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Supplementary Materials

Improving the Structural Ordering and Particle-Size Homogeneity of Li-Rich Layered $Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$ Cathode Materials through Microwave Irradiation Solid-State Synthesis

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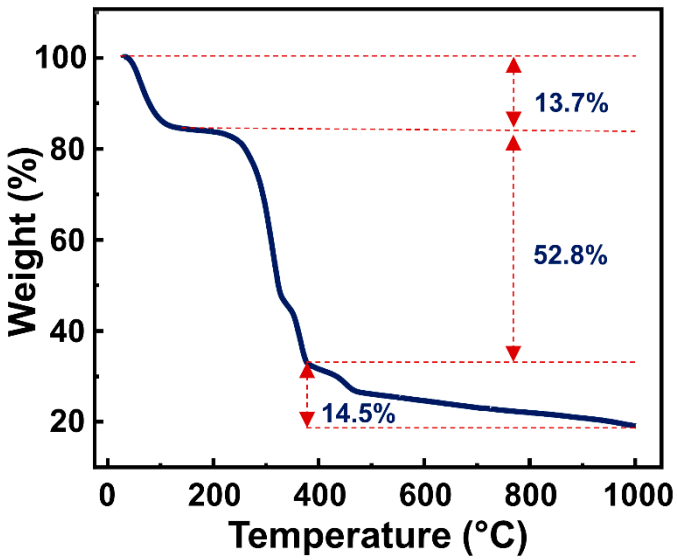

Figure S1. TG curve of the precursor of the LNCM sample.

The TG measurement of the precursor mixture was conducted in the range of 30 – 1000 °C. In the first stage, in the range of 30 – 100 °C, 13.7%

weight loss is related to the removal of adsorbed water in the precursor. In the temperature range of 300 – 395 °C, the weight loss of around 52.8% corresponds to the decomposition of organic components (i.e., acetate) [57]. The process is completed around 400 °C. The third stage of the weight loss (14.5%) at 395 – 1000 °C corresponds to the decomposition of

Table S1. Comparison of Structural ordering with the other works.

No.	Synthesis method / strategy	Type of Li-rich cathode	Intensity ratio $I_{(003)}/I_{(104)}$	R factor	Reference
1.	Ultrasonic/microwave-assisted co-precipitation method	$Li_{1.1}Ni_{0.233}Co_{0.233}Mn_{0.433}O_2$	1.538	-	[28]
2.	Microwave-assisted hydrothermal method	$xLi_2MnO_3\cdot(1-x)LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	1.270	-	[29]
3.	Microwave hydrothermal method	$Li_{1.2}Ni_{0.16}Co_{0.08}Mn_{0.56}O_2$	1.454	0.2561	[30]
4.	Coprecipitation / Fe and PO_4^{3-} co-doped	$Li_{1.2}Mn_{0.56}Ni_{0.16}Co_{0.08}O_2$	1.400	-	[59]
5.	Coprecipitation / S doped	$Li_{1.2}Ni_{0.2}Mn_{0.6}O_2$	1.320	-	[60]
6.	Sol-gel / Mg-Al codoped	$Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$	1.330	-	[61]
7.	Solid-state/ C coated	$Li_{1.15}(Ni_{0.23}Co_{0.08}Mn_{0.54})O_2$	1.580	0.3000	[62]
8.	Coprecipitation / $LaNiO_3$ coating	$Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$	1.370	0.3000	[63]
9.	Microwave-assisted solid-state method	$Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$	2.010	0.4011	**This work**

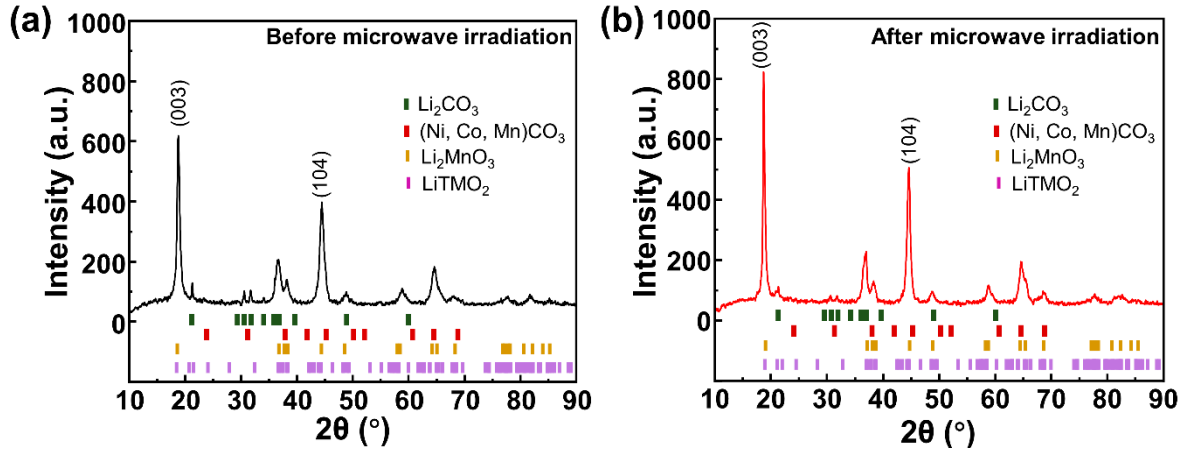

Figure S2. XRD patterns of LNCM sampel after the first calcination step, (a) before microwave treatment and (b) after microwave treatment for 20 minutes.

Table S2. Full-width at half maximum (FWHM) of the typical XRD diffraction peak of LNCM sample after the first calcination step, before and after microwave treatment for 20 minutes.

Sample	FWHM	
	(003)	(104)
Before microwave treatment	0.643°	0.902°
After microwave treatment	0.365°	0.713°

The microwave irradiation treatment affects to the structure of the LNCM samples. Figure S2 shows the amorphorous oxide of both samples. The microwave treatment can reduce the containing of Li_2CO_3 and $(Ni,Co,Mn)CO_3$ that is showed by the clear reduction peak of Li_2CO_3 and $(Ni,Co,Mn)CO_3$ after the treatment. The decomposition of Li_2CO_3 and $(Ni,Co,Mn)CO_3$ products probably increases the formation $Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$ (LNCM) after microwave irradiation as indicated by the increasing the intesity of dominant peaks and crystalinity (showed by the narrowing of FWHM, as shown in Table S2) [64]. Furthermore,

microwave irradiation treatment potentially increase the cation ordering of Li^+/Ni^{2+} indicated by the increasing of intensity ratio of $I_{(003)}/I_{(104)}$ (intensity ratio of $I_{(003)}/I_{(104)}$ before and after microwave treatment = 1.515 and 1.563, respectively), as shown by the Figure S2.

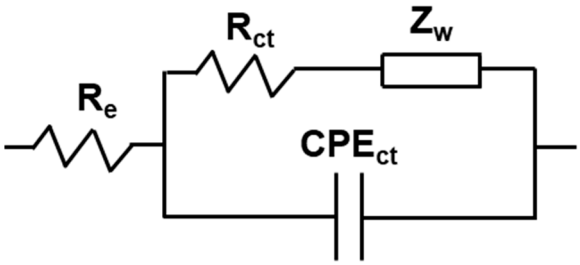

Figure S3. Basic equivalent circuit used to model the EIS results.

Table S3. Electrochemical impedance spectroscopy (EIS) fitting results of LNCM cathode materials.

No.	Sample	R_s (Ω)	R_{ct} (Ω)
1.	w/o microwave	3.27	234.89
2.	10 minutes	3.56	218.24
3.	20 minutes	3.89	186.57
4.	30 minutes	3.13	275.44

Table S4. Comparison of electrochemical performance with the other works.

No.	Synthesis method	Type of Li-rich cathode	Discharge capacity (Current density)	Capacity retention	Reference
1.	Ultrasonic/microwave-assisted co-precipitation method	$Li_{1.1}Ni_{0.233}Co_{0.233}Mn_{0.433}O_2$	260 mAh/g (0.1 C)	79.61% 50 cycles	[28]
2.	Microwave-assisted hydrothermal method	$xLi_2MnO_3\cdot(1-x)LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$	325 mAh/g (0.1 C)	72.2% 50 cycles	[29]
3.	Microwave hydrothermal method	$Li_{1.2}Ni_{0.16}Co_{0.08}Mn_{0.56}O_2$	159.3 mAh/g (2000 mA/g)	88.6% 300 cycles	[30]
4.	Coprecipitation and Microwave pre-activation	$Li(Li_{0.21}Ni_{0.131}Co_{122}Mn_{0.538})O_2$	268.6 mAh/g (25 mA/g)	81.1% 200 cycles	[65]
5.	Sol-gel	$Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$	245 mAh/g (20 mA/g)	68.9 % 100 cycles	[66]
6.	Microwave-assisted solid-state method	$Li_{1.2}Ni_{0.13}Co_{0.13}Mn_{0.54}O_2$	259.84 mAh/g (20 mA/g)	80.57% 200 cycles	**This work**