Supporting information

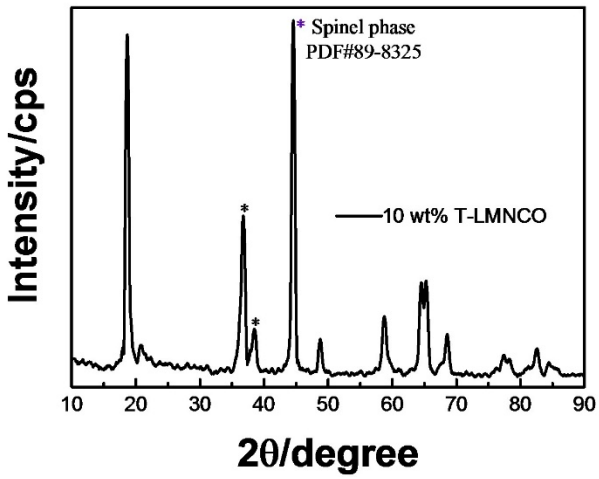

Fig.S1 XRD patterns of 10 wt% T-LMNCO sample

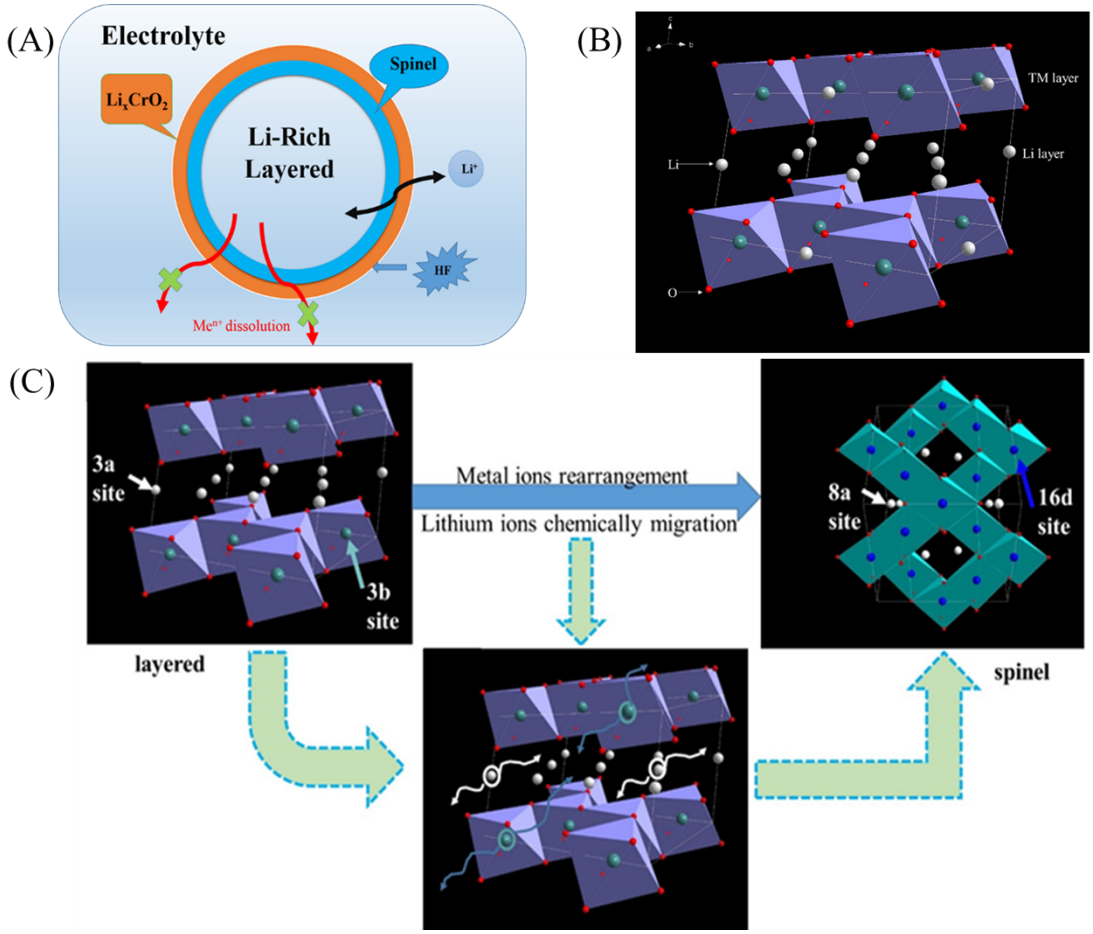

Fig. S2 Schematic view of the Layered@Spinel@Coating layer heterostructure (A); schematic

diagram of the lithium-rich layer cathode material (B); scheme of layered-to-spinel structural transformation (C).

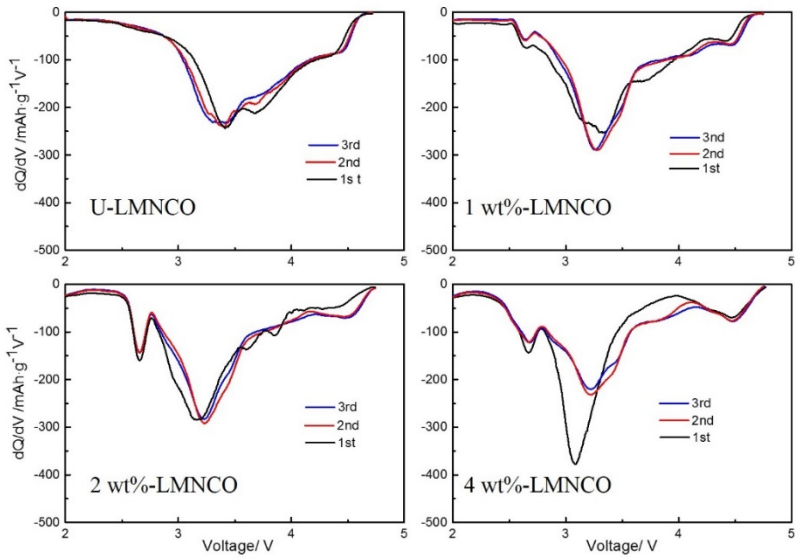

Fig. S3 dQ/dV curves of all the samples for three cycles at 0.1 C rate

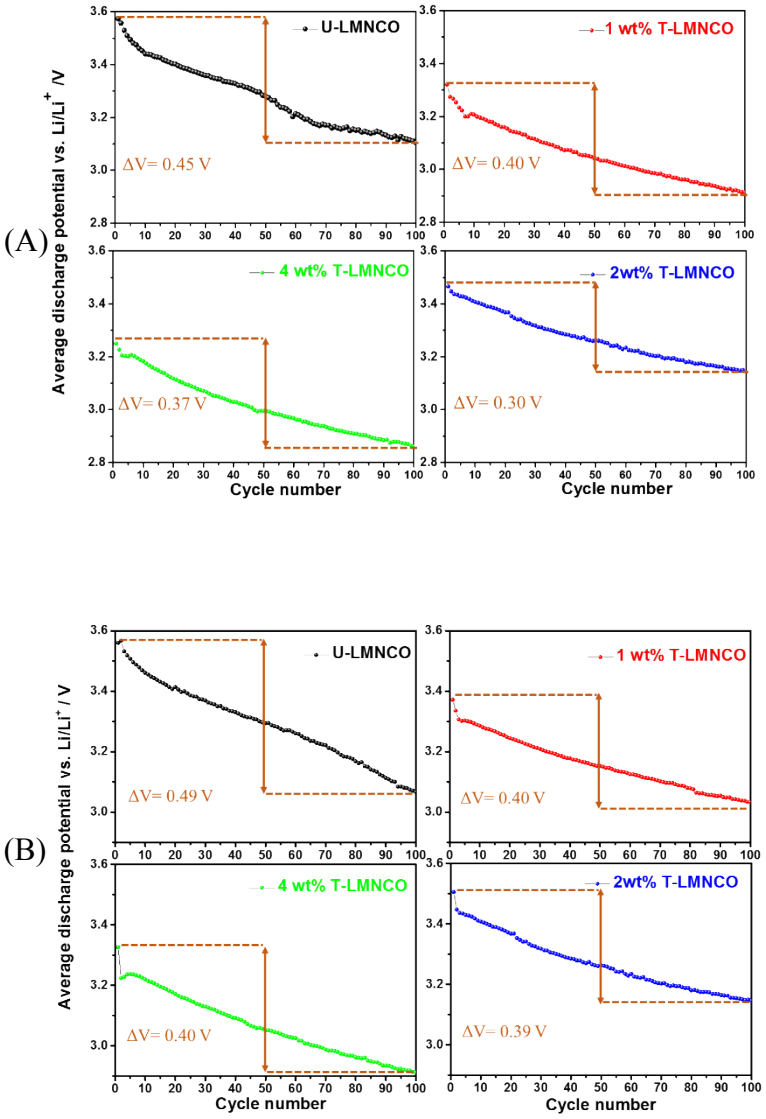

Fig. S4 Comparison of voltage fading rates of the cells with the synthesized cathode materials during cycling at 0.5 C (A) and 1 C rate (B).

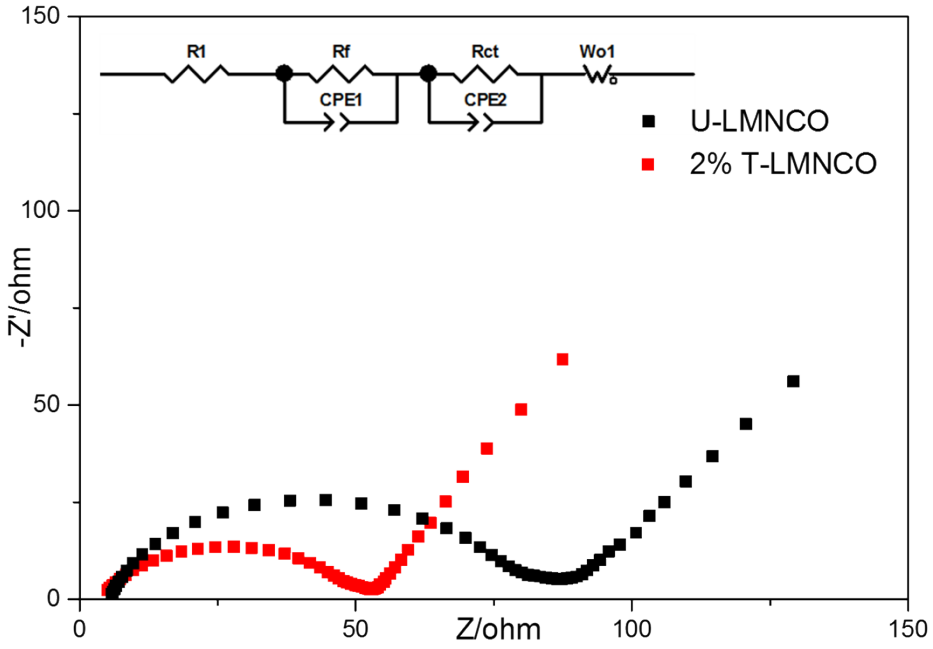

Fig. S5 EIS spectrum of U-LMNCO and 2% T-LMNCO charged to 4.8 V after first cycle at 0.1 C

Two overlapped semicircles were observed due to different contributions of interfacial resistances. According to previous studies [1-2], the semicircle at high to middle frequency represents the resistance of the surface film (R_f), whereas the semicircle at middle to low frequency is attributed to charge transfer resistance at the electro-electrolyte interface (R_{ct}). The cells assembled with surface-modified $Li_{1.2}Mn_{0.52}Ni_{0.13}Co_{0.13}O_2$ exhibited much lower interfacial resistances than the cell with pristine $Li_{1.2}Mn_{0.52}Ni_{0.13}Co_{0.13}O_2$.

References:

1. C.H. Chen, J. Liu, K. Amine, Journal of Power Sources, 2001, 96, 321

2. Y.W. T. A. Arunkumar, A. Manthiram, Chem. Mater, 2007, 19, 3067

To investigate the amount of metal dissolved into the electrolyte, the experiment proceed is as follows. The CR2016 cell battery is disassembled in the glove box after 30 cycles at 1 C rate, and then, soaked into the right amount of DMC (Dimethyl carbonate) for several hours. Take a certain amount of this DMC mixed solution, treated by same acid. The last, the data is obtained by ICP test. The value calculated is based on the active materials.

Table.S1 The amount of metal dissolved into the electrolyte

Dissolution after 30 cycles	U-LMNCO	1 wt% T-LMNCO	2 wt% T-LMNCO	4 wt% T-LMNCO
Mn/ %	0.40	0.12	0.02	0.02
Co/ %	0.19	0.1	0.01	0.01
Ni/ %	0.1	0.06	0	0
Cr/ %	0	0	0	0