Supporting Information

Graphene-Based Smart Thermal Conductive System Regulated by Reversible Pressure-Induced Mechanism

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EXPERIMENTAL SECTION

Synthesis of the GO Dispersion. 2 g expanded graphite and 92 g concentrated sulfuric acid were poured into a dry beaker (1000 mL), and the mixture was stirred for 24 h. Then 1 g $NaNO_3$ was poured into the beaker with 5 minutes stirring. After that, 6 g $KMnO_4$ was poured into the beaker very slowly with 5 minutes stirring under 0 °C and then 30 minutes stirring under 35 °C. Then 10 mL water was added to the beaker at twice with two-time 5 minutes stirring. After that, 40 mL water was added to the beaker with 30 minutes stirring, and then 400 mL water was added to the beaker with 5 minutes stirring. Afterwards, 20 mL H_2O_2 was poured into the beaker with 5 minutes stirring. The mixture was washed with 5wt % HCl solution at twice and then centrifuged to obtain the GO dispersion.

Preparation of the GO and rGO Foams. The GO aqueous dispersion was calibrated to 3 mg mL^{-1}, and 15 mL dispersion was disposed with ultrasound (SB-5200DTDN, 90 W, 40 KHz) under cool environment (below 10 °C) for 1 h. Then the dispersion was freezed at -80 °C for 2 h with a following freeze-dried process for 40 h. The dried GO foam was reduced at 800 °C for 4 h under Ar/H_2 atmosphere (H_2: 5 vol%) to obtain the rGO foam.

Material Characterizations. The Optical microscopy images of GO and rGO foam were collected by Nikon Eclipse Ts2. The morphological, elemental-mapping, and microstructural characterizations of GO and rGO foams were characterized by scanning electron microscopy (SEM, S4800, Hitachi). The surface chemical analyses of GO and rGO foams were implemented by X-ray photoelectron spectroscopy (XPS, Axis Ultra, Kratos Analytical Ltd.). The Fourier transform infrared (FTIR) spectra of GO and rGO foams were recorded by using a Nicolet 6700 FTIR system. The Raman spectra of GO and rGO foams were collected by using a Raman

spectrometer (ARAMIS, HORIBA Jobin Yvon, 532 nm excitation wavelength). The thermal diffusivity (D) was collected by laser flash method with a LFA-1000 apparatus (Laser Flash Analyses). The specific heat (C_p) was measured by differential scanning calorimetry (METTLER DSC1).

Thermal Measurement of the rGO Foam. The thermal diffusivity (D) of the fabricated rGO foam was detected under different pressures (from 0 to 1.3 KPa) by using a laser flash method with a LFA-1000 apparatus. Then, the specific heat (C_p) was measured by a differential scanning calorimetry with a heating rate of 10 K/min from 25 to 60 °C. Finally, thermal conductivity (k) of rGO foam under different pressures can be calculated by using the followed equation: $k = D\rho C_p$, in which ρ represent the density of the rGO foam.

Thermal Regulation of the rGO Foam. The rGO foam was placed on a hot stage with the temperatures of 50 and 100 °C. Thermal regulation of the rGO foam can be represented visually by an IR thermometer under different pressures (326, 402, and 495 Pa) using the hollow lead cylinders. Match-ignition experiments were conducted with an alcohol burner. The rGO foam was placed on the asbestos network with the alcohol lamp burning. Then, the match was put on the surface of rGO foam. Meanwhile, another match was directly put on the asbestos network with the alcohol lamp burning as comparison. The result can be obtained through comparing the recorded ignition time of these two tests.

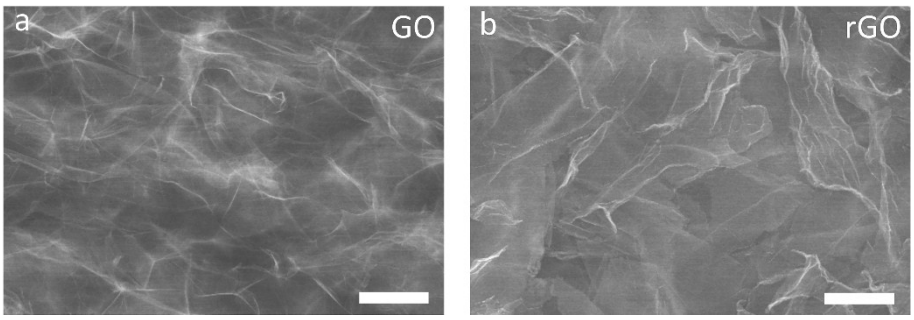

Fig. S1 Corresponding SEM images of the EDX elemental mapping images for the GO (a) and rGO (b) foams. The scale bars in panels (a,b) are both 20 μm.

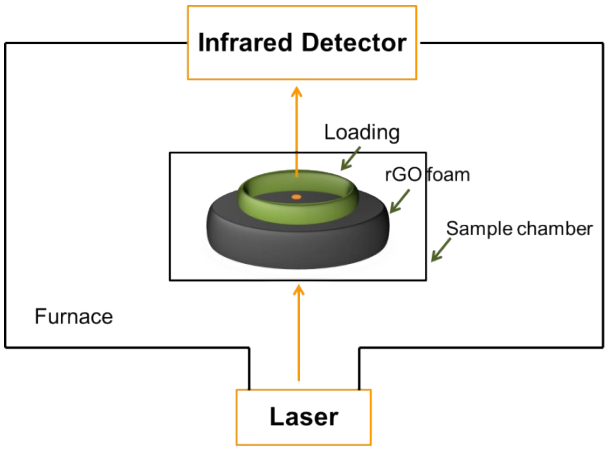

Fig. S2 Schematic of thermal conductivity measurement under compression.

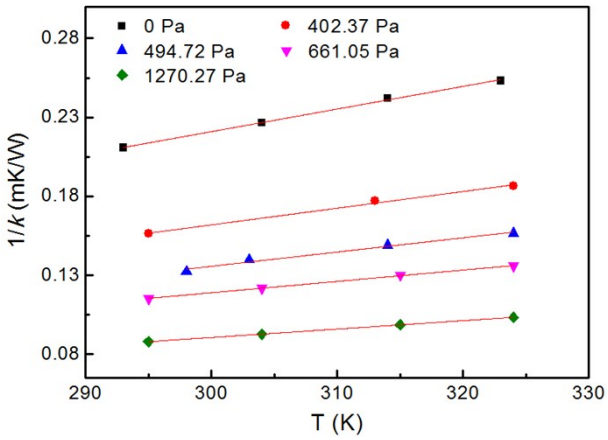

Fig. S3 Linear relationship between the inverse of the calculated thermal conductivity k^{-1} and the temperature under different pressures.

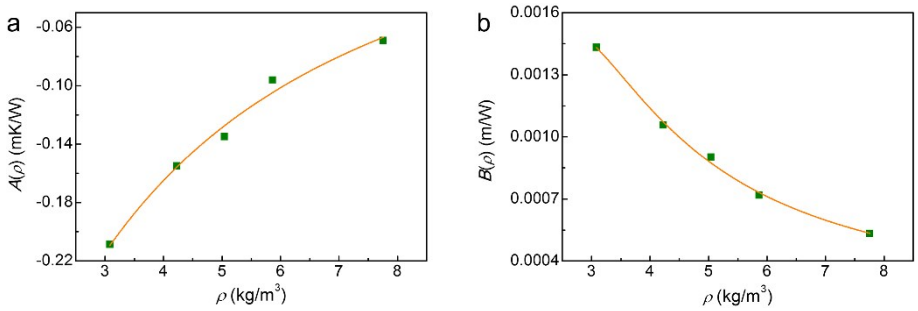

Fig. S4 Fitting curves of density (ρ)-based $A(\rho)$ (a) and $B(\rho)$ (b). $A(\rho)$ and $B(\rho)$ are positively and negatively associated with ρ, respectively.

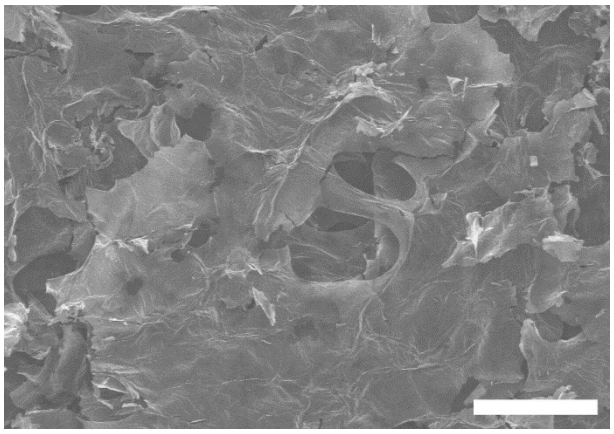

Fig. S5 Low-resolution SEM image of the rGO foam after hundreds of cycles of pressure. The scale bar is 200 μm.

Table S1 Fitting terms $A(\rho)$ and $B(\rho)$ in the equation of $k^{-1}(T,\rho) = A(\rho) + B(\rho)T$ at different pressures.

P (Pa)	**ρ (kg/m^3)**	***A* (mK/W)**	***B* (m/W)**
0	3.085	-0.20879	0.00143
402.37	4.228	-0.15498	0.00106
494.72	5.042	-0.13491	9.02116E-4
661.05	5.864	-0.09612	7.17051E-4
1270.27	7.760	-0.06903	5.32291E-4

Table S2 Fitting terms a_0-a_2 and b_0-b_2 in the equations of $A(\rho) = a_0 + exp(a_1 + a_2/\rho)$ and $B(\rho) = exp(b_0 + b_0/\rho + b_0/\rho^2)$.

a_0	a_1	a_2	b_0	b_1	b_2
-0.3804	-0.7596	-3.1003	-8.8647	12.4070	-16.2406