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Electronic Supplementary Information

Fabrication and Molecular Dynamics Analyses of Highly Thermal Conductive Reduced Graphene Oxide Films at Ultra-high Temperature

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1. Preparation of graphene oxide (GO)

The as-obtained GO sheets were further investigated by Atomic Force Microscope (AFM, Shimadzu SPM-960). As shown in **Figure S1**, the GO sheet in view is more than 10μm×10μm in planar size and the average thickness is 0.80 nm. The AFM observations prove that high quality GO sheets have been acquired by modified Hummer's method and provide a good foundation for subsequent fabrication of rGO films with high thermal conductivity.

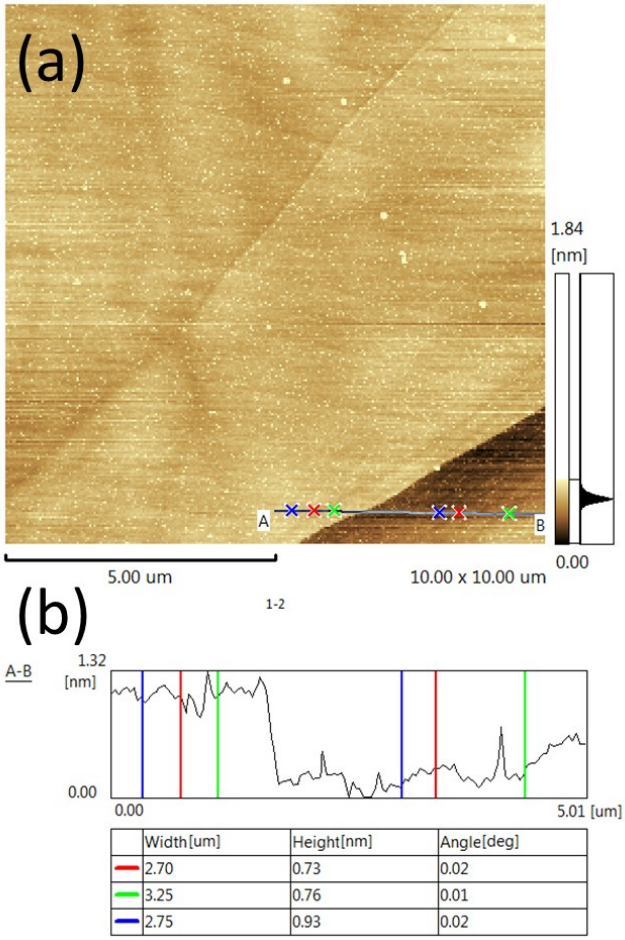


Figure S1. AFM image of GO sheets (a) and its thickness data (b).

2. Modified roller coating method

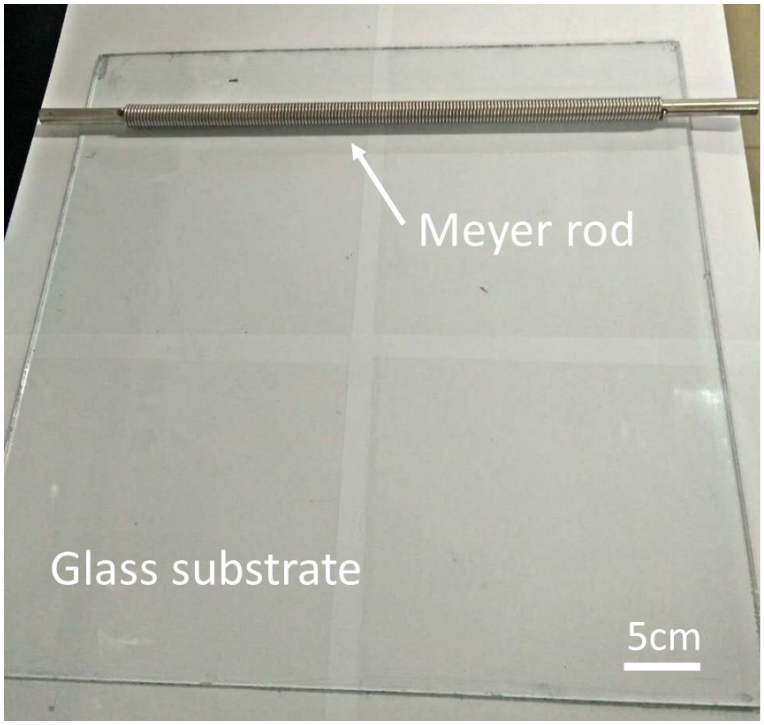


Figure S2. Image of the Meyer rod we used in the modified roller coating method.

3. Characterization of as-obtained GO and rGO films

Scanning electron microscope (SEM) was used to probe the microstructure of samples by Hitachi Limited SU-8010 SEM. Energy Dispersive Spectroscopy (EDS) were also adopted to determine the elemental compositions of samples **(Table S1)**. It could be confirmed that 1200 ^{o}C is sufficient for thermal reduction of GO.

X-ray diffraction (XRD) patterns were recorded by an X-ray diffractometer (Rigaku Ultima IV) at 40kV and 40mA with Cu Kα radiation. The interlayer distance **(Table S1)** was determined through theoretical calculation by the Scherer equation (2):

$$d = \frac{\lambda}{2\sin\theta} \quad (2)$$

where λ was wavelength of Cu Kα radiation (~1.5406 Å), θ was Bragg diffraction angle (°). The decreasing of interlayer spacing of the samples was proportional to the increasing of annealing temperature, which was due to the higher graphite degree, higher reduction level, the decomposition of oxygen-containing functional groups and a better ordering of the two-dimensional sheets.

Raman spectroscopic measurements were performed with a Jobin Yvon HORIBA Raman spectrometer, using a He-Ne laser operating at 632.8nm as the excitation source. The detailed values of I_D/I_G were shown in **Table S1** and the results suggested that with the annealing temperature rising, the in-plane defects in the GO or rGO sheets were decreased.

Table S1. Summary of the elemental compositions of samples

Sample	C[a] (wt%)	O[a] (wt%)	d-spacing[b] (Å)	I_D/I_G[c]
GO	72	28	8.06	1.01

rGO-1200	99.9	0.1	4.12	1.37
rGO-2000	100	0	3.37	0.12
rGO-2400	100	0	3.36	0.10
rGO-2800	100	0	3.35	0.05

[a] Determined by EDS. [b] Determined by XRD. [c] Determined by Raman.

4. Thermal measurement details

To investigate the thermal conductivity (κ) of the as-obtained samples, we adopted the laser flash method (Netzsch, LFA-467) with specially designed accessories (**Figure S3**), which enabled the measurement of in-plane thermal diffusivity (α) of the sample. The testing process was in accordance with ASTM E1461-92, i.e., ‘‘Standard Test Method for Thermal Diffusivity of Solids by the Flash Method’’, which was for the measurement of thermal diffusivity. **Figure S3a** shows the schematic diagram of this modified LFA method and in order to measure the in-plane thermal conductivity of thin films, we applied a specially designed sample holder as shown in **Figure S3b**.

The IR detector, which was in a fixed distance from the hot spot, could obtain the continuous temperature variation signals, and through the time-temperature curves (**Figure S3**c) the thermal thermal diffusion coefficient (α) could be calculated by equation: $\alpha = 0.1388 \times \frac{h^2}{t_{50}}$, in which h was the thickness of sample and t_{50} was the time needed for the temperature rising to half of the maximum temperature rise. After that, the thermal conductivity (κ) (W m^{-1} K^{-1}) was determined by calculating with the equation: $\kappa = \alpha \times C_p \times \rho$.

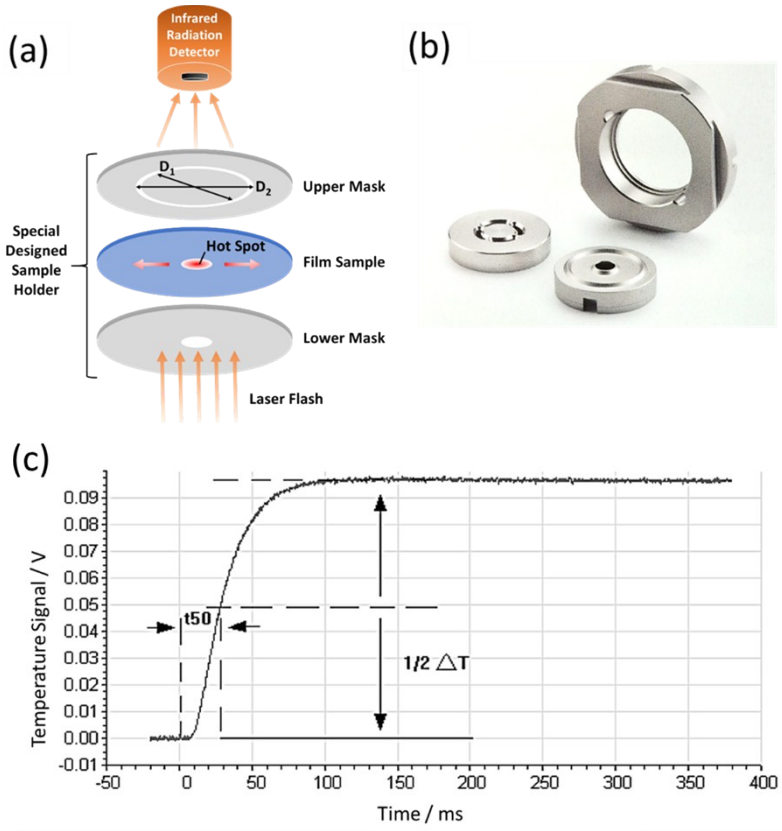


Figure S3. Schematic diagram of modified LFA method to measure in-plane thermal conductivity of films (a) and the real special designed sample holder for films (b). (c) An example of time-temperature rise curves with LFA method.

Table S2. (a) Summary of average quantitative values of the samples

Sample	α (mm^2 s^{-1})	ρ (g cm^{-3})	C_p (J g^{-1} K^{-1})	κ (W m^{-1} K^{-1})
GO	20.48	1.33	1.541	41.64
rGO-1200	103.06	1.23	0.801	101.54
rGO-2000	422.17	1.20	0.751	380.46
rGO-2400	561.59	1.18	0.751	457.79
rGO-2800	678.96	1.62	0.751	826.03

Table S2. (b) Summary of average quantitative values of the density of samples

Sample	m (mg)	h (μm)	a (mm)	b (mm)	ρ (g cm^{-3})
GO	23.30	13	41.72	32.34	1.328
rGO-1200	39.80	16	63.20	32.00	1.230
rGO-2000	67.60	20	64.40	43.88	1.196
rGO-2400	62.10	18	65.00	45.04	1.178
rGO-2800 (with mechanical compaction)	28.40	9	54.84	35.52	1.620

5. Molecular dynamics simulations

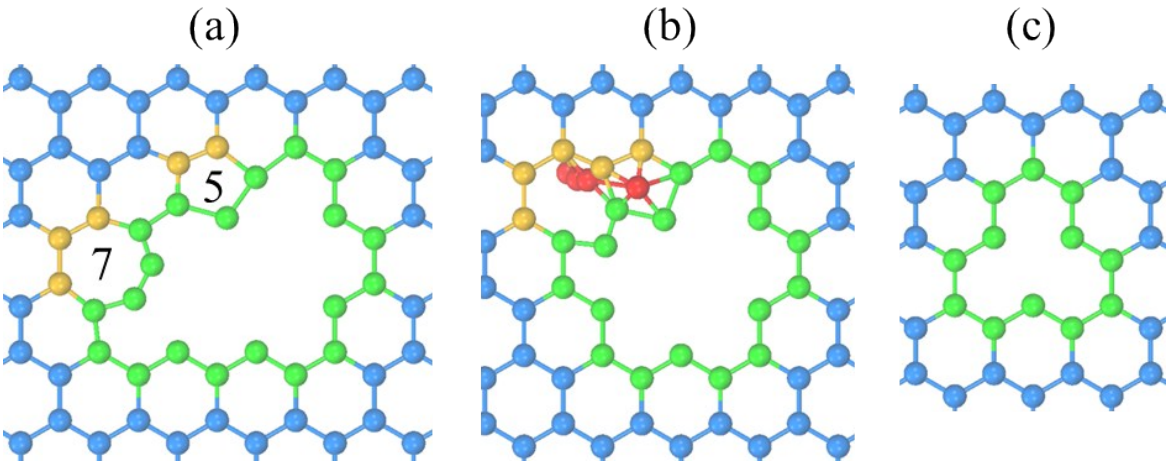
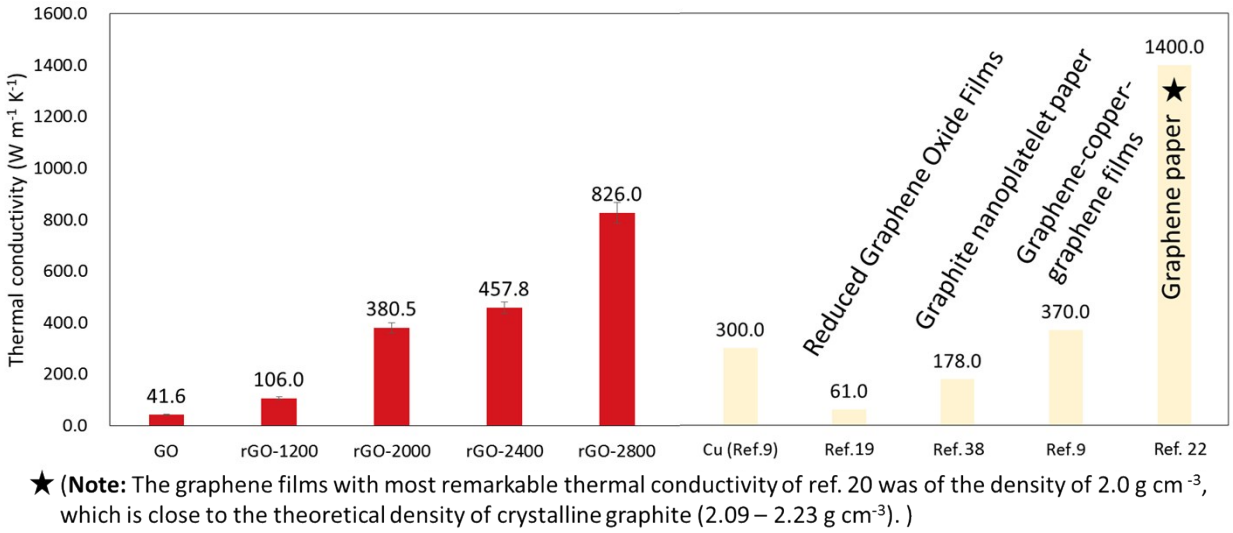


Figure S4. Snapshots of defects introduced into the initial structure. (a) no-hexagonal rings around a hole; (b) dangling bonds around a hole (the red atoms are hanging); (c) mono-vacancy.

6. Comparison of thermal conductivities



★ (**Note:** The graphene films with most remarkable thermal conductivity of ref. 20 was of the density of 2.0 g cm $^{-3}$, which is close to the theoretical density of crystalline graphite (2.09 – 2.23 g cm^{-3}).)

Figure S5. Comparison of thermal conductivities for as-prepared samples and those cited from the references.