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Electronic Supplementary Information

Self-assembled hollow urchin-like $NiCo_2O_4$ microspheres for aqueous asymmetric supercapacitors

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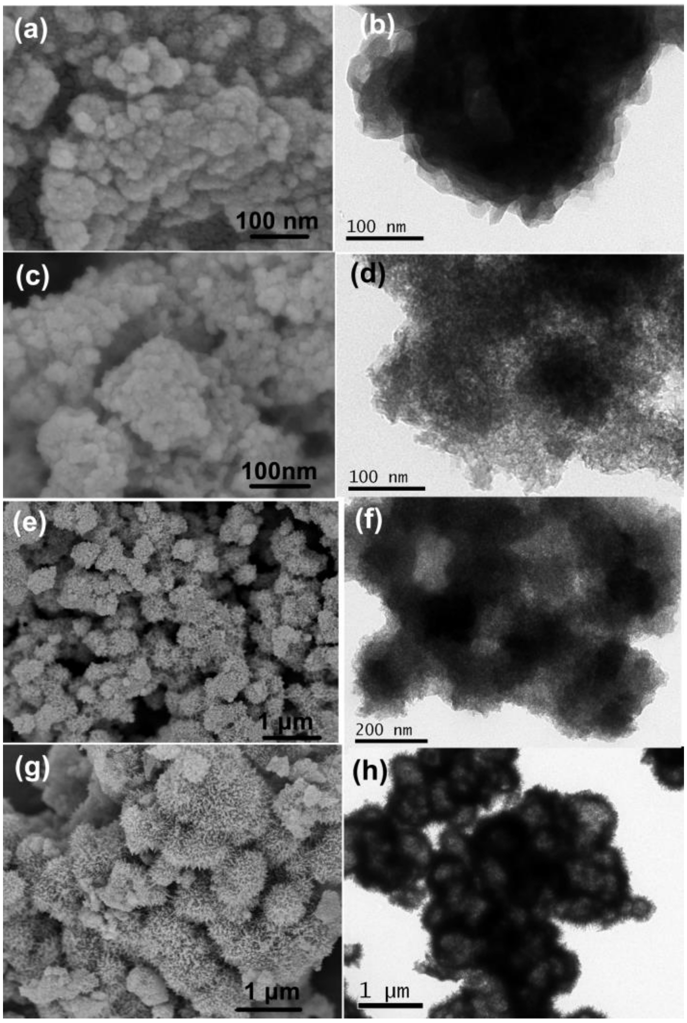

Fig. S1 SEM and TEM images of the precursor samples of the NC-0 collected after different reaction time: (a, b) 10 min, (c, d) 30 min and (e, f) 2 h without aging process, and (g, h) 6 h followed by aging for 12 h.

Time-dependent experiments are carried out to investigate the evolution of the hollow urchin-like microspheres. Fig. S1† shows the SEM and TEM images of the precursor samples without CTAB harvested after different reaction durations. At the early stage of the reaction (10 min, Fig. S1a,b), agglomerations from nanoparticle subunits are formed. When the reaction duration is extended to 30 min (Fig. S1c,d), the product containing a mountain of irregular particles forms initially. After 2 h (Fig. S1e, f), some primary microspheres start to come into being. After further prolonged reaction time (Fig. S1g, h), the microspheres will develop into complete and well-rounded hollow superstructures.

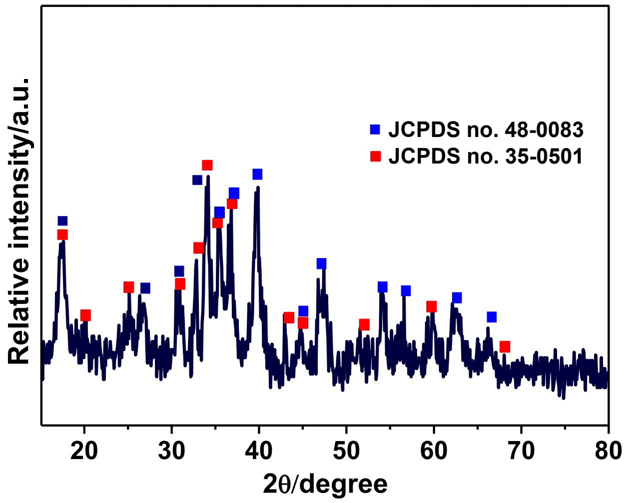

Fig. S2 XRD pattern of the as-synthesized precursor without addition of CTAB

Fig. S2 shows the XRD pattern of the uncalcined precursor. The XRD pattern can be indexed as the cobalt/nickel hydroxide carbonate salts. The result shows that the coprecipitate is a mixture of $Co(CO_3)_{0.5}(OH).nH_2O$ (JCPDS card no. 48-0083) and $Ni_2CO_3(OH)_2$ (JCPDS card no. 35-0501).

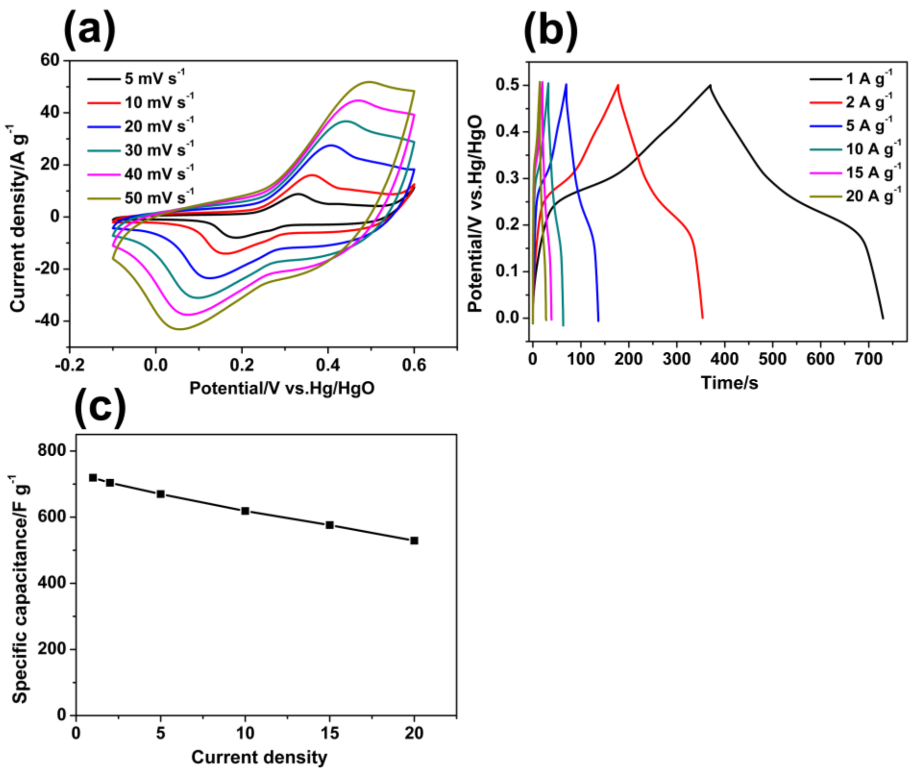

Fig. S3 Electrochemical properties of the hollow NC-0 microspheres: (a) CV curves at different scan rates, (b) GCD curves at different current densities, (c) the corresponding specific capacitance as a function of current density.

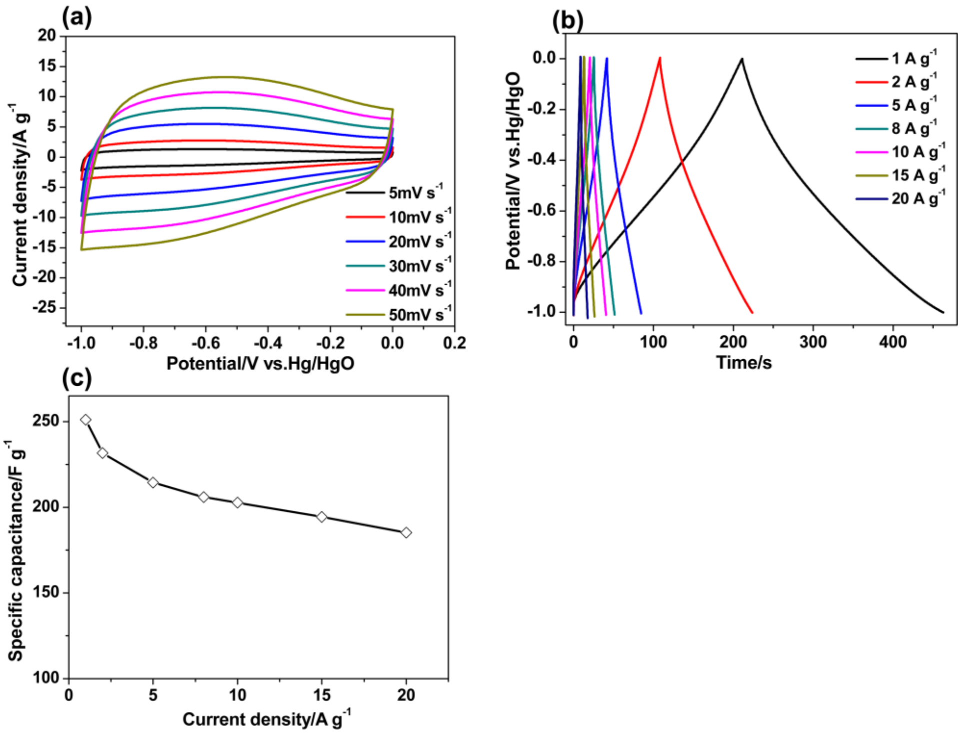

Fig. S4 Capacitance properties of the laboratory-prepared AC: (a) CV curves in a voltage window of -1.0–0.0V at different scan rates from 5 of 50 mV s^{-1}. (b) GCD curves at different current densities, and (c) the corresponding specific capacitance as a function of current density.

As depicted in the Fig. S3c, the AC delivers a specific capacitance of 251, 217, 205 and 188 F g^{-1} at a current density of 1, 5, 10 and 20 A g^{-1}, respectively.

Optimization of mass ratio of $NiCo_2O_4$ and activated carbon (AC)

In order to obtain a well electrochemical performance for supercapacitor, the charge balance between the positive and negative electrodes should be follow the relationship $q_+=q_-$ (q_+ and q_- represent the charge stored by the positive and negative electrodes, respectively). The discharge specific capacitance of the as-prepared $NiCo_2O_4$ (Fig. 5c) and AC (Fig. S4c) are 945 F g^{-1} and 251 F g^{-1} at the current density of 1 A g^{-1}, respectively. According to the following equation [1, 2]

$$\frac{m_+}{m_-} = \frac{C_- \times \Delta E_-}{C_+ \times \Delta E_+}$$

where m is the mass (g) of electrode, C is the specific capacitance (F g^{-1}) and ΔE is the potential range (V) for the charge/discharge process, the mass ratio between positive and negative electrodes of the $NiCo_2O_4$-AC-based asymmetric capacitor should be 0.531. Typically, the total mass of an ASC 8.1 mg is composed of the positive active material mass 2.8 mg and negative electrode active material mass 5.3 mg.

Reference

[1] H. Li, M. Yu, F. Wang, P. Liu, Y. Liang, J. Xiao, C. Wang, Y. Tong and G. Yang. *Nat. Commun.* **2013,** *4,* 1894.

[2] P. C. Gao, A. H. Lu, and W. C. Li. *J. Power Sources* **2011,** *196,* 4095-4101.