

Supplementary Information

**Reduction intermediates of graphene oxide for low  
temperature curable electrode material**

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## Materials and methods

**Preparation of e-GO.** A sample of e-GO was prepared from synthetic graphite (Aldrich, < 20  $\mu\text{m}$ ) by modification of the Hummers method. First, 4 g of graphite was dispersed into a mixture of 150 ml of  $\text{H}_2\text{SO}_4$  (Dae Jung, 96%) dissolved with 2 g of  $\text{NaNO}_3$  (Aldrich, 99%) in an ice bath and stirred using a magnetic stirrer. Second, 12 g of  $\text{KMnO}_4$  (Aldrich, 99%) was added carefully to the mixture over a period of 15 min, with regular checks on the temperature of the reaction vessel to avoid a vigorous oxidative reaction. Then, the reaction temperature was adjusted to room temperature and maintained for 1 h. The reaction was stopped by adding 300 ml of distilled water slowly for and then  $\text{H}_2\text{O}_2$  (Dae Jung, 30 wt %) until bubbles in the mixtures disappeared, which indicated the disappearance of un-reacted  $\text{KMnO}_4$ . The oxidized graphite was centrifuged and cleaned repeatedly with distilled water (more than 16 times), until the centrifuged solution was about pH 7. The residues of graphite oxide were centrifuged and cleaned three times with absolute ethanol. Then the residues dispersed into 150 ml of 98% ethanol with 2% distilled water in a beaker and sonicated for 5 min using a sonicator (40 kHz and 200 W). After that the mixture was centrifuged and the supernatant containing the exfoliated GO was transferred to vials (e-GO). The residues was centrifuged and cleaned three times with distilled water. Then the residue was dispersed into 150 ml of distilled water and sonicated for 30 min. After that the mixture was centrifuged and the supernatant containing the exfoliated GO (ew-GO) was transferred to vials. For comparison, fresh graphite oxide were dispersed into 150 ml of distilled water in a beaker and sonicated for 30 min, centrifuged, and then transferred to vials (Ref. GO).

**Preparation of rGO.** We cooled the 30 ml solution with 0.02 wt% Ref. GO in a beaker using ice bath and then added 0.034 (rGO1), 0.064 (rGO2), 0.102 (rGO3), 0.136 (rGO4), 0.170 (rGO5), and 0.34 ml (rGO6) of 0.8 wt% hydrazine (80 wt%, Daejung), respectively. The reaction mixtures were stirred for 20 min and then kept for 24 days at room temperature. For

the resistivity test of rGO (in addition, a stability test of higher concentration rGO), we prepared a 30 ml of 0.2 wt% Ref. GO and reacted it with a corresponding amount of 8 wt% of hydrazine described above (namely, rGO1', rGO2', rGO3', rGO4', rGO5', and rGO6').

**Preparation of thin films of rGO' and e-GO.** For rGO films, as soon as hydrazine was added to the Ref. GO dispersion, it was diluted with 30 ml of ethanol (99.9%, Daejung). Then, after 20 min stirring, it was coated on glass substrate with a size of 4.5 cm x 4.5 cm using a spin coater at 850 rpm for 10 s. Then, each sample was dried at 120 °C for 3 min on a hot plate. The remained solutions were kept at room temperature. The same coating processes were carried out after 24 and 48 h, respectively. For e-GO films, 30 ml of 0.1 wt% of e-GO dispersion was diluted with 20 ml of distilled water and then coated on the same glass substrate and by the same method as for the preparation of rGO film. Transmittance and sheet resistance of the rGO' and e-GO films on glass were measured using a UV-Vis spectrometer (K-MAC, 2100V), and by the four-point probe method, respectively.

**Characterization of GO.** Photoluminescence (PL; Sinco, Fluomate FS-2) and excitation spectra were recorded to observe structural variation of the GO samples. In the case of rGO samples, PL was recorded periodically over 24 days to estimate the progress of reducing GO. All the samples were characterized using Raman (Horiba, LabRam HR), Fourier transform infrared spectroscopy (FT-IR; Bruker, IFS66v/S), X-ray photoelectron spectroscopy (XPS; VG Scientific), and transmission electron microscopy (TEM; JEOL JEM-2100F).

## Periodic exfoliated GO

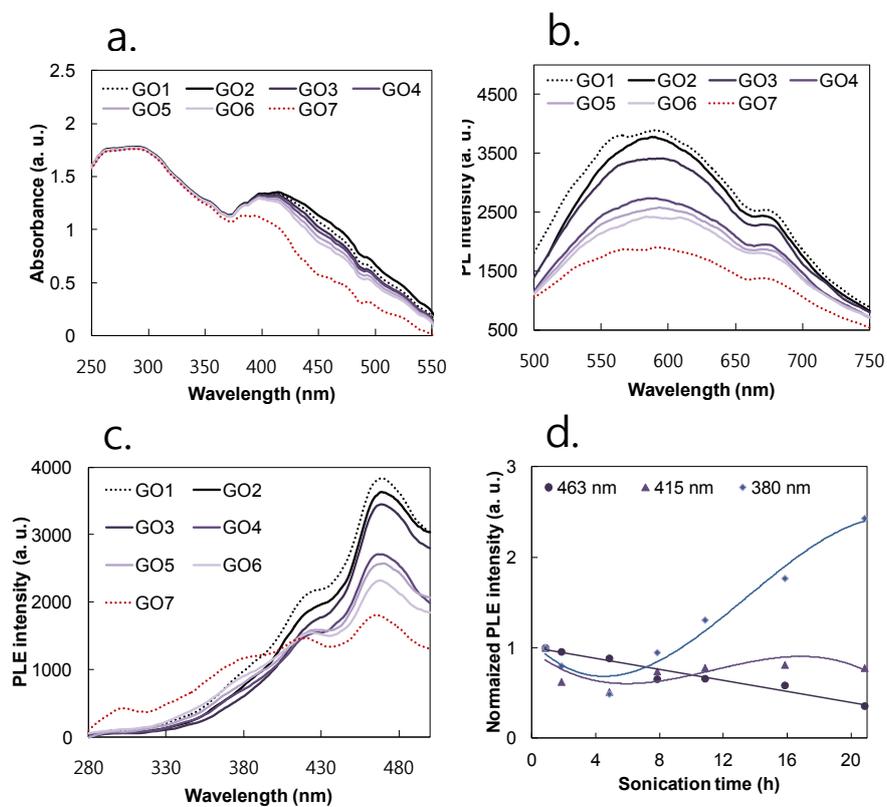


Fig. S1. (a) Quantitatively normalized absorption spectra of various GOs sonicated for different periods to below 300 nm peaks, (b) PL spectra of normalized GO samples excited at 463 nm, (c) PL excitation spectra of the normalized GO samples, (d) Relative peak intensity in the PLE spectra of (c).

## XPS spectra of N-e-GO and N-ew-GO

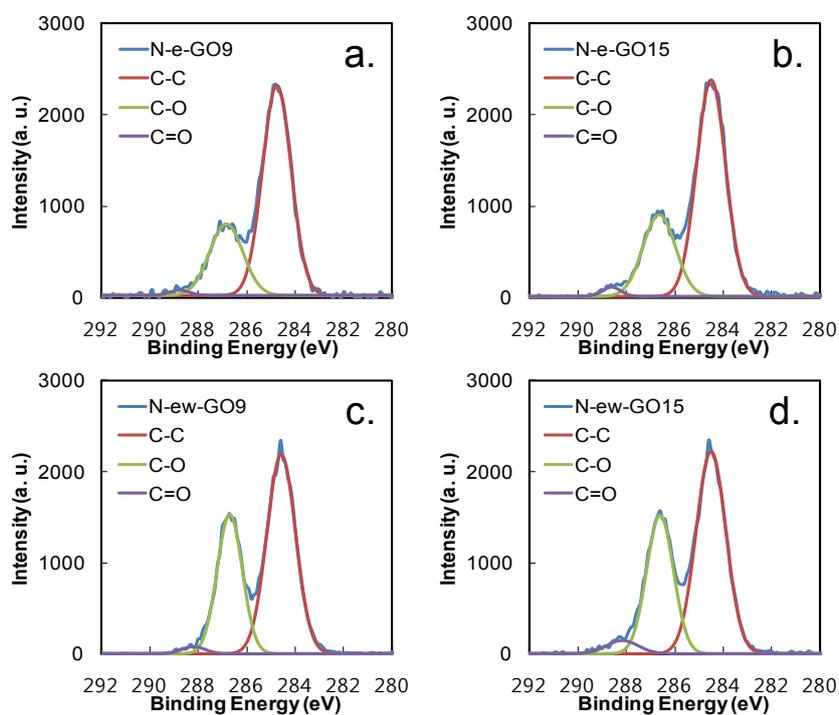


Fig. S2. XPS spectra of C1s of various e-GO and ew-GO samples prepared at different oxidation condition: (a) N-e-GO9, (b) N-e-GO15, (c) N-ew-GO9, (d) N-ew-GO15. All the XPS spectrum are fitted after a Shirley background correction. N-e-GO and N-ew-GO were prepared using the same method as for e-GO and ew-GO varying the content of oxidant of  $\text{KMnO}_4$  (9g: N-e-GO9 and N-ew-GO9, 15g: N-e-GO15 and N-ew-GO15).

## PL and PLE spectra of rGO samples to estimate the reaction rate of GO reduction using hydrazine

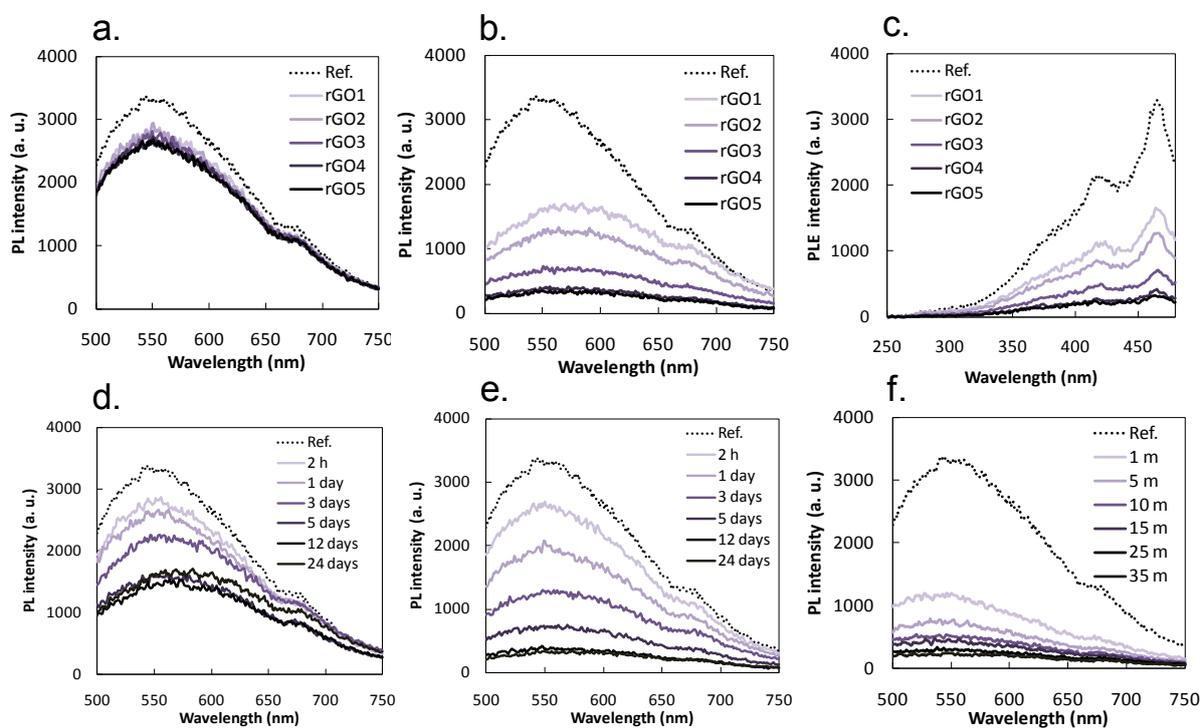


Fig. S3. PL and PLE spectra of rGO excited at 463 nm: (a) Measured after 2 h, (b) measured after 24 days, (c) PLE spectra of (b), (d) Selected PL spectra of rGO1 according to the reaction time, (e) Selected PL spectra of rGO5 according to the reaction time, (f) PL spectra of rGO6 measured *in situ*, low concentration of GO solution, 0.02 wt%, was used to avoid coagulation of rGO sheets as shown in Fig. 3.