

## SUPPORTING INFORMATION

### **Exceptionally high thermal and electrical conductivity of three dimensional graphene foam based polymer composites**

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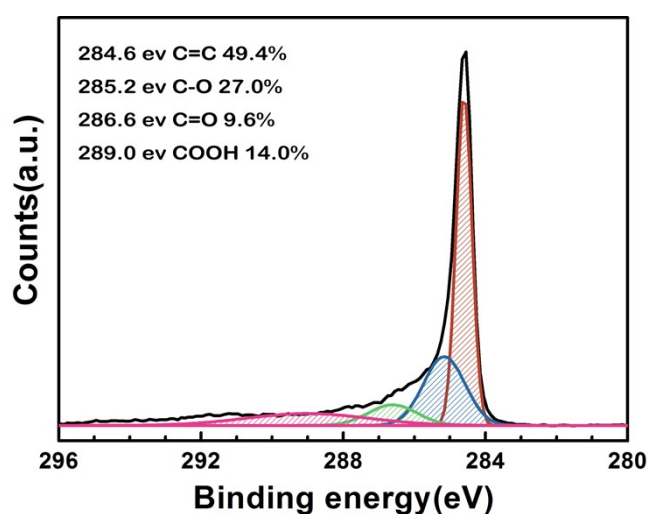
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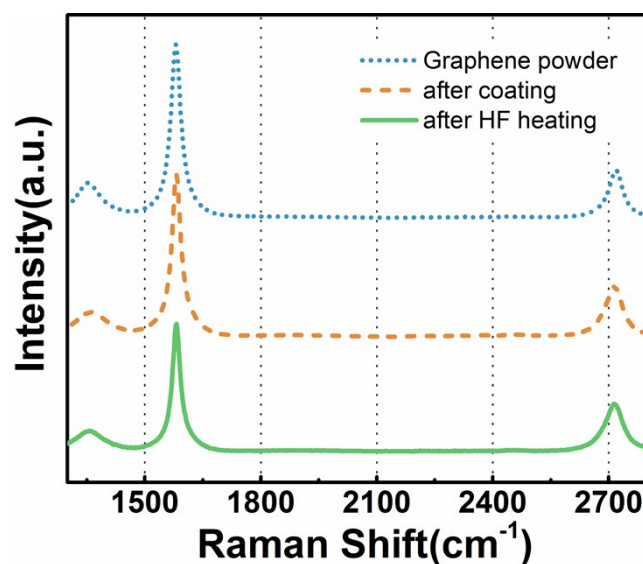
*# These authors contributed equally to this study.*

## SUPPLEMENT

The binding energy profile measured by X-ray photoemission spectroscopy, which was used to further explore the chemical compositions of GNPs, as demonstrated in **Figure S1**. This figure shows  $C_{1s}$  binding energies at 285.2 eV (C-OH), 286.6 eV (C=O), and 289 eV (O=C-OH), respectively, suggesting that some oxygen-containing functional groups are present, where the atomic percentages of various carbon bonds derived from the fitted peak areas are indicated in **Figure S1**.



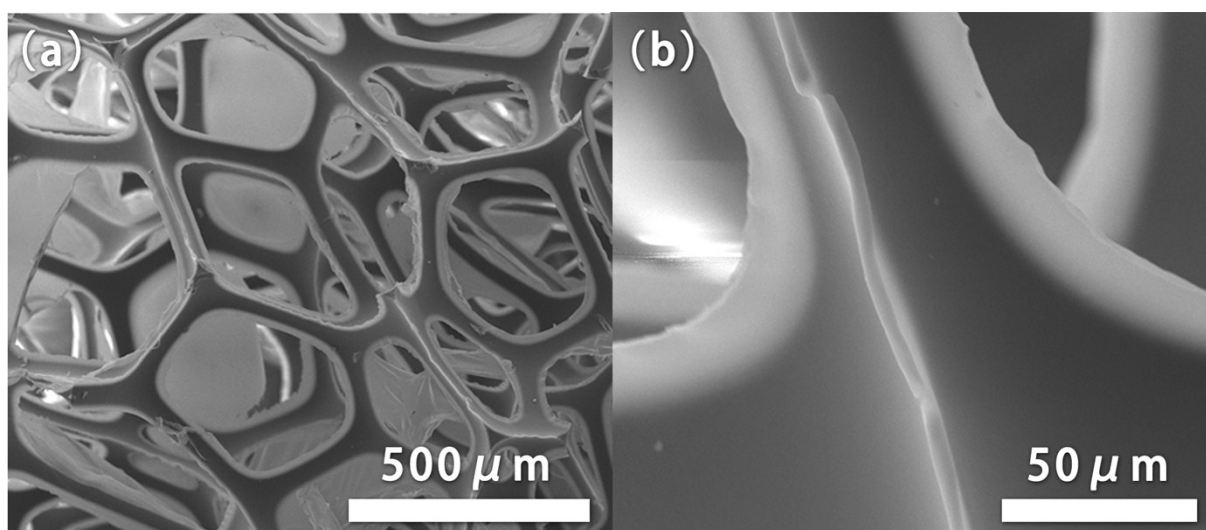
**Figure S1.** XPS spectra of GNP.



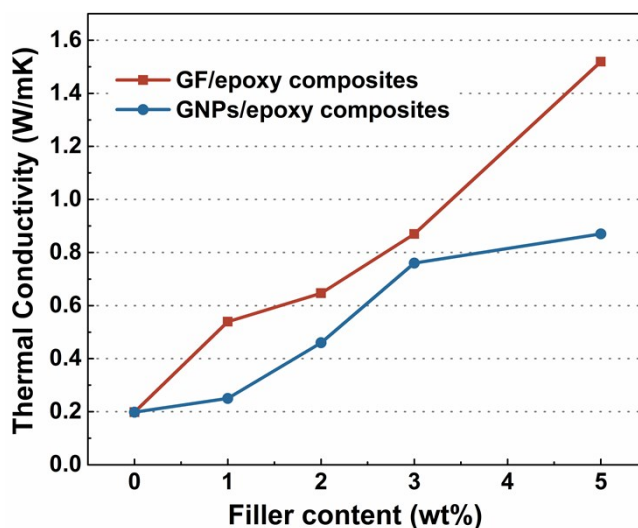
**Figure S2.** Raman spectra of GNP.

Raman spectra were often assigned to be an effective method to characterize carbon-related materials. As can be seen in **Figure S2**, the prominent G peak at 1579  $cm^{-1}$  corresponds to the high-frequency  $E_{2g}$  phonon at  $\Gamma$ . In addition, the D-band (1345  $cm^{-1}$ ) intensity is related to the defect,  $sp^3$ -

hybridized carbon which can be activated at the edges. The  $I_D/I_G$  ratio reflects the degree of the defects in the GNPs structure. The Low  $I_D/I_G$  ratio means high crystalline quality of graphene, which has good thermal conduction property. The decline of  $I_D$  from the graphene powder to the GF after PU decomposition indicates that the defects have been recovered to some extent. The 2D band ( $2708\text{ cm}^{-1}$ ) has been known to be very sensitive to the graphene layer number. From the position and the shape of 2D peak, we can draw a conclusion that graphene flake are constituted by few-layer graphene.



**Figure S3.** (a) Low- and (b) high-magnification SEM images of neat PU foam.



**Figure S4.** Thermal conductivity of the epoxy composites.