### Supporting Information

# One-step Fabrication of High Performance Tremella-like Fe<sub>3</sub>S<sub>4</sub>/C Magnetic Adsorbent with Easy Recovery and Regeneration Properties

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#### **TG and XPS measurements**

TG measurement was performed for the Fe<sub>3</sub>S<sub>4</sub>/C composite with heating rate 5 °C/min and nitrogen flow 25 ml/min, as shown in Fig.S1. The weight loss before  $160^{\circ}$ C is ascribed to the adsorbed water in the sample. In the temperature range from  $160^{\circ}$ C-500 °C, the weight loss is mainly attributed to decomposition and further carbonization of the carbonaceous materials in the sample, which release CO, CO<sub>2</sub> and H<sub>2</sub>O gases, etc.,[S1, S2]. Meanwhile, the Fe<sub>3</sub>S<sub>4</sub> is decomposed to FeS<sub>2</sub> [S3], due to the surface-adsorbed oxygen [O], or the reaction

$$3Fe_3S_4 \xrightarrow{[O]} 6FeS_2 + Fe_3O_4 \tag{S1}$$

would take place. When the temperature is above 500  $^{\circ}$ C, the FeS<sub>2</sub> is further transformed to Fe<sub>1-x</sub>S and sulphur [S3], or

$$(1-x)FeS_2 \longrightarrow Fe_{1-x}S + (1-2x)S \tag{S2}$$

resulting in a rapid weight loss in the range from 500 °C to 560 °C owing to evaporation of

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sulphur. The final slight weight loss stage occurs from 560  $^{\circ}$ C due to the continuing carbonization of carbonaceous materials.

XPS measurements were conducted for the Fe<sub>3</sub>S<sub>4</sub>/C composite. The binding energy spectrum of O1s is illustrated in Fig. S2. The peaks at 533.03, 531.37 and 529.71 eV correspond to C-O, C=O and –OH groups, respectively [S4], indicating the existence of functional groups at surface of Fe<sub>3</sub>S<sub>4</sub>/C composites. Also, the binding energy spectrum of C1s is shown in Fig. S3. The peaks at 284.2 eV, 285.0 eV, 286.0 eV, 287.3 eV and 288.5 eV correspond to C=C, C-C/C-H, C-O, C=O and O-C=O groups, respectively [S4]. Obviously, the C1s signal in Fig.S3 indicates that the hydrophobic groups(C-H, C=C, C-C) account for a decent proportion in total carbon containing groups on the adsorbent's surface.

Both measurements have further confirmed existence of functional groups (C-O, C=O and -OH) at surface of Fe<sub>3</sub>S<sub>4</sub>/C composites.

#### References

[S1] C. E. Byrne and D. C. Nagle, *Carbon*, 1997, **35**, 259-266.

- [S2] I.C. Lewis, Carbon, 1982, 20, 519-529
- [S3] M. J. Dekkers, H. F. Passier and M. A. A. Schoonen, *Geophys. J. Int.*, 2000, 141, 809-819.
- [S4] C.G Chen, B. Liang, Di Lu, A. Ogino, X. K. Wang and M. Nagatsu, *Carbon*, 2010, 48, 939 -948.



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Fig. S1 TG curve of Fe<sub>3</sub>S<sub>4</sub>/C composite with heating rate 5 °C/min and nitrogen flow 25ml/min.

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Fig. S2 The binding energy spectrum of O1s in the  $Fe_3S_4/C$  composite.

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**Fig. S3.** The binding energy spectrum of C1s in the  $Fe_3S_4/C$  composite.

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Fig. S4 A magnified area cut from Fig.4(d) in the manuscript (with different contrasts in different areas)



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Fig. S5 The schematic atomic structure of the crystal plane (111) (View along [111]).





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Figure S6. The FESEM images of the as-prepared products after solvothermal reaction for (a) 1 h and (b) 4h.



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Fig. S7 FESEM image (a) and XRD (b) of the Fe<sub>3</sub>S<sub>4</sub>/C composites after MB adsorption. (Note: The adsorption experiment was conducted using 10 mg Fe<sub>3</sub>S<sub>4</sub>/C composites in the 5 ml solution with MB 10mg/L for 10min. After separation from the solution, the composites were washed with deionized water and dried before FESEM and XRD characterization)