

## Supporting Information

### **Bio-inspired Nano-Engineering of Ultrahigh Loading 3D Hierarchical Ni@NiCo<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>S<sub>2</sub> electrode for High Energy Density Supercapacitors**

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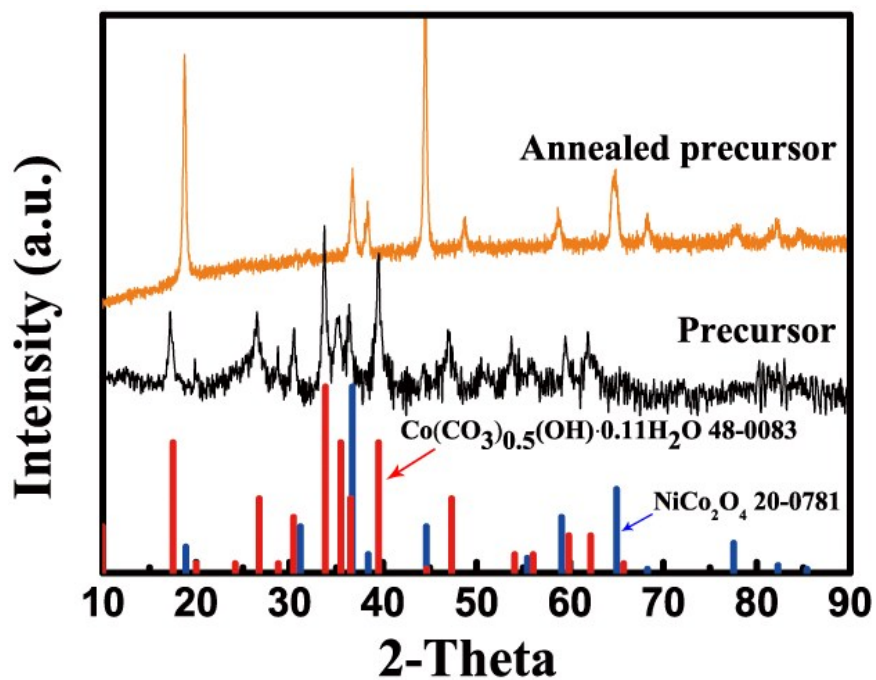


Figure S1. The XRD patterns of precipitate and annealed precipitate.

The precipitate takes the crystal structure of  $\text{Co}(\text{CO}_3)_{0.5}(\text{OH}) \cdot 0.11\text{H}_2\text{O}$  (JCPDS 48-0083) due to the similarity between  $\text{Ni}^{2+}$  and  $\text{Co}^{2+}$ . And the annealed precipitate could be assigned to  $\text{NiCo}_2\text{O}_4$  (JCPDS 20-0781), which reflects that the precipitate is  $\text{Ni}_{1/3}\text{Co}_{2/3}(\text{CO}_3)_{0.5}(\text{OH}) \cdot 0.11\text{H}_2\text{O}$ .

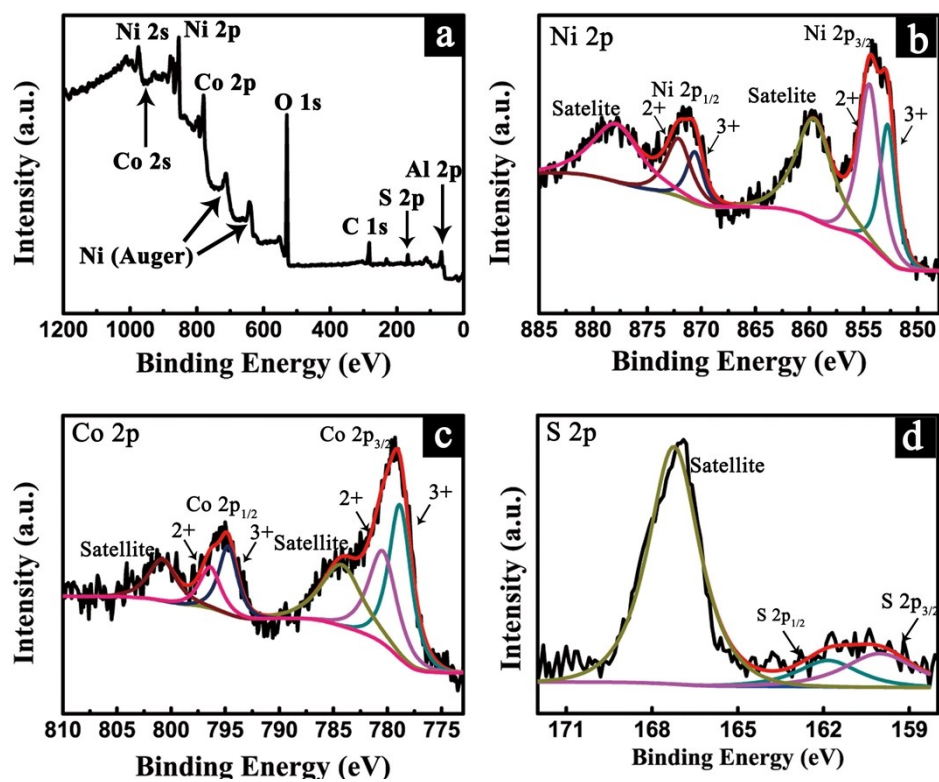
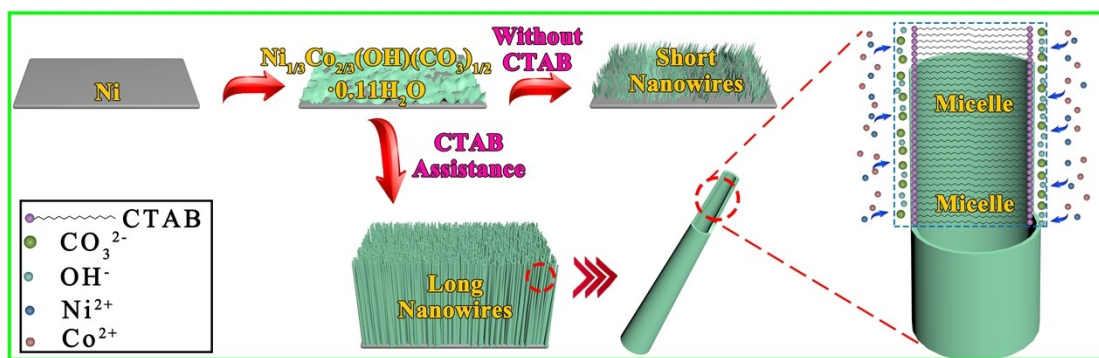
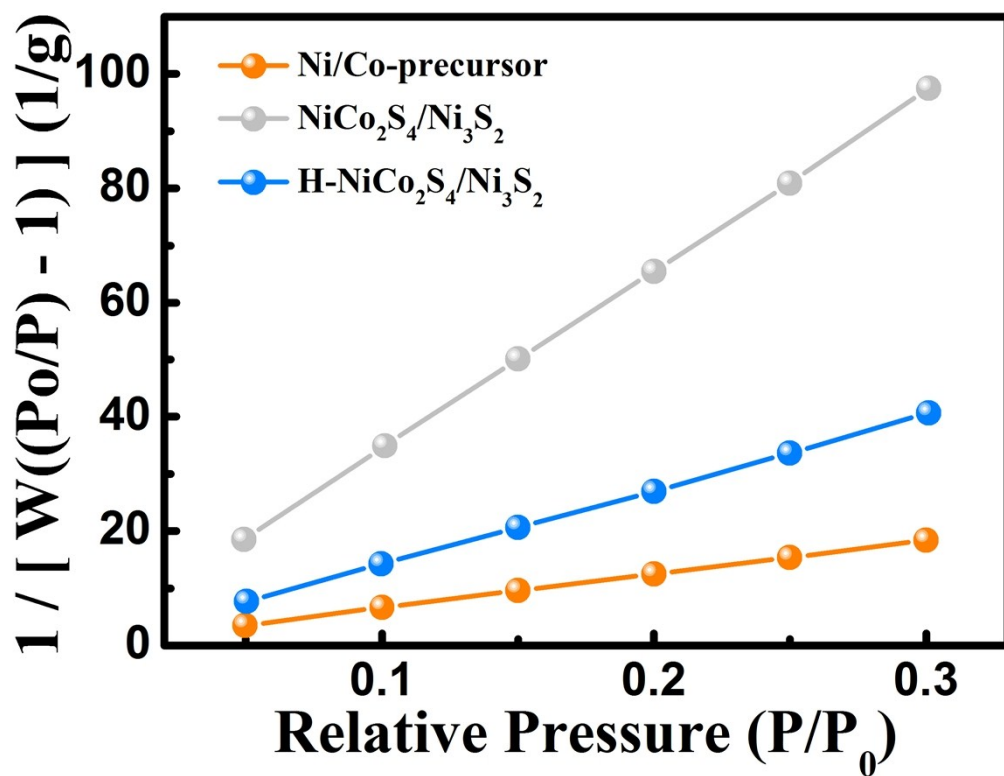


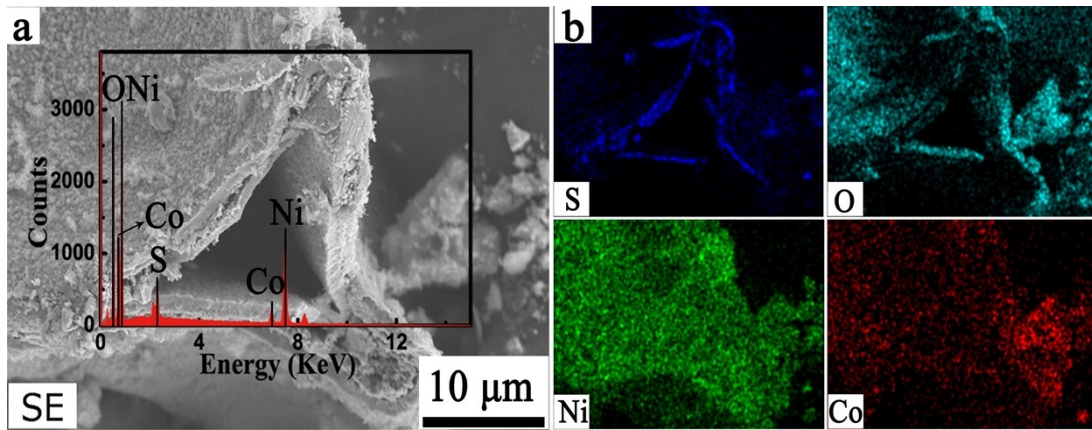
Figure S2. XPS spectra of H-Ni@NiCo<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>S<sub>2</sub>



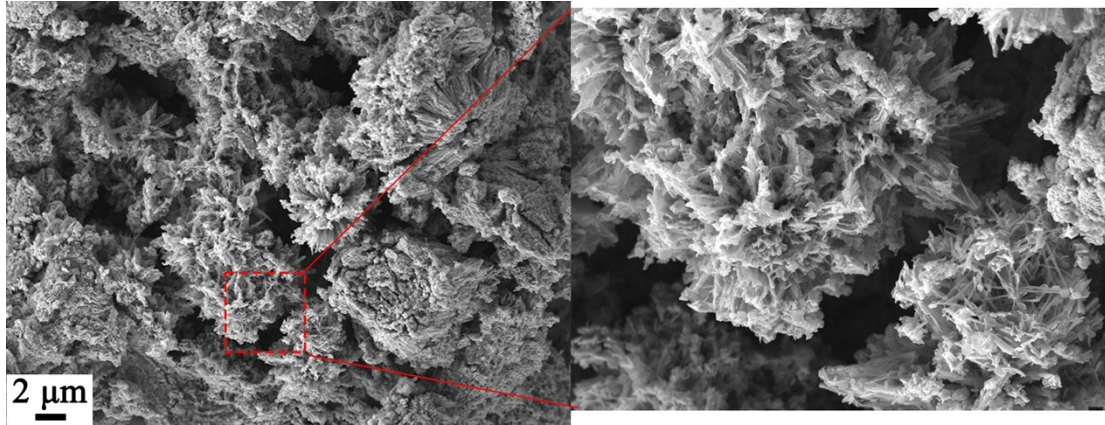
**Figure S3.** The schematically formation process of the nanowire arrays on Ni foam with/without CTAB.



**Figure S4.** The BET plots of the scraping Ni/Co-precursor, NiCo<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>S<sub>2</sub> and H-NiCo<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>S<sub>2</sub> powders.



**Figure S5.** The SEM (a) and mapping images (b) of H-Ni@NiCo<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>S<sub>2</sub> (the inset of SEM image shows the EDX spectrum).



**Figure S6.** After 5000 cycles, SEM images of the H-Ni@NiCo<sub>2</sub>S<sub>4</sub>/Ni<sub>3</sub>S<sub>2</sub> electrodes.

**Table S1.** Energy densities and corresponding power densities of devices assembled by Ni, Co-base electrodes.

Devices	Energy density (Wh m <sup>-2</sup> )	Power density (W m <sup>-2</sup> )	Mass Loading (mg cm <sup>-2</sup> )	Reference
Ni-Co-S/GF	0.79	8.25	1.00	[1]
Ni <sub>3</sub> S <sub>2</sub> @Ni	2.18	18.36	1.08	[2]
Ni <sub>3</sub> S <sub>2</sub> @CdS core-shell	1.91	14.93	1.50	[3]
CNFs/NiCo <sub>2</sub> S <sub>4</sub> @PPy	0.71	11.50	1.92	[4]
Vanadium-modified NiCo <sub>2</sub> S <sub>4</sub> hybrid supercapacitors	0.98	18.97	2.18	[5]
PPy@NiCo <sub>2</sub> S <sub>4</sub> core-shell	1.52	4.80	3.00	[6]
FeCo <sub>2</sub> S <sub>4</sub> -tube	2.28	22.65	3.00	[7]
ZIF-NiCo <sub>2</sub> S <sub>4</sub> //AC	1.34	23.84	3.00	[8]
NiCo <sub>2</sub> S <sub>4</sub> @Ni(OH) <sub>2</sub> @PPy	1.11	3.86	3.21	[9]
C-NiCo <sub>2</sub> S <sub>4</sub>	1.15	24.00	3.0±0.5	[10]
NiCo <sub>2</sub> S <sub>4</sub> /N,S-MGA	3.66	24.00	3.0-5.0	[11]
NiCo <sub>2</sub> S <sub>4</sub> @PPy-50/NF	2.38	8.26	6.87	[12]
<b>NiCo<sub>2</sub>S<sub>4</sub>@ NiCo<sub>2</sub>O<sub>4</sub> //AC ASC</b>	<b>4.69</b>	<b>10.33</b>	<b>10.33</b>	<b>This work</b>

\* AC: Activated Carbon

ASC : Asymmetric Supercapacitors

## Reference

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