Supporting Information:

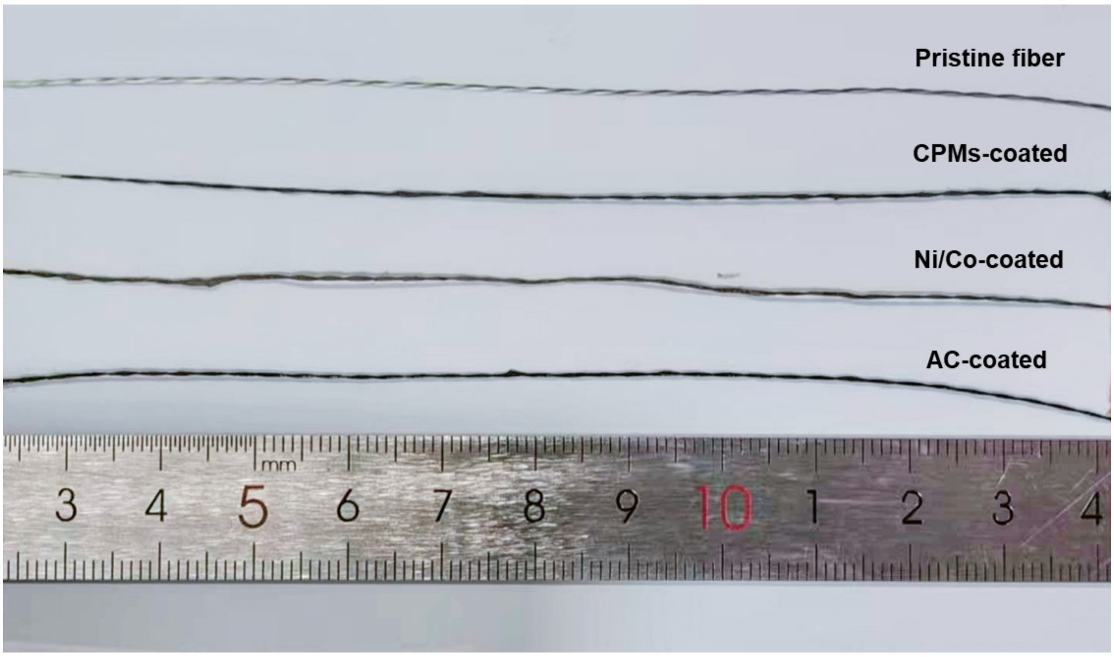


Figure S1. The photo image of four fiber samples with different coatings: pristine SSY yarn, SSY@CPMs, NiCo-SSY@CPMs, and an AC-coated.

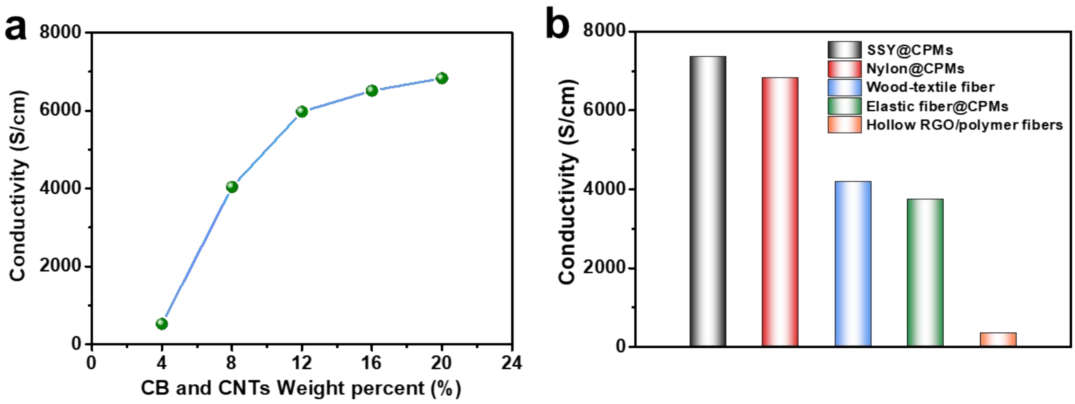


Figure S2. a) The electrical conductivity of CPCs-coated nylon fiber (Nylon@CPMs) as a weight function of CB and CNTs. The weight percent was relative to the matrix and the mass ratio of CB and CNTs was set as 1:1. b) The electrical conductivity of SSY@CPMs, Nylon@CPMs, Elastic fiber@CPMs and reported conducting fibers.

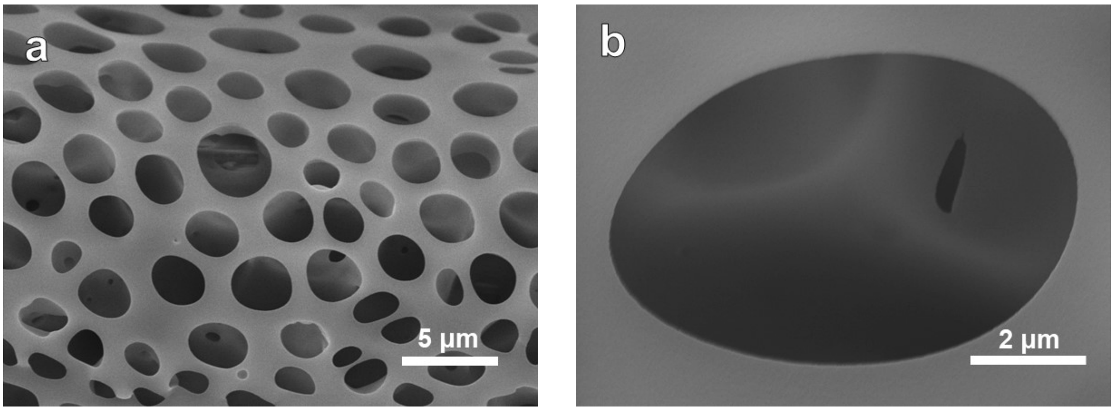


Figure S3. a) Typical SEM images of the pristine SBS-coated fiber. b) The magnified view of (a).

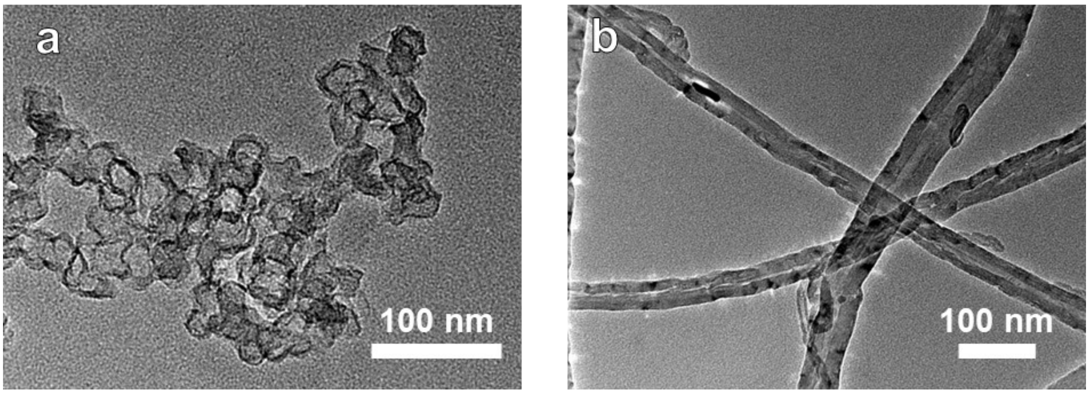


Figure S4. The TEM images of the pristine a) CB and b) CNTs.

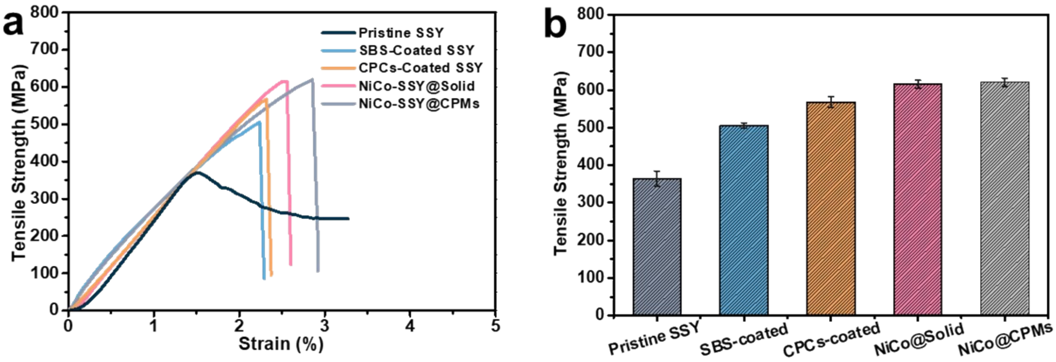


Figure S5. a) The typical strength-strain curves of pristine fibers and fiber electrodes that were fabricated under different conditions. b) The comparison of tensile strength of pristine fibers and fiber electrodes that were fabricated under different conditions.

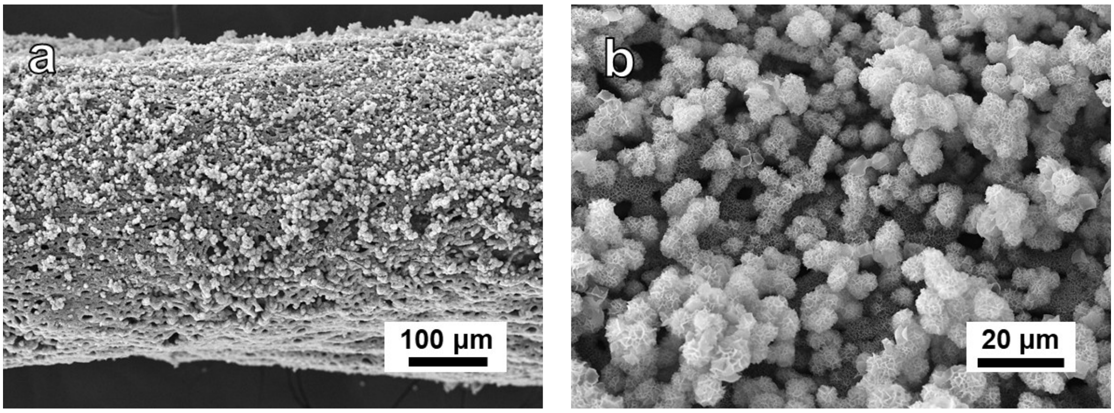
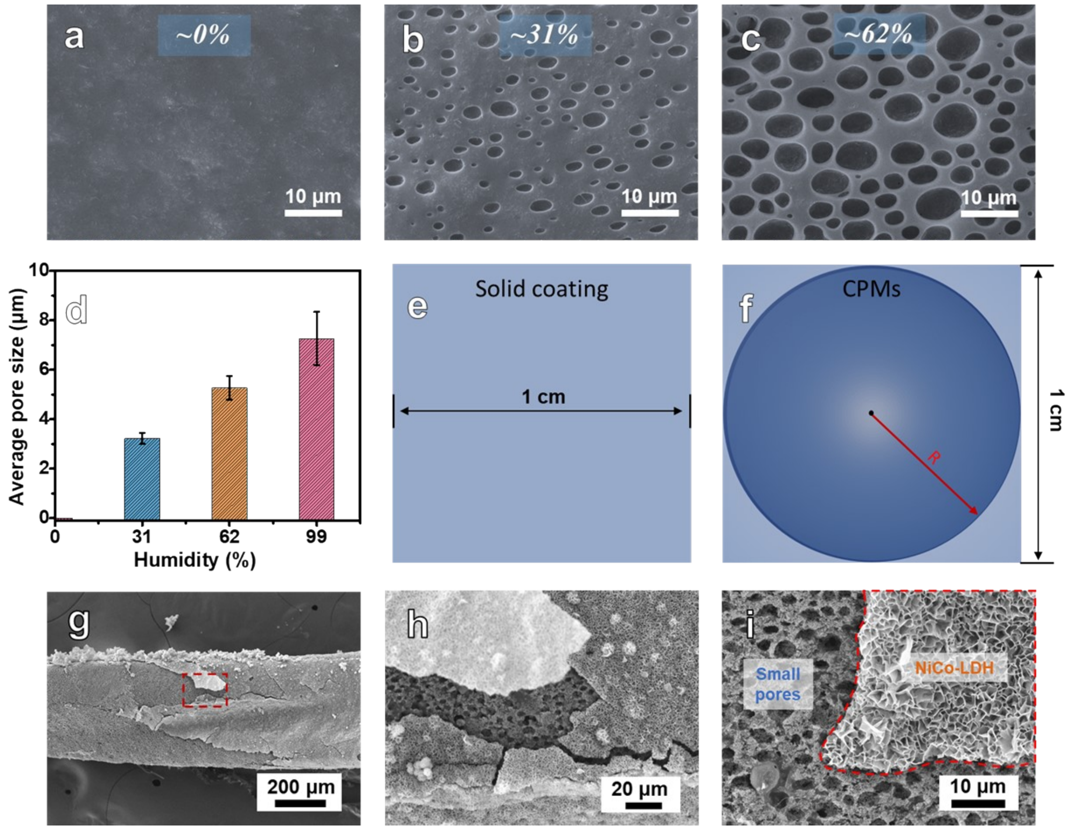


Figure S6. Typical SEM images of the SSY@CPMs@NiCo-LDH under the concentration of 25 mM ($Ni(NO_3)_2 \cdot 6H_2O$) and 50 mM ($Co(NO_3)_2 \cdot 6H_2O$) during hydrothermal process. (b) is the magnified views of (a).



Area calculation of (e) and (f):

Area of e): A_{Solid}= 1 cm × 1 cm= 1 cm^2

Area of f): A_{CPMs}= (Outer region) + (Spherical region)

$= (1\ cm^2 - \pi \times R^2) + (2 \times \pi \times R^2)$

$= 1 + \pi \times R^2\ cm^2$

Figure S7. a-c) Typical SEM images of SSY@Solid and SSY@CPMs prepared at different relative humidity: a) dry environment, b) 31%, c) 62%. The SEM image of SSY@CPMs prepared at ~99% RH can be found at Figure 2a. d) Average pore size of SSY@CPMs as a function of humidity. e, f) Area comparison of the solid coating and CPMs coating under a 1 cm × 1 cm unit cell. g-i) Typical SEM image of NiCo-SSY@ CPMs

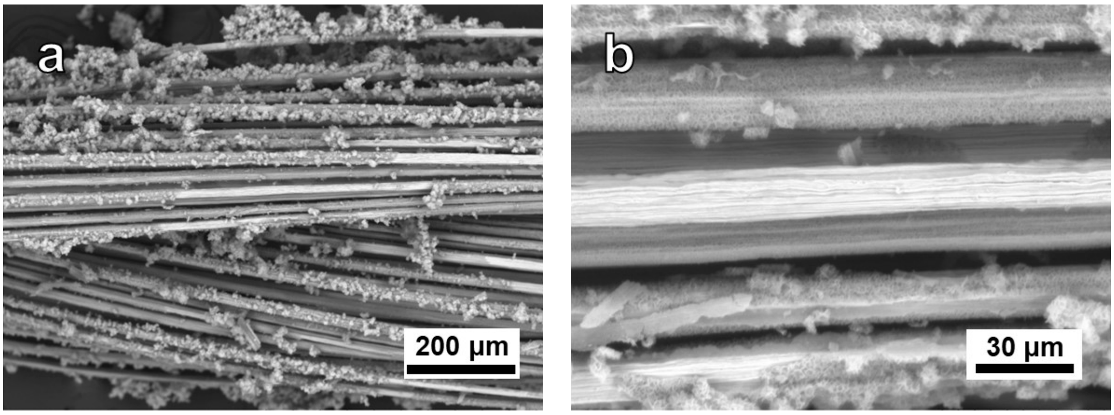


Figure S8. Typical SEM images of NiCo-LDH hydrothermal growth on pristine SSY yarn. (b) is the magnified views of (a). Concentration of $Ni(NO_3)_2 \cdot 6H_2O$ and $Co(NO_3)_2 \cdot 6H_2O$: 5 and 10 mM, respectively.

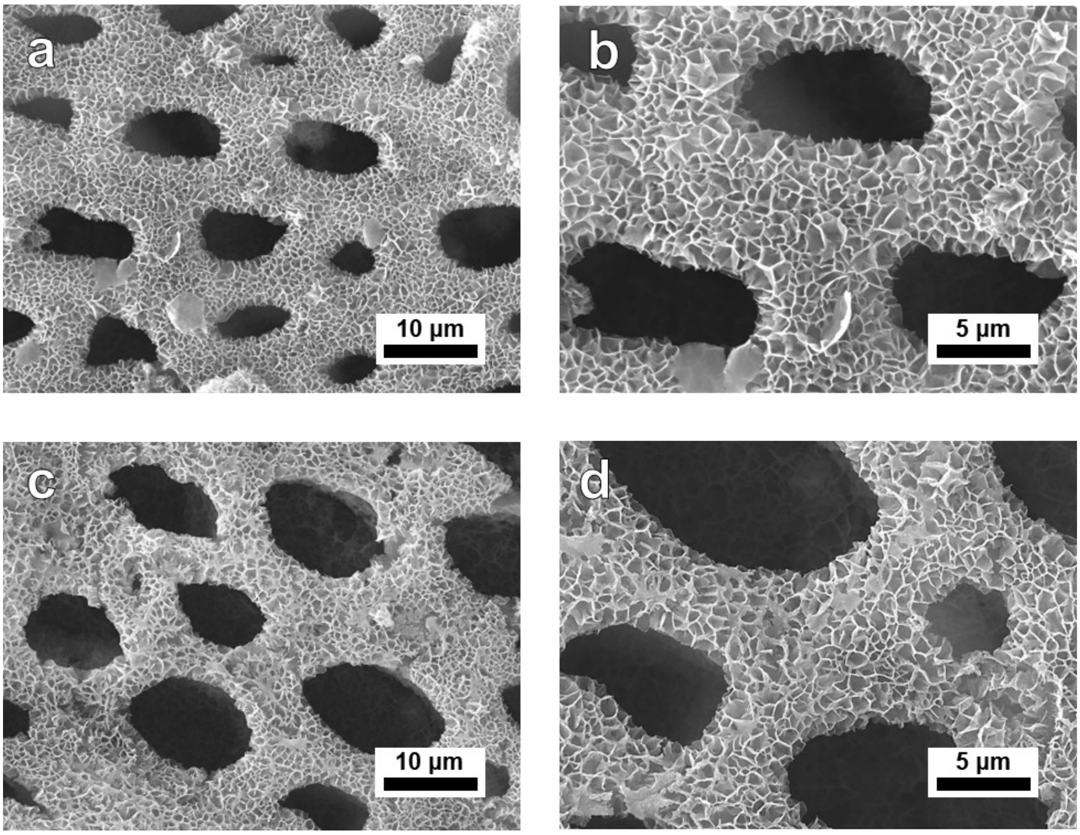


Figure S9. The SEM image of NiCo-SSY@CPMs electrode before and after the testing cycles. (b) and (d) are the magnified view of (a) and (c), correspondingly.

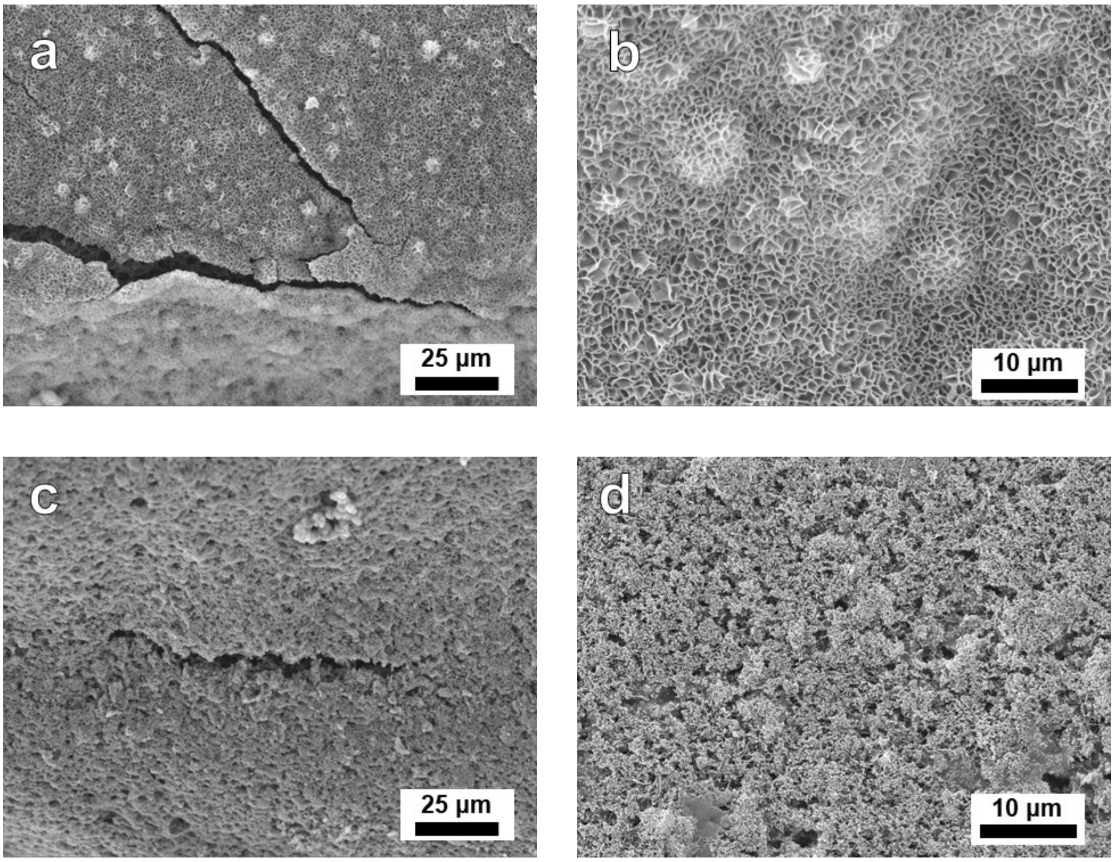


Figure S10. The SEM image of NiCo-SSY@Solid electrode before and after the testing cycles. (b) and (d) are the magnified view of (a) and (c), correspondingly

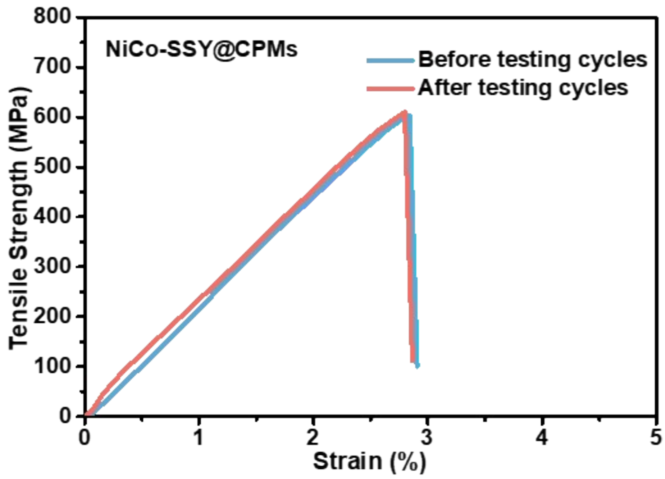


Figure S11. The typical strength-strain curves of NiCo-SSY@CPMs electrode before and after 1500 GCD testing cycles.

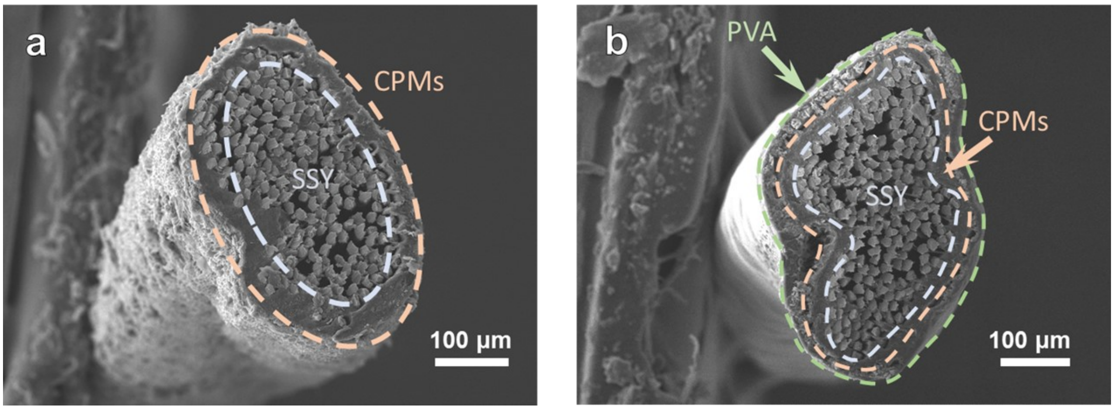


Figure S12. Cross section images of NiCo-SSY@CPMs electrode and the solid-state PVA/KOH-coated NiCo-SSY@CPMs.

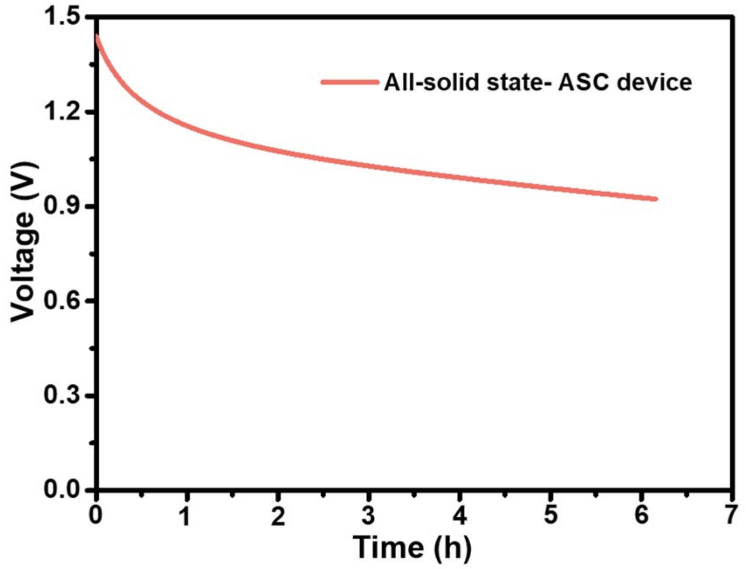


Figure S13. Self-discharging property of the all-solid-state ASC device.

Table S1. Comparison of the energy density and power density of the device with that in the literature.

Electrode materials	Current collector	P_A (mW cm^{-2})	E_A (μWh cm^{-2})	Reference
Hierarchical self-assembly NiCo-LDH	CPCs-coated SSY	7.43	22.52	This work
rGO/PPy	PET	0.03	11	*Electrochim. Acta*[1]
RGO/CNT	Carboxymethyl cellulose yarn	0.02	3.84	*Nat. Commun.*[2]
PET/Au/Ni-MOF	Carbon yarn	0.034	5.41	*Electroanal. Chem.*[3]
rGO	Conducting polymer composite fiber	0.17	6.8	*Adv. Mater.*[4]
PPy/PEDOT:PSS	Cotton yarn wrapped SSY	2	22.7	*Biosens. Bioelectron.*[5]
PEDOT:PSS	Hydrogel	0.40	15.73	*Chem. Eng. J.*[6]
PEDOT:PSS	Cellulose/ polyester cloth	0.4	1.63	*Adv. Mater.*[7]
MXene ink	Polymer gel	0.11	0.32	*Nat. Commun.*[8]

PEDOT:PSS/Ag nanofibers	NOA 63	0.93	0.09	*Chem. Eng. J.*[9]
Cellulose-based ionic hydrogel	Carbon fiber	5.33	0.017	*ACS Appl. Energy Mater.*[10]

P_A: area power density; E_A: area energy density.

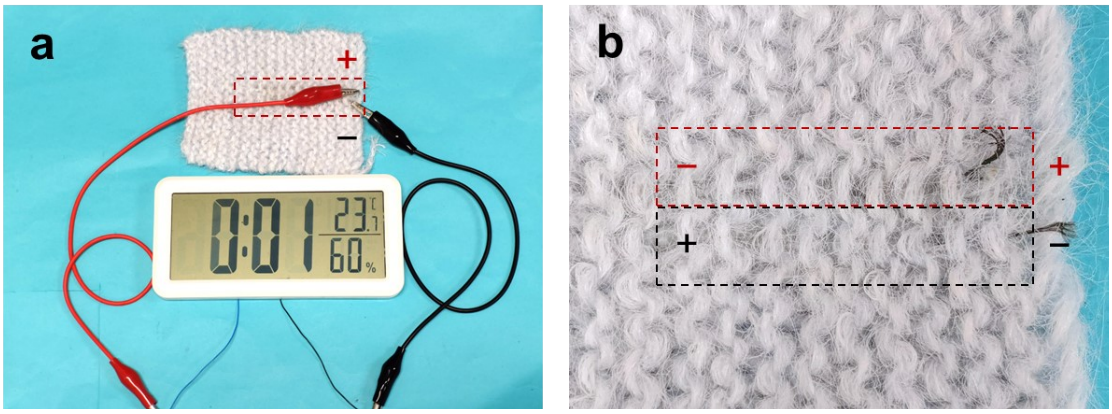


Figure S14. a) The photograph of two serially connected ASC devices woven into a fabric and powered a thermo-hygrometer. b) The closer view of the energy storage fabric.

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