Electronic Supplementary Information

Holey graphene nanosheets: large-scale rapid preparation and their application toward high-effective water cleaning

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Experimental section

Materials

Fuming HNO_3, activated carbon, graphite powder, $NaClO_3$, CR, MB, toluene, ethylene glycol and pump oil were purchased from Aladdin Ltd. (Shanghai, China). All chemical reagents were used without further purification.

Preparation of h–G NSs and graphene nanosheets

Oxidized graphite powder was prepared according to a modified Brodie method.[15] In brief, graphite (20 g), $NaClO_3$ (85 g) and fuming HNO_3 (200 ml) were mixed together and stirred for 30 min at room temperature. The resulting mixture was washed with deionized water and the dried oxidized graphite powder in glass beaker was then transferred to a chamber with a temperature-programmed furnace and heated at 300 ºC for 30 min in air to obtain the h–G NSs. Graphene nanosheets were prepared according to our previous method.[1]

Characterizations

SEM measurements were made on a XL30 ESEM FEG microscope. Transmission electron microscopy (TEM) measurements were made on a HITACHI H–8100 microscopy (Tokyo, Japan). AFM characterizations were performed on MultiMode–V (Veeco Metrology, Inc.). The BET surface area, pore volume and pore size were measured on a Quantachrome NOVA 1000 system at liquid N_2 temperature. Raman spectra were obtained on J–Y T64000 Raman spectrometer with 514.5 nm wavelength incident laser light. Nicolet 560 FTIR spectrometer was applied to collect the FTIR spectrum. XPS data were collected on an ESCALAB MK II X–ray photoelectron spectrometer using Mg as the exciting source. UV–vis absorption spectra were obtained on a UV5800 Spectrophotometer (Shimadzu, Japan).

Oil and organic solvent removal tests

We measured the absorption capaticy of h–G NSs for four different kinds of organic solvents and oils, including toluene, ethylene glycol, pump oil and used pump oil. The weighted samples were left submerged in oil or solvent fully for overnight to ensure full saturation. The saturated samples were weighed quickly to avoid evaporation of absorbed solvents. The absorption capacity values, W (wt/wt) %, were obtained by calculating the differences of mass. The graphene nanosheets, commercial activated carbon and graphite were performed with the same procedure as used for comparison purpose.

Dye removal tests

Different concentrations of dye solutions (CR and MB) were prepared by dissolving desired amounts of CR and MB into deionised water, respectively. In a typical adsorption procedure of CR, the h–G NSs (10 mg) were added to CR solution (130 mg L^{-1}, 25 ml) under magnetic stirring and the concentrations of CR (496 nm) in the solution at different time intervals were determined using UV–vis absorption spectra. The adsorption capacity of MB was also studied under the similar procedure. The Langmuir adsorption model is used to calculate the maximal adsorption capacity by varying the initial dye concentration:[23,24a] $Q_e= Q_m bC_e/(1+bC_e)$, Where Q_e (mg g^{-1}) is the amount of dyes adsorbed at equilibrium concentration, C_e (mg L^{-1}) is the equilibrium concentration in solution, Q_m (mg g^{-1}) is the maximum adsorption capacity corresponding to complete monolayer covering on the adsorbents, and b is the equilibrium constant (L mg^{-1}).

Reference

1. W. Lu, S. Liu, X. Qin, L. Wang, J. Tian, Y. Luo, A. M. Asiri, A. O. Al-Youbi and X. Sun, *J. Mater. Chem.,* 2012, **22**, 8775.

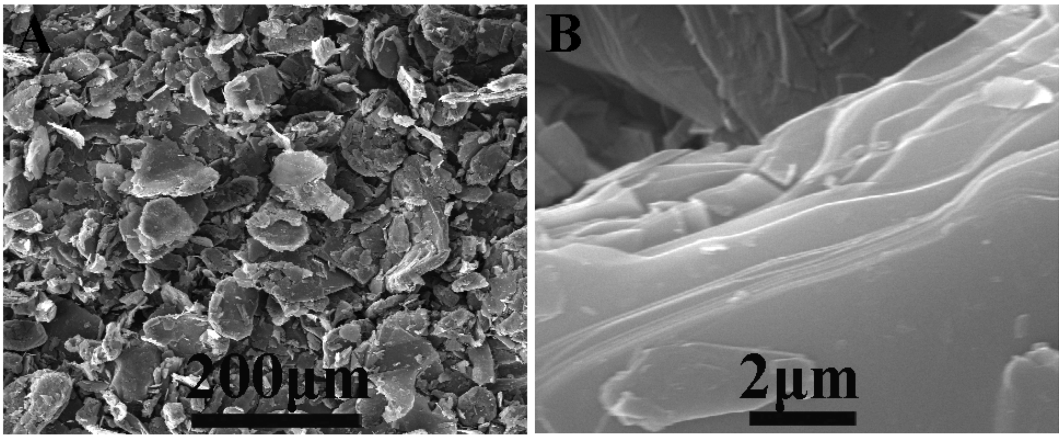

Fig. S1 (A) Low and (B) high magnification SEM images of the commercial graphite.

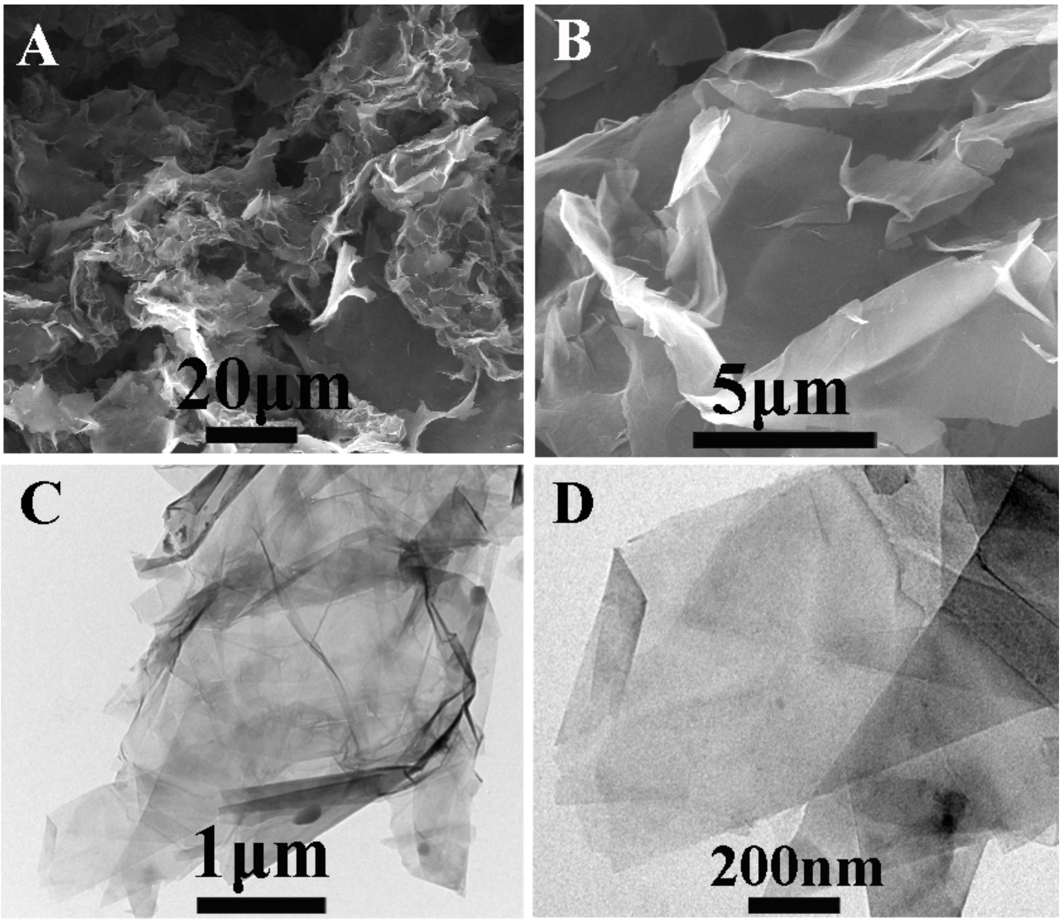

Fig. S2 (A) Low and (B) high magnification SEM images, (C) low and (D) high magnification TEM images of graphene nanosheets.

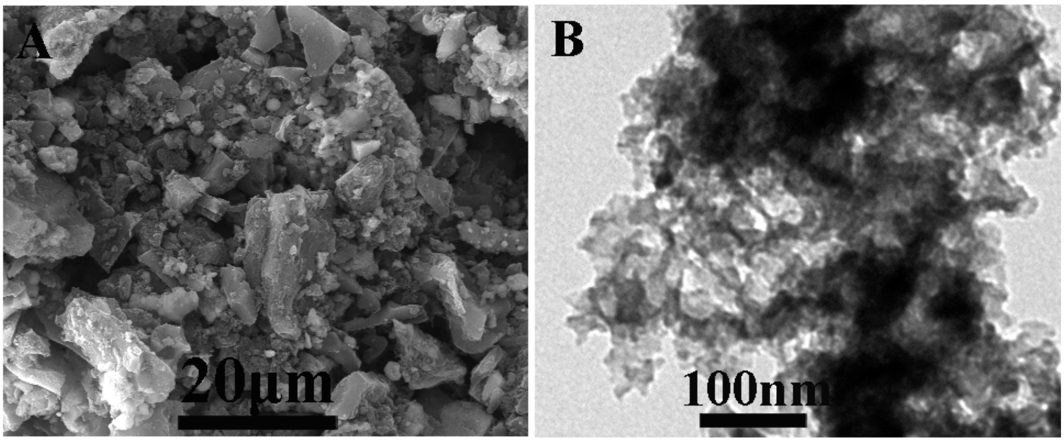

Fig. S3 (A) SEM and (B) TEM images of activated carbon.

Table S1 Summary of the BET surface area, pore volume and BJH pore-size distribution of the adsorbents used in this work.

Adsorbents	BET area (m^2 g^{-1})	Pore volume (cm^3 g^{-1})	BJH pore–size distribution (nm)
h–G NSs	1053.3	3.39	1.4–16.3
Graphene nanosheets	123.3	0.16	0.7–15.1
Activated carbon	380.7	0.393	1.4–16.5
Commercial graphite	90.0	0.109	0.7–6.2

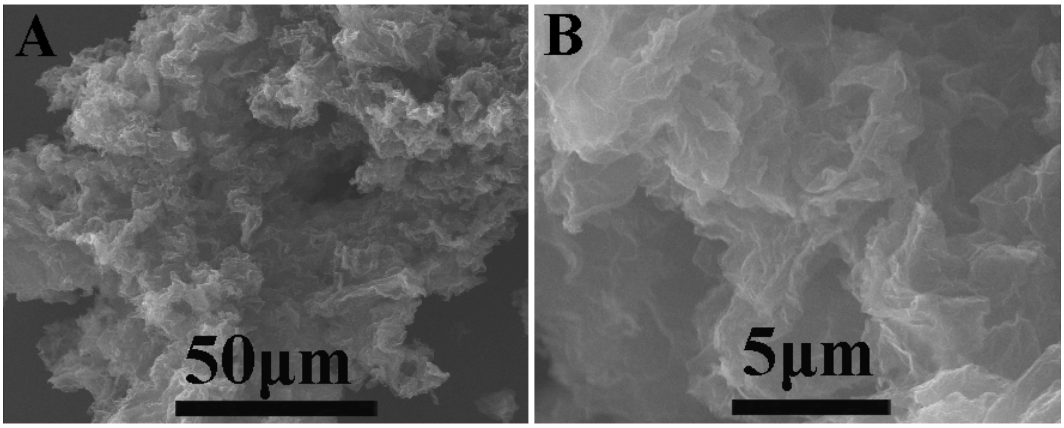

Fig. S4 (A) Low and (B) high magnification SEM images of the h–G NSs after recycling adsorption.

Table S2 Summary of oil maximum absorption capacities (Q_m) on various adsorbents.

Adsorbents	Type of oil	Absorption capacity (W%)	Reference
Graphene aerogels	Vegetable oil	1,700	20
Boron nitride nanosheets	Used pump oil	2,900	22
Carbon nanofiber aerogels	Vegetable oil	8,000	16
Magnetic carbon nanotubes sponges	Diesel oil	6,600	21
Exfoliated graphite	Heavy oil	8,300	18d
Carbon nanotube sponges	Gasoline	13,500	23b
Carbon aerogels	Pump oil	32,100	23a
3D graphene framework	Gasoline	27,700	17
h–G NSs	Used pump oil	10,700	present work

Table S3 Summary of Congo red maximum adsorption capacities (Q_m) on various adsorbents.

Adsorbents	Adsorption capacity (mg g^{-1})	Reference
MnO_2 hierarchical hollow nanostructures	80	24b
Multishelled Co_3O_4–Fe_3O_4 hollow spheres	125	24c
FeOOH hierarchical nanostructures	240	24d
α–FeOOH hollow spheres	275	24a
Boron nitride nanosheets	782	22
h–G NSs	907	This work