Supporting Information for

Mitigating the capacity and voltage decay of lithium-rich layered oxide cathodes by fabricating Ni/Mn graded surface

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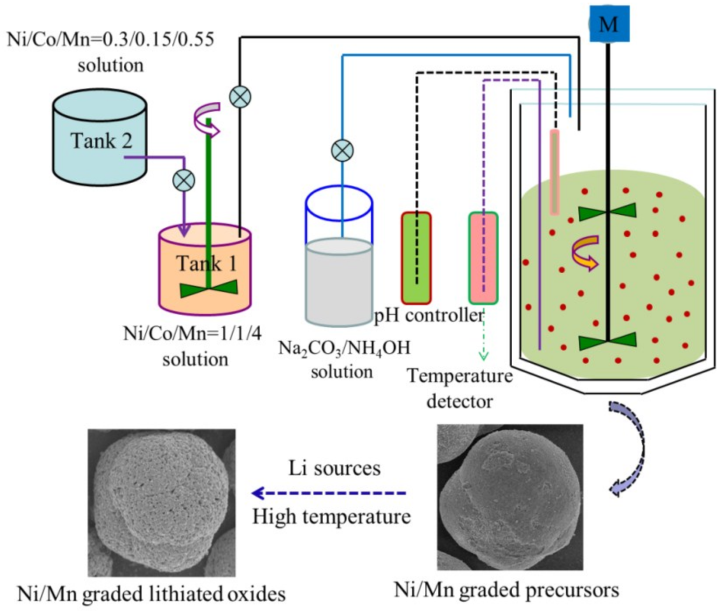

Figure S1 Schematic illustration of preparing the aimed Ni/Mn graded precursors and lithium-rich layered oxides *via* a developed co-precipitation route and solid-state reactions.

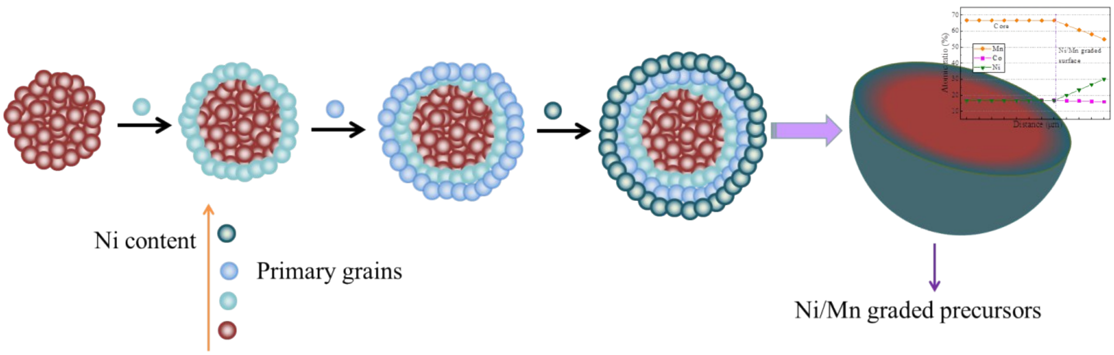

Figure S2 The growth mechanism of Ni/Mn graded micron-sized spherical secondary particles as precursors during co-precipitation process in the CSTR.

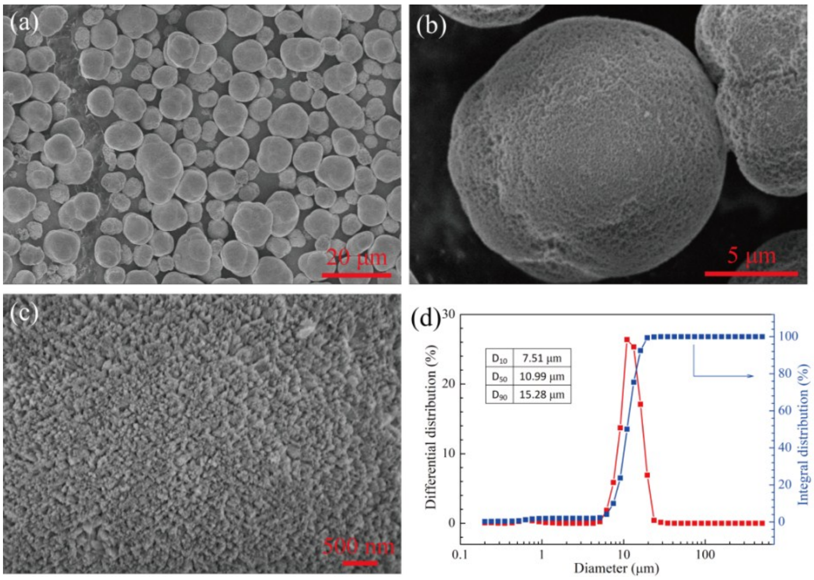

Figure S3 (a-c) SEM images and (d) particle size distribution of the normal precursors $[Ni_{0.183}Co_{0.167}Mn_{0.65}]CO_3$.

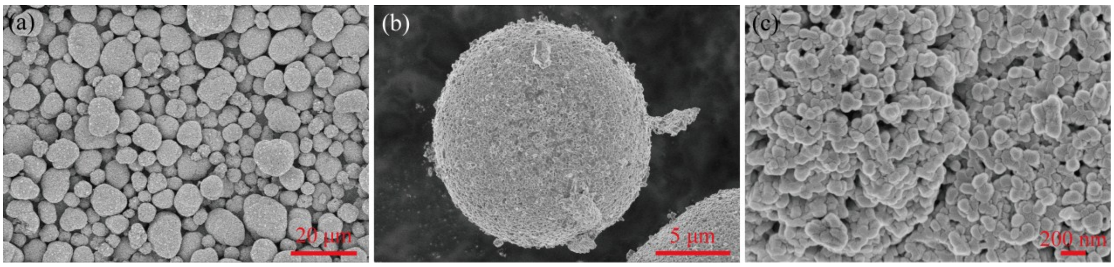

Figure S4 SEM images of the normal LLOs.

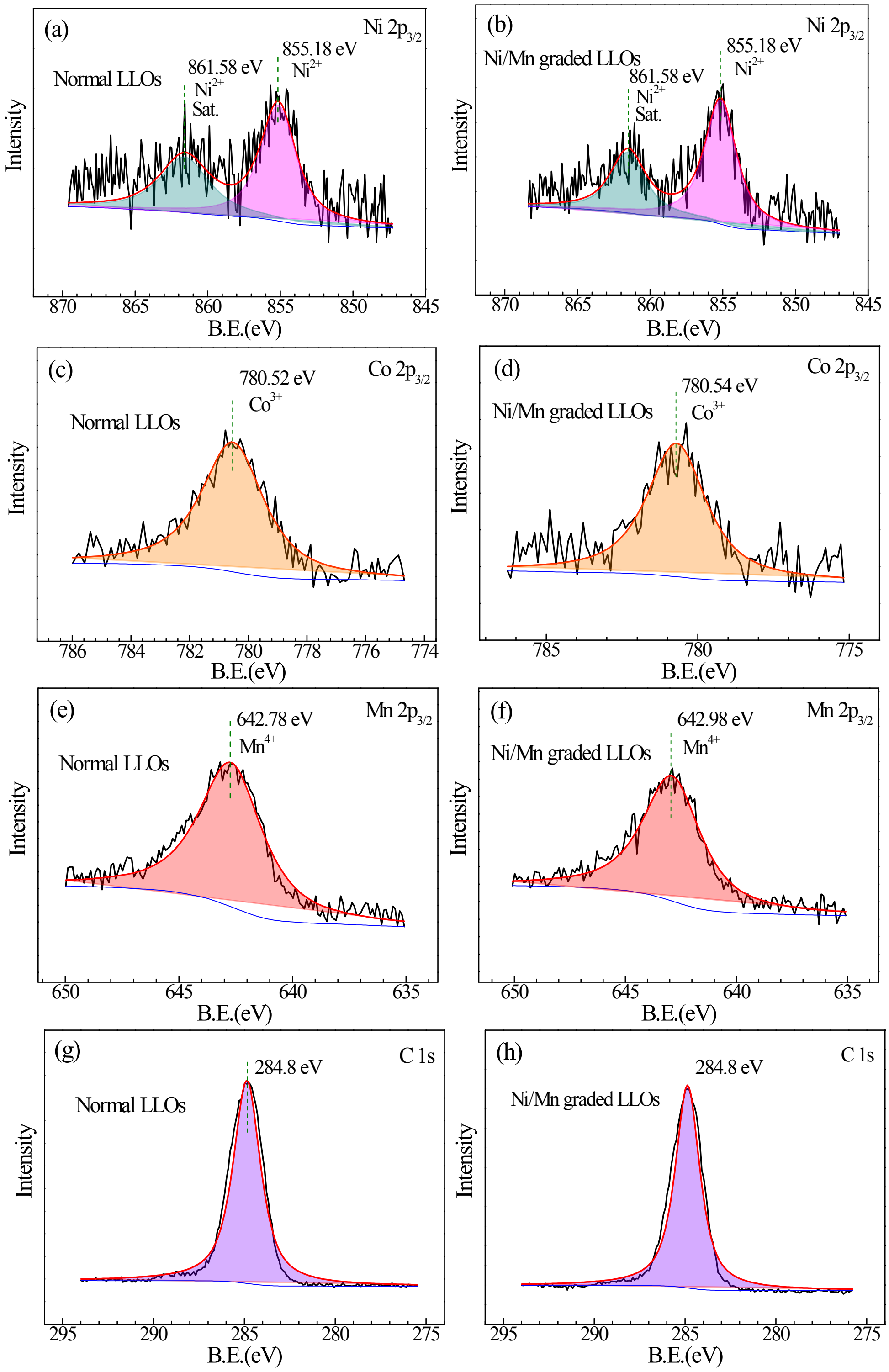

Figure S5 XPS of the (a,b) Ni $2p_{3/2}$, (c,d) Co $2p_{3/2}$, (e,f) Mn $2p_{3/2}$ and (g,h) C1s for the both pristine LLOs.

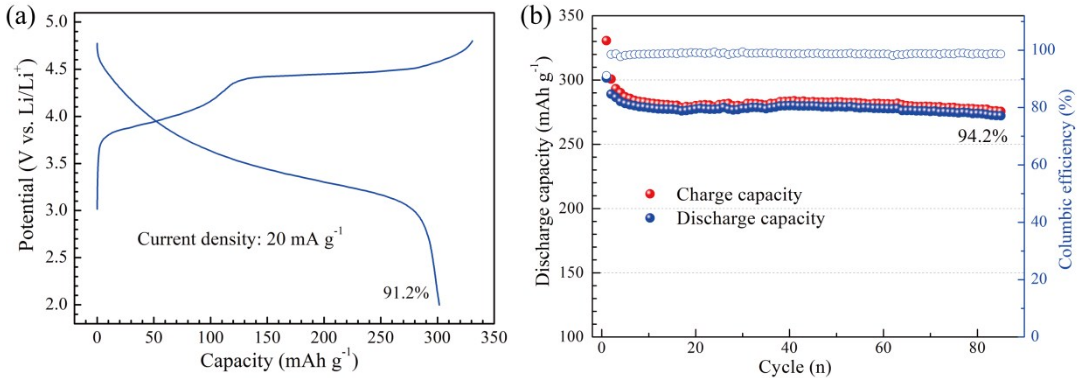

Figure S6 (a) The initial charge/discharge curves and (b) the cycling stability of the Ni/Mn graded LLO electrodes at high working temperature of 50 ℃.

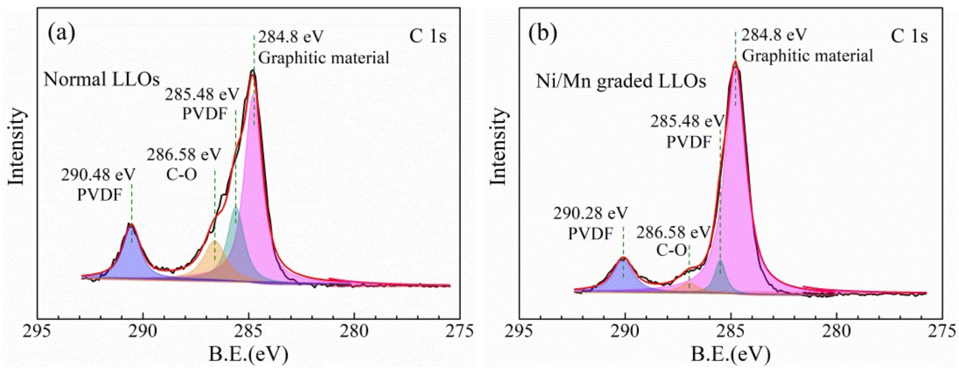

Figure S7 XPS of the C1s for both the cycled LLO electrodes in the fully discharged state after 200 cycles at a rate of 0.5C and 25 °C.

Table S1 The molar ratio of Ni/Co/Mn by ICP−AES for both as-prepared precursors.

Samples	Ni	Co	Mn
Normal precursors	0.183	0.167	0.650
Ni/Mn graded precursors	0.184	0.168	0.648