

***Electronic Supplementary Information for:***

***Scalable fabrication of high quality graphene with excellent  
supercapacitor performance by exfoliation of edge sulfonated graphite***

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## Experimental section

### 1. Detailed procedure for the functionalization of graphite with 4-toluene sulfonyl chloride.

All chemicals were of analytical grade and used without further purification. In a typical procedure, to a 100 mL flask equipped with a magnetic stir bar, graphite (100 mg), 4-toluene sulfonyl chloride (Ts-Cl) (2.0 g),  $\text{AlCl}_3$  (3.0 eq. based on Ts-Cl) were sequentially added in fresh 1,2-dichloroethane (30 mL). The reaction mixtures were stirred at 100 °C under air for 48 h. After cooling down, the mixtures were diluted with EtOH, filtered and washed with EtOH and deionized (DI) water. The obtained precipitates were dispersed in NMP and sonicated with low-power. Then, by the means of centrifugation at 10000 rpm, the supernatant was dumped and collected, subsequently, a large number of flocculent precipitate that is TsG occurred, when the appropriate amount of DI water was added. The precipitate was filtered, washed using DI water, and vacuum freeze-dried (85% yield according to the formula:  $m_{\text{TsG}}/(m_{\text{G}}+m_{\text{Ts}})$   $m_{\text{TsG}}$  is the total mass of TsG obtained,  $m_{\text{G}}$  is the total mass of pristine graphite,  $m_{\text{Ts}}$  is the total mass of Ts added).

### 2. Characterization

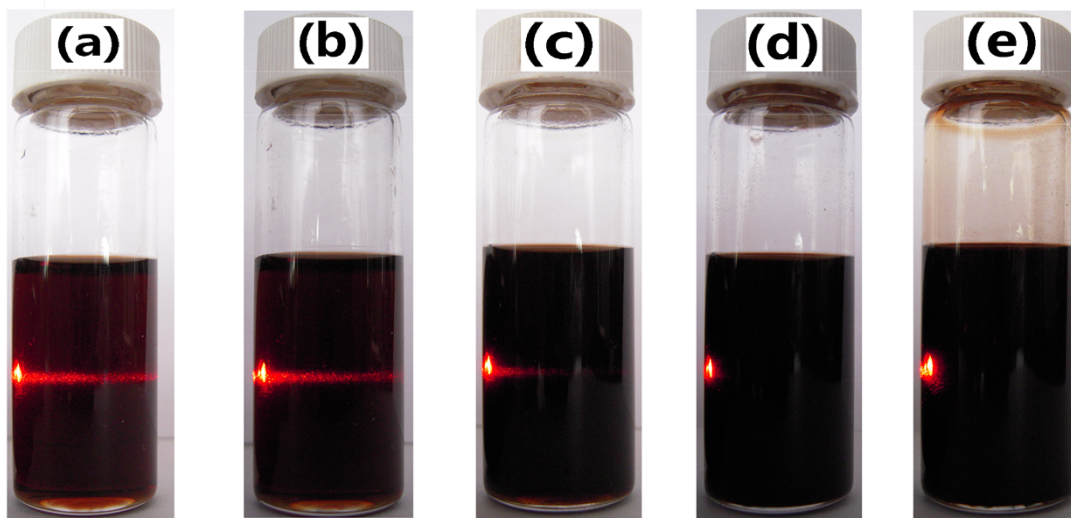
Microstructural characteristics of pristine graphite and TsG were characterized by transmission electron microscope (TEM, FEI Tecnai G2 F20) and high-resolution TEM working at 200 kV accelerating voltage and the lattice structure was identified by selected area electron diffraction (SAED) technique. The height of TsG was tested on atomic force microscope (AFM, VEECO Nanoscope IIIa). The crystallographic structure of the sample was analyzed by X-ray diffraction (XRD, Rigaku D/Max-2400 diffractometer using  $\text{Cu-K}\alpha$  radiation and graphite monochromator,  $\lambda = 1.54056 \text{ \AA}$ ) over the  $2\theta$  ranging from 5° to 80°. A Fourier transform infrared spectroscope (FTIR, IFS 66 V/S Bruker, Germany) was used for recording the FTIR spectra of TsG ranging from 400 to 4000  $\text{cm}^{-1}$ . Raman spectroscopy was accomplished at Renishaw Raman microscope with 633 nm line of an Ar ion laser as an excitation source. Nitrogen adsorption-desorption measurements were conducted at Micromeritics

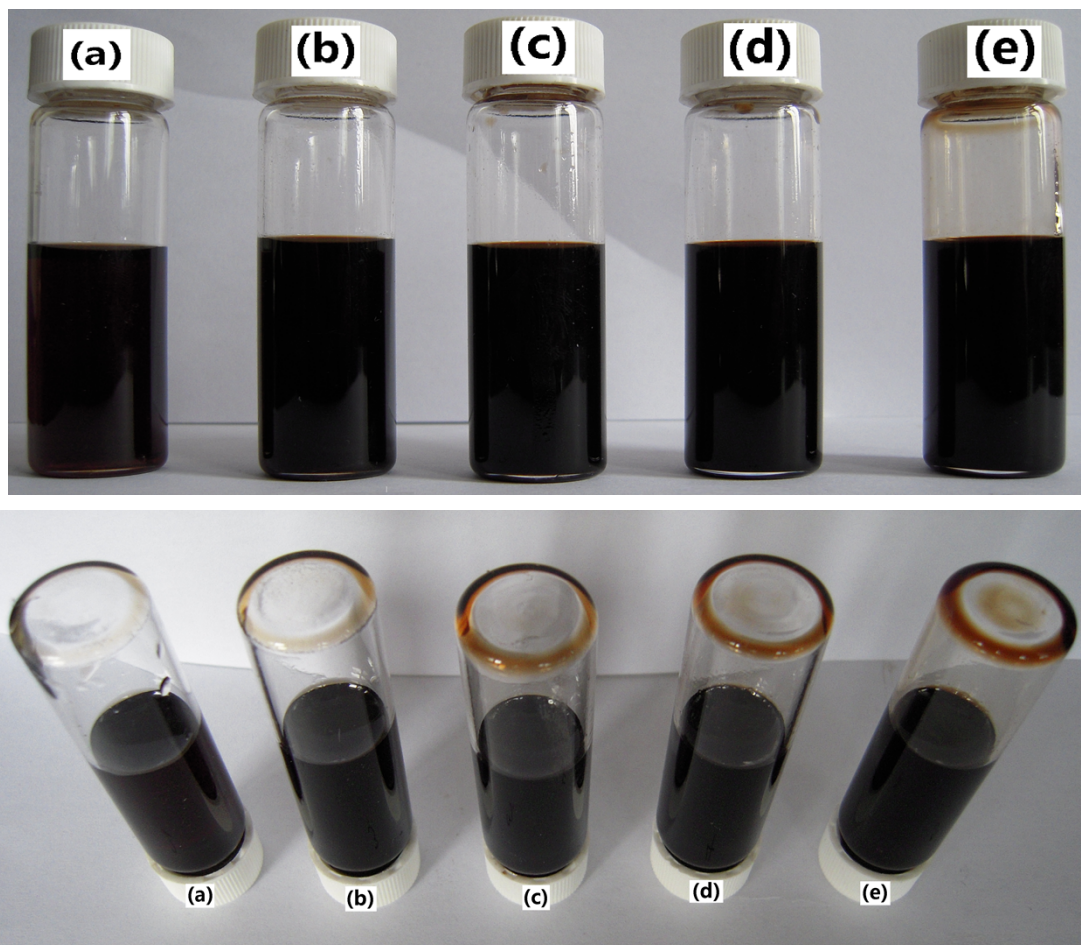
ASAP 2020 apparatus. Specific surface areas were determined according to the Brunauer-Emmet-Teller analysis.

### 3. Electrochemical measurement

The working electrodes were fabricated by mixing the as-prepared S-graphene (4 mg), acetylene black, and polytetrafluoroethylene (1% wt) with a mass ratio of 85 : 10 : 5. The mixture was ground adequately to form a slurry, then was coated onto stainless steel, pressed at 10 MPa, and dried under vacuum at 60 °C for 24 h. A Pt sheet and a saturated Ag/AgCl electrode were employed as the counter and reference electrodes, respectively. 6 M KOH aqueous solution was used as the electrolyte. All the electrochemical characterization was performed in a standard three-electrode system controlled by CHI 660C electrochemical workstation.

### 4. Digital images of TsG dispersed in NMP at various concentrations

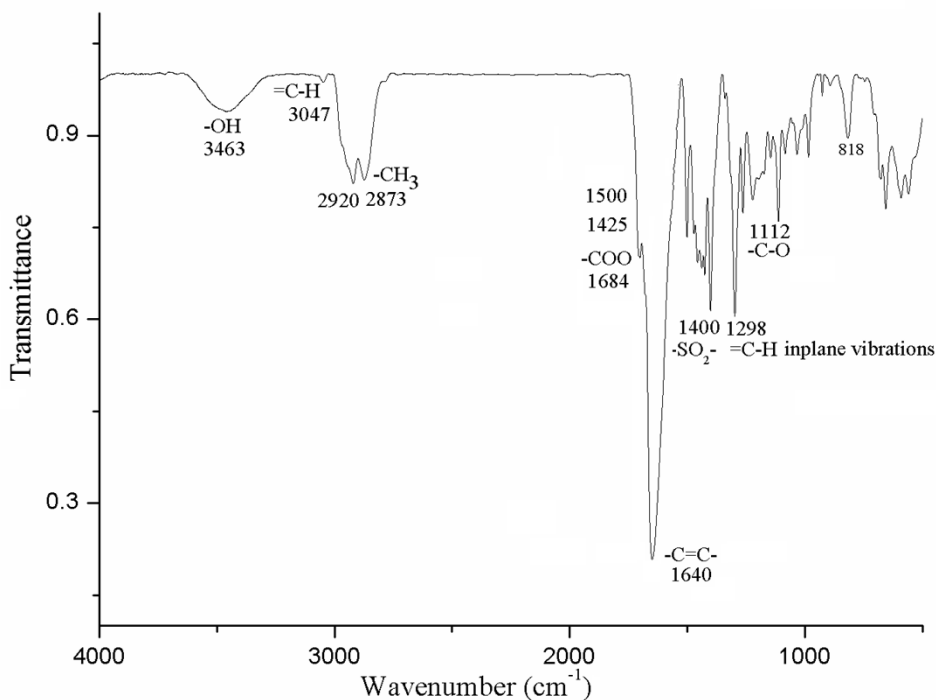




**Figure S1.** Digital photographs of different concentrations of TsG in NMP with a red beam from hand-held laser pointer (top), and without one (middle), and after one month (bottom). (a) 2 mg/mL, (b) 4 mg/mL, (c) 8 mg/mL, (d) 16 mg/mL, and (e) 20 mg/mL.

As shown above the concentrations of TsG in NMP, when the concentration of TsG reaches up to 20 mg/mL, visually discernable precipitations are observed (Fig. S1e). Additionally, a little of precipitations are generated with the increase of concentration, when the samples are settled after one month, indicating TsG enjoys good stability and dispersibility in NMP medium.

## **5. FTIR spectrum and functional group analyses of TsG**



**Figure S2.** FT-IR spectrum of TsG.

The peak at  $3463\text{ cm}^{-1}$  is attributed to -OH and the peaks at  $2920$  and  $2873\text{ cm}^{-1}$  are designated as the asymmetric stretching and symmetric vibrations of  $-\text{CH}_3$ . The peaks at  $3047\text{ cm}^{-1}$  and  $1298\text{ cm}^{-1}$  are  $=\text{C-H}$  stretching vibrations and in-plane vibrations,<sup>[1]</sup> respectively. The bands at  $1500$  and  $1425\text{ cm}^{-1}$  are assigned to typically aromatic ring skeleton vibrations. The peaks at  $1684\text{ cm}^{-1}$  and  $1112\text{ cm}^{-1}$  represent the  $-\text{COO}$  (asymmetric) and  $-\text{C-O}$  stretching vibrations, respectively. The peak at  $1640\text{ cm}^{-1}$  is the  $\text{C}=\text{C}$  backbone stretching vibrations. The peak at  $1400$  is corresponding to  $-\text{SO}_2-$  stretching vibrations.<sup>[2]</sup> The peak at  $818\text{ cm}^{-1}$  indicated the group  $-\text{CH}_3$  is located at *para*-orienting on the aromatic ring.

## References

- [1] C. H. Xu, J. Sun and L. Gao, *J. Mater. Chem.*, 2011, **21**, 11253-11258.  
 [2] R. Bogoczek and E. K. Balawejder, *Reactive Polymers*, 1987, **7**, 57-60.