

Supporting Information

Laser-Induced Reduction of Graphene Oxide by Intensity-Modulated Line Beam for Supercapacitor Applications

Tung Xuan Tran^a, Hayelin Choi^a, Cuong Huu Che^a, Ji Hwan Sul^b, In Gyoo Kim^b, Seung-Mo Lee^c, Jae-Hyun Kim^c, Jung Bin In^{a,*}

^a Laser Thermal Nanoengineering Laboratory, School of Mechanical Engineering, Chung-Ang University, Seoul, 06974, Republic of Korea

^b ICT Materials & Components Research Laboratory, Electronics and Telecommunications Research Institute (ETRI), Daejeon 34129, Republic of Korea

^c Department of Nanomechanics, Korea Institute of Machinery and Materials, Daejeon 34113, Republic of Korea

* Corresponding author. E-mail: jbin@cau.ac.kr (JB In)

Calculation of specific capacitance

To calculate a mass-specific (gravimetric) capacitance C_g (F/g) from the galvanostatic charge-discharge (CC) curve, we used the following equation:

$$C_g = \frac{2I}{m} \cdot \frac{\Delta t}{\Delta V}$$

where I (A) is the discharge current; Δt (s) is the discharge time; ΔV (V) is the potential window; and m (g) is the mass of the single electrode. Similarly, an areal capacitance was calculated by the following equation:

$$C_A = \frac{2I}{A} \cdot \frac{\Delta t}{\Delta V}$$

where A is the area of the single electrode. The gravimetric capacitance was also obtained from CV curves, and the equation follows:

$$C_g = \frac{2}{mv(V_b - V_a)} \int_{V_a}^{V_b} IdV$$

where v (V/s) is the scan rate; I (A) is the discharge current; V_b and V_a (V) are the upper and the lower potential limits of the CV test, respectively.



Figure S1. Structure of an rGO supercapacitor.

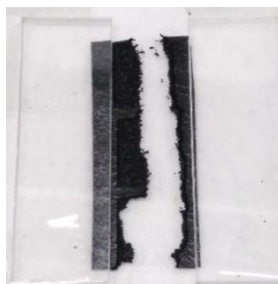


Figure S2. Severely burned rGO film by laser-induced combustion under an air flow.

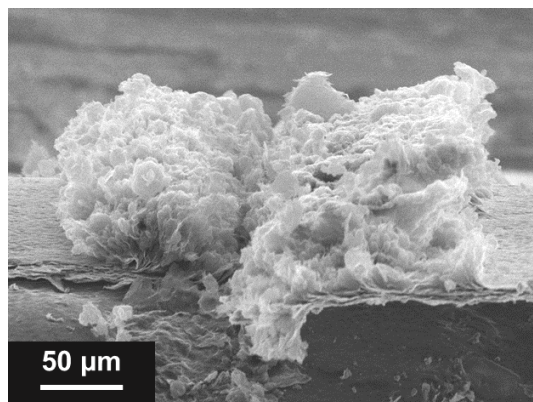


Figure S3. Cross-sectional SEM image of a single reduction spot.

Characteristics of rGO strips

A series of rGO strips were prepared by varying the laser power (10, 12, 15, 20, 25 W) at a scanning speed of 200 mm/s and a chopper frequency of 1 kHz. As the rGO surface is highly rough due to the periodic laser pulses, a Raman spectroscopy instrument (QE Pro-Raman; Ocean Optics, Inc.) equipped with a large probing laser beam (diameter: ~ 0.48 mm; wavelength: 785 nm) was used to capture the averaged characteristics. Fig. S4 shows the Raman spectra obtained from the strip centers and their I_D to I_G ratios.

The dimensions (width and thickness) of the rGO strips are provided in Fig. S5(a) and (b). The size of the rGO strip was found to increase with laser power. The thickness or the height of the rGO structure was determined from its cross-sectional SEM image. However, it should be noted that the surface of the rGO is not flat but highly corrugated because of periodic laser pulses. Therefore, the thickness estimated from the cross-sectional SEM image corresponds to the maximum height value. Fig. S5(c) shows the electrical resistance per unit length of the rGO strip, which was measured by using the 2-point probe method. Fig. S5(d) shows the resistivity calculated from the measured resistance and the thickness information. The rGO strips are highly porous, and thus the calculated resistivity is an effective value that does not directly indicate the intrinsic resistivity of the individual rGO layers.

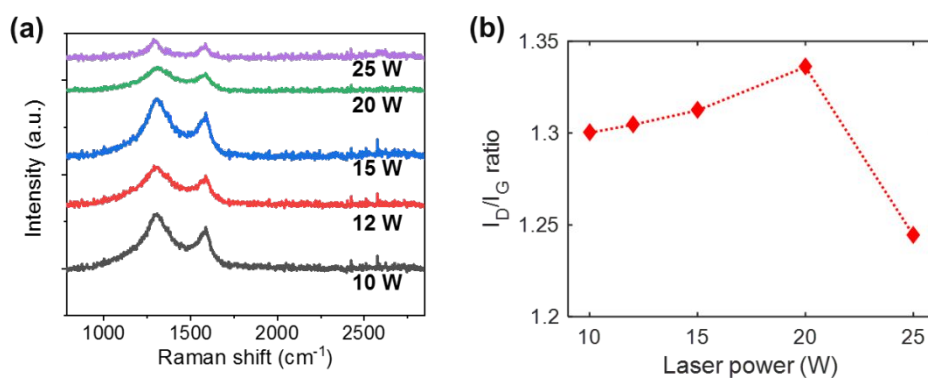


Figure S4. (a) Raman spectra of rGO produced at different powers. (b) I_D/I_G ratio calculated from the spectra data.

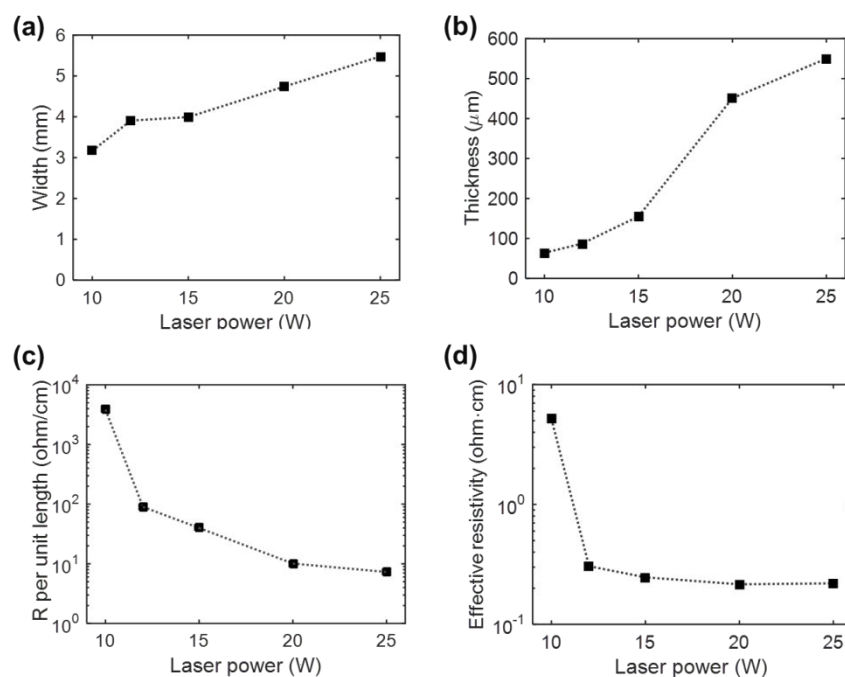


Figure S5. (a) Width, (b) thickness, (c) electrical resistance per unit length, and (d) calculated effective resistivity of the rGO strips produced at different powers.

Electrochemical Impedance Spectroscopy (EIS) of rGO supercapacitors

Fig. S6(a) and S6(b) show Nyquist plots and Bode plots of phase angle versus frequency for sample #8 and #9, respectively. The characteristic frequencies (f_0) for a phase angle -45 deg. are 10 Hz for sample #8 and 18 Hz for sample #9. The obtained frequencies are comparable to f_0 (30 Hz) of the DVD laser-scribed rGO reported by El-Kady et al.¹ However, the series resistance of our rGO, which can be estimated from the Nyquist plot, is significantly lower than that of their rGO. As mentioned in their report, this is possibly because of the use of our gold current collector.

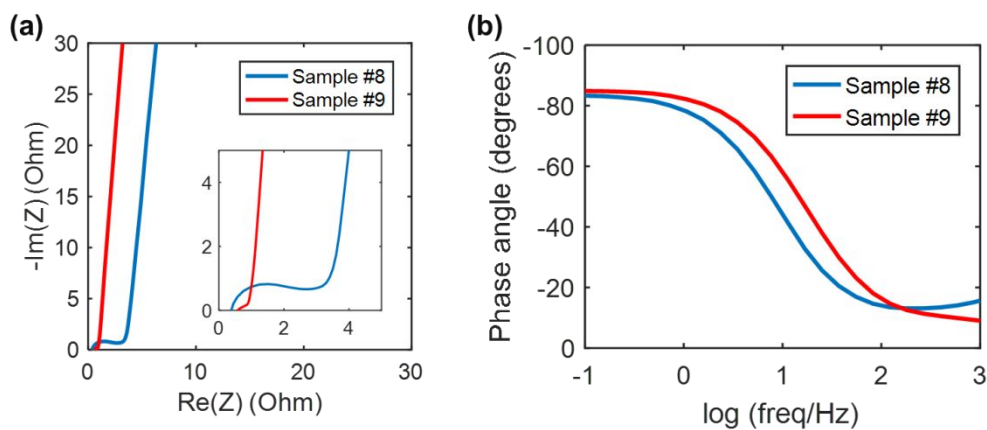


Figure S6. EIS measurement results. (a) Nyquist plots and (b) Bode plots of phase angle versus frequency for sample #8 (18.6 W, 200 mm/s) and sample #9 (26.5 W, 100 mm/s).

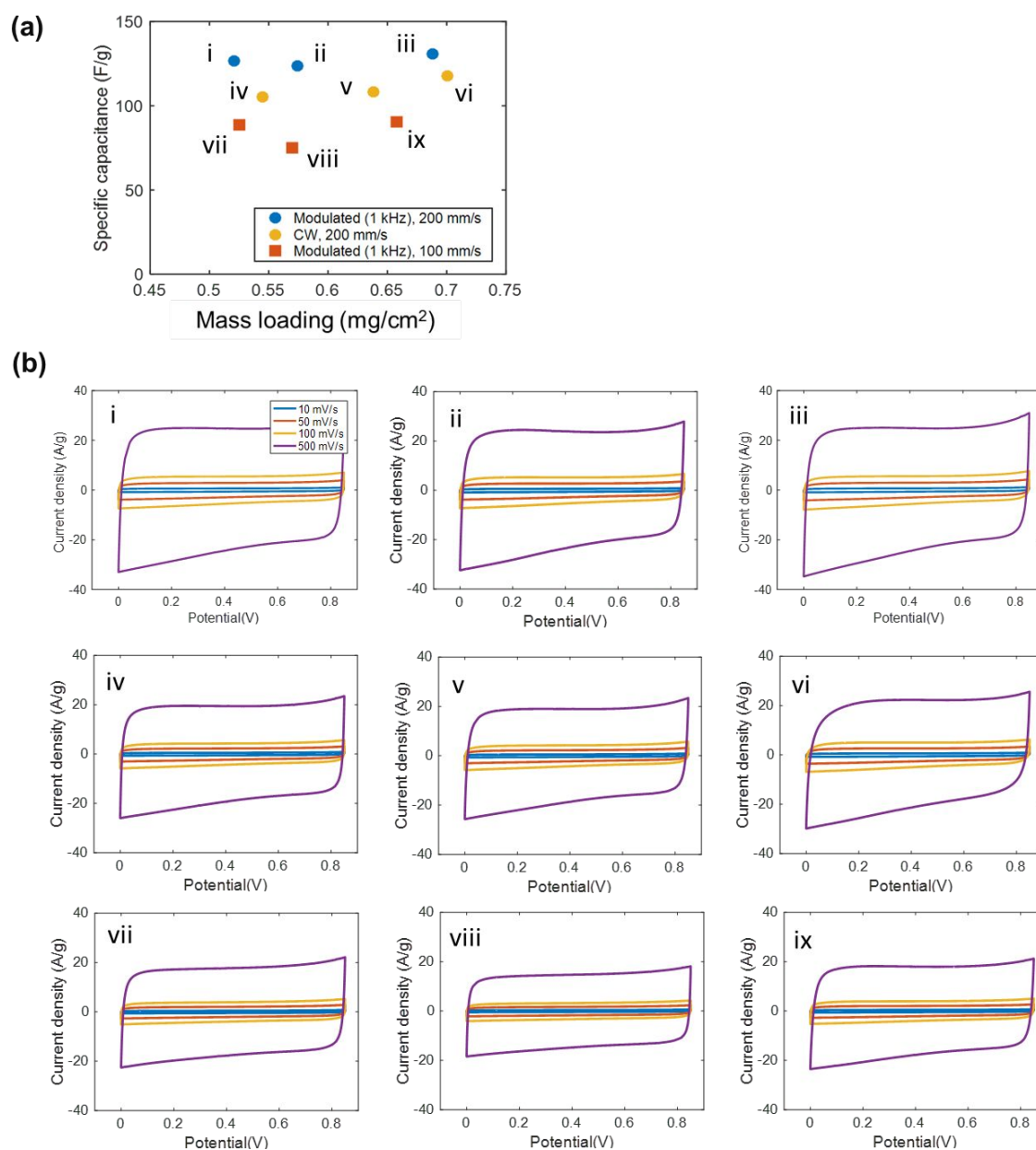


Figure S7. (a) Specific capacitance versus mass loading plot (figure 7) of the rGO samples marked with indices from i to ix. (b) Corresponding CV curves (scan rate: 10, 50, 100, 500 mV/s) of sample i–ix.

References

- (1) El-Kady, M. F.; Strong, V.; Dubin, S.; Kaner, R. B. Laser Scribing of High-Performance and Flexible Graphene-Based Electrochemical Capacitors. *Science* **2012**, *335*, 1326-1330.