Supporting Information

Understanding room-temperature metastability of graphene oxide utilizing hydramines from a synthetic chemistry view

Yueyue Qian,^a Shupeng Zhang^{*a}, Juanjuan Gao^a and Haiou Song^{*b}

^{a.} School of Chemical Engineering, Nanjing University of Science and Technology, Nanjing, Jiangsu,

210094, China. E-mail: shupeng_2006@126.com

^{b.} State Key Laboratory of Pollution Control and Resource Reuse, School of the Environment, Nanjing

University, Nanjing, Jiangsu, 210023, China, E-mail:songhaiou2011@126.com

1 Experimental

1.1 Materials

Natural graphite was obtained from Qingdao Zhongtian Company. Ethanol amine (AE), amino-1propanol (AP), 4-amino-1-butanol (AB) and others chemical reagents were analytical grade and used as received without further purification.

1.2 Fabrication of HA functionalized graphene oxide nanomaterials

Graphene oxide used in this work was synthesized by oxidation of natural graphite power using the modified Hummers method. As a typical example, the sample prepared with powdery GO and Ethanol amine (AE) is labeled as AE-mGO for easy expression. The synthesis procedure for a typical well-dispersed AE-mGO compound was carried out as follows: 200 mg of GO was added into 15 mL of *N*, *N*-dimethylforamdie under ultrasonication for 7 h and in this process GO was completely exfoliated into

individual sheets to form a stably dispersed GO/DMF solution. 1 g of AE was also dissolved in CHCl₃ (5 mL). Subsequently, a dispersion of AE was added dropwise to the GO solution and stirred vigorously for 2 d at room temperature. The mixture was then washed several times with CHCl₃ and ethanol to remove any impurities by filtration. Finally, the product was dried at 70 $^{\circ}$ C for 7 h in vacuum oven.

For the preparations of the AP-mGO and AB-mGO, all procedures are identical except the raw material is switched from AE to AP and AB respectively.

1.3 Fabrication of the PVA/1 wt%(HA-mGO) polymer nanocomposites

In our case, PVA and HA-mGO can all be readily dispersed in D.I. water, which will help to achieve molecular level dispersion by solution blending. For the preparation of PVA/1 wt% (HA-mGO) polymer nanocomposites, the 1 % *wt*. HA-mGO nanofillers were first dispersed in water by ultrasonication. 1 g of PVA was also dissolved in water. Then, a dispersion of HA-mGO was dropwise added to the PVA solution and stirred vigorously for 10 h. At last, the composite solution was casted and dried at room temperature for 48 h and then in a vacuum oven for 7 h at 70 °C.

1.4 Characterization

FT-IR spectroscopy of GO-based hybrid materials were collected on a Nicolet IS-10 FT-IR spectrometer equipped with a Smart OMNI sampler with a high purity Ge crytal. Raman spectra of samples were measured from 1000 to 2000 cm⁻¹ on a LabRAM ARAMIS (HORIBA Jobin Yvon S.A.S.) using an excitation wavelength of 532 nm. XRD analyses were performed on a Bruker D8 Advance diffractometer with Cu-K α radiation. The diffraction data was recorded for 2 θ angles between 5° and 60°. The structure and morphology of the products were investigated by a JEOL JSM- 6380LV SEM. The elemental mappings were performed on a Zeiss Supra 40 field emission scanning electron microscope. Thermogravimetric analyses (TGA) were preformed on a Mettler TGA/SDTA851e thermogravimetric

analyzer over the temperature range of 50-800 °C at a heating rate of 20 °C/min in a dry nitrogen atmosphere. X-ray photoelectron spectroscopy (XPS) analysis was obtained using PHIQuantera II with C60 high resolution spectrometer with a monochromatized Al Ka X-ray source (1486.71 eV photons) to analyze thechemical composition of the materials.



Fig. S1 FT-IR spectra (b) of GO, AE, AP, AB, AB-mGO, AP-mGO and AE-mGO.



Fig. S2 The clear layer shape structure of AP-mGO.



Fig. S3 Photographs of AB-mGO, AP-mGO, AE-mGO and GO dispersed in water and 8 organic solvents through bath ultrasonication; Approximately 1.5 mg of powder was added to a given volume of solvent (~1.5 mL), in such a way that the resulting nominal concentration was adjusted to 1 mg•mL⁻¹ for all cases. The solvents are listed below, empty bottle as a comparison; water, dimethylsulfoxide, dimethyl formamide, acetone, n-propanol, isobutanol, tetrahydrofuran, methanol, ethanol.



Fig. S4 XRD patterns of PVA-based nanocomposites with different HA-mGO as nanofillers.