Supplementary Information (SI) for New Journal of Chemistry.
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Supporting Information

High-performance asymmetric supercapacitor electrode materials based on $CoSe_2$@NiCo-LDH@Ni Foam

Yan Gu [a*], XiaoShan Liu[a], ShuYan Xie[b], HanXin Chen[a], Hao Guo[a], YunYi Feng[a]

a College of Material Science and Engineering, Yangtze Normal University，Chongqing 408100, People's Republic of China.

b the Faculty of Education, Yunnan Normal University, Yunnan 650500, People's Republic of China.

Corresponding author: Yan Gu

Tel: +86 023 72792223

E-mail address: guyan1000@ccut.edu.cn

Experimental section

Materials

All substances, including $Ni(NO_3)_2{\cdot}6H_2O$、$Co(NO_3)_2{\cdot}6H_2O$、$Cu(NO_3)_2{\cdot}4H_2O$、$C_4H_6N_2$、SeO_2 and $CoCl_2{\cdot}6H_2O$ were purchased from Aladdin and utilized without additional purification. And foamed nickel (NF) was provided by Zhihongxuan Electronic Materials Management Department, Yushan Town, Kunshan City, China. Clean the NF (1cm × 1.2cm) with 3m of HCl, ethanol and deionized water to remove impurities and oxide layers on the surface.

Preparation of NiCo-LDH@$CoSe_2$ heterostructure grown on a NF substrate

To prepare NiCo-LDH electrode material, 2 mmol of $Ni(NO_3)_2$ and 2 mmol of $Co(NO_3)_2$ were dissolved in 20 mL of CH_3OH and stirred at room temperature for 1 hour (named solution A). And 6 mmol of $C_4H_6N_2$ were dissolved in 20 mL of CH_3OH (named solution B). The mixed solution was poured into a high-pressure reactor (50 mL), and the foamed nickel was immersed in the mixed solution and sealed. The high-pressure reaction vessel was placed at 120 ℃ for 10 hours. The prepared material was washed with deionized water and dried at 70 ℃ for 12 hours. Subsequently, the obtained NiCo-LDH electrode material was electrodeposited in an $CoSe_2$ solution, which composed of 4 mmol of SeO_2 and 4 mmol of $CoCl_2$. The electrodeposition was carried out by constant potential deposition method with a voltage of -1 V and time of 200 s, 400 s, and 600 s. In the end, NiCo-LDH@$CoSe_2$ heterostructure was obtained.

Assembly of an Asymmetric Supercapacitor

An Asymmetric Supercapacitor was assembled using NiCo-LDH@$CoSe_2$ and

Activated carbon as the cathode and anode, respectively, with KOH serving as the electrolyte. And the cathode was prepared by a slurry coating method. In short, activated carbon, acetylene black and polyvinylidene fluoride are mixed in 2 ml NMP at a mass ratio of 8:1:1. After being completely dispersed, the slurry was dropped onto clean nickel foam (1cm x 1.2 cm) and dried at 80 °C.

Materials characterization

The experiment was conducted using an X-ray photoelectron spectroscopy (XPS) instrument (Thermo Fisher Scientific ESCALAB 250Xi). The analysis chamber was maintained at a vacuum level of 1 x 10^{-9} mBar, and the excitation source employed Al Kα radiation (hv = 1486.68 eV), with an X-ray spot size of 500 μm. The operating voltage was set to 15 kV with a filament current of 10 mA, and signal accumulation was performed over 5 to 10 cycles. The passing energy for the measurement was set at 30 eV, with a step size of 0.05 eV. Charge correction was performed using C 1s = 284.80 eV as the binding energy reference. And the XRD testing adopted SmartLab, Rigaku Corporation. The microstructure and morphology of different samples were tested by Scanning Electron Microscope（SEM; JEOL Ltd.）and transmission electron microscopy (TEM; Philips CM200 apparatus).

Calculation methods

The specific capacity (C_s, mF cm^{-2}) and specific capacitance (C_m, F g^{-1}) of the materials was calculated from the GCD curves using Eq. S1 and Eq. S2, as follow:

$$C_s = \frac{I \times \Delta t}{s \times \Delta V} \quad \text{(S1)}$$

$$C_m = \frac{I \times \Delta t}{m \times \Delta V} \quad \text{(S2)}$$

where *I* refer to the discharge current density (A g^{-1}); *m* represents the mass of active materials grown on NF for three-electrode configurations (g); *Δt* is the discharge time (s); and *ΔV* is the potential window (V).

In order to achieve the optimum mass ratio of as-prepared positive electrode to AC negative electrodes, we examined the charge balance using the given equation, $q^+ = q^-$. The mass of the positive electrode and negative electrode was tuned according to the following Eq. S3 and Eq. S4:

$$Q = C \times \Delta V \times m \quad \text{(S3)}$$

$$\frac{m^+}{m^-} = \frac{C_- \times \Delta V_-}{C_+ \times \Delta V_+} \quad \text{(S4)}$$

Where *C* is the specific capacity (investigated through the three-electrode system); + and – represents positive and negative electrodes.

The energy density (*E*, Wh kg^{-1}) and power density (*P*, W kg^{-1}) of the ASCs were investigated by Eq. S5 and Eq. S6, respectively, as follows:

$$E = \frac{C_m \times \Delta V^2}{2 \times 3.6} \quad \text{(S5)}$$

$$P = \frac{3600 \times E}{\Delta t} \quad \text{(S6)}$$

Typically, the current (i) measured at different scan rates (v) adheres to an empirical power-law expression, as represented by the following equation, where a and b are constants. The b-value takes on specific values, specifically 0.5 and 1, which are associated with diffusion-controlled and capacitance-controlled mechanisms, respectively.

$$i = av^b \tag{S7}$$

In this equation, the terms (k_1v) and ($k_2v^{1/2}$) correspond to the capacitive-controlled and diffusion-controlled capacities, respectively.[4]

$$i = k_1v + k_2v^{1/2} \tag{S8}$$

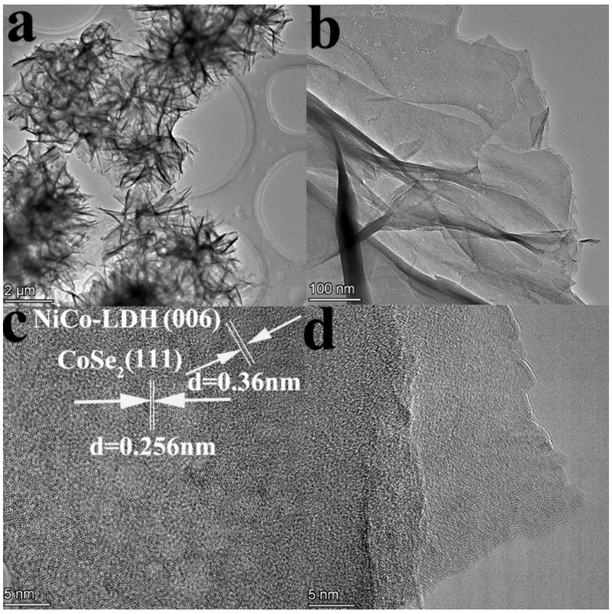

Fig. S1. TEM image of the composite material.

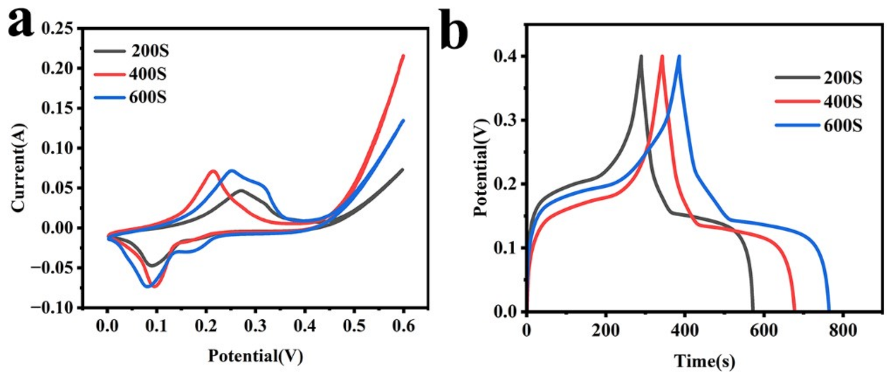

Fig. S2. CV versus GCD curves of composites with different deposition durations.

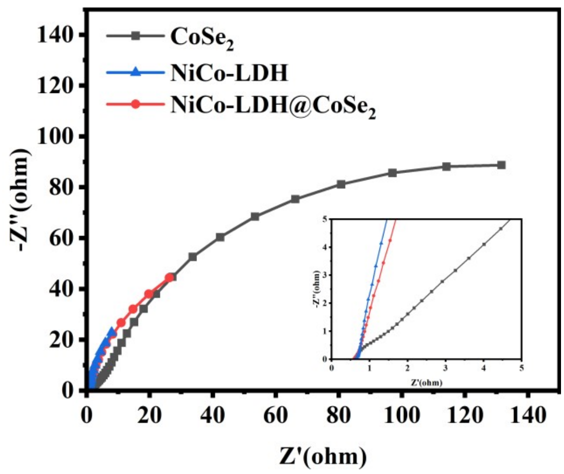

Fig. S3. Impedance testing of different materials.

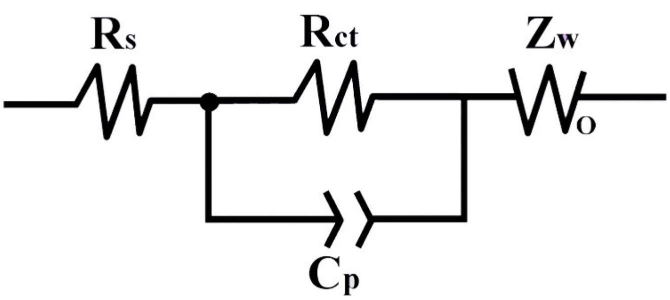

Fig. S4. Equivalent circuit diagram of the EIS curve.

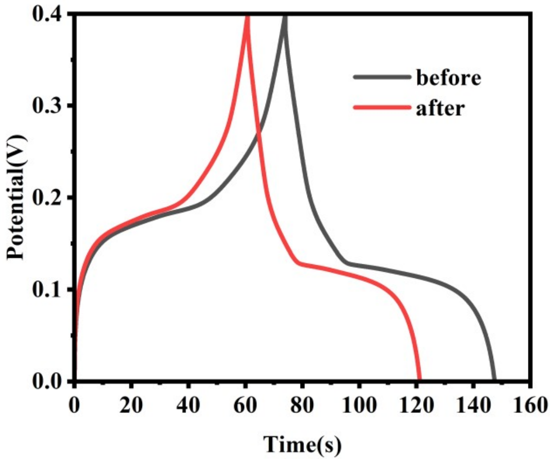

Fig. S5. GCD curves before and after cycling.

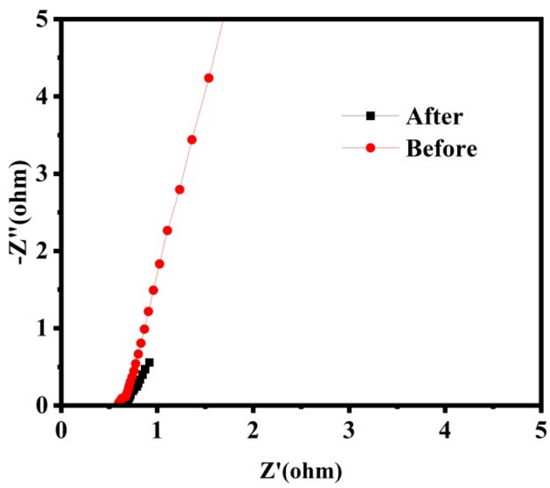

Fig. S6. Impedance curves before and after cycling.

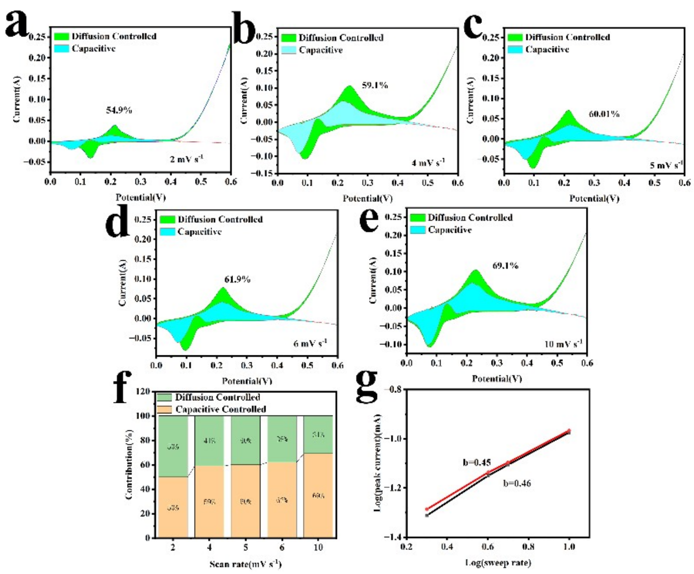

Fig. S7 Pseudocapacitance contribution ratio and b-value plot.

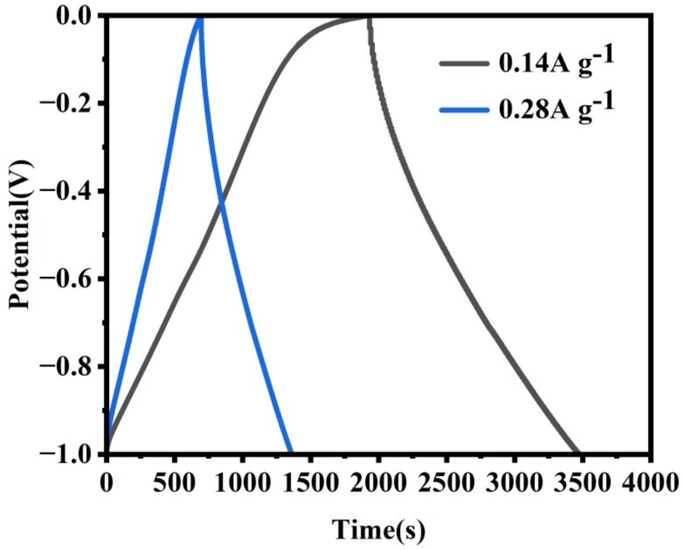

Fig. S8. GCD curve of activated carbon.

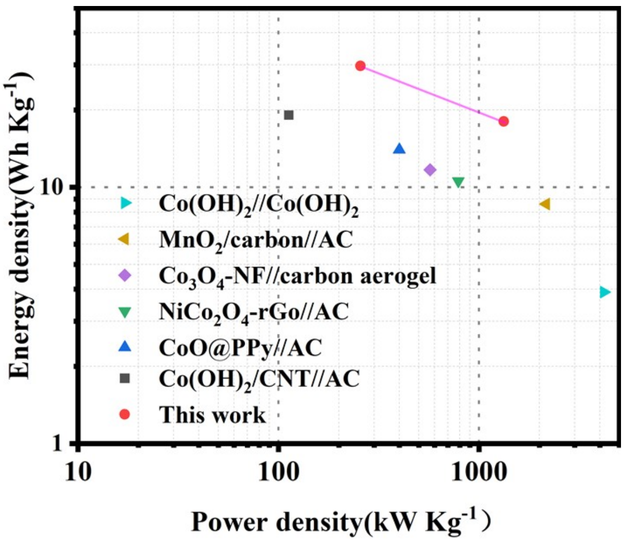

Fig. S9. Ragone plot of the device.

Table 1. Comparison of composite electrode materials with other electrode materials

Material	Fabrication Method	Specific Capacitance	Reference
$Ni_3(PO_4)_2 \cdot 8H_2O$	**hydrothermal**	**1053 F g^{-1}**	*ACS Applied Materials & Interfaces* **2022,** *14* (2), 2674-2682.
$FeMoO_4$	**hydrothermal and calcination**	**493 F g^{-1}**	*Ceramics International* **2022,** *48* (19), 29144-29151
NiCo-LDH@NCF	**hydrothermal**	**1512 F g^{-1}**	*Diamond and Related Materials* **2021,** *113*.
$NPC_{0.5}$-700	**carbonization**	**180.2 F g^{-1}**	*Chinese Chemical Letters* **2017,** *28* (12), 2227-2230.
$NiCo_2O_4$	**hydrothermal and calcination**	**667.6 F g^{-1}**	*Journal of Materials Science* **2022,** *57* (9), 5566-5576.
NPC	**double crucible method**	**261 F g^{-1}**	*RSC Advances* **2017,** *7* (18), 10901-10905.
Cu-Doped NiCo-LDH/CuO	**hydrothermal**	**1237 F g^{-1}**	Advanced Functional Materials, 2019, 29(24): 1809004.
Cu_2S	**combustion**	**677 F g^{-1}**	*Journal of Energy Storage* **2022,** *45*.
NiCo-LDH@$CoSe_2$	**hydrothermal and electrodeposition**	**1100 F g^{-1}**	**This work**