Supporting Information

Yolk-Shelled $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ Cathode for High-Performance Lithium Ion Batteries：A General Synthetic Strategy

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Experimental Section

Materials synthesis. The nickel cobalt manganese complex was synthesized using the solvothermal method. Firstly, a stoichiometric amount of $Ni(NO_3)_2·6H_2O$, $Co(NO_3)_2·6H_2O$ and $Mn(NO_3)_2·4H_2O$ were dissolved in 60 ml isopropanol solution, and magnetic stirring was carried out for about 30 minutes to form a transparent and clear solution. After dissolving 0.9 g anhydrous glucose, the transparent solution is stirred for about 20 minutes. The obtained solution was transferred to the 100 mL Teflon-lined stainless-steel autoclave and heated to 180 °C for 12 h at 300 rpm. The product collected by centrifugation and washing for 3 times was dried in an oven with a temperature of 60 oC overnight. The product was named as NCMC. The product was sintered in a muffle furnace, slowly heated to 600 °C at a heating rate of 2 °C /min in air atmosphere for 2 h, and then cooled naturally to obtain nickel cobalt manganese oxide (NCMO). Lithium carbonate and nickel cobalt manganese oxide (NCMO) are weighed respectively and mixed according to a certain molar ratio (Li_2CO_3 : NCMO = 0.625 : 1). Then 100 μl polyvinyl alcohol aqueous solution with the concentration of 2 g L-1 is added to fully mix them. After grinding for 20 minutes, the samples were pre-sintered at 480 °C at the rate of 2 °C/min for 5 h in oxygen atmosphere. $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ materials was obtained by calcining at different temperatures for 15 hours under oxygen atmosphere. $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ materials with different temperatures (730, 780, and 830 °C) were marked NCM622-P2-730, NCM622-P2-780 and NCM622-P2-830. For comparison purposes, samples marked as NCM622-P0-780 was prepared by the same procedure with 100 μl deionized water and sintering temperature of 780 °C.

Structural characterization. The microstructures of the products were characterized

by scanning electron microscopy (SEM, FEI, Quanta 250F) and (high-resolution) transmission electron microscopy ((HR)TEM, Thermo Fisher, Talos F200X). The phase structures of the products were characterized by X-ray diffraction (XRD) under Cu Ka radiation (k = 1.5418 Å) at 30 kV and 10 mA in the 2θ range of 10-80°. The elemental composition and chemical state of the products were analyzed by X-ray photoelectron spectroscopy (XPS) using an ESCALAB Xi+ Thermo Fisher XPS instrument.

Electrochemical measurements. The electrochemical properties of $LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$ were investigated in CR2025 coin-type half-cells. Firstly, the working electrodes were fabricated by casting a slurry on the aluminum foil, and the slurry was prepared by mixing the active materials ($LiNi_{0.6}Co_{0.2}Mn_{0.2}O_2$), conductive carbon black (ECP-600JD) and polyvinylidene fluoride (PVDF, HSV900, Arkema) with the weight ratio of 8:1:1 and 1-methyl-2-pyrrolidinone (NMP, C5H9NO99.5%, Macklin) as a solvent. The slurry coated aluminum foils were then dried under vacuum at 60 ℃ for 12 h. The working electrodes were punched into round sheets with a diameter of 14 mm, and the loading amount of the active material was about 1.8 mg cm^{-2}. LIB cells were assembled in Ar-filled glove box (H_2O < 0.1 ppm, O_2 < 0.1 ppm). For lithium ion half cells, the aluminum foils were used as the counter electrodes, and the Celgard 2400 as the separators. 1M LiPF6 in EC/DEC (1:1 in volume) was used as the electrolyte. The galvanostatic discharge-charge tests of LIBs were performed on a battery test system (Neware BTS, China).

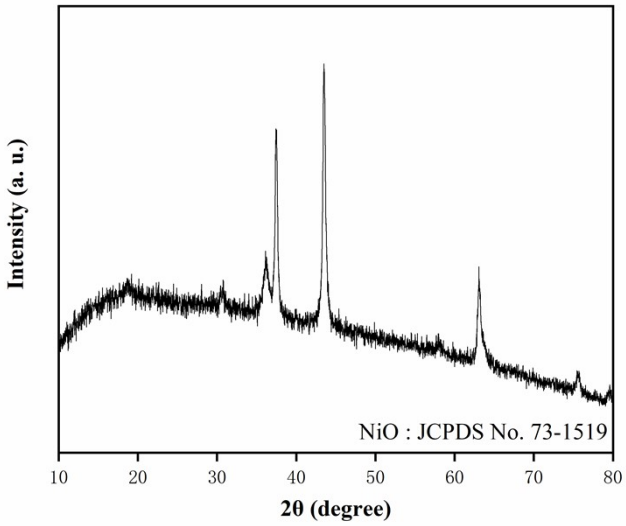

Fig. S1. XRD pattern of the NCMO.

Table S1. The ICP result of NCM622-P2-780.

Elements	**Li**	**Ni**	**Co**	**Mn**
Atomic ratio	2.94	1.55	0.55	0.57

Table S2. Lattice parameters of different NCM622 products.

Sample	**c (Å)**	**c (Å)**	**c/a**	$\mathbf{I_{(003)}/I_{(104)}}$
NCM622-P0-780	2.8650	14.0759	4.91	1.57
NCM622-P2-730	2.8559	14.0434	4.92	1.67
NCM622-P2-780	2.8568	14.0460	4.92	1.81
NCM622-P2-830	2.8655	14.0583	4.91	1.77

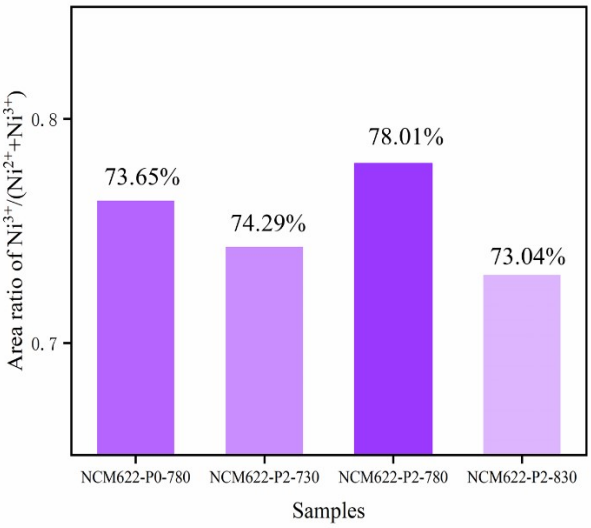

Fig. S2. Area ratios of $Ni^{3+}/(Ni^{2+}+Ni^{3+})$ of different NCM622 products.

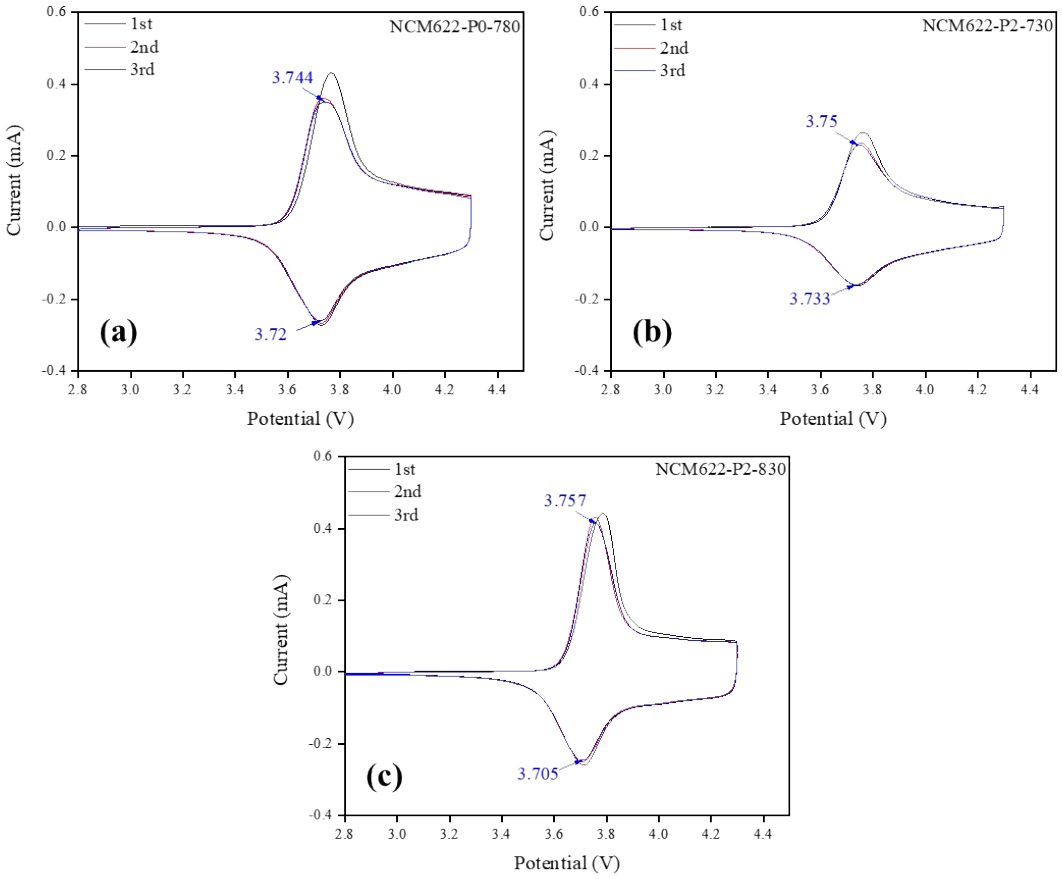

Fig. S3. CV curves of NCM622-P0-780, NCM622-P2-730 and NCM622-P2-830 at 0.1 mV/s.

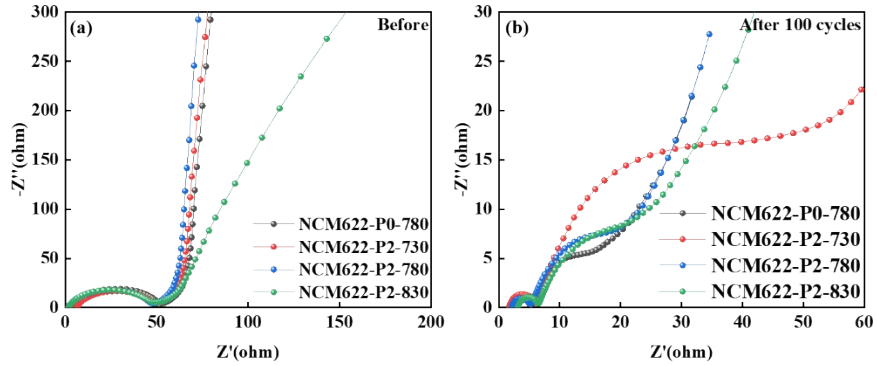

Fig. S4. Nyquist plots of the different NCM622 products.

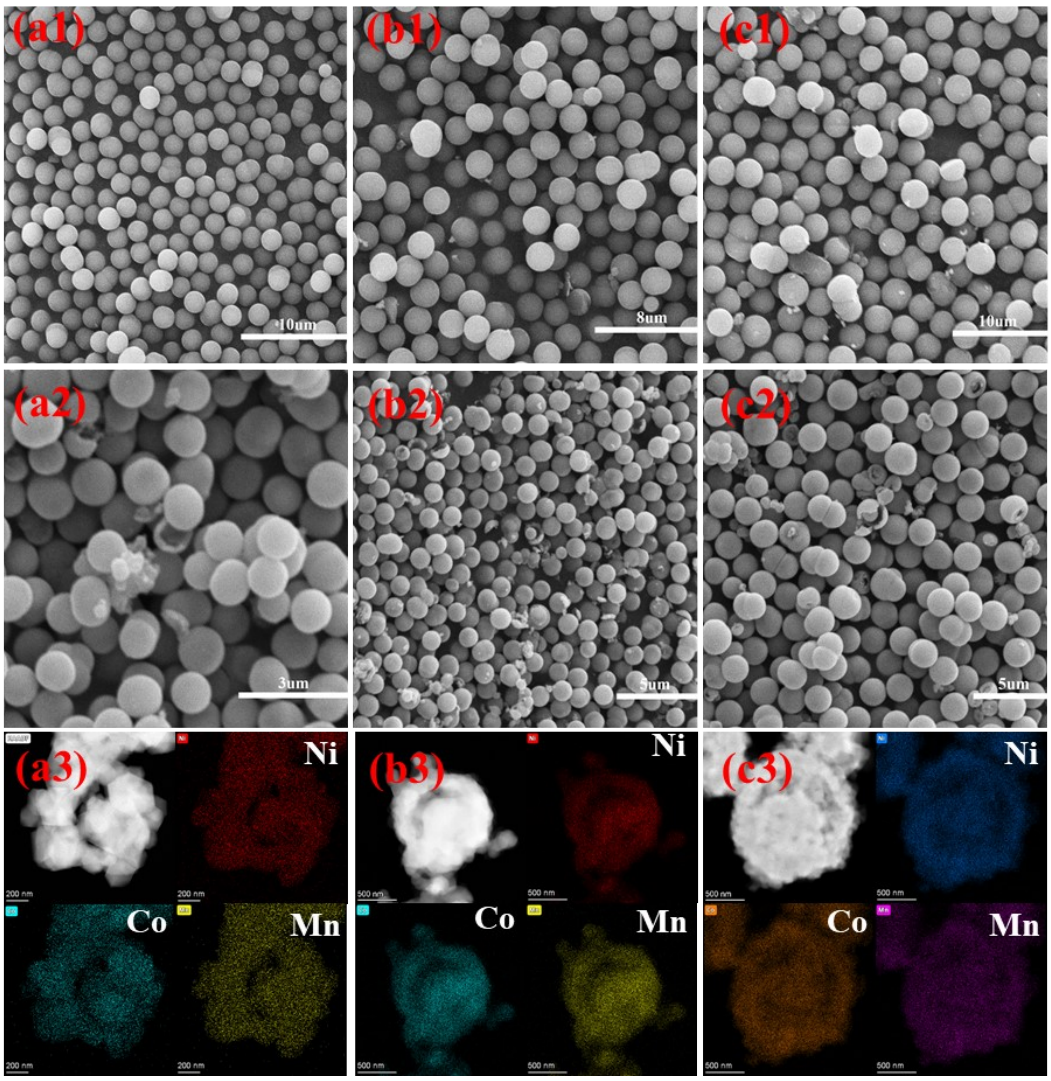

Fig. S5. (a1-2) SEM images of the NCM111 precursor (a1) and oxide (a2), and (a3) HAADF STEM image of the yolk-shelled NCM111 cathode with corresponding EDS maps of Ni/Co/Mn elements. (b1-2) SEM images of the NCM523 precursor (b1) and oxide (b2), and (b3) HAADF STEM image of the yolk-shelled NCM523 cathode with corresponding EDS maps of Ni/Co/Mn elements. (c1-2) SEM images of the NCM811 precursor (c1) and oxide (c2), and (c3) HAADF STEM image of the yolk-shelled NCM811 cathode with corresponding EDS maps of Ni/Co/Mn elements.

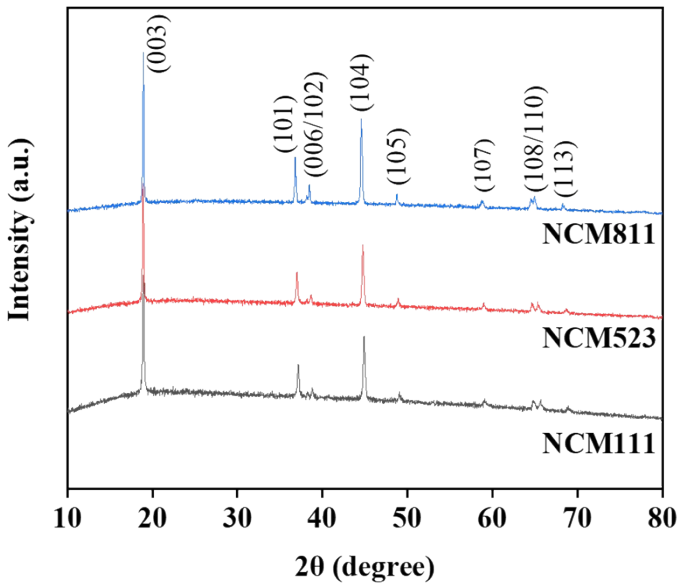

Fig. S6. XRD patterns of the NCM11, NCM523 and NCM811 products.

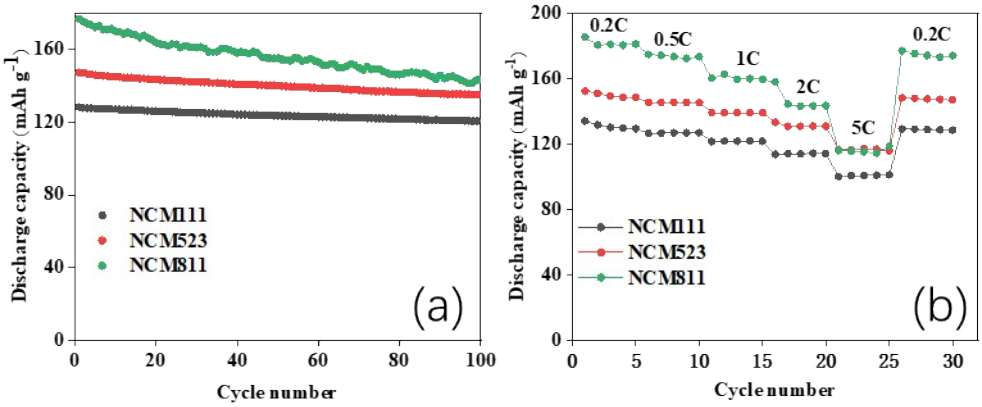

Fig. S7. (a) Cycle and (b) rate performances of the NCM11, NCM523 and NCM811 cathodes.

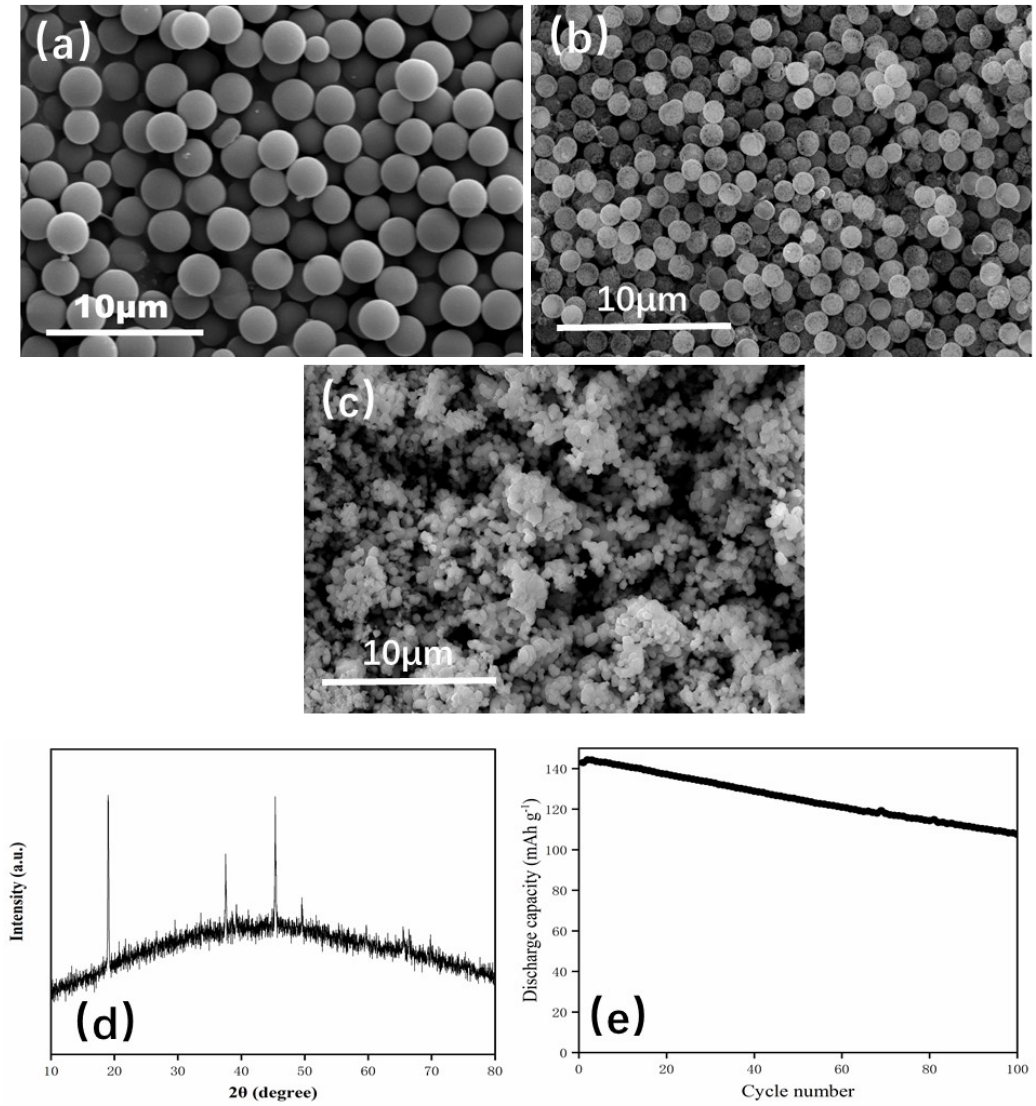

Fig. S8. (a-c) SEM images of the (a) Co-gluconate, (b) CoOx and (c) $LiCoO_2$. (d) XRD pattern and (e) cycle performance of $LiCoO_2$ cathdode.

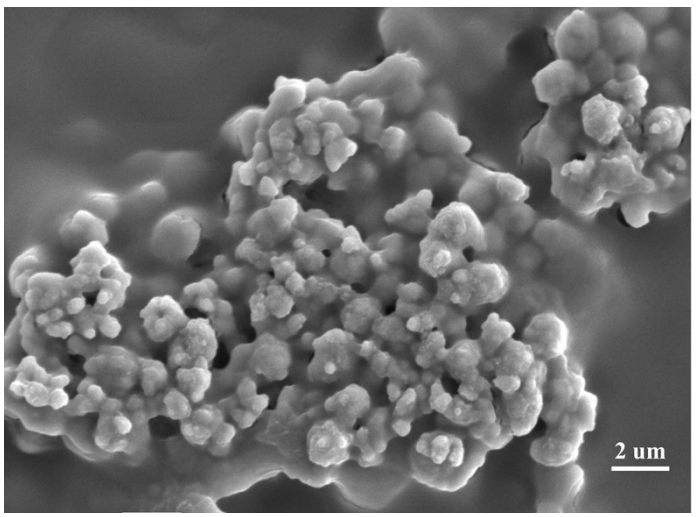

Fig. S9. SEM images of NCM622 electrode after the cycling test.

Table S3. Comparison of the electrochemical performances of various NCM622 cathodes.

Literature	Voltage range	1st discharge capacity	Cycle number	Capacity retention
This work	2.8-4.3V	179.5 mAh g^{-1}(0.2C) 177.4 mAh g^{-1}(1C)	100(1C)	88.3% (177.4/156.7 mAh g^{-1})
[1]	2.8-4.3V	143 mAh g^{-1}(1C)	100(1C)	87% (150/130.5 mAh g^{-1})
[2]	2.8-4.3V	152.0 mAh g^{-1}(1C)	100(1C)	81.5% (123/100.7 mAh g^{-1})
[3]	2.8-4.3V	189.1 mAh g^{-1}(0.1C)	100(1C)	96.5%(144.2/139.2 mAh g^{-1})
[4]	2.7-4.4V	165.1 mAh g^{-1}(1C)	200(1C)	84.9%(165.1/140.1 mAh g^{-1})
[5]	2.8-4.4V	171.2 mAh g^{-1}(0.5C)	300(0.5C)	99.7%(171.2/170.7 mAh g^{-1})
[6]	2.8-4.3V	153.8 mAh g^{-1}(0.5C)	100(0.5C)	99.1%(153.8/152.4 mAh g^{-1})

References.

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