Supporting information

1. Method

1.1 Materials

All chemicals were used without any further purification. Terephthalic acid (H_2BDC), Sodium sulfate (Na_2SO_4), Sodium hydroxide (NaOH) and Nickel(II) nitrate hexahydrate ($Ni(NO_3)_2 \cdot 6H_2O$) were acquired from Tianjin Yongda Chemical Reagents. N,N-Dimethylformamide (DMF) and Carboxymethylcellulose (CMC) were purchased from Sinopharm Chemical Reagent Co., Ltd.

1.2 Synthesis of C-Ni/NiO/NiBDC

3 mmol H_2BDC and 6 mmol NaOH were dissolved in 15 mL DI water, and then 265 mg CMC was added into the solution. The above mixture was stirred for 3 h at room temperature. The mixture was precooled in the refrigerator, and then was treated by freeze-drying process for 8 hours. Subsequently, 5 mmol $Ni(NO_3)_2 \cdot 6H_2O$ and the obtained precursor were put into 10 mL DMF and transferred into Teflon-lined stainless steel autoclave at 140 °C for 12 hours. After cooling down the room temperature, the samples were carefully rinsed several times with DMF and ethanol, and dried at 60 °C under vacuum. Finally, the samples were calcined at 350 °C for 3 h, the obtained samples were donated as C-Ni/NiO/NiBDC.

1.3 Synthesis of C-Ni-CMC

For comparison, the similar synthesis procedure was carried out merely the

different sequence of ligands and metal ions. Typically, 5 mmol Ni(NO$_3$)·6H$_2$O and 6 mmol NaOH were dissolved in 15 mL DI water, and then 265 mg CMC was added into the solution. The above mixture was stirred for 3 h at room temperature. The mixture was precooled in the refrigerator, and then was treated by freeze-drying process for 8 hours. Subsequently, 3 mmol H$_2$BDC and the obtained precursor were put into 10 mL DMF and transferred into Teflon-lined stainless steel autoclave at 140 °C for 12 hours. After cooling down to the room temperature, the samples were carefully rinsed several times with DMF and ethanol, and dried at 60 °C under vacuum. Finally, the samples were calcined at 350 °C for 3 h, the obtained samples were donated as C-NiCMC.

1.4 Synthesis of C-NiBDC

0.7 mmol H$_2$BDC and 0.7 mmol Ni(NO$_3$)·6H$_2$O were dissolved in 10 mL DMF and 0.6 mL H$_2$O, the solution was stirred for 15 min at room temperature to form a homogenous solution. Then, the above solution was transferred into a Teflon-lined stainless autoclave and maintained at 140 °C for 12 hours. After cooling down to room temperature, the sample was washed with DMF and ethanol and dried at 60 °C under vacuum. The samples were calcined at 350 °C for 3 h, the obtained samples were donated as C-NiBDC.

1.5 Synthesis of C-NiBDC-CMC

The as-prepared NiBDC was dispersed in 15mL DI water, and then 265 mg CMC was added into the solution. The above solution was stirred for 3 h at room temperature. The mixture was precooled in the refrigerator, and then was treated by freeze-drying process for 8 hours. The samples were calcined at 350 °C for 3 h, the obtained samples were donated as C-NiBDC-CMC.

1.6 Characterization

The morphology, compositions and size of the all samples were investigated using a scanning electron microscope (SEM, HITACHI, SU8010, Japan). Transmission electron microscopy (TEM) images and high-resolution transmission electron microscopy (HRTEM) images were taken on a Tecnai G2 F20 S-Twin electron microscope. The crystal structures of the samples were characterized by an X-ray powder diffractometer (XRD, PANalytical, X'Pert Pro, Netherlands). XPS measurements were performed on a Thermo K-Alpha XPS instrument (Thermo Fisher Scientific, USA).

1.7 Electrochemical measurements

All electrochemical tests were performed on the Shanghai Chenhua 660E workstation. The as-prepared samples were used as working electrode, saturated calomel as the reference electrode, Pt foil as the counter electrode, and 1 M Na_2SO_4 as the electrolyte. The as-prepared samples were electrochemically tested in a three-electrode system by means of

cyclic voltammetry (CV), galvanostatic charge/discharge (GCD) and electrochemical impedance spectroscopy (EIS). The method for preparing electrode is as follows: the as-prepared samples, PVDF and super P in the ratio of 7:2:1 were mixed and ground, and then the N-methyl-2-pyrrolidone(NMP) was added to form slurry. Afterwards, the slurry was coated on the graphite foil (1×1 cm^2), and the obtained graphite foil was dried at 80° for 8 hours. The mass of each electrode was around 0.7 mg.

The specific capacitance of the supercapacitors can be calculated by galvanostatic discharge-charge (GCD) test as following equation:

$$C_m = \frac{I * \Delta t}{m * \Delta V}$$

Where C_m is the specific capacitance of the capacitors (F g^{-1}); I is the current of the charge/discharge process; Δt is the discharging time period for the potential changeΔV; m is the mass loading of the active material on the working electrode.

Energy density and power density of the supercapacitors can be calculated as following equation:

$$E = \frac{1}{2} C \Delta V^2$$

$$P = \frac{E}{\Delta t}$$

Where E (Wh kg^{-1}) is the energy density, C (F g^{-1}) is the mass specific capacitance, ΔV (V) is the voltage window, P (W kg^{-1}) refers to the power
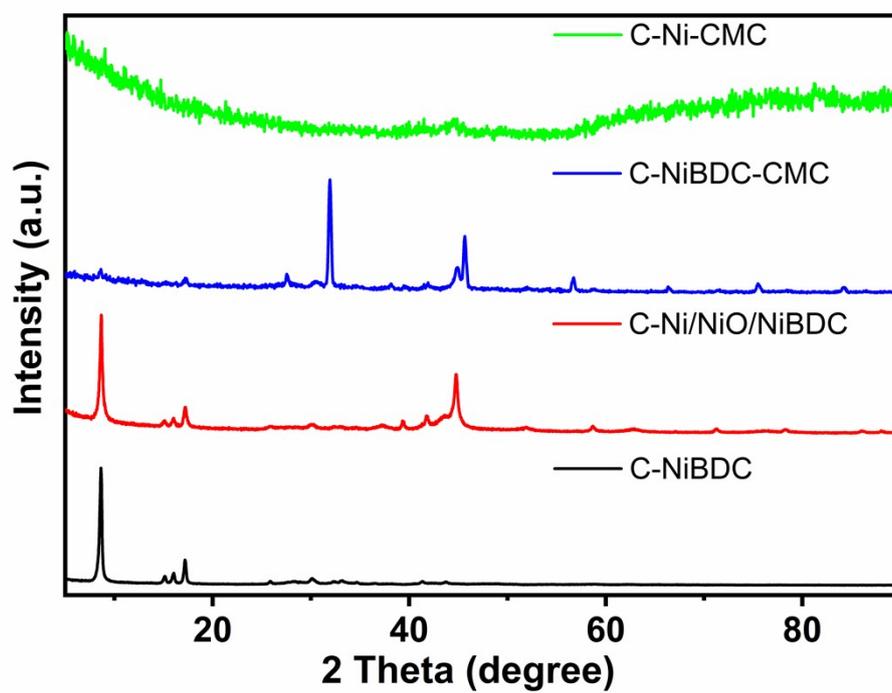
density and △t is the discharge time.

Fig. S1. XRD patterns of C-Ni/NiO/NiBDC, C-NiBDC, C-Ni-CMC and C-NiBDC-CMC.
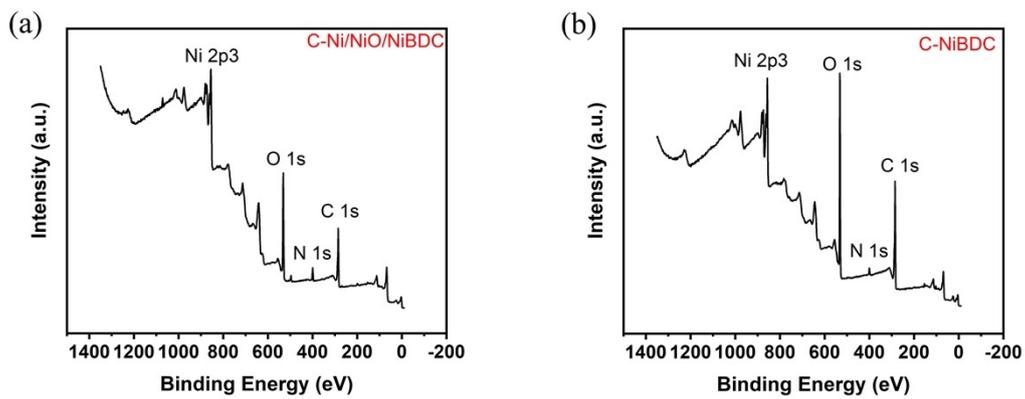
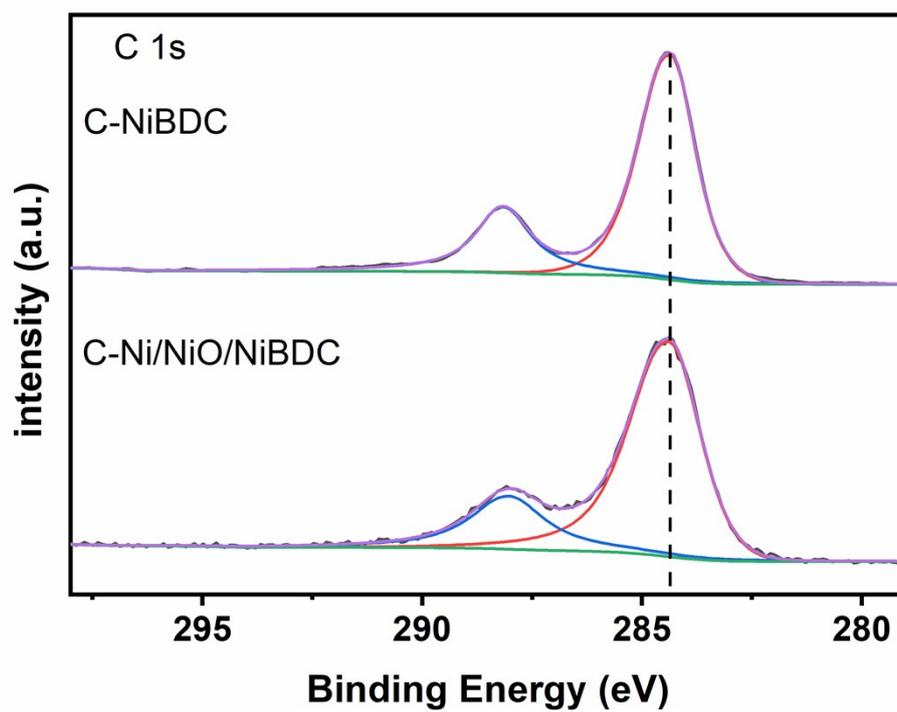**Fig. S2.** XPS survey spectra of C-Ni/NiO/NiBDC (a) and C-NiBDC (b)

Fig. S3. High-resolution XPS spectra of C 1s for C-Ni/NiO/NiBDC and C-NiBDC.
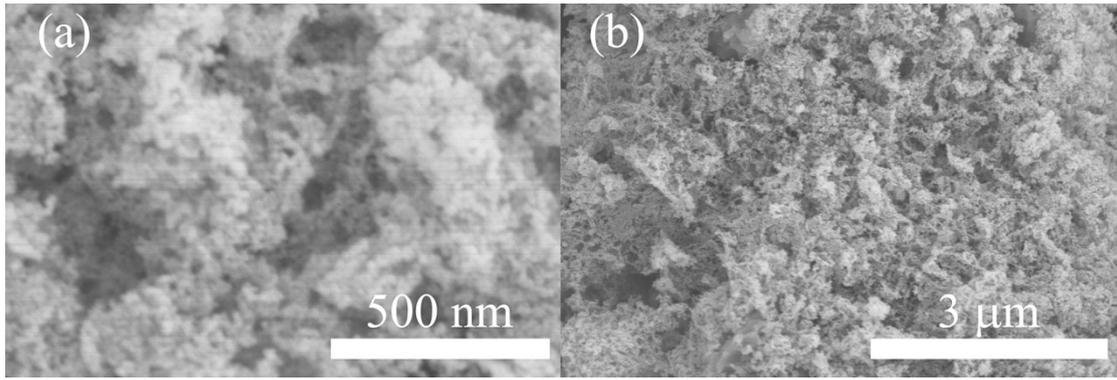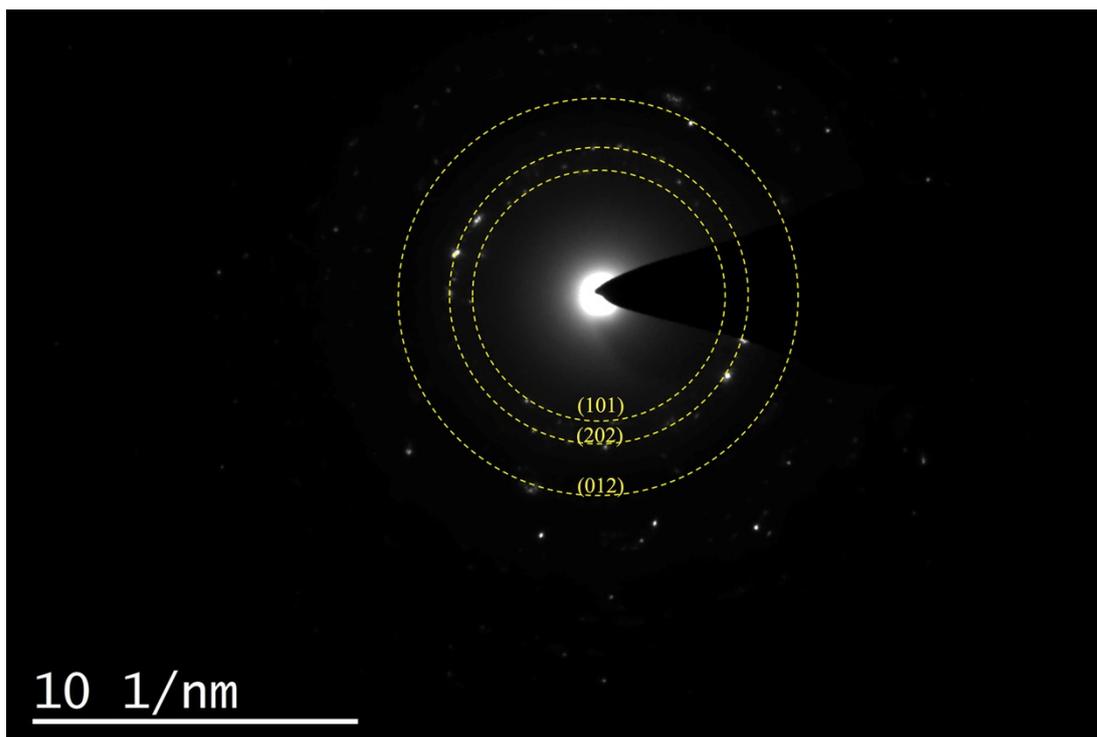
Fig. S4. SEM images of C-NiBDC.
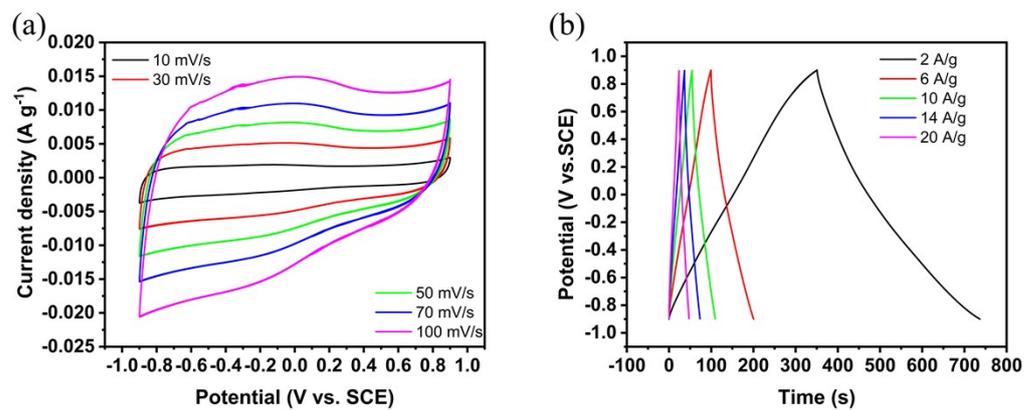
Fig. S5. SAED pattern of the C-Ni/NiO/NiBDC.

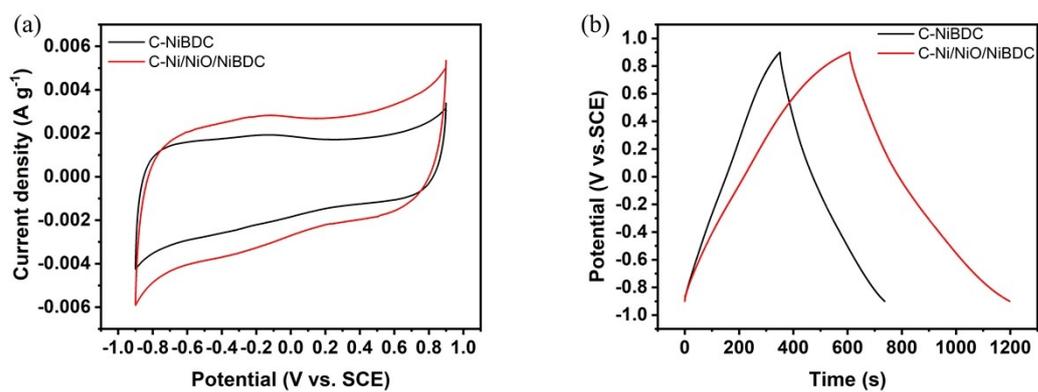**Fig. S6.** (a) CV curves and (b) GCD curves of C-NiBDC.

Fig. S7. (a) CV curves at 10 mV s^{-1} and (b) GCD curves at 2 A g^{-1} of C-NiBDC and C-Ni/NiO/NiBDC.
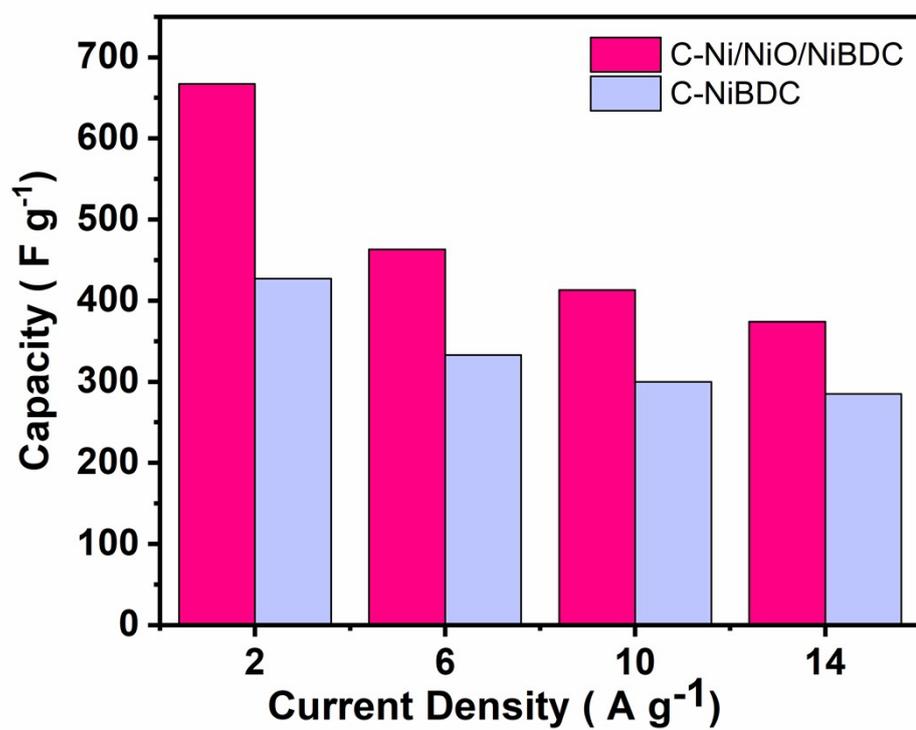
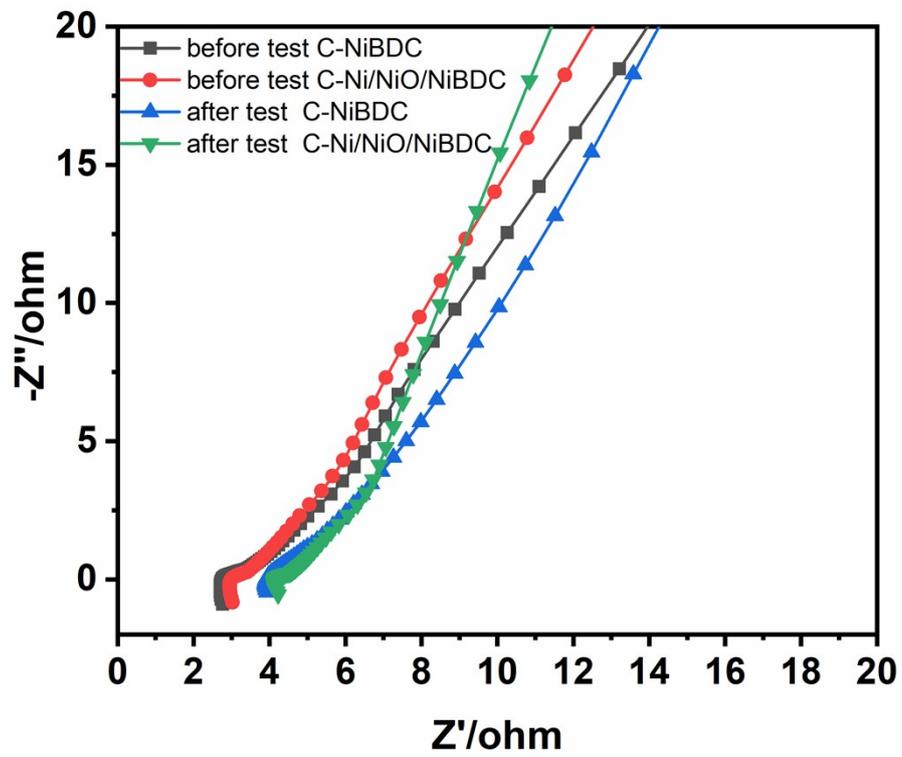**Fig. S8.** Rate performance of C-Ni/NiO/NiBDC and C-NiBDC

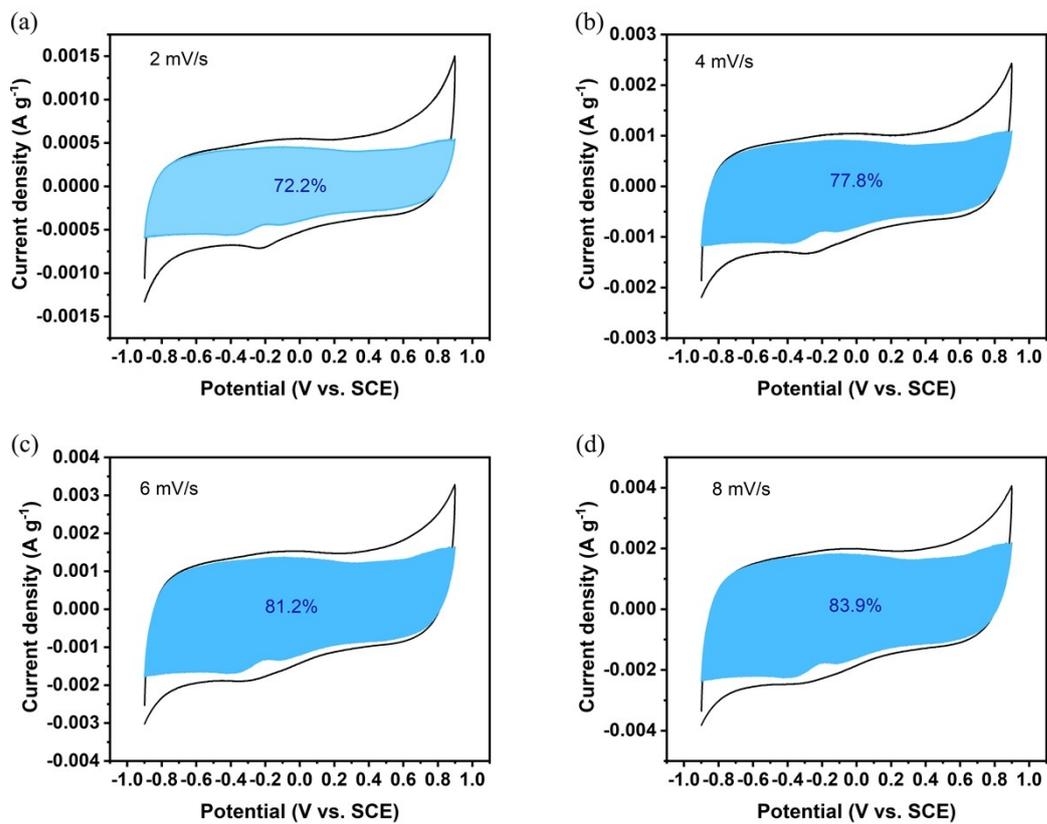**Fig. S9.** Nyquist plots of before and after C-Ni/NiO/NiBDC and C-NiBDC testing

Fig. S10. Capacitive and diffusion of contribution of C-Ni/NiO/NiBDC at dfferent scan rates: 2 mV s^{-1}(a); 4 mV s^{-1}(b); 6 mV s^{-1} (c) and 8 mV s^{-1}(d).
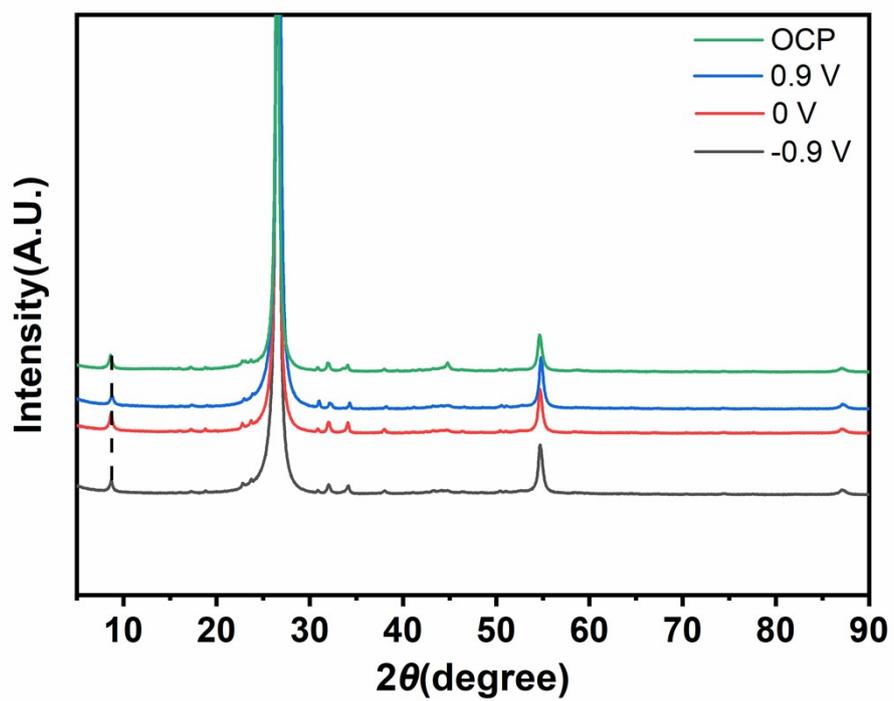
Fig. S11. XRD patterns of C-Ni/NiO/NiBDC during the discharge process.
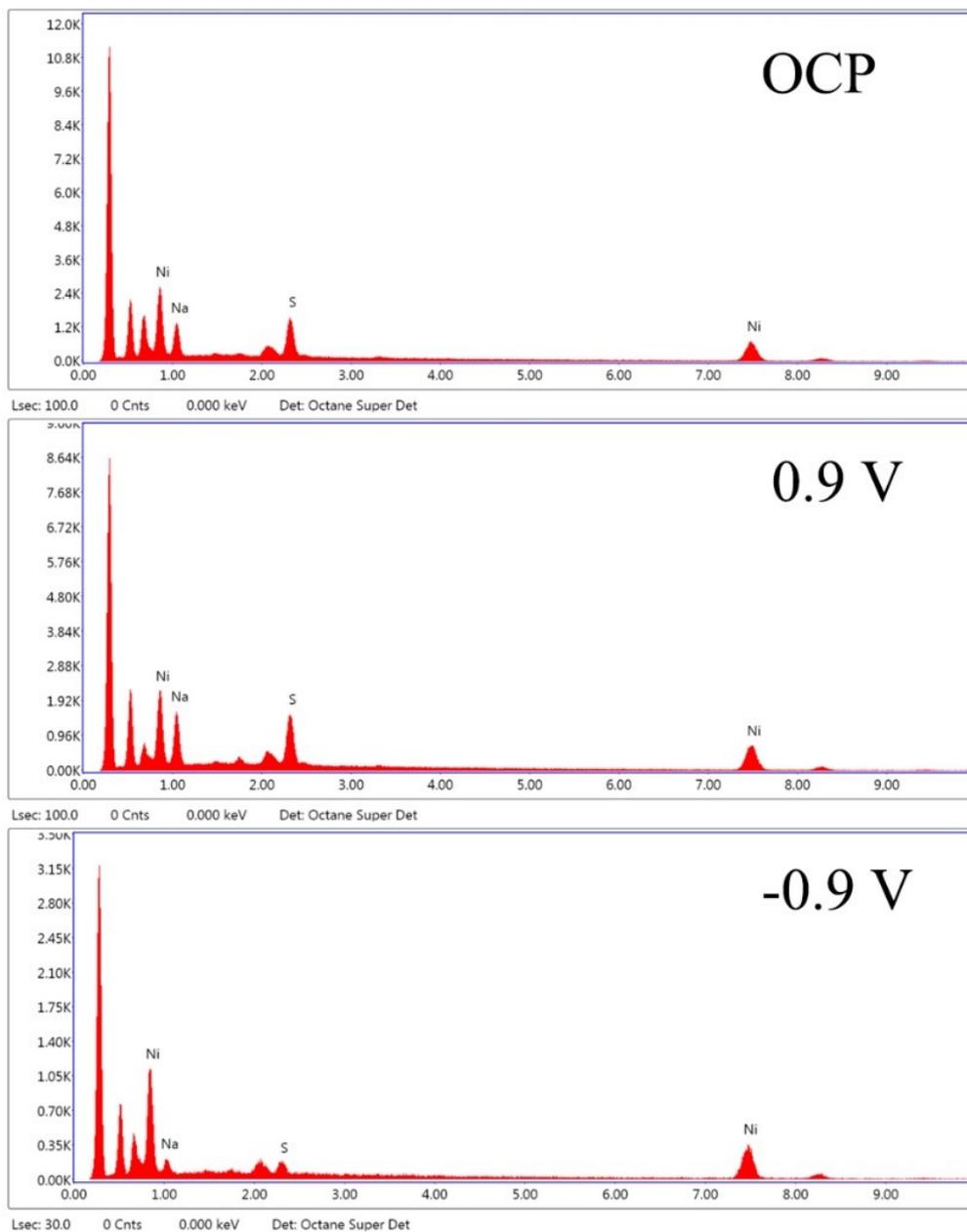
Fig. S12. EDX of C-Ni/NiO/NiBDC during the discharge process

Table S1. Element proportion of C-Ni/NiO/NiBDC during the discharge process

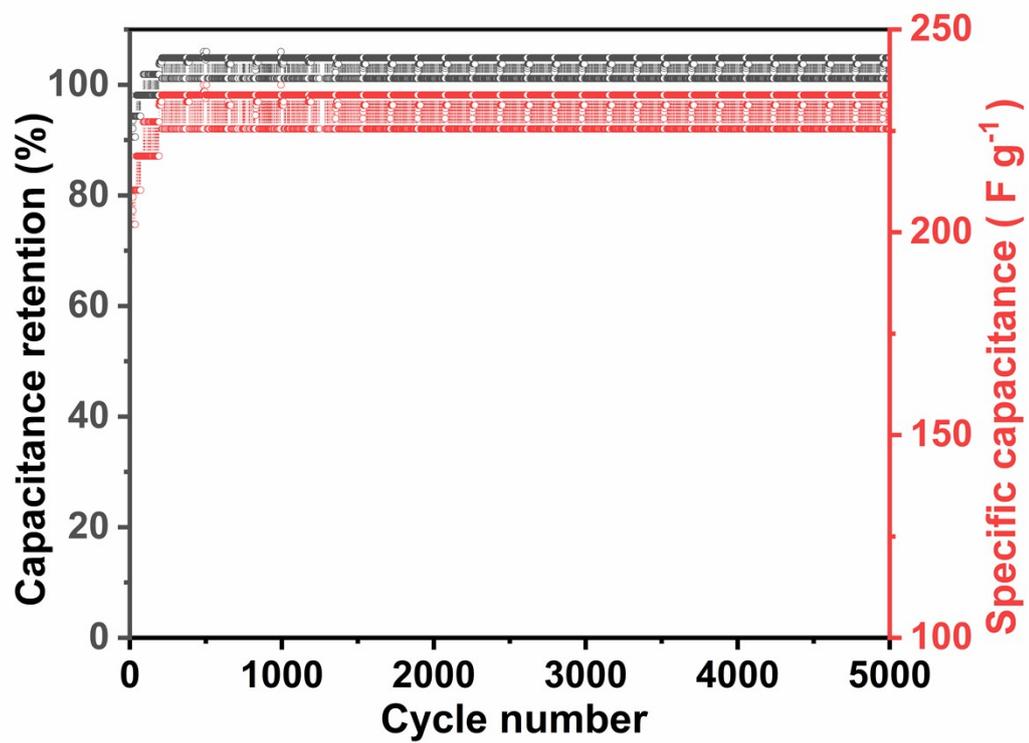|	OCP(%)	0.9 V(%)	-0.9 V(%)
Na	27.95	28.99	15.69
S	13.56	13.22	3.46
Ni	58.49	57.79	80.85

Fig. S13. Capacitance retention plot of the supercapators for 5,000 cycles.

Table S2. Comparison of carbon-based composites contianning Ni species as electrodes as supercapacitors.

Samples	Capacitance (F/g)	References
NiO/C	260	1
NiO/CMC	294	2
N/C	381	3
C	315	4
NiNOG	532	5
NiCo-MOF	134	6
TOCN/GO	398	7
C	274	8
C/S	63.6	9
C-Ni/NiO/NiBDC	667	This Work

References

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