Supporting Information

Single-Layer Graphene-Based Transparent and Flexible Multifunctional Electronics for Self-Charging Power and Touch-Sensing System

Sungwoo Chun[1†], Wonkyeong Son[2†], Gwangyeob Lee[3], Shi Hyeong Kim[4], Jong Woo Park[5], Seon Jeong Kim[5], Changhyun Pang[1,6], and Changsoon Choi[2*]

[1]Department SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon, Gyeonggi-do 16419, Republic of Korea

[2]Division of Smart Textile Convergence Research, DGIST, Daegu 42988, Republic of Korea

[3]Department of Materials Science and Engineering, Yonsei University, Seoul 03722, Republic of Korea

[4]Research Institute of Industrial Technology, Ansan, 15588, Korea

[5]Center for Self-powered Actuation, Department of Biomedical Engineering, Hanyang University, Seoul 04763, Republic of Korea

[6]School of Chemical Engineering, Sungkyunkwan University (SKKU), Gyeonggi-do 16419, Republic of Korea

[†] S. C. and W. S. contributed equally to this work.

*To whom correspondence should be addressed. E-mail: cschoi@dgist.ac.kr (C.C.)

Supporting Methods

Single-layer graphene growth and transfer

The SLG sheet was obtained using a direct growing method involving custom-designed thermal chemical vapor deposition (CVD). The catalytic metal of 25 μm copper foil (purity: 99.8%, Alfa Aesar Co.) is chosen because of its low solubility with carbon which is expecting self-limiting process required for large area graphene formation. The catalytic metal foil is placed on a quartz holder and then heated up to the growth temperature (1000 °C) for 5 min in an argon atmosphere after pre-pumping down to 10^{-6} Torr. After an interval to allow the stabilization of substrate temperature, a mixture of methane (40 sccm) and hydrogen (10 sccm) gases is introduced into the quartz tube. Transparent and flexible PEN and PTFE were chosen as the substrates for coatings of SLG sheets. After spin coating of polymethyl methacrylate (Micro Chem. Co. 950K PMMA C4) in chlorobenzene solution on the grown SLG, the copper foil below the SLG was removed by treatment with $FeCl_3$ solution for 3 h. Cleaning was followed by washing with a diluted HCl solution to remove Cu residues, and the SLG film was then baked for 30 min at 85°C. After removing PMMA using acetone for 10 min, the SLG film was etched to the effective area (3.5×3.5 cm^2) with O_2 plasma at a power of 30 W after lithographic patterning. Finally, electrodes were formed with a sputtered Pt film (80 nm) using a stencil mask.

Preparation of SLG-based supercapacitor and capacitive touch sensor as top panel

The PAN electrospun mat was fabricated by spinning PAN solution using electrospinning system (ESR200RR2D, Nano NC). The PVA–LiCl gel and LiCl solutions (0.4 M) were used as electrolytes for electrochemical performance characterization. More specifically, the gel electrolyte was prepared by mixing 3 g PVA (Sigma Aldrich, USA) and 6 g LiCl (Sigma-Aldrich, USA) in 30 ml deionized water and heating it at 90 °C until it became transparent. The

PAN electrospun mat functions as a transparent separator. Accounted fiber diameter of the spun mat exhibited that more than 80% were in range of 500~900 nm, indicating relatively uniform size distribution. The SLG films on the PEN substrates fabricated above were used as electrode. By sandwiching the quasi-solid state PAN mat electrolyte and separator between the SLG-coated plates, the SLG-based supercapacitor and capacitive pressure sensor was finally completed.

Preparation of TENG device as bottom panel

As dielectric materials for friction, the PEN and PTFE surfaces are selected because those show opposite and far difference of electron affinity. After coating of SLG sheet on each surface, the PEN/SLG panel and PTFE/SLG panel were assembled with an acryl bumper (~ 1 mm height) to isolate the facing frictional surfaces (PEN and PTFE) with no pressure.

Electrochemical measurements

The capacitance of the two-electrode system was calculated from CV curves. From $C = I/(dV/dt)$, where I is average discharge current from the CV curve and the dV/dt is the scan rate, the single-electrode specific areal capacitance was calculated from the following Equation Areal capacitance (F/cm^2) = $2C/A_{surface}$ where $A_{surface}$ is the area of the single layer graphene. Electrochemical measurement utilized a two-electrode configuration using an electrochemical analyzer (Vertex EIS, Ivium).

Electromechanical measurements

For measuring electrical responses by dynamically applied pressures in TENG devices and SLG capacitive touch sensor, a homemade measurement apparatus equipped with z-axis stage pressure inductor to supply vertical strain was used. The NI precision system SMU (PXIe-4139) was used to collect electrical data from the two contact channels of the devices.

Characterizations

To obtain images of the SLG film on PEN, multifunctional electronic devices a cell phone camera (Samsung Galaxy Note 8) was used. The field-emission scanning electron microscopy (FE-SEM) (Hitachi S-4800) was employed for morphology of PI electrospun mat. Double monochromator (Perkin Elmer Co.) with a wavelength range from 200 to 800 nm and a resolution of 0.15 nm was used for the transmittance analyses.

Supporting Figures

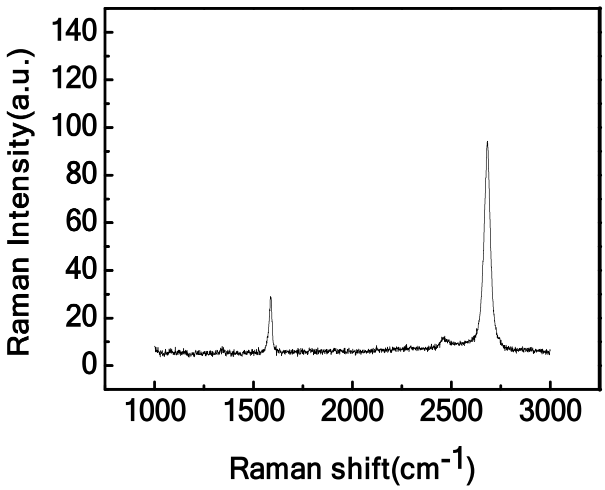

Figure S1. Raman spectrum of SLG film. The excitation wavelength is 532 nm and the laser power is 2 mW. The Raman resonance observations indicate typical single-layer graphene with a reasonably small ratio of intensities (~0.31) of the G band (1587 cm^{-1}) to the 2D band (2682 cm^{-1}).

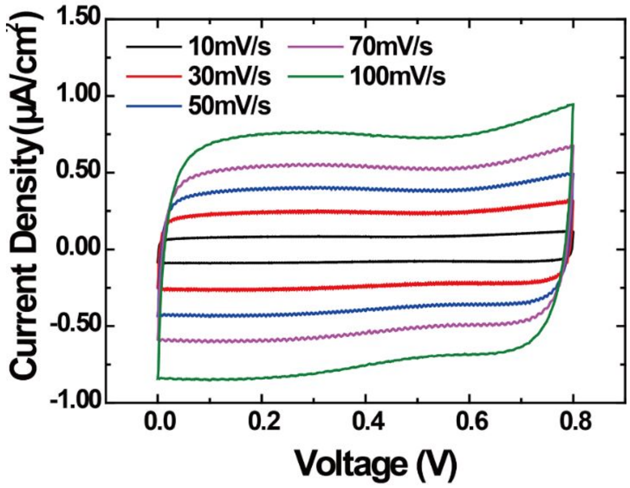

Figure S2. CV curves for transparent supercapacitor of bottom panel measured from 100 to 1000 mV/sec scan rates

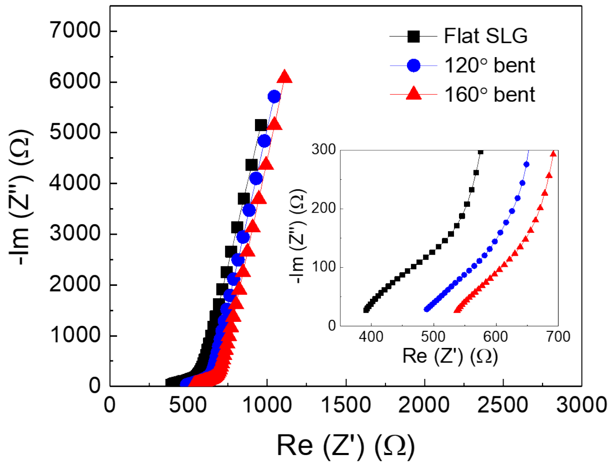

Figure S3. Nyquist curves (20 ~ 50000 Hz) of SLG electrode with various bending degrees are measured in three electrode system using 0.4M LiCl electrolyte. And Ag/AgCl, and Pt mesh were used as reference, and counter electrodes, respectively (Inset shows magnified Nyquist curves at high frequency range).

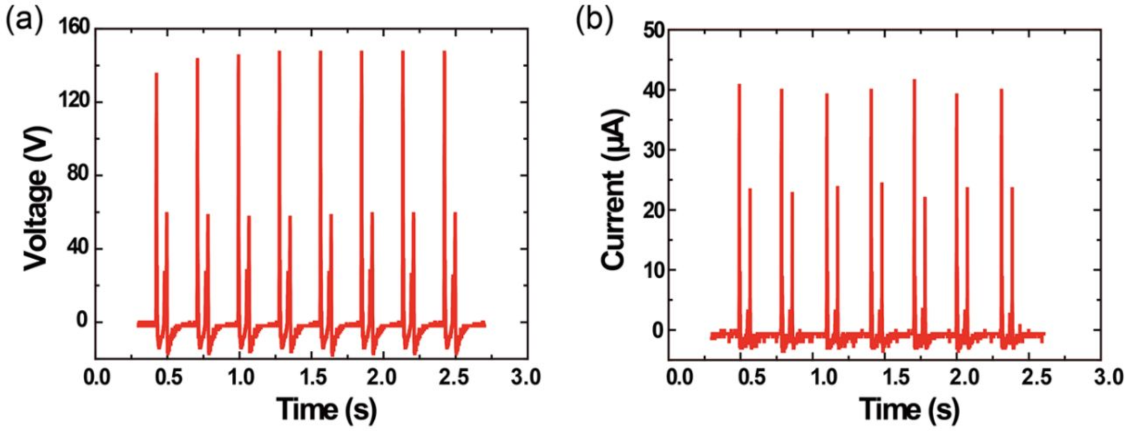

Figure S4. Rectified (a) voltage and (b) current output performance of transparent TENG bottom panel when vertical pressure (17 kPa) was applied and released.

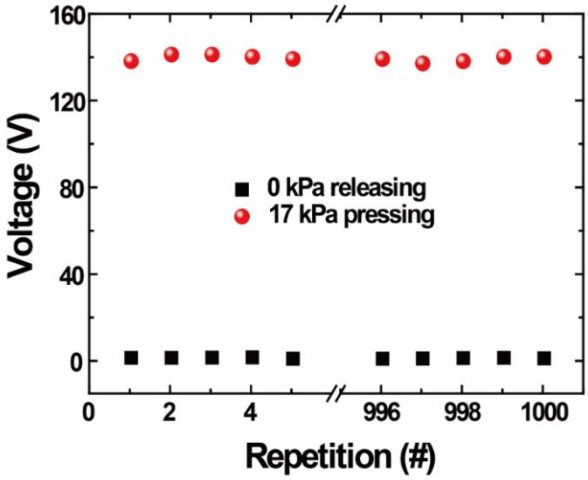

Figure S5. Consistent output performances for output voltage during the 1,000 repetitions with vertical pressure of 17 kPa.

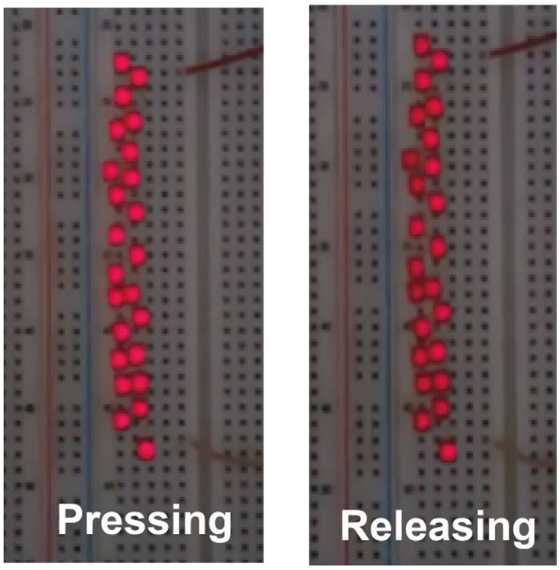

Figure S6. Photo images showing 24 red LED powering by transparent TENG bottom panel when pressing and releasing of vertical pressure (17 kPa).