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Synthesis of magnetic Fe₃O₄/activated carbon nanocomposites with high surface area as recoverable adsorbents

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ABSTRACT

The magnetic Fe₃O₄/activated carbon nanocomposites with high surface area were synthesized as recoverable adsorbents by chemical binding of Fe₃O₄ nanoparticles on activated carbon (AC) powders. The component AC and Fe₃O₄ in this nanocomposite possesses amorphous non-graphitic structure and cubic crystal structure, respectively. All composite samples presented superparamagnetic properties. The saturation magnetization of Fe₃O₄/AC nanocomposites was significantly lower than that of bare Fe₃O₄ particles, indicating that Fe₃O₄ particles were truly attached on AC surface. The microstructure image indicated that the Fe₃O₄ particles were uniformly dispersed on AC surface and thus maintained high specific surface area. The adsorption capacity of methyl orange (MO) at 30 °C slightly decreased from 384 mg/g on AC powders to 324 mg/g on Fe₃O₄/AC nanocomposites, which was reduced by 15% after magnetic fabrication. It was found that MO adsorption on Fe₃O₄/AC nanocomposites followed the pseudo-second order kinetic model and the isotherms could be described by the Langmuir model. The easy recovery of magnetic adsorbents from aqueous solution demonstrated their application potential to remove toxic pollutants in water and wastewater treatment.

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1. Introduction

Industrial activities increase the convenience in the human life since the 18th century. Inevitably, lots of industrial wastewaters containing organic compounds such as dyes from printing, paper-making, textile, and food industries were produced and discharged into the environments. Most of industrial organic pollutants are colored, toxic, carcinogenic, or teratogenic. However, most of colored substances such as dyes are very hard to remove naturally. Textile and dyeing effluents hugely affect human health and endanger thousands of animals and plants. People try to remove dyes from wastewater, but it is very difficult when their concentrations in the effluents are less than 1 mg/L [1,2]. Thus, efficient treatment of textile effluents becomes an important task. Several methods such as adsorption [3,4], centrifugation [5,6], ultrafiltration [7,8], ion exchange [9,10], and electrochemistry [11,12] are usually used to separate dyes from aqueous solutions. Among these methods,

adsorption is powerful and comparatively cheap, and therefore most widely applied for this purpose [13–17].

Activated carbon (AC) is the most common materials used to remove different organic pollutants and has been widely applied to industrial wastewater treatment in practice [18]. AC has a high specific surface area and various functional groups to adsorb colored dyes, particularly in the powdered form [19,20]. However, it is still hard to remove and recover the used AC powders from water, resulting in secondary pollution [21]. Because there is no polarity on AC powders, they cannot be controlled by electric and/or magnetic fields. Centrifugation [5] and filtration [7] are usually adopted to separate the used AC powders from liquid solutions; however, both methods are costly. If AC has the polarity on its surface, we may apply electric and/or magnetic fields to trap, restore, and recycle to avoid secondary pollution after adsorption processes. The idea of combining AC powders and magnetic nanoparticles to adsorb organic pollutants is hence considered [15,22–31], which provides a low-cost, simple, and quick way for practical applications. However, the combination of AC powders and magnetic nanoparticles will create another problem; that is, most pore space within AC powders may be occupied by

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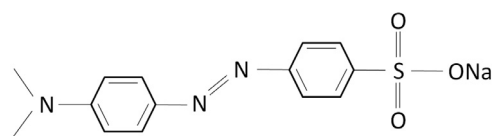


Fig. 1. Chemical structure of methyl orange (MO).

magnetic nanoparticles, leading to a sharp decrease in surface area [29–31]. For example, Ma et al. [31] prepared magnetic ACs from their powdered forms by chemical co-precipitation. The BET surface areas and total pore volumes of the samples changed from 1106 m²/g and 0.97 cm³/g to 254 m²/g and 0.29 cm³/g, respectively, after magnetic fabrication. Although AC powders are easily recovered after connecting with magnetic materials, the loss of massive pore space is accompanied by a significant decrease of adsorption ability for organic pollutants. This is particularly the case when magnetic nanoparticles themselves do not show certain adsorption ability for the pollutants of interest [29–31].

To remove organic pollutants from wastewater more efficiently, a large amount of magnetic AC powders is required. Consequently, it is desired to develop a facile way to prepare magnetic ACs with relatively high surface area. It was reported that the loss of surface area of graphene is minimized when Fe₃O₄ nanoparticles are attached because such nanoparticles will be uniformly distributed on graphene nanosheets [32]. These implied that the decrease of surface area of AC powders could be minimized if Fe₃O₄ nanoparticles were uniformly distributed on AC powder surface. Therefore, we systematically adjusted the weight ratio of FeCl₃ to NaOH used (F/N ratio) to modify the size and distribution of Fe₃O₄ nanoparticles on AC powder surface in this study. The polyol-mediated solvothermal reduction method [32,33] was adopted to connect AC powders and magnetic Fe₃O₄ nanoparticles (the so-called Fe₃O₄/AC nanocomposites), which were used to remove the model pollutant methyl orange (MO) from aqueous solution.

2. Experimental

2.1. Materials

AC powders with a size ranging from 20 to 40 μm were purchased from Showa Chemical Co. (Gyoda, Saitama, Japan). Ferric chloride (FeCl₃) is the precursor of Fe₃O₄, supplied by Riedel-de Haën (Morristown, NJ, USA). Sodium hydroxide and diethylene glycol (DEG) were obtained from Macron Fine Chemical Co. (Center Valley, PA, USA) and Sigma-Aldrich Co. (St. Louis, MO, USA), respectively. Methyl orange (MO, C₁₄H₁₄N₃NaO₃S) purchased from Acros Organics (Geel, Belgium) was chosen as the model adsorbate because it is one of azo dyes that are those constituting more than 50% used in industrial applications [34]. Fig. 1 shows the structure of MO. The commercially-available magnetic Fe₃O₄ nanoparticles with a size of 20–40 nm were purchased from the Nanostructured and Amorphous Materials Inc. (Houston, TX, USA). Their magnetic properties and adsorption performance were also measured and compared in this study.

2.2. Synthesis of Fe₃O₄/AC nanocomposites

Fig. 2 presented the synthetic process of magnetic Fe₃O₄/AC nanocomposites. First at all, NaOH (*y* g) was added into DEG (100 mL) at 120 °C for 1 h and then cooled down to 80 °C for 30 min (the so-called NaOH/DEG stock solution). Meanwhile, AC powders were dispersed in DEG (40 mL) by sonicating for 1 h as shown in step (a), which was the so-called AC/DEG solution. The AC surface had -OH and -COOH groups originated from its activation process. When *x* g of FeCl₃ was added into the AC/DEG

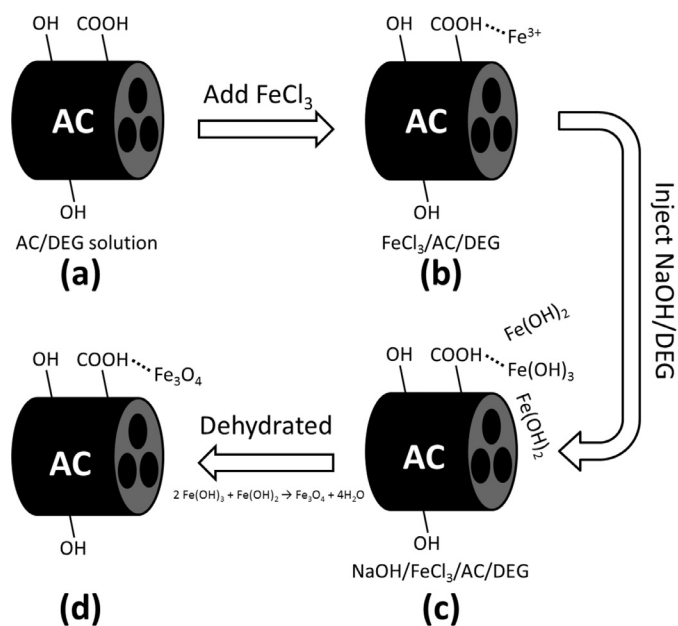
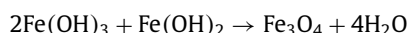


Fig. 2. Synthetic process of Fe₃O₄/AC nanocomposites.

solution at 220 °C for 30 min, the Fe³⁺ ions were coordinated to the -COOH groups of AC as shown in step (b). At step (c), NaOH/DEG stock solution was rapidly injected into FeCl₃/AC/DEG solution, and the reaction took place at 220 °C for 1 h. The Fe³⁺ ions on AC were hydrolyzed to Fe(OH)₃. Some Fe(OH)₃ would be transformed to Fe(OH)₂ in the solution. Then, Fe(OH)₃ was conjugated with Fe(OH)₂ and was dehydrated to form Fe₃O₄ and H₂O according to the reaction:



All reactions occurred under N₂ atmosphere. At final step (d), the Fe₃O₄/AC nanocomposites were produced. The wet products were dried at 60 °C in a vacuum oven for 24 h. In this work, Fe₃O₄/AC nanocomposite particles were prepared with different parameters, where F_{*x*}N_{*y*} denote *x* g of FeCl₃ and *y* g of NaOH [32], and F/N ratio means the value of (*x*/*y*).

2.3. Characteristic of AC powders and Fe₃O₄/AC nanocomposite

The magnetic hysteresis loops of bare AC powders and Fe₃O₄/AC nanocomposites were measured using a vibrating sample magnetometer, VSM (DMS model 1660, ADE Technologies Inc., MA, USA) in an applied field range of ±10 kOe. Crystal structures of the powders were analyzed by D2 PHASER X-ray diffractometer, XRD (Bruker, Germany) with the scanning region of 2θ from 10° to 70°. The surface morphology of bare AC powders and the prepared Fe₃O₄/AC nanocomposites were determined by the JEOL scanning electron microscope, SEM (JSM-7800F Prime, Tokyo, Japan). The selected area electron diffractions (SAED) and microstructures of the powders were observed by a JEOL transmission electron microscope, TEM (JSM-2010, Tokyo, Japan) with accelerating voltage at 200 keV. The BET surface areas of the samples were measured from N₂ adsorption-desorption isotherms at -196 °C using a porosimeter (ASAP2020, Micromeritics Instrument Co., GA, USA). Prior to measurement, sample was dried overnight in an oven at 130 °C and then placed in the sample tube. After that, the tube was heated to 230 °C and evacuated for 4 h until the pressure was less than 1.3 × 10⁻⁷ bar. The total pore volumes of bare AC powders and Fe₃O₄/AC nanocomposites were determined from N₂ adsorption isotherms, according to the manufacturer's software (based on the Kelvin equation), and the pore size distribution

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