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Electronic Supplementary Information

Buckypaper templating Ni-Co hydroxide nanosheets as free-standing electrode for ultrathin and flexible supercapacitors †

Xinxue Wang,[a,b] Tianshu Song[*a]

[a]School of Aerospace and Civil Engineering, Harbin Engineering University, Harbin, P.R.China 150001.

[b]Mudanjiang Normal University, Mudanjiang, P.R.China 157011.

*Corresponding author: xiwang_hit@126.com

Carbon materials are usually by nature hydrophobic. Thus, for the supercapacitor applications, bare BP was activated to be hydrophilic. The bare BP is electrochemically oxidized in 1M H_2SO_4 solution through a three-electrode configuration using BP as working electrode, Pt mesh as counter electrode and Ag/AgCl as reference electrode to functionalize the surface with oxygen-containing functional groups. The current and voltage transients during the electrochemical oxidization process are shown in Figure S1.

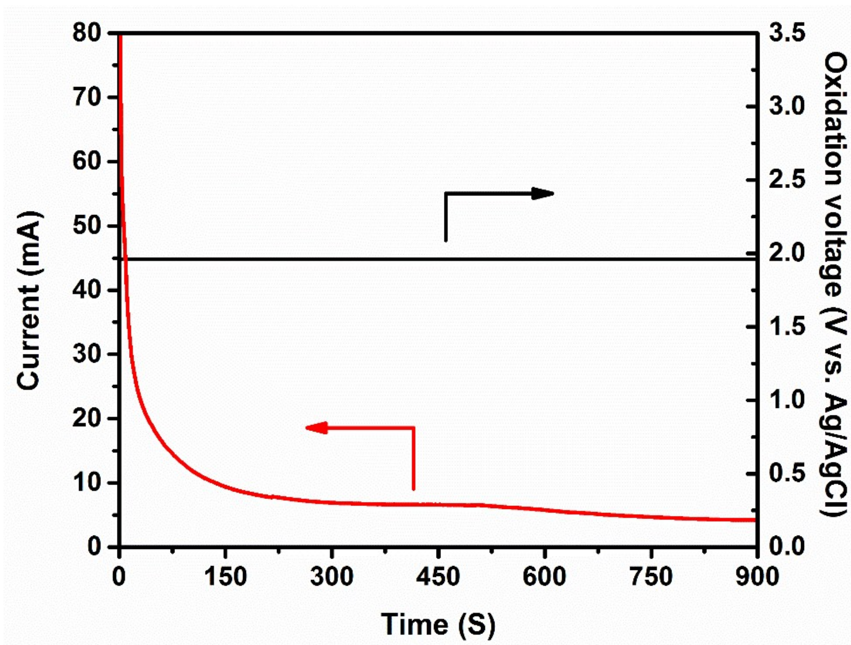

Fig. S1 Electrochemical oxidization process at 1.9 V in 1 M H_2SO_4 for 15 min.

The EIS of the bare BP has also been measured and the result is shown in Fig. S2. From the Nyquist plot for the bare BP, the R_e value calculated from the EIS spectrum at high frequencies is relatively low (approx. 4 Ω). The absence of a clear semicircle in the high frequency region indicates a low charge transfer resistance R_{ct} for the bare BP.

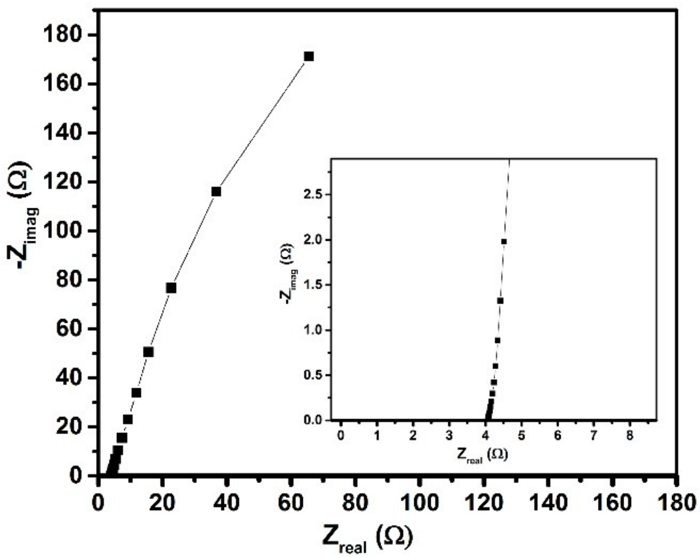

Fig. S2 Nyquist plot for the bare BP recorded from 0.1 Hz to 1 MHz

The elemental components in NCHPs prepared in this work are estimated from EDX maps and shown in Table S1. The atomic ratio of Ni: Co in the as-prepared NCHPs is calculated to be 2:1, which is consistent with the atomic ration in the precursor solution. The composition is expected to be tunable by adjusting the concentrations of raw chemicals in the precursor solution.

Table S1. The atomic ratio of Ni: Co: O elements in NCHPs

Elements	Ni	Co	O	C
Atomic ratio (%)	15.74	7.68	52.54	24.07

Fig. S3 displays galvanostatic charge/discharge profiles of NCHS/BP electrodes at higher current densities ranging from 15 mA cm^{-2} to 50 mA cm^{-2} between 0 V to 0.4 V vs. SCE.

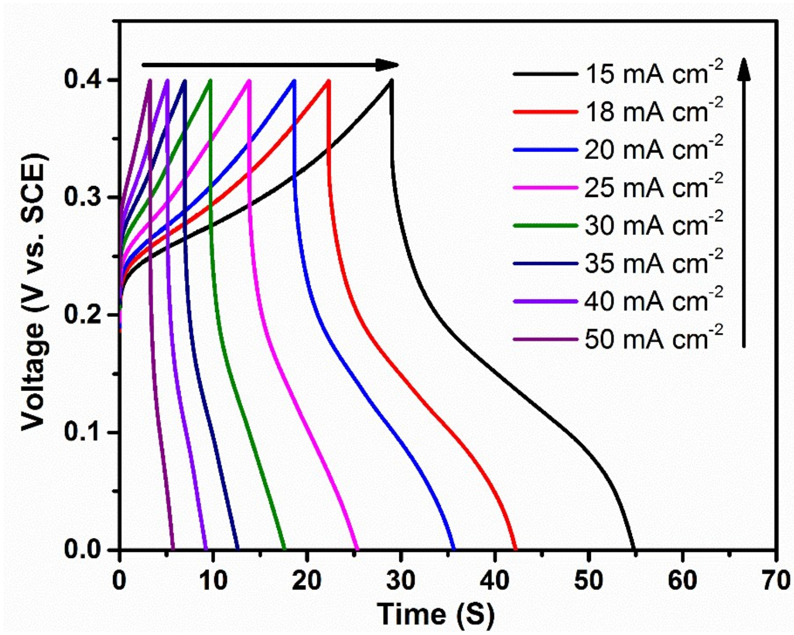

Fig. S3 Galvanostatic charge/discharge profiles of NCHS/BP electrodes at current densities ranging from 15 mA cm^{-2} to 50 mA cm^{-2} between 0 V to 0.4 V vs. SCE.

To further evaluate the electrochemical properties and estimate the stable potential windows of the asymmetric supercapacitor devices, we performed CV measurements on the two individual (positive and negative) electrodes separately using a three-electrode system with a platinum mesh as counter electrode, a saturated calomel electrode (SCE) as reference electrode and 2 M KOH as electrolyte. The bare BP electrodes were measured within a potential window of –0.8 to 0 V vs. SCE, while the NCHS/BP electrodes were measured within a voltage window of –0.2 to 0.5 V vs. SCE at a scan rate of 20 mV s^{-1}, as shown in Fig. S4. Two obvious peaks existing in the CV curve for bare BP, which are related to the redox reactions between oxygen-containing functional groups (endowed by the EO process) and KOH electrolyte. Due to the relatively high scan rate, the anodic peak on the CV curve of NCHS/BP electrode is not obvious compared to that under the low scan rates. These two electrode materials are stable in a different range of potentials. Consequently, the voltage of the as-prepared asymmetric supercapacitor can be extended up to 1.5 V with NCHS/BP as positive electrode and BP as negative electrode, since the total device voltage can be possibly expressed as the sum of the potential range for NCHS/BP and BP electrodes.

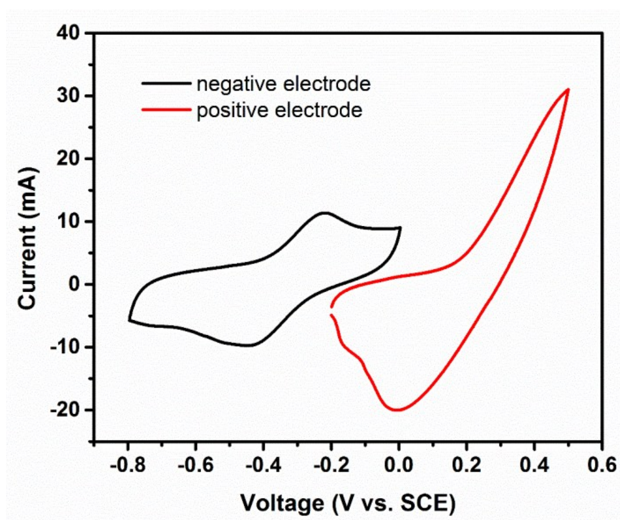

Fig. S4 The CV curves of BP and NCHS/BP electrodes at a scan rate of 20 mV s^{-1}.