Supporting Information

Twist-Stabilized, Coiled Carbon Nanotube Yarns with Enhanced Capacitance

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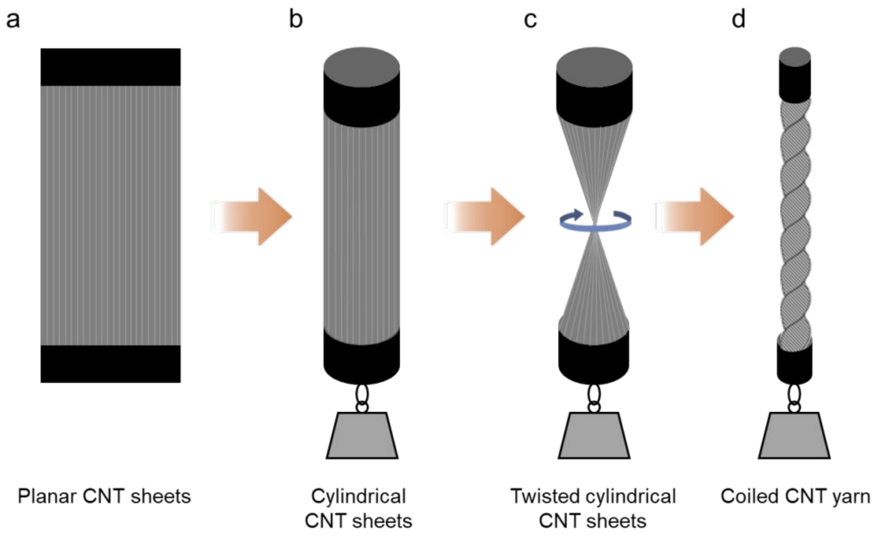

Figure S1. (a) A stack of forest-drawn CNT sheets was rolled into (b) a cylinder and twisted under an applied load to produce (c) a twisted yarn, which was subsequently twisted under the same applied load to produce (d) a coiled CNT yarn.

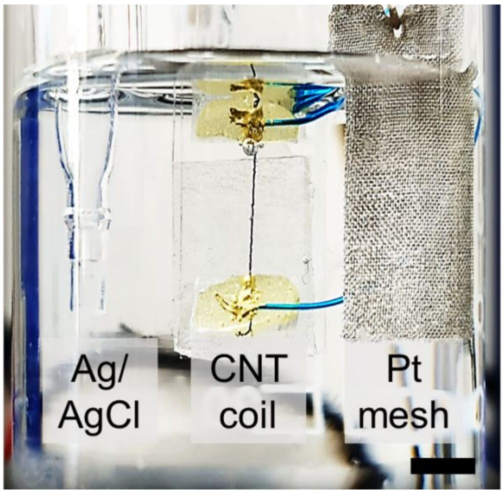

Figure S2. A photograph of the experimental setup for electrochemical oxidation (ECO) treatment using a three-electrode system consisting of a 10 mm-long coiled CNT yarn (the working electrode), Ag/AgCl (the reference electrode), Pt mesh (the counter electrode), and 0.1 M Na_2SO_4 (the electrolyte) (scale bar = 5 mm).

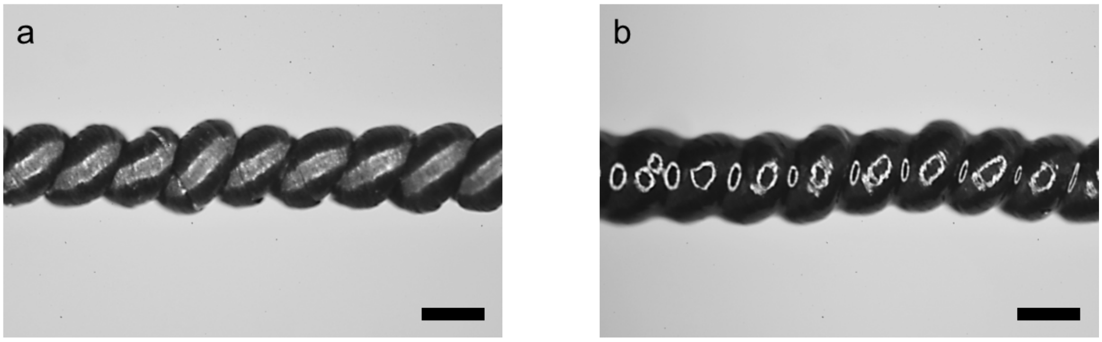

Figure S3. Optical images showing (a) pristine coiled carbon nanotube (CNT) and (b) electrochemical oxidation (ECO)-treated coiled CNT yarns after immersion in water (scale bar = 200 μm), respectively.

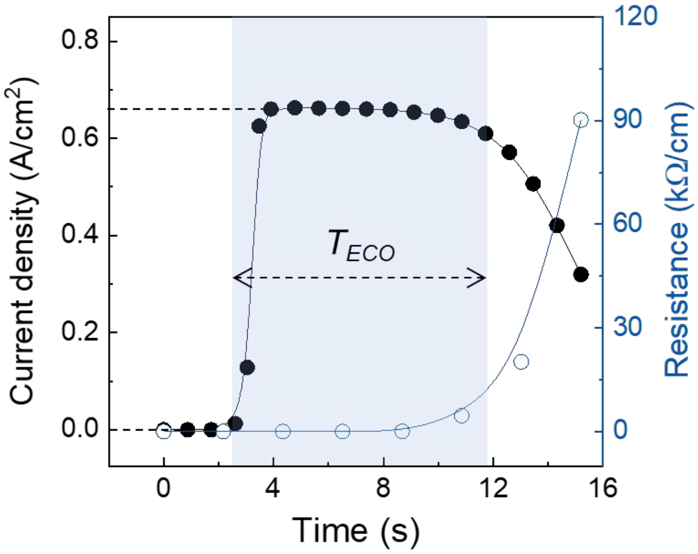

Figure S4. Current density and yarn resistance versus oxidation time under a constant voltage of 4.5 V (vs. Ag/AgCl).

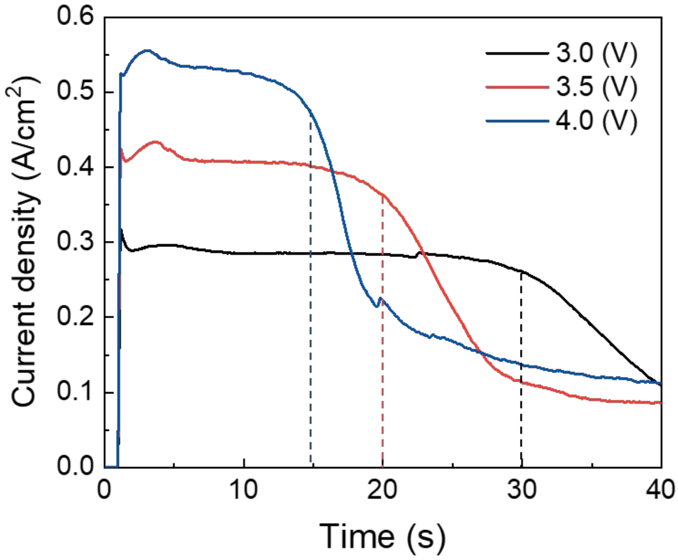

Figure S5. Current density (normalized to the external surface area of electrochemical oxidation (ECO)-treated coiled carbon nanotube (CNT) yarn) versus oxidation time for various voltage applications ranging from 3.0 to 4.0 V (vs. Ag/AgCl). The dotted lines indicate the endpoints of the optimized oxidation time (T_{ECO}) at each voltage.

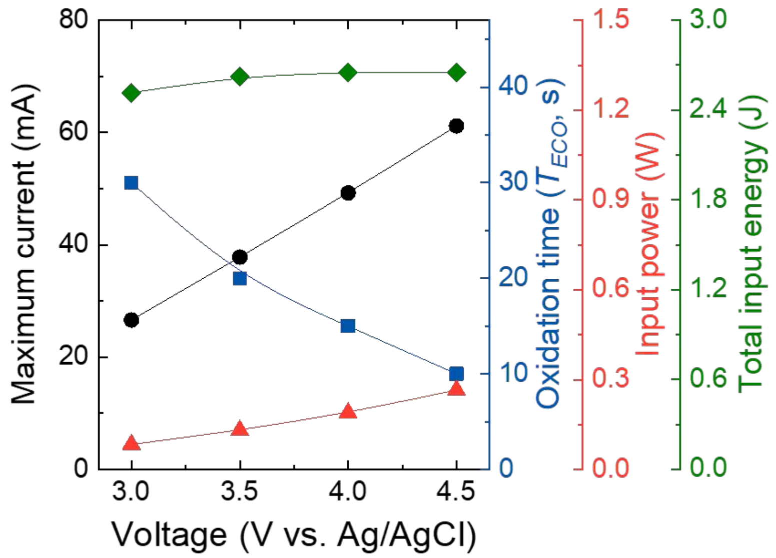

Figure S6. Maximum current, oxidation time, input power, and total input energy versus applied voltage.

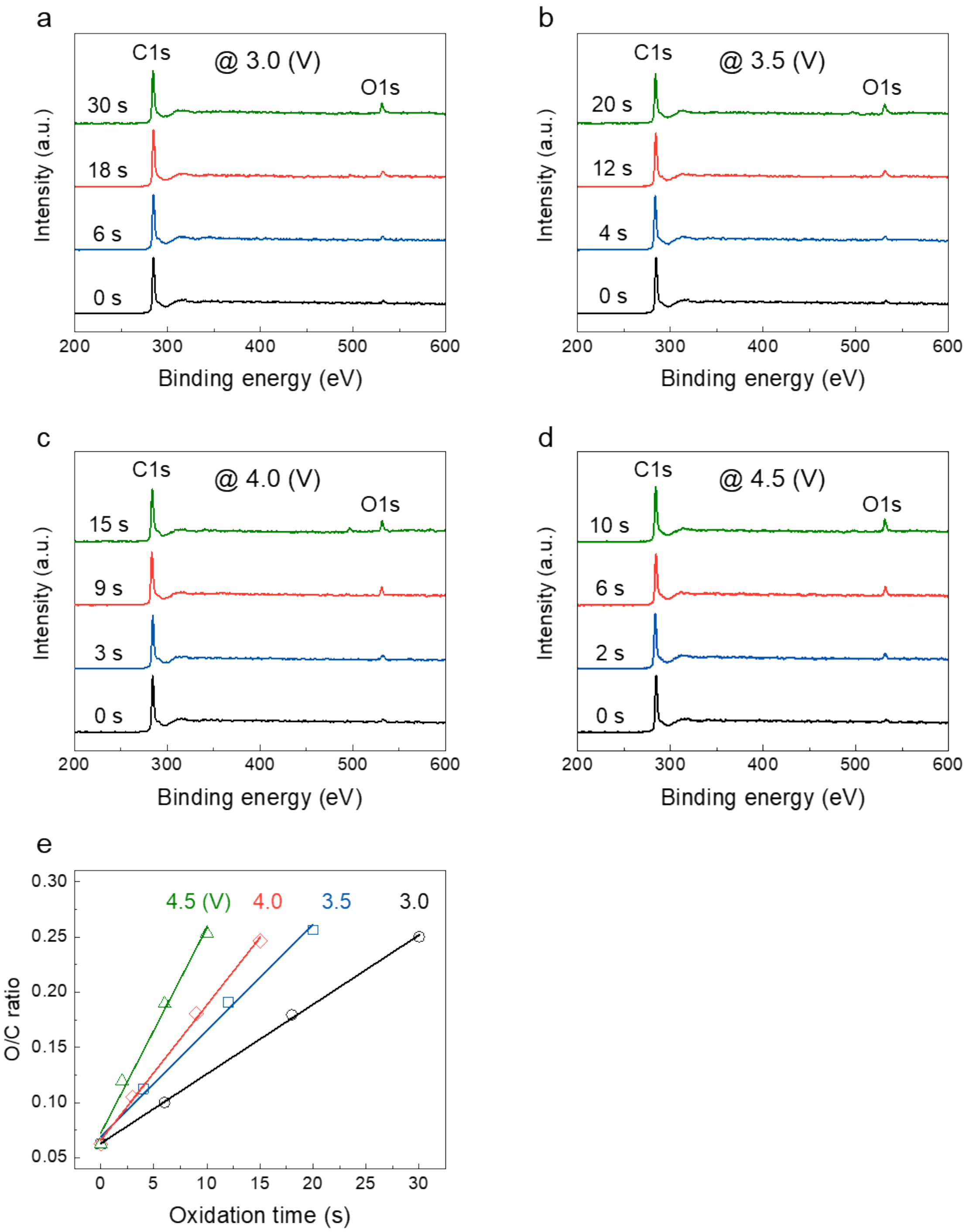

Figure S7. (a-d) XPS spectra of coiled yarns under different electrochemical treatment conditions (oxidation time and applied voltage). (e) The oxygen/carbon (O/C) ratios with various oxidation time for each applied voltage (3.0–4.5V).

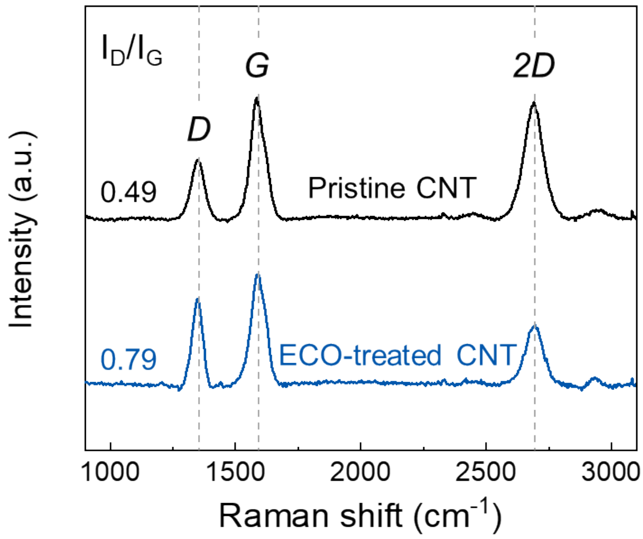

Figure S8. Raman spectra of the pristine and electrochemical oxidation (ECO)-treated coiled carbon nanotube (CNT) yarns.

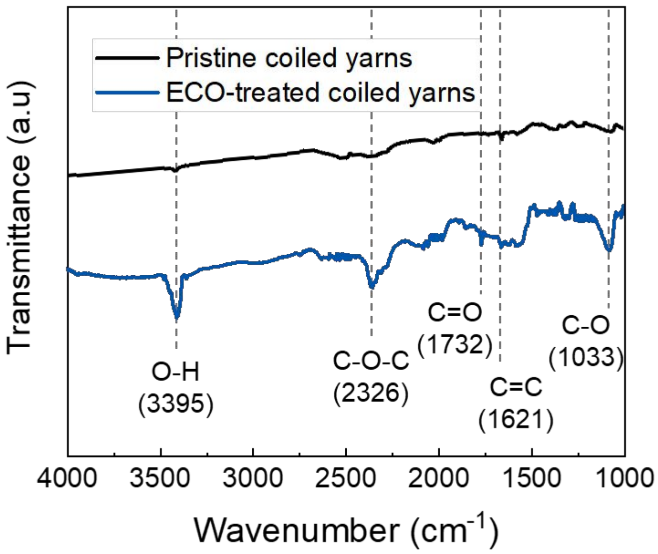

Figure S9. Fourier-transform infrared (FTIR) spectra of pristine coiled yarns and ECO-treated coiled yarns.

Table S1. Characteristics of the functional groups identified in the deconvoluted C 1s spectra.

	C=C		C-C		C-OH		C-O-C		COOH		π-π		O/C Ratio
	Peak (eV)	Area (%)	Peak (eV)	Area (%)	Peak (eV)	Area (%)	Peak (eV)	Area (%)	Peak (eV)	Area (%)	Peak (eV)	Area (%)	
Pristine CNT	234.3	53.9	284.9	29.4	286.5	6.7	287.7	1.5	288.9	3.8	290.6	4.7	0.07
ECO-treated CNT	284.4	45.9	284.9	27.6	286.1	11.6	287.4	6.8	288.8	4.3	290.4	3.6	0.26

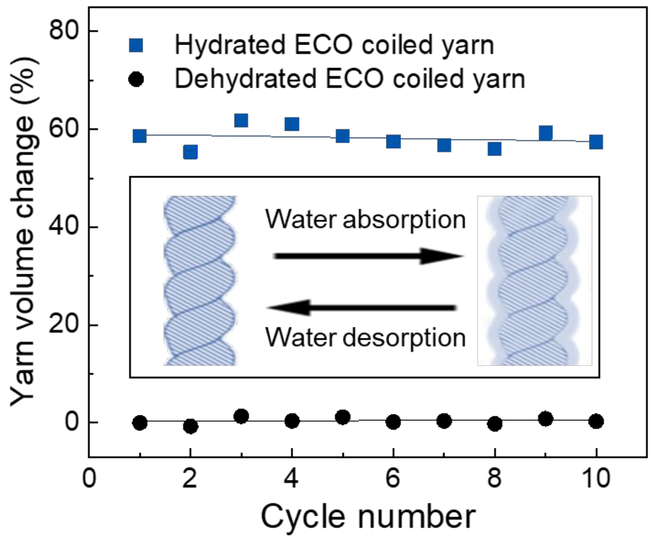

Figure S10. Hydro-volume change (%) of the electrochemical oxidation (ECO)-treated coiled carbon nanotube (CNT) yarns. The yarns were repeatedly hydrated with water droplets (absorption) and dehydrated in an oven (desorption) for the volume change measurements (the inset schematic images show the reversible volume change of an ECO-treated coiled CNT yarn due to water absorption/desorption).

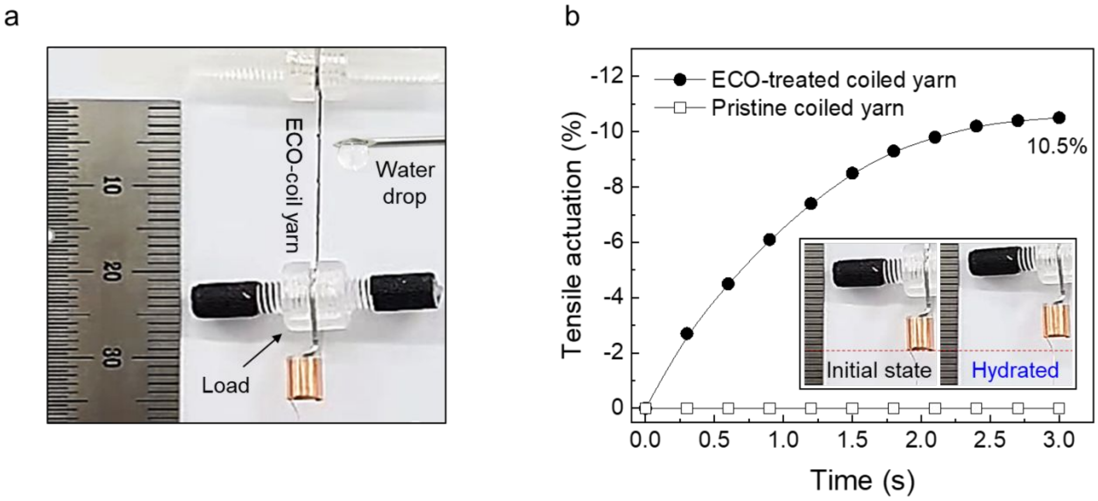

Figure S11. (a) Photograph showing ECO-treated coiled yarn under a 290 kPa load. (b) The time dependence of tensile strokes driven by water absorption. Inset images show weight, hanged by ECO-treated coiled yarn, before and after hydration.

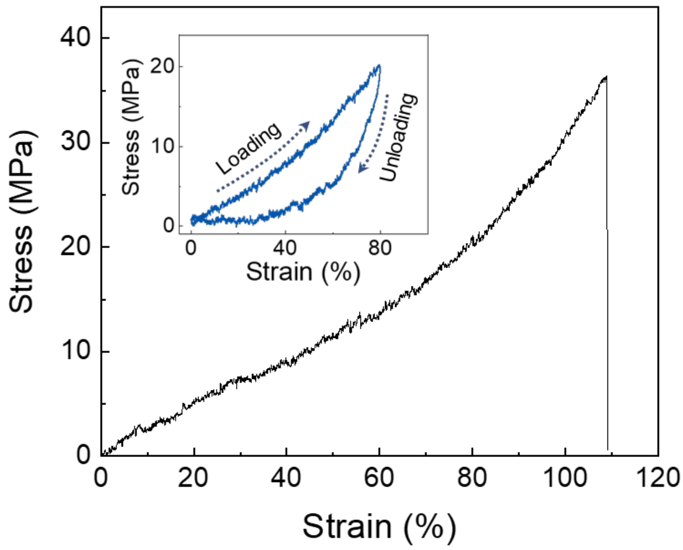

Figure S12. Engineering stress–strain curves for ECO-treated coiled CNT yarns. The inset shows stress loading-unloading curves for 80% strain, showing negligible residual strain and elastic property.

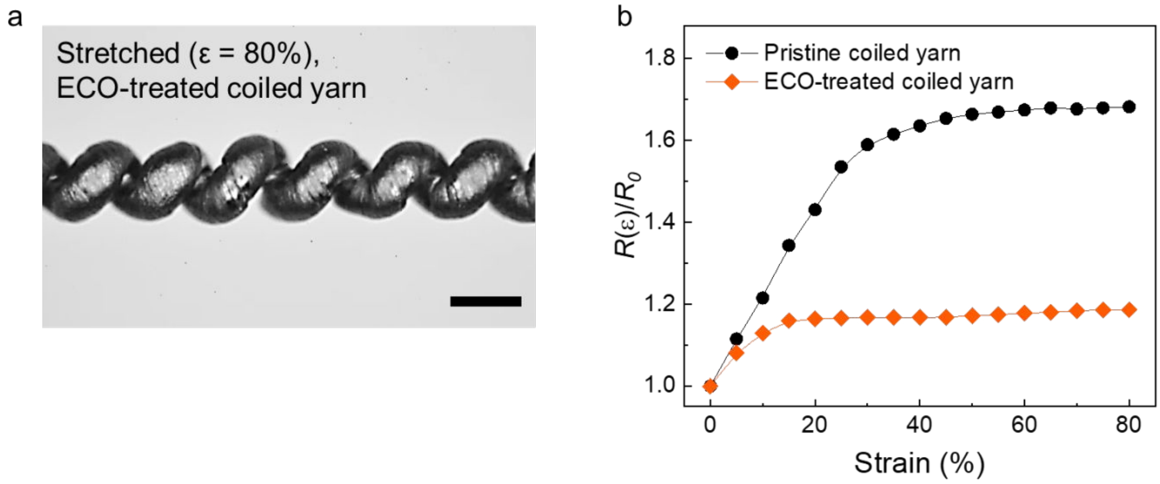

Figure S13. (a) Optical image showing an electrochemical oxidation (ECO)-treated coiled yarn after strain application ε = 80% (scale bar = 150 μm). (b) Resistance changes versus strain for pristine and ECO-treated coiled yarns.

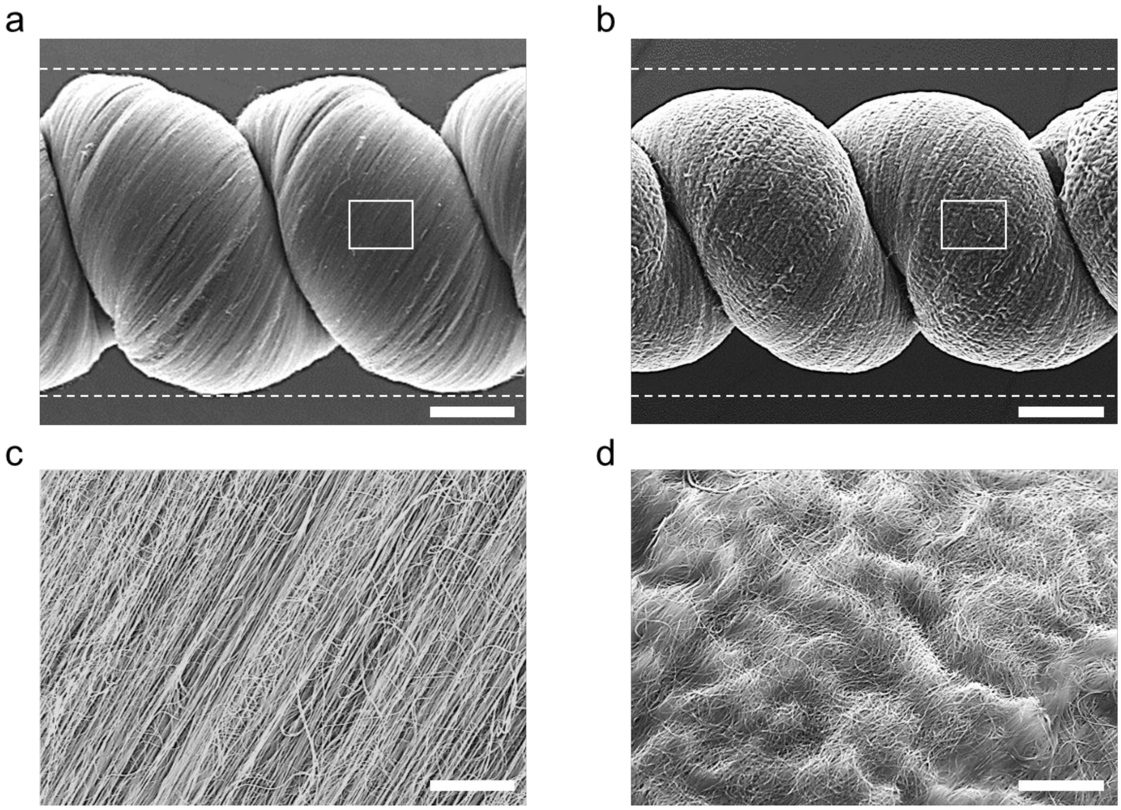

Figure S14. SEM images of (a) a pristine and (b) electrochemical oxidation (ECO)-treated coiled carbon nanotube (CNT) yarn (scale bar = 70 μm) and (c, d) magnification of the center part marked with a square in (a) and (b), respectively (scale bar = 5 μm). The initial diameter for the pristine coiled CNT yarns is denoted by the dash lines in (a) and (b), respectively.

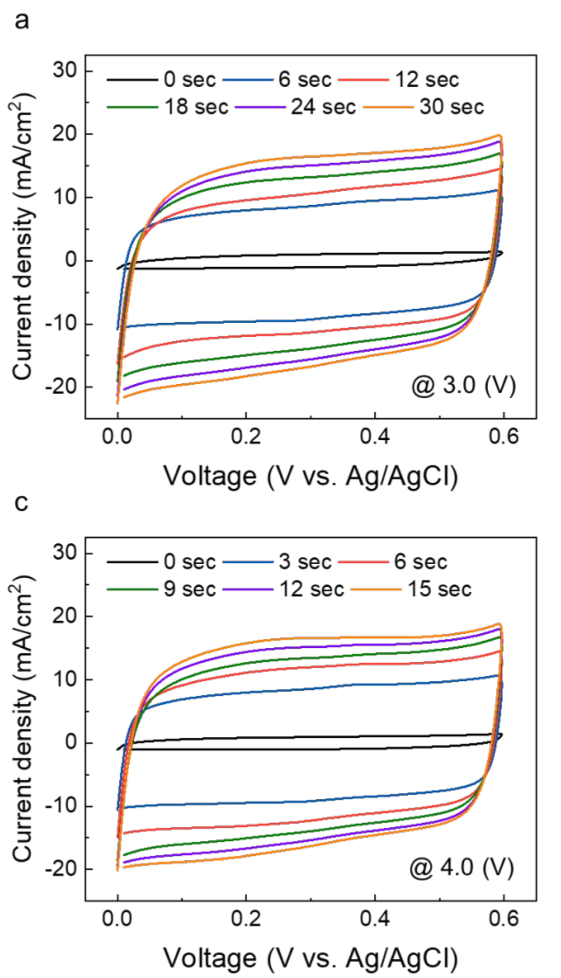

Figure S15. Cyclic voltammetry (CV) curves for ECO-treated coiled CNT yarns with applied voltages of (a) 3.0, (b) 3.5, and (c) 4.0 V, versus Ag/AgCl; the optimized oxidation time (T_{ECO}) for each curve was 30, 20, and 15 s, respectively.

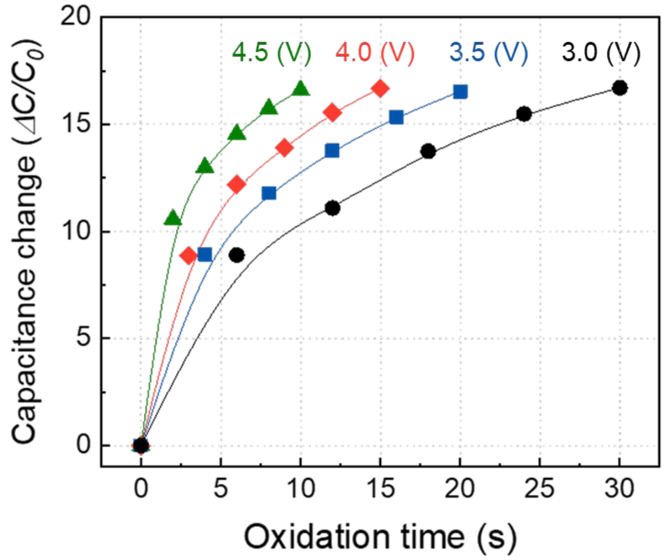

Figure S16. Capacitance enhancement ratio (the increase in the area under the curve (AUC) of the CV plot / ΔC normalized to the initial CV AUC, C_0) versus standard oxidation time (T_{ECO}) for each applied voltage (3.0–4.5V).

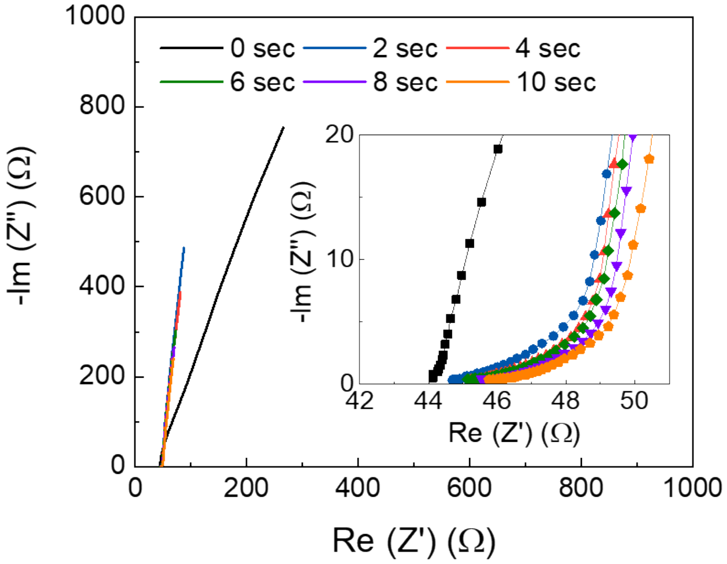

Figure S17. Nyquist curves (0.2 Hz–100 kHz) for pristine (0 s) and electrochemical oxidation (ECO)-treated coiled carbon nanotube (CNT) yarn supercapacitors for various periods (2–10 s) at a given voltage of 4.5 V (vs. Ag/AgCl). The inset shows the high-frequency region.

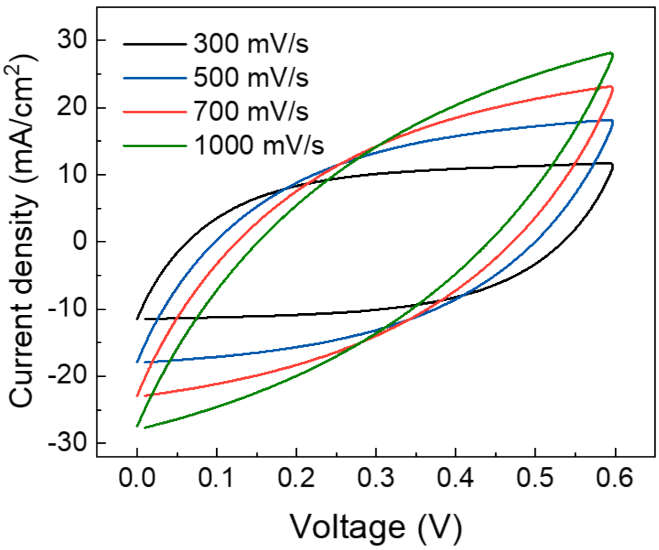

Figure S18. CV AUCs measured at high scan rates (300–1,000 mV/s). The measurements were performed with a two-electrode system consisting of two symmetrical ECO-treated coiled CNT yarn electrodes.

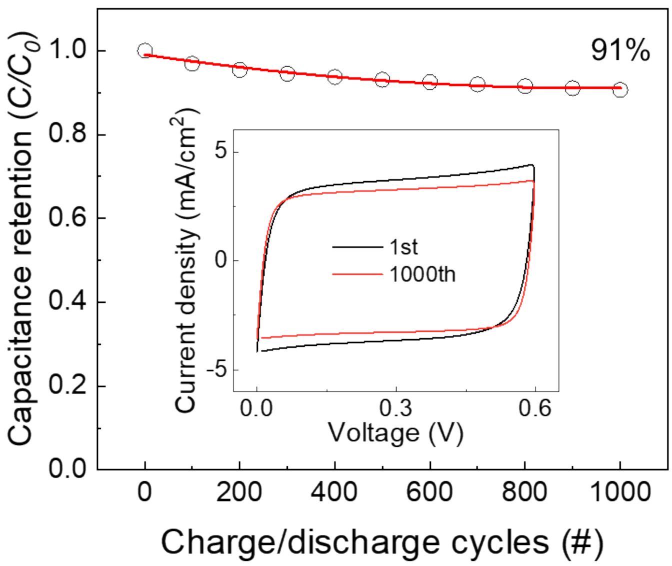

Figure S19. Capacitance retention versus charge/discharge cycles. The inset shows a comparison of the 1st and 1,000th CV curves measured at a 100 mV/s scan rate showing a retention performance of 91%.

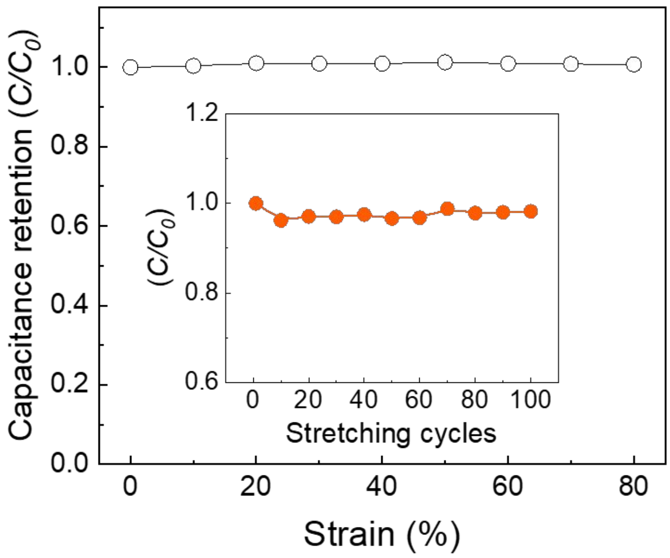

Figure S20. Capacitance retention versus strain for the electrochemical oxidation (ECO)-treated coiled yarn supercapacitor and (inset) its cyclic performance against repeated stretch/release deformations.

Table S2. Comparison of electrical conductivity, specific capacitances, and stretchability of present work and prior-art various supercapacitors.

Electrode Materials (Ref. No.)	Electrical conductivity [S m^{-1}]	Areal capacitance [mF cm^{-2}]	Volumetric capacitance [F cm^{-3}]	Stretchability [%]
ECO-treated coiled yarn (this work)	16064	172.93	30.06	80
Screen-printed graphene film (1)	3600	1	-	-
Nitrogen-doped graphene fiber (2)	30785	1132	-	4.2

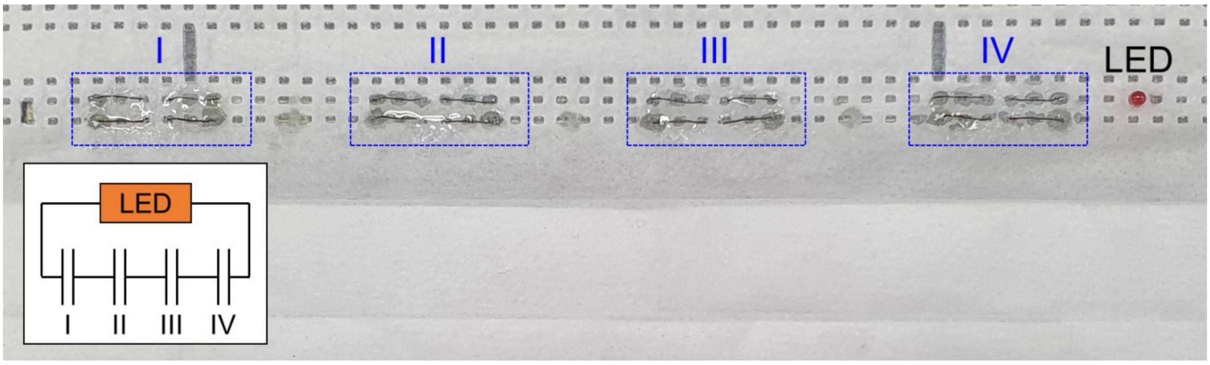

Figure S21. A photograph showing four ECO-treated coiled CNT yarn supercapacitors woven into a commercially available mask with a red LED, each comprising symmetrical two 1.5 cm-long ECO-treated coiled CNT yarns. Inset: Circuit diagram.

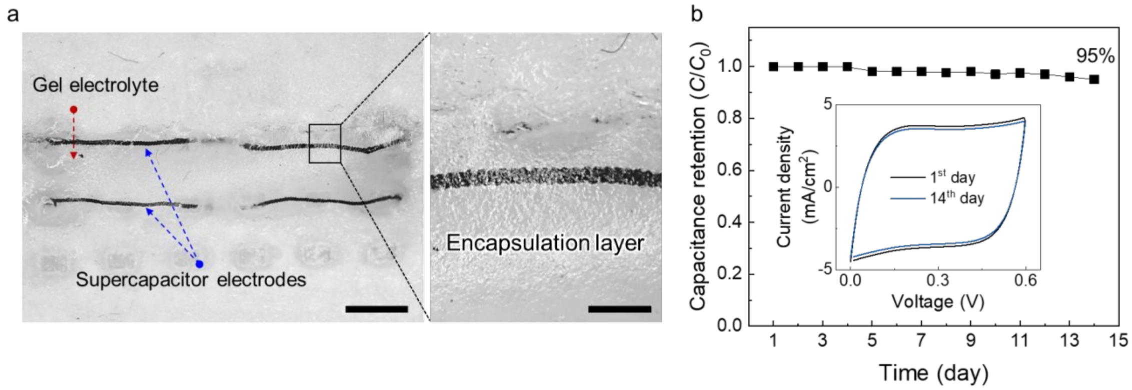

Figure S22. (a) Optical images of encapsulation layer (Ecoflex 00-30)-coated mask supercapacitors and its magnification (scale bars from left to right = 3 and 1 mm). (b) Capacitance retention versus time for the encapsulated mask supercapacitor. The inset shows a comparison of the 1st day and 14th day CV curves (at 100 mV/s) showing a capacitance retention of 95%.

Supplementary References

1. Shi, X.; Pei, S.; Zhou, F.; Ren, W.; Cheng, H.-M.; Wu, Z.-S.; Bao, X. Ultrahigh-Voltage Integrated Micro-Supercapacitors with Designable Shapes and Superior Flexibility. *Energy & Environmental Science* **2019**, 12, 1534-1541.

2. Wu, G.; Tan, P.; Wu, X.; Peng, L.; Cheng, H.; Wang, C.-F.; Chen, W.; Yu, Z.; Chen, S. High-Performance Wearable Micro-Supercapacitors Based on Microfluidic-Directed Nitrogen-Doped Graphene Fiber Electrodes. *Advanced Functional Materials* **2017**, 27, 1702493.