

Supplementary information for:

Solution-processable reduced graphene oxide films as broadband terahertz wave impedance matching layers

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1. THz time-domain system :

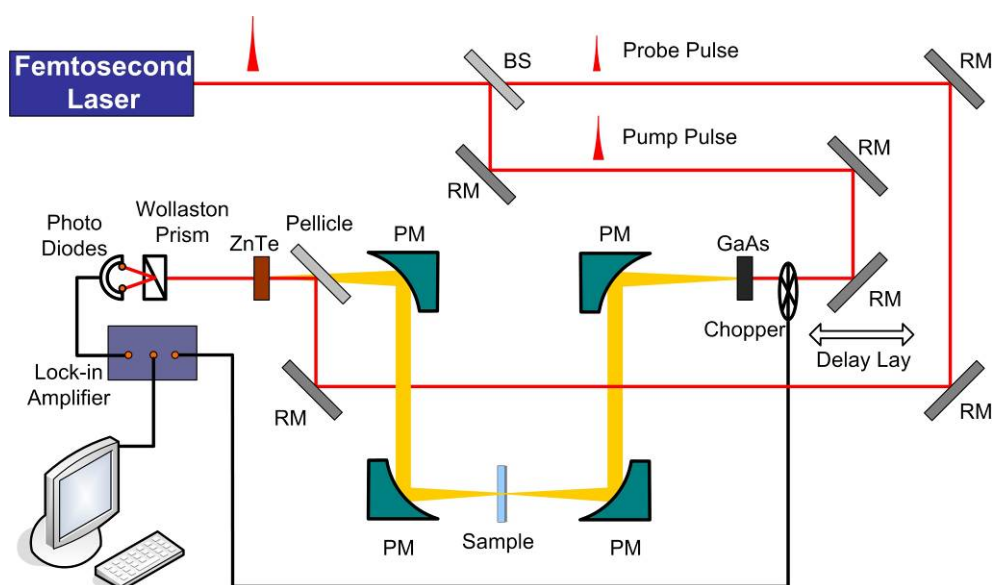


Fig. S1 Schematic illustration of our custom-designed THz TDS system (BS: beam splitter, RM: reflective mirror, PM: parabolic mirror)

THz characterization of all our samples are made by a custom-designed THz time domain spectroscopy (THz-TDS), and the setup is shown in Fig. S1. In general, THz pulses are generated by a photoconductive antenna under the excitation of a 70-fs laser (Ti:Sapphire femtosecond laser: Maitai Spectra-Physics) with a center wavelength of 800 nm and a repetition rate of 80 MHz. THz pulses are detected by electro-optic sampling with a ZnTe(110) crystal. The optical path from THz generation to THz detection is purged with N₂ to avoid the effect of the humidity.

2. X-ray photoelectron spectroscopy (XPS)

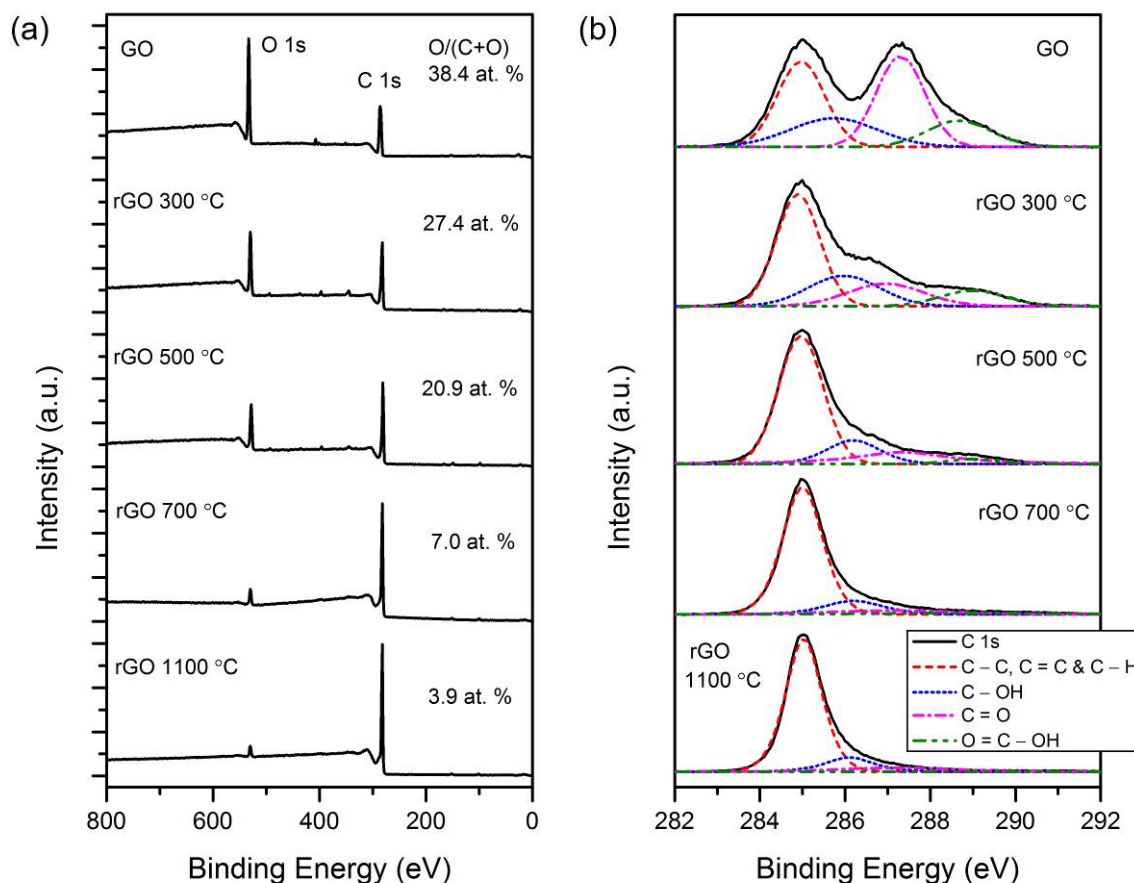


Fig. S2 (a) XPS spectra of GO and rGO (300, 500, 700 and 1100 °C) films. (b) C 1s peaks of GO and rGO (300, 500, 700 and 1100 °C) films.

Figure S2 shows typical XPS spectra of GO and rGO samples with thermal treatment at 300, 500, 700 and 1100 °C. From Figure S2a, we can see that the atomic concentration of oxygen for GO is approximately 38.4 at. %. After thermal reduction at 300, 500, 700 and

1100 °C, it decreases to 27.4 at. %, 20.9 at. %, 7.0 at. % and 3.9 at. %, correspondingly. From Figure S2b, the calculated atomic concentration of sp² carbon has an increased value with the treatment temperature as: 32.1 at. % (as-deposited), 49.8 at. % (300 °C), 66.0 at. % (500 °C), 77.2 at. % (700 °C), and 81.3 at. % (1100 °C). These results suggest that the sp³ graphitic domain decreases from 67.9 at. % to 18.7 at. % and the concentration of oxygen drops dramatically from 38.4 at. % to 3.9 at. % at 1100 °C, which clearly reveals the reduction effects induced by thermal treatments.

3. Raman spectra

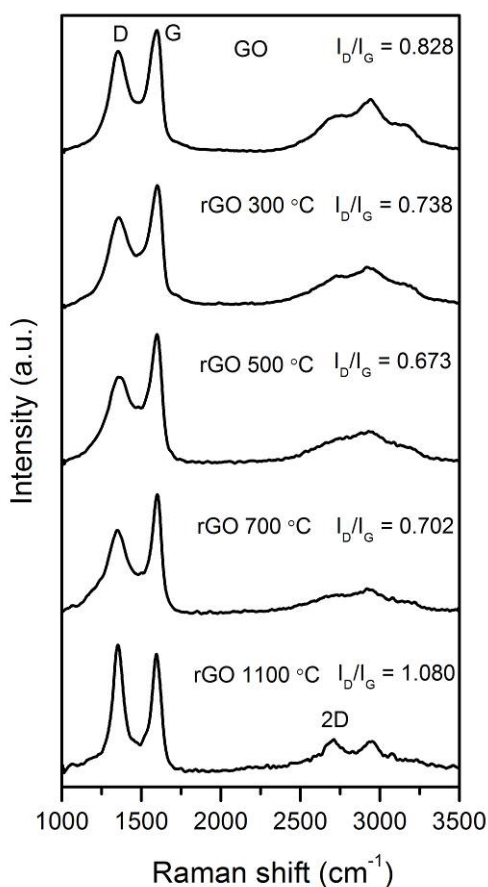


Fig. S3 Raman spectra of GO and rGO (300, 500, 700 and 1100 °C) films with 514 nm laser excitation.

Figure S3 presents the Raman spectra of the pristine and annealed GO films. In the partially reduced samples below 700 °C, the 2D band at 2694~2716 cm⁻¹ is merged with other peaks and is not obvious with respect to the D and G peaks. With Tuinstra–Koenig (TK)

relation: $L_a(nm) = (2.4 \times 10^{-10}) \lambda^4 (I_D / I_G)^{-1}$. The I_D/I_G intensity ratio of rGO slightly decreases from 0.83 ($L_a = 20.2$ nm) to 0.67 ($L_a = 25.0$ nm) along with the reduction temperature elevated to 500 °C. It then increases to 1.08 ($L_a = 15.5$ nm) for rGO at 1100 °C. The decrease of sp^2 graphitic domain in rGO is also observed in some previous results with different reduction methods.^{1,2} From the XPS results we can confirm the expansion of the total sp^2 graphitic domain along with the rising temperature. Thus the decrease of L_a might be attributed to the different structural changes in rGO with temperature-dependence. For example, with 1100 °C reduction, the sp^2 domains in rGO are smaller in size but more numerous in number. However, the shape changes in the Raman results of rGO below 700 °C are not very obvious. This also implies that to some extent Raman spectroscopy is not sufficient to distinguish the reduction degrees of rGO films.

4. Atomic force microscopy (AFM)

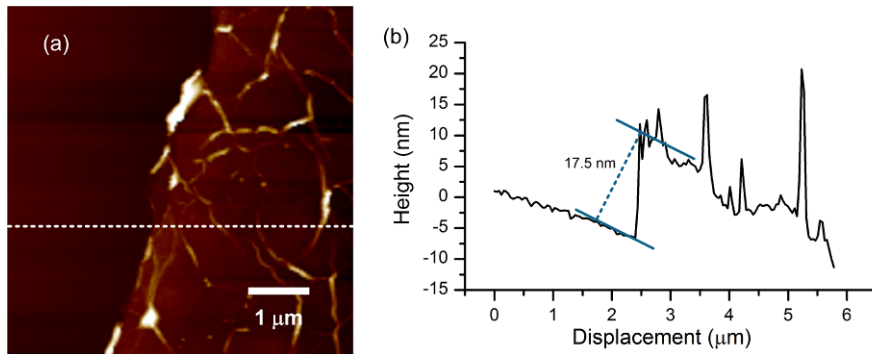


Fig. S4 (a) AFM image of 40 mL 1100 °C rGO film. (b) Line scans for 40 mL films along the dashed lines on the images in (a).

Surface morphology of the thin films is studied by AFM images obtained by using a Dimension Icon (Bruker AXS Inc.). Height profiles are obtained in a tapping mode. As shown in Fig. S4a, 40 mL 1100 °C rGO film has a surface structure of multilayer stack. And step height of most flakes for this film was found to be approximate 17.5 nm (Figure S4b).

5. THz refractive index of the fused quartz substrate

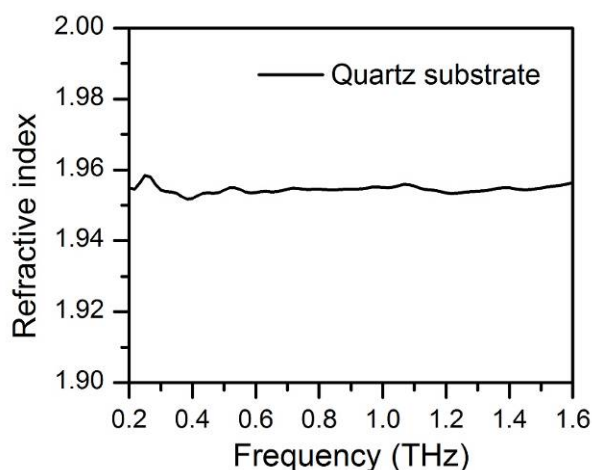


Fig. S5 Refractive index of the fused quartz substrate.

The refractive index of the SiO_2 substrate is measured by the THz–TDS. Fig. S5 shows a frequency-independent value of about 1.955, which coincides with the previous measurement for fused silica.³ This value is used in the calculation in this work.

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