetic ordered mesoporous carbon (Fe/OMC), and then followed by doping with nitrogen atoms using aniline as a nitrogen precursor and ammonium persulfate as an oxidant. After comprehensive characterization of its physicochemical properties, N-Fe/OMC is firstly used to investigate the removal behaviors of Pb(II) and phenol from wastewater. The single and binary adsorption systems were applied to investigate the distinct interaction of Pb(II) and phenol with N-Fe/OMC, and the complexation and competition between the two target pollutants, respectively. The effects of pH, ionic strength and temperature were studied, and the adsorption isotherms, kinetics and thermodynamics were proposed to analyze the adsorption mechanisms. The actual application was

investigated by combining with the regeneration of N-Fe/OMC and its application in different water samples.

2. Materials and methods

2.1. Preparation of Fe/OMC

Mesoporous silicon templates (SBA-15) were prepared by hydrothermal synthesis method [20]. Fe/OMC was synthesized by following a co-impregnation method with slight alterations [21]. Typically, 1.0 g as-synthesized SBA-15 was impregnated with 10 mL multi-component alcohol solution containing 0.721 g $Fe(NO_3)_3 \cdot 9H_2O$ and 0.05 g oxalic acid, and then 2.0 mL furfuryl alcohol was added dropwise into the above mixture. After that, the mixture thus prepared was heated at 90 °C for 10 h in air and calcinated at 900 °C for 2 h under flowing nitrogen atmosphere. After dissolving the silica framework with 2.0 mol/L NaOH solution at 90 °C, the resultant solid was filtered, washed, dried and then stored in a nitrogen-filled glovebox until required. For comparison, OMC was also prepared without iron.

2.2. Preparation of N-Fe/OMC

N-Fe/OMC was synthesized as follows: 1.0 g Fe/OMC nanocomposite was first dissolved into 300 mL 0.1 mol/L HCl under 3.0 h mechanically stirring, and subsequently 1.0 mL aniline was in situ polymerized using 2.5 g ammonium persulfate as the initiator and HCl as the catalyst. The polymerization was conducted in an ice bath (<5 °C) for 24 h. Then the samples were repeatedly washed with 50 wt% ethanol solution and dried at 60 °C overnight. After carbonization the immature nanocomposites at 950 °C for 3 h under nitrogen atmosphere, the synthetic N-Fe/OMC was acquired.

2.3. Characterization

Transmission electron microscopy (TEM, JEOL JEM-1230) and Scanning electron microscope (SEM, JEOL JSM-6700) images were used to investigate the morphology and structure of N-Fe/OMC. The Energy dispersive X-ray (EDX) analysis in the SEM image was applied to detect the element composition of the resultant sample. X-ray diffraction (Rigaku D/max-II B) and FTIR spectrometer (Nicolet NEXUS 670) were applied to analyze the likely existence of compositions and functional groups. Nitrogen adsorption measurements at 77 K were performed using an ASAP 2020 volumetric adsorption analyzer. Raman spectroscopy was mounted by using a LabRam HR800 Raman spectrometry. X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific, UK) was proposed to analyze the surface elemental composition. Zetasizer Nano (ZEN3600, Malvern) and vibrating sample magnetometer (VSM National Institute of Metrology) were used to determine the zeta potential and magnetization of the samples, respectively.

2.4. Batch adsorption experiments

Adsorption of Pb(II) and/or phenol on N-Fe/OMC were performed in 50-mL stoppered conical flasks undergoing shaking at 150 rpm in a water bath shaker. 0.1 mol/L HCl or NaOH was used to adjust the solution pH, and NaCl was used as a function of ionic strength. Before each experiment, 5 mg of specific mesoporous adsorbent was dispersed into 10 mL of homogeneous aqueous solution containing certain amount of NaCl. At each preselected time point, the solid and liquid phases were separated using an external magnet for magnetic OMC or centrifugation for OMC, and then the supernatant was filtered through 0.45- μ m membrane for measurement of the residual pollutant concentrations.

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