Electronic supplementary information

Hierarchical $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ plates with exposed {010} active planes as high performance cathode material for Li-ion battery

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Part 1 Experimental Section

Material Synthesis

Synthesis of Flower-like Precursor

Flower-like precursor was prepared via a solvolthermal method. Specific experimental process is as follows: using 3 mmol of $Ni(NO_3)_2\cdot 6H_2O$ and $Co(NO_3)_2\cdot 6H_2O$ (the cationic ratio of Ni: Co = 80: 15) as the solutes, 0.1 g of polyvinylpyrrolidone (PVP, M_w = 29,000 g mol^{-1}) as the template and surfactant, 60 ml of a mixture containing ethanol and deionized water (the volumetric ratio of ethanol and deionized water = 1:1) as the solvent. The mixed solution was transferred to a 100 ml teflon-lined stainless steel autoclave after stirred for 30 minutes and then heated at 180 °C for 12 h. The prepared precipitant was washed using distilled water and dried at 50°C for 12 h in a vacuum oven to obtain the precursor. In order to avoid breaking apart of the secondary particles for target $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, the as-synthesized precursor was then heated at 300 °C for 3 h.

Synthesis of Hierarchical $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ Plates (HP-NCA)

Hierarchical $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ plates were prepared via a solid state method. The heated precursor was mixed with a stoichiometric amount of $Al(OH)_3$ and an excess amount of $LiOH\cdot H_2O$ (5 wt%). Then the mixture was heated in oxygen atmosphere at 500 °C for 5 h and extra 750 °C for 18 h to obtain the final product.

Synthesis of $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ Spherical Micro-particles (SP-NCA)

The commercial precursor purchased from Henan New Taihang Power Source Co., Ltd was also heated at 300 °C for 3 h. The $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ spherical micro-particles were synthesized via the same procedure described in the synthesis of HP-NCA.

Characterization

The surface morphology and microstructure were examined using scanning electronic microscopy (SEM, Hitachi S-4500) and transmission electronic microscopy (TEM, JEOL-2100). The elemental analysis was conducted by EDX transmission electronic microscopy (TEM, JEOL-2100). The crystal phases of HP-NCA and SP-NCA were characterized by X-ray diffraction (XRD, Philips, X' pert TRO MPD) using Cu Kα radiation ($\lambda = 1.5418$ Å) at room temperature. The compositions of HP-

NCA and SP-NCA were measured by inductively coupled plasma (ICP, Perkin Elmer, Optima 8000).

Electrochemical Measurements

The electrochemical properties of HP-NCA and SP-NCA were tested by CR2032 coin-type lithium half-cells. To prepare the positive electrode, 80 wt% cathode materials, 13 wt% acetylene black and 7 wt% polyvinylidene fluoride (PVDF) were dissolved in N-methyl-2-pyrrolidone (NMP). The slurry was coated uniformly onto the aluminum foil which was selected as the current collector of the positive electrode. The cathode film was dried at 120 °C overnight in a vacuum oven. The electrodes were cut into disks with a diameter of 1.6 cm. The average mass loading was about 3.0 mg cm^{-2}. A lithium metal foil was used as the negative electrode. A porous polyethylene film (Celgard 2500) was selected as the separator. The electrolyte is a mixed solution of $LiPF_6$ in ethylene carbonate, dimethyl carbonate and ethyl methyl carbonate ($V_{\mathrm{EC}}:V_{\mathrm{DMC}}:V_{\mathrm{EMC}} = 1:1:1$). The CR2032 type coin cells were assembled in an argon-filled glove box. The charge and discharge performances of HP-NCA and SP-NCA were conducted on an Arbin instrument (BT-2043) at room temperature. The electrochemical impedance spectra of HP-NCA and SP-NCA were gained on a PARSTAT 4000 workstation in the frequency range from 0.01 Hz to 0.1 MHz and in the perturbation amplitude of 10 mV. Nyquist plots were collected and analyzed using ZSimpWin software.

Part 2 Supporting Figures and Tables

Table S1 Element molar ratios of HP-NCA and SP-NCA from ICP analysis.

	Element	Li	Ni	Co	Al
HP-NCA	Content (ppm)	2.8529	19.159	3.5778	0.5602
	Molar ratio	1	0.7935	0.1498	0.0567
SP-NCA	Content (ppm)	2.5473	17.075	3.2155	0.5091
	Molar ratio	1	0.7911	0.1493	0.0596

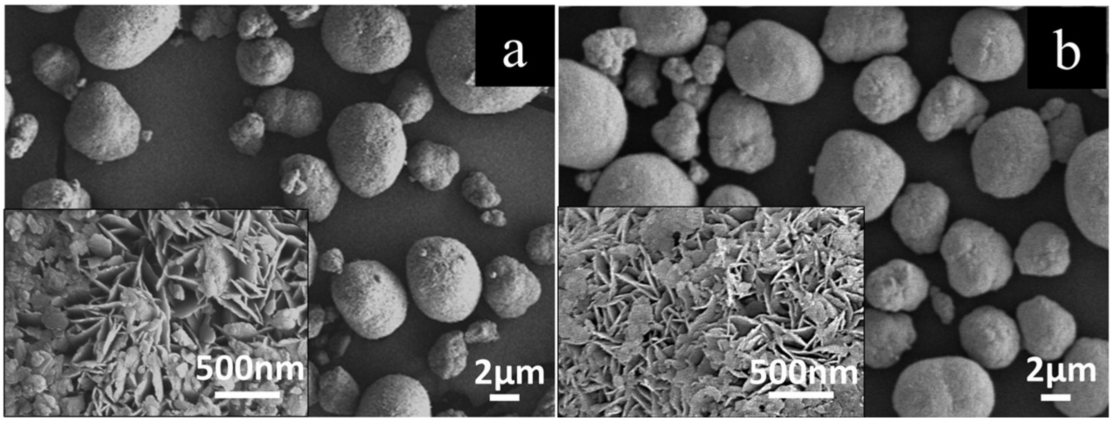

Fig. S1 SEM images of (a) commercial precursor, (b) heated commercial precursor. Insets are the SEM images at high magnification.

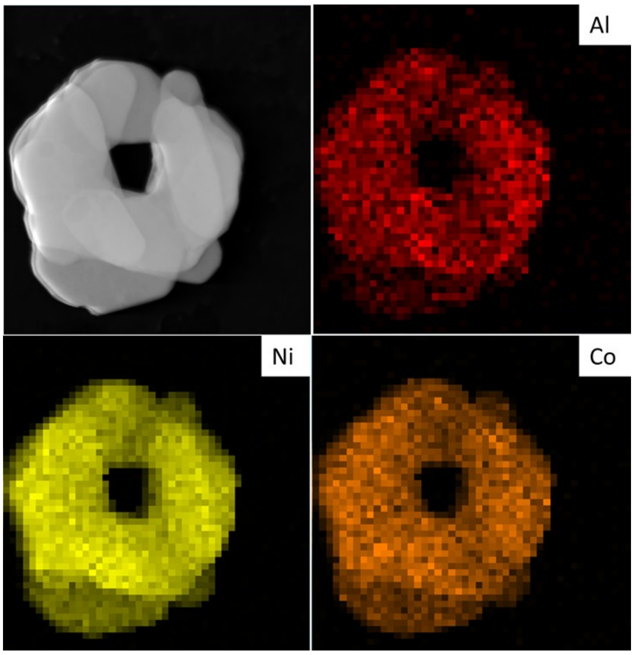

Fig. S2 EDX-mapping images for Ni, Co and Al in HP-NCA.

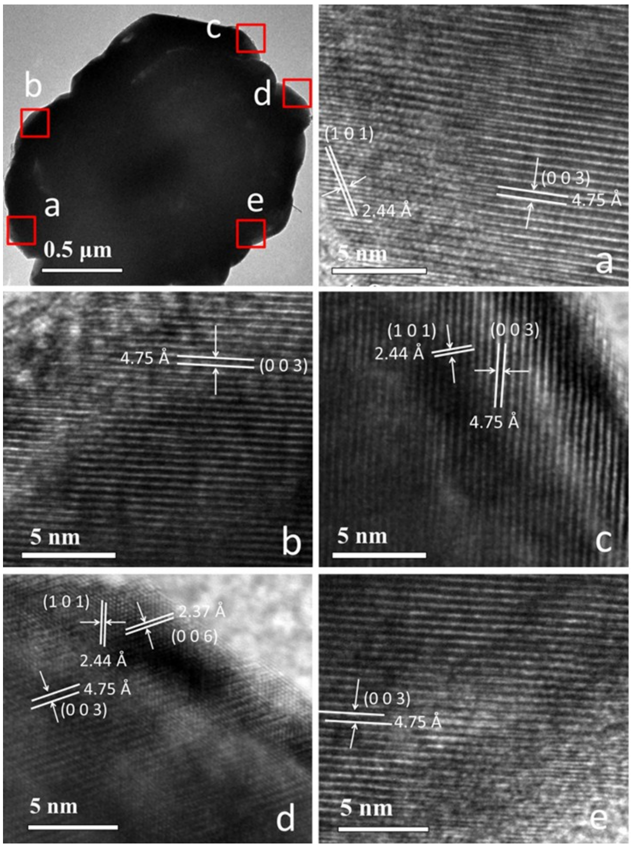

Fig. S3 TEM and HR-TEM images of SP-NCA.

Table S2 The values of R_e, R_{SEI}, R_{ct} and D_{Li^+} of HP-NCA and SP-NCA

	R_e / Ω	R_{SEI} / Ω	R_{ct} / Ω	D_{Li^+} / cm^2 s^{-1}
HP-NCA	5.795	8.064	80.51	5.53×10^{-15}
SP-NCA	5.022	7.538	192.5	1.40×10^{-15}