Graphene oxide interpenetrated polymeric composite

hydrogels as highly effective adsorbents for water treatment

Chong Cheng,^{a,b} Zhengyang Liu,^a Xiaoxiao Li,^a Baihai Su,^c Tao Zhou,^{a,*} Changsheng Zhao,^{a,*}

^a College of Polymer Science and Engineering, State Key Laboratory of Polymer Materials Engineering, Sichuan University, Chengdu 610065, China

^b Department of Chemical Engineering, Department of Biomedical Engineering, University of Michigan, Ann Arbor, Michigan, 48109, USA

^c Department of Nephrology, West China Hospital, Sichuan University, Chengdu 610041, China

Tel: +86-28-85400453, Fax: +86-28-85405402, E-mail: (T. Zhou) zhoutaopoly@scu.edu.cn; (C.S. Zhao) zhaochsh70@163.com or zhaochsh70@scu.edu.cn

1. Preparation of graphene oxide (GO)

Graphene oxide (GO) was prepared from natural graphite flakes by a modified Hummers method ²⁹. Briefly, 5 g graphite and 3.75 g NaNO₃ were placed in a flask. Then, 150 mL H₂SO₄ was added with stirring in an ice-water bath, and then 20 g KMnO₄ was slowly added for over 1 h. The mixture was stirred in an ice water bath for 2 h, followed by a vigorously stirring for 3 days at room temperature. Then, the mixture was diluted with DI water (500 mL) slowly, and the excessive KMnO₄ was decomposed by H₂O₂ (30 wt. %, 15 mL). The insoluble precipitations were removed by centrifugation. The resulted GO solution was filtered and washed with HCl (10 wt. %, 1 L) and DI water for several times to remove the metal ions. The pristine brown GO solution was dialyzed with DI water for 1 week before use to remove any residual salts and acids.



Fig. S1. (a) Typical AFM images and cross-section analyses of GO; (b) TEM images and corresponding SAED patterns of GO; (c) Chemical structure of GO; (d) Photographs of the prepared PAA2 and PAA2/GO interpenetrated hydrogels and the gelation formation tests.

Materials	AA (g)	BIS (mg)	APS (mg)	Water (g)	GO(mg)	GO/AA(%)
PAA1-GO-0	0.52	26	2.13	10	0	0
PAA1-GO-1	0.52	26	2.13	10	16	3.16
PAA1-GO-2	0.52	26	2.13	10	30	6

Table S1 The synthetic chemical components of PAA1/GO composite hydrogels.



Fig. S2. The SEM images for the PAA1/GO hydrogels: PAA1 (a-1), (a-2), and (a-3); PAA1-GO-1 (b-1), (b-2), and (b-3); PAA1-GO-2 (c-1), (c-2), and (c-3).



Fig. S3. The adsorbed MB amount per unit mass of PAA1, PAA1-GO-1, and PAA1-GO-2



Fig. S4. FTIR spectra for the PAA2, PAA2-GO-1, PAA2-GO-2, and PAA2-GO-3.

To characterize the chemical structure of PAA2/GO hydrogels, FTIR was applied to detect the difference, as shown in Fig. S4. Compared with pure PAA2 hydrogels, multiple characteristic peaks for the mixed GO were observed in the range of 900 to 1750 cm⁻¹, which could be assigned to the functional groups of GO, including the C–O (vC–O at 1060 cm⁻¹) and C–O–C (vC–O–C at 1240 cm⁻¹); and the peak intensity increased when more GO was added. The broad band at 1720 cm⁻¹ associated with v(C=O) in carboxylic acid and carbonyl moieties became sharp peak after the adding of GO, indicating that GO had great influence on the hydrogel structures. The broad peak usually derived from the random polymer structures, while, when GO was added, the PAA was polymerized on the GO skeleton, thus became more ordered; thus the vibration peak of C=O became stronger than the pure PAA2 hydrogel.



Fig. S5. The time-dependent MB adsorbed amounts per unit mass of PAA2-GO-3 at various initial concentrations.



Fig. S6. (a) Langmuir adsorption isotherm of MB onto PAA2/GO hybrid hydrogel. (b) Freundlich adsorption isotherm of MB onto PAA2/GO hybrid hydrogel.