

Supporting Information

Atomic Layer Deposition of Lithium Zirconium Oxides for Improved Performance of Lithium-ion Batteries

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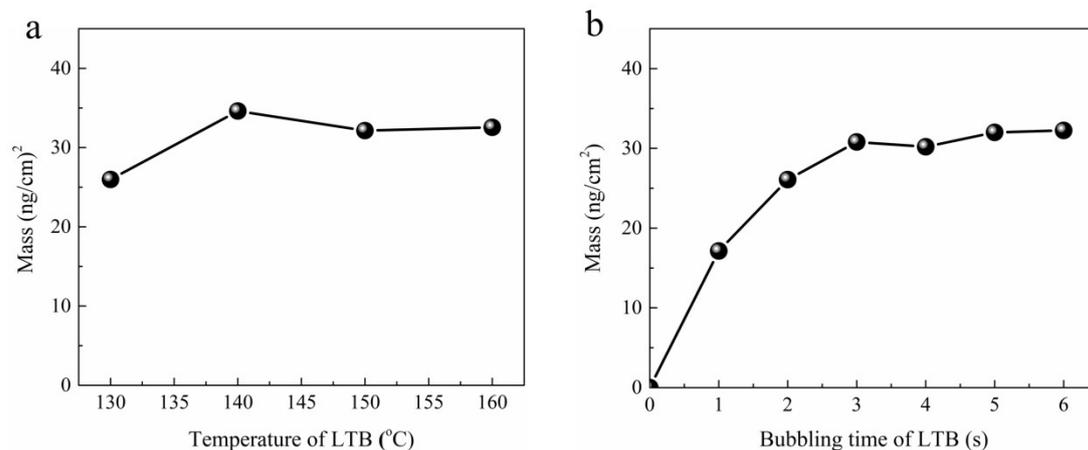


Figure S1. The effects of (a) heating temperatures of LTB precursor and (b) dosing times of LTB on the mass gain per cycle of the ALD-LiOH at 225 °C. The timing sequence is LTB (t_l) – purge (10 s) – H₂O (0.015 s) – purge (10 s) for (a) and (b), where $t_l = 5$ s for (a).

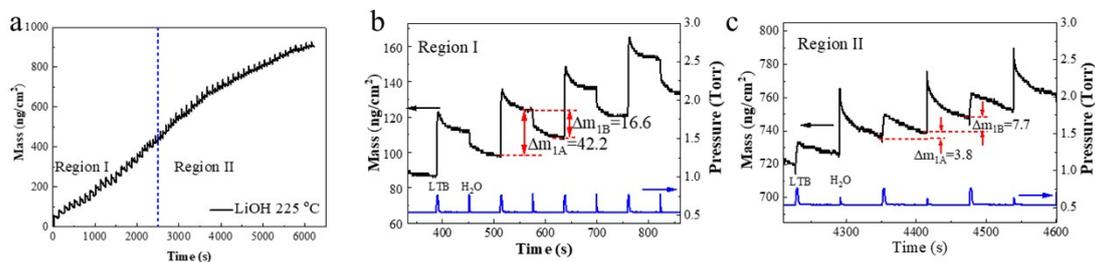


Figure S2. *In-situ* QCM measurements of ALD-LiOH using the timing sequence of 3 - 60 - 0.015 - 60 s in correspondence to LTB-purge-H₂O-purge, respectively, at 225 °C: (a) 50 consecutive cycles, (b) an enlarged view of 3 consecutive cycles in Region I of (a), and (c) an enlarged view of 3 consecutive cycles in Region II of (a).

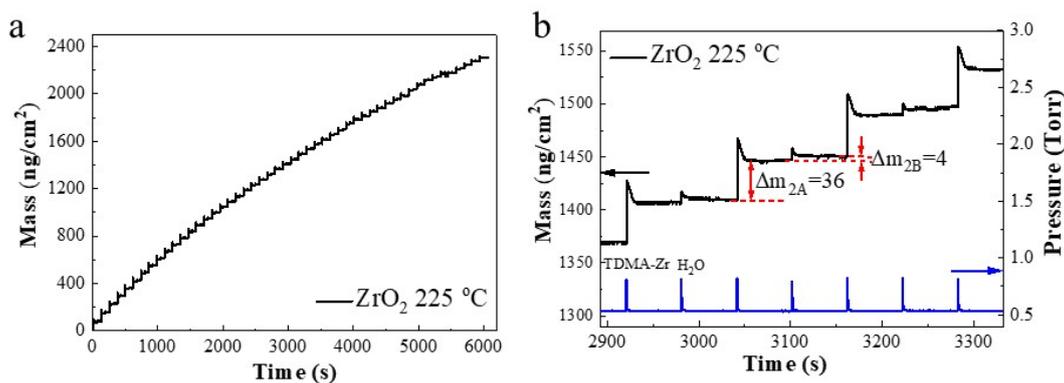


Figure S3. *In-situ* QCM measurements of ALD-ZrO₂ using the timing sequence of 0.03 - 60 - 0.015 - 60 s in correspondence to TDMA-Zr-purge-H₂O-purge, respectively, at 225 °C: (a) 50 consecutive 50 cycles and (b) an enlarged view of 3 consecutive cycles.

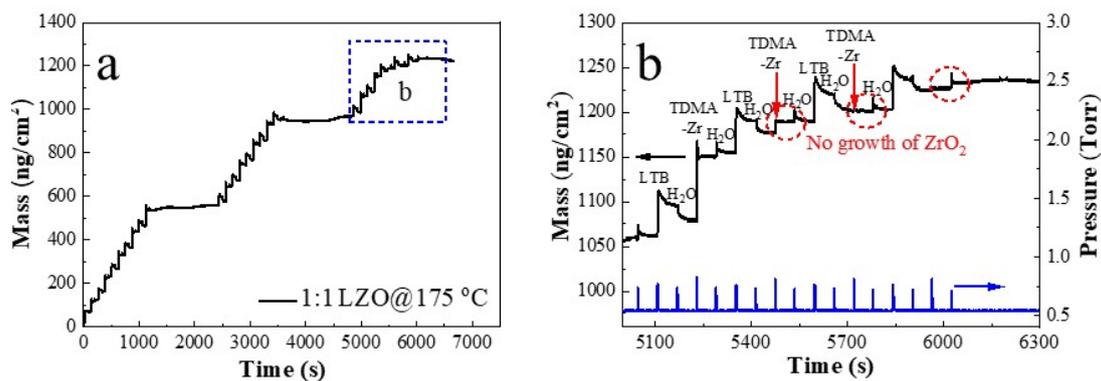


Figure S4. *In-situ* QCM measurements of 1:1 ALD-LZO using the timing sequence of LTB (3 s) - 60 s - H₂O (0.015 s) - 60 s - TDMA-Zr (0.03 s) - 60 s - H₂O (0.015 s) - 60 s at 175 °C. (a) Consecutive 30 cycles, (b) enlarged view of 3 cycles. The difference from the previous experiment is that after every 10 ALD cycles, a 20 min purging process is added.

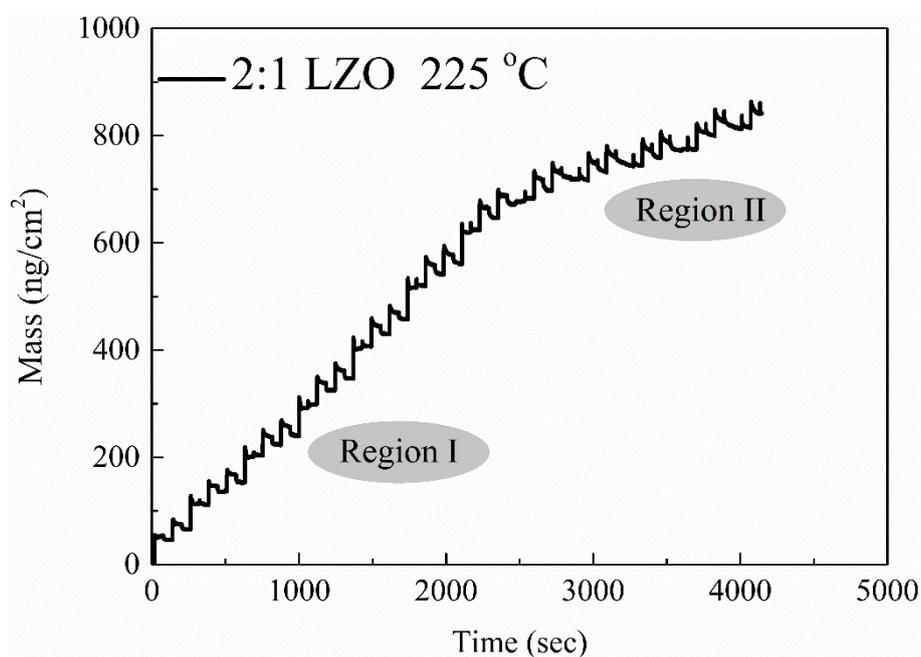


Figure S5. *In-situ* QCM measurements of 2:1 ALD-LZO for 10 ALD cycles using the timing sequence of LTB (3 s) - 60 s - H₂O (0.03 s) - 60 s - LTB (3 s) - 60 s - H₂O (0.03 s) - 60 s - TDMA-Zr (0.03 s) - 60 s - H₂O (0.015 s) - 60 s at 225 °C.

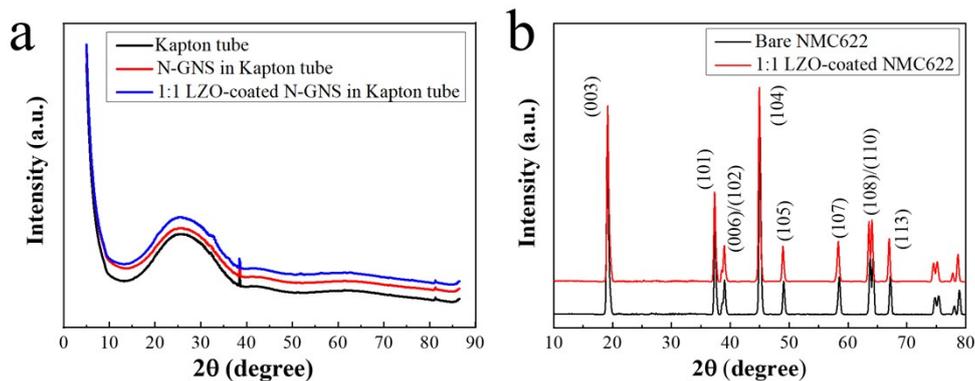


Figure S6. (a) XRD spectra of Kapton tube, N-GNS powders in Kapton tube, and 200-cycle 1:1 LZO-coated N-GNS in Kapton tube. (b) XRD spectra of NMC622 powders and 60-cycle 1:1 LZO-coated NMC622 powders.

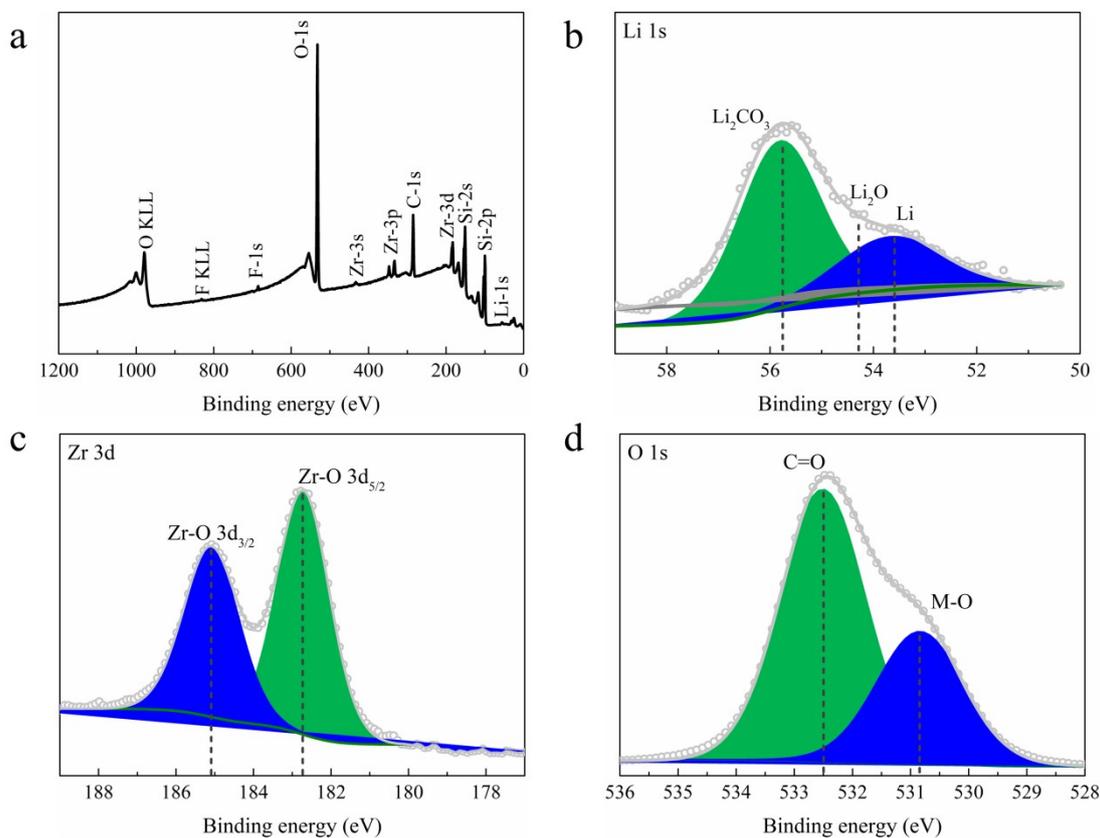


Figure S7. (a) X-ray photoelectron spectroscopy (XPS) full spectrum scanning of 1:2 ALD-LZO. (b), (c) and (d) are high-precision XPS spectra of Li1s, Zr3d and O1s respectively.

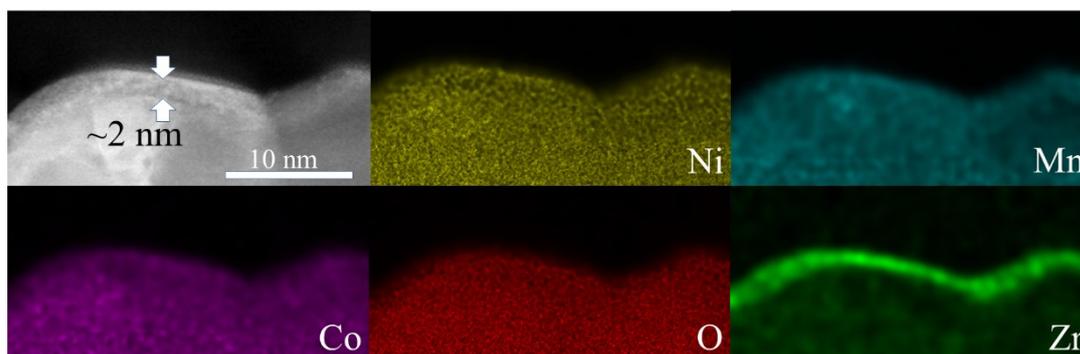


Figure S8. STEM-EDS image and elemental mapping of LZO-coated NMC622.

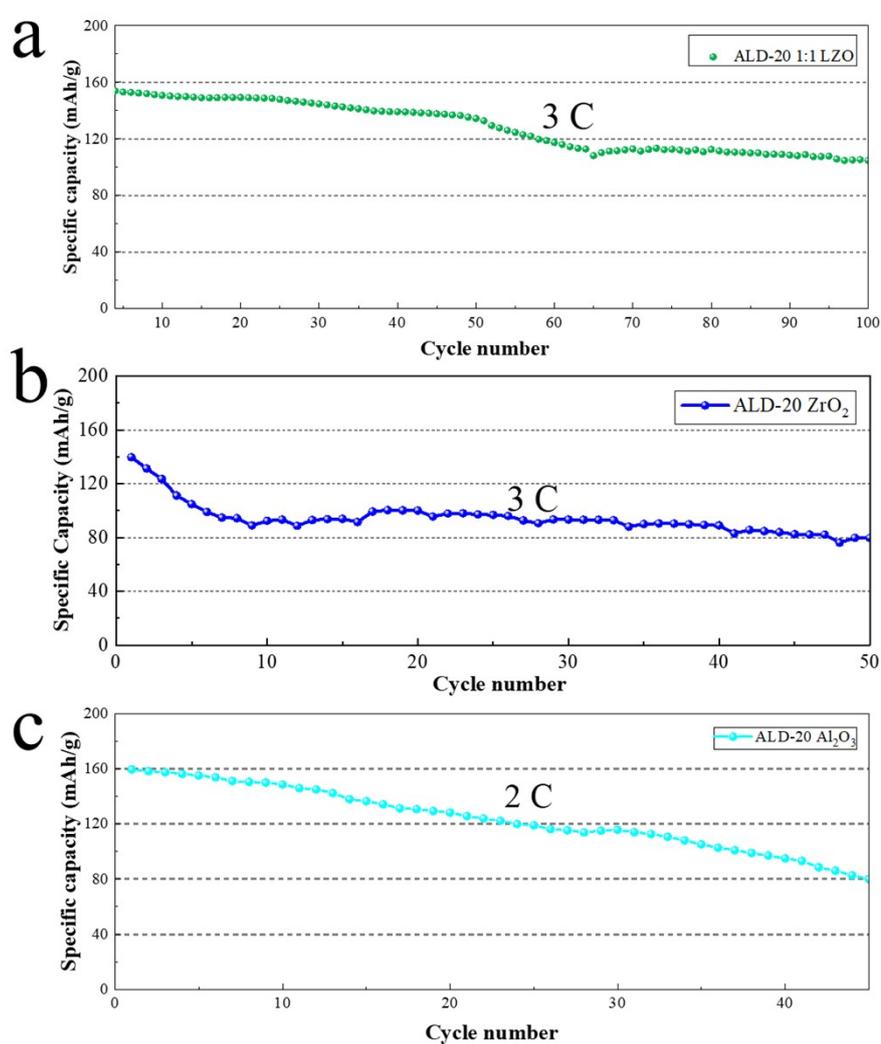


Figure S9. Comparison of the effects of 20-cycle ALD (a) 1:1 LZO, (b) ZrO₂, and (c) Al₂O₃ coatings on NMC622 electrodes in the voltage window of 3.0 – 4.5 V in the 3:7 EC:EMC electrolyte. The first two were tested under 3 C while the last was tested under 2 C.

Table S1. Element composition and quantitative analysis of 1:2 ALD-LZO

<i>Name</i>	<i>Peak BE</i>	<i>FWHM eV</i>	<i>Area (P) CPS.eV</i>	<i>Atomic %</i>
O1s	532.11	3.38	1736798.41	36.79
Si2p	100.26	3.22	600596.23	30.59
C1s	285.06	2.98	393932.52	20.17
Zr3d	183.57	3.09	273934.79	1.46
F1s	685.39	3.09	39810.34	0.67
Li1s	55.69	3.73	14055.84	10.31

Table S2. The charge-discharge data of bare and 1:1 ALD-LZO coated NMC622 cathodes at first cycle.

<i>Sample</i>	<i>Charge capacity mAh/g</i>	<i>Discharge capacity mAh/g</i>	<i>Irreversible capacity%</i>
Bare	224.7	184.4	17.9%
ALD-10	219.9	194.9	11.3%
ALD-20	223.9	189.0	15.6%
ALD-40	219.3	193.9	11.6%
ALD-60	219.5	188.6	14.1%