

**Electronic Supplementary Information**

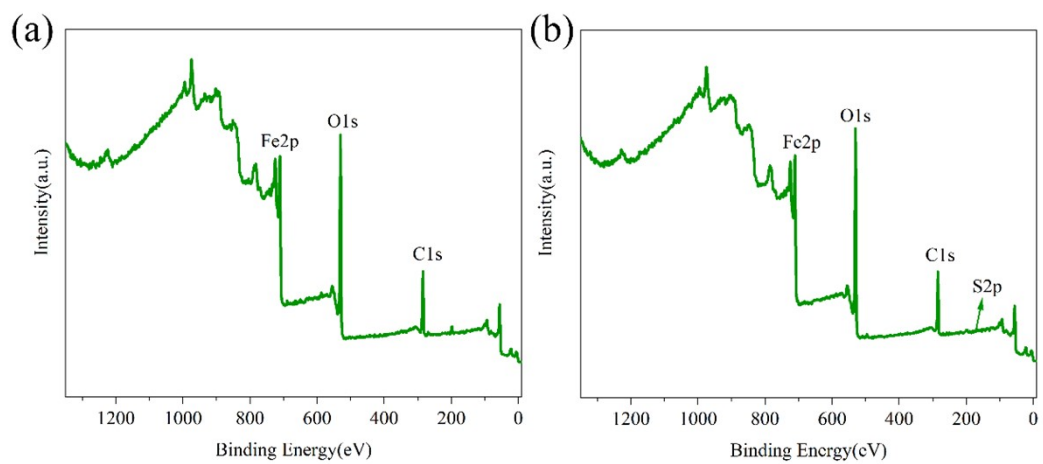
**Decorated reduced graphene oxide could transfer sulfides into sulfur  
and sulfone in wastewater**

Zheng Wang<sup>a,b</sup>, Hongtao Cui<sup>a</sup>, Hengduo Xu<sup>b</sup>, Yanqing Sheng<sup>b\*</sup>

<sup>a</sup> School of Chemistry and Chemical Engineering, Yantai University, Yantai 264005,  
China

<sup>b</sup> Key Laboratory of Coastal Zone Environmental Processes, Yantai Institute of  
Coastal Zone Research, Chinese Academy of Sciences, Yantai 264003, China

\* Corresponding author. *E-mail address:* [yqsheng@yic.ac.cn](mailto:yqsheng@yic.ac.cn) (Y. Sheng); *Tel:* +86-  
0535-2109265



**Fig.S1.** XPS spectra of fresh (a) and used (b) RGO/Fe<sub>3</sub>O<sub>4</sub>.

### **Text S1** Preparation of graphene oxide.

The modified Hummer's method<sup>1</sup> was used to prepare Graphene Oxide (GO) from graphite powder. Briefly, graphite powder (1.0 g) and NaNO<sub>3</sub> (0.5 g) were mixed with 23 mL H<sub>2</sub>SO<sub>4</sub> (98%) in a 500 mL round-bottom flask. The mixture was stirred for a time to dissolve completely in an ice-water bath. KMnO<sub>4</sub> (3.0 g) was added gradually in portions under quickly stirring, while the temperature was closely supervised so as not to make the mixture to go above 10 °C. The ice bath was then removed and the mixture was heated at 35 °C for 30 min. Next, ultrapure water (50 mL) was slowly added, and the mixture was stirred for 20 min. Subsequently, the temperature was raised to 98 °C and the reaction was kept for 15 min, at which time of ultrapure water (140 mL) was added. In the end, 30% H<sub>2</sub>O<sub>2</sub> (10 mL) was added dropwise to the reaction mixture, and the color of the mixture gradually changed to bright yellow with bubbles. The resulting mixture was settled at room temperature at least 5 h after which the clear supernatant was decanted. The remaining mixture was centrifuged and washed with a total of 500 mL 5% HCl solution until there was no sulfate ion in the filtrate, and then washed repeatedly with ultrapure water until it was neutral. The resulting solid was freeze-dried for 24 h to obtain solid GO.

1. W. Zhang, X. Zou and J. Zhao, *J. Mater. Chem. C*, 2015, **3**, 2788-2791.

**Text S2** Methylene blue method.

Sodium sulfide reacts with N, N-dimethyl-p-phenylenediamine oxalate to form blue colored complex which is evaluated by UV–vis spectrophotometer. The intensity of the blue color and the sulfide concentration are in the direct ratio.

**Text S3** The detailed experimental procedures of main experimental parameters.

**Mass ratio of GO to FeCl<sub>2</sub> · 4H<sub>2</sub>O (1:1-1:4):** Briefly, 10 ml of sulfide with concentration of 19.98 mg/L were added to four 50 mL stopper conical flasks, respectively. Then RGO/Fe<sub>3</sub>O<sub>4</sub>-1(1:1), RGO/Fe<sub>3</sub>O<sub>4</sub>-2(1:2), RGO/Fe<sub>3</sub>O<sub>4</sub>-3(1:3), RGO/Fe<sub>3</sub>O<sub>4</sub>-4(1:4) was weighed 25 mg respectively and was added to solution. The system was shaken in a constant temperature oscillator for 180 min. 1.0 mL of the dispersion solution was pipetted after the reaction reached equilibrium and filtered quickly with a 0.45 μm filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined before and after adsorption. The test was executed repeatedly. The adsorption temperature was 298 K, the oscillation frequency was 160 rpm. When the sulfide concentration is 44.60 mg/L, 83.01 mg/L, 111.55 mg/L and 134.54 mg/L, the procedure is the same as above.

**Contact time (0-180min):** Briefly, 10ml sulfide solution (133.14 mg/L) were added to eight 50mL stopper conical flasks, respectively. Then 25 mg adsorbent was added to each solution. The system was shaken in a constant temperature oscillator for 1,5,10,15,30,60,120 and 180 min. 1.0 mL of the dispersion solution was pipetted at specified moment and filtered quickly with a 0.45 μm filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined before and after adsorption. The test was executed repeatedly. The adsorption temperature was 298 K, the oscillation frequency was

160rpm.

**pH value (2.0-12.0):** Firstly, 10ml sulfide solution (138.56 mg/L) were added to six 50mL stopper conical flasks, respectively. Then the pH of the solution was adjusted to 1.97, 3.87, 5.4, 7.43, 9.57, 11.85 with 0.1MHCl and 0.1MNaOH. Subsequently, 25mg adsorbent was added to each solution. The system was shaken in a constant temperature oscillator for 180 min. 1.0 mL of the dispersion solution was pipetted after the reaction reached equilibrium and filtered quickly with a 0.45  $\mu$ m filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined after adsorption. The test was executed repeatedly. The adsorption temperature was 298 K, the oscillation frequency was 160 rpm.

**Adsorbent dosage (0-50.0 mg):** Briefly, 10ml sulfide solution (135.88 mg/L) were added to ten 50mL stopper conical flasks, respectively. Then 0.005, 0.010, 0.015, 0.020, 0.025, 0.030, 0.035, 0.040, 0.045, 0.050 g of adsorbent were weighed and added to the solution, respectively. The system was shaken in a constant temperature oscillator for 180 min. 1.0 mL of the dispersion solution was pipetted after the reaction reached equilibrium and filtered quickly with a 0.45  $\mu$ m filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined before and after adsorption. The test was executed repeatedly. The adsorption temperature was 298 K, the oscillation frequency was 160 rpm.

**Initial sulfide concentration (0-350 mg/L):** Briefly, 10 ml of sulfide with concentration of 50, 100, 150, 200, 225, 250, 300, 350 mg/L were added to eight 50mL stopper conical flasks, respectively. Then 25 mg adsorbent was added to each solution. The system was shaken in a constant temperature oscillator for 180 min. 1.0 mL of the dispersion solution was pipetted after the reaction reached equilibrium and filtered quickly with a 0.45  $\mu\text{m}$  filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined before and after adsorption. The test was executed repeatedly. The adsorption temperature was 298 K, the oscillation frequency was 160 rpm.

**Temperature (25–45°C):** Briefly, 10ml of sulfide with concentration of 130.23 mg/L were added to three 50mL stopper conical flasks, respectively. Then, 25 mg adsorbent was added to each solution. The system was shaken in a constant temperature oscillator for 180 min. The temperature of constant temperature oscillator is set to 298 K, 308 K and 318 K, respectively. 1.0 mL of the dispersion solution was pipetted after the reaction reached equilibrium and filtered quickly with a 0.45  $\mu\text{m}$  filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined before and after adsorption. The test was executed repeatedly. The oscillation frequency was 160 rpm.

**Ionic strength:**

NaCl: Briefly, sulfide solution (116.72 mg/L) was prepared with NaCl solution (0,0.25,0.5,0.75,1 M), respectively. Then, 10 ml above sulfide solution were added to five 50mL stopper conical flasks, respectively. Then 25 mg adsorbent was added to each solution. The system was shaken in a constant temperature oscillator for 180 min. 1.0 mL of the dispersion solution was pipetted after the reaction reached equilibrium and filtered quickly with a 0.45  $\mu\text{m}$  filter membrane. Next, the equilibrium concentrations of sulfide were quantified by methylene blue method. The pH of the solution was determined before and after adsorption. The test was executed repeatedly. The adsorption temperature was 298 K, the oscillation frequency was 160 rpm.

CaCl<sub>2</sub>: The sulfide solution (116.72 mg/L) was prepared with CaCl<sub>2</sub> solution (0,0.25,0.5,0.75,1 M), respectively. The other steps are the same as those for NaCl.