Supporting Information

Graphene-based surface modification on layered Li-rich cathode for high-performance Li-ion batteries

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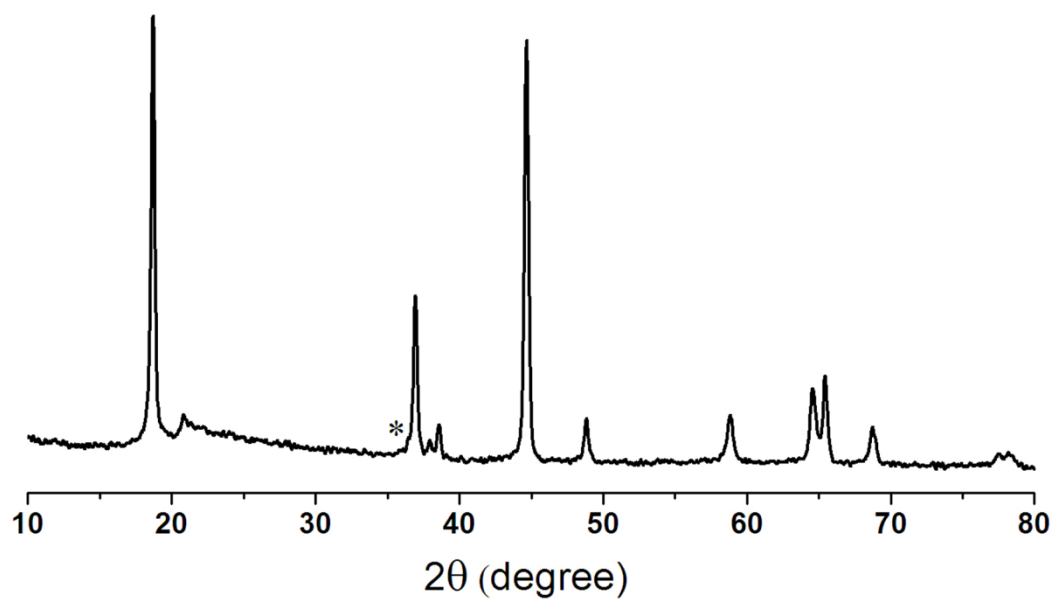
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Figure S1. XRD pattern of LLNCM/LAA-350 sample including a spinel-like phase labeled as *.
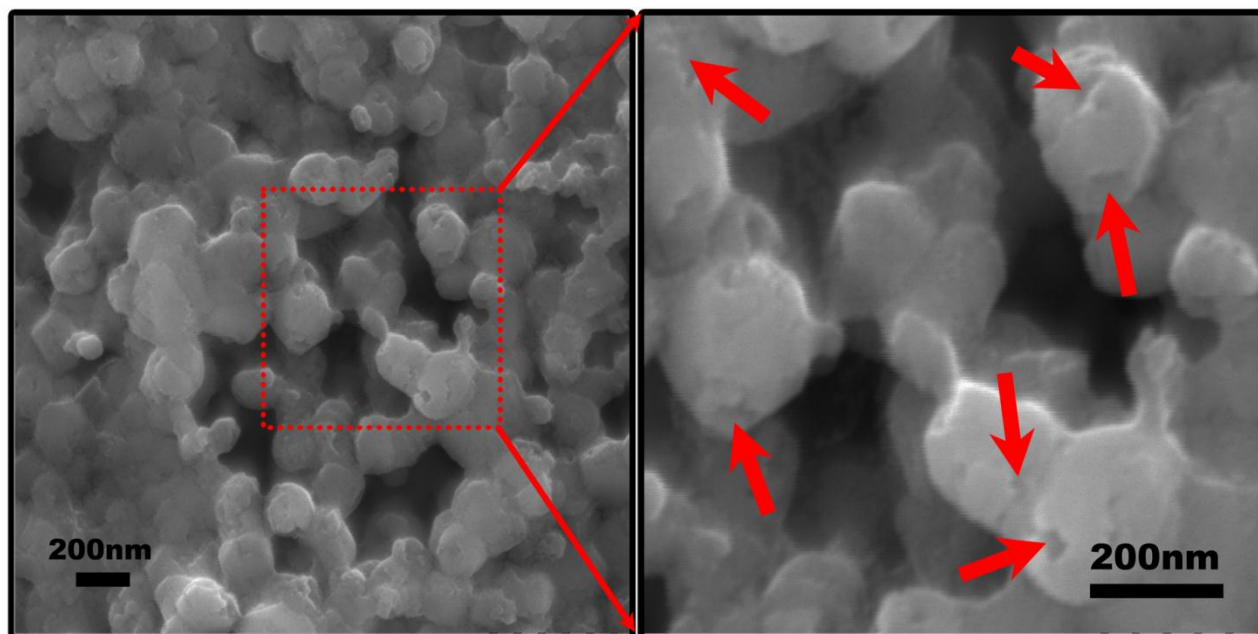
Figure S2. SEM images of LLNCM/LAA-350 powders in which the arrows indicate the collapse regions of particle surfaces after LAA and the following 350 °C heat treatment.
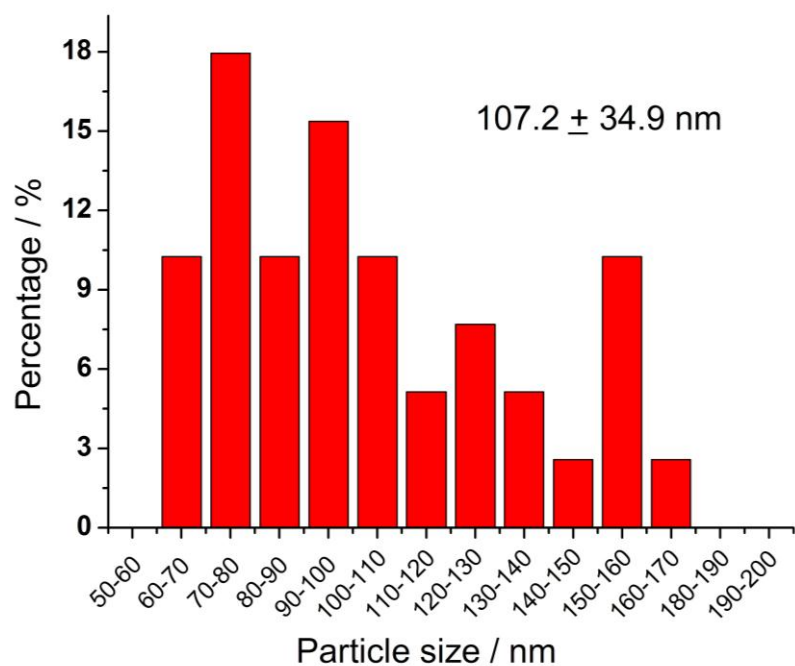
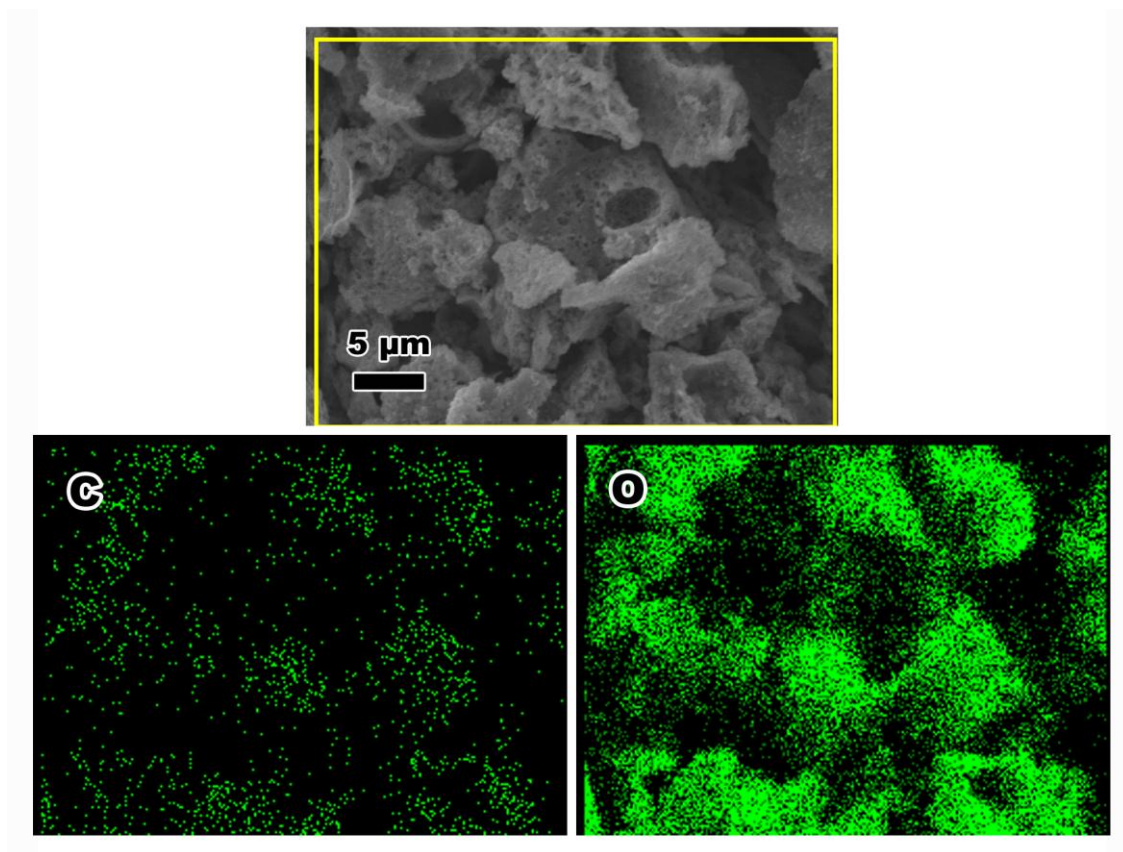**Figure S3.** Particle size distributions of bare $Li(Li_{0.2}Mn_{0.54}Ni_{0.13}Co_{0.13})O_2$.

Figure S4. SEM image and corresponding carbon and oxygen elemental mapping of LNCMO/GO composite.
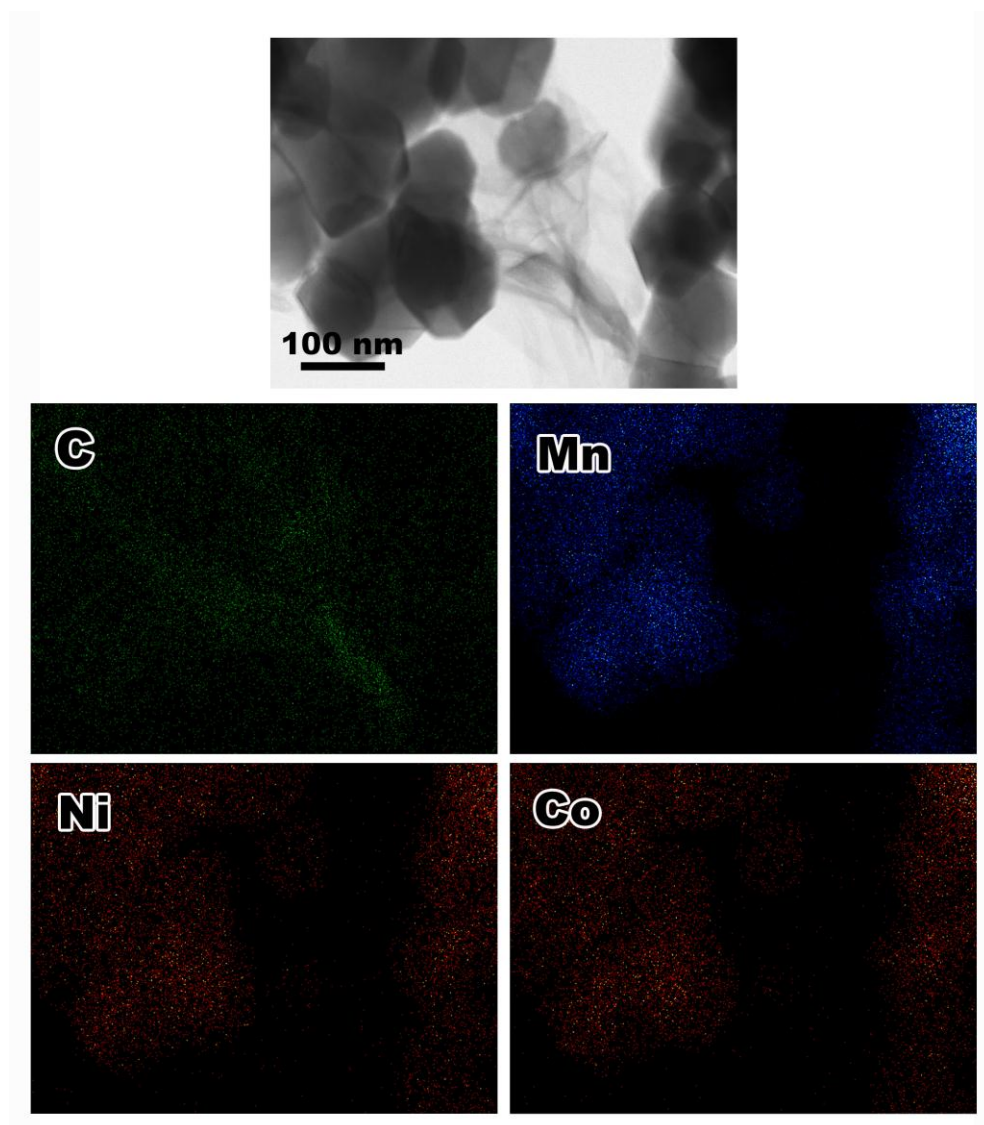
Figure S5. TEM image and corresponding carbon, manganese, nickel and cobalt elemental mapping of LLNCM/GO composite.
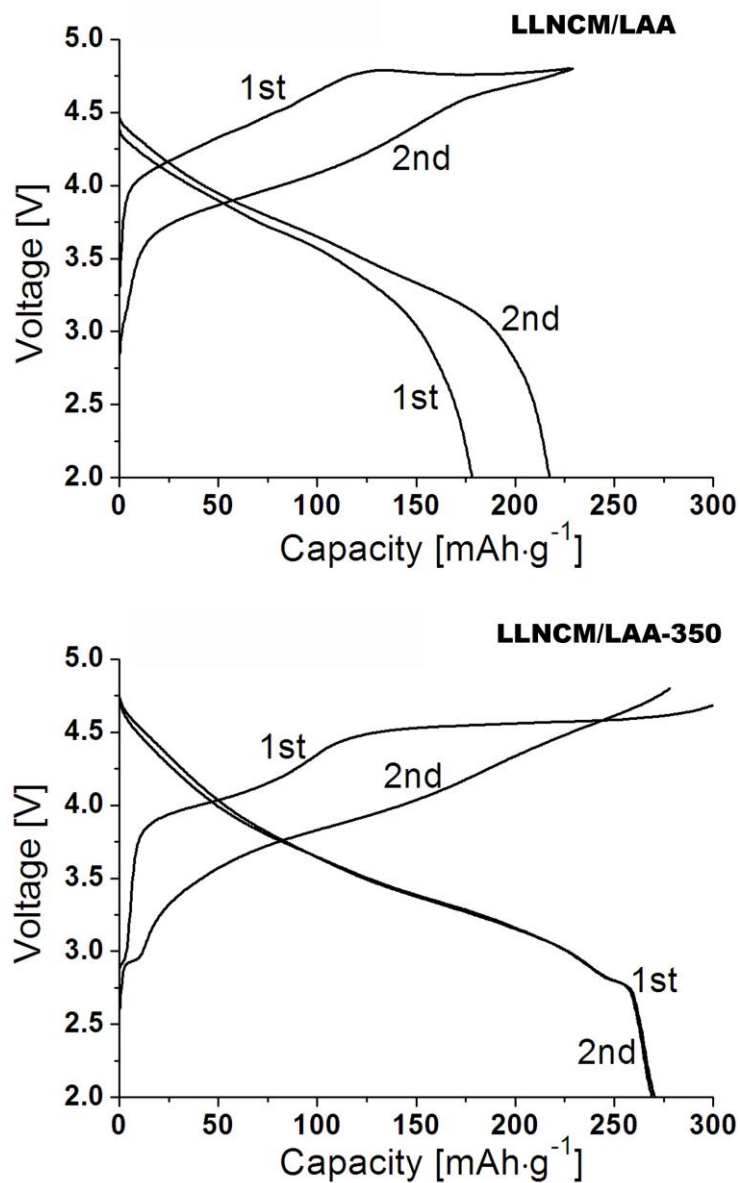
Figure S6. First and second charge/discharge curves of LLNCM/LAA and LLNCM/LAA-350 samples

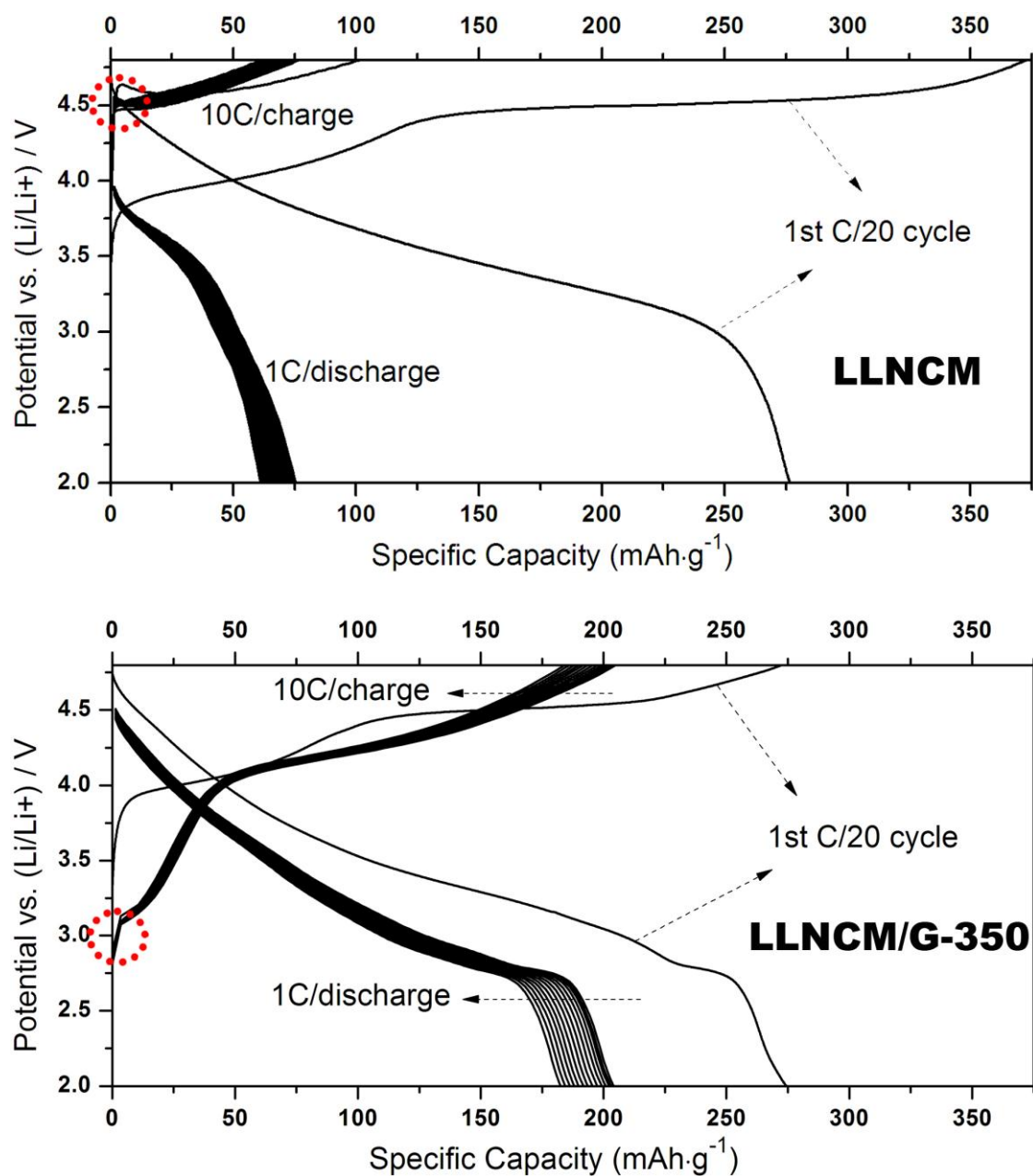cycled between 2.0 and 4.8 V at 50 mA·g^{-1} (0.2 C).

Figure S7. 10C-charge/1C-discharge curves after a first forming cycle of LLNCM/G-350 and LLNCM

samples cycled between 2.0 and 4.8 V where 1 C corresponds to 250 mA·g^{-1}. The red dot cycles

indicate the various polarization effects caused by the high charging rate of 10 C.
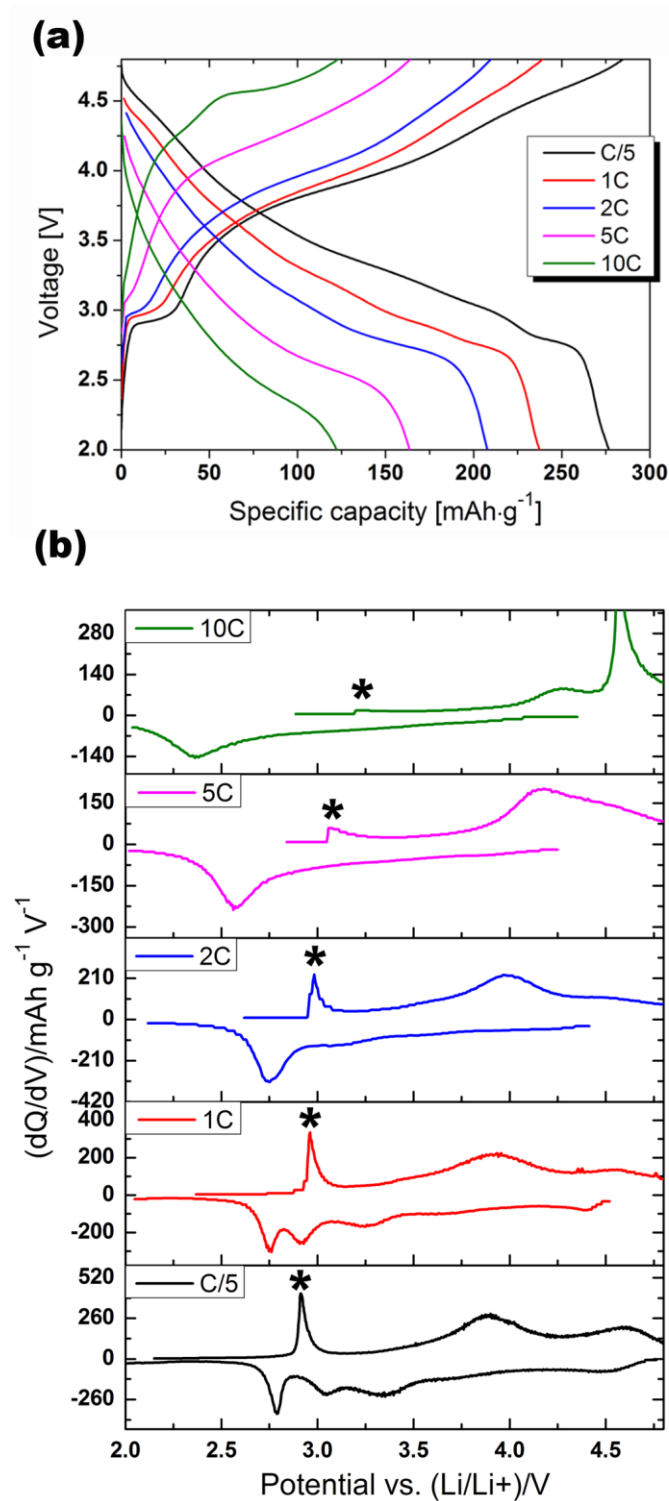
Figure S8. Charge/discharge curves (a) and corresponding dQ/dV plots (b) of LLNCM/G-350 sample obtained from the same testing of Fig. 9 (d). Symbol ∗ indicates the charging peaks resulted from the Li reinsertion into surface-spinel framework.

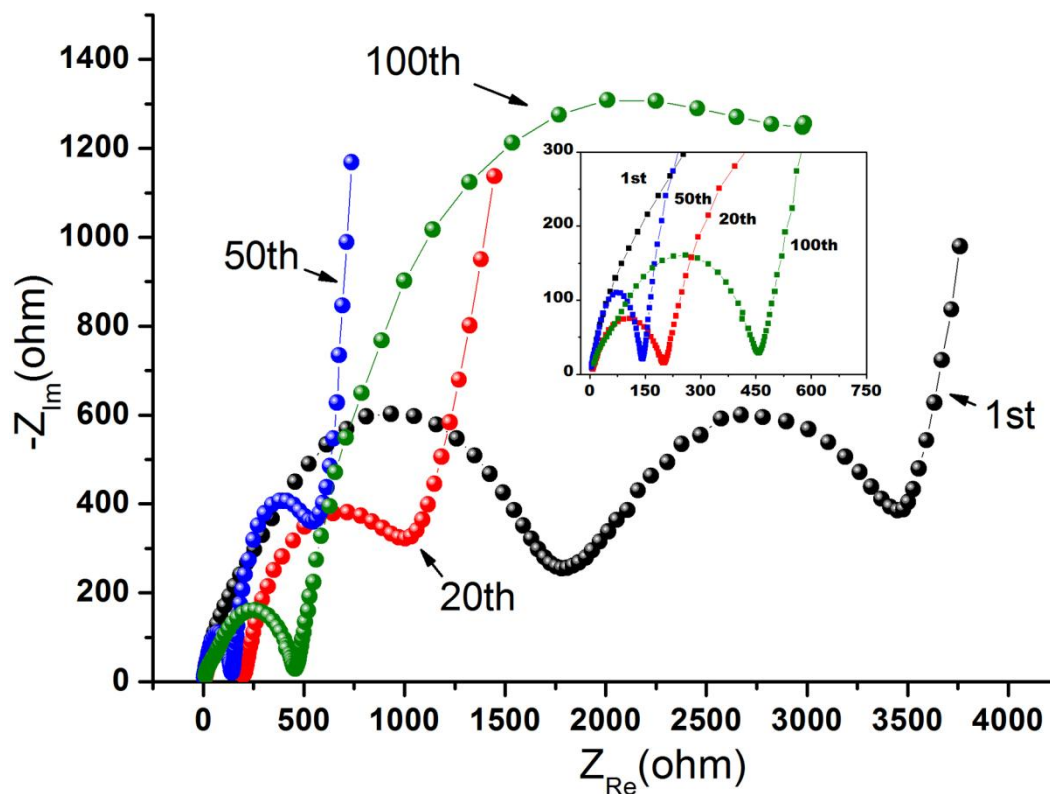**Figure S9.** EIS spectra of cycled LLNCM/GO cathode material at 1st, 20th, 50th and 100th cycles. Before

the EIS measurement, the half cell was charged to the cutoff voltage of 4.8 V, then discharged to 3.5 V

at 0.05 C, and held at 3.5 V for another 3 h. In between of two individual cycles, 2 C current density

was used to cycle the half cell where 1 C stands for 250 mA·g^{-1}.

Table S1. Surface quantification of chemical composition for LLNCM, LLNCM/GO, LLNCM/G-250

and LLNCM/G-350 samples, as analyzed from XPS results.

	Li	Transition metals				O
		Mn	Ni	Co	Total	
LLNCM	**1.01**	0.53	0.23	0.23	**0.99**	2.8
LLNCM/GO	**0.76**	0.68	0.34	0.22	**1.24**	5.3
LLNCM/G-250	**0.66**	0.94	0.25	0.16	**1.35**	3.1
LLNCM/G-350	**0.64**	1.1	0.15	0.13	**1.38**	3.4

Table S2. Charge/discharge capacities at various cycling states of LLNCM, LLNCM/GO, LLNCM/G-250 and LLNCM/G-350 cathodes, which were cycled between 2.0 and 4.8 V at 50 mA·g^{-1}.

Sample	1st cycle (mAh·g^{-1})		3rd cycle (mAh·g^{-1})		5th cycle (mAh·g^{-1})		20th cycle (mAh·g^{-1})		50th cycle (mAh·g^{-1})	
	charge	discharge	charge	discharge	charge	discharge	charge	discharge	charge	discharge
LLNCM	300	253	247	243	247	243	217	216	181	181
LLNCM/GO	263	238	250	244	241	237	203	201	191	189
LLNCM/G-250	261	262	262	257	257	252	232	229	197	194
LLNCM/G-350	260	268	270	264	266	260	240	236	206	203