

Supporting Information

Three-dimensional elastic macroscopic graphene network for thermal management application

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Experimental Details

Preparation for 3D-GN and SA-GN composites

SiO₂ powder (AR, SinoReag) was pressed into ϕ 13 and ϕ 26 mm disks by adding 0.5 wt.% PVA under 5 MPa for 2 min. The ceramic green bodies were placed in the centre of a horizontal Al₂O₃ tube mounted inside a high temperature of 1200 °C at a heating rate of 10 °C/min with a gas flow of H₂ : Ar = 40 : 450 sccm. A small amount of methane (25 sccm) was introduced into the tube at 1200 °C for 15~60 min. After the graphene growth, the sample was cooled down to 400 °C at a rate of 10 °C/min under the flow of H₂ and Ar. SiO₂ substrate was etched in diluted HF solution overnight, and the final free-standing macroscopic 3D-GN was washed repeatedly in distilled water and obtained by freeze drying method.

The phase change material used in the experiment was SA, with a thermal conductivity of SA was 0.15 W m⁻¹ K⁻¹ and a melting temperature is around 56 °C. A certain volume (the weight ratio of 3D-GN to SA is 1: 15) of 3D-GN was putted into the melted SA at 80 °C for 4h, the system was

allowed to cool rapidly under ambient atmosphere to obtain SA-GN composites.

Characteristics and measurement

The morphology of samples was observed by a Hitachi S-4800 field emission scanning electron (FE-SEM) microscope. Transmission electron microscope (TEM) and high resolution TEM (HR-TEM) images and the selected area electron diffraction (SAED) patterns of 3D-GN were investigated by JEOL 2100F. Raman spectra were collected on a thermal dispersive spectrometer using a laser with an excitation wavelength of 532 nm at a laser of power of 8 mW. The electrical transport properties were measured by Van der Pauw method with an Accent HL5500. The electric conductivities were measured for five times to obtain an average value. A thermal imager (SC305, Flir system USA) was used to record temperature distribution images of our samples. The DSC curve was measured by the Q100 Thermal Analyser. The thermal conductivity was measured using LFA 427 laser conductometer. The mechanism property was measured on Instron-5566-5500R with a compression speed of 2 mm/min.

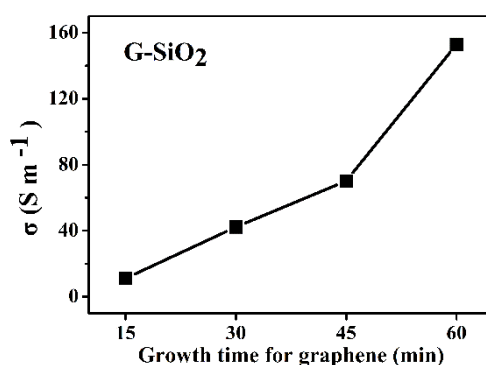


Fig. S1 Electric conductivity of G-SiO₂ for different growth time.

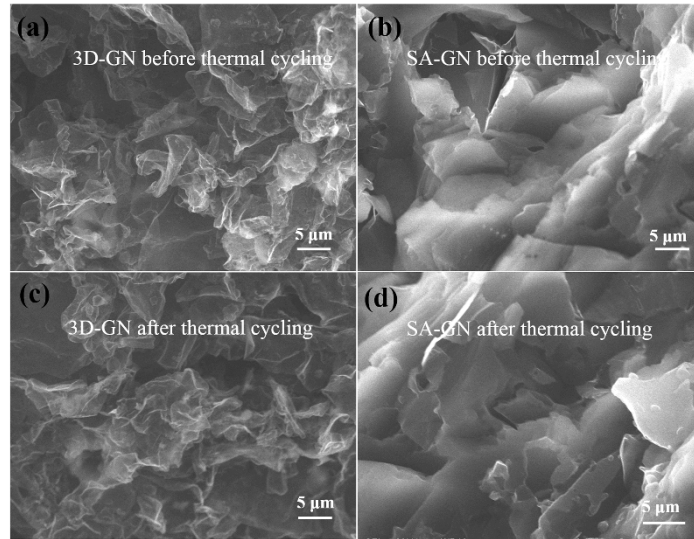


Fig. S2 (a, c) SEM image of unfilled 3D-GN before and after thermal cycling (SA was removed by hot ethanol), and (b, d) SEM images of the fracture surface of SA-GN composites before and after thermal cycling.

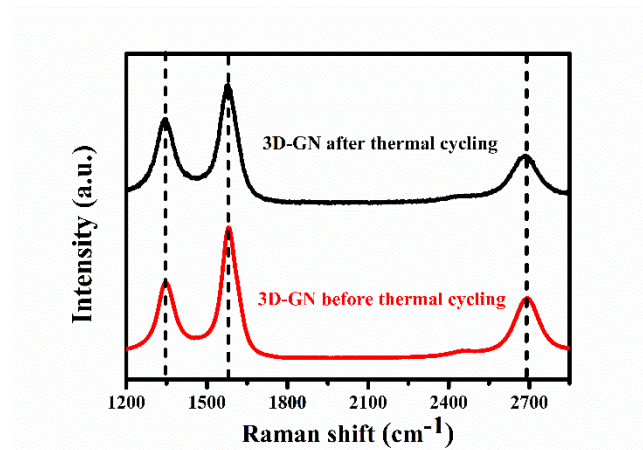


Fig. S3 Raman spectrum of 3D-GN before and after thermal cycling.

Tab. S4 Thermophysical properties (thermal diffusivity α ; thermal conductivity κ) of various composites (3D-GN prepared for different time).

Sample	Layer number	C_p (J g ⁻¹ K ⁻¹)	α (mm ² s ⁻¹)	κ (W m ⁻¹ K ⁻¹)
SA	-	2.30 ± 0.12	0.07	0.15
SA-GN (15min)	~ 3	1.74 ± 0.08	0.349	0.19 ± 0.01
SA-GN (30 min)	~ 9	1.70 ± 0.08	0.632	0.44 ± 0.02
SA-GN (45 min)	~ 15	1.68 ± 0.08	0.701	0.62 ± 0.03