Supporting Information

Design of CNTs@$Ni_{1/3}Co_{2/3}(CO_3)_{1/2}(OH)$ $0.11H_2O$ *in-situ* Compounded in Nanoscale for All-Solid-State Supercapacitor

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Fabrication of the All-Solid-State Supercapacitor Devices

The synthesis of polyvinyl alcohol (PVA)/KOH gel electrolyte as follows: 3 g of PVA was added into 20 mL deionized water and stirred in 85 °C oil bath for 1.5 hours until a uniform solution was obtained. Then, after cooling to room temperature, the KOH (3.366 g) solution mixed in the viscous solution under continually stirring. The all-solid-state supercapacitor devices of NCC and CNTs@NCC were assembled as follow: The counter electrode was obtained by adding active carbon (AC) and polyvinylidene fluoride (PVDF) with mass ratio of 8:2. The mixture was dispersed in absolute ethyl alcohol and isopropanol with a 1:1 volume ratio. Then, it was casted by drop-by-drop

addition of the pulp suspension onto a piece of foam nickel (1.5 cm × 1.5 cm). The preparation method of working electrode was similar to that of the counter electrode. The working electrode was composed of 80 Wt% active material, 10 Wt% PVDF and 10 Wt% acetylene black, the dispersed solvent and coating methods were exactly the same the counter electrode. The active substance (including CNTs) in the working electrode was 5 mg; the activated carbon (AC) in the counter electrode was 30 mg. The fabrication of all-solid-state supercapacitor devices as follows: The PVA/KOH gel electrolyte was used as solid electrolyte; a piece of non-woven was used as separator. The working electrode and counter electrode were fully immersed into the PVA/KOH gel electrolyte for 30 mins; then, they were assembled with a non-woven separator in between, left 1 hour until the electrolyte solidified. The entire device was wrapped by plastic wrap while a small part of nickel strap kept outside.

Materials Characterization

The X-ray diffraction (XRD) patterns of the products were recorded on a Bruker D8 X-ray power diffractometer equipped with a Cu-Kα irradiation at a scan rate of 0.1^{o}/s. The surface microstructure of products were obtained with field emission scanning electron microscope (FESEM; ZEISS, MERLIN COMPACT) equipped with energy dispersive X-ray spectroscopy (EDS). The X-ray photoelectron spectroscopy (XPS) date of the sample were collected using an X-ray photoelectron spectroscopy (K-Alpha, Thermo Fisher) with a monochromatic Al-Kα X-ray source, and the binding energy was calibrated with C1s=284.4 eV.

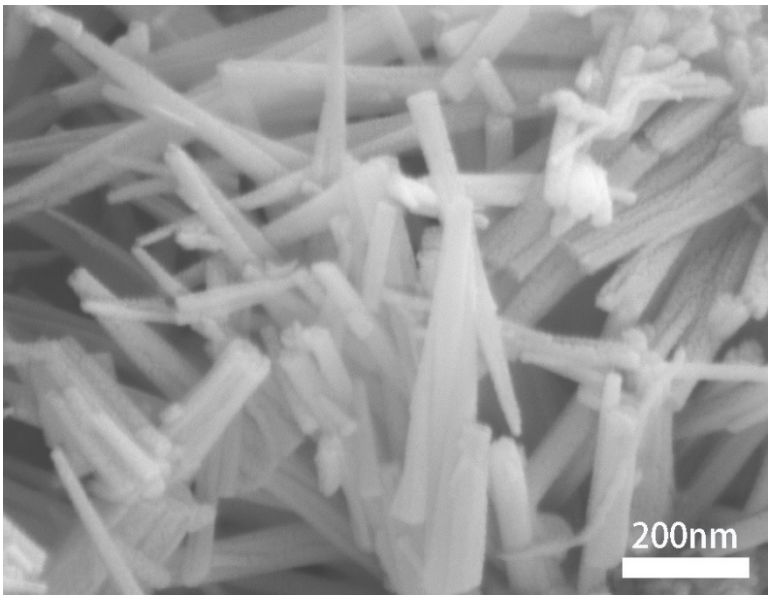

Fig. S1 The SEM image of NCC.

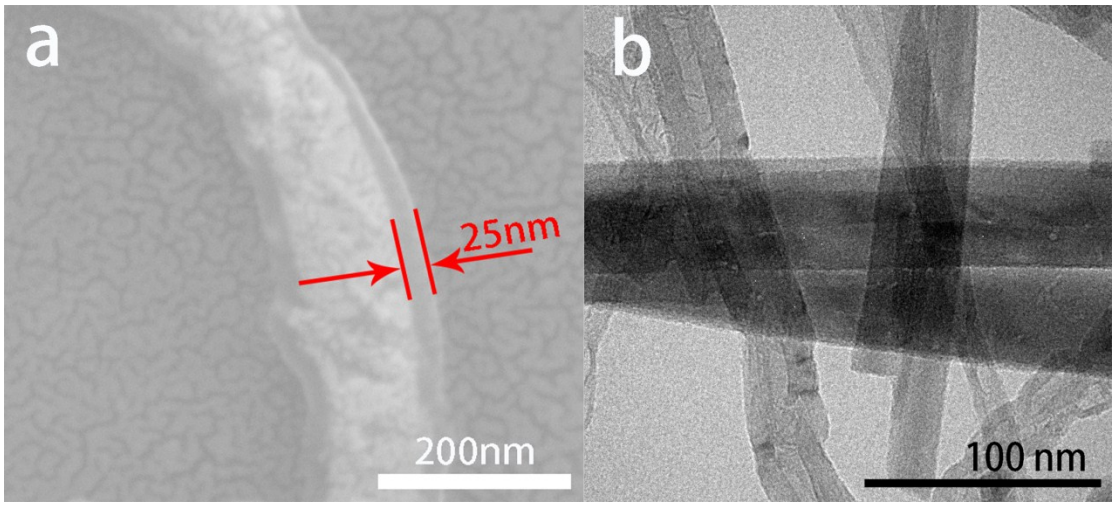

Fig. S2 (a) the high-magnification SEM image of CNTs@NCC; (b) low-magnification TEM image of CNTs@NCC.

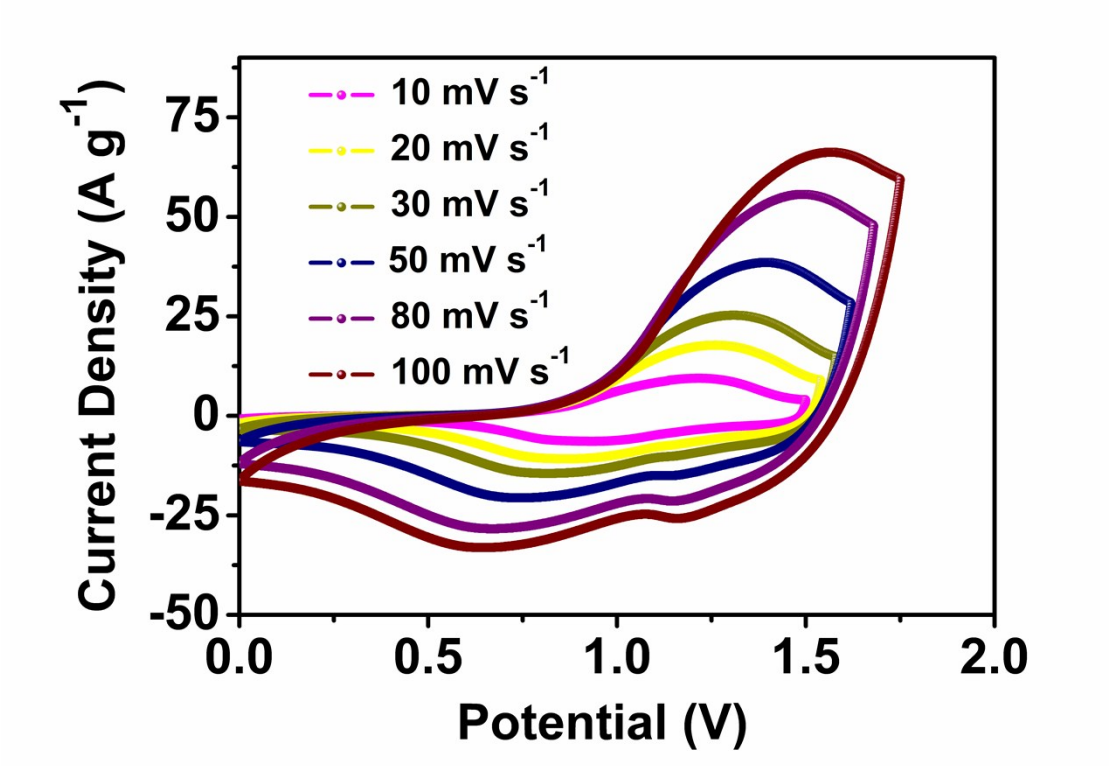

Fig. S3 the cyclic voltammetry date of CNTs@NCC//AC device in 2 M KOH electrolyte at scanning rate of 10-100 mV s^{-1}.

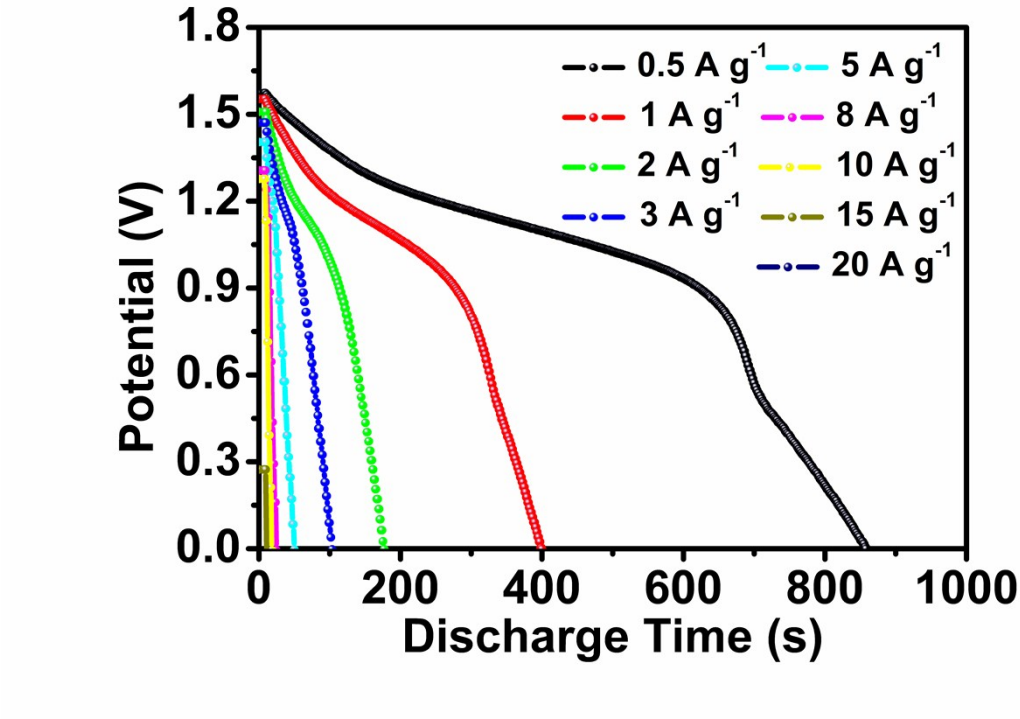

Fig. S4 The discharge time of NCC in 2 M KOH electrolyte.

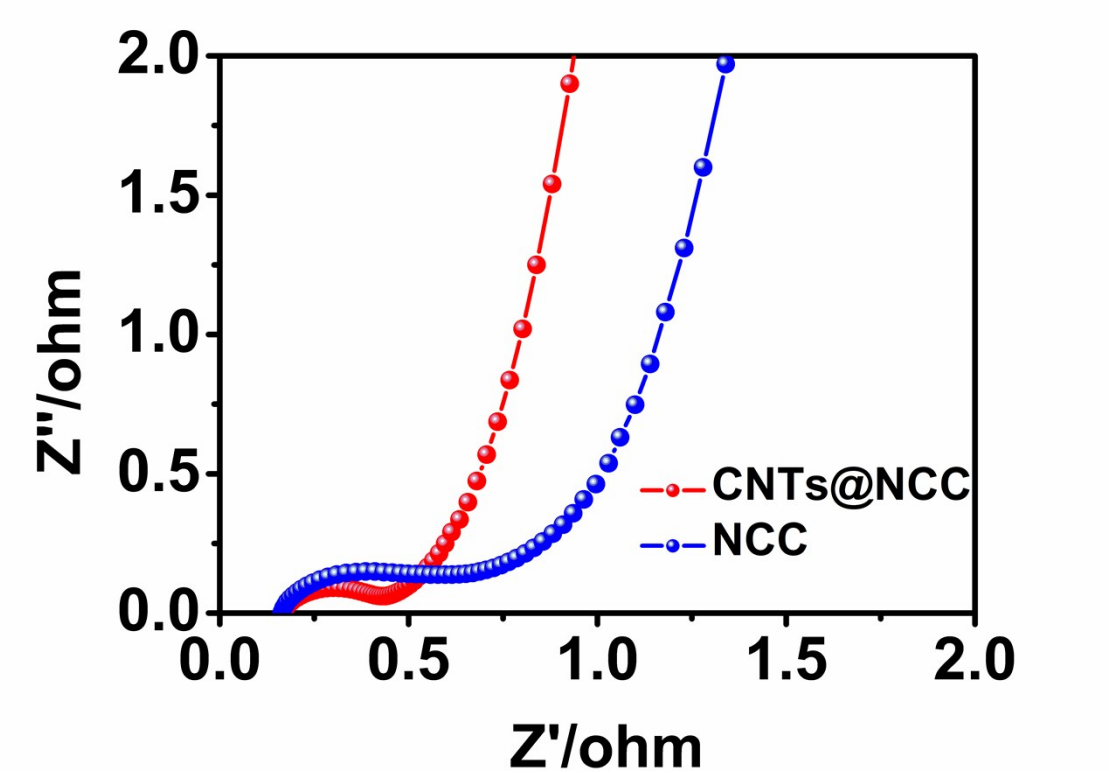

Fig. S5 The Nyquist plot of CNTs@NCC//AC and NCC//AC devices in 2 M KOH electrolyte.

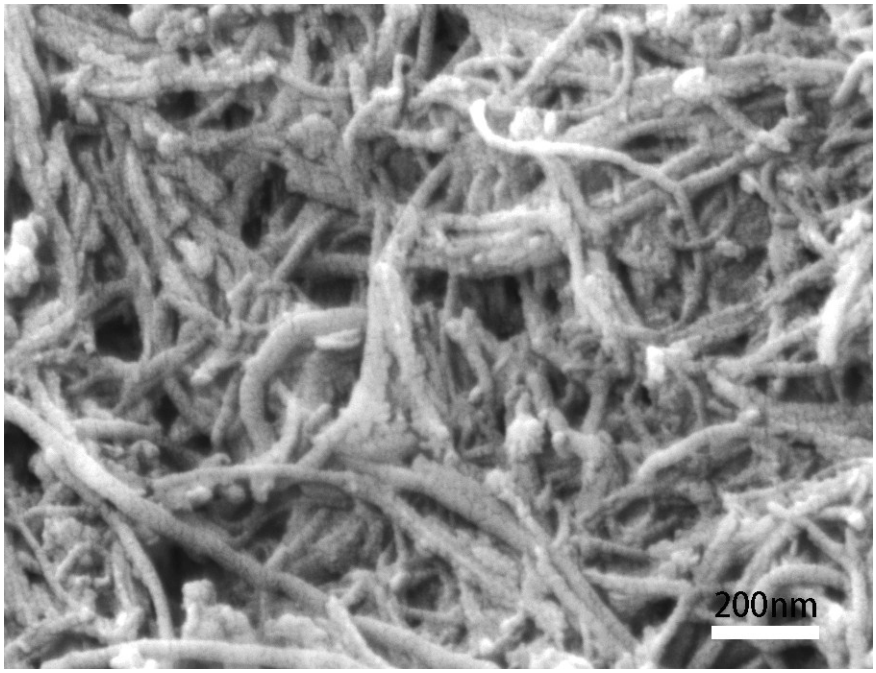

Fig. S6 The FESEM image of CNTs@NCC electrode material after cycles.

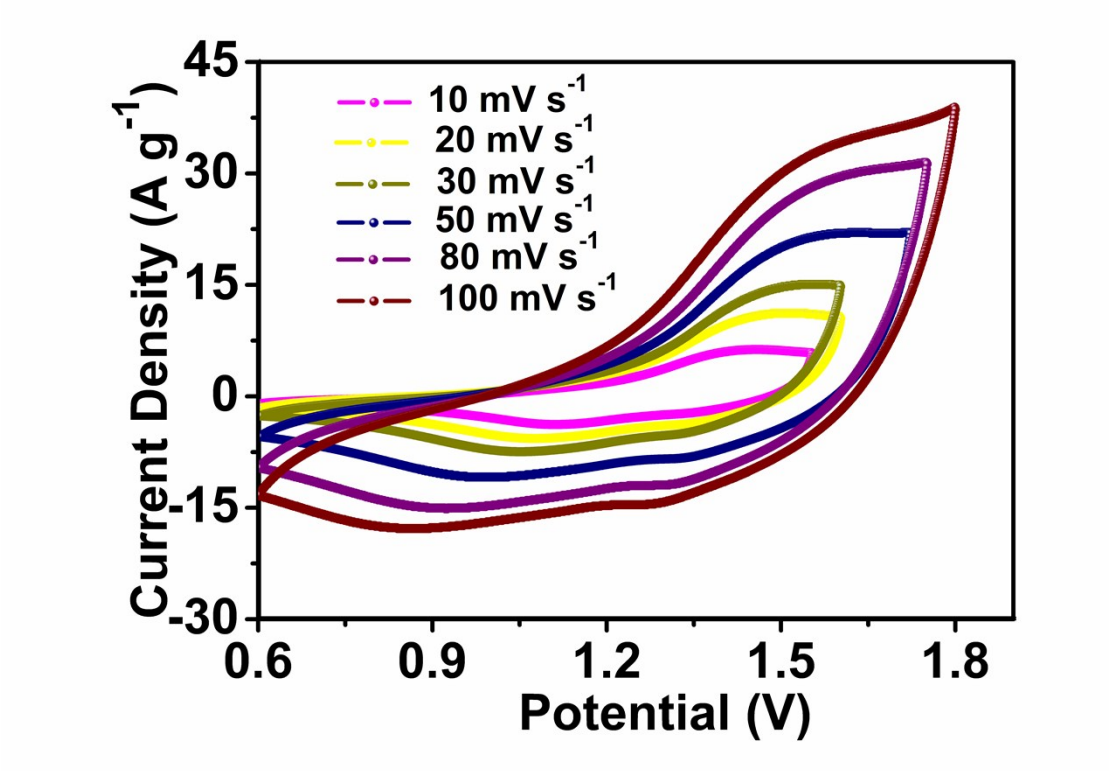

Fig. S7 The cyclic voltammetry date at a scan rate from 10-100 mV s^{-1} in PVA/KOH gel electrolyte.