

## Supporting information

### One-pot, aqueous-phase synthesis of graphene oxide functionalized with heterocyclic groups to give increased solubility in organic solvents

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#### Experimental Section

All chemicals were purchased from Sigma-Aldrich and used without further purification.

#### Preparation of GO

GO was prepared by a modified Hummers method. In a typical procedure, graphite (1.0 g) was added into concentrated H<sub>2</sub>SO<sub>4</sub> (98%, 50 ml) under stirring in an ice bath. Then 6.0 g of KMnO<sub>4</sub> was added slowly to the suspension. The mixture was stirred at 30 °C for 1 hour. Subsequently, 80 ml

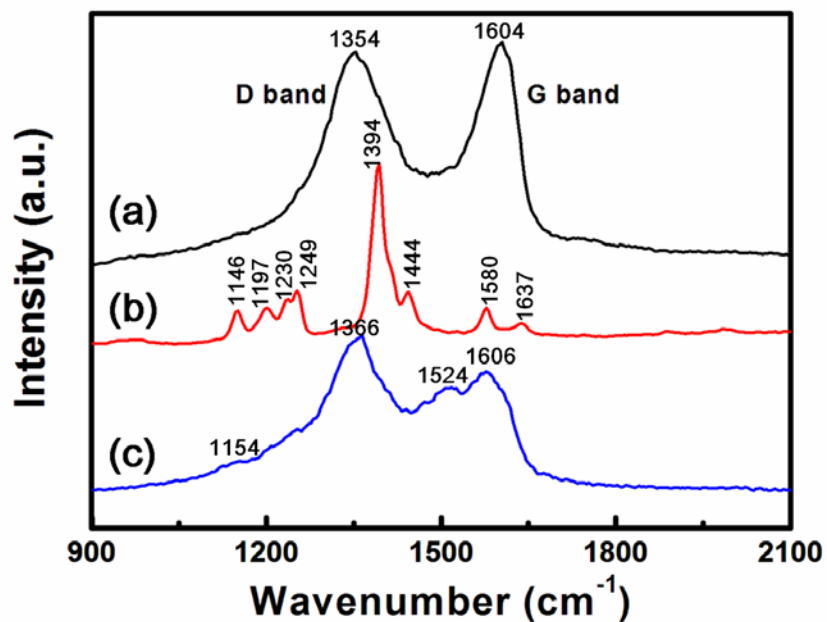
deionized water was added under vigorous stirring with increasing the temperature to 90 °C, and further stirred for 30 min. Finally, 200 ml deionized water and 6 ml of H<sub>2</sub>O<sub>2</sub> (30%) was added, along with the color of solution changing from dark brown to yellow. The diluted suspension was stirred for additional 10 min, graphite oxide was obtained by repeated washing (in water) and centrifugation step more than 9 times, and finally freeze-dried in a freeze drying machine.

### **Preparation of OAP and OPD functionalized GO**

160 mg GO was suspended in 80 ml deionized water to form a homogeneous suspension (2 mg/ml) by sonication for 30 min, then 7 mmol OAP or OPD was added with further sonication for 5 min. After that, 1 ml PPA was dropwised by vigorous stirring for 7 days under the darkness condition at room temperature. The product was collected by repeated filtration and washed with HCl/H<sub>2</sub>O (V/V= 1:2) (this procedure was repeated for 4 times), and subsequently washed with deionized water for six times. The benzoxazole and benzimidazole covalently grafted GO was obtained by drying under vacuum condition for 24 hours.

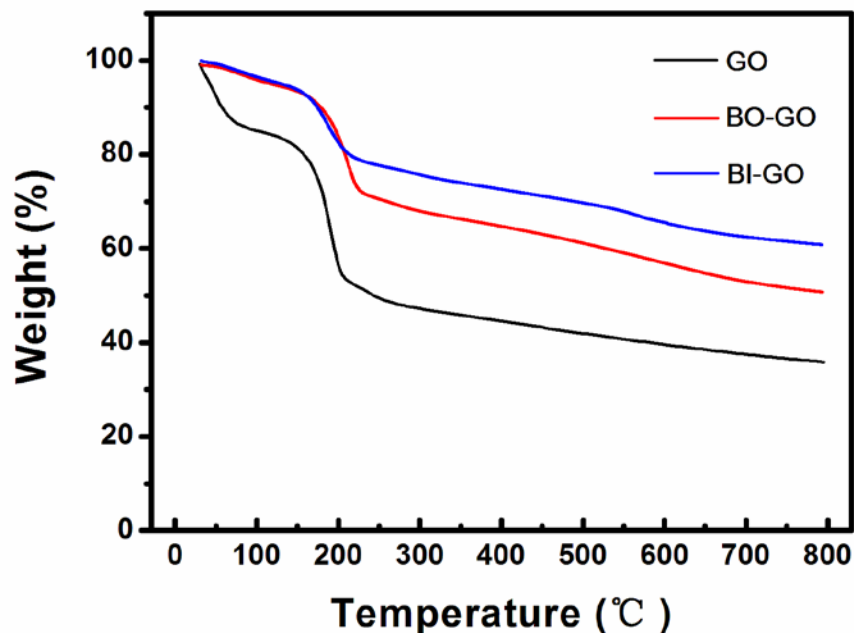
### **Characterization**

Fourier-transformed infrared spectra (FT-IR) were measured using the KBr-pellet method in transmission mode on a NEXUS 670 FT-IR spectrometer. X-ray photoelectron spectroscopy (XPS) analysis was performed on an ESCALAB MK II X-ray photoelectron spectrometer using Al K $\alpha$  (1486.6eV) X-ray source. Scanning electron microscopy (SEM) analysis was conducted with a JEOL JSM-6700F electron microscope. Atomic force microscopy (AFM) was carried out using a Digital Nanoscope V in the tapping mode. Powder X-ray diffraction (XRD) analysis was performed using a D8 Advanced diffractometer with Cu-K radiation ( $\lambda = 1.54056 \text{ \AA}$ ). Raman spectra were collected using a WITEC CRM200 Raman system with 532 nm excitation laser. Thermogravimetric analysis (TGA) was recorded by a Shimadzu DTG-60H under a heating rate of 10°C/min and a nitrogen flow rate of 50 cm<sup>3</sup>/min.



**Fig. S1** Raman spectra of GO (a), BO-GO (b) and BI-GO (c).

Raman spectra of GO, BO-GO and BI-GO are shown in Fig. S1. The Raman spectrum of GO shows an intense tangential mode (G band) at 1604 cm<sup>-1</sup>, with a disordered-induced peak (D band) at 1354 cm<sup>-1</sup> (Fig. S1a). After the functionalization with the *o*-aminophenol (OAP), BO-GO exhibits the G at 1580 cm<sup>-1</sup> and D bands at 1394 cm<sup>-1</sup> (Fig. S1b). The BO-GO shows six typical peaks at 1146, 1197, 1230, 1249, 1444 and 1637 cm<sup>-1</sup>, which are attributed to the Raman bands of oxazole rings grafted on GO sheets.<sup>1</sup> However, after its functionalization with the *o*-phenylenediamine (OPD), BI-GO exhibits the G at 1606 cm<sup>-1</sup> and D bands at 1366 cm<sup>-1</sup> (Fig. S1c). Furthermore, the BI-GO also shows two typical peaks at 1154 and 1524 cm<sup>-1</sup>, which are attributed to the vibrational frequencies of imidazole rings grafted on GO sheets.<sup>2</sup> The value of I(D)/I(G) is ~0.99 for GO while the value of I(D)/I(G) is increased to ~3.93 for BO-GO and ~1.13 for BI-GO. The increased value is due to the covalent functionalization of GO with OAP and OPD that cause many defects or de-oxygenation, that resulting in partially restore of the graphene network.<sup>3,4</sup>



**Fig. S2** Thermogravimetric curves of GO, BO-GO and BI-GO at a heating rate of  $10\text{ }^{\circ}\text{C min}^{-1}$  under nitrogen.

The thermogravimetric analysis (TGA) was also performed on the functionalized GO to assess their thermal stabilities at a heating rate of  $10\text{ }^{\circ}\text{C min}^{-1}$  under  $\text{N}_2$  atmosphere as shown in Fig. S2. It can be seen that both GO and functionalized GO (BO-GO and BI-GO) start to lose mass even below  $100\text{ }^{\circ}\text{C}$  due to the removal of adsorbed water molecular trapped between the sheets.<sup>5</sup> However, a relatively large loss of mass during the period of  $150\text{--}200\text{ }^{\circ}\text{C}$  was observed, which is attributed to the decomposition of the labile oxygen-contained groups in GO-based materials.<sup>6</sup> Furthermore, compared to GO, the mass loss rate with temperature for BO-GO and BI-GO is significantly lower and more residues are observed, indicating the improved thermal stability of functionalized GO.

## References

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