Supporting information

Chemicals and reagents

Cobalt nitrate hexahydrate ($Co(NO_3)_2 \cdot 6H_2O$, ≥ 99.0 %), Copper nitrate hexahydrate ($Cu(NO_3)_2 \cdot 6H_2O$, ≥ 99.0 %), nickel nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$, ≥ 99.0 %), thioacetamide (TAA, C_2H_5NS, ≥ 99.0 %) and trimesic acid (BTC, $C_9H_6O_6$, ≥ 98.0 %) were purchased from Aladdinlk. Polyvinylalcohol ($[C_2H_4O]n$) was purchased from Sinopharm. N, N-Dimethylformamide (DMF, analytical grade), absolute ethanol (C_2H_6O, ≥ 95.0 %), hydroxide (KOH, ≥ 99.0 %), polyvinyl pyrrolidone (PVP, K-30) were purchased from Kermel.Activated carbon (AC, 968.16 $m^2\ g^{-1}$), Carbon cloth, and nickel foam (NF) were obtained from Keliyuan. Deionized water was used for the entire process. Each reagent was of analytical grade and used directly.

Synthesis of flexible wearable electrodes

The carbon cloth (1x4 cm) was cleaned with acetone solution for 10 minutes, and soaked in 3mol/L hydrochloric acid for 30 minutes. Afterwards, the sample was washed multiple times with ethanol and water, and then subjected to vacuum drying at a temperature of 60 ℃ for a duration of 12 hours.

To prepare PVA/KOH gel, 2 g PVA was added to 30 ml water and heated at 100 °C for 1 hour until fully dissolved. Natural cooling was followed by adding and stirring 15 mL 6 mol/L KOH and placed in a petri dish to cool overnight.

The resulting slurry from the working electrode preparation method was applied onto the treated carbon cloth, while PVA/KOH gel was the diaphragm and then wrapped with insulating tape.

Preparation of the working electrode

The obtained sample was used as electrode material and prepared into a slurry with acetylene black and Nafion solution (wt 5 %) according to the mass ratio of 8:1:1. As a binder, Nafion solution was utilized while Acetylene black served as a conductive agent. After application to the nickel foam, the paste was weighed to 1.25 mg. Nickel foam was dried under vacuum at 60 °C for 6 h, and then pressed under 8 Mpa pressure for 3 min to yield the working elect Co-Ni-Cu/S rode.

Electrochemical calculation formula

The specific capacitance (C, F g^{-1}) was calculated from the discharge curves using the following equation.

$$C = \frac{2i_m \int V dt}{V_1^2 - V_0^2} \qquad (S1)$$

the specific capacitance Q_s (C g^{-1}) was calculated from the discharge curves using the following equation.

$$Q_s = C \times \Delta V \qquad (S2)$$

The energy density (E, Wh kg^{-1}) and power density (P, W kg^{-1}) against two electrodes in the device were calculated using the following equations.

$$\mathrm{E} = \frac{1/2\mathrm{C}(\Delta \mathrm{V})^2}{3.6} \qquad (S3)$$

$$\mathrm{P} = \frac{\mathrm{E} \times 3600}{\Delta \mathrm{t}} \qquad (S4)$$

The capacitance contribution and diffusion control contribution of the charge storage process are according to the following equation.

$$i(v) = k_1 v + k_2 v^{1/2} \quad \text{(S5)}$$

Where $i(v)$ is the peak current at a specific voltage, k_1 and k_2 represent constants, and v represents the scan rate.

The contribution of surface and diffusion energy storage processes to charge storage is according to the following equation.

$$i = av^b \quad \text{(S6)}$$

$$\log(i) = b\log(v) + \log(a) \quad \text{(S7)}$$

Supplementary figures

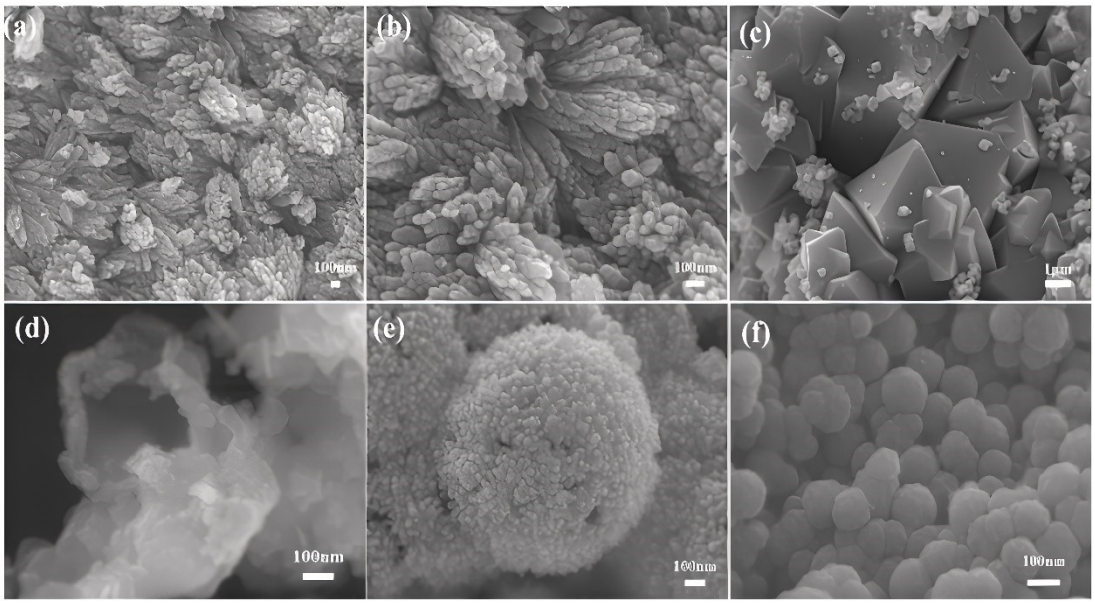

Fig. S1 . (a, b) SEM images of Co-Ni/BTC, (c) SEM images of Co-Ni-Cu/OH, (d, e, f) Co-Ni-Cu/S, Co-Ni-Mn/S, and Co-Ni-Fe-S, respectively.

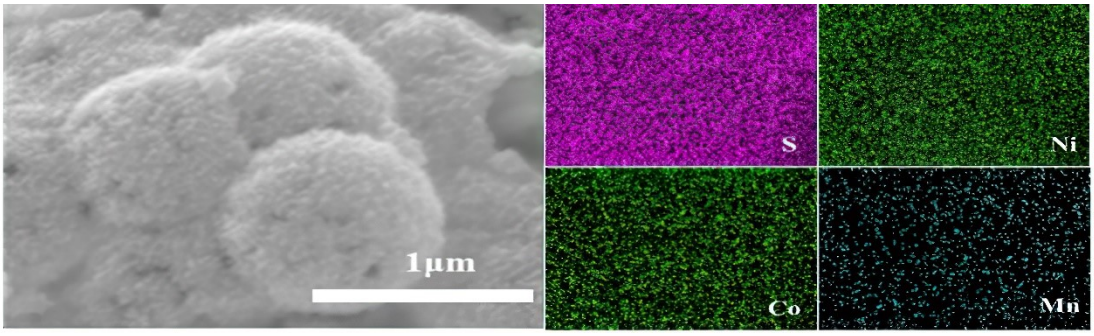

Fig. S2. FESEM image and corresponding EDS elemental mapping of Co-Ni-Mn/S.

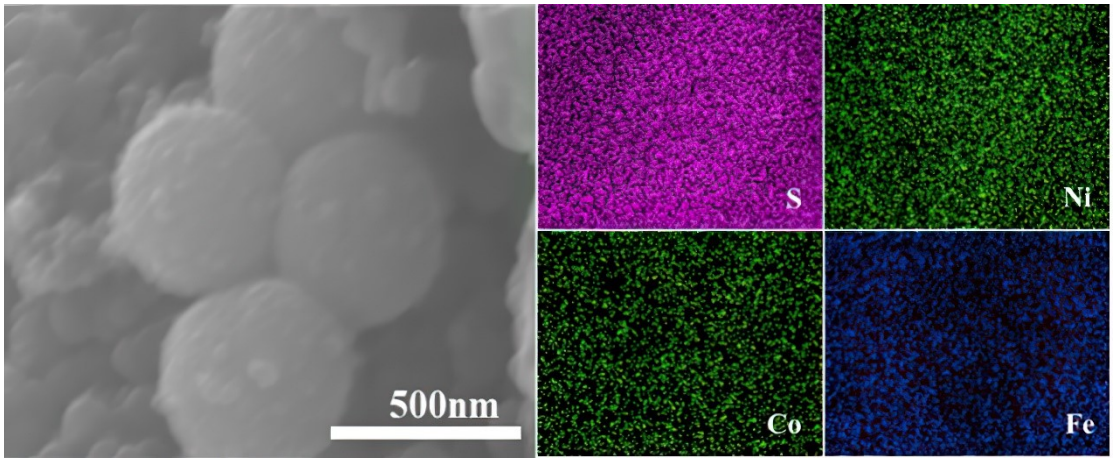

Fig. S3 . FESEM image and corresponding EDS elemental mapping of Co-Ni-Fe/S.

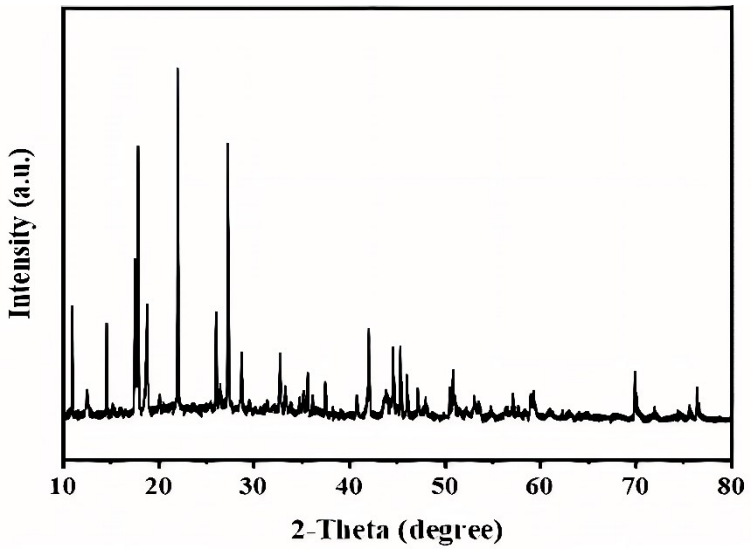

Fig. S4 . XRD patterns of Co-Ni/BTC.

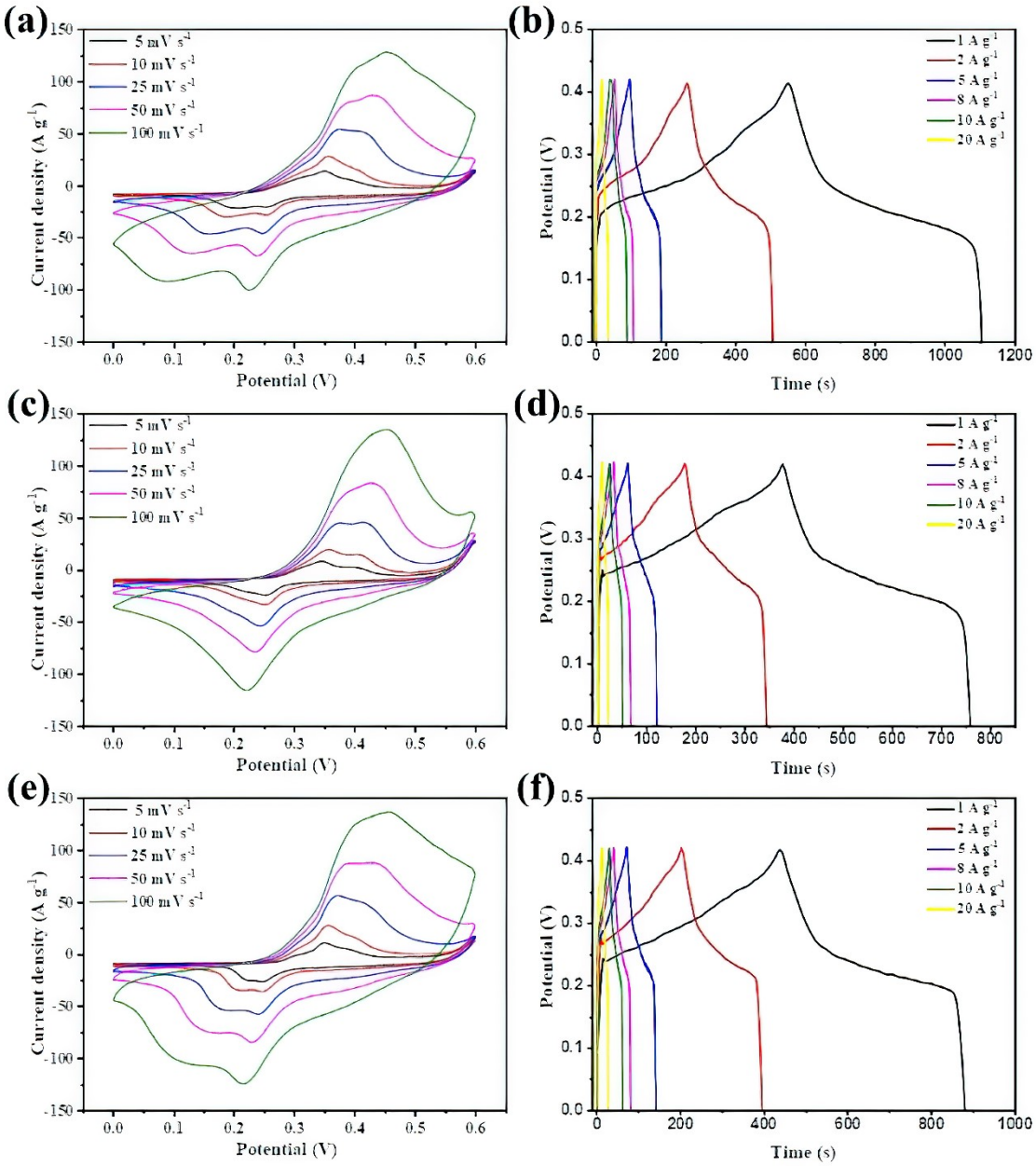

Fig. S5. (a, c, e) CV curves at various scan rates and (b, d, f) GCD curves at various densities of Co-Ni/BTC, Co-Ni-Cu/OH, and Co-Ni-Mn/OH,respectively.

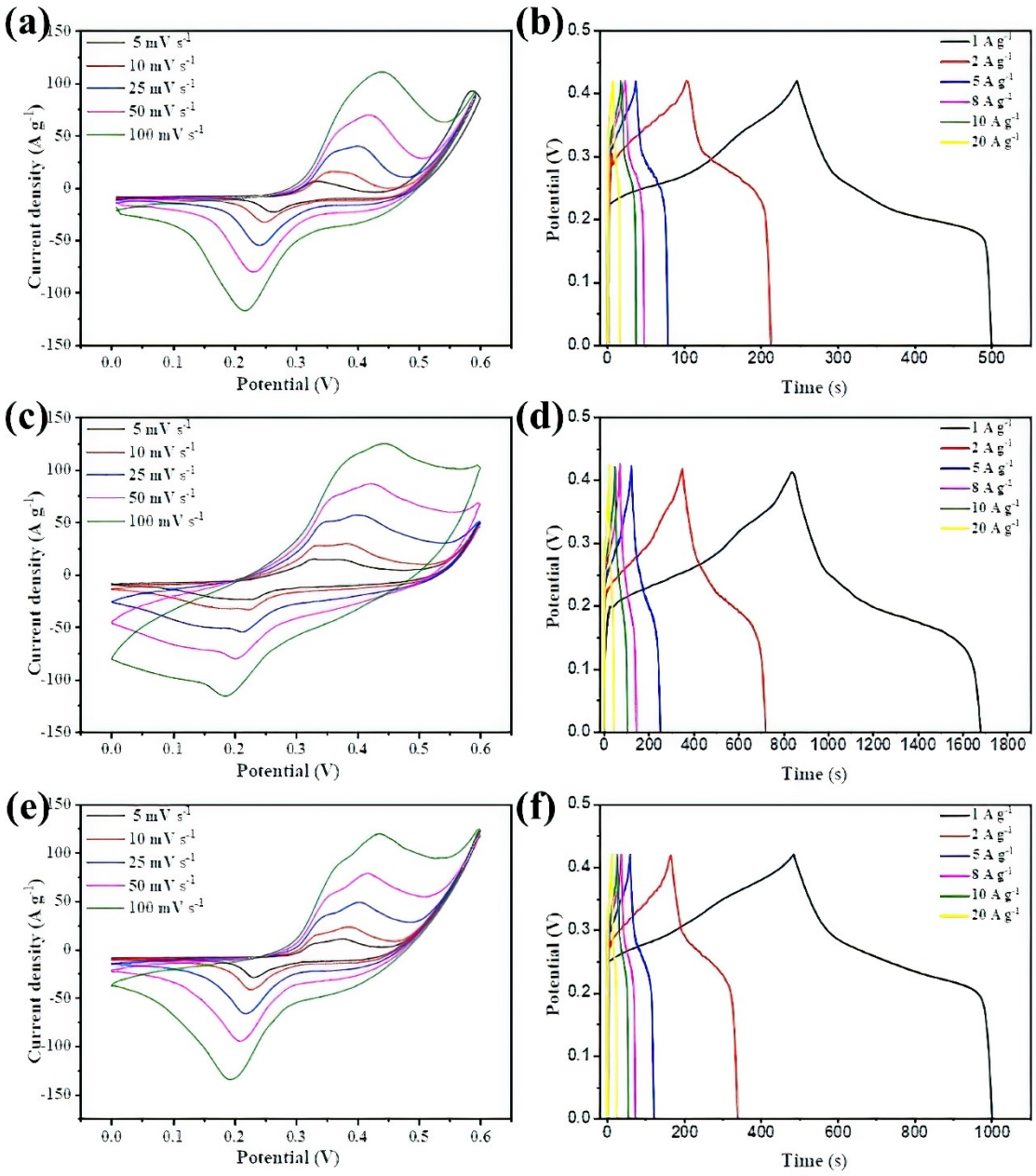

Fig. S6. (a, c, e) CV curves at various scan rates and (b, d , f) GCD curves at various densities of Co-Ni-Fe/OH, Co-Ni-Mn/S, and Co-Ni-Fe/S.

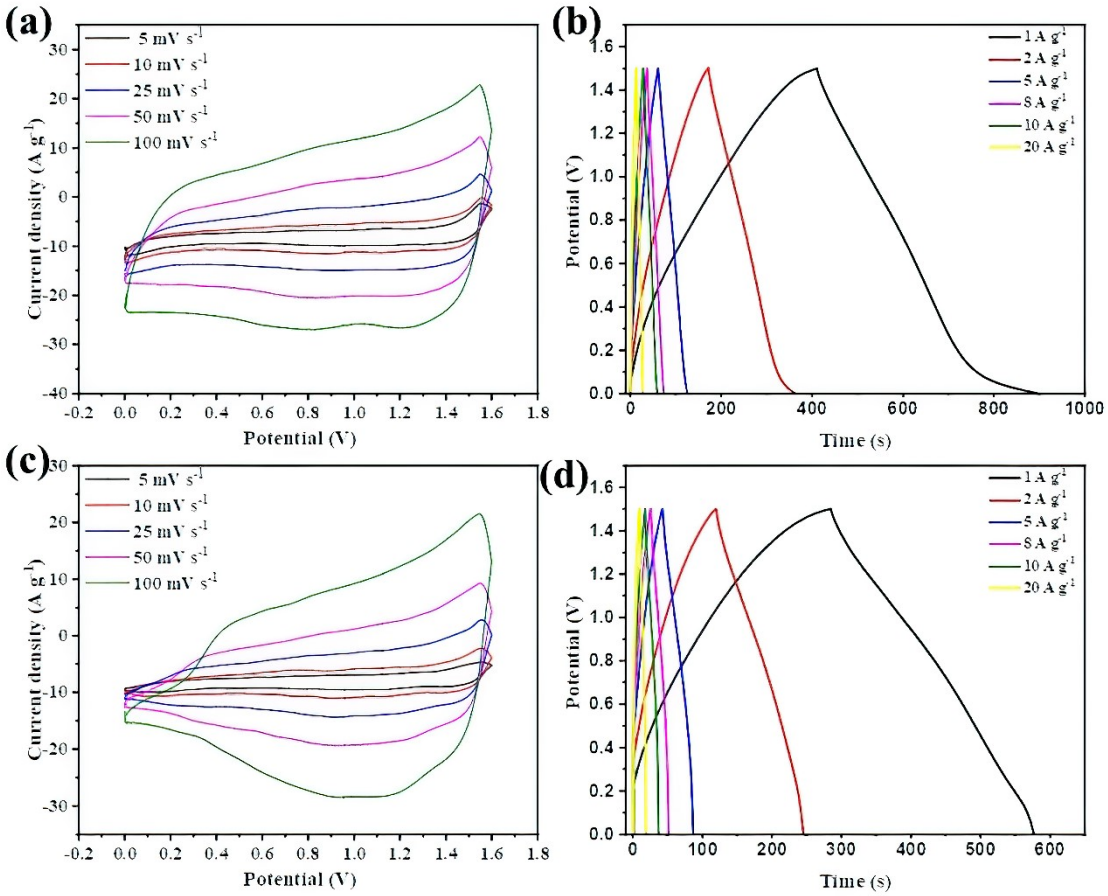

Fig. S7. (a, c) CV curves at various scan rates and (b, d) GCD curves at various densities of Co-Ni-Mn/S//AC and Co-Ni-Fe/S//AC ASC devices.

Supplementary Table 1. Metal contents in different samples determined by ICP-OES.

NO.	Samples	Element	Theoretical values (wt.%)	Measured values by ICP-OES (wt.%)
1	Co-Ni-Cu/S	Co	1.00 %	0.89 %
		Ni	3.00 %	2.76 %
		Cu	1.00 %	0.92 %
2	Co-Ni-Mn/S	Co	1.00 %	0.92 %
		Ni	3.00 %	2.88 %
		Fe	1.00 %	0.74 %
3	Co-Ni-Fe/S	Co	1.00 %	0.94 %
		Ni	3.00 %	2.79 %
		Mn	1.00 %	0.83 %