Supporting Information for

Mesoporous Layered Graphene Oxide/Fe₃O₄/C₃N₃S₃ Polymer Hybrids for Rapid Removal of Pb²⁺ and Cd²⁺ from Water

Hongjun Yang, ¹ Hongwen Yu, ² Jidun Fang, ¹ Jingkuan Sun, ¹Jiangbao Xia, ¹ Wenjun Xie, *¹ Shoucai

Wei, ¹ Qian Cui, ¹ Chunlong Sun, ¹ and Tao Wu¹

¹ Shandong Key Laboratory of Eco-Environmental Science for the Yellow River Delta, Binzhou University, NO. 391, 5th Yellow River Road, Binzhou City 256603, Shandong Province, China.

² Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, 4888 Shengbei Rd, Changchun 130102, China.

(⊠) To whom correspondence should be addressed. E-mail: sfyanghongjun@126.com, xwjeric@163.com (W. Xie).

. Experimental

Materials

Chemicals (Iron (III) chloride anhydrous, Iron (II) chloride tetrahydrate, nitric acid, lead nitrate, cadmium nitrate, Na₃C₃N₃S₃ monomer, calcium chloride anhydrous, sodium hydroxide, ammonium hydroxide, Concentric sulfuric acid, potassium permanganate, expanded graphite, magnesium chloride hexahydrate, etc.) were purchased from Beijing Chemical Works, China. All the chemical reagents used in this paper are of analytical reagent grade (AR Grade). Simulated surface water contain K⁺ (6 mg/L), Na⁺ (125 mg/L), Ca²⁺ (7 mg/L), Mg²⁺ (9 mg/L). Simulated ground water contain K⁺ (5 mg/L), Na⁺ (106 mg/L), Ca²⁺ (24 mg/L), Mg²⁺ (7 mg/L). Simulated seawater contain K⁺ (500 mg/L), Na⁺ (9600 mg/L), Ca²⁺ (400 mg/L), Mg²⁺ (1280 mg/L).

All reagents were commercially available and were used without further purification. Deionized water with a resistivity of >18 M Ω cm was used in this study.

Preparation of Fe₃O₄ NPs

Magnetite (Fe₃O₄) nanoparticles (NPs) sized 8-12 nm were obtained by a modified Massart Method. In a typical procedure, FeCl₂·4H₂O (2.9813 g) and FeCl₃ (4.87 g) were mixed together in 380 mL of H₂O at 90 °C and NH₃·H₂O (25%, 20 mL) was added, magnetite formation was visible as a black precipitate. The particles were washed with 500 mL of deionized water several times until the pH value of the supernatant was unchanged. The Fe₃O₄ NPs were then peptized with a 2M HNO₃ solution (500 mL). The magnetite was recovered with a magnet and was dried in an oven at 50 °C.

Results and discussion

Characterization

Scanning electron microscope (SEM) images of powders were taken by using a Hitachi S-3400N–II system (with 25.0 kV acceleration voltages, a 60 s acquisition time).Transmission electron microscopy (TEM) images were taken by using a TECNAI G2 high- resolution transmission electron microscope with an accelerating voltage of 200 kV. All TEM samples were created by depositing a drop of diluted suspensions in water s2 on a carbon-film-coated copper grid. Infrared spectra were collected on a VERTEX 70 Fourier transform infrared (FT-IR) spectrometer (Bruker). Samples were degassed under vacuum at 60°C over night prior to analysis. X-Ray diffraction (XRD) patterns of the samples were collected on a Bruker D8ADVANCE diffractometer (Germany) using Cu-Ka (1.5406 Å) radiation. Surface area was determined using Brunauer-Emmett-Teller (BET) method. Nitrogen adsorption-desorption isotherm measurements were conducted at 77 K (ASAP 2010). X-Ray photoelectron spectrum (XPS) measurements were performed on an ESCALAB-MKII spectrometer (VG Co., United Kingdom) with Al Ka X-ray radiation as the X-ray source for excitation. Thermogravimetric analysis (TGA) measurements were performed by using a Pyris Diamond TG/DTA Thermogravimetric Analyzer (Perkin-Elmer Thermal Analysis). The samples were heated under dry air from room temperature to 850 °C at a rate of 10 °C/min. The concentration of Fe and H⁺ was measured by atomic absorption spectrophotometer (AA-6300C, Shimadzu) and pH meter.



Figure S1. SEM images of (a) GP2 and (b) GFP2.



Figure S2. TGA curves of P, GP and GFP.



Figure S3. The N_2 sorption and desorption isotherm of (a) GFP1, (c) GFP2, (e) GFP3 and the pore size distribution of (b) GFP1, (d) GFP2, (f) GFP3.



Figure S4. Freundlich isotherms of (a) GFP2 (Pb^{2+}) and (b) GFP2 (Cd^{2+}).



Figure S5 Adsorption-regeneration cycles of Pb^{2+} (Cd²⁺) onto GFP2.



Figure S6. Magnetic separation of GFP2 (adsorption liquid). (Photograph courtesy of Hongjun Yang. Copyright 2019 and the image is free domain)