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Electronic Supplementary Information for

Highly conductive reduced graphene oxide *via* pressure-assisted reduction at mild temperature for flexible and transparent electrodes

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1. Experimental Section

Water-soluble GO powder was prepared by modified Hummer's method.[1] A mixture of graphite (1 g), $K_2S_2O_8$ (0.5 g) and P_2O_5 (0.5 g) was placed in H_2SO_4 (1.5 mL) solution at 80°C for 6 h (all chemicals were purchased from Sigma Aldrich, St. Louis, MO, USA). Then, the mixed solution was diluted, filtered and washed sequentially on the filter with deionized water. The washed mixture was dried in a vacuum oven for overnight. Next, the peroxidated graphite (1 g) and $NaNO_3$ (0.5 g) were added to H_2SO_4 (23 mL) solution and placed in an icebath at 0°C for 30 min. Then, $KMnO_4$ (3 g) was injected into the mixed solution and carefully stirred overnight. Deionized water was added to the mixed solution over 2 h and the temperature was increased to 98°C slowly. Subsequently, 30 wt% aqueous H_2O_2 solution was added to the mixture and stirred for 3 h, producing a brown-colored slurry. The slurry was washed with 10wt% HCl. Additional washing was performed with deionized water to neutralize the solution. Finally, GO powder was obtained after drying and a GO suspension was prepared by dispersing the GO powder in deionized water. The fabricated GO suspension was stable for several months without an external stimulus.

A final concentration of GO solution (2 mg/mL) was obtained by mixing of the GO suspension with ethanol at a volume ratio of 50% ethanol to water. Before the spin-coating process, the PES substrates were plasma-treated (200 W, 8 SCCM O_2) to modify their surface wettability. The substrates were completely covered with a sufficient amount of the GO suspension.

Hydrazine vapor reduction was performed prior to thermal reduction. The spin-coated GO thin films were placed in a clean glass chamber containing 1 mL of hydrazine (30%, Sigma Aldrich). The chamber was sealed and placed in an oven at 100°C for 1 h. The color of the GO thin films changed from brown to metallic gray by chemical reduction. Thermal

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reduction was carried out following hydrazine vapor reduction. The sample was heated to 180°C at a heating rate of 5°C min^{-1} in a tubular furnace under an argon atmosphere, and held the temperature for 30 min. Pressure-assisted reduction was carried out inside a glovebox. The sample was placed in a hot press between stainless-steel plates at 180°C for 30 min.

The OTFTs were fabricated with a bottom-gate and top-contact configuration. The as-prepared PRGO thin film on the PES substrate was used as a gate electrode. Polyvinylpyrrolidone (PVP), a polymeric dielectric material, and poly(melamine-co-formaldehyde), a cross-linking agent, were dissolved in propylene glycol monomethyl ether acetate (PGMEA, 10 mL) at a molar ratio of 2:1. The PVP solution was spin-casted at 4000 rpm for 30 s and subsequently cross-linked at 130°C for 15 min and 200°C for 5 min in N_2 atmosphere, forming an about 300 nm PVP gate dielectric. On top of the cross-linked PVP dielectric layer, pentacene film was thermally evaporated in a vacuum chamber by a shadow mask under a pressure of 5×10^{-6} Torr at room temperature at a deposition rate of 0.4 Å s^{-1}. The pentacene semiconductor was about 60 nm thick. The gold S/D electrode (50 nm) was thermally deposited on the PES substrate at a deposition rate of 1.0 Å s^{-1}.

X-ray photoelectron spectroscopy (XPS) was performed using an ASCALab220i-XL electron spectrometer form VG Scientific (Cedex, France) with 300 W Al Kα radiation at a base pressure of 3×10^{-9} mbar. High power X-ray diffraction (XRD) measurements were carried out using an M18XHF-SRA (Mac Science, Yokohama, Japan) diffractometer equipped with a Cu Kα radiation source (k = 1.5406 A) at 40 kV and 300 mA (12 kW). The XRD peaks were collected between 10° and 40° at a scan rate of 4 min^{-1}. The plasma reactor was a parallel-electrode type with a 13.56 MHz radio-frequency generator. Contact angles were determined with a DSA10 contact angle analyzer (Krüss, Matthews, NC, USA) interfaced to drop shape analysis software. Electrical resistance was measured using a 2400

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sourcemeter (Keithley, Cleveland, OH, USA) at 25°C by a four-probe method. Scanning electron microscopy (SEM) images were acquired with JSM-6700F microscope (JEOL, Tokyo, Japan) at an acceleration voltage of 10 keV. Film transmittance was measured using a UV–visible spectrometer (Lamda-20; Perkin-Elmer, Waltham, MA, USA) at a resolution of 1 nm. Atomic force microscopy (AFM, XE-70; Park Systems, Suwon, Korea) in tapping mode was used to examine the surface roughness of RGO thin films. Transmission electron microscopy (TEM) images were acquired with a JEOL JEM-200CX microscpoe at an acceleration voltage of 200 kV. For the microtome process, SPURR's kit resin (Electron Microscopy Sciences, Fort Washington, PA, USA) was mixed with the powder MSNF, and cured for 24 h at 70°C. Transistor performance was characterized using an Agilent HP4155C semiconductor parameter analyzer (Agilent, Santa Clara, CA, USA). Capacitance voltage characteristics of the metal–insulator–metal (MIM) capacitors were measured using an HP 4284 precision LCR meter (Agilent, Santa Clara, CA, USA).

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2. X-ray photoelectron spectroscopy (XPS) analysis

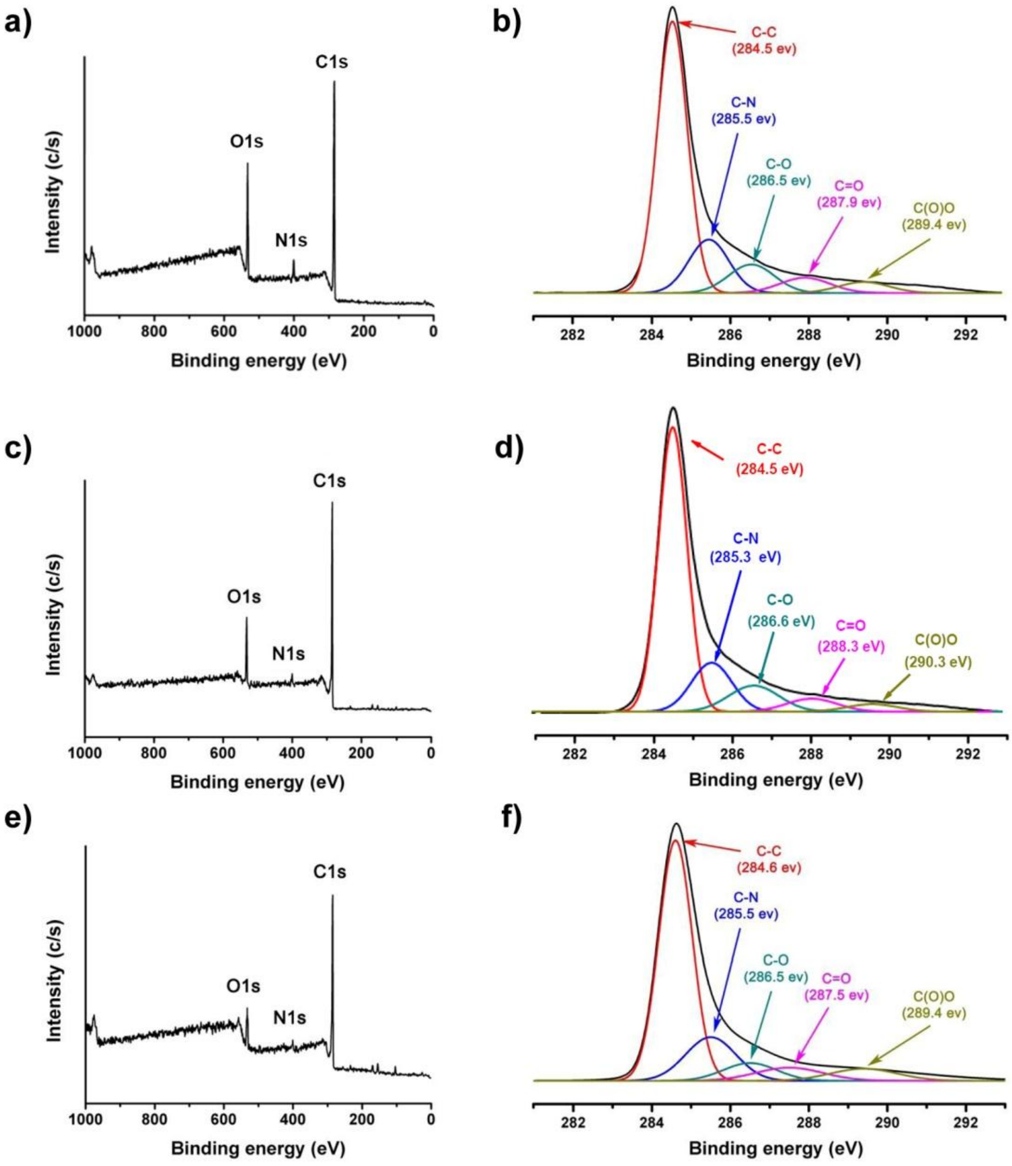

Fig. S1 High-resolution XPS analysis of the effect of different reduction treatments on rGO thin films. (a), (c), (e) XPS survey spectra and (b), (d) (f) XPS C1s spectra of CRGO, TRGO, and PRGO, respectively.

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Table S1. C/O atomic ratios of GO and rGO according to reduction method.

	GO	CRGO	TRGO	PRGO
C/O ratio	3.0	6.9	8.6	10

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3. X-ray diffraction (XRD) analysis

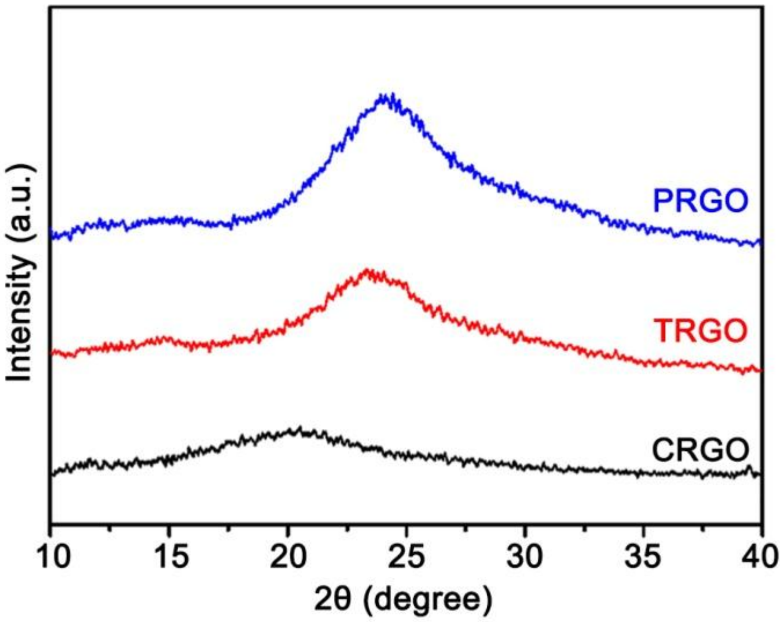

Fig. S2 XRD patterns of CRGO, TRGO, and PRGO.

X-ray diffraction (XRD) analysis was used to measure the interlayer spacing between graphene multi-layers, which corresponded to the degree of reduction of rGO. In Fig. S2, the XRD patterns of rGO prepared using different reduction methods exhibited a peak at around 24°, which is a typical characteristic of rGO. The rGO peak appeared at a higher angle in PRGO than in CRGO and TRGO. The interlayer spacings of CRGO, TRGO, and PRGO, calculated based on their 2θ position using Bragg's law, were 0.430, 0.382 and 0.364 nm, respectively. The relatively small interlayer spacing of PRGO suggests a denser packing of multi-layers compared to CRGO or TRGO.[2] These results are similar to the intrinsic interlayer spacing of graphite, indicating good reduction of GO by pressure-assisted process.[3] Thus, a small interlayer spacing indicates the removal of oxygen- and nitrogen-containing functional groups from GO and subsequent restoration of the sp^2 carbon network.

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4. Transmission spectra

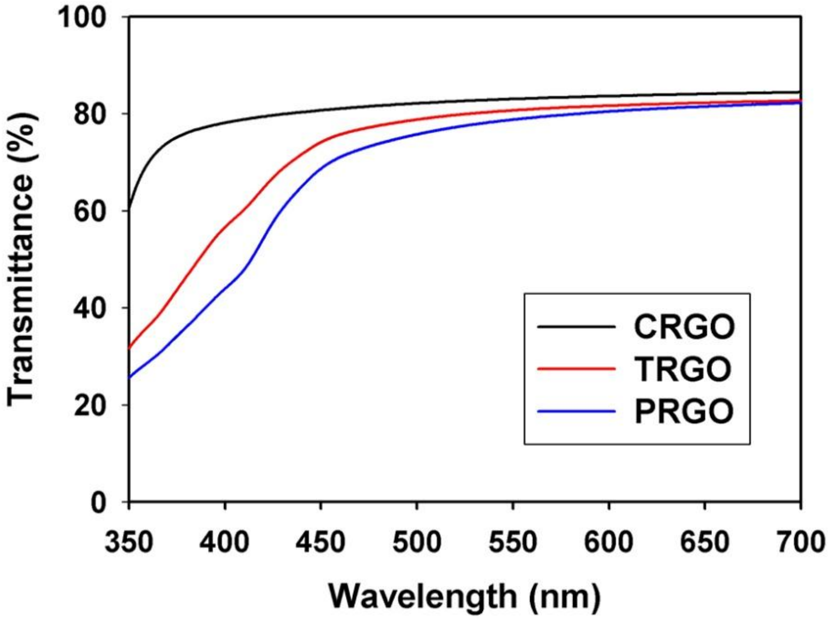

Fig. S3 UV-Vis spectra of CRGO, TRGO, and PRGO. Transmittances of each material at 550 nm are 83.0, 80.6, and 78.7 %, respectively.

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5. Bending fatigue test

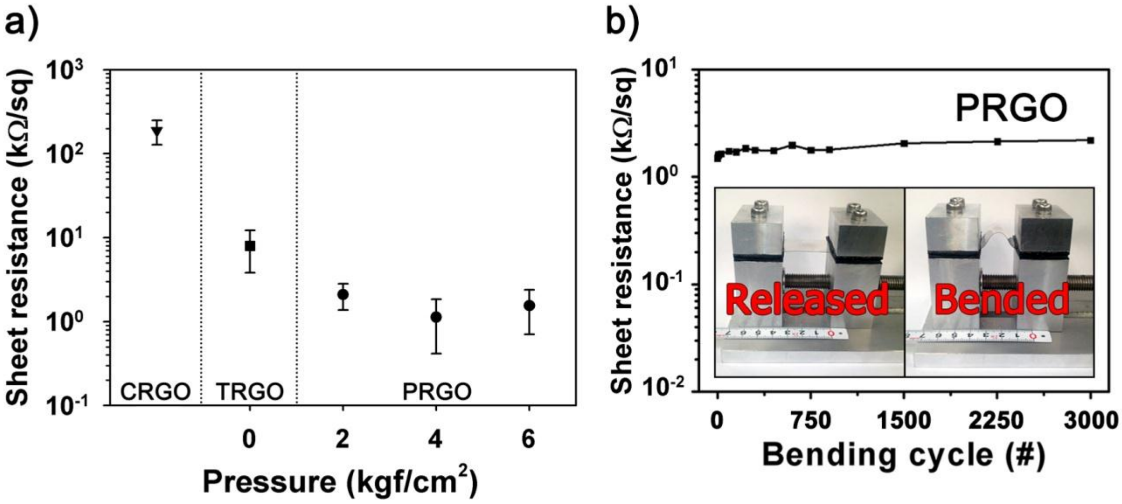

Fig. S4 (a) Comparison of surface resistance for different reduction methods according to pressure. (b) Continuous bending fatigue test of the PRGO thin film.

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6. Photograph of PRGO-gated OTFT

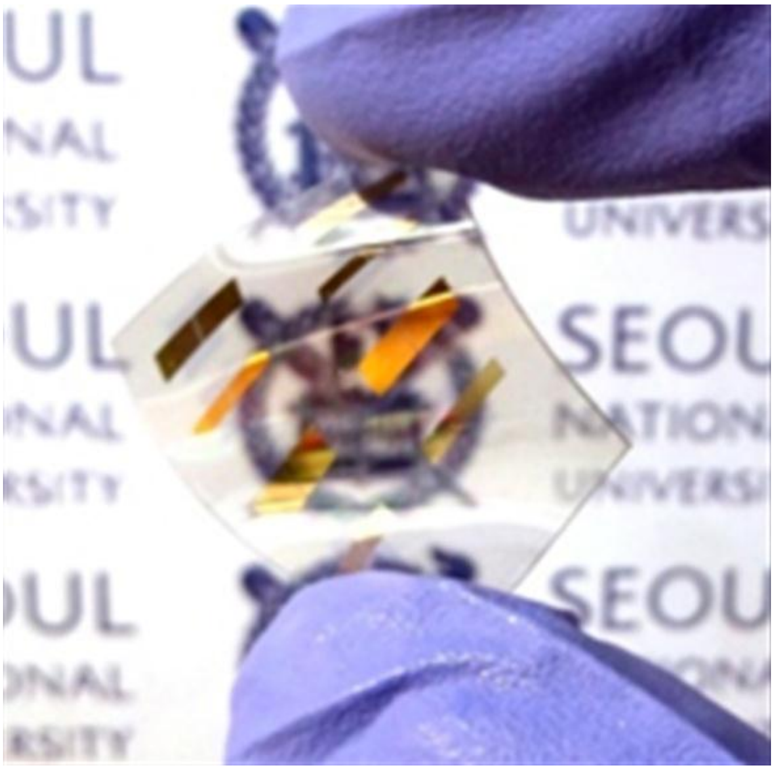

Fig. S5 Digital photograph of the PRGO-gated OTFTs.

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7. References

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