Supporting Information for:

Ultrafast, Dry Microwave Synthesis of Graphene Sheets

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GO synthesis

Graphite flakes 230U were obtained from Asbury. All the chemicals were of analytical reagent grade, and used without further purification. GO was synthesized by Hummer's method.[1] Graphite (0.5 g) was put into a $NaNO_3$ (0.5 g)/concentrated H_2SO_4 (50 mL) solution in a 1 L flask seated in an ice bath. 3 g of $KMnO_4$ was slowly added to the flask and the temperature was kept below 20 ^oC. The mixture was stirred in the ice bath for 2 h and then in water bath at 35 ^oC for 0.5 h. 46 mL of 70 ^oC DI water was then added into the flask drop by drop and the generated heat kept the solution temperature around 98 ^oC. The mixture was diluted by 140 mL 70 ^oC DI water, into which 10 mL (30 wt.%) of aqueous solution of hydrogen peroxide was added to terminate the reaction. The mixture was filtrated and washed several times with hot water. The resulting GO solid was dried overnight at 55 ^oC in a vacuum (1 inch Hg) and peeled off the filtration membrane to form a GO film.

Rate of temperature rise

2mL 1mg/mL GO solution was added in a beaker and dried at 55 ^oC. Then the GO sample was placed in the microwave field of 500 W, 6.425 GHz$\pm$1.150GHz with temperature of the GO surface recorded by an infrared thermometer. For each cycle, the sample was heated to 200 ^oC and then the MW was stopped to let the sample cool down to room temperature. The heating and cooling cycle was repeated until the rate of temperature rise fell back to a similar value of the starting GO (Figure S5). For each cycle, a linear fit was performed on the temperature-time curve and the slope of the fitted line was determined as the rate of temperature ($\frac{\Delta Temp}{\Delta t}$). The dielectric properties may subject to change at elevated temperatures.[2] Therefore, the cycling process characterized the rate of temperature change with little effect on dielectric properties.

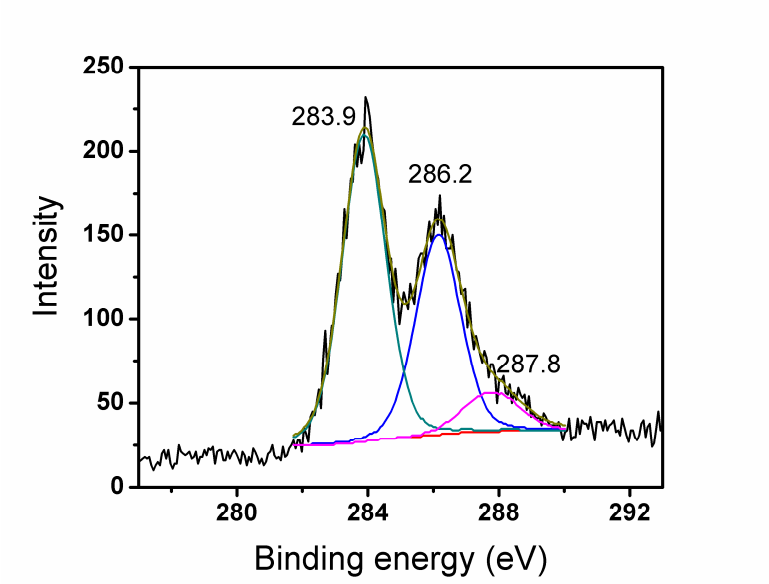

Fig. S1 XPS survey result of GO. Curve fitting of the C1s spectra was performed using a Gaussian-Lorentzian peak shape after performing a Shirley background correction. The binding energy of the C–C and C–H bonding are assigned at 284.5–285 eV and chemical shifts of +1.5, +2.5 and +4.0 eV are typically assigned for the C–OH, C=O, and O=C–OH functional groups, respectively.[3] The epoxide group (C-O-C) should have similar binding energy with C-OH, but it is also possible that the C-O-C emission shifted to the range of C=O. The C1s spectra have a main peak at 283.9 and 285 for GO and rG respectively, assigned to C-C and C-H bonding; there is another peak at higher binding energy, that can be fitted into 286.2, 287.8, 289 eV, representing the functional groups C-OH, C=O, and O=C–OH respectively. The assignment of a small fitted peak at 287 in rG cannot be determined, but the mean square error for a fit without this peak was much larger.

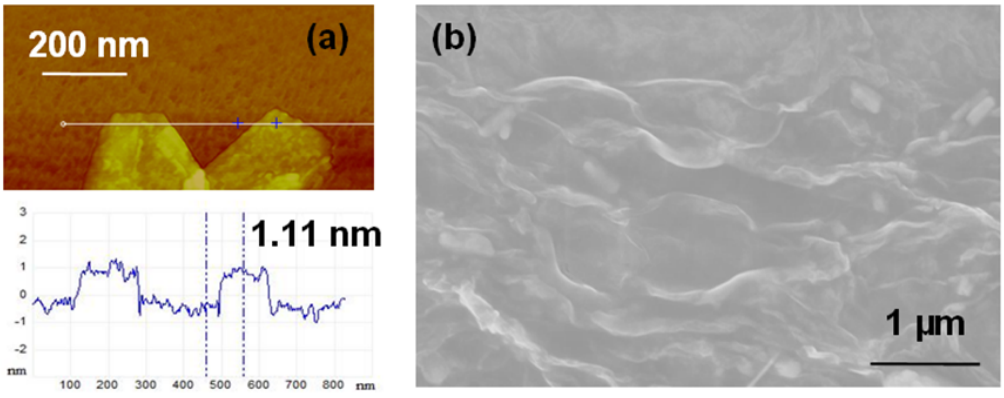

Fig. S2 (a) Typical AFM image of rG and its height profile in the bottom. (b) SEM image of rG. During the MW, the decomposition rate of oxygen functionalized groups exceeded the diffusion rate of the evolved gases, generating huge gas pressure to exfoliate rG.

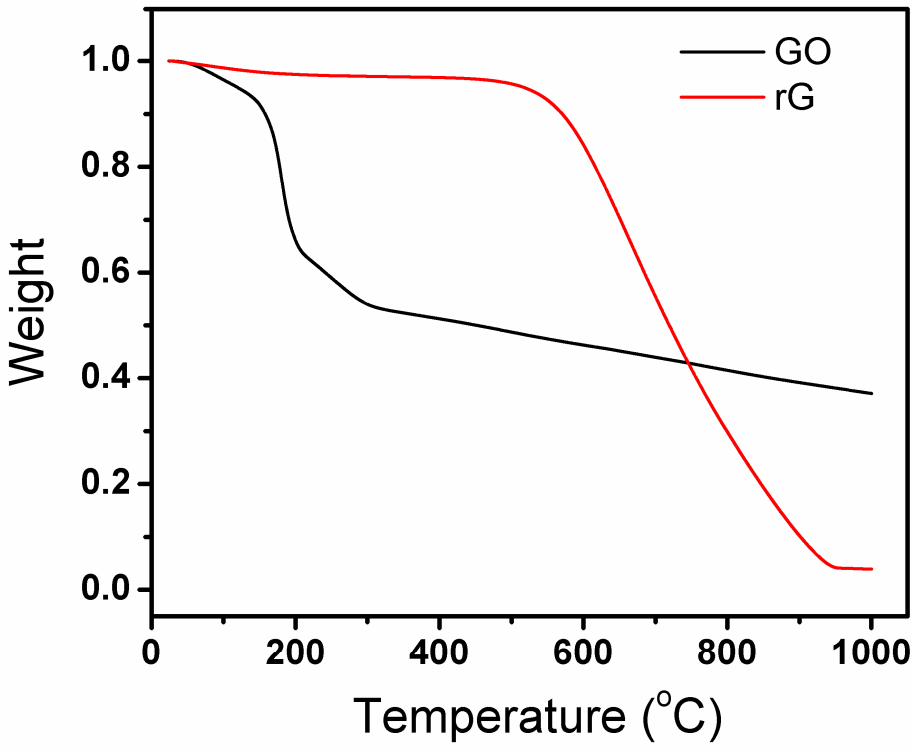

Fig. S3 TGA data of GO (black curve) and rG by MW (red curve).

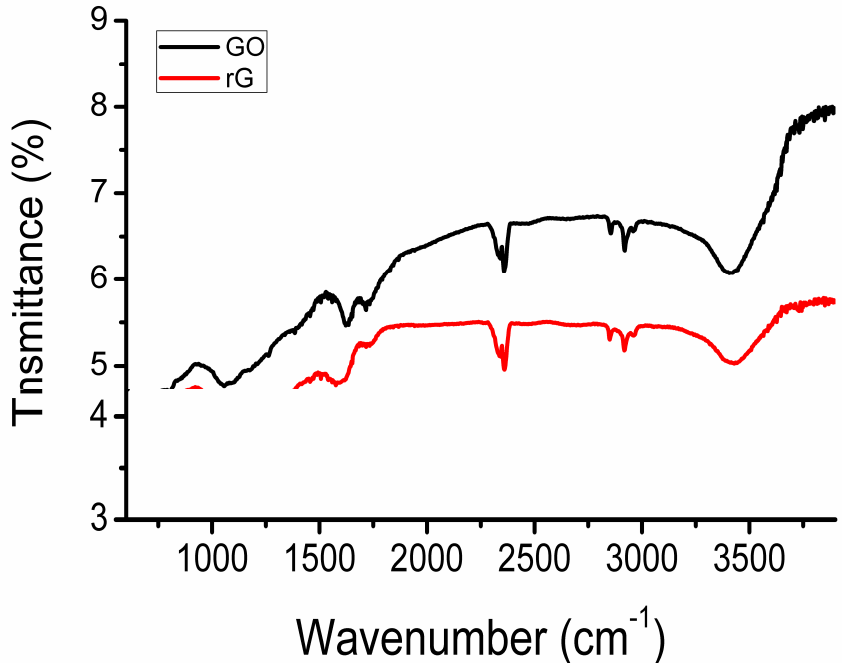

Fig S4. IR data of rG (red curve) and GO (black curve).

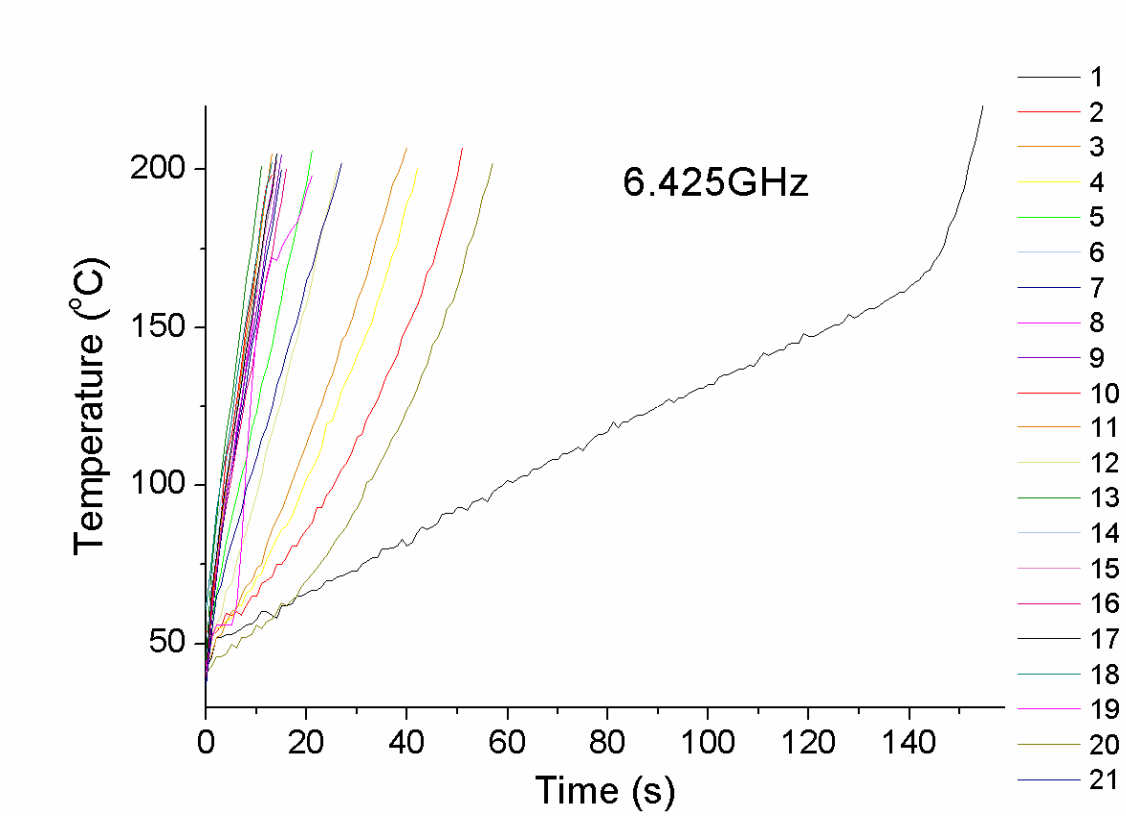

Fig. S5 Heating cycles at 500 W, 6.425 GHz±1.150 GHz microwave irradiation.

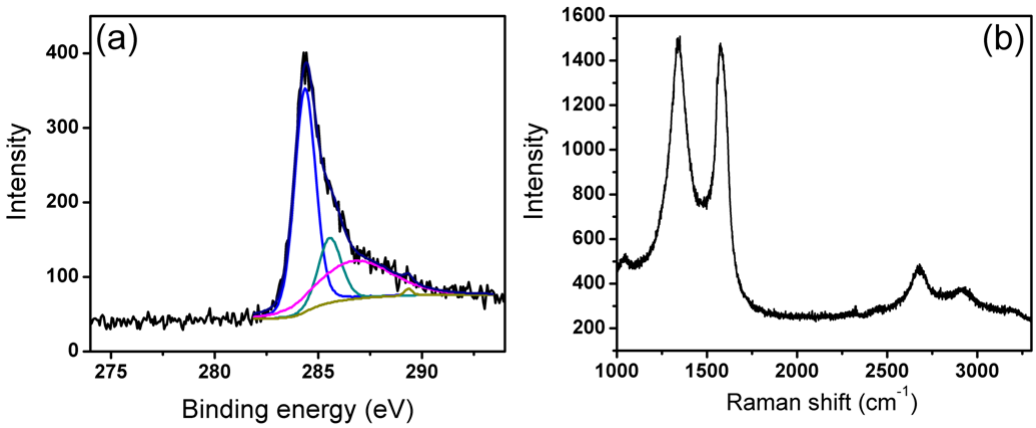

Fig. S6 XPS survey result (a) and Raman spectrum (b) of re-oxidized rG. In XPS, a shoulder became pronounced around 287 eV, probably due to the formation of carbonyl or peroxide groups in the re-oxidation process. At the same time, the D/G ratio in Raman spectra increased back to around 1.

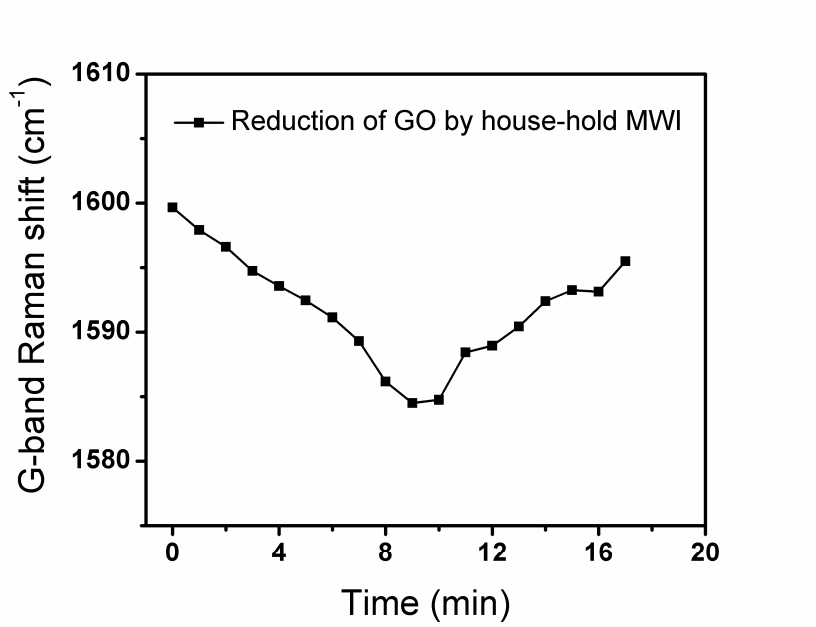

Fig. S7 G-peak position in Raman spectra of rG samples by house-hold MW as a function of MW irradiation time. From the Raman shift of G-band, it is obvious that there is an optimal irradiation time for the synthesis of rG and too long time MW in air would re-oxidize the graphene structure.

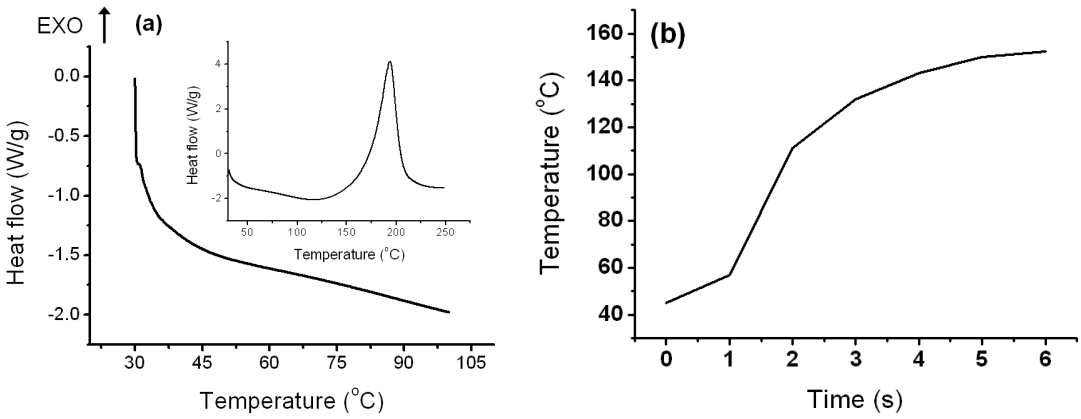

Fig. S8 Energy analysis on MW reduction of GO. (a) The close-up of differential scanning calorimetry (DSC) heating up from 30 °C to 100 °C. The DSC was performed under N_2 at a ramping rate of 5 °C/min from 30 °C to 250 °C (inset figure). A peak at 193 °C is the exothermal peak of GO degradation. The input energy for heating GO up to 100 °C can be calculated by integration of the area under the heating curve and gives an energy of 1354 J/g. (b) A piece of GO film (16.3 mg) was heated up by MW from 45 °C to 110 °C within 2 seconds. The forwarded power is 500 W and reflected power is fluctuating from 40 W to 100 W. The average adsorbed power is 430 W. Therefore, the microwave energy absorbed by GO to heating up to 110 °C is estimated by (430W*2 s)/0.0163g= 52,760 J/g, which is almost 50 times higher than the thermal energy needed to heat GO up to 100 °C and approximately 10 times higher than the photo energy provided by Xenon flash.[4, 5]

References:

1 W.S. Hummers and R.E. Offeman, *Journal of the American Chemical Society*, 1958, **80,** 1339.

2 A.C. Metaxas and R.J. Meredith, *Industrial microwave heating*. IEE power engineering series 4. 1983: Peter Peregrinus Ltd, London, The United Kingdom.

3 D. Yang, A. Velamakanni, G. Bozoklu, S. Park, M. Stoller, R.D. Piner, S. Stankovich, I. Jung, D.A. Field, C.A. Ventrice Jr, and R.S. Ruoff, *Carbon*, 2009, **47,** 145.

4 G. Scott, D. Sergey, B. Alireza, F. Jabari, A.D. Stephen, and B.K. Richard, *Adv. Mater.*, 2010, **22,** 419.

5 L.J. Cote, R. Cruz-Silva, and J.X. Huang, *J. Am. Chem. Soc.*, 2009, **131,** 11027.