# Mechanical, thermal and swelling properties of poly(acrylic acid)/graphene oxide composite hydrogels

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# **Electronic Supplementary Information**

## 1. FTIR analysis

Figure S1 shows the FTIR spectra of raw graphite and GO. Comparing with raw graphite, it can be seen that the peak at 3450 cm<sup>-1</sup> of GO is according to the -OH vibrational stretching. Besides, it also shows bands due to carboxyl C=O and C-O (1650 and 1400 cm<sup>-1</sup>), epoxy C-O (1250 cm<sup>-1</sup>) and alkoxy C-O (1100 cm<sup>-1</sup>) groups situated at the edges of the GO sheets. FTIR analysis of GO confirms that there are many functional groups on the surface of GO.



Figure S1. FTIR spectra of raw graphite and GO

## 2. Raman analysis

Raman spectroscopy offers an efficacious tool to probe the structural characteristics and properties of graphene and graphene- based materials. Figure S2 shows the Raman spectra of raw graphite and GO. In the spectrum of pristine graphite,

the peak at 1580 cm<sup>-1</sup> (G band) corresponds to an  $E_{2g}$  mode of graphite and is related to the vibration of sp<sup>2</sup>-bonded carbon atoms in a 2D hexagonal lattice. The peak at 1350 cm<sup>-1</sup> (D band, the breathing mode of  $\kappa$ -point phonons of  $A_{1g}$  symmetry) is associated with vibrations of carbon atoms with dangling bonds in plane terminations of disordered graphite. Comparing with raw graphite, G mode of GO becomes weaker and broader, suggesting a higher level of disorder of the graphene layers. Besides, the ratio of the intensities ( $I_D/I_G$ ) for GO samples is markedly increased, indicating the formation of some sp<sup>3</sup> carbon by functionalization. This phenomenon can be attributed to the significant decrease of the size of the in-plane sp<sup>2</sup> domains due to oxidation and ultrasonic exfoliation, and partially ordered graphite crystal structure of graphene nanoplatelets.<sup>1,2</sup>



Figure S2. Raman spectra of raw graphite and GO

#### 3. TGA analysis

TGA is a complementary technique that can reveal the composition and changes in thermal stability of the tested samples. Figure S3 shows the TGA curve of raw graphite and GO. It can be seen that TGA trace of raw graphite shows little weight loss, which is about 3% below 700 °C. Comparing with the raw graphite, GO shows much lower thermal stability, which is because of the lowered thermal stability due to the reduced van der Walls interaction. Besides, the onset temperature becomes dramatically lower, presumably due to pyrolysis of the labile oxygen-containing functional groups, yielding CO, CO<sub>2</sub> and steam. The main mass loss takes place around 100~400 °C and is ascribed to the decomposition of labile oxygen functional groups present in the material.<sup>3</sup> There is also a mass loss (~ 3%) below 100°C attributed to the removal of adsorbed water and a slower, steady mass loss (~10%) over the whole temperature range between 400°C~700 °C, which can be assigned to the removal of more stable oxygen functionalities.



Figure S3 TGA curves of raw graphite and GO

#### 4. XRD analysis

Figure S4 shows powder XRD result of raw graphite and GO. For the pristine graphite (figure S4a), the sharp and intensive peak positioned at  $26^{\circ}$  (002) is corresponding to an interlayer *d*-spacing of 0.34 nm, which indicates a highly organized crystal structure. However, the feature diffraction peak appears at 10.6° for GO (figure S4b), corresponding to an interlayer *d*-spacing of 0.78 nm.



Figure S4 XRD patterns of raw graphite and GO

# 5. SEM analysis

From the SEM image of GO (figure S5), it can be found that solid GO shows rough surface, which might be due to the oxidation of sheets.



Figure S5 SEM image of GO

## 6. TEM analysis

From TEM image of GO (figure S6), the layered structure of the solid GO sample with stacked GOS can be clearly seen. It appears to have rough surface which might be due to the oxidation of sheets. In addition, large sheets are observed to be situated on the top of the grid, where they resemble silk veil waves, illustrating the flake-like shapes of graphene.



Figure S6 TEM image of GO (1mg/mL)

## 7. AFM analysis

AFM characterization has been one of the most direct methods of quantifying the

degree of exfoliation to graphene level after the dispersion of the powder in a solvent. We can find that GO consists of flat sheets with an average thickness of about 1.0 nm. It is known that the basal planes of the GO sheets are decorated mostly with epoxide and hydroxyl groups, in addition to carbonyl and carboxyl groups. These oxygen functionalities will alter the van der Walls interactions between the layers and make them hydrophilic, thus facilitating their exfoliation in aqueous media. As a consequence, GO readily forms stable colloidal dispersions in water. Since a pristine graphene sheet is atomically flat with a well-known van der Waals thickness of 0.34nm, GO are expected to be "thicker" due to the presence of covalently bonded oxygen and the displacement of the sp<sup>3</sup>-hybridized carbon atoms slightly above and below the original graphene plane. Considering the XRD results, we can expect that these sheets are uniformly monolayers and bilayers.



Figure S7 AFM image of GO (1mg/mL)

# Reference

- 1. Y. Xu, H. Bai, G. Lu, C. Li, G. Shi, J. Am. Chem. Soc. 2008,130, 5856-5857.
- S. Stankovich, D. A. Dikin, R. D. Piner, K. A. Kohlhaas, A. Kleinhammes, Y. Jia, Y. Wu, S. T. Nguyen, R. S. Ruoff, *Carbon* 2007, 45, 1558-1565.
- E. Yoo, J. Kim, E. Hosono, H. Zhou, T. Kudo, I. Honma, *Nano. Lett.* 2008, 8, 2277-2282.