Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A

Electronic supplementary information

In situ Controllable Synthesis of Magnetic Prussian Blue/Graphene Oxide Nanocomposites for Removal of Radioactive Cesium in Water

Hongjun Yang, [a,b] **Lei Sun,** [a] **Jiali Zhai ,** [a] **Haiyan Li,**[a] **Yan Zhao**[a] **and Hongwen Yu**[* a]

[a]Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, 4888 Shengbei Rd,Changchun130102, China.

[b]University of Chinese Academy of Sciences, Beijing, 100049, China.

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*Phone: +86 431 85542290; Fax: +86 431 85542290; E-mail: yuhongwen@neigae.ac.cn

Experimental section

Materials. All the chemical reagents used in this study are of analytical reagent grade (AR Grade). Iron (Ⅲ) chloride anhydrous($FeCl_3$, Sinopharm Chemical Reagent Co. , Ltd, 97%), Iron (□) chloride tetrahydrate($FeCl_2{\cdot}4H_2O$, Beijing Chemical Works, 98%), potassium hexacyanoferrate(□)trihydrate($K_4[Fe(CN)_6]$ $\cdot 3H_2O$, Beijing Chemical Works, 98%), cesium chloride(CsCl, Beijing Chemical Works, 99%), ammonia solution(25%)($NH_3{\cdot}H_2O$, Beijing Chemical Works, 25-28%), nitric acid(HNO_3, Beijing Chemical Works, 65-68%), simulated seawater contain Na^+ (9.6 g/ L), Mg^{2+} (1.28 g/L), Ca^{2+} (0.4 g/L), K^+ (0.5 g/L).

Preparation of Fe_3O_4 NPs. 8-12 nm magnetite particles were synthesized by a slightly modified Massart Method.[21] In a typical synthesis, $FeCl_3$ (4.87 g) and $FeCl_2{\cdot}4H_2O$ (2.9813 g) were mixed together in H_2O (380 mL) at 90 ℃ and $NH_3{\cdot}H_2O$ (25%; 20 mL) was added, magnetite formation was visible as a black precipitate. The particles were washed with H_2O (500 mL) many times until the pH value of the supernatant was constant. The magnetite particles were then peptized with a 2M HNO_3 solution (500 mL) . The precipitate was recovered with a magnet and then dried in an oven at 50 ℃ .

Preparation of graphene oxide (GO). The graphite oxide were prepared by chemical exfoliation of graphite powder based on a modified Hummer's method.[22] Then, exfoliation of graphite oxide to GO was achieved under ultrasonic treatment for 60 min. Subsequently, the as-prepared brown dispersion was subjected to centrifugal separation for 5 min at 2000 rpm to eliminate the black precipitate, and a homogeneous GO aqueous dispersion was obtained. Finally, the graphene oxide were collected by centrifugation at 8000 rpm for 20 min , and then GO was dried via evaporation of water at 50 ℃.

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 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℃ over night prior to analysis. Surface area was determined using Brunauer-Emmett-Teller (BET) method. X-Ray diffraction (XRD) patterns of the samples were collected on a Bruker D8ADVANCE diffractometer (Germany) using Cu-Kα (1.5406 Å) radiation. 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 Kα 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 950℃ at a rate of 10℃/min. The concentration of Cs^+, K^+, Fe and H^+ was measured by atomic absorption spectrophotometer (AA-6300C, Shimadzu) and pH meter.

Results and discussion

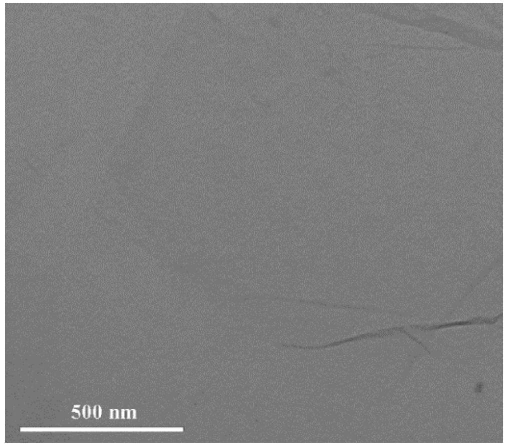

Figure S1 TEM images of GO

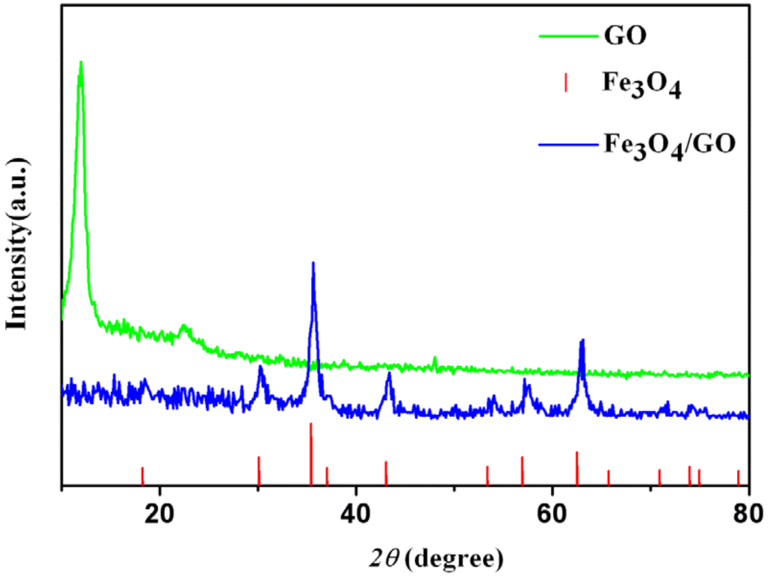

Figure S2 XRD patterns of GO, Fe_3O_4 and Fe_3O_4/GO

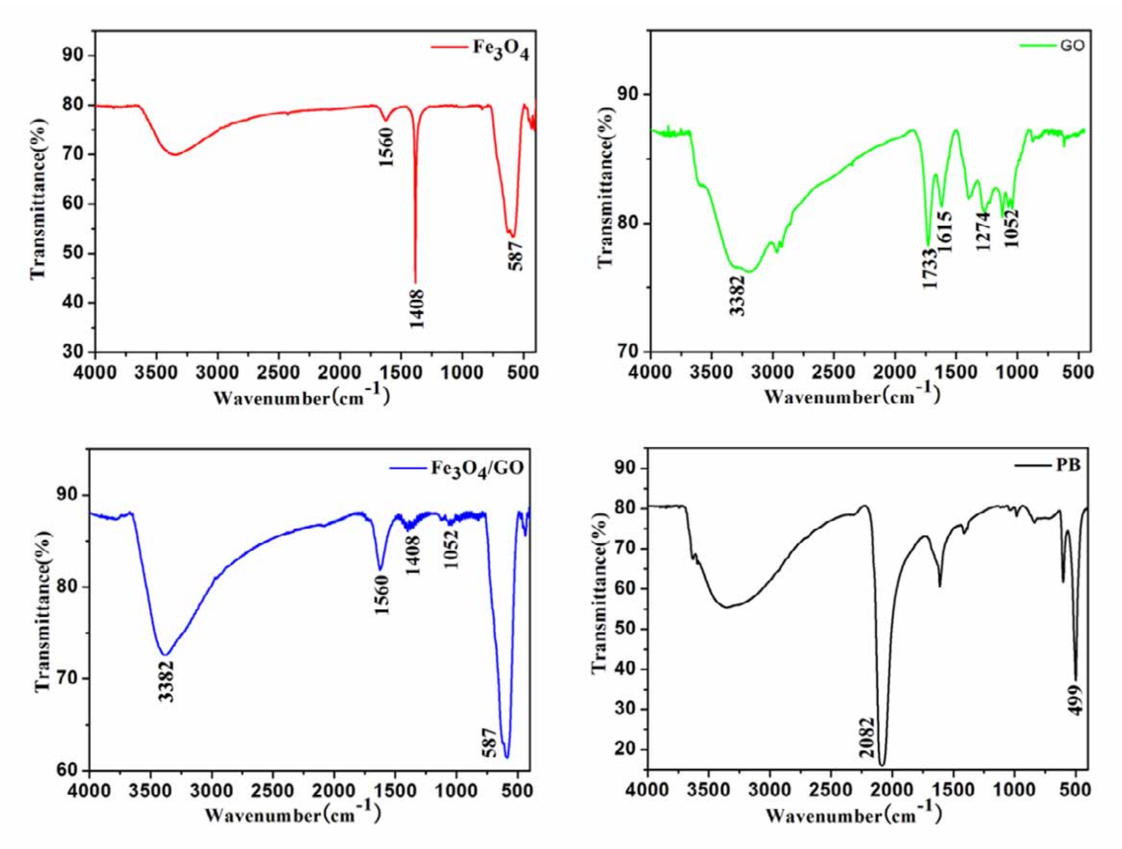

Figure S3 FTIR images of Fe_3O_4, GO, Fe_3O_4/GO and PB

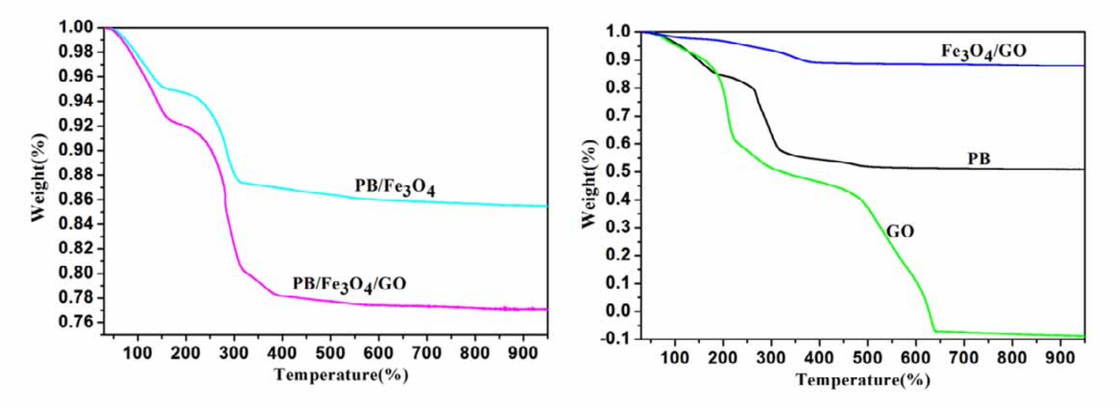

Figure S4 TGA curves of GO, PB, Fe_3O_4/GO, PB/Fe_3O_4 and PB/Fe_3O_4/GO

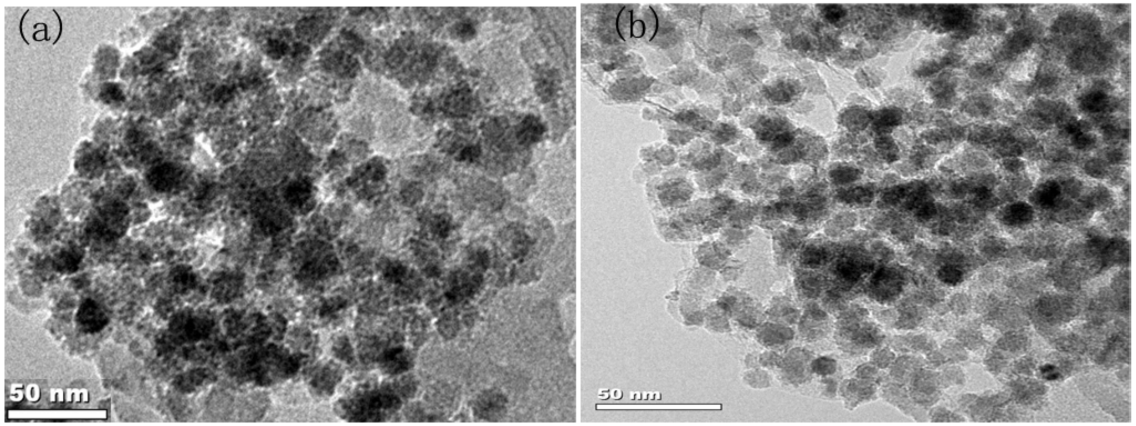

Figure S5 TEM images of PB/Fe_3O_4 and PB/Fe_3O_4/GO (after adsorption)

Table S1. Leaching of Fe after suspending 50 mg of adsorbent in 30 mL of different fluids for 24 h

water matrix	pH=4(water)	pH=5(water)	pH=6(water)	pH=7(water)	pH=8(water)	pH=9(water)	pH=10(water)	seawater
%Fe leached per total Fe of PB/Fe_3O_4	0.0008	0.0021	0.0023	0.0026	0.0024	0.0037	0.0061	0.9530
%Fe leached per total Fe of PB/Fe_3O_4/GO	0.0427	0.0354	0.0317	0.0555	0.0441	0.0269	0.0609	0.6051