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Supporting Information

Integrated Self-Charging Power Unit with Flexible Supercapacitor and Triboelectric Nanogenerator

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Figure S1. Schematic diagram of the fabrication of wrinkle structure PDMS. Detailed process with (a) PET/ITO film, (b) spin coating PDMS, (c) uncured PDMS treated with a C_4F_8 plasma treatment and (d) wrinkle structure formed with PDMS cured.

The detailed fabrication diagram of the wrinkle structure^[1] is shown in Figure S1. Firstly, PDMS mixture was spin coated in the PET/ITO film. Then with the C_4F_8 plasma treatment process, the uncured PDMS surface was deposited by the fluorocarbon polymer with a velocity. Considering the fact that deposited fluorocarbon polymer is a stiff layer compared to the PDMS layer, both of the stress mismatching and the velocity would deform PDMS. Finally, the PDMS layer would recover to its original state after getting cured, leading to the occurrence of the wrinkle structure on this multilayered film.



Figure S2. Illustration of simulated CNT network percolation. In the first drop-dry process, CNT coated (a) and connected (b) distribution in the paper substrate. After several drop-dry processes, CNT coated (c) and connected (d) redistribution in the paper substrate.

Monte Carlo simulation was used to validate the percolation of CNT in the paper substrate.^[2] For the 2D simulation square cell measuring 1000 μ m × 1000 μ m, the CNT ink with 1 mg/mL was firstly dropdried, in which assumed 100 virtual lines of CNTs were randomly distributed in blue line. These lines had a fixed length of 100 μ m, with the percolation probability being determined from the formations of fully connected CNTs (red line), where no conductive path was formed in the cell. After several dropdry processes, cell was almost coated with CNTs, resulting in several conductive paths formation and prove the capability to work as supercapacitor electrode.



Figure S3. (a) SEM image of CNT/paper electrode. (b) Photograph of a vial containing an aqueous dispersion of 1 mg ml⁻¹ CNTs.



Figure S4. The normalized voltage output of the wrinkle TENG shows good stability after 30,000 cycles.



Figure S5. Capacitance retention of the solid-state supercapacitor during 4,000 cycles at the scan rate of 100 mV/s.



Figure S6. Two upward and downward peaks of output voltage signal under 2 Hz compressive stress of two TENGs connected in parallel.



Figure S7. Initial charging curves of the sandwiched SC charged by single TENG at various frequencies



Figure S8. A calculator could be driven by self-charging power unit continuously over 40 minutes.



Figure S9. Schematic diagram of two-electrode electrochromic device with (a) black state and (b) transparent state.

Detailed fabrication of electrochromic device (ECD) was as follows:

Firstly, the gel electrolyte for the ECD was prepared with 0.5 mmol of AgNO₃ (85 mg) as electrochromic material, 2.5 mmol of Tetra-n-butylammonium bromide (TBABr) (806 mg) as supporting electrolyte, and 0.1 mmol of CuCl₂ (13 mg) as electrochemical mediator.^[3] After all of them were dissolved in 10 mL of Dimethyl sulfoxide (DMSO) evenly, 10 wt% of Polyvinyl butyral (PVB) as host polymer was then mixed into the DMSO-based electrolyte solution. Subsequently, the ECD was assembled by sandwiching PVB-based gel electrolyte between the two flat ITO electrode as working electrode and counter electrode with a Surlyn spacer (30 um), the window area of which was patterned with "EC" words. After the ECD was heated in 130°C for 10 minutes, the ECD was eventually fabricated.

When the SCPU changed to the energy supply mode, it could form a positive potential between the two electrodes of the ECD device. During the voltage sweeping from zero to positive voltage, an auxiliary redox process would occur in the electrode surface. The voltage above 2.0 V to the ITO electrode caused the dissolved Ag^+ ion to be deposited on the ITO electrode, resulting in a black color state. Then with the SCPU turn off the energy supply mode, there would be no voltage potential between two electrodes. The Ag began to dissolve in the electrolyte and the ECD device changed back to the transparent state.

References:

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