

Supporting information

Partially Sulfurized Nickel-Cobalt LDH on CNT-coated Aramid

Fiber for High-performance Yarn-shaped Supercapacitors

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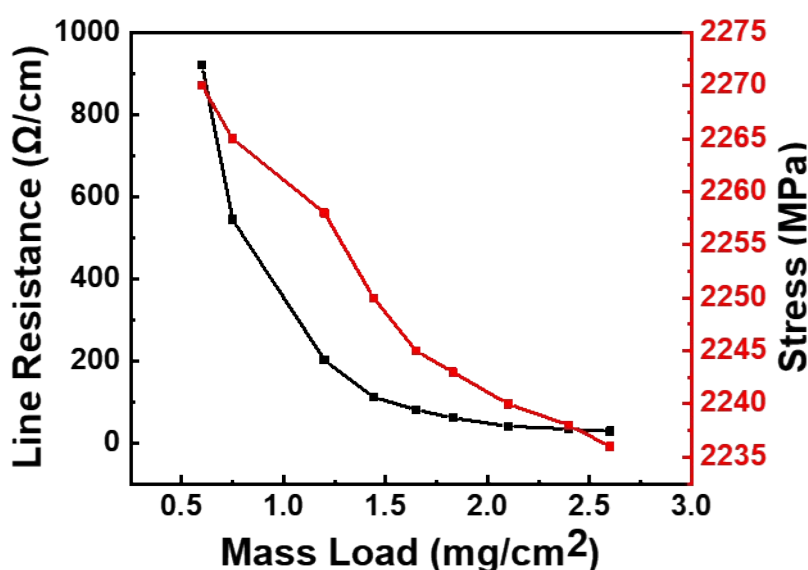


Fig. S1. Relationships between line resistance and strength and mass load of CNTs.

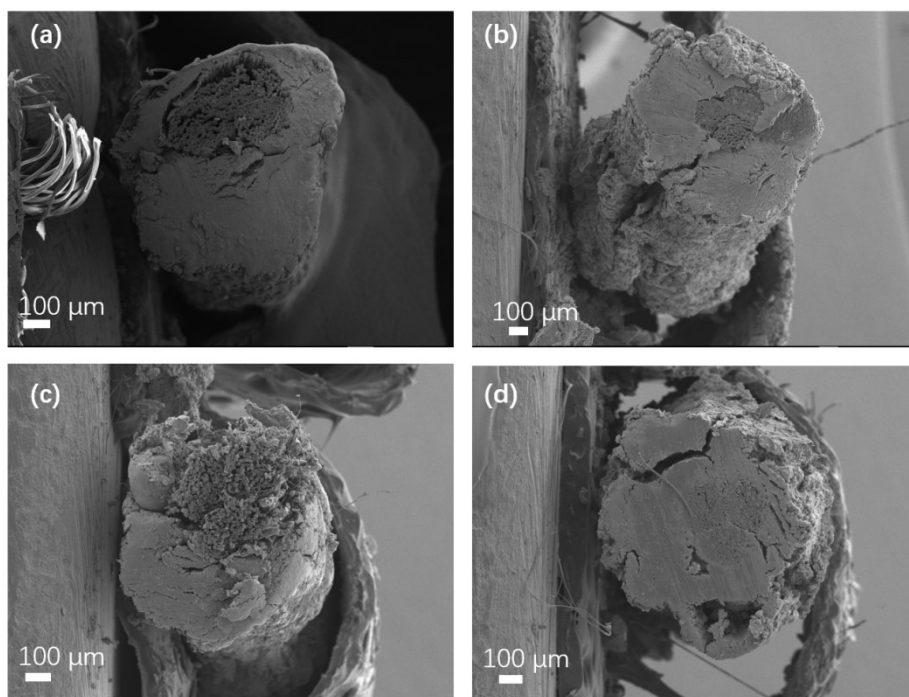


Fig. S2. The cross-sectional SEM images of (a) NiCo-LDH@CAY, (b) NiCo-LDH-S₁@CAY, (c) NiCo-LDH-S_{1.5}@CAY, (d) NiCo-LDH-S₂@CAY.

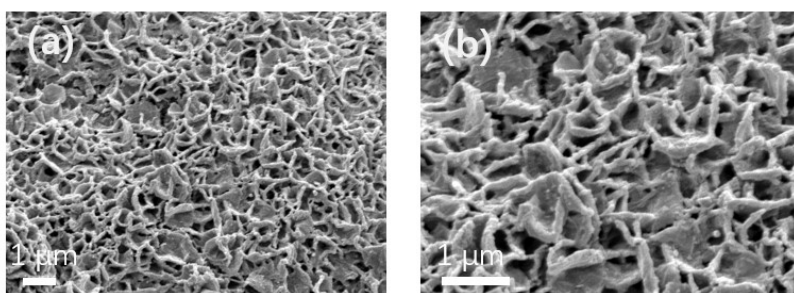


Fig. S3. Scanning electron microscope image of nickel-cobalt hydrotalcite after 5h of sulfurization

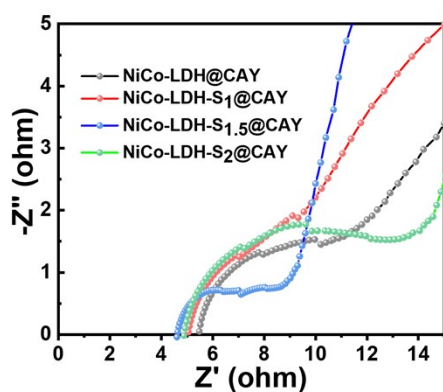


Fig. S4. The Nyquist plots of the four types in the high-frequency region (the magnified zone of Figure 4e).

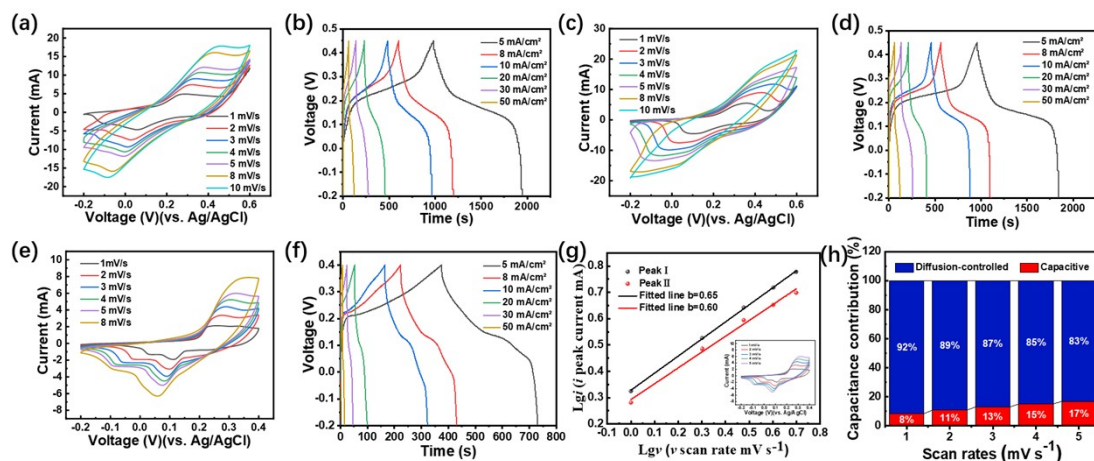


Fig. S5. (a) CV and (b) GCD curves of the NiCo-LDH-S₁@CAY composite electrode material at various scan rates and current densities, respectively. (c) CV and (d) GCD curves of the NiCo-LDH-S₂@CAY composite electrode material at various scan rates and current densities, respectively. (e) CV and (f) GCD curves of the NiCo-LDH@CAY composite electrode material at various scan rates and current densities, respectively. (g) The relationship between the peak current and the scanning rate of NiCo-LDH@CAY. (h) The detailed contributions of diffusion-controlled processes and surface capacity effect in NiCo-LDH@CAY.

Table S1. Equivalent circuit simulation values of the EIS results.

Electrode	Impedance values (Ω)		
	R_s	R_l	R_2
NiCo-LDH@CAY	5.32	4.42	20.19
NiCo-LDH-S ₁ @CAY	5.01	4.24	18.29
NiCo-LDH-S _{1.5} @CAY	4.27	1.12	2.19
NiCo-LDH-S ₂ @CAY	4.86	2.55	2.89

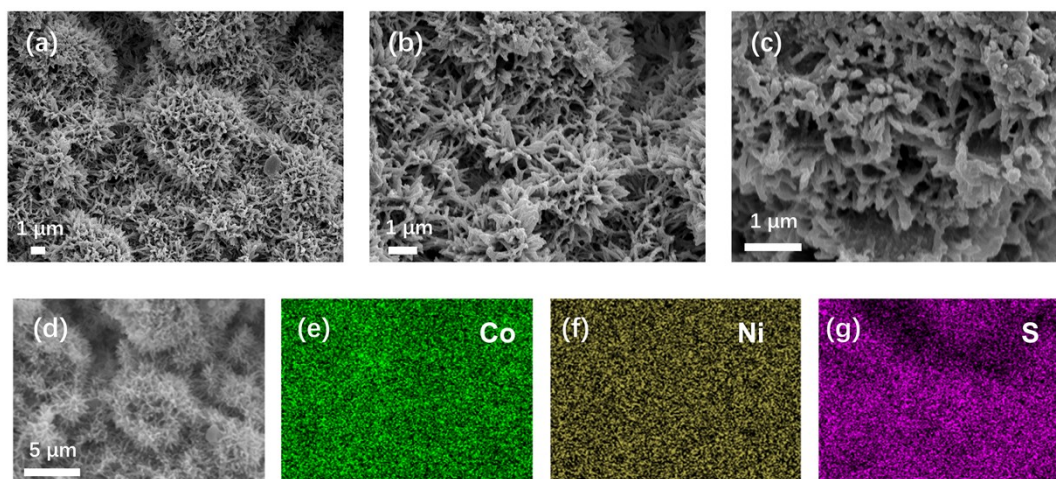


Fig. S6. (a-c) SEM images of post-cycling NiCo-LDH-S_{1.5}@CAY, (d) SEM image for (e-g) EDS mappings of the post-cycling sample.

Table S2. Comparison of the capacitance, energy density, and power density of the device with those in the literature.

Electrode materials	Current collector	Electrolyte	V	<i>i</i>	C _A	P _A	E _A
PPy nanotubes ^[1]	cotton yarn	PVA/H ₂ SO ₄	1.8V	0.42 mA cm ⁻²	74.0 mF cm ⁻²	22.40 μW cm ⁻²	7.5 μWh cm ⁻²
Ni-Co LDH ^[2]	Ni/cotton yarn	PVA/KOH	0.75V	0.04 mA cm ⁻²	221 mF cm ⁻²	43.99 μW cm ⁻²	9.3 μWh cm ⁻²
Co ₃ O ₄ ^[3]	CNT yarn	PVA/H ₂ SO ₄	0.8V	0.053 mA cm ⁻²	52.6 mF cm ⁻²	10 μW cm ⁻²	1.10 μWh cm ⁻²
NiO ^[3]	CNT yarn	PVA/H ₂ SO ₄	0.8V	0.053 mA cm ⁻²	15.2 mF cm ⁻²	5 μW cm ⁻²	0.2 μWh cm ⁻²
NiCo-OH/ZnO-NR ^[4]	CNT-yarn	PVA/LiOH	/	/	/	117 μW cm ⁻²	1.38 μWh cm ⁻²
MoS ₂ /MnS/GR ^[5]	PI fibers	PVA/Na ₂ SO ₄	1.0V	0.05 mA cm ⁻²	50.2 mF cm ⁻²	49.9 μW cm ⁻²	7.0 μWh cm ⁻²
NiCo ₂ S ₄ @Ni-Co LDH ^[6]	Au/cotton yarn	PVA/KOH	0.8V	0.4 mA cm ⁻²	37.2 mF cm ⁻²	163.2 μW cm ⁻²	3.6 μWh cm ⁻²
CuO PPy ^[7]	Cu wire Carbon fiber	PVA/KOH	1.3V	/	39.67 mF cm ⁻²	/	9.31 μWh·cm ⁻²
Co ₃ O ₄ @MnO ₂ Graphene ^[8]	Ni wire Carbon fiber	PVA/KOH	1.5V	0.1 mA cm ⁻²	13.9 mF cm ⁻²	75 μW cm ⁻²	4.34 μWh·cm ⁻²
MnO ₂ ^[9]	AgNP/AgNW/ Au/ PVDF fibers	PVA/H ₃ PO ₄	0.85V	0.1 mA cm ⁻²	24.5 mF cm ⁻²	10.6 μW cm ⁻²	0.63 μWh·cm ⁻²
RGO ^[10]	cotton yarn	PVA/H ₂ SO ₄	1.0V	0.06 mA cm ⁻²	9.54 mF cm ⁻²	31.84 μW cm ⁻²	1.32 μWh·cm ⁻²
This work	CNT/aramid yarn	PVA/KOH	1.6V	0.5 mA cm ⁻²	526.64 mF cm ⁻²	401.49 μW cm ⁻²	151.67 μWh cm ⁻²

References

- [1] C. Wei, Q. Xu, Z. Chen, W. Rao, L. Fan, Y. Yuan, Z. Bai, J. Xu, An all-solid-state yarn supercapacitor using cotton yarn electrodes coated with polypyrrole nanotubes, Carbohydr. Polym. 169 (2017) 50-57.

- [2] H.-T. Wang, C. Jin, Y.-N. Liu, X.-H. Kang, S.-W. Bian, Q. Zhu, Cotton yarns modified with three-dimensional metallic Ni conductive network and pseudocapacitive Co-Ni layered double hydroxide nanosheet array as electrode materials for flexible yarn supercapacitors, *Electrochim. Acta* 283 (2018) 1789-1797.
- [3] F. Su, X. Lv, M. Miao, High - performance two - ply yarn supercapacitors based on carbon nanotube yarns dotted with Co₃O₄ and NiO nanoparticles, *Small* 11(7) (2015) 854-861.
- [4] T.S. Le, T.K. Truong, J. Bae, D. Suh, Synergetic design of enlarged surface area and pseudo-capacitance for fiber-shaped supercapacitor yarn, *Nano Energy* 67 (2020) 104198.
- [5] C. Zhou, M. Hong, Y. Yang, C. Yang, N. Hu, L. Zhang, Z. Yang, Y. Zhang, Laser-induced bi-metal sulfide/graphene nanoribbon hybrid frameworks for high-performance all-in-one fiber supercapacitors, *J. Power Sources* 438 (2019) 227044.
- [6] Y.-F. Wang, H.-T. Wang, S.-Y. Yang, Y. Yue, S.-W. Bian, Hierarchical NiCo₂S₄@ nickel-cobalt layered double hydroxide nanotube arrays on metallic cotton yarns for flexible supercapacitors, *ACS Appl. Mater. Interfaces* 11(33) (2019) 30384-30390.
- [7] R. Zhang, C. Chen, H. Yu, S. Cai, Y. Xu, Y. Yang, H. Chang, All-solid-state wire-shaped asymmetric supercapacitor based on binder-free CuO nanowires on copper wire and PPy on carbon fiber electrodes, *J. Electroanal. Chem.* 893 (2021) 115323.
- [8] X. Niu, G. Zhu, Z. Yin, Z. Dai, X. Hou, J. Shao, W. Huang, Y. Zhang, X. Dong, Fiber-based all-solid-state asymmetric supercapacitors based on Co₃O₄@ MnO₂ core/shell nanowire arrays, *J. Mater. Chem. A* 5(44) (2017) 22939-22944.
- [9] P.T. Nguyen, J. Jang, Y. Lee, S.T. Choi, J.B. In, Laser-assisted fabrication of flexible monofilament fiber supercapacitors, *J. Mater. Chem. A* 9(8) (2021) 4841-4850.
- [10] L. Li, D. Du, C. He, L. Yu, W. Tang, S. Hu, X. Wang, Z. Fu, L. Xia, W. Xu, Cotton yarns decorated with hydrothermally reduced graphene oxide for flexible supercapacitors, *Ind Crop Prod* 205 (2023) 117547.