Supplementary Information

Remarkable electrochemical properties of novel $LaNi_{0.5}Co_{0.5}O_3/0.333Co_3O_4$ hollow spheres with mesoporous shell

Ni Wang,[ab] Mengqi Yao,[a] Peng Zhao,[a] Wencheng Hu[a]* and Sridhar Komarneni[b]*

[a]Center for Applied Chemistry, University of Electronic Science and Technology of China, Chengdu, 610054, China

[b]Materials Research Institute and Department of Ecosystem Science and Management, Materials Research Laboratory, The Pennsylvania State University, University Park, Pennsylvania, 16802, United States

*Corresponding Authors: huwc@uestc.edu.cn (Wencheng Hu), sxk7@psu.edu (Sridhar Komarneni)

1. The formation mechanism of hollow spheres of $LaNi_{0.5}Co_{0.5}O_3/0.333Co_3O_4$

Precursor solution was atomized to form liquid droplets with a size of several hundred nanometers, and the liquid droplets were introduced by gas flow into quartz tube of a tubular furnace and heated at a high temperature. The solvent on the surface of droplets was firstly evaporated and the residual inorganic salts transformed into desiccated oxide shells. The solvent within a micro-sphere was subsequently volatilized though the pore passageways of the desiccated shell, and the precipitated inorganic salts thickened the shell. Due to the wetting mechanism, the residual solution remained on the inner surface of the shell until the liquid was completely evaporated to form hollow micro-spheres.

2. Preparation of electrode for measurement

The slurry containing active material ($LaNi_{0.5}Co_{0.5}O_3$/$0.333Co_3O_4$), carbon black PVDF binder and isopropanol was ground in an agate mortar for 20min, and then pressed onto carbon felt. After vacuum-drying at 120 °C for 12 h, the working electrode was weighted to calculate the mass of active material on the basis of the weight ratio described in manuscript. The typical mass loading of the active materials on single electrode was about 1 mg cm^{-2}.

3. The electrical resistivity of the prepared samples

Test method: The prepared samples were poured in a carbon steel mould. An external pressure up to 50 MPa was applied to the pistons in order to compress the sample to make pellets. Here, pellets of about l mm in thickness and 12 mm in diameter were prepared. The electric conductivities of the pellets were measured in a Four Probe Tester (RTS-8, Uncommon Technology Development Co., Ltd, China) and the results are listed in Table S1.

Table S1Electrical conductivities of different samples using four-probe measurement method.

Sample	LNCO	CO	LNCO/CO
Electrical conductivity ($S\ cm^{-1}$)	121.7	1.88×10^{-6}	94.2

4. The effect of mass loading on specific capacity

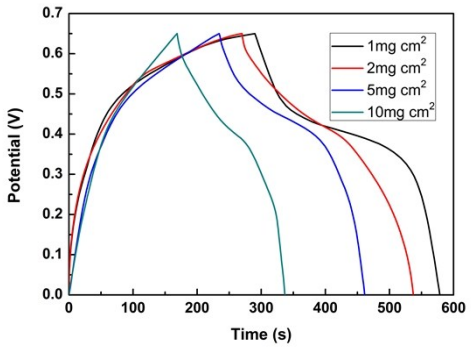

Fig.S1 GCD curves of $LaNi_{0.5}Co_{0.5}O_3/0.333Co_3O_4$ with different mass loadings

Table S2 Specific capacity of $LaNi_{0.5}Co_{0.5}O_3/0.333Co_3O_4$ with different mass loadings

Mass loading ($mg\ cm^{-2}$)	Specific capacity ($C\ g^{-1}$)	Ratio* (%)
1	537.8	108
2	498.0	**100**
5	423.8	85.1
10	315.2	63.3
*Based on the specific capacity with a mass loading of 2 $mg\ cm^{-2}$.		

5. Preparation method of the N-doped mesoporous carbon

In a typical synthesis, 1.3g of iron (III) chloride hexahydrate was dissolved in 50ml of deionized water to form a transparent solution. Subsequently, 2g of pyrrole was dropwise added to the above solution under magnetic stirring at 5 ^{o}C. The polymerization reaction was carried out for 24 h so that all the pyrrole molecules were transformed into polypyrrole (PPy). After filtering, washing and drying, the PPy was mixed with 1.5g KOH and ground for 30min. The mixture was carbonized at 900 °C for 3 h under a continuous flow of high purity Ar gas. The carbonized product was rinsed thoroughly with hydrochloric acid solution, deionized water and ethyl alcohol, dried at 100^{o}C under vacuum for 4h to finish the synthesis of N-doped mesoporous carbon.

6. FESEM and HRTEM images of mesoporous carbon

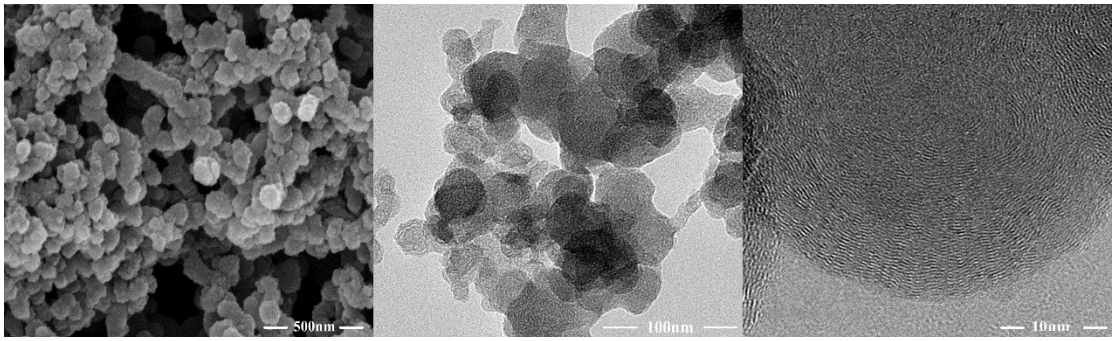

Fig. S1 FESEM and HRTEM images of N-doped mesoporous carbon derived from PPy

7. XPS of NMC

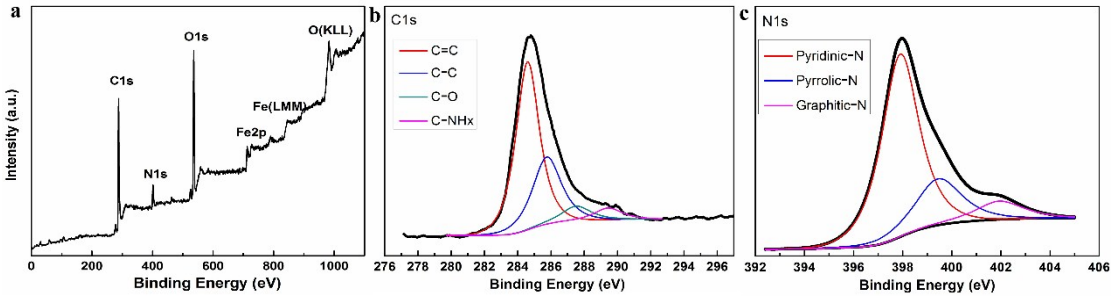

Fig. S2 (a) survey XPS spectrum of NMC; (b) C1s XPS spectrum (c) N1s XPS spectrum.

The N concentration, defined as N/(C+N) atomic ratio % and estimated by the area ratio of the N 1s and C 1s bands, was 9.72%. The high-resolution N1s spectrum presents three kinds of nitrogen: pyridinic, pyrrolic, and graphitic nitrogen.

8. N_2 adsorption/desorption isotherms

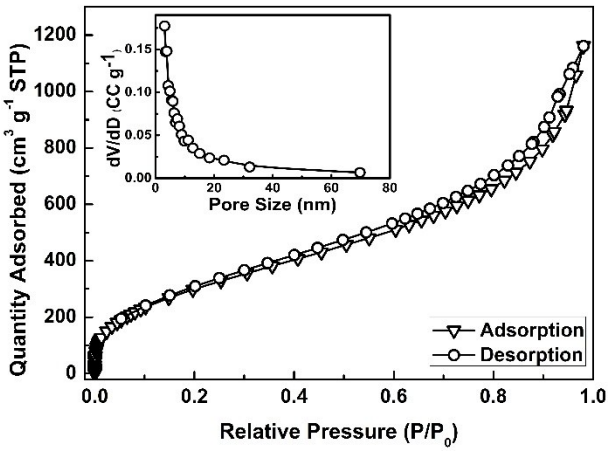

Fig. S3. Nitrogen adsorption and desorption isotherms. The BET surface area was calculated to be 1131 m²/g and the pores were mainly in the size range of 3 to 10 nm.

9. Mass ratio of the hybrid supercapacitor

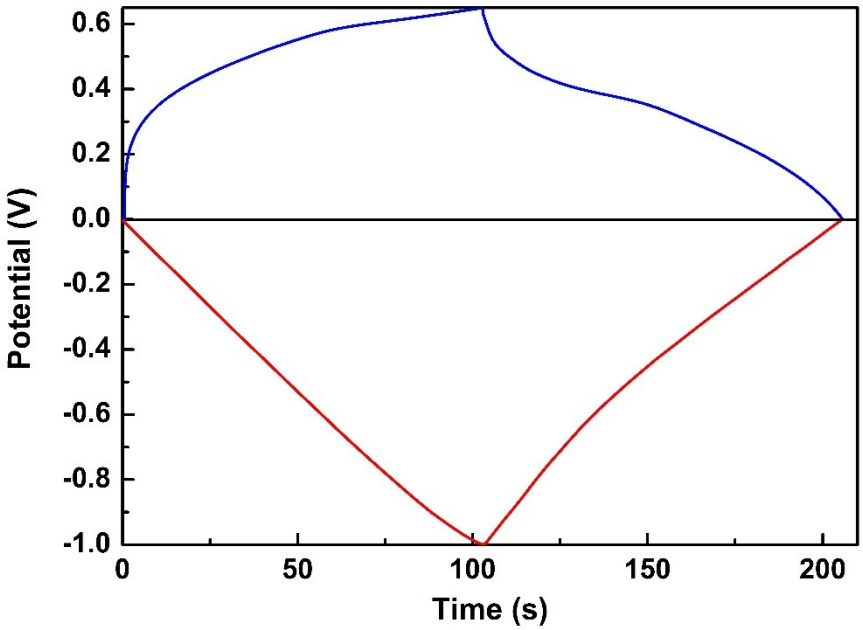

Fig. S4. The matched capacitance between LNCO/CO positive electrode and NMC negative electrode by GCD measurement at 5A g^{-1}. The mass ratio between LNCO/CO and NMC was determined to be 0.56 as calculated approximately by GCD measurement.

5. EIS measurements of different samples

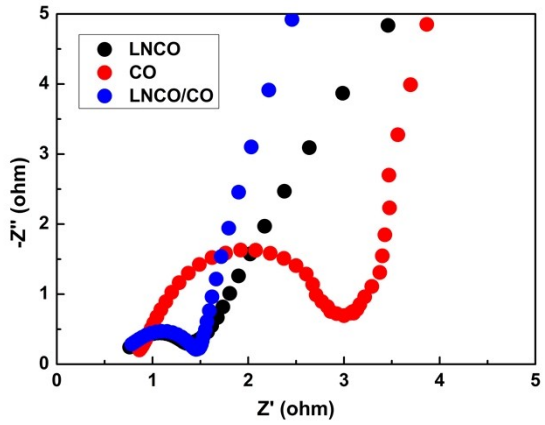

Fig.S2. Nyquist plots of the hybrid supercapacitor with difference anode materials

Table S3 Rs and ESR values of the hybrid supercapacitor with difference anode materials

Sample	LNCO	CO	LNCO/CO
Solution resistance (Rs, Ω)	0.76	0.83	0.78
Equivalent series resistance (ESR, Ω)	0.98	2.27	1.02