Supplementary Information

Experimental

The measurement of Ni and Mn contents in $LiNi_{0.5}Mn_{1.5}O_4$ sample

A chemical titration method was adopted to measure the content of Ni and Mn in $LiNi_{0.5}Mn_{1.5}O_4$ sample. First, the total amount of Ni and Mn was determined by using EDTA titration. Second, the Mn content was titrated through ferrous ammonium sulfate titration. Finally, the Ni content can be dealt with the subtraction method. The specific procedures are as follows.

(i) Determination of Ni and Mn content: About 0.5 g of $LiNi_{0.5}Mn_{1.5}O_4$ powder was weighed accurately and dissolved in 20 ml concentrated hydrochloric acid under heating condition. The solution was fixed in 200 ml volumetric flask, and 20 ml solution was removed with pipette to 250 ml beater, and 3 ml ascorbic acid solution and 10 ml NH_3-NH_4Cl buffer solution (pH=10) were added to the beater. Add water to 150 ml, shake well, heat to ~ 40 °C and add 0.05~0.1 g ammonium purpurate as indicator. The thus-obtained solution was titrated with EDTA standard solution with color changing from yellow to pale purple, which is the end point of titration.

(ii) Determination of Mn content: Another 20 ml solution was removed with pipette from the above 200 ml volumetric flask to 250 ml conical flask. 5 ml phosphoric acid was added and then heated to ~ 50 °C. 5 ml perchlorate was added to the solution, which was heated up to take a lot of smoke and then removed to cool a little bit (to make all Mn^{2+} ions be oxidized to Mn^{3+} ions). Add 60 ml dilute sulphuric acid solution (H_2SO_4:H_2O=1:19, v:v), shake well, and then cool to room temperature. With ammonium ferrous sulfate standard solution titration to reddish color, 2 drops of N-phenylanthranilic acid was added as indicator, and ammonium ferrous sulfate standard solution was continued for titration with color changing from cherry red to light yellow, which is the end point of titration.

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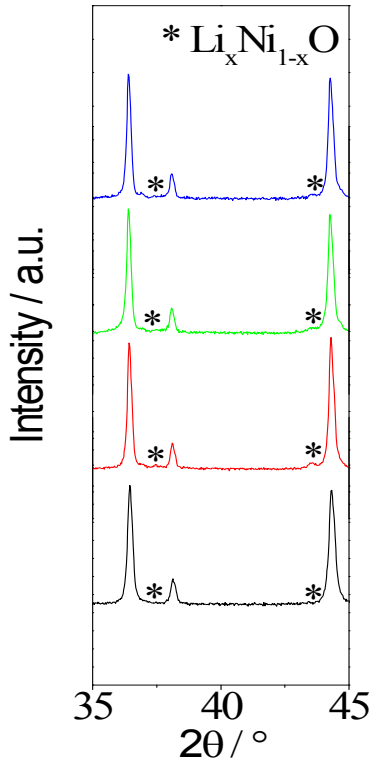


Fig. S1 Local enlarged image of XRD pattern between 35° and 45°

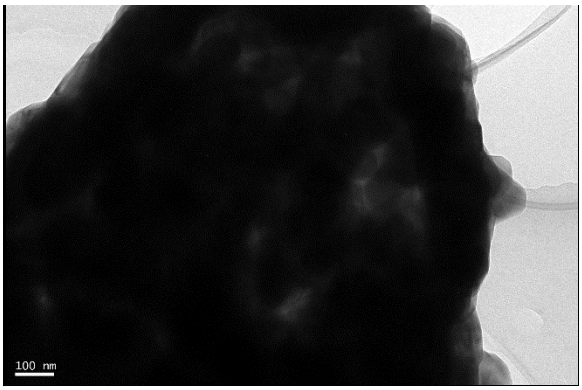


Fig. S2 TEM image of LNMO-2.0 sample

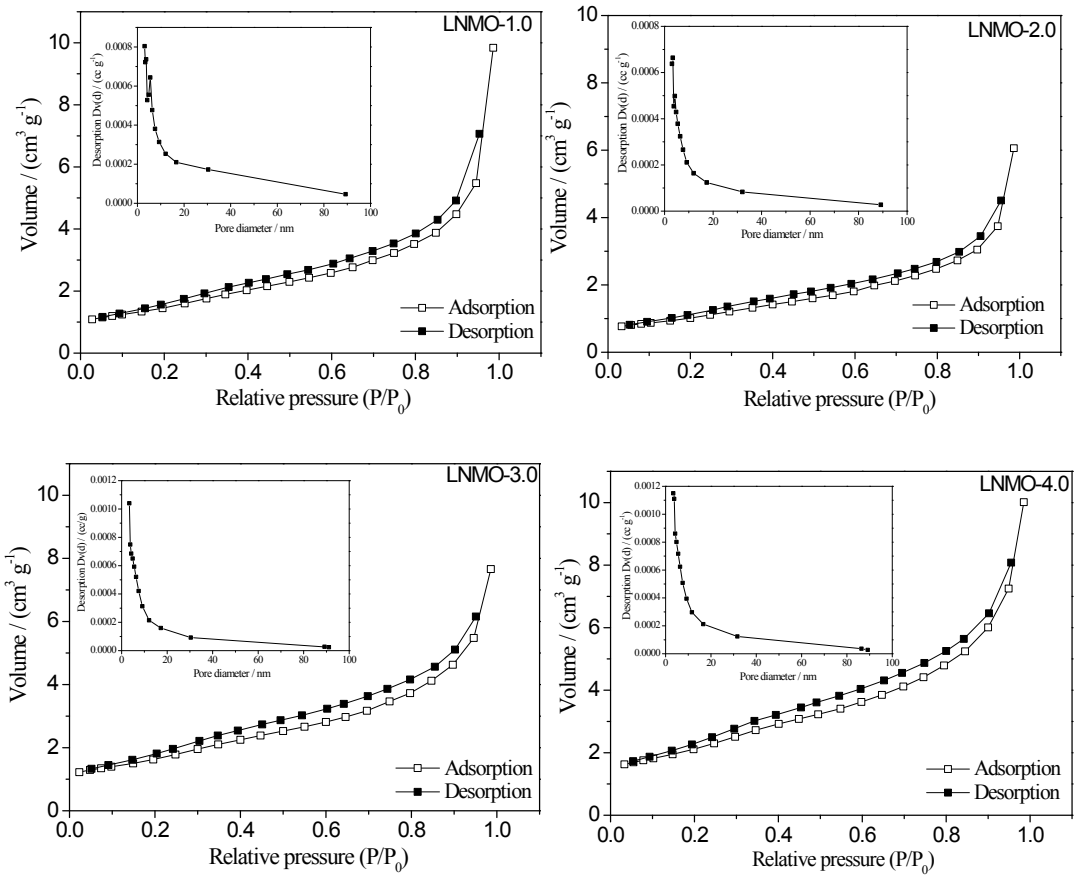


Fig. S3 Nitrogen adsorption-desorption isotherms and pore size distribution for $LiNi_{0.5}Mn_{1.5}O_4$ samples synthesized at different U/TM ratios

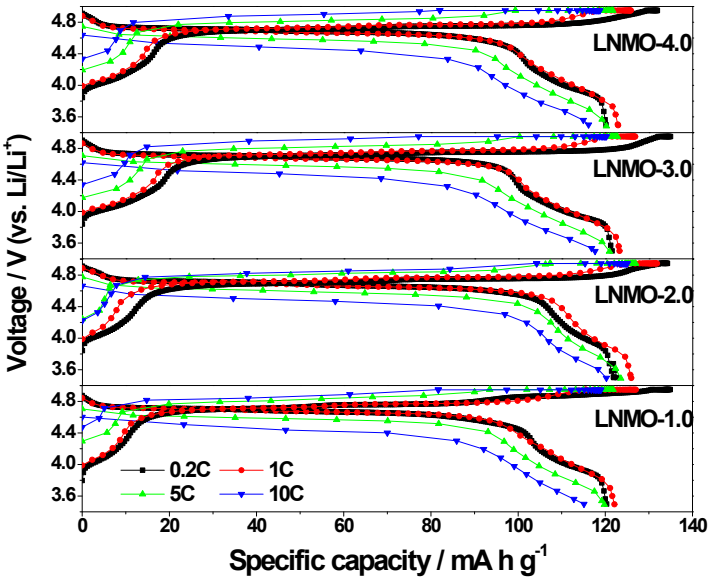


Fig. S4 Charge/discharge curves of all samples at various rates

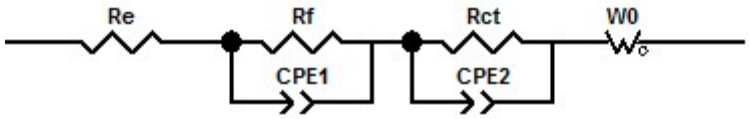
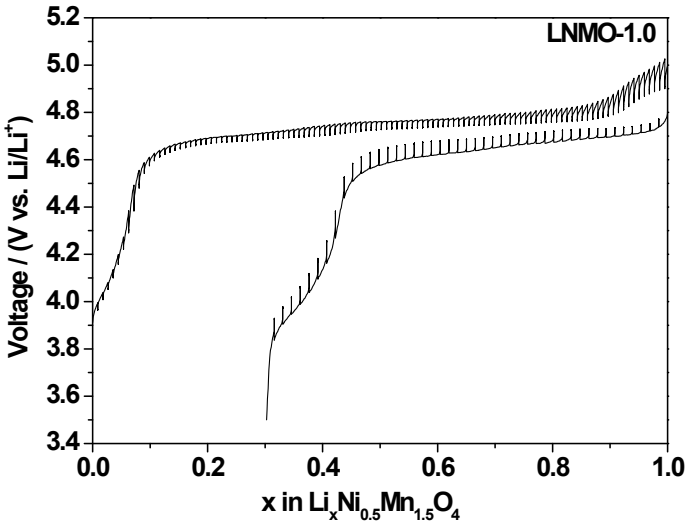
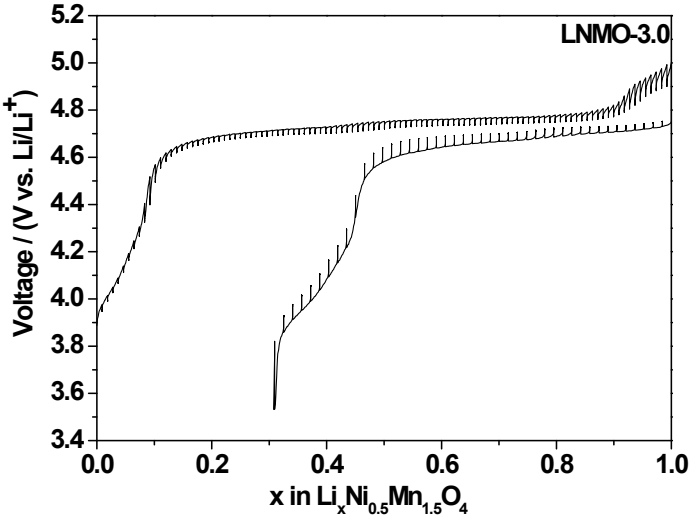


Fig. S5 Equivalent circuit for EIS spectra





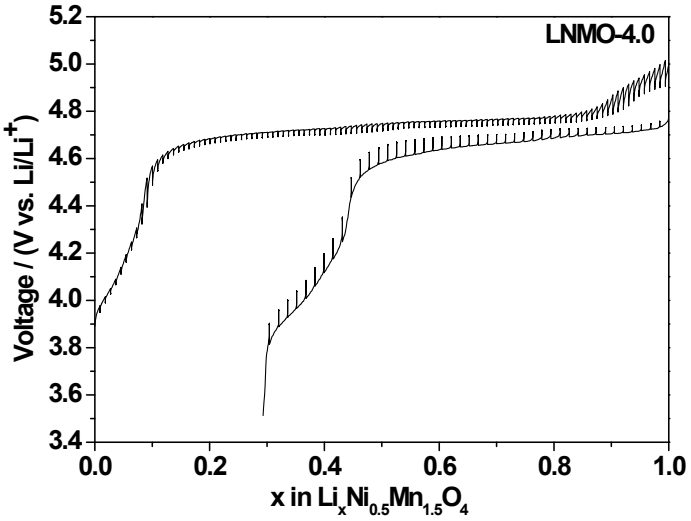


Fig. S6 GITT curves of LNMO-1.0, LNMO-3.0 and LNMO-4.0 samples

Table S1 The molar ratio results of Ni and Mn obtained by chemical titration method

Sample	LNMO-1.0	LNMO-2.0	LNMO-3.0	LNMO-4.0
Ni	0.4690	0.4952	0.4940	0.4874
Mn	1.5	1.5	1.5	1.5

Table S2 Values of the CV peaks for all samples

Sample		φ_a / V	φ_c / V	$\Delta\varphi$ / mV
LNMO-1.0	Ni^{2+}/Ni^{3+}	4.746	4.646	125
	Ni^{3+}/Ni^{4+}	4.796	4.662	146
LNMO-2.0	Ni^{2+}/Ni^{3+}	4.745	4.649	96
	Ni^{3+}/Ni^{4+}	4.795	4.679	116
LNMO-3.0	Ni^{2+}/Ni^{3+}	4.763	4.638	108
	Ni^{3+}/Ni^{4+}	4.798	4.671	121
LNMO-4.0	Ni^{2+}/Ni^{3+}	4.762	4.645	115
	Ni^{3+}/Ni^{4+}	4.804	4.674	130