

## Supporting Information

### **Ni, S co-doped Cu dendrites decorated with core-shell architecture assisted by MOF and Fe<sub>0.92</sub>Co<sub>0.08</sub>S nanoflakes on nanocellulose/graphene fibers for fabrication of flexible wire-type micro-supercapacitor**

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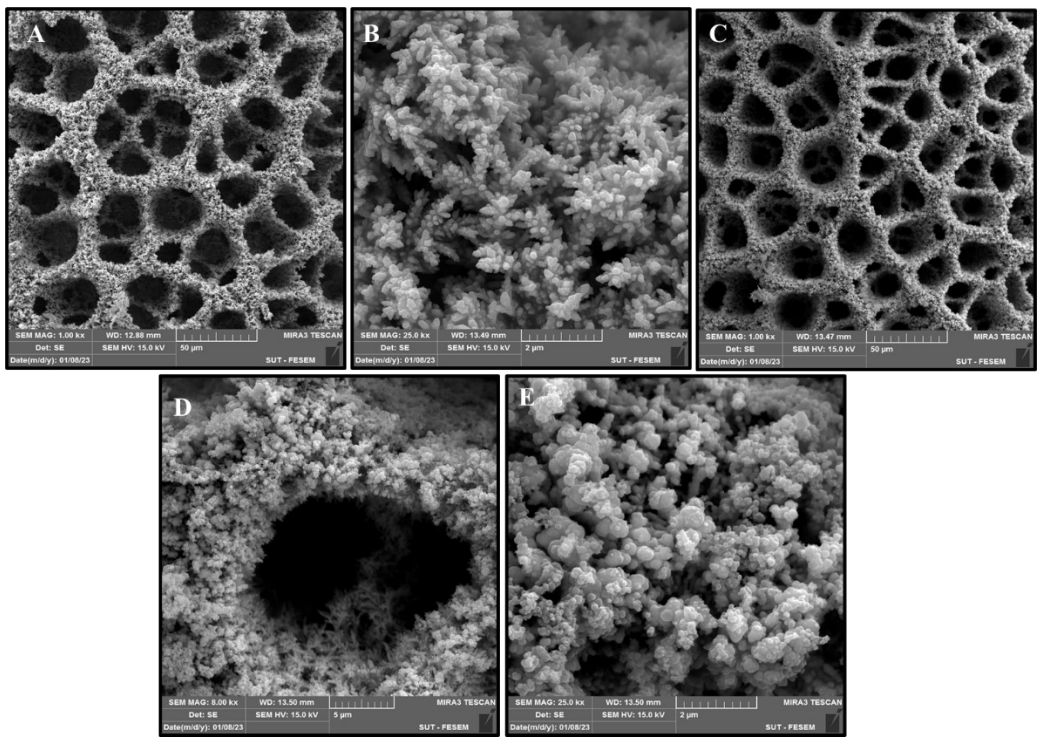
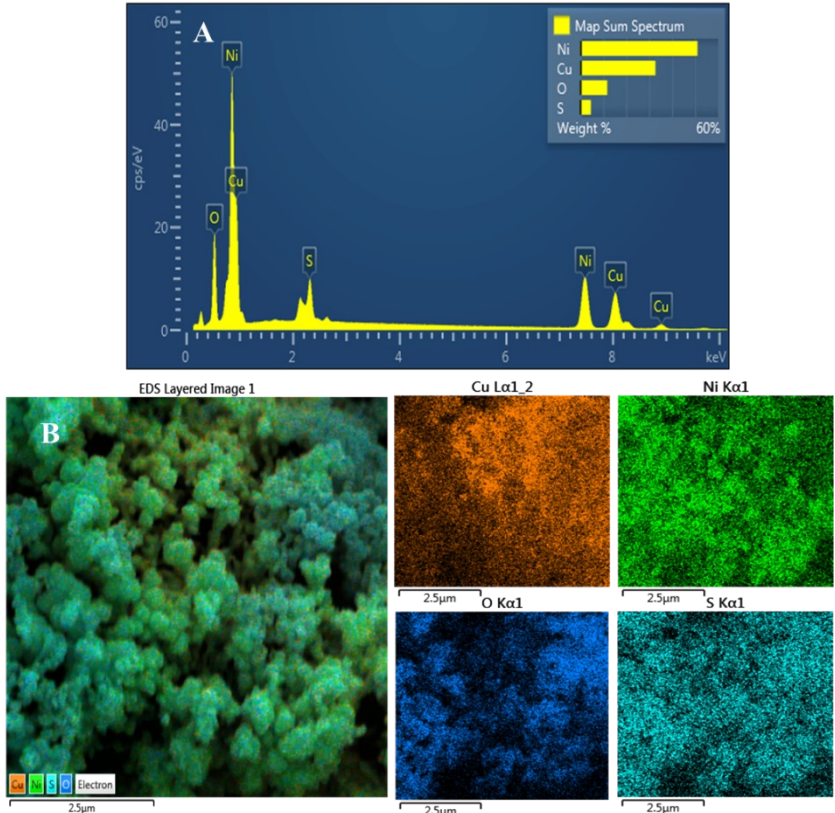
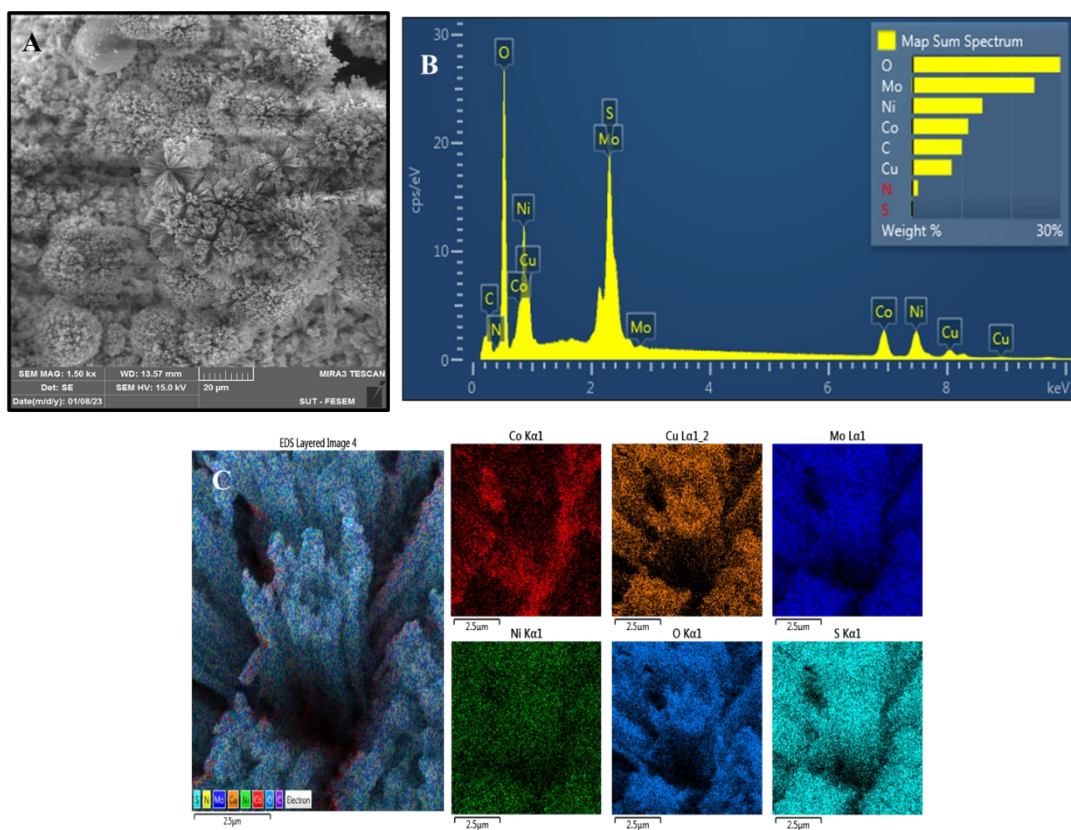


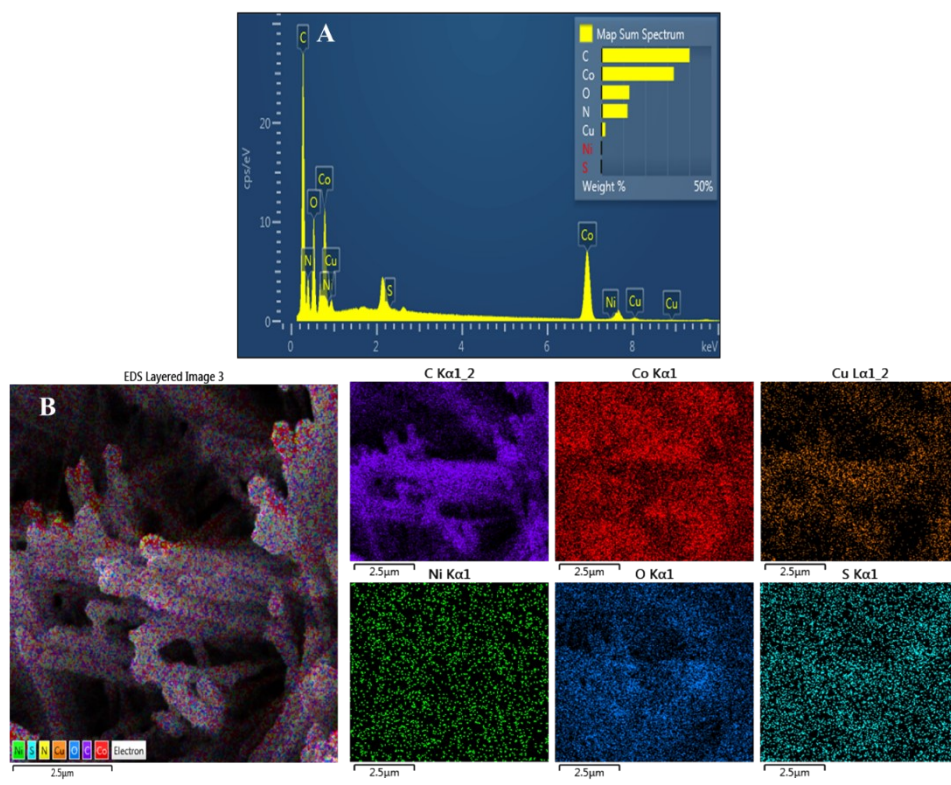
Fig. S1. FE-SEM images of the (A, B) Cu film@CW, (C-E) Ni,S-doped Cu@CW



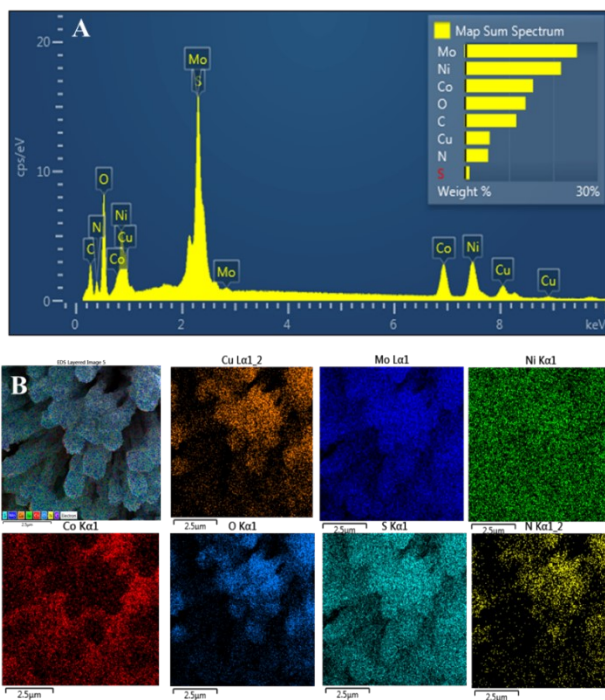
**Fig. S2.** (A) EDX spectra, (B) Elemental mapping of the Ni,S doped Cu@CW



**Fig. S3.** (A) FESEM images, (B) EDX spectra, (C) elemental mapping of the NiMoCo-LTH/Ni,S-doped Cu@CW

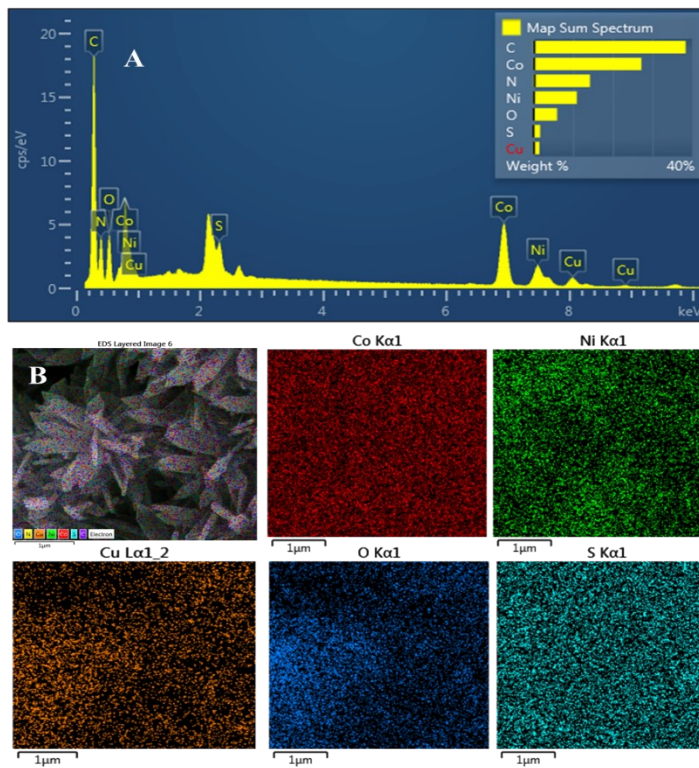


**Fig. S4.** (A) EDX spectra, (B) Elemental mapping of the ZIF-67@Ni,S-doped Cu@CW

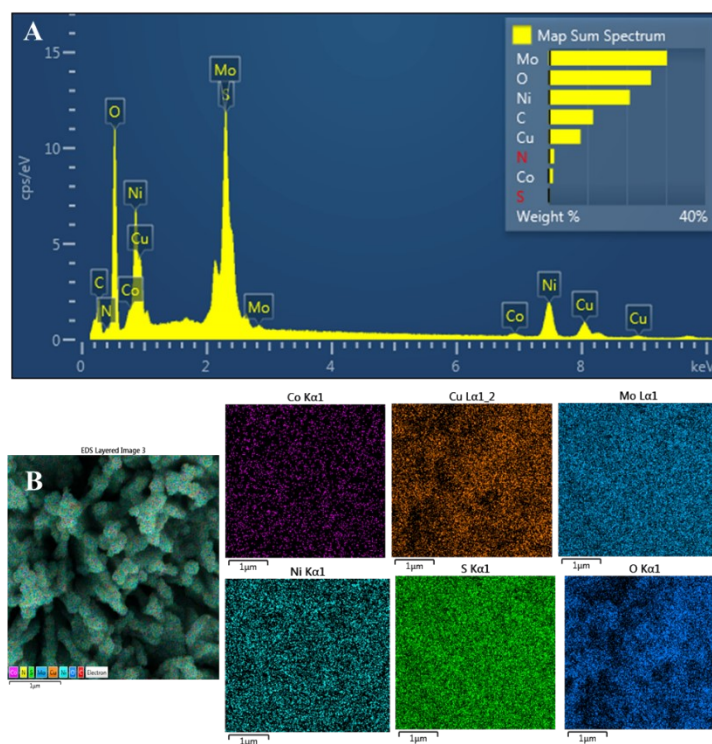


**Fig. S5.** (A) EDX spectra, (B) elemental mapping of the Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu@CW

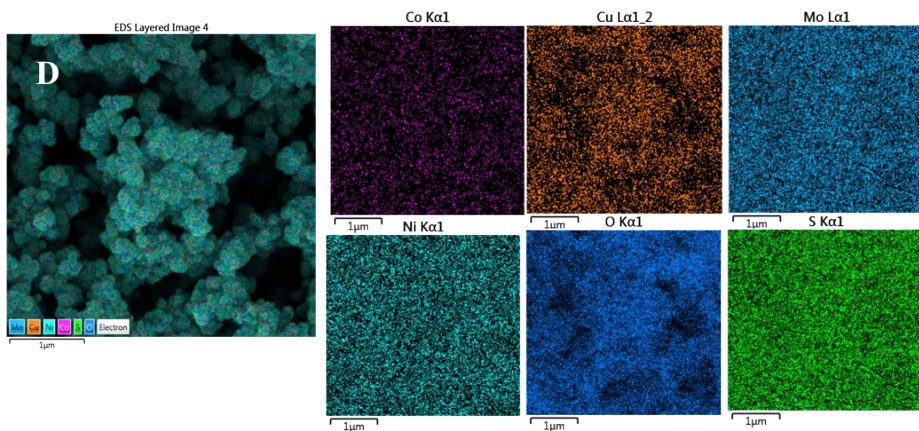
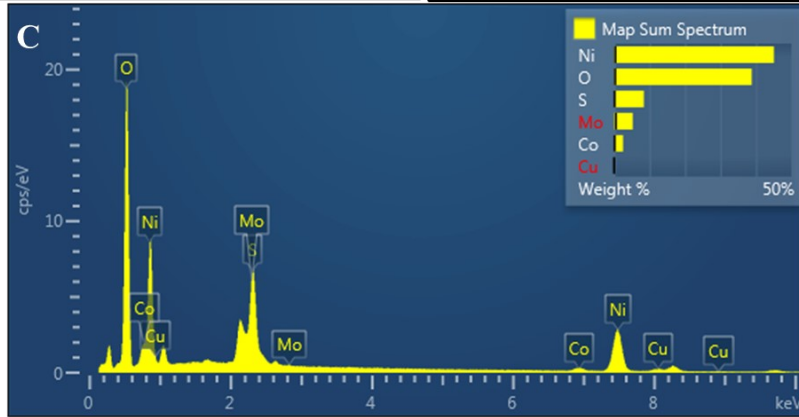
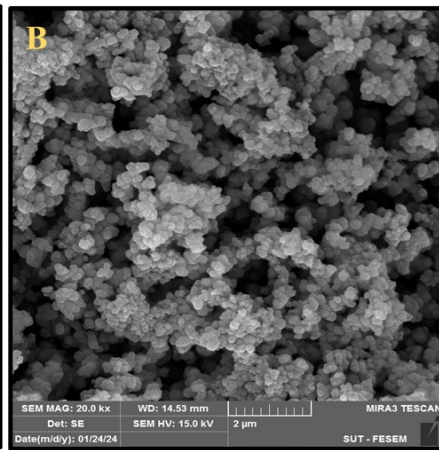
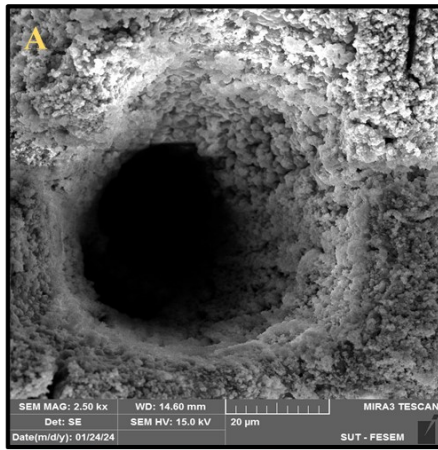




**Fig. S6.** (A) EDX spectra, (B) elemental mapping of the ZIF-Co leaf-like/Ni,S-doped Cu@CW

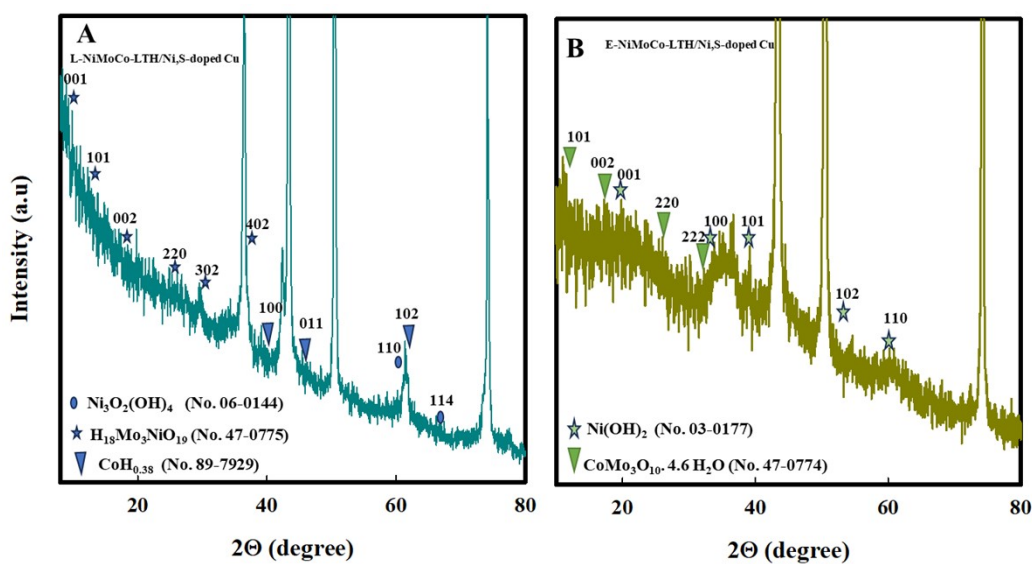


**Fig. S7.** (A) EDX spectra, (B) elemental mapping of the L-NiMoCo-LTH/Ni,S-doped Cu@CW



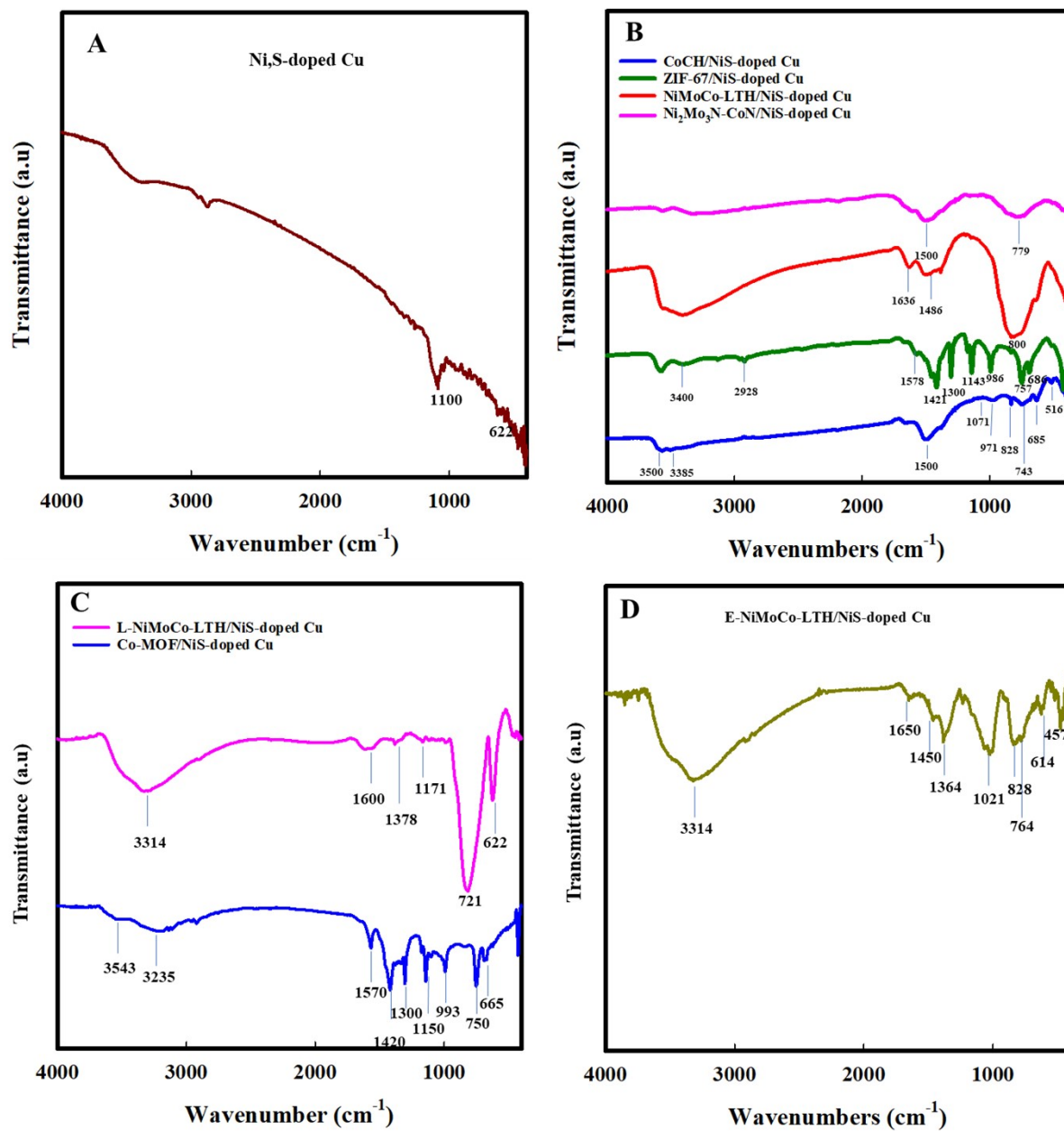


**Fig. S8.** (A, B) FE-SEM images, (C) EDX spectra, (D) elemental mapping of the E-NiMoCo-LTH/Ni,S-doped Cu@CW

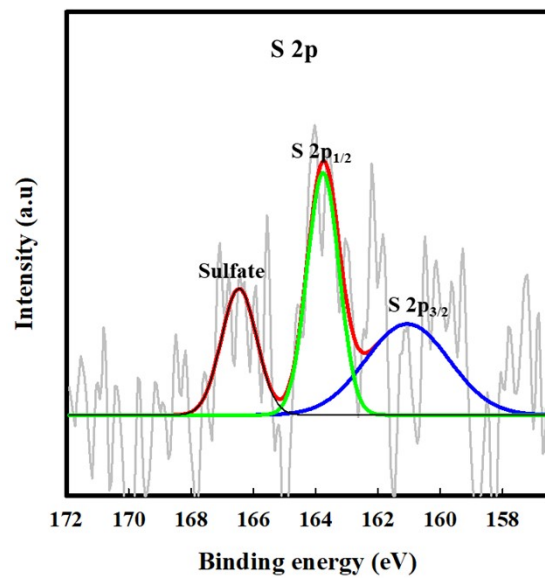


**Fig. S9.** XRD patterns of the (A) L-NiMoCo-LTH/Ni,S-doped Cu@CW, and (B) E-NiMoCo-LTH/Ni,S-doped Cu@CW



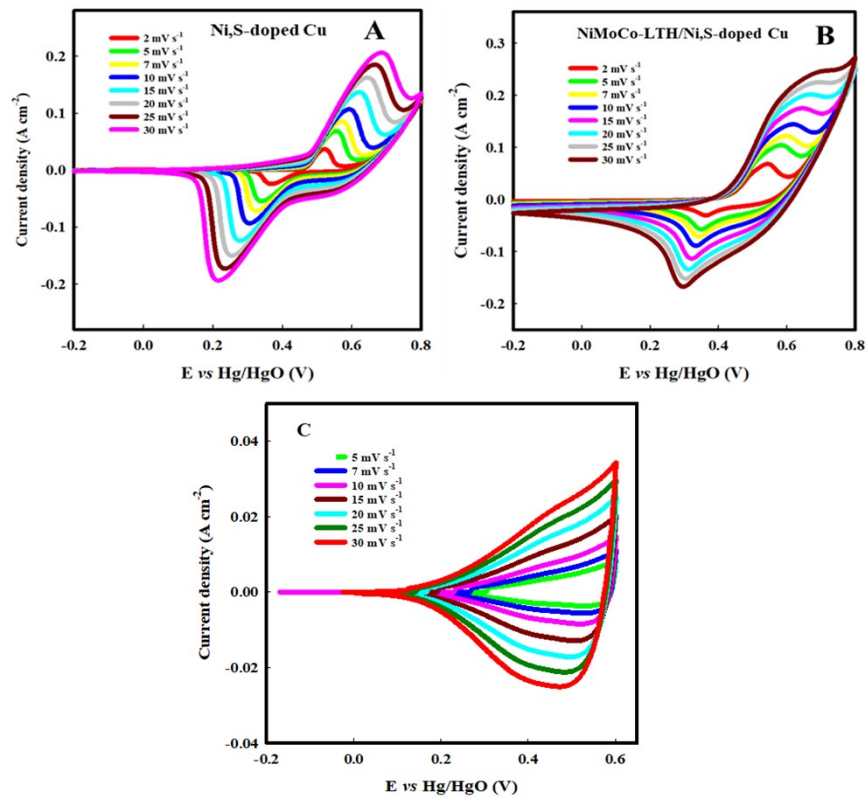


**Fig. S10.** FT-IR spectra of the (A) Ni,S-doped Cu, (B) CoCH/Ni,S-doped Cu, ZIF-67/Ni,S-doped Cu, NiMoCo-LTH/Ni,S-doped Cu, Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu, (C) ZIF-Co leaf-like/Ni,S-doped Cu, L-NiMoCo-LTH/Ni,S-doped Cu and (D) E-NiMoCo-LTH/Ni,S-doped Cu

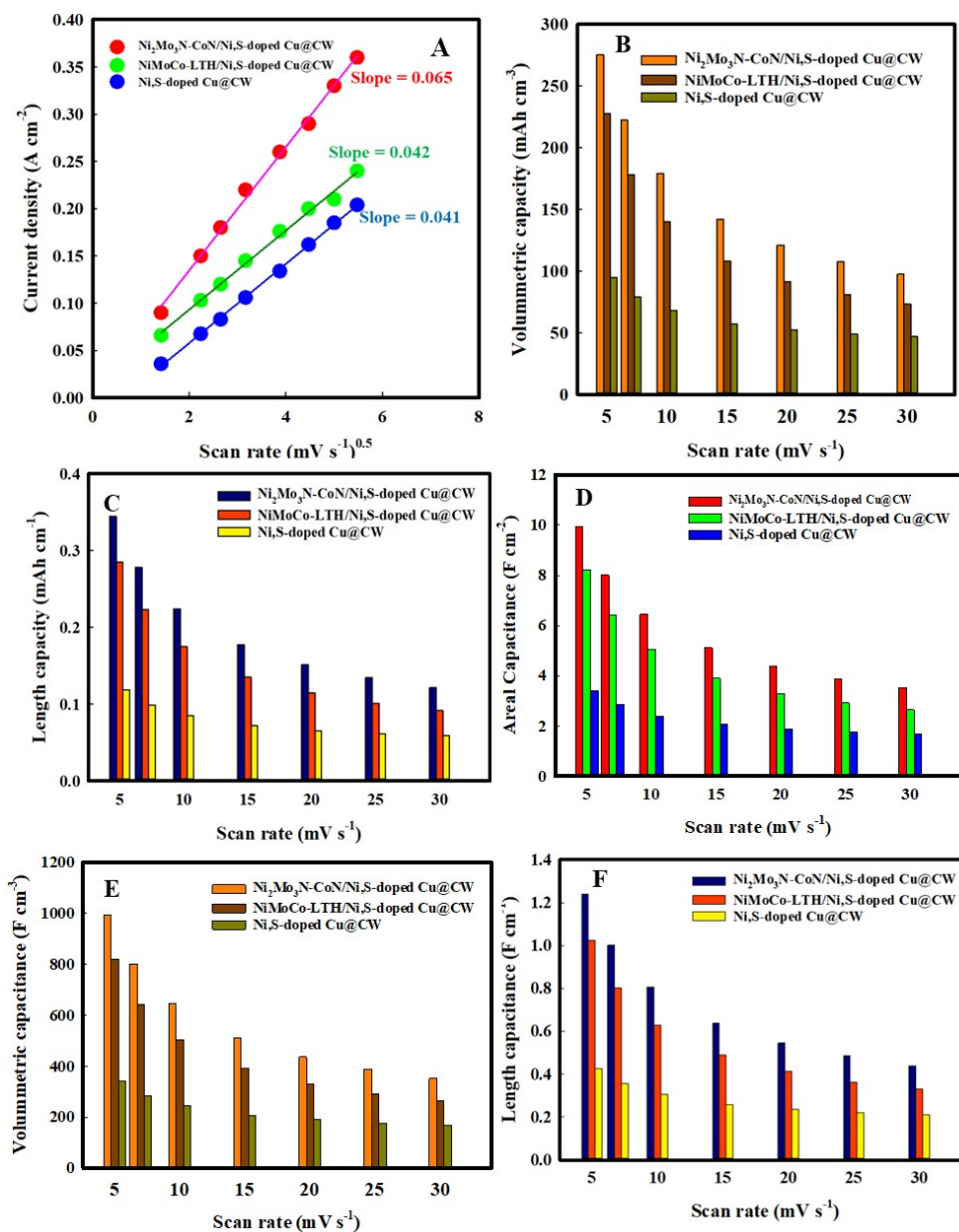


**Fig. S11.** XPS spectra of the S 2p

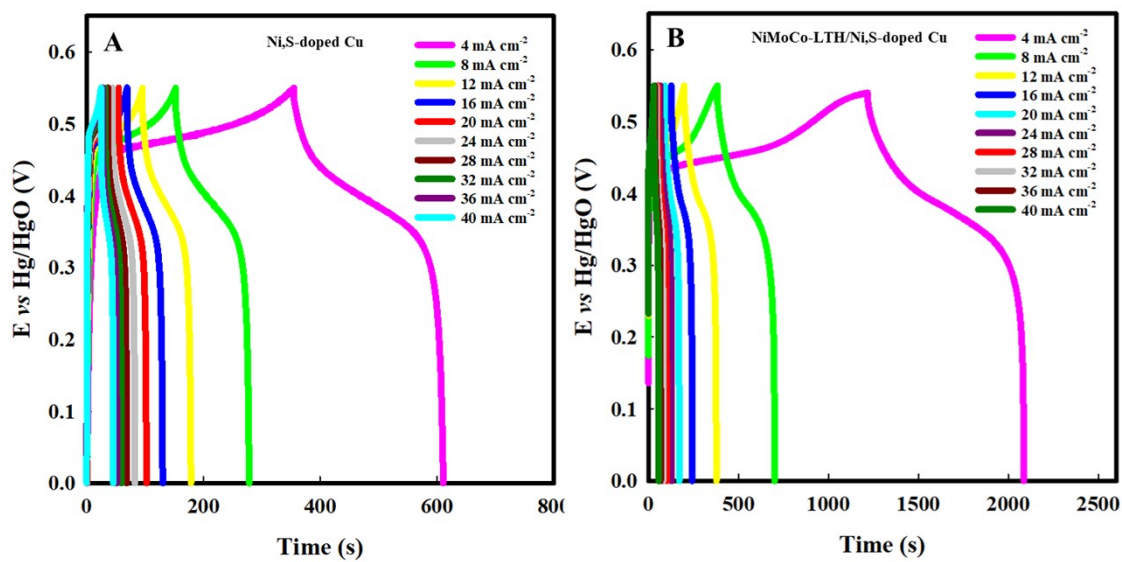




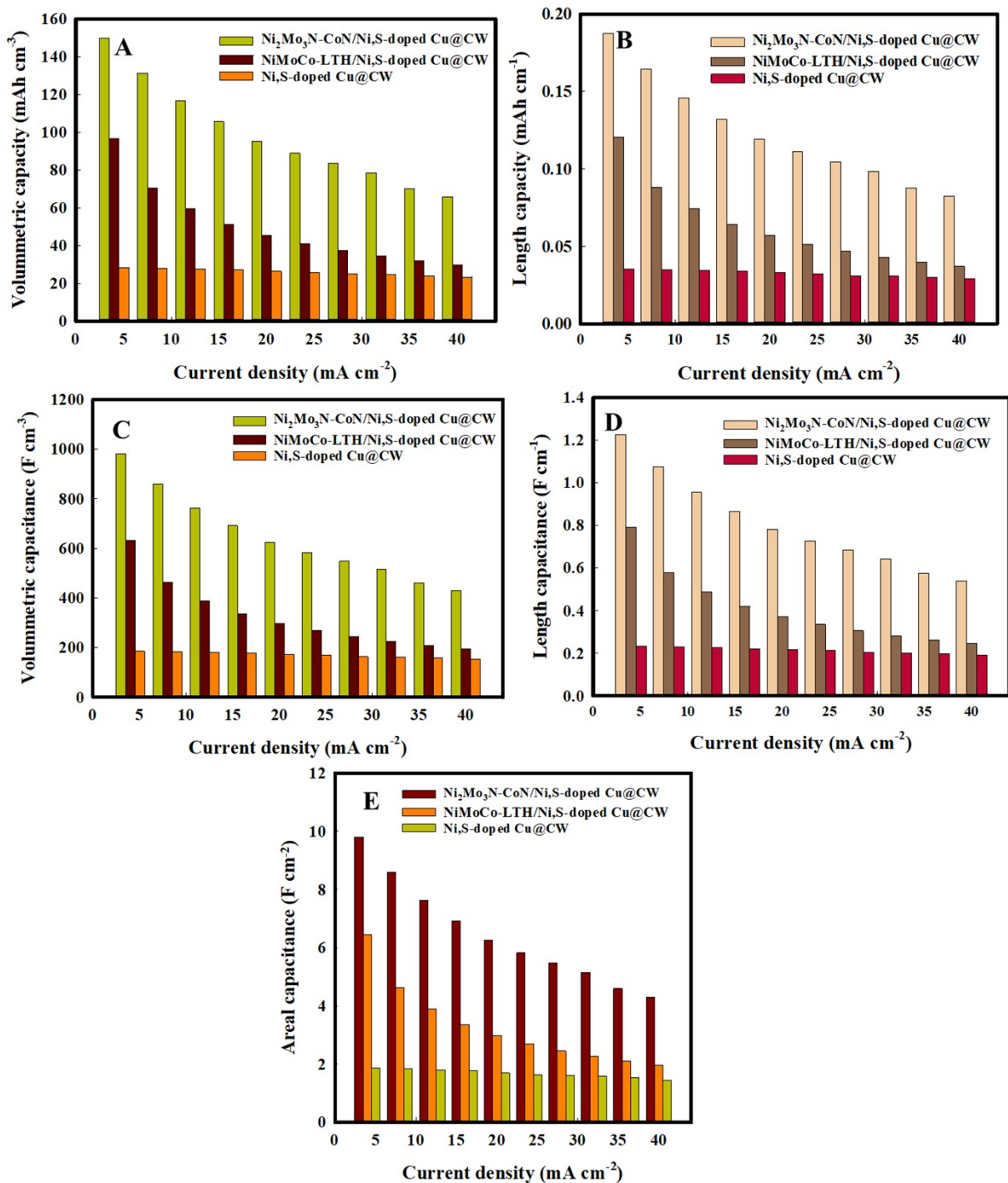
**Fig. S12.** CV curves of the (A) Ni,S-doped Cu@CW, (B) NiMoCo-LTH/Ni,S-doped Cu@CW and (C) Cu film at different scan rates



**Fig. S13.** (A) Plot of the current density as function of the scan rate square root, (B) Plot of the volumetric and (C) length capacities, (D) Areal, (E) volumetric and (F) length capacitance calculated from CV curves for the  $\text{Ni,S-doped Cu/CW}$ ,  $\text{NiMoCo-LTH/Ni,S-doped Cu@CW}$ ,  $\text{Ni}_2\text{Mo}_3\text{N-CoN/Ni,S-doped Cu@CW}$  microelectrodes at different scan rates.

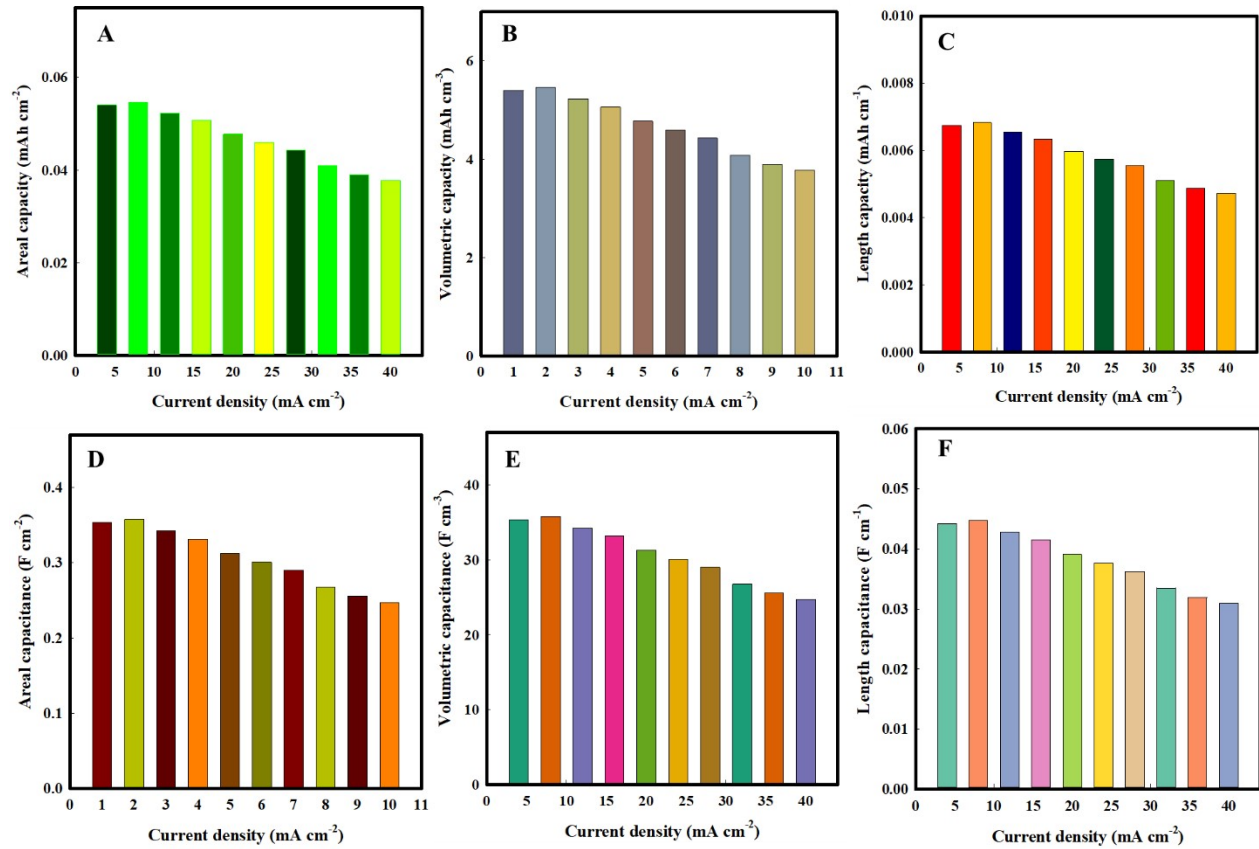


**Fig. S14.** GCD curves of the (A) Ni,S-doped Cu and (B) NiMoCo-LTH/Ni,S-doped Cu at different current densities

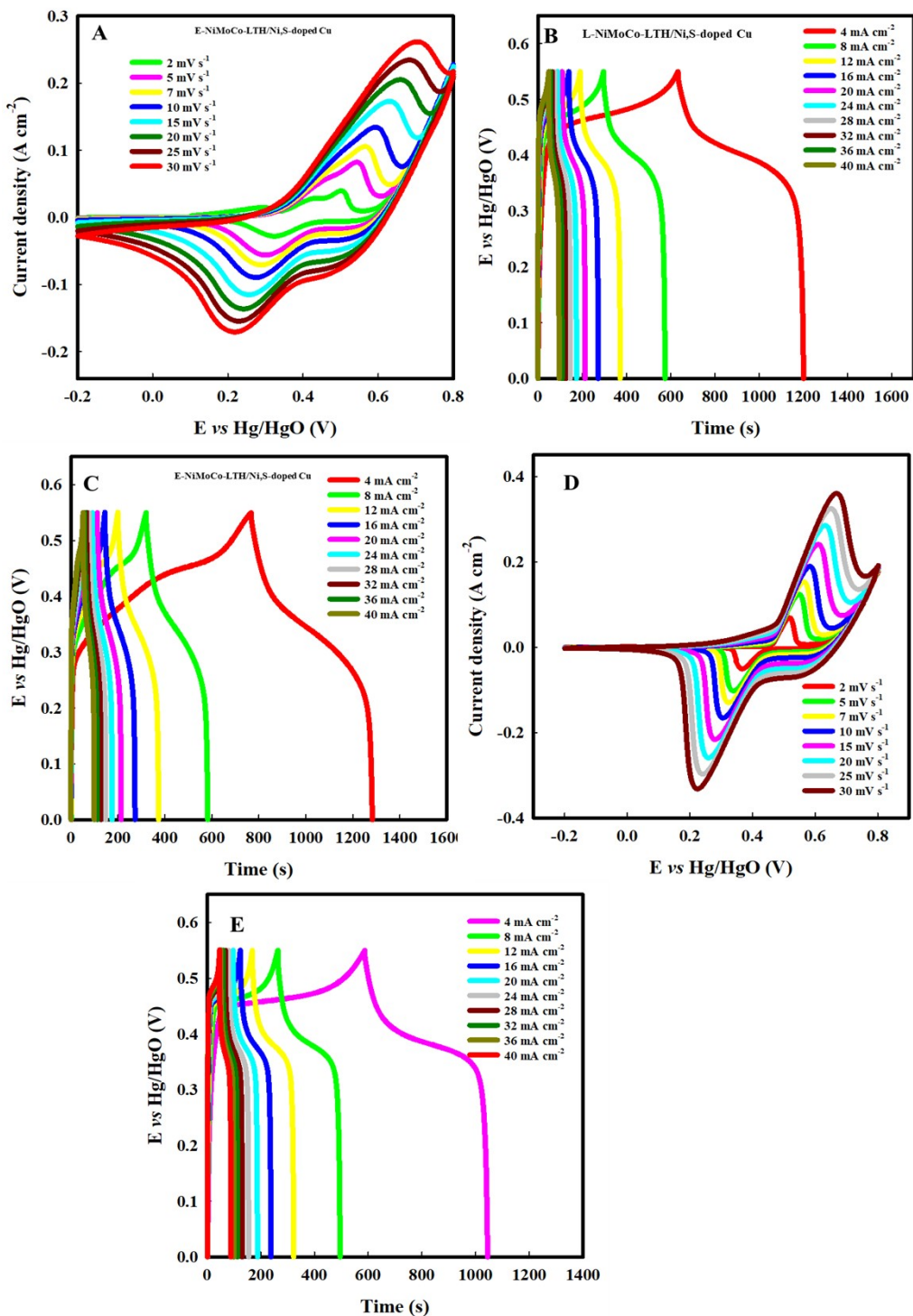


**Fig. S15.** Plot of the (A) volumetric, (B) length capacities and (C) volumetric, (D) length, (E) areal capacitances calculated from GCD curves for the Ni,S-doped Cu/CW, NiMoCo-LTH/Ni,S-doped Cu@CW, Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu@CW microelectrodes at different current densities.

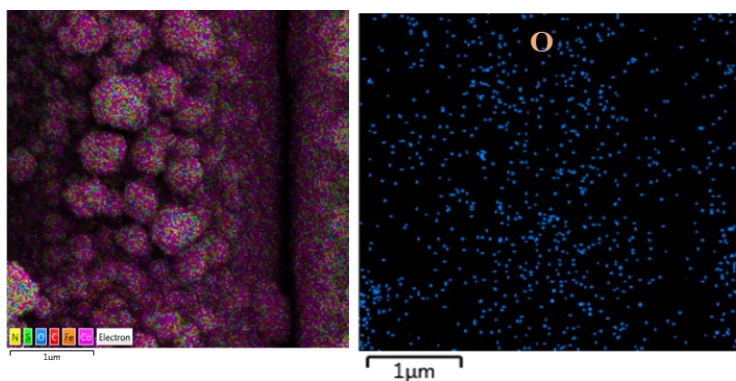




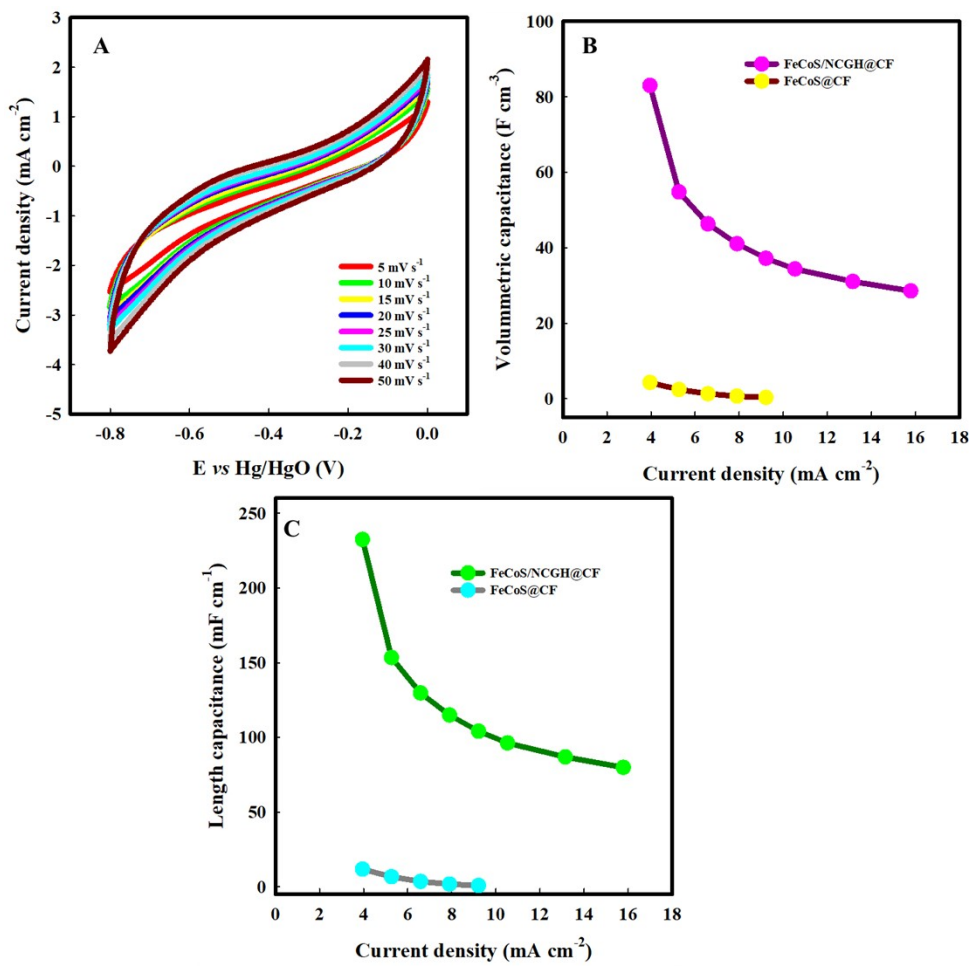
**Fig. S16.** Plot of the (A)Areal, (B) volumetric and (C) length capacities and (D) Areal, (E) volumetric and (F) length capacitances, calculated from GCD curves for the Cu film@CW microelectrode at different current densities.



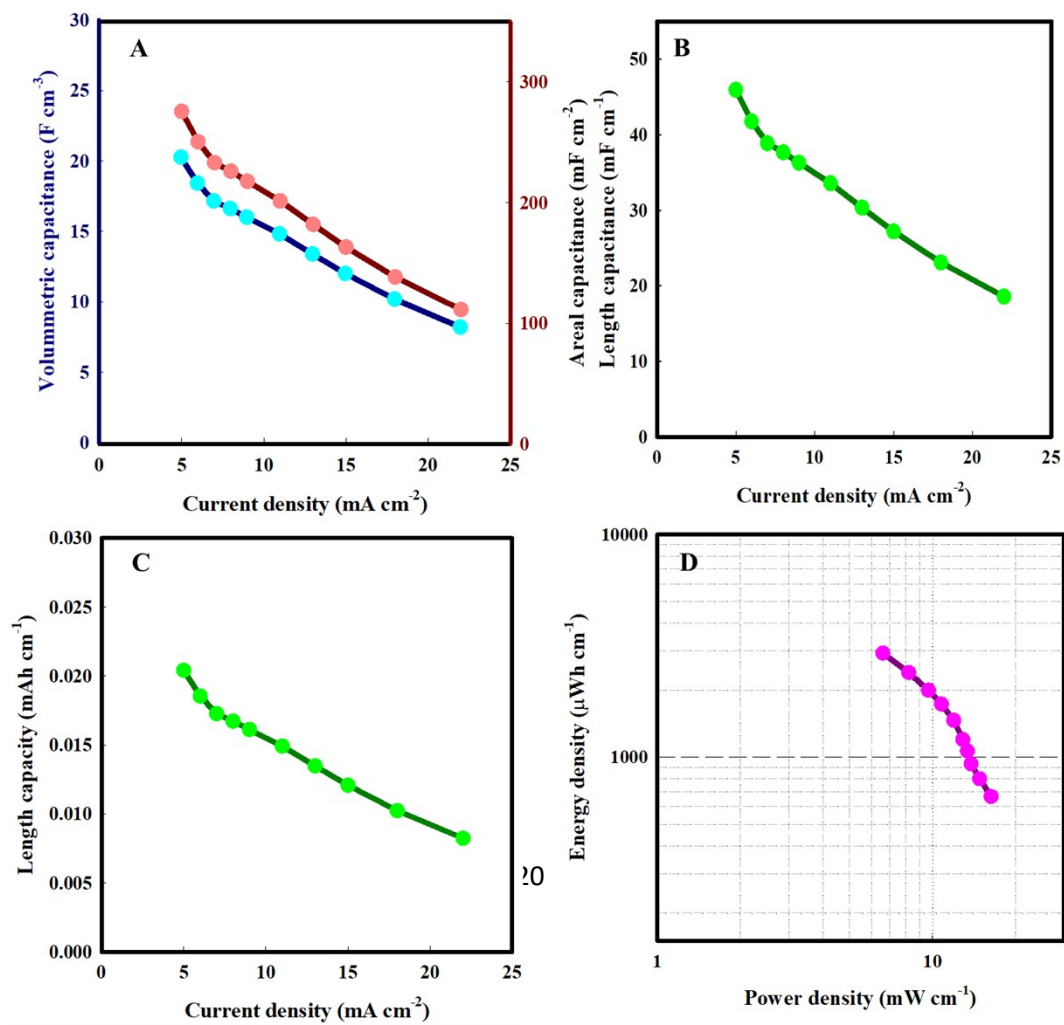
**Fig. S17.** (A) CV curves of the E-NiMoCo-LTH/Ni,S-doped Cu@CW at different scan rates, (B) GCD curves of the L-NiMoCo-LTH/Ni,S-doped Cu@CW at different current densities, (C) GCD curves of the E-NiMoCo-LTH/Ni,S-doped Cu@CW at different current densities, (D) CV curves of the NiMo-LDH/Ni,S-doped Cu@CW at different scan rates, (E) GCD curves of the NiMo-LDH/Ni,S-doped Cu@CW at different current densities



**Fig. S18.** Elemental mapping of the FeCoS/NCGH@CF

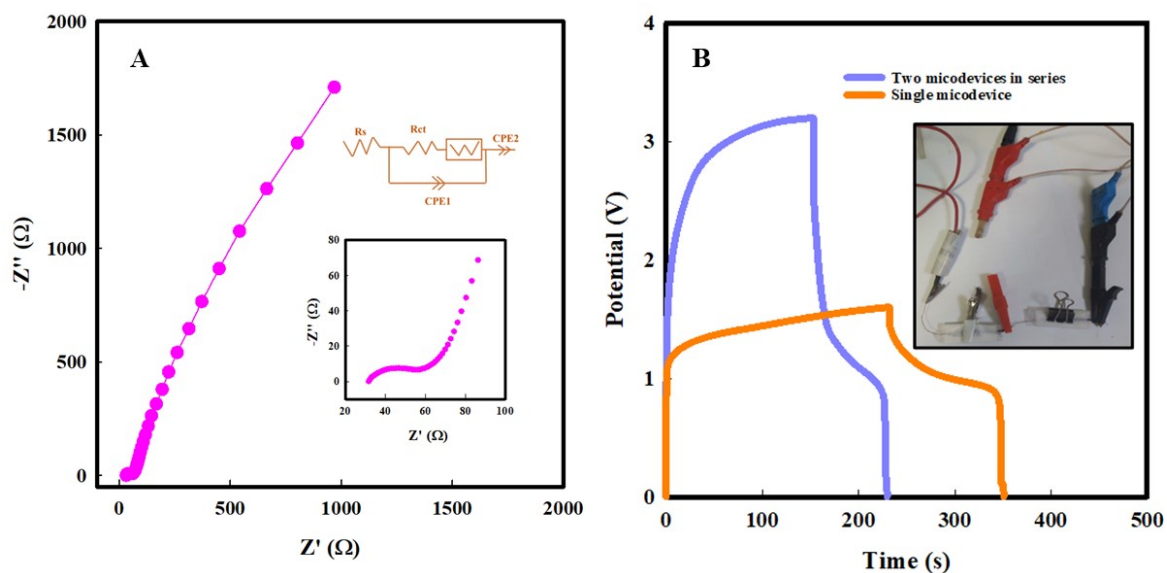


**Fig. S19.** (A) CV curves of the FeCoS@CF at different scan rates, (B) plot of the volumetric capacitance calculated from GCD curves for FeCoS@CF and FeCoS/GNCH at different current densities, (C) plot of the length capacitance calculated from GCD curves for FeCoS@CF and FeCoS/GNCH at different current densities.





**Fig. S20.** Plot of the (A) Areal, volumetric capacities, and length (B) capacity and (C) capacitance calculated from GCD curves 1D microdevice at different current densities, (B) length Ragon plot for 1D microdevice



**Fig. S21.** (A) EIS spectra of the assembled microdevice, (B) GCD profiles of two micro-devices connected in series at 1.25 mA.

Electrode material	Areal capacity (capacitance)	Volumetric capacity (capacitance)	Length capacity (capacitance)
<b>Ni,S-doped Cu</b>	0.947 mAh cm <sup>-2</sup> (3.41 F cm <sup>-2</sup> )	94.74 mAh cm <sup>-3</sup> (341.06 F cm <sup>-3</sup> )	0.118 mAh cm <sup>-1</sup> (426.3 mF cm <sup>-1</sup> )
<b>NiMo-LDH/Ni,S-doped Cu</b>	1.34 mAh cm <sup>-2</sup> (4.84 F cm <sup>-2</sup> )	134.4 mAh cm <sup>-3</sup> (483.76 F cm <sup>-3</sup> )	0.168 mAh cm <sup>-1</sup> (604.7 mF cm <sup>-1</sup> )
<b>NiMoCo-LTH/Ni,S-doped Cu</b>	2.279 mAh cm <sup>-2</sup> (8.204 F cm <sup>-2</sup> )	227.9 mAh cm <sup>-3</sup> (820.4 F cm <sup>-3</sup> )	0.285 mAh cm <sup>-1</sup> (1025.5 mF cm <sup>-1</sup> )
<b>L-NiMoCo-LTH/Ni,S-doped Cu</b>	1.31 mAh cm <sup>-2</sup> (4.72 F cm <sup>-2</sup> )	131.1 mAh cm <sup>-3</sup> (472 F cm <sup>-3</sup> )	0.164 mAh cm <sup>-1</sup> (590 mF cm <sup>-1</sup> )
<b>E-NiMoCo-LTH/Ni,S-doped Cu</b>	1.92 mAh cm <sup>-2</sup> (6.916 F cm <sup>-2</sup> )	192.1 mAh cm <sup>-3</sup> (691.6 F cm <sup>-3</sup> )	0.240 mAh cm <sup>-1</sup> (864 mF cm <sup>-1</sup> )
<b>Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu</b>	2.755 mAh cm <sup>-2</sup> (9.92 F cm <sup>-2</sup> )	275.5 mAh cm <sup>-3</sup> (992 F cm <sup>-3</sup> )	0.344 mAh cm <sup>-1</sup> (1240 mF cm <sup>-1</sup> )

**Table S1.** Areal, volumetric and length capacity and capacitance values of the different microelectrodes at scan rate of 5 mV s<sup>-1</sup>

**Table S2.** Areal, volumetric and length capacity and capacitance values of the different microelectrodes at current density of  $4 \text{ mA cm}^{-2}$

Electrode material	Areal capacity (capacitance)	Volumetric capacity (capacitance)	Length capacity (capacitance)
<b>Ni,S-doped Cu</b>	0.284 mAh cm <sup>-2</sup> (1.86 F cm <sup>-2</sup> )	28.4 mAh cm <sup>-3</sup> (185.89 F cm <sup>-3</sup> )	0.0355 mAh cm <sup>-1</sup> (232.4 mF cm <sup>-1</sup> )
<b>NiMo-LDH/Ni,S-doped Cu</b>	0.508 mAh cm <sup>-2</sup> (3.32 F cm <sup>-2</sup> )	50.8 mAh cm <sup>-3</sup> (332.5 F cm <sup>-3</sup> )	0.063 mAh cm <sup>-1</sup> (415.6 mF cm <sup>-1</sup> )
<b>NiMoCo-LTH/Ni,S-doped Cu</b>	0.966 mAh cm <sup>-2</sup> (6.32 F cm <sup>-2</sup> )	96.6 mAh cm <sup>-3</sup> (632.14 F cm <sup>-3</sup> )	0.121 mAh cm <sup>-1</sup> (790.2 mF cm <sup>-1</sup> )
<b>L-NiMoCo-LTH/Ni,S-doped Cu</b>	0.631 mAh cm <sup>-2</sup> (4.13 F cm <sup>-2</sup> )	63.1 mAh cm <sup>-3</sup> (413.23 F cm <sup>-3</sup> )	0.079 mAh cm <sup>-1</sup> (516.5 mF cm <sup>-1</sup> )
<b>E-NiMoCo-LTH/Ni,S-doped Cu</b>	0.573 mAh cm <sup>-2</sup> (3.75 F cm <sup>-2</sup> )	57.33 mAh cm <sup>-3</sup> (375.27 F cm <sup>-3</sup> )	0.0716 mAh cm <sup>-1</sup> (469 mF cm <sup>-1</sup> )
<b>Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu</b>	1.5 mAh cm <sup>-2</sup> (9.81 F cm <sup>-2</sup> )	149.92 mAh cm <sup>-3</sup> (981.3 F cm <sup>-3</sup> )	0.187 mAh cm <sup>-1</sup> (1230 mF cm <sup>-1</sup> )

**Table S3.** Performance comparison of the Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu@CW with other reports

Electrode materials	Current collectors	Current density	Specific capacitance	Ref
<b>Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu</b>	Cu wire	4 mA cm <sup>-2</sup>	9.81 F cm <sup>-2</sup> 981.3 F cm <sup>-3</sup> 1.22 F cm <sup>-1</sup>	This work
<b>S-doped CoZnNi-OH/CuCoP</b>	Cu wire	4 mA cm <sup>-2</sup>	2.9 F cm <sup>-2</sup> 290.1 F cm <sup>-3</sup> 0.348 F cm <sup>-1</sup>	1
<b>MnCo<sub>2</sub>O<sub>4</sub>@Co<sub>3</sub>O<sub>4</sub></b>	Ni foam (using binder)	1 mA cm <sup>-2</sup>	3199.24 mF cm <sup>-2</sup>	2

<b>CuO/CF@ NiCoMn-OH</b>	Cu foam	8 mA cm <sup>-2</sup> 1 A g <sup>-1</sup>	26.8 F cm <sup>-2</sup> 2866 F g <sup>-1</sup>	3
<b>Ni-P@NMC-LDH</b>	Ni foam	1 A g <sup>-1</sup>	2980 F g <sup>-1</sup>	4
<b>NiCoMo oxide</b>	Ni foam	1 A g <sup>-1</sup>	3.31 F cm <sup>-2</sup>	5
<b>NiMo-LDH</b>	Ni foam	4 mA cm <sup>-2</sup>	4.4 F cm <sup>-2</sup>	6
<b>CuO@NiMoO<sub>4</sub></b>	Cu foam	3 mA cm <sup>-2</sup>	3.9 F cm <sup>-2</sup>	7
<b>Cu<sub>3</sub>N@NiCo-N/Cu</b>	Cu foam	1 mA cm <sup>-2</sup>	8.49 F cm <sup>-2</sup>	8
<b>Co<sub>9</sub>S<sub>8</sub></b>	Ni foam	1 A g <sup>-1</sup>	369.1 mAh g <sup>-1</sup>	9
<b>Cu<sub>x</sub>O NW@CoS<sub>2</sub></b>	Cu foam	1 mA cm <sup>-2</sup>	2.46 mAh cm <sup>-3</sup>	10
<b>MnCo<sub>2</sub>O<sub>4</sub>/porous Ni/Ni</b>	Cu wire	2 mA cm <sup>-2</sup>	1.798 F cm <sup>-2</sup>	11
<b>3D-NiCo<sub>2</sub>S<sub>4</sub></b>	Ni wire	0.2 mA cm <sup>-1</sup>	199.74 F cm <sup>-3</sup> 98 mF cm <sup>-1</sup> 1.248 F cm <sup>-2</sup>	12
<b>3D-NiCoO<sub>4</sub></b>	Ni wire	2.5 mA	38.84 F cm <sup>-3</sup>	13

**Table S4.** the comparative of the electrochemical properties of electrodes based on fiber substrates and similar electroactive material for supercapacitor

Supercapacitor	Specific capacitance	Current density	Voltage	Maximum energy density	Maximum power density	Stability	Ref
<b>Ni<sub>2</sub>Mo<sub>3</sub>N-CoN/Ni,S-doped Cu@CW//FeCoS/GNCH@CF</b>	275.6 mF cm <sup>-2</sup> 20.3 F cm <sup>-3</sup>	5 mA cm <sup>-2</sup>	1.6 V	98 μWh cm <sup>-2</sup> 7.2 mWh cm <sup>-3</sup> 16.3 μWh cm <sup>-1</sup>	17.6 mW cm <sup>-2</sup> 1294 mW cm <sup>-3</sup> 2933 μW cm <sup>-1</sup>	94% after 5000 cycles At 10 mA cm <sup>-2</sup>	<b>This work</b>
<b>CuO/CF@NiCoMn-OH//AC</b>	19.3 F g <sup>-1</sup>	2 A g <sup>-1</sup>	1.5 V	37.28 Wh kg <sup>-1</sup>	At 170 W kg <sup>-1</sup>	62.5% even at 3000 cycles at 8 A g <sup>-1</sup>	3
<b>Ni-Co-N/GP GOP Substrate: graphene paper</b>	4.24 F/cm <sup>3</sup> 42.3 mF/cm <sup>2</sup>	0.5 mA/cm <sup>2</sup>	1.6 V	4.78 mWh cm <sup>-3</sup>	1.26 W/cm <sup>3</sup>	89% after 8000 cycles at 5 mA cm <sup>-2</sup>	14
<b>0.075 Mo-NiCo-LDH@C//RGO Substrate: CC</b>	235.1 C g <sup>-1</sup>	1 A g <sup>-1</sup>	1.6 V	52.2 Wh kg <sup>-1</sup>	32.2 Wh kg <sup>-1</sup>	84.2 % after 10000 cycles at 10 A g <sup>-1</sup>	15
<b>Cu<sub>3</sub>N@NiCo-N/Cu//rGO Substrate: Cu foam</b>	351.3 mF cm <sup>-2</sup>	2 mA cm <sup>-2</sup>	1.6 V	124.9 μWh cm <sup>-2</sup> 12.49 mWh cm <sup>-3</sup>	8 mW cm <sup>-2</sup> 0.8 W cm <sup>-3</sup>	After 8000 cycles at 20 mA cm <sup>-2</sup> 86.68%	8
<b>NF/Ni-P@NMC-LDH//AC Substrate: NF</b>	202.67 F g <sup>-1</sup>	0.8 A g <sup>-1</sup>	1.8 V	91.2 Wh kg <sup>-1</sup>	1812.3 W·kg <sup>-1</sup>	86.97 % after 5000 cycles at 3 A g <sup>-1</sup>	4
<b>MnCo<sub>2</sub>O<sub>4</sub>@Co<sub>3</sub>O<sub>4</sub>/AC Substrate: NF</b>	1338 C cm <sup>-2</sup> 148.7 C g <sup>-1</sup>	2.5 mA cm <sup>-2</sup>	1.5 V	31 Wh kg <sup>-1</sup>	3326.6 Wkg <sup>-1</sup>	101.23% after 8000 cycles at 130 mA cm <sup>-2</sup>	2
<b>Co<sub>3</sub>S<sub>4</sub>@NiO//AC Substrate: powder pasted on NF</b>	164.80 F g <sup>-1</sup>	1 A g <sup>-1</sup>	1.55 V	54.99 Wh kg <sup>-1</sup>	19.10 kW kg <sup>-1</sup>	86.1% after 10,000 cycles at 5 A g <sup>-1</sup>	16
<b>NiCoMoO-P//N-rGO Substrate: NF</b>	109.1F g <sup>-1</sup>	0.5 A g <sup>-1</sup>	1.6 V	45.3 Wh Kg <sup>-1</sup>	4000 W Kg <sup>-1</sup>	60% after 2000 Cycles at 1 A g <sup>-1</sup>	17
<b>CoNi<sub>2</sub>S<sub>4</sub>/E-NZP//rGO (solid state) Substrate: CW and CF</b>	241 mF cm <sup>-2</sup> 18.54 F cm <sup>-3</sup>	4 mA cm <sup>-2</sup>	1.8 V	108.4 μWh cm <sup>-2</sup> 8.34 mWh cm <sup>-3</sup>	9280 μW cm <sup>-2</sup> 716.9 mW cm <sup>-3</sup>	88.89% after 5000 cycles at 5.6 mA cm <sup>-2</sup>	18
<b>CoZnNiS@CNTs/rGO //carbon spheres integrated graphene</b>	185.1 F cm <sup>3</sup>	1.5 A g <sup>-1</sup>	1.6 V	65.2 Wh h L <sup>-1</sup>	at 1308 W L <sup>-1</sup>	90.6% after 10000 cycles	19
<b>CoVSe/NiCuSe@CW//PPy/RGO@CF</b>	351.7 mF cm <sup>-2</sup> 56.73 F cm <sup>-3</sup>	4.8 mA cm <sup>-2</sup>	1.6 V	111.4 μWh cm <sup>-2</sup> 20.17 mWh cm <sup>-3</sup>	12900 μW cm <sup>-2</sup> 2081.14 mW cm <sup>-3</sup>	96.7% after 5000 cycles at 6.4 mA cm <sup>-2</sup>	20
<b>Symmetric Cu@Ni/porous Ni/MnCo<sub>2</sub>O<sub>4</sub></b>	54.8 mF cm <sup>-2</sup> -	0.18 mA cm <sup>-2</sup>	1.3 V	4.8 μWh cm <sup>-2</sup>	1040 μW cm <sup>-2</sup>	-	11
<b>NiCo<sub>2</sub>S<sub>4</sub>//N-rGO</b>	120 mF cm <sup>-2</sup> 19.57 F cm <sup>-3</sup>	0.2 mA	1.4 V	32.67 μWh cm <sup>-2</sup> 5.33 mWh cm <sup>-3</sup>	5352.92 μW cm <sup>-2</sup> 855.69 mW cm <sup>-3</sup>	92% after 1000 cycles at 3 mA	12

## References



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