

## Supporting Information

### ZIF-67-derived NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P honeycomb nanosheets on carbon cloth for high-performance asymmetric supercapacitors

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## Summary

S1. Supporting experimental section.

S2. Supporting Figure S1~S11.

S3. Supporting Table S1.

### Electrochemical Calculation

#### Capacitances of single electrode

The areal capacitance of a single electrode in three-electrode system can be calculated based on galvanostatic charge-discharge experiments according to equations:

$$C_s = \frac{I\Delta t}{S\Delta V} \quad (1)$$

Where  $C_s$  ( $\text{F cm}^{-2}$ ) represents the areal capacitance,  $I$  is the discharge current (A),  $\Delta t$  represents the discharge time (s),  $S$  ( $\text{cm}^2$ ) is the apparent area of actives materials loaded in working electrode, and  $\Delta V$  is the potential window (V).

#### Capacitances of ASC devices

The volume specific capacitance of  $\text{NiCo}_2\text{O}_4@/\text{Co}_2\text{P}/\text{Ni}_2\text{P}-\text{CC}/\text{AC}/\text{CC}$  devices can be calculated based on galvanostatic charge-discharge experiments according to equations:

$$C_v = \frac{I\Delta t}{V\Delta V} \quad (2)$$

Where  $C_v$  ( $\text{F cm}^{-3}$ ) represents the volume specific capacitance,  $I$  is the discharge current (A),  $\Delta t$  represents the discharge time (s),  $V$  ( $\text{cm}^3$ ) represents the apparent volume of actives materials loaded in working electrode and  $\Delta V$  is the potential window (V).

In order to obtain more stable electrochemical performance, the positive and negative electrodes should follow the principle of equal capacitance. The relationship between positive and negative capacitance can be determined by the following equations:

$$Q = C_s \times \Delta V \times S \quad (3)$$

$$Q_+ = Q_- \quad (4)$$

$$\frac{C_{s,-}}{C_{s,+}} = \frac{S_+ \times \Delta V_+}{S_- \times \Delta V_-} \quad (5)$$

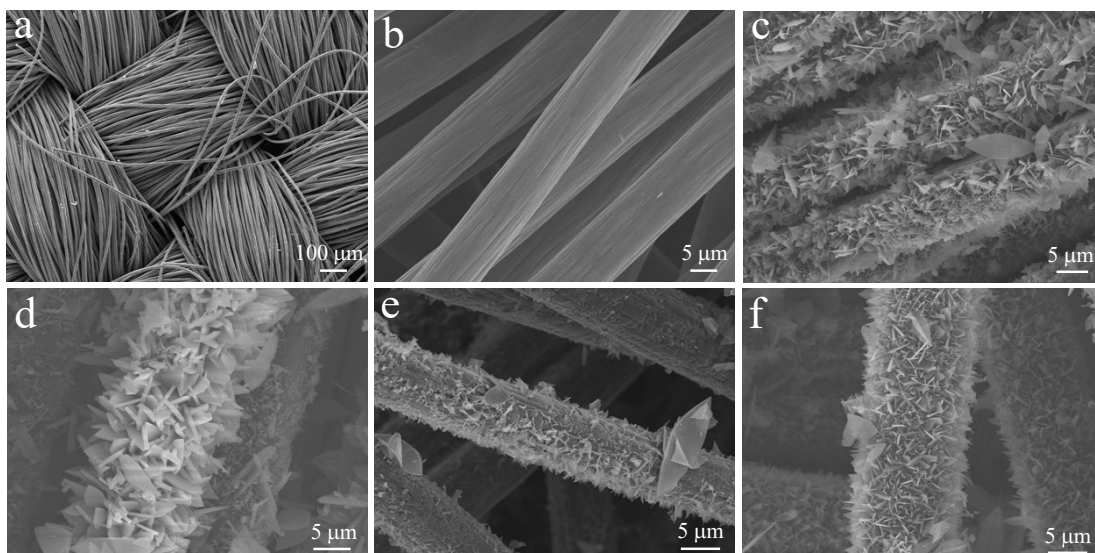
Where  $Q$  is the charge of the electrode,  $C_s$  represents the areal capacitance of the electrode,  $\Delta V$  represents the potential window of the electrode, and  $S$  is the area of the electrode material. In this work, the active area of AC/CC and NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC was 1 cm<sup>2</sup>. In addition,  $\Delta V_- = 1$  V and  $\Delta V_+ = 0.6$  V. According to the calculation (5), the ratio of areal capacitance of AC/CC and NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC was 0.6. Therefore, the areal capacitance of AC/CC should be adjusted to 1729.13 mF cm<sup>-2</sup> at a current density of 2 mA cm<sup>-2</sup>.

The energy density ( $E$ , mWh cm<sup>-3</sup>) and power density ( $P$ , mW cm<sup>-3</sup>) are calculated by the following equations:

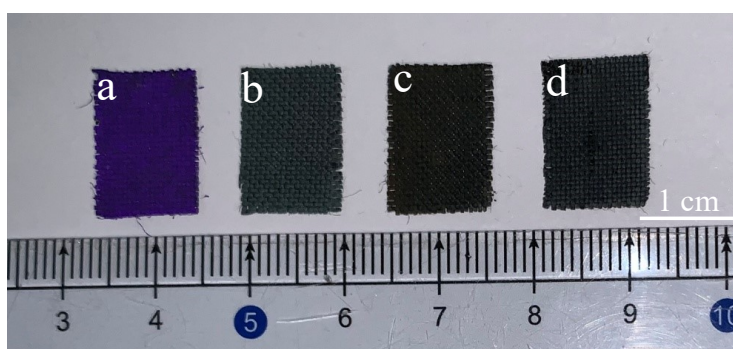
$$E = \frac{\int IV dt}{V_v} \quad (6)$$

$$P = \frac{E}{\Delta t} \quad (7)$$

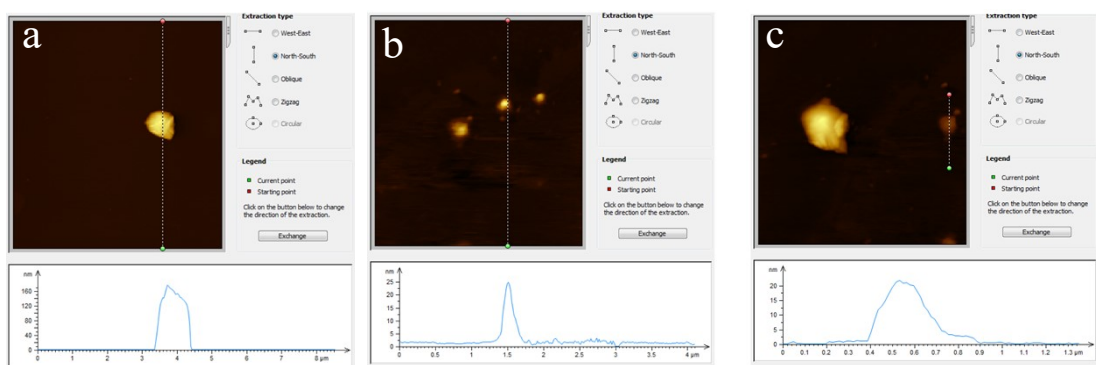
Where  $V$  represents the potential window,  $V_v$  represents the volume of device and  $\Delta t$  represents the discharge time.



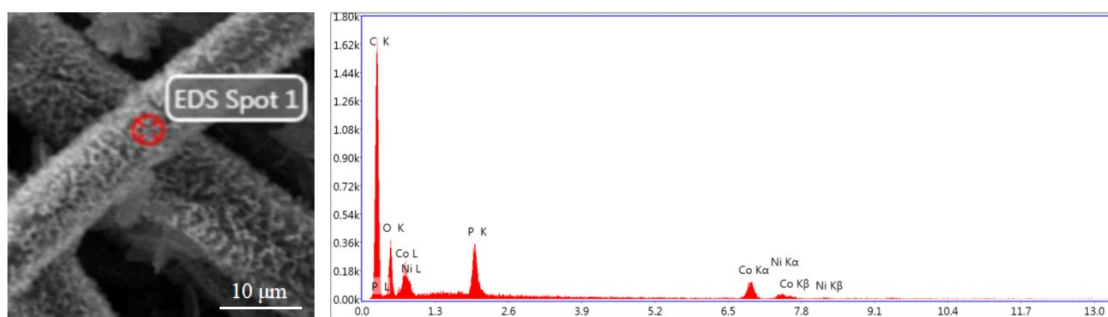
**Figure S1.** (a, b) SEM images of CC. (c) SEM images of ZIF-67-CC- I . (d) SEM images of ZIF-67-CC- II . (e) SEM images of ZIF-67-CC-III. (f) SEM images of ZIF-67-CC-IV.



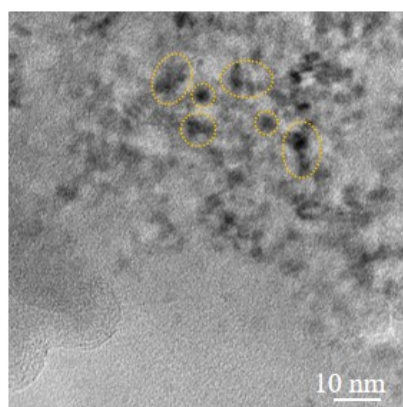
**Figure S2.** (a) Picture of ZIF-67-CC. (b) Picture of Co-Ni LDH-CC. (c) Picture of NiCo<sub>2</sub>O<sub>4</sub>-CC. (d) Picture of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC.



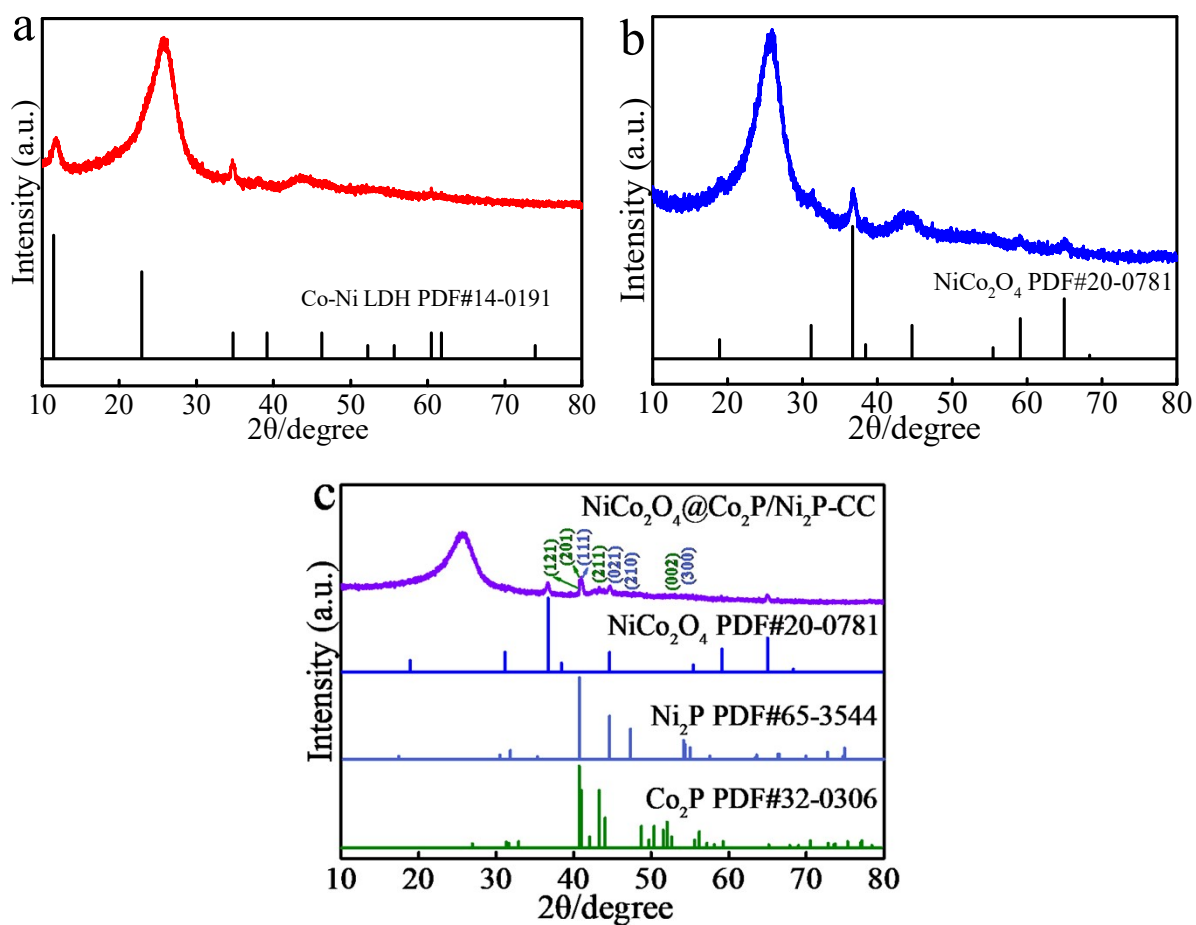
**Figure S3.** (a) AFM images of ZIF-67-CC. (b) AFM images of Co-Ni LDH-CC. (c) AFM images of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC.



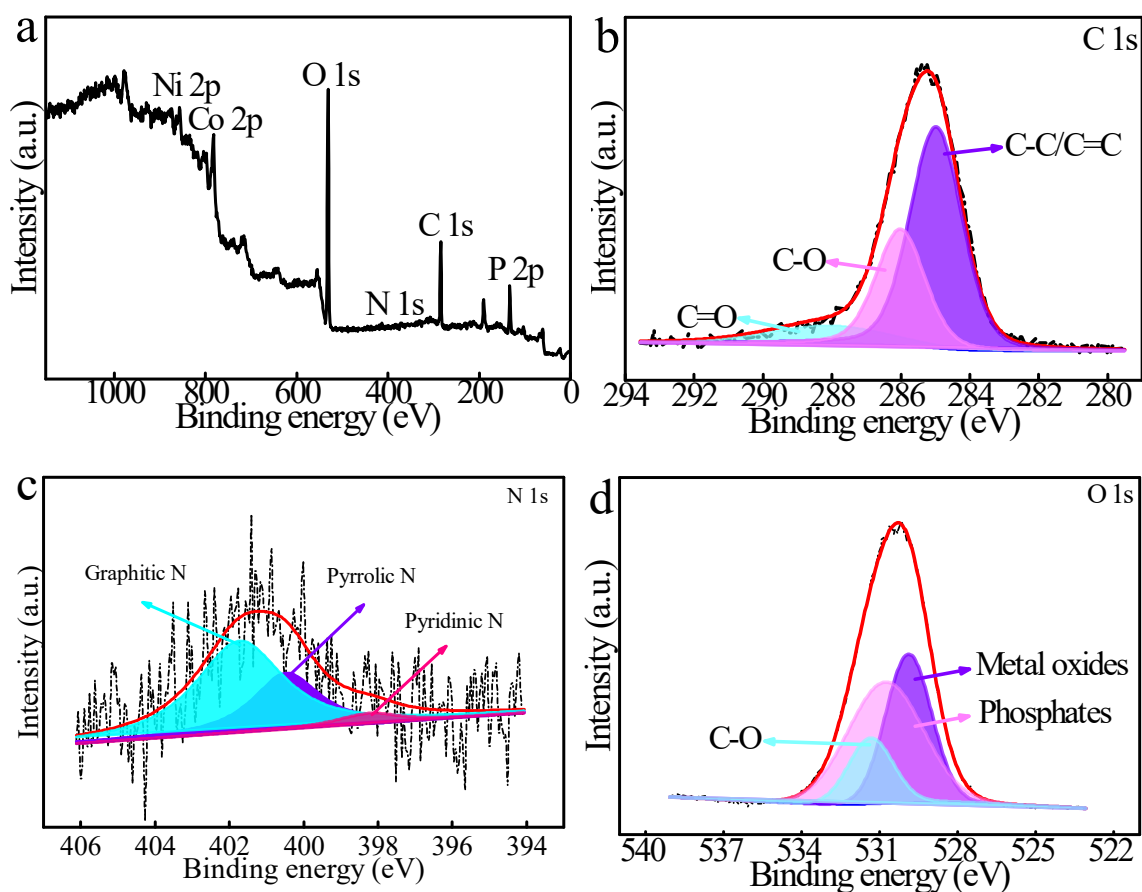
**Figure S4.** EDS point scan test results of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC.



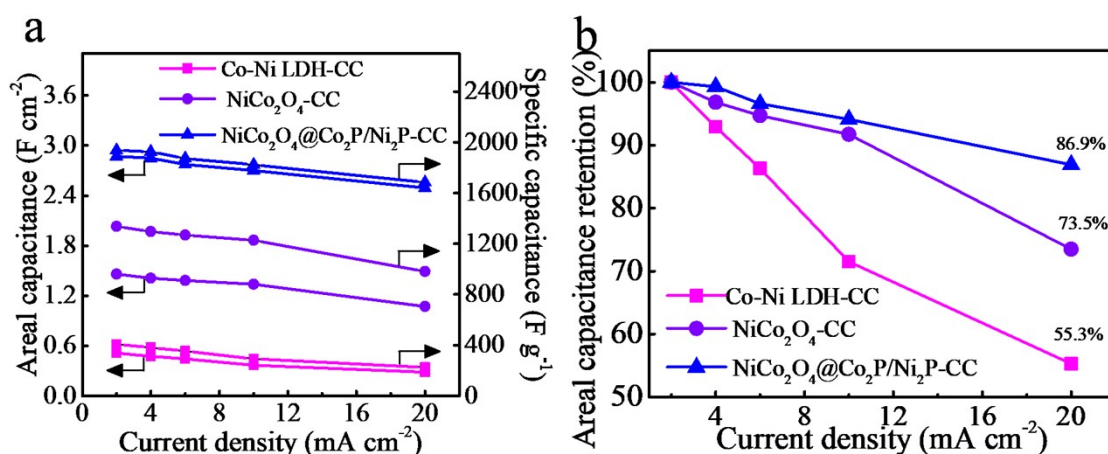
**Figure S5.** HRTEM images of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC.



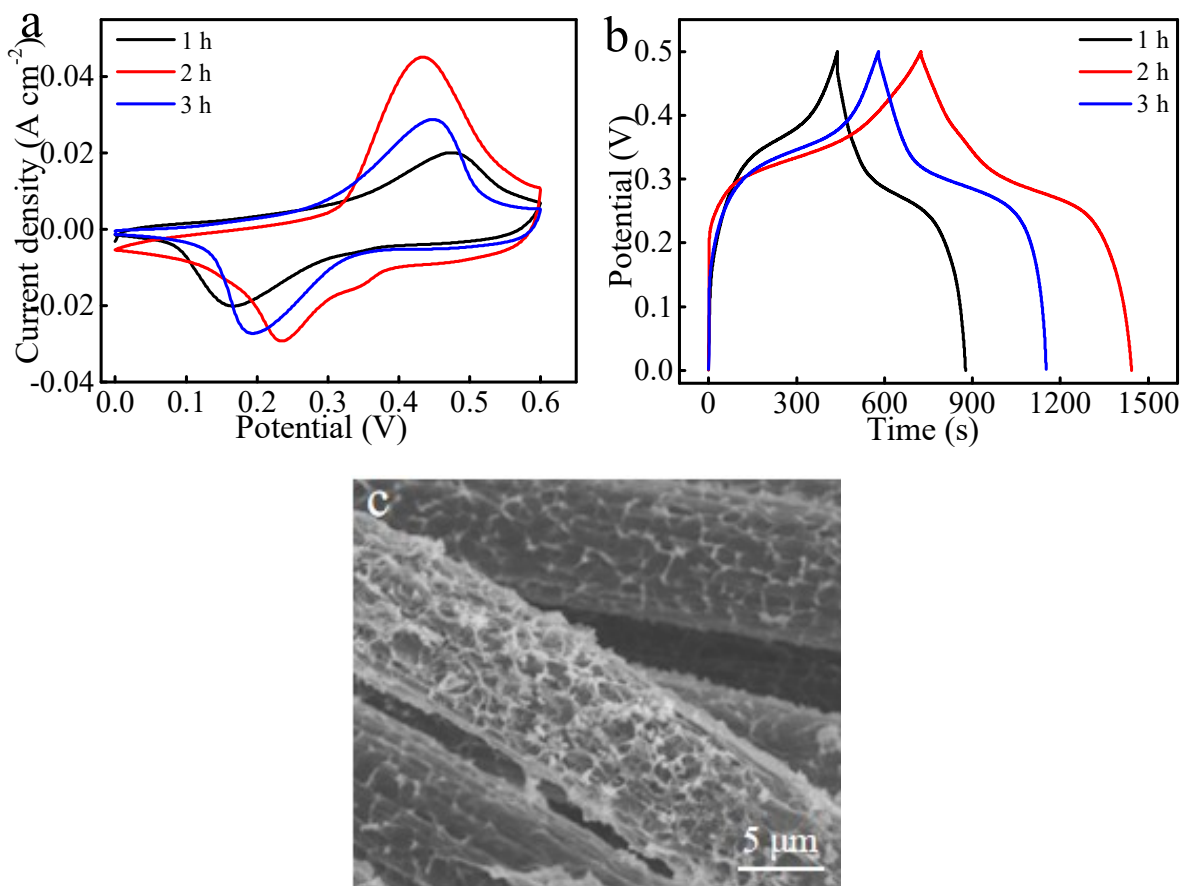
**Figure S6.** (a) XRD patterns of Co-Ni LDH-CC. (b) XRD patterns of NiCo<sub>2</sub>O<sub>4</sub>-CC. (c) XRD patterns of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC.



**Figure S7.** (a) Survey spectra of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC. (b, c and d) C 1s, N 1s and O 1s high-resolution XPS spectra of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC.

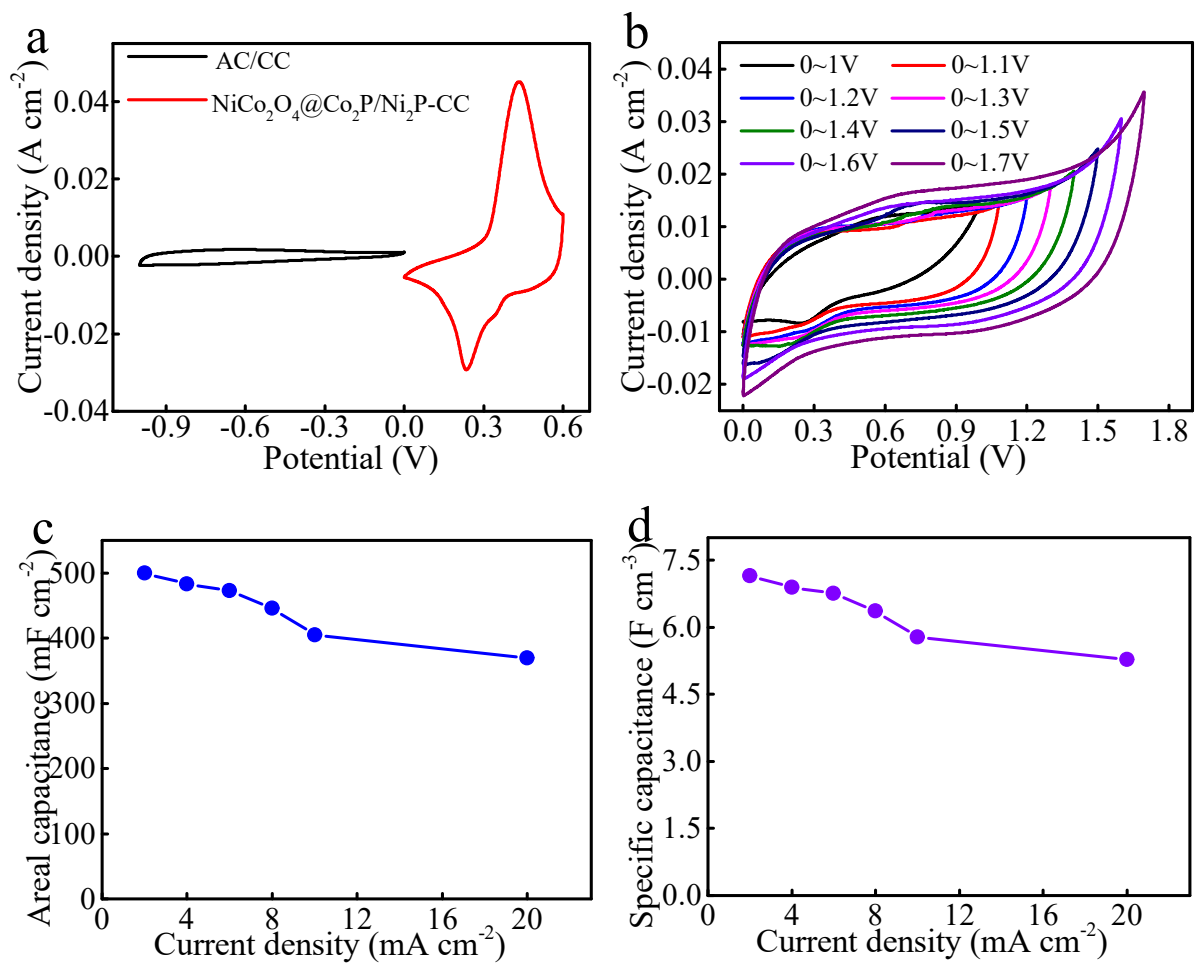


**Figure S8.** (a) The areal capacitance and specific capacitance of Co-Ni LDH-CC, NiCo<sub>2</sub>O<sub>4</sub>-CC and NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC at different current density from 2 to 20 mA cm<sup>-2</sup>. (b) The areal capacitance retention of Co-Ni LDH-CC, NiCo<sub>2</sub>O<sub>4</sub>-CC and NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC at different current density from 2 to 20 mA cm<sup>-2</sup>.

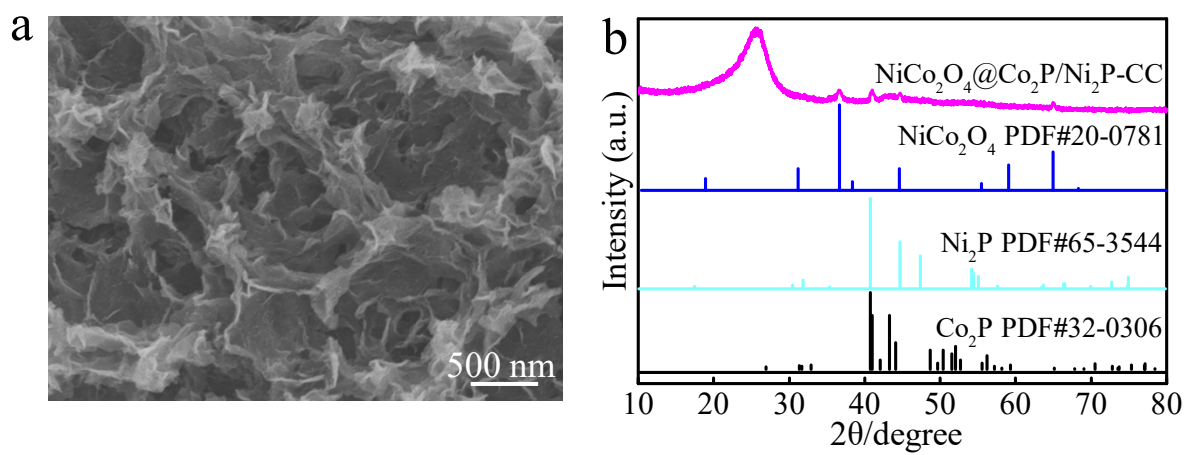


**Figure S9.** (a) CV curves of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC on different phosphatization time from 1 h to 3 h at a scan rate of 10 mV s<sup>-1</sup>. (b) GCD curves of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC on different phosphatization time from 1 h to 3 h at a current density of 2 mA cm<sup>-2</sup>. (c) SEM images of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC on phosphatization time of 3 h.





**Figure S10.** (a) CV curve of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC and AC/CC at scan rate of 10 mV s<sup>-1</sup>. (b) CV curves of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC and AC/CC at different voltage window at a scan rate of 10 mV s<sup>-1</sup>. (c) Areal capacitance of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC/AC/CC at different current from 2 to 20 mA cm<sup>-2</sup>. (d) Specific capacitance of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC/AC/CC at different current density from 2 to 20 mA cm<sup>-2</sup>.



**Figure S11.** (a) SEM and (b) XRD images of  $\text{NiCo}_2\text{O}_4@\text{Co}_2\text{P}/\text{Ni}_2\text{P-CC}$  electrode material after 10000 cycles by charge-discharge cycle at 2 mA  $\text{cm}^{-2}$ .

**Table S1.** Comparison of NiCo<sub>2</sub>O<sub>4</sub>@Co<sub>2</sub>P/Ni<sub>2</sub>P-CC with various Ni/Co based electrodes materials for supercapacitor.

Type	Morphology	Electrolyte	Scan rate/ current density	Capacitance	Ref.
CoO@MnO <sub>2</sub>	Nanosheets arrays	6 M KOH	2 mA cm <sup>-2</sup>	2.4 F cm <sup>-2</sup>	1
Zn-Ni-Co TOH	Nanowire array	1 M KOH	3 mA cm <sup>-2</sup>	2.14 F cm <sup>-2</sup>	2
CoO/Co <sub>9</sub> S <sub>8</sub> @CN	Nanocage cluster	6 M KOH	0.5 A g <sup>-1</sup>	303.3 F g <sup>-1</sup>	3
Ni(OH) <sub>2</sub>	Nanosheets	6 M KOH	5 mA cm <sup>-2</sup>	0.863 F cm <sup>-2</sup>	4
Ni Co-LDH@Au- CuO/Cu	Array	3 M KOH	1.5 mA cm <sup>-2</sup>	1.97 F cm <sup>-2</sup>	5
NiNW@NiCo-DH/NF	Nanosheets @Nanowire	6 M KOH	5 mA cm <sup>-2</sup>	2.25 F cm <sup>-2</sup>	6
NiCo <sub>2</sub> O <sub>4</sub> @MnO <sub>2</sub>	Nanosheets	1 M KOH	5 mA cm <sup>-2</sup>	2.85 F cm <sup>-2</sup>	7
Co-Ni LDH-CC	Honeycomb nanosheets	6 M KOH	2 mA cm <sup>-2</sup>	0.512 F cm <sup>-2</sup>	<b>This work</b>
NiCo <sub>2</sub> O <sub>4</sub> -CC	Honeycomb nanosheets	6 M KOH	2 mA cm <sup>-2</sup>	1.458 F cm <sup>-2</sup>	<b>This work</b>
NiCo <sub>2</sub> O <sub>4</sub> @Co <sub>2</sub> P/Ni <sub>2</sub> P- CC	Honeycomb nanosheets	6 M KOH	2 mA cm <sup>-2</sup>	2.88 F cm <sup>-2</sup>	<b>This work</b>

## References

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