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Electronic Supplementary Information

Highly stretchable hybrid nanomembrane supercapacitors

Keon Jung Kim[a], Jae Ah Lee[a,b], Márcio D. Lima[b], Ray H. Baughman[b], Seon Jeong Kim*[a]

*E-mail: sjk@hanyang.ac.kr

Materials and method

Materials

Spinable aligned arrays of multi-walled carbon nanotubes forests were grown on a silicon wafer by the chemical vapour deposition method[27]. Iron(III) p-toluenesulfonate hexahydrate (Fe(III)PTS, M_w = 677.52), pyridine (anhydrous, 99.8%), 1-butanol (for molecular biology, ≥99%), and 3,4-ethylenedioxythiophene (EDOT) monomer (97%), poly(vinyl alcohol) (PVA) (M_w = 146,000, 186,000), and LiCl (M_w = 42.39) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Ecoflex 0050 and Sil-Poxy were from Smooth-On.

Fabrication of hybrid nanomembranes

A 8 wt % Fe(III)PTS/butanol solution was used as an oxidizing agent. Pyridine (1.6 vol %) was added to the 8 wt % Fe(III)PTS/butanol solution. The oxidizing solutions were slowly flowed over two-layer CNS. The CNS were allowed to dry at 60 °C for 20 min to completely evaporate the solvent. The EDOT monomer was cast next to the densified CNS in a VPP chamber, and the samples were then exposed to EDOT vapor at 60 °C for 1 h. After polymerization, the samples were rinsed several times in ethanol to remove unreacted oxidant[11].

Fabrication of stretchable supercapacitor electrodes

An Ecoflex rubber layer (2.6 cm × 2.6 cm, 1.25 mm thick) was fully stretched and then held to a homemade template (8.0 cm × 8.0 cm). The 15-layer CNS (width: ~2.1 cm) to be used as a current collector were alternatively stacked onto the pre-stretched Ecoflex rubber and densified by ethanol. The PEDOT/CNS hybrid nanomembrane was transferred onto the densified CNS current collector in an ethanol bath. After densifying, the pre-stretched electrode was coated with a PVA/LiCl gel electrolyte. The 4.5 M PVA/LiCl gel electrolyte was fabricated by mixing PVA (3 g), LiCl (6 g), and deionized water (30 mL) using a stirring bar for 3 h at 90 °C until it became transparent.

Supercapacitor assembly

Two symmetric electrodes of a hybrid nanomembrane supercapacitor were separately prepared and then assembled. A solid electrolyte-absorbed nylon panty hose was used as a stretchable separator to avoid electrical shorting between two electrodes during stretching/releasing. Al foils were respectively connected with the CNS current collectors of both electrodes and then the two sandwich structures were sealed by Sil-Poxy glue.

Characterization

Surface morphology and height profiles of the hybrid nanomembranes were obtained using SEM (S4700; Hitachi, Tokyo, Japan). Potentiostatic and electrochemical impedance measurements were performed using a Reference 600 potentiostat (Gamry Instruments, Warminster, PA USA). Two-probe uniaxial stretchability changes were measured using a homemade device.

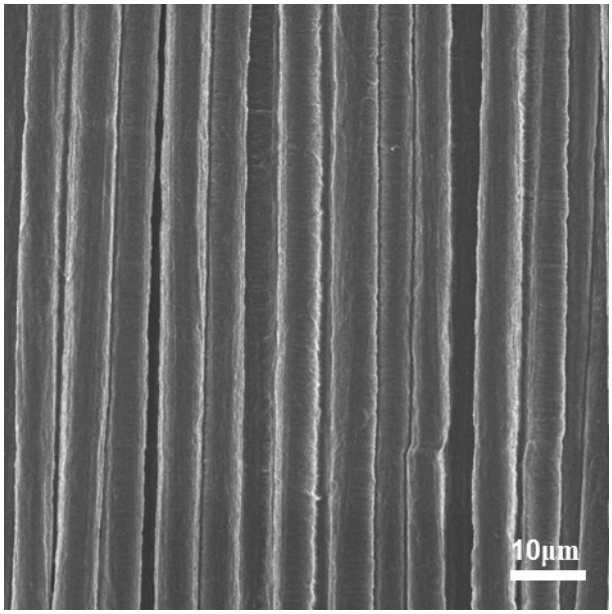

Fig S1. The SEM image of uniaxial-wrinkled structure of hybrid nanomembrane after releasing (200% by x-axis direction only).

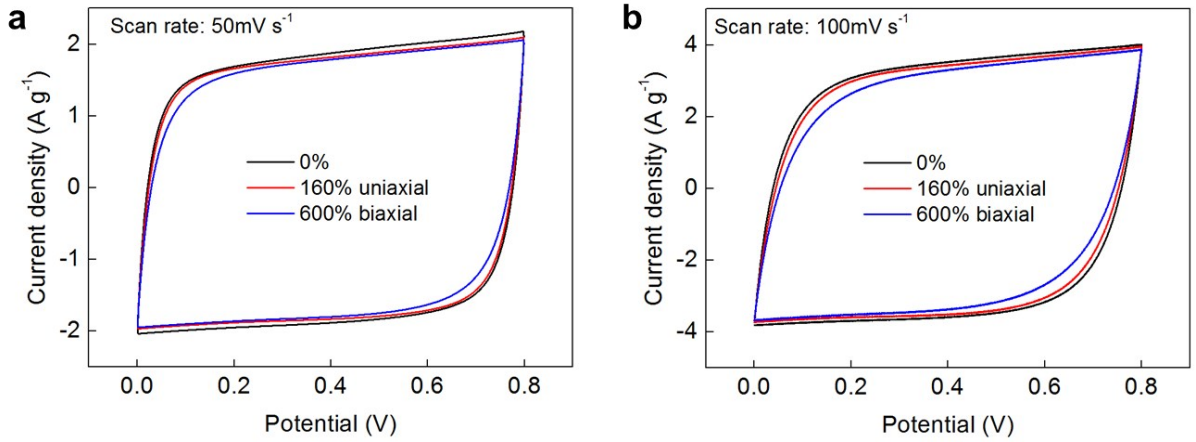

Fig S2. a) Cyclic voltammetry curves at strains of 0%, 160%, and biaxial 600% with scan rate of 50 mV s^{-1} and b) 100 mV s^{-1}.

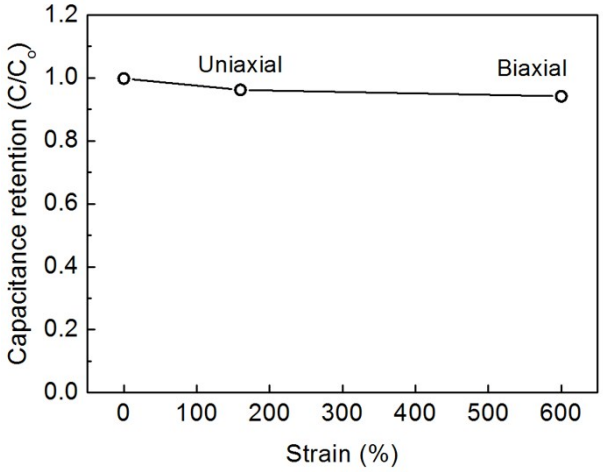

Fig S3. Capacitance retention of supercapacitor with increasing strain. (Scan rate: 10 mV s^{-1})

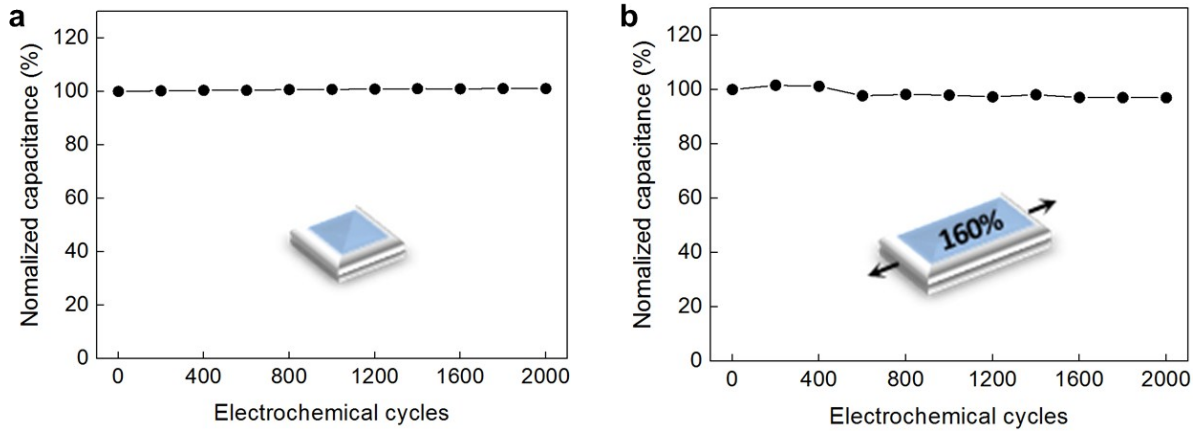

Fig S4. a) Dependence of capacitance ratio on electrochemical cycle number with 0% strain and b) 160% strain.

Table S1. Summary of our highly stretchable supercapacitor results and comparison with other stretchable pseudocapacitors plotted in Figure 3f.

Active materials	Energy density, E_{max} (Wh kg^{-1})	Strain (%)	Ref.
PEDOT/CNS	7.28	0	☆This work
	7.02	160	
	6.87	biaxial 600	
Polypyrrole /Nylon Lycra fabric	6.7	0	□ ref.15
	8.7	20	
	9.4	40	
	11.1	60	
Polyaniline /multi-walled carbon nanotubes	11	0	○ ref.16
	10.78	50	
MnO_2/carbon nano particles	4.8	0	△ ref.9