Supplementary Information for

Solvent Evaporation Induced Self-assembly of Graphene Foam for Thermally Conductive Polymers

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Figure S1. (a) Raman spectrum of and (b) XPS spectrum of raw graphene material. (c) Raman spectrum and (d) XPS spectrum of GF.

The peak intensity ratio of D band to G band in the Raman spectrum of raw graphene material is 0.29, indicating that the graphene sample has few defects. Deconvoluted XPS C 1s spectra shows that the peak intensity of the sp^2 C=C bonds is much higher than both the C-C and C-O bonds, also verifies the high quality of the raw graphene material. Both the Raman spectrum and XPS spectrum of GF are almost identical to those of the raw graphene material, suggesting that the conjugated structure of graphene was well preserved during the solvent evaporation induced self-assembly of GF.



Figure S2. (a) Photographs of GFs prepared under different proportions of ethanol to water and (b) densities of graphene GFs and graphene film prepared by evaporation of graphene suspension in pure water.

The test method of density is as follows. In the first step, the mass of GF (m) was weighed. Afterwards, the GF was immersed in molten paraffin for four hours. The GFs was then taken out from paraffin and the excess paraffin on the surface of GF was wiped off. The volume (V) of paraffin perfused GF was measured by the water displacement method. The density of the GF sample can be calculated by dividing m by V.



Figure S3. SEM images of GF/1 (a), GF/3 (b), GF/5 (c) and GF/9 (d).



Figure S4. SEM images of GF/E-3 (a), GF/E-5 (b) and GF/E-9 (c). (d) SEM image of graphene/epoxy composite obtained by mechanical mixing. The severely aggregated graphene sheets can be observed in the red square in (d).



Figure S5. Thermogravimetric analysis of GF/epoxy composites and pure epoxy. The higher weight retention after 500 °C indicates higher weight content of graphene in the GFs.



Figure S6. SEM image of the GF after the addition of PVP



Figure S7. Thermogravimetric analysis of PVP.