



Biomass-derived multifunctional magnetite carbon aerogel nanocomposites for recyclable sequestration of ionizable aromatic organic pollutants

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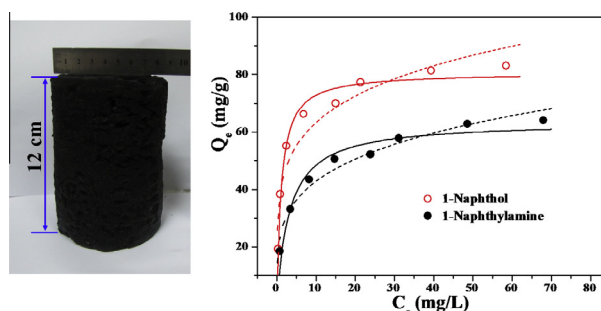
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HIGHLIGHTS

- A novel magnetite carbon aerogel (MCA) was synthesized by using crude biomass and watermelon.
- The porous MCA has large surface area of $323.8 \text{ m}^2 \text{ g}^{-1}$ and can be easily lift by a magnet.
- The MCA exhibited excellent adsorption capacity toward both 1-naphthol and 1-naphthylamine.
- The MCA can be easily regenerated by a simple Fenton reaction.

GRAPHICAL ABSTRACT



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ABSTRACT

In this article, a novel composite material based on biomass-derived carbon aerogel and iron oxide were prepared by a facile hydrothermal treatment of watermelon and stepped by incorporating iron oxide nanoparticles into the networks of the carbon aerogel. The as prepared magnetite carbon aerogel (MCA) was characterized by Fourier transformed infrared (FTIR), field emission scanning electron microscopic (FE-SEM) and X-ray diffraction (XRD). The MCA has high surface area of $323.8 \text{ m}^2 \text{ g}^{-1}$ and can be easily lift by a magnet. The multifunctional MCA was further applied as adsorbents for the removal of ionizable aromatic organic pollutants from aqueous solution. 1-Naphthol and 1-naphthylamine were used as model of the ionizable organic pollutants. The MCA exhibited excellent adsorption capacity toward both 1-naphthol and 1-naphthylamine and can be easily separated from aqueous solution by an external magnet. More importantly the adsorbed MCA can be easily regenerated by a simple Fenton reaction and still retained high adsorption capacity after cycles of reuse. The results imply that the MCA can be potential candidate as cost-effective adsorbents for the preconcentration and degradation of organic pollutants from aqueous solutions in environmental pollution cleanup.

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1. Introduction

Water contamination by aromatic compounds is recognized as a global environmental problem and has drawn growing public

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concerns. Phenols and anilines are large class of ionizable aromatic compounds which are widely found in the effluents from the pharmaceuticals, petrochemicals, dyestuffs, pesticides and other industries [1]. Both phenols and anilines are classified as priority contaminants due to their toxicity to organisms at low concentrations. Besides, they are relatively high solubility in water and can easily transport in natural environment which may boost human

risk for cancer and acute toxicity by contact and uptake. Therefore, the removal of aromatic compounds from the discharged effluent has become a major focus of environmental protection and research. Many techniques including adsorption [1,2], biological treatment [3,4], chemical oxidation [3] and photochemical degradation [5] have been applied for the remove of aromatic pollutants from aqueous solution. Among these techniques, adsorption is one of the most effective technologies due to its operational simplicity, effectiveness, low energy requirements and low cost. In the past decade, the development of nanoscience and nanotechnology open up new avenues for the remediation of environmental problems. Nanomaterials based adsorbents have been studied extensively and showed much faster rates and higher efficiency toward pollutants as compared with the conventional adsorbents [6–9]. Various carbonaceous nanomaterials have been adopted as adsorbents for the removal of organic pollutants such as mesoporous carbon [10], carbon nanotubes [1,11], graphene [8] and carbon aerogels [12]. Among them, carbon aerogels were considered as one of the cost-effective carbon materials for water decontamination due to their low cost and excellent capability.

Aerogels are unique three-dimensional (3D) solid networks with open pores and majority of the volume is air [13,14]. Carbon aerogels are large class of aerogels which have recently triggered many research activities due to their multifunctional properties such as high surface area, porous structure, low mass densities and high electrical conductivity [15,16]. Because of these special chemical and textural characteristics, carbon aerogels possess promising applications in adsorbents, catalysis and supercapacitors [17]. In recent years, growing attentions have been paid to the design and fabrication of carbon aerogels based composite materials with multifunctional properties for environmental applications. For example, TiO_2 /carbon aerogels composites have been synthesized and used as electrosorption–photocatalysis synergistic electrode for the degradation of organic pollutants in aqueous solutions [18]. Wang and co-workers prepared ferrite–carbon aerogel composites and applied as Fenton catalysis for the oxidation degradation of metalaxyl [14]. Conventional carbon aerogels are prepared by the pyrolysis of organic aerogels such as resorcinol and formaldehyde polymer aerogels. However, these methods are usually associated with problems of multiple step process and using expensive and poisonous chemical reagents. Thus, to develop a simple and environmental friendliness approach to carbon aerogels for environmental applications is still challenging.

In this work a green and template free route was developed for the synthesis of low-cost carbon aerogels and further modified with iron oxide nanoparticles. The obtained magnetite carbon aerogel (MCA) composite material was applied as adsorbent for the efficient removal of ionizable aromatic compounds from aqueous solution. 1-Naphthol and 1-naphthylamine are used as model compounds of the harmful and water soluble aromatic organic pollutants, which are widely used as industrial intermediates [19]. The adsorption properties of 1-naphthol and 1-naphthylamine on MCA are investigated and the mechanisms were also discussed. To the best of our knowledge, it is the first example of using carbon aerogels based nanocomposites for the efficient removal of toxic ionizable aromatic compounds from aqueous solution. And the results showed that MCA can be suitable candidate as cost-effective adsorbents for practical application.

2. Experimental

2.1. Chemicals

Chemicals including ferric chloride (anhydrous, FeCl_3), ferrous sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), ammonium hydroxide (25%,

$\text{NH}_3 \cdot \text{H}_2\text{O}$), trisodium citrate (Na_3Cit), sodium hydroxide (NaOH) and hydrochloric acid (38%, HCl) were purchased from Sinopharm Chemical Reagent Co., Ltd. (China). 1-Naphthol and 1-naphthylamine were purchased from Sigma–Aldrich (St Louis, MO). All above chemicals were used as received without further purification. Milli-Q ultrapure water ($18.2 \text{ M}\Omega \text{ cm}^{-1}$) was used for all the experiments.

2.2. Sample preparation

The original carbonaceous monolith was prepared by a well-established hydrothermal carbonization method by using crude biomass, watermelon, as the carbon source. Watermelon was first cut and put in into a Teflon-lined stainless-steel autoclave and hydrothermal treated at 180°C for 12 h. After that, the 3D black monolith was taken out and washed with Milli-Q water several times. For preparing magnetite carbon aerogel (MCA), Fe_3O_4 nanoparticles were first loaded in the above carbonaceous monolith by using a simple solution process [20]. Briefly, a piece of carbonaceous monolith with dimension of about $2 \times 3 \times 3 \text{ cm}^3$ was fully immersed in 20 mL aqueous solution of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (1.25 g, 4.5 mmol) and FeCl_3 (1.30 g, 8 mmol) for 15 min under N_2 atmosphere. The mixture was heated to 80°C under N_2 protection and 10 mL of 30% ammonia solution was quickly added into the mixture. After that, the mixture was kept at 80°C for 30 min and 3.0 g of trisodium citrate was added to the solution while the temperature rising to 95°C . The obtained Fe_3O_4 loaded carbon monolith was rinsed with Milli-Q water and dried in a vacuum oven at 70°C over night. The magnetite carbon aerogel (MCA) was obtained by calcination of the above Fe_3O_4 loaded carbon monolith in nitrogen atmosphere at 550°C for 4 h.

2.3. Characterization and analysis

Transmission electron microscopy (TEM) (JEOL-2010) and field emission scanning electron microscopic (FE-SEM) (JEOL JSM-6330F) were applied to observe the microstructure of the original carbon monolith and MCA. X-ray diffraction (XRD) pattern was performed on a Philips X'Pert X-ray diffractometer (Cu $\text{K}\alpha$ source ($\lambda = 1.54178 \text{ \AA}$)) in the range of $2\theta = 5\text{--}65^\circ$. The N_2 adsorption–desorption curve of MCA was measured by a Micromeritics ASAP 2010 system at 77 K utilizing Barrett–Emmett–Teller (BET) methods to calculate the specific surface area. The pore size distribution of the MCA was calculated by applying the Barrett–Joyner–Halenda (BJH) model by using the desorption branch of the N_2 adsorption–desorption isotherm. Magnetic measurement of the MCA was carried out on a (Quantum Design MPMS XL) magnetometer at 300 K with an applied magnetic field of 20 KOe. The UV–vis measurements were performed on a UV-3000 spectrophotometer (Japan).

2.4. Adsorption experiments

The batch adsorption experiments of 1-naphthol and 1-naphthylamine on MCA were carried out at $\text{pH} = 6.0$ in glass vials. Before adsorption, calibration curves of 1-naphthol and 1-naphthylamine was obtained from the UV–vis spectra of the standard 1-naphthol and 1-naphthylamine solutions ($5\text{--}60 \text{ mg L}^{-1}$) at $\text{pH} 6$, respectively. The stock suspensions of MCA, 1-naphthol or 1-naphthylamine solutions were added in the glass vials to achieve the desired concentrations of different components. The desired pH of the suspension in each vial was adjusted by adding negligible volumes of 0.01 or 0.1 M NaOH or HCl with a pH meter. After the suspensions were oscillated for 48 h, the solid and liquid phases were separated by centrifugation at 10,000 rpm for 5 min. The concentration of the aromatic organic compounds in the supernatant was determined by UV–vis spectrophotometer. The adsorbed

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