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## Synthesis of carbon-coated magnetic nanocomposite ( $\text{Fe}_3\text{O}_4@\text{C}$ ) and its application for sulfonamide antibiotics removal from water

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### ABSTRACT

The occurrence of antibiotics in the environment has recently raised serious concerns regarding their potential threat to human health and aquatic ecosystem. A new magnetic nanocomposite,  $\text{Fe}_3\text{O}_4@\text{C}$  ( $\text{Fe}_3\text{O}_4$  coated with carbon), was synthesized, characterized, and then applied to remove five commonly-used sulfonamides (SAs) from water. Due to its combinational merits of the outer functionalized carbon shell and the inner magnetite core,  $\text{Fe}_3\text{O}_4@\text{C}$  exhibited a high adsorption affinity for selected SAs and a fast magnetic separability. The adsorption kinetics of SAs on  $\text{Fe}_3\text{O}_4@\text{C}$  could be expressed by the pseudo second-order model. The adsorption isotherms were fitted well with the Dual-mode model, revealing that the adsorption process consisted of an initial partitioning stage and a subsequent hole-filling stage. Solution pH exerted a strong impact on the adsorption process with the maximum removal efficiencies (74% to 96%) obtained at pH 4.8 for all selected SAs. Electrostatic force and hydrogen bonding were two major driving forces for adsorption, and electron-donor-acceptor interactions may also make a certain contribution. Because the synthesized  $\text{Fe}_3\text{O}_4@\text{C}$  showed comprehensive advantages of high adsorptivity, fast magnetic separability, and prominent reusability, it has potential applications in water treatment.

## Introduction

As a class of important synthetic sulfanilamide derivatives, sulfonamides (SAs) are widely used for treatment of bacterial, protozoal and fungal infections in human therapy, livestock production, and aquaculture. In China, the total consumption of antibiotics reached over 120,000 tons in 2009 (Guo, 2009). Residual antibiotics discharged from municipal wastewater treatment plants and confined animal feeding operations have been detected worldwide in surface, ground, and even drinking waters (Boxall et al., 2003; Nikolaou et al., 2007; Schwab et al., 2005).

Long-term exposure to low levels of antibiotics may induce chronic allergic reactions, toxic effects and potential development of antibiotic-resistant bacteria (Boxall et al., 2003), so the occurrence of antibiotics in aquatic environments has raised an increasing concern in recent years. Consequently, there is an urgent need to develop efficient and cost-effective technologies for the removal of antibiotics from water.

A number of oxidation technologies utilizing potassium permanganate, ozone, Fenton's reagent, and chlorine have been attempted to decompose antibiotics in water (Ben et al., 2009; Chamberlain and Adams, 2006; Hu et al., 2010; Gómez-Pacheco et al., 2011; Nasuhoglu et al., 2012). Comparatively, adsorption is a conventional but effective technology for water treatment due to its relatively low

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cost, easy application, and no production of harmful by-products. Some adsorbents, such as activated carbon, carbon nanotubes and sludge-derived materials, have been used to remove antibiotics from water (Choi et al., 2008; Ji et al., 2009; Ocampo-Pérez et al., 2012); however, new adsorbents, which have a high adsorption capacity and can be rapidly separated and easily regenerated, are still needed.

Functionalized magnetic nanocomposites have distinct advantages over conventional adsorbents because of their selective adsorptivity, strong magnetic responsiveness, favourable water dispersibility, and benign biocompatibility. Magnetic nanocomposites have been applied in many fields including magnetic resonance imaging, bioscience, analytical chemistry, and pollutants removal (Arsalani et al., 2010; Erathodiyil and Ying, 2011; Guo et al., 2009; Zhang et al., 2010). Carbon-coated magnetic nanocomposite, Fe<sub>3</sub>O<sub>4</sub>@C, has a special structure with functionalized carbon as an outer shell and magnetite as an inner core. The polysaccharide surface layer contains abundant functional sites, such as carboxylic, formyl and hydroxyl groups; moreover, this dense outer layer can protect well the inner Fe<sub>3</sub>O<sub>4</sub> microspheres from being dissolved in acidic solutions (Wang et al., 2010). Previous studies about Fe<sub>3</sub>O<sub>4</sub>@C nanocomposite focused on its synthetic methods (Wang et al., 2006), while few have addressed its application in water treatment.

The present study aimed to investigate the adsorptive removal of five representative SAs from water using Fe<sub>3</sub>O<sub>4</sub>@C. Regular-shaped Fe<sub>3</sub>O<sub>4</sub>@C nanocomposite was synthesized in laboratory and comprehensively characterized. Thereafter, the adsorption kinetics of SAs on Fe<sub>3</sub>O<sub>4</sub>@C was determined through batch experiments, and the impact factors including the initial concentration of SAs, temperature and solution pH were examined in detail. Based on the surface properties of the synthesized Fe<sub>3</sub>O<sub>4</sub>@C and the functional groups of the studied SAs, possible adsorption mechanisms were proposed.

## 1 Materials and methods

### 1.1 Chemicals

Five commonly-used SAs, including sulfathiazole (STZ, > 98%), sulfamethoxazole (SMX), sulfamethizole (SML) and sulfadimethoxine (SDM, > 99%) were purchased from Sigma-Aldrich (St. Louis, MO, USA), and sulfamethazine (SMN) from Acros (Fair Lawn, NJ, USA). The chemical structures and major physicochemical properties of the selected SAs are provided in **Table S1** (Supporting materials). The stock solution of each SA was prepared individually at a concentration of 100 mg/L in ultrapure water, which was produced by a Milli-Q system (Advantage A10, Millipore, Billerica, MA) with a resistivity of

at least 18.2 MΩ-cm. The organic solvents, methanol and acetonitrile, were of high performance liquid chromatography grade. Other chemicals were purchased from Beijing Chemical Reagents Company (Beijing, China) with at least analytical grade.

### 1.2 Preparation of Fe<sub>3</sub>O<sub>4</sub>@C

Fe<sub>3</sub>O<sub>4</sub> nanoparticles (NPs) were prepared according to a sonochemical method with minor modification (Bang and Suslick, 2007). In brief, FeCl<sub>2</sub>·4H<sub>2</sub>O and FeCl<sub>3</sub>·6H<sub>2</sub>O were dissolved in pre-degassed ultrapure water with a molar ratio of Fe<sup>2+</sup> to Fe<sup>3+</sup> controlled at 1:2. NaOH solution (1.5 mol/L) was degassed for 15 min and then added dropwise to the above solution in nitrogen atmosphere under vigorous mechanical agitation (ca. 500 r/min) and ultrasonication conditions. The glass flask containing the reaction solution was heated at 30°C for 1 hr. The resulting Fe<sub>3</sub>O<sub>4</sub> NPs were separated by an external magnetic field (2300 G), repeatedly rinsed with ultrapure water till the solution pH was close to neutral, and then dried overnight at 60°C.

Afterwards, Fe<sub>3</sub>O<sub>4</sub>@C nanocomposite was prepared by a hydrothermal method with certain modification (Wang et al., 2006). Oleyl amine (1 mL) was added into an aqueous suspension of Fe<sub>3</sub>O<sub>4</sub> NPs (50 mL, 10 g/L) at 80°C under vigorous mechanical agitation conditions, and the reaction was allowed to proceed for 1 hr. The resulting oleic acid-stabilized Fe<sub>3</sub>O<sub>4</sub> NPs were separated magnetically, and further suspended in 200 mL ultrapure water. Glucose (20 g) was dissolved in this aqueous suspension by mechanical agitation assisted with ultrasonication for 30 min. The mixture was transferred to a stainless steel autoclave (200 mL, with Teflon lining) and heated at 155°C for 3 hr. The solid product was magnetically separated, repeatedly rinsed with an ethanol solution (95%, V/V) for 5 times, and then dried overnight at 65°C.

### 1.3 Characterization of Fe<sub>3</sub>O<sub>4</sub>@C

Powder X-ray diffraction (XRD) patterns of Fe<sub>3</sub>O<sub>4</sub> and Fe<sub>3</sub>O<sub>4</sub>@C NPs were analyzed on a Bruker D8 Advance X-diffractometer using Cu K<sub>α</sub> radiation (λ = 0.15406 nm). Surface element valences, morphology, and surface properties (i.e., specific surface area, pore volume, and pore size) were determined with X-ray photoelectron spectroscopy (XPS, Escalab 250, Thermo Fisher Scientific), scanning electron microscopy (SEM, S-3000N, Hitachi), transmission electron microscopy (TEM, H-7500, Hitachi), and a surface area analyzer (ASAP2000, Micromeritics), respectively. Hysteresis loops of Fe<sub>3</sub>O<sub>4</sub> and Fe<sub>3</sub>O<sub>4</sub>@C NPs were measured with a vibrating specimen magnetometer (VSM, BHV-50HTI, Riken Keiki) at 300 K. The pH<sub>pzc</sub> of Fe<sub>3</sub>O<sub>4</sub>@C was determined by the mass titration method (Babić et al., 2009). The surface functional groups of Fe<sub>3</sub>O<sub>4</sub>@C were examined by Fourier transform infrared spectroscopy (FT-IR, Tensor 27, Bruker

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