## **Electronic Supplementary Information (ESI)**

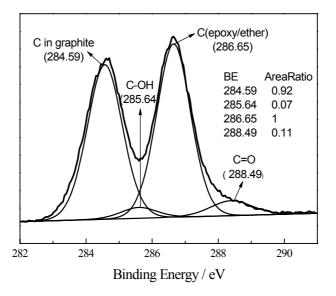
# Convenient Preparation of Tunably Loaded Chemically Converted Graphene oxides/ Epoxy Resin Nanocomposite from Graphene Oxide Sheets through

## **Two-phase Extraction**

## **Experimental Section**

**Preparation of graphene oxide (GO).** Graphene oxide (GO) was prepared by oxidizing natural graphite powder (SP, 320 mesh) based on a modified Hummers method as originally presented by Kovtyukhova and colleagues.<sup>1,2</sup> As-prepared graphene oxide was suspended in ultra-pure water to give a brown dispersion, which was subjected to dialysis to completely remove residual salts and acids for 4 days.<sup>3</sup> The resulting purified graphene oxide powders were collected by centrifugation and air-dried. Graphene oxide powders were dispersed in water to create a 0.05 wt% dispersion. Then the dispersion were exfoliated through ultrasonication for 1 h, which the bulk graphene oxide powders were transformed into GO nanoplatelets.

**Preparation of water-soluble chemically converted graphene nanosheets.** water-soluble chemically converted graphene nanosheets was synthesized from as-purified graphene oxide nanosheets.<sup>3,4</sup> Briefly, 20 mL graphene oxide sheets (in water, 0.05 wt%) was mixed with 20 mL water, 0.012 mL hydrazine solution (50% in water, Beijing Yili Chemicals, China) and 0.142 mL ammonia solution (25% in water, Beijing Chemicals, China) in a 100 mL glass vial. After being vigorously shaken or stirred for a few minutes, the GO nanosheets were reduced to graphene nanoplatelets by refluxing the mixture for 1 h under a oil bath (95 °C). The final homogenous graphene sheets dispersion was ready for use, in control experiment.



### Fig. S1. C1s XPS spectra of GO sheets.

To assess the degree of oxidation, XPS analysis was firstly carried out on GO films on silicon wafers. The C1s XPS spectra of GO (as shown in Fig. S1) clearly indicates a considerable degree of oxidation with four components corresponding to carbon atoms in different functional groups <sup>4,5</sup>: the C in graphite (BE, 284.58 eV), the C in C-OH (BE, 285.64 eV), the C in C-O epoxy/ether groups (BE, 286.65 eV), and the carbonyl C (BE, 288.49 eV). Based on the area ratios of these groups, we can calculate the degree of oxidation and the degree is about 56.2%. Among these groups, the degree of oxidation of it is clear that the C-O epoxy/ether group be in the majority. To further assess the functional groups present in the GO sheets, FTIR spectrum was acquired (as shown in Fig. S2), revealing that the characteristic band of the carboxylic group in GO appeared at ca. 1729 cm<sup>-1</sup> (C=O stretching)<sup>3,6</sup> and the C-O vibrations of epoxy groups in GO appeared at ca. 1139 and 873 cm<sup>-1</sup>. This is consistent with our XPS analysis.

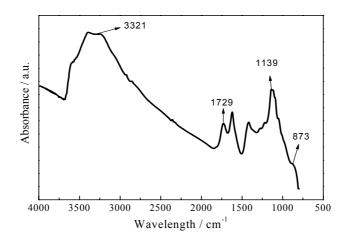


Fig. S2. FTIR spectrum of GO sheets.

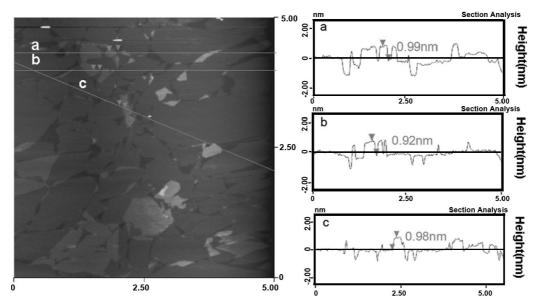


Fig. S3. AFM image and height profile of GO sheets.

The well-exfoliated samples of GO sheets were imaged using tapping mode AFM as shown in Fig. S3. On average, height of the GO sheets is ca.0.96 nm. While a pristine graphite sheet is atomically flat with a well-known van der Waals thickness of ca. 0.34 nm<sup>7</sup>, the GO is expected to be thicker mainly owing to the presence of epoxy groups on the GO sheets.

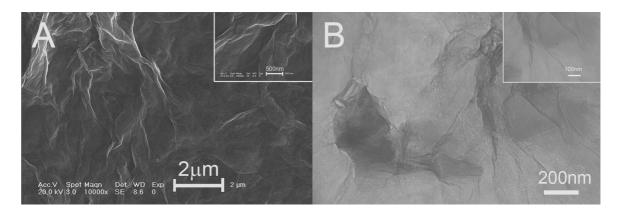
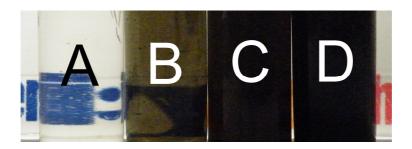
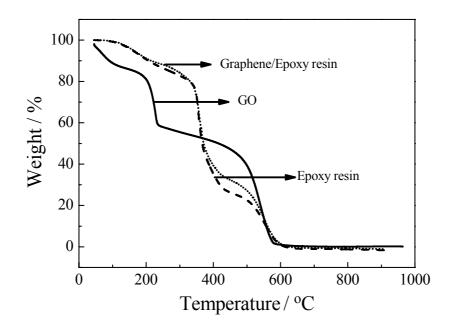


Fig. S4. (A) SEM and (B) TEM images of GO sheets.



**Fig. S5.** Photos of neat epoxy resin monolith and graphene sheets/epoxy resin monoliths. From B to D, three graphene sheets/epoxy resin bulk monoliths were prepared at different graphene sheets weights (0.0075wt%, 0.01875wt%, and 0.0375wt%, respectively).



**Figure S6.** TGA curves of GO (solid), Epoxy resin (dashed), and Graphene/Epoxy resin (dotted).

### References

- 1. Hummers W-S, Offeman R-E (1958) Preparation of Graphitic Oxide. *J Am Chem* Soc 80: 1339.
- 2. Kovtyukhova N-I, *et al.*(1999) Layer-by-layer assembly of ultrathin composite films from micron-sized graphite oxide sheets and polycations. *Chem Mater* 11:771-778.
- 3. Li D, Muller M-B, Gilje S, Kaner R-B, Wallace G-G (2008) Processable aqueous dispersions of graphene nanosheets. *Nat Nanotech* 3: 101-105.
- Stankovich S, *et al.* (2006) Stable aqueous dispersions of graphitic nanoplatelets via the reduction of exfoliated graphite oxide in the presence of poly(sodium 4-styrenesulfonate). *J Mater Chem* 16: 155-158.
- Becerril H-A, *et al.* (2008) Evaluation of Solution-Processed Reduced Graphene Oxide Films as Transparent Conductors. *ACS Nano* 2: 463-470.
- Szabo T, *et al.* (2006) Evolution of surface functional groups in a series of progressively oxidized graphite oxides. *Chem Mater* 18: 2740-2749.
- 7. Liu N, et al. (2008) One-step ionic-liquid-assisted electrochemical synthesis of

ionic-liquid-functionalized graphene sheets directly from graphite. Adv Funct Mater

18: 1518-1525.