

## Supporting Information

### Electric field-induced ball-cactus like $\text{CuCo}_2\text{S}_x(\text{OH})_y$ nano-heterostructure towards high-performance supercapacitors

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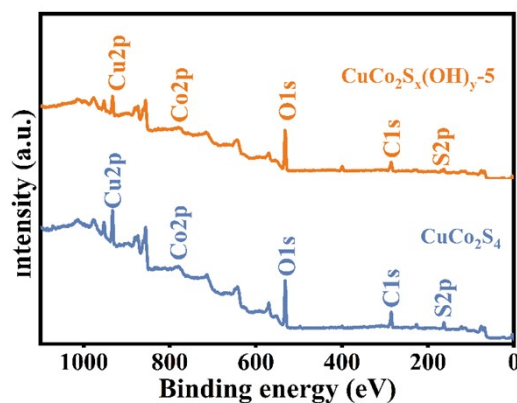


Fig. S1 XPS survey spectrum of the as-synthesized  $\text{CuCo}_2\text{S}_4$  and  $\text{CuCo}_2\text{S}_x(\text{OH})_{y-5}$

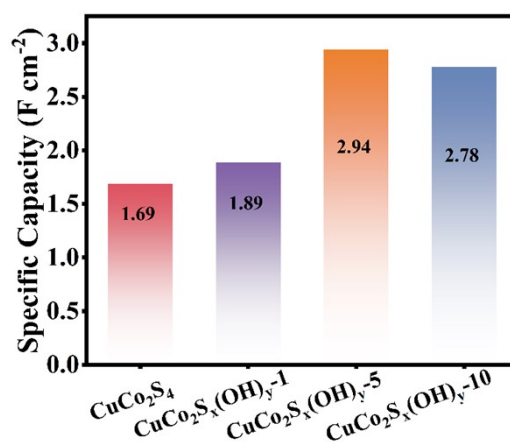


Fig. S2 The comparison of specific capacitance of different electrode materials at the current density of  $5 \text{ mA cm}^{-2}$ .

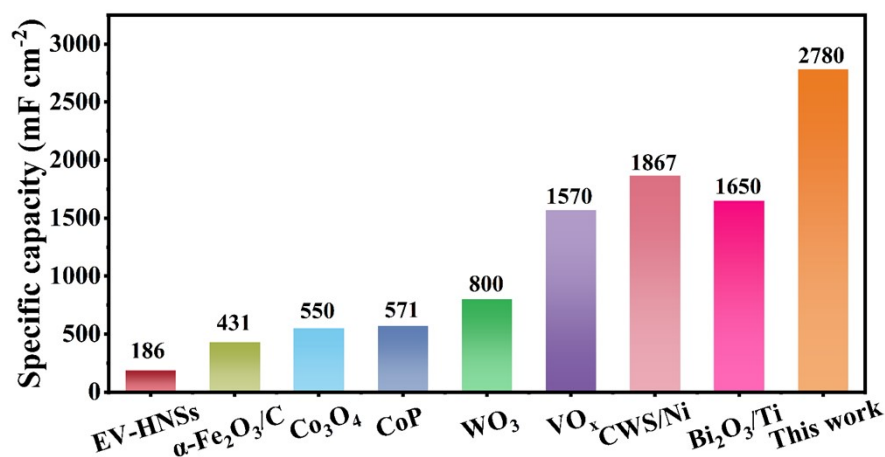


Fig. S3 Comparison of area specific capacitance (mF cm<sup>-2</sup>) with other reported negative electrode materials.

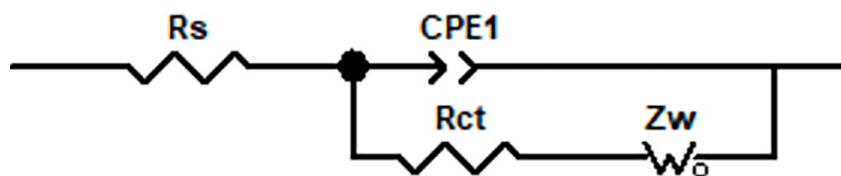


Fig. S4 The equivalent circuit model fitting Nyquist diagram.

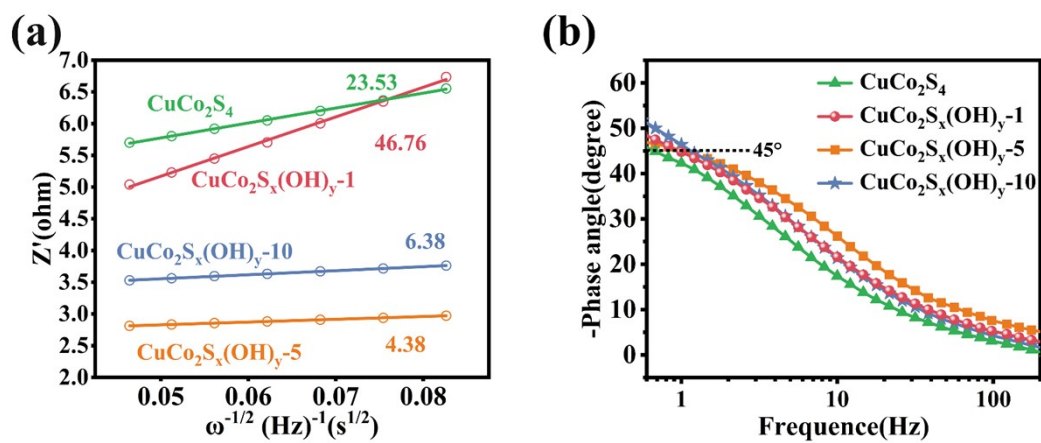
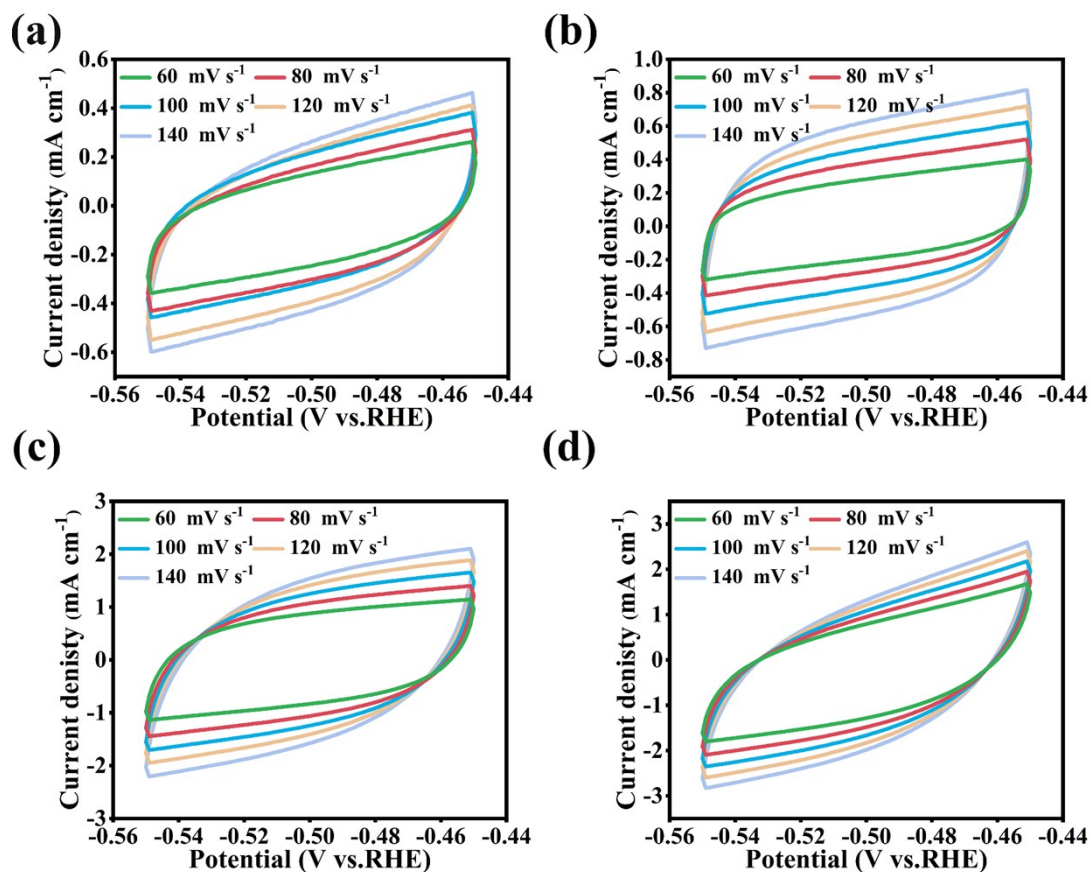
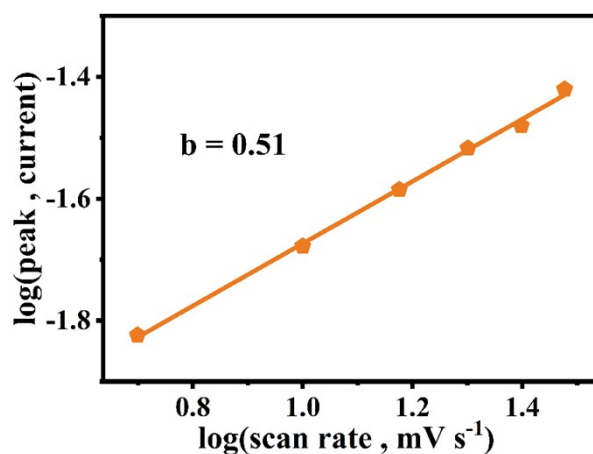


Fig. S5 (a) The linear relation between real part ( $Z'$ ) and the square root of frequency ( $\omega^{-1/2}$ ) at low frequency region; (b) The bode plots of phase angle versus frequency.



**Fig. S6** CV curves of (a)  $\text{CuCo}_2\text{S}_4$ , (b)  $\text{CuCo}_2\text{S}_x\text{OH}_y-1$ , (c)  $\text{CuCo}_2\text{S}_x\text{OH}_y-5$  and (d)  $\text{CuCo}_2\text{S}_x\text{OH}_y-10$  at different scan rates (from 60 to  $140 \text{ mV s}^{-1}$ ) with the potential range of  $-0.55 \text{ V}$  to  $-0.45 \text{ V}$ .



**Fig. S7** Plot of the anodic peak current density against the scan rate for  $\text{CuCo}_2\text{S}_x\text{OH}_y-5$  electrode.

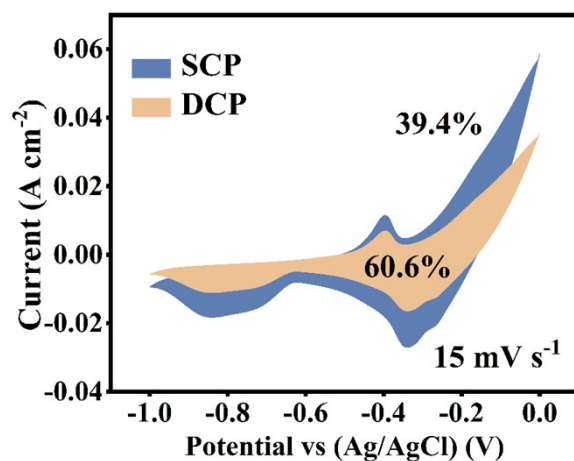


Fig. S8 The surface contribution and diffusive contribution of the  $\text{CuCo}_2\text{S}_x\text{OH}_y\text{-5}$  electrode at the scan rate of  $15 \text{ mV s}^{-1}$ .

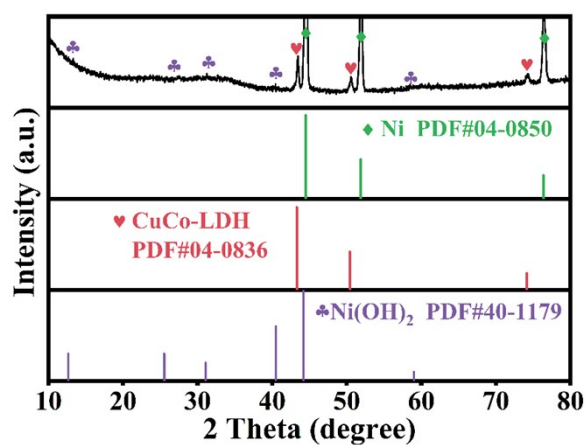


Fig. S9 XRD patterns of the as-synthesized  $\text{CuCo-LDH@Ni(OH)}_2$ .

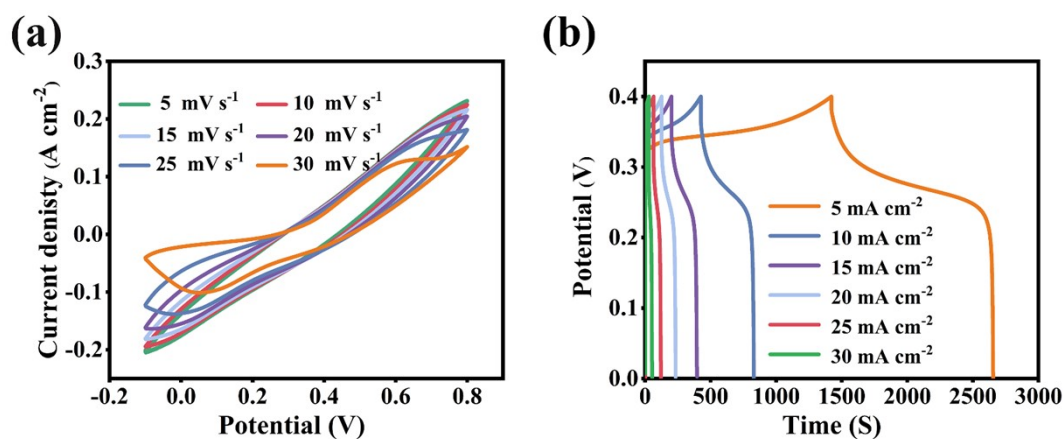
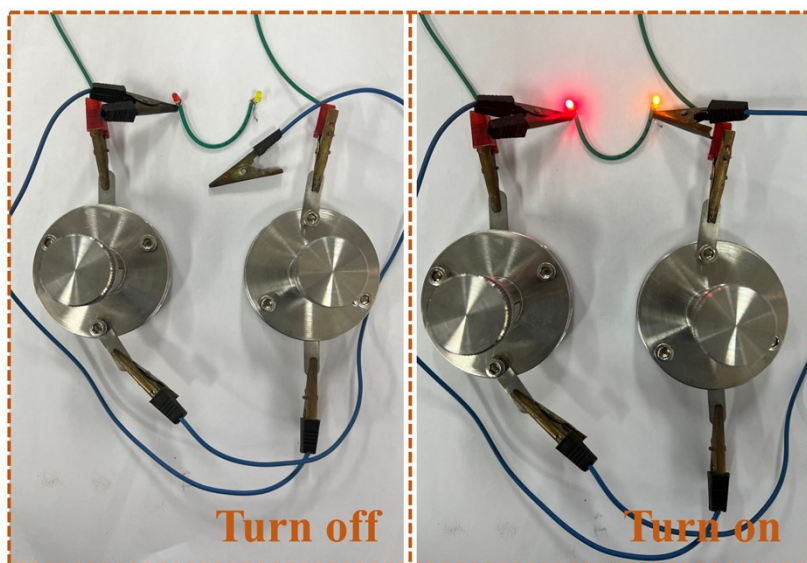


Fig. S10 (a) CV curves of  $\text{CuCo-LDH@Ni(OH)}_2$  electrode at different scan rates from  $5 \text{ mV s}^{-1}$  to  $30 \text{ mV s}^{-1}$  and (b) the corresponding

GCD curves at different current densities from 5 mA cm<sup>-2</sup> to 30 mA cm<sup>-2</sup>.



**Fig. S11** Two LED lights powered by two series connected HSC devices.

**Table S1** Comparison of the internal resistance ( $R_s$ ) value and charge-transfer resistance ( $R_{ct}$ ) value of  $\text{CuCo}_2\text{S}_4$  and  $\text{CuCo}_2\text{S}_x\text{OH}_y$ -1,  $\text{CuCo}_2\text{S}_x\text{OH}_y$ -5 and  $\text{CuCo}_2\text{S}_x\text{OH}_y$ -10.

Sample	$R_s$ ( $\Omega$ )	$R_{ct}$ ( $\Omega$ )
$\text{CuCo}_2\text{S}_4$	3.5	2.8
$\text{CuCo}_2\text{S}_x\text{OH}_y$ -1	3.2	2.1
$\text{CuCo}_2\text{S}_x\text{OH}_y$ -5	2.2	1.0
$\text{CuCo}_2\text{S}_x\text{OH}_y$ -10	2.7	2.1