Electronic Supplementary Information

Atomic Layer Deposition of Solid State Electrolyte Coated Cathode Materials with Superior High-Voltage Cycling Behavior for Lithium Ion Battery Application

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Table S1 The atomic ratio between Ta and Ni in the coated cathode for different ALD cycles.

ALD-Cycles	**2**	**5**	**10**	**50**
Ta/Ni	0.0028	0.0043	0.0074	0.0337

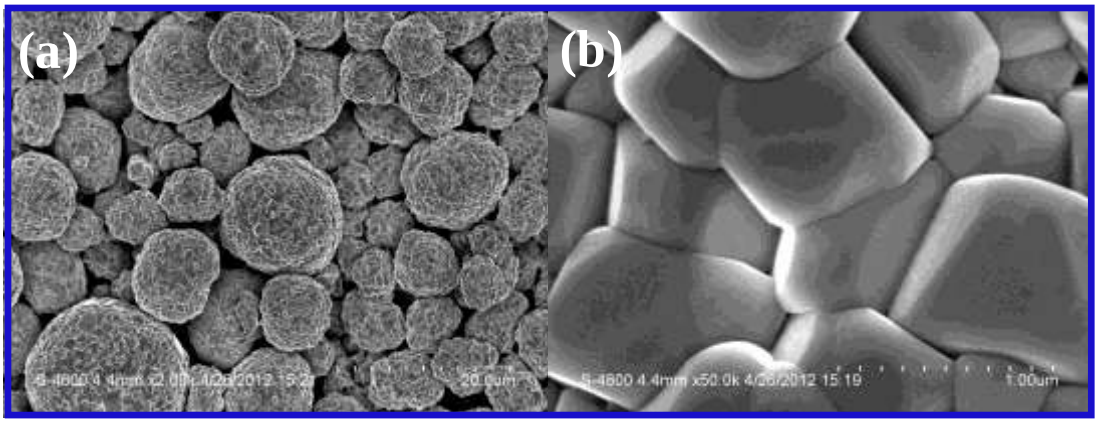

Figure S1 the typical morphologies of the pristine NMC cathode

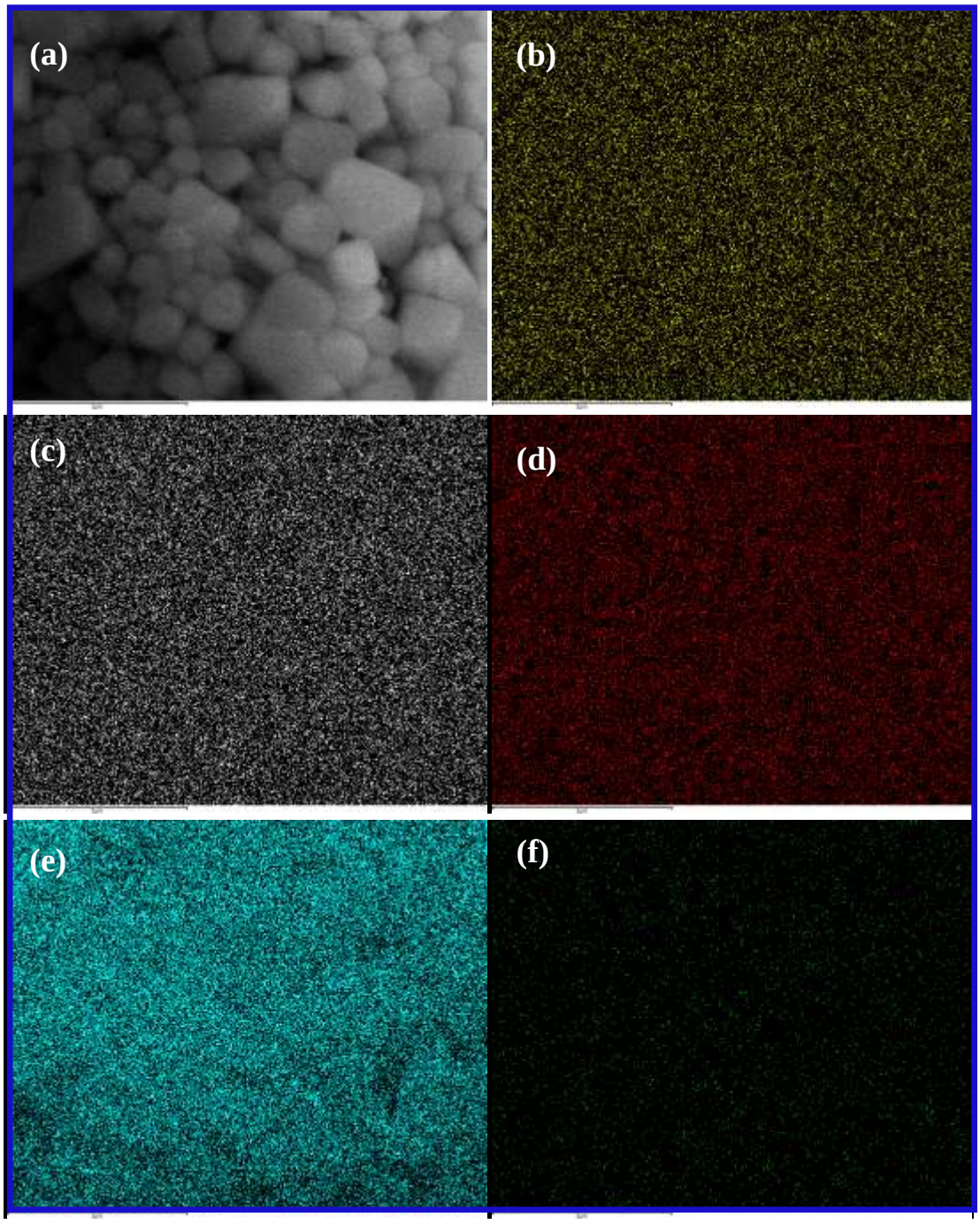

Figure S2 (a) SEM image and elemental mapping of NMC-2: (b) Ni, (c) Mn, (d) Co, (e) O, and (f) Ta.

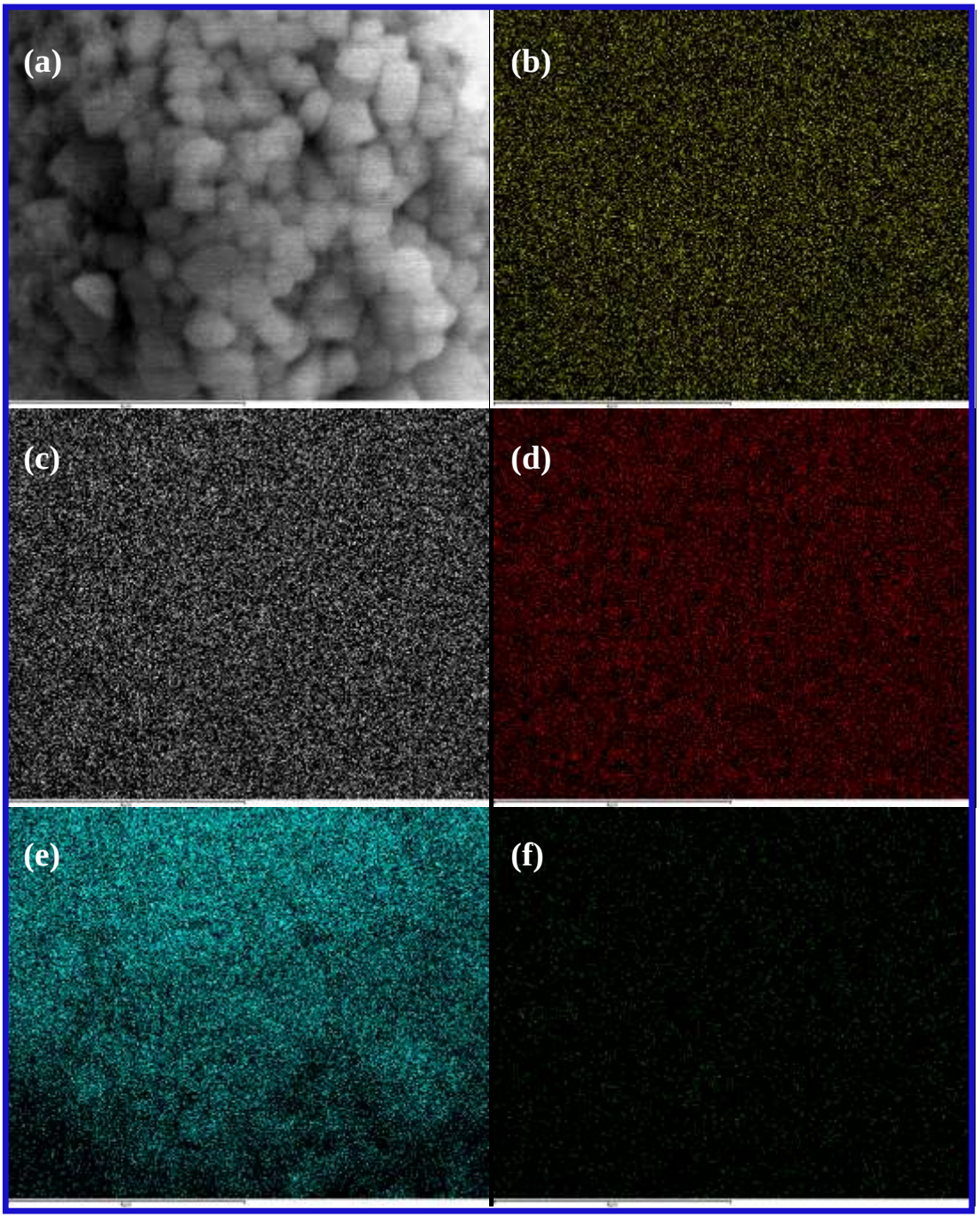

Figure S3 (a) SEM image and elemental mapping of NMC-5: (b) Ni, (c) Mn, (d) Co, (e) O, and (f) Ta.

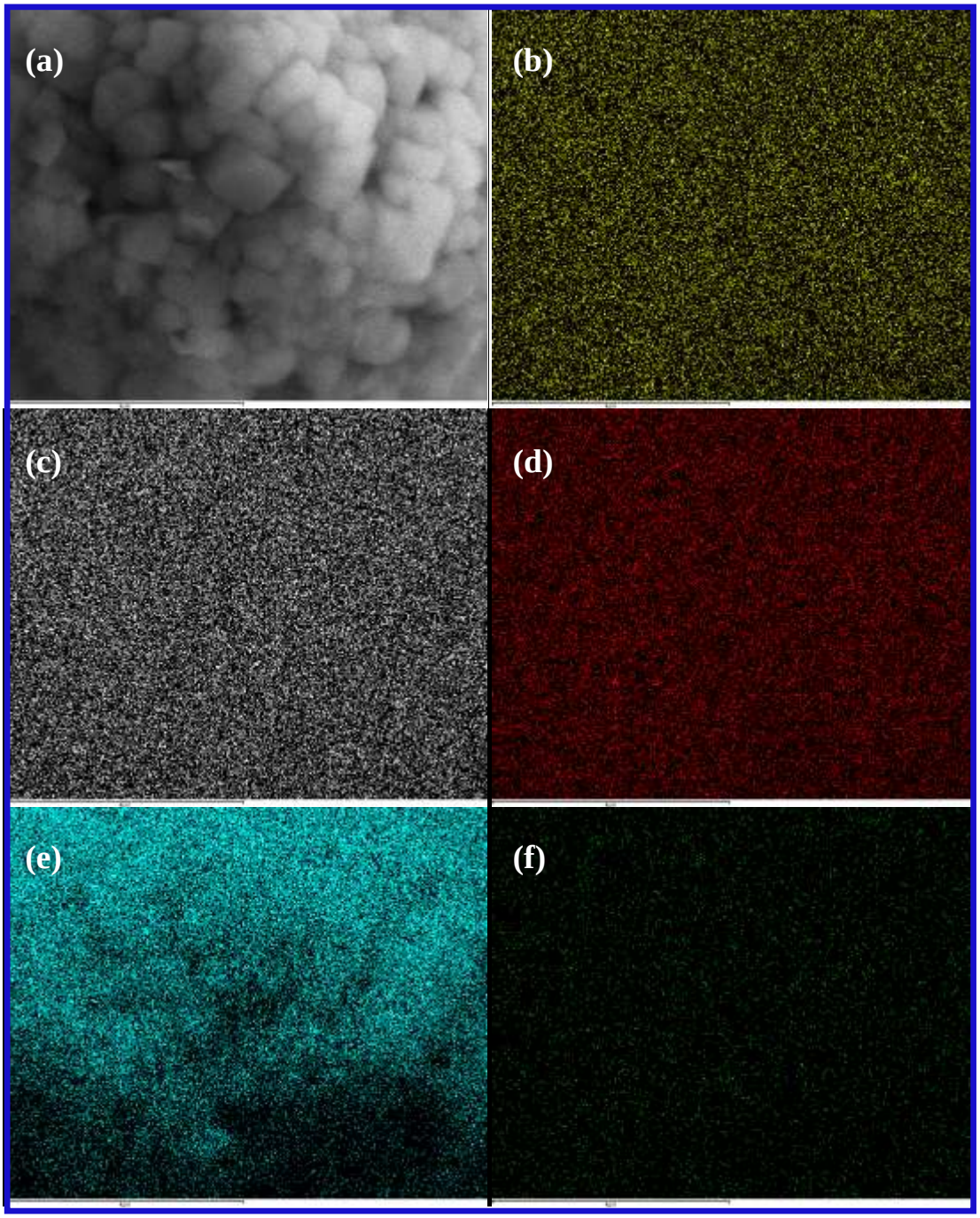

Figure S4 (a) SEM image and elemental mapping of NMC-10: (b) Ni, (c) Mn, (d) Co, (e) O, and (f) Ta.

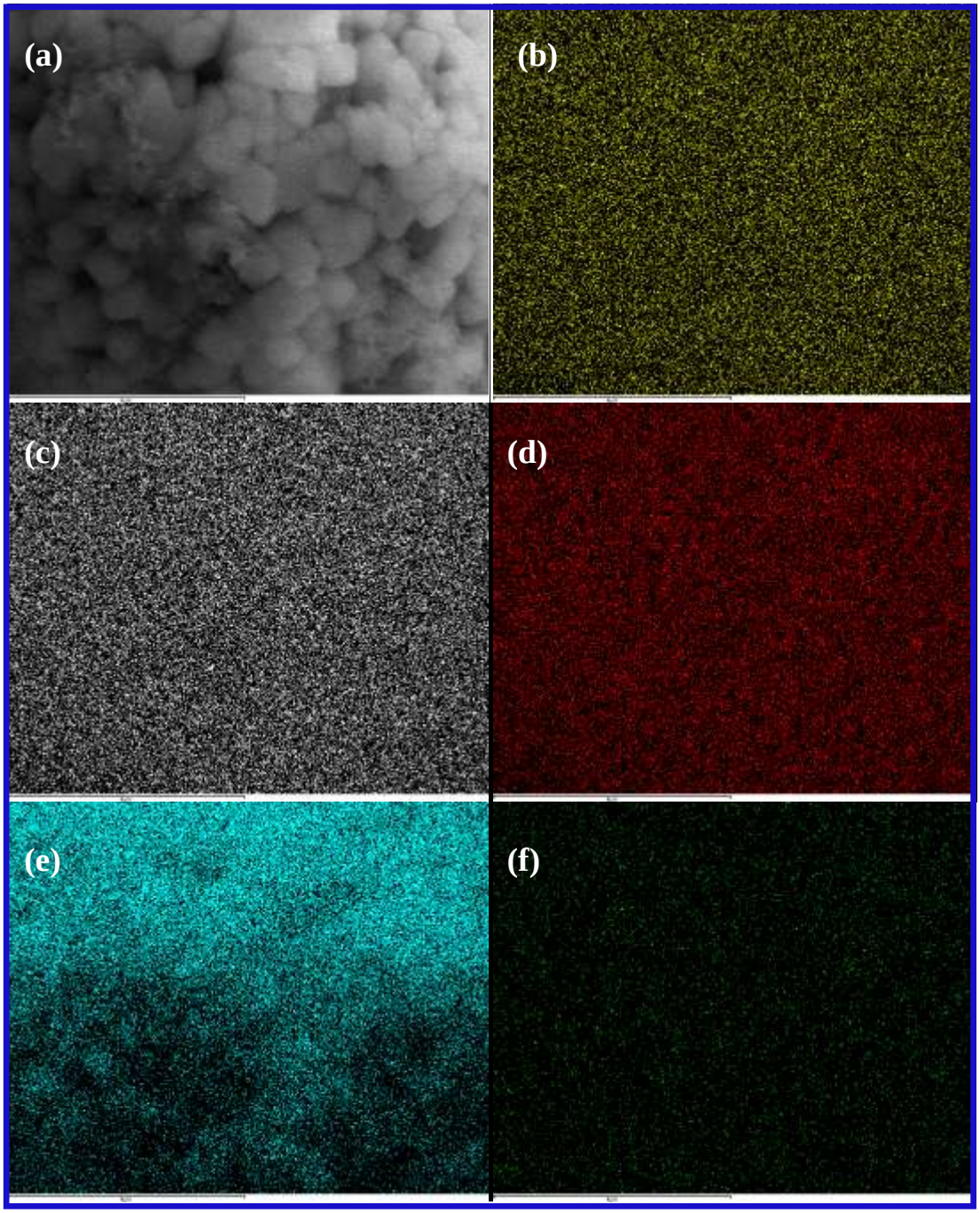

Figure S5 (a) SEM image and elemental mapping of NMC-20: (b) Ni, (c) Mn, (d) Co, (e) O, and (f) Ta.

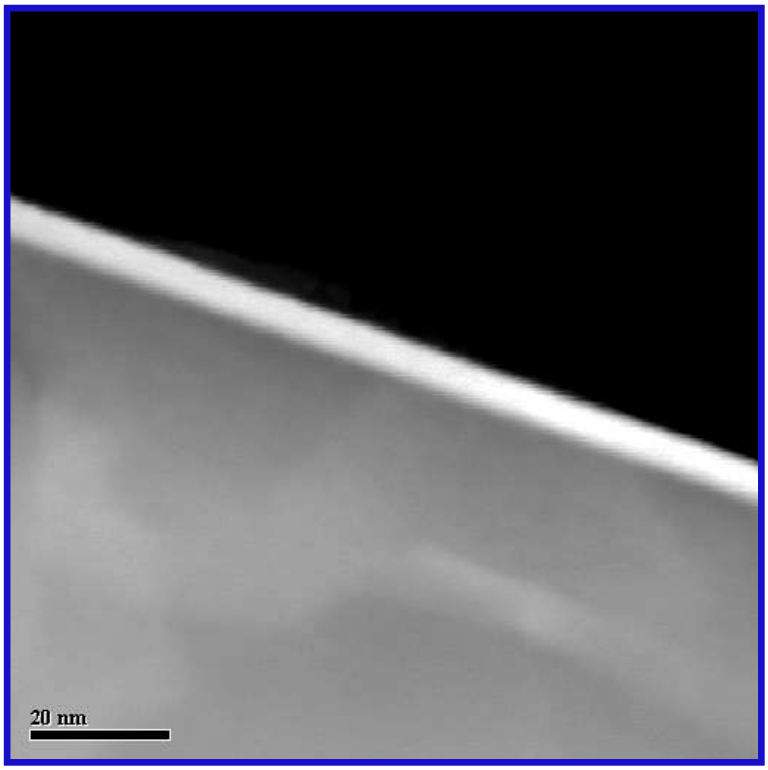

Figure S6 Dark field image of HRTEM of NMC-20 at low magnification.

Table S2. The potentials of the redox peaks in the 1st, 2nd, and 6th scans for the NMC electrodes coated by $LiTaO_3$ with different ALD cycles.

ALD Cycles	1st Scan			2nd Scan			6rd Scan		
	O	R	△	O	R	△	O	R	△
0	3.890	3.696	0.194	3.857	3.694	0.163	3.840	3.692	0.148
2	3.894	3.697	0.197	3.874	3.695	0.179	3.842	3.690	0.152
5	3.938	3.700	0.238	3.894	3.698	0.196	3.870	3.690	0.180
10	3.969	3.707	0.262	3.898	3.700	0.198	3.888	3.685	0.203

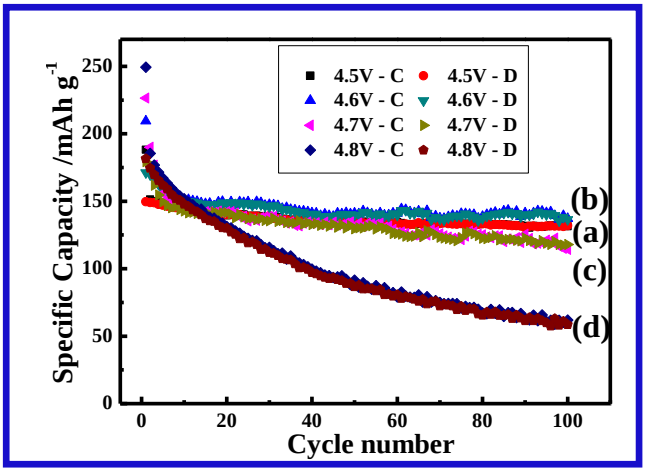

Figure S7 Cyclic performance of the NMC cathode at different potential ranges: (a) 3.0~ 4.5 V, (b) 3.0~ 4.6 V, (c) 3.0~ 4.7 V, and (d) 3.0~ 4.8 V.

Table S3 The discharge capacities and capacity retention of NMC cathode at different potential ranges employed in Figure S6.

Cut-off Potentails (V)	**3.0~ 4.5**	**3.0~ 4.6**	**3.0~ 4.7**	**3.0~ 4.8**
The first discharge capacity (mAh g^{-1})	150	171	179	182
The 100th discharge capacity (mAh g^{-1})	132	136	118	59
Capacity Fade (%)	12	20	34	68

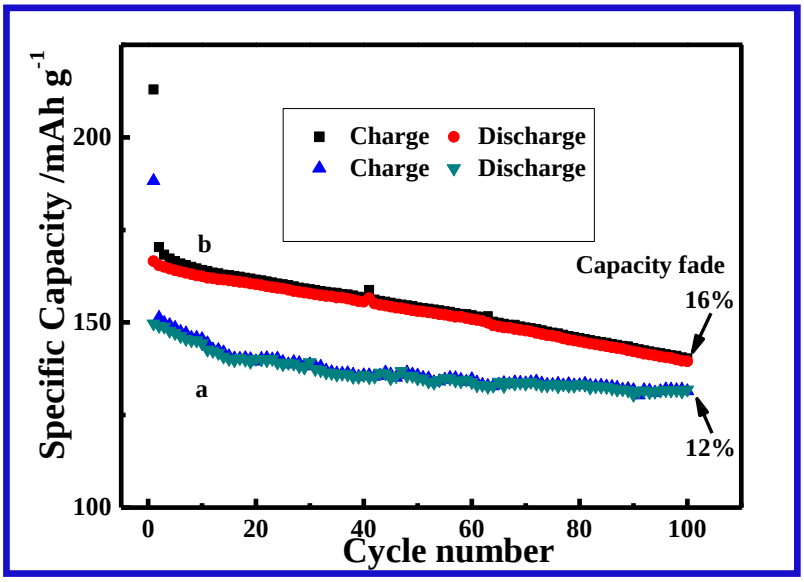

Figure S8 The comparison of cyclic performance of the NMC cathodes at (a) room temperature and (b) 55 ^{o}C in a potential range of 3.0~ 4.5 V at a current density of 160 mA g^{-1}.

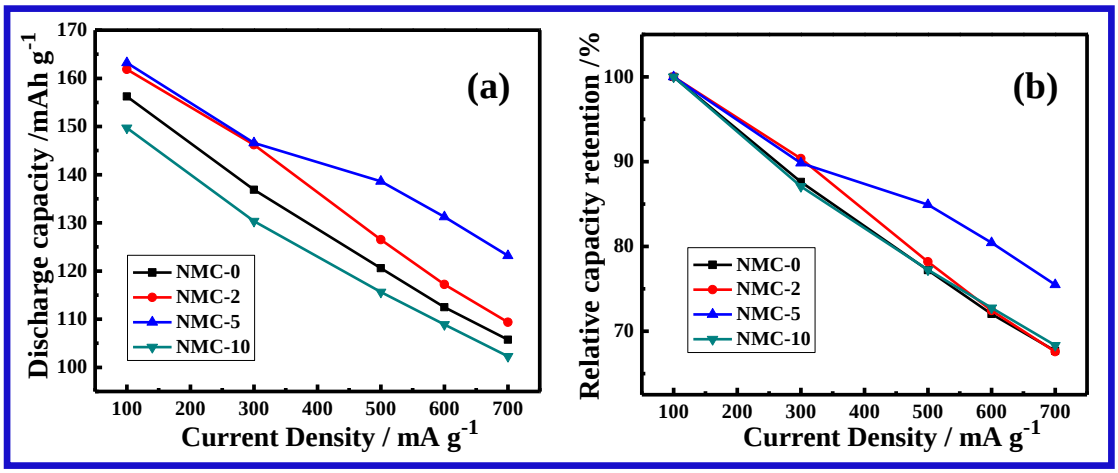

Figure S9 (a) The discharge capacities of NMC-0, NMC-2, NMC-5, and NMC-10 as a function of current densities from 100 mA g^{-1} to 700 mA g^{-1}; (b) The comparison of discharge capacities of the second cycle at various current densities with the capacity of the second cycle at 100 mA g-1 for NMC-0, NMC-2, NMC-5, and NMC-10.

Table S4. Comparison of Lithium diffusion coefficient (cm^2 s^{-1}) of the NMC cathodes with 2-ALD-cycle derived coatings of solid-state electrolyte ($LiTaO_3$) and metal oxide (ZrO_2).

$LiTaO_3$		ZrO_2	
Charge	Discharge	Charge	Discharge
2.912×10^{-10}	7.543×10^{-11}	2.474×10^{-10}	5.009×10^{-11}

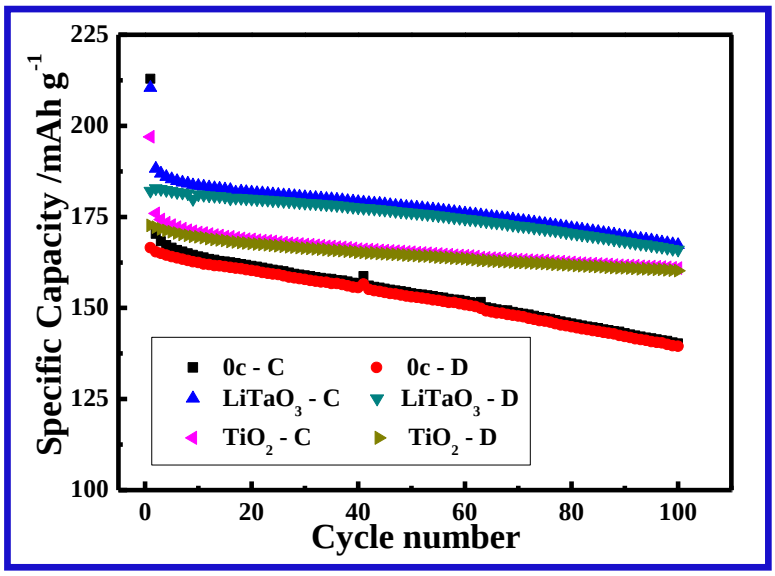

Figure S10 The comparison of cyclic performance of the NMC cathodes with 2-ALD-cycle derived coatings of solid-state electrolyte ($LiTaO_3$) and metal oxide (TiO_2) at 55 ^oC in a potential range of 3.0~ 4.5 V at a current density of 160 mA g^{-1}.