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ORIGINAL ARTICLE

Preparation of magnetic separable $CoFe_2O_4$/PAC composite and the adsorption of bisphenol A from aqueous solution

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Abstract $CoFe_2O_4$/PAC composite adsorbent has been prepared via an immersing-calcination process, using ethylene diamine tetraacetic acid (EDTA) and citric acid (CIT) ligands containing sol as the $CoFe_2O_4$ precursor. The microstructure characterization and magnetic property of as-prepared sample were performed by means of XRD and VSM measurements. The adsorption kinetics, isotherms and thermodynamic process toward Bisphenol A molecules (BPA, which is considered as one of the typical endocrine disrupting chemicals) occurred on as-prepared magnetic adsorbent which were investigated by the pseudo-second order kinetic/intraparticle models, the Langmuir/Freundlich adsorption isothermal models and basic chemical thermodynamics principles, respectively.

1. Introduction

Bisphenol A (BPA), a monomer of various polymeric materials, is widely used for the production of polycarbonate plastics and epoxy resins and also as stabilizer or antioxidant for many types of plastics such as polyvinyl chloride (Staples et al., 1998). However, BPA is also considered as a typical kind of endocrine disrupting chemicals (EDCs) which may influence the living system (Kang et al., 2006). Recently, various techniques have been put forward by researchers in order to remove BPA molecules from the environmental matrices, involving adsorption separation (Tsai et al., 2006; Dong et al., 2010), biochemical (Zhao et al., 2008), electrochemical (Kuramitz et al., 2001), sonochemical (Torres et al., 2007) and photochemical reactions (Huang and Huang, 2009), etc.

Among the processes being proposed and/or developed, powdered-activated-carbon (PAC) based adsorption method is one of the most promising techniques owing to its low-cost, highly efficiency and less time consumption (Esteves et al., 2008; Mahmudov and Huang, 2010). However, the drawback

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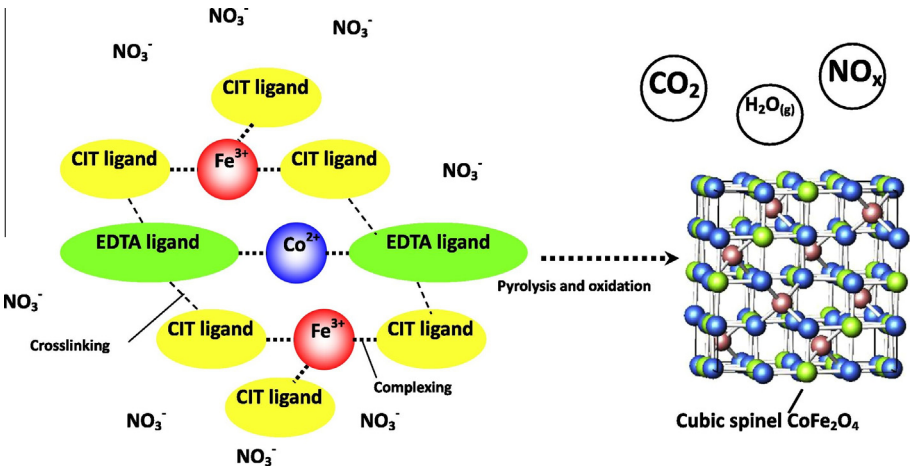

Figure 1 Schematic illustration of $CoFe_2O_4$ precursor (sols) and basic formation process of the complex oxides (Note: the picture of $CoFe_2O_4$ crystal cell was copied directly from the website of http://wikis.lib.ncsu.edu/index.php/Image:Size21.png).

regarding this technique is the separation of activated carbon which limits its further applications. Magnetic separation has been recognized as a quick and effective technique for separating magnetic particles with multi-composition. In recent years, the application of magnetic particle technology to solve environmental problems has received considerable attention (Tseng et al., 2007; Ao et al., 2008; Afkhami and Rasoul, 2009).

In the present work, cobalt ferrite ($CoFe_2O_4$), a typical kind of multiple complex oxide with hard magnetic property (Sun et al., 2010) was induced into PAC through a sol-gel chemical route. Magnetic PAC was used for the adsorption of BPA from aqueous solutions and was then separated from the process following a simple magnetic process. The interfacial adsorption property of magnetic PAC for the removal of BPA was systematically investigated and the possible process and mechanism was discussed as well.

2. Materials and methods

All the commercial reagents were of analytical grade and used as received without further purification. The reagents and PAC were purchased from Sinopharm Group Chemical Reagent Co., Ltd. The excellent complexing ability toward metal ions was noticed and the roles as buffer reagents to keep the constant pH values of the solution, Citric acid (CIT) and ethylene diamine tetraacetic acid (EDTA) were used as ligands of Ferric and Cobalt ions, respectively. Cobalt and ferric were adopted as corresponding nitrate, namely, cobalt nitrate ($Co(NO_3)_2{\cdot}6H_2O$) and ferric nitrate ($Fe(NO_3)_3{\cdot}9H_2O$), respectively. Mole ratio of Co to Fe was 1:2. Co-EDTA and Fe-CIT complex solutions were prepared by mixing proper amount of cobalt nitrate, ferric nitrate and their corresponding ligands. The mole ratio of Co/EDTA and Fe/CIT was controlled at 1:1.5. After both of the pH of the solutions were adjusted by $NH_3{\cdot}H_2O$/HNO_3 to between 3 and 4, Co-EDTA and Fe-CIT solutions were then mixed with the Co/Fe mole ration of 1:2, and the pH of the final solution was adjusted between 3 and 4 by using $NH_3{\cdot}H_2O/HNO_3$.

To obtain the $CoFe_2O_4$/PAC composite adsorbent, a certain amount of PAC was added into the complex compound solution above, ensuring the mass ration of PAC/$CoFe_2O_4$ was 10:1. The stable gels were prepared after stirring at 333 K for 5 h and then heated at 413 K in the oven for 48 h. The final product of PAC/$CoFe_2O_4$ composite was formed though a heating treatment process at 673 K under nitrogen atmosphere. Fig. 1 schematically illustrates the precursor of $CoFe_2O_4$ (sols) and the formation of complex oxides.

The phase structure characterization of PAC/$CoFe_2O_4$ composite was carried out by X-ray diffraction (XRD) in the 2θ range of 20–80 using Cu Kα X-ray source (λ = 0.154, 18 nm) with voltage and current of 40 kV and 100 mA, respectively. The magnetic properties were measured by vibrating sample magnetometer (VSM, Lakeshore, Model 7300 series).

To examine the behaviors of adsorption kinetic/isotherm and calculate the thermodynamics parameters, the adsorption experiments were carried out in 200 ml of 90 mg/L BPA solution at 150 rpm in a thermostatic shaker at 25 °C for 24 h at pH 7.0 after 0.01 g amount of adsorbent was added unless otherwise indicated. The concentrations of BPA were determined at 276 nm, using a UV–vis spectrophotometer (VARIAN Cary 100) (Dong et al., 2010).

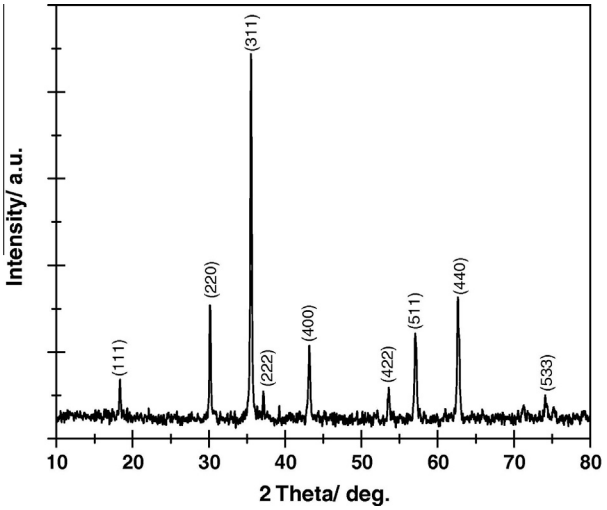

Figure 2 XRD pattern of as-prepared $CoFe_2O_4$/PAC adsorbent.