

Supporting online materials for

## **Solution-processable graphene nanomeshes with controlled pore structures**

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## 1. Synthesis of GO, rGO and GNMs

**GO.** GO was prepared by the oxidation of natural graphite powder (325 mesh) according to a modified Hummers' method. Briefly, graphite (3.0 g) was added to concentrated sulfuric acid (70 mL) under stirring at room temperature, then sodium nitrate (1.5 g) was added, and the mixture was cooled to 0 °C. Under vigorous agitation, potassium permanganate (9.0 g) was added slowly to keep the temperature of the suspension lower than 20 °C. Successively, the reaction system was transferred to a 35–40 °C water bath for about 0.5 h, forming a thick paste. Successively, 140 mL of deionized (DI) water was added, and the solution was stirred for another 15 min. An additional 500 mL of DI water was added and followed by a slow addition of 20 mL of H<sub>2</sub>O<sub>2</sub> (30%), turning the color of the solution from brown to yellow. The mixture was filtered and washed with 1:10 HCl aqueous solution (250 mL) to remove metal ions followed by repeated washing with water and centrifugation to remove the acid. The resulting solid was dispersed in water by ultrasonication for 1 h to make a GO aqueous dispersion (1 mg mL<sup>-1</sup>). The obtained brown dispersion was then subjected to 30 min of centrifugation at 4000 rpm to remove any aggregates. Finally, it was purified by dialysis for 1 week to remove the remaining salt impurities.

**rGO.** rGO was prepared by reducing 20 mL aqueous dispersion of GO (0.25 mg mL<sup>-1</sup>) by 10 μL hydrazine hydrate (35 wt % , Sigma-Aldrich) with 70 μL ammonia solution (28 wt% in water) at 95°C for 1 h. Successively, the dispersion was filtrated through glass cotton to remove the precipitate and yield a stable black aqueous dispersion of rGO.

**GNMs.** 50 mL rGO dispersion ( $0.1 \text{ mg mL}^{-1}$ ) was mixed with a certain amount of  $\text{HNO}_3$  solution to form a mixture containing  $8 \text{ mol L}^{-1} \text{ HNO}_3$  and then refluxed at  $100^\circ\text{C}$  for 4, 6, 9 or 11 h. Then, the mixture was cooled to room temperature and diluted with 50 mL de-ionized (DI) water. The diluted mixture was filtered through a porous polytetrafluoroethylene membrane with pore size of  $0.2 \text{ }\mu\text{m}$ . The GNMs on the surface of filtrating membrane were washed thoroughly with DI water and ethanol. Finally, they were dispersed in DI water to form a suspension with a typical concentration of  $2 \text{ mg mL}^{-1}$ .

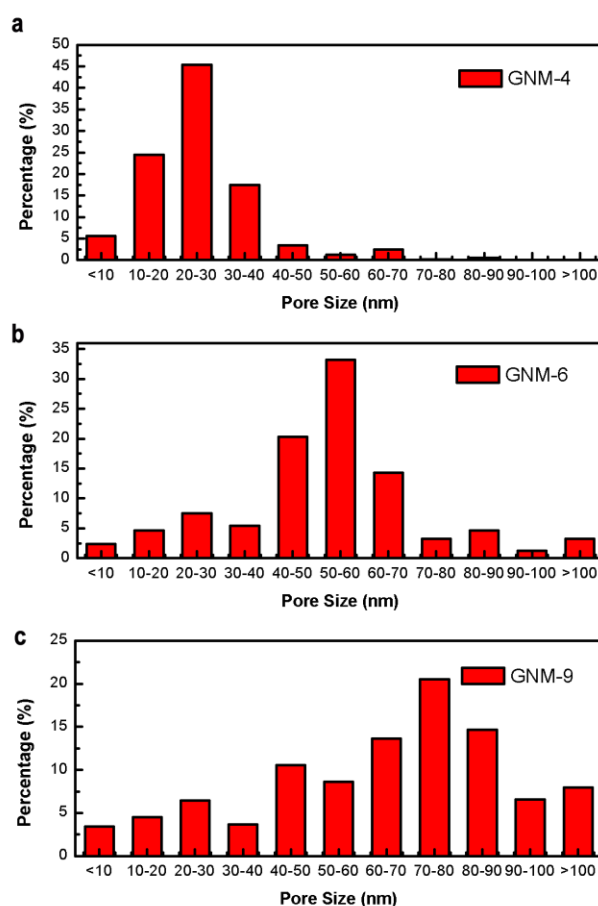
## **2. Fabrication of rGO and GNM films**

A  $10 \text{ mg L}^{-1}$  suspension of rGO or GNMs was used to prepare the desired thin films. To obtain thin films with diverse thicknesses, a specific volume of rGO or GNMs dispersion was diluted to 40 mL with DI water and sonicated for 5 min. The diluted suspension was subjected to vacuum filtration, leading to the deposition of an rGO or GNMs film on the surface of the porous anodic aluminum oxide (AAO, pore size =  $0.2 \text{ }\mu\text{m}$ ) membrane. After drying at  $60^\circ\text{C}$ , the AAO membrane was dissolved with  $1 \text{ mol L}^{-1} \text{ NaOH}$  solution. Subsequently, rGO or GNM film was released from AAO substrate and floated on the solution surface. The NaOH solution was exchanged with copious amount of DI water, and the rGO or GNMs film was transferred to the surface of a poly(ethylene terephthalate) (PET) or quartz substrate and dried at  $60^\circ\text{C}$ .

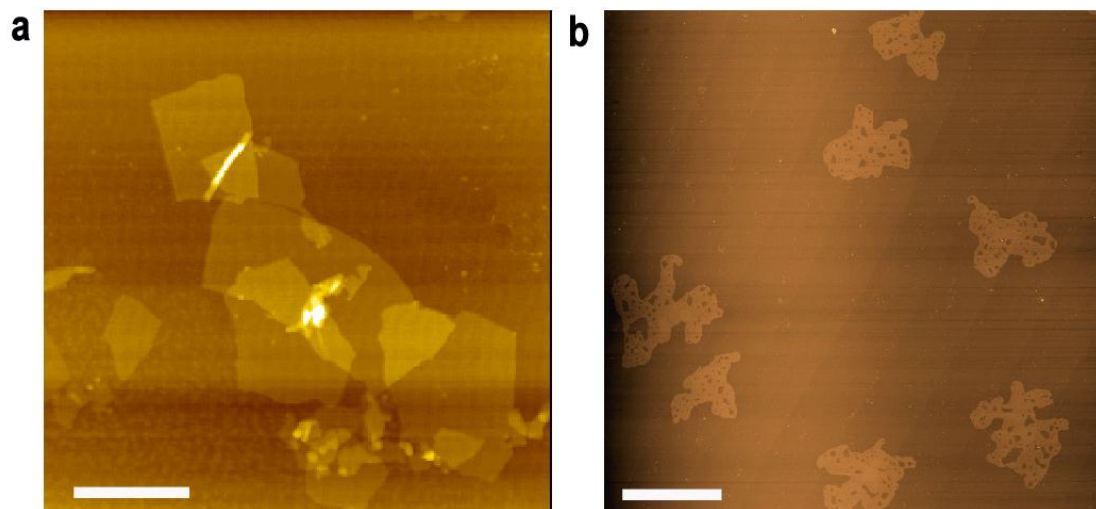
A 21 nm thick GNM film coated on a quartz substrate was further thermally

annealed at 400°C under a vacuum condition. This annealing process was performed in a tube oven equipped with a quality vacuum system. The oven was heated to 100°C under ambient pressure, then the turbo pump was switched on and a vacuum of  $< 10^{-5}$  Torr was established before heated to 400 °C. The heating rate was controlled to be 20°C min<sup>-1</sup>. After annealing at 400 °C for 3 h, the system was cooled automatically to room temperature. Under the ambient condition, the annealed GNM-6 film was collected for characterization.

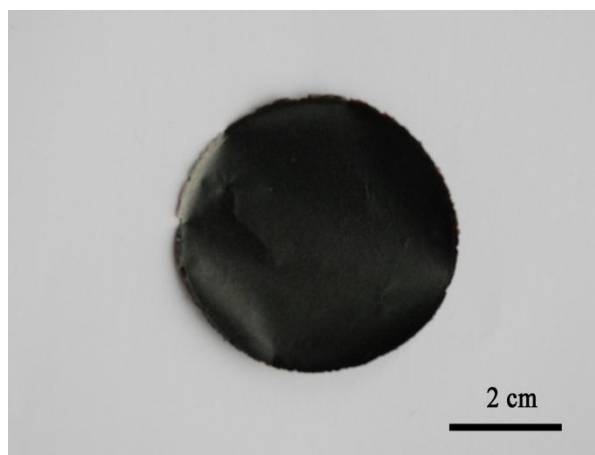
### 3. Supplementary Figures



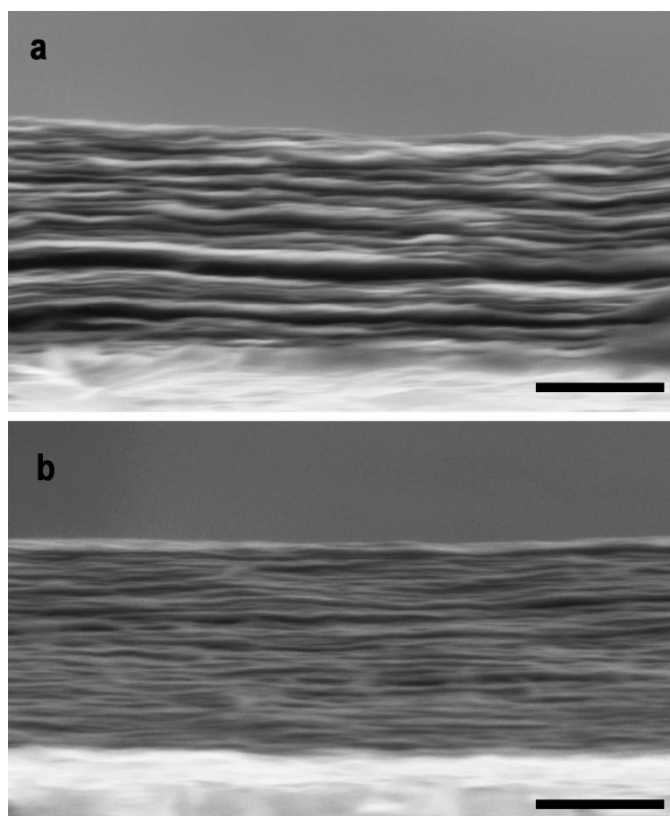
**Figure S1** | The size distribution of nanopores in (a) GNM-4, (b) GNM-6 and (c) GNM-9 sheets.



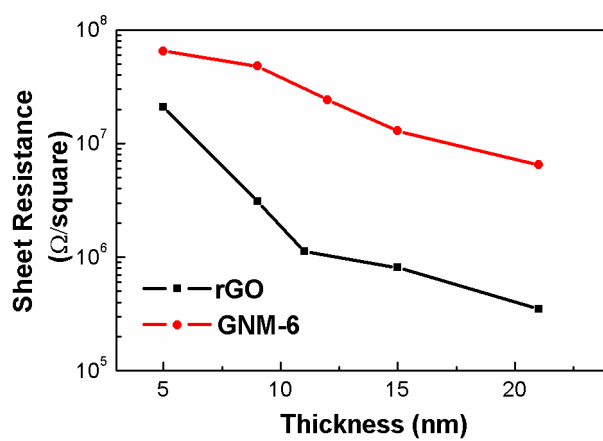
**Figure S2** | Typical AFM images of rGO (a) and GNM-6 sheets (b). Scale bar = 5  $\mu\text{m}$ .



**Figure S3**| Photograph of a GNM-6 paper.



**Figure S4** | Cross-section SEM image of an rGO (a) or a GNM-6 (b) paper. Scale bar = 2 μm.



**Figure S5** | Sheet resistances of rGO or GNM-6 films as a function of film thickness.