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Supplementary Information

Controllable Crystalline Preferred Orientation in Li–Co–Ni–Mn Oxide Cathode Thin Film for All-Solid-State Lithium Batteries

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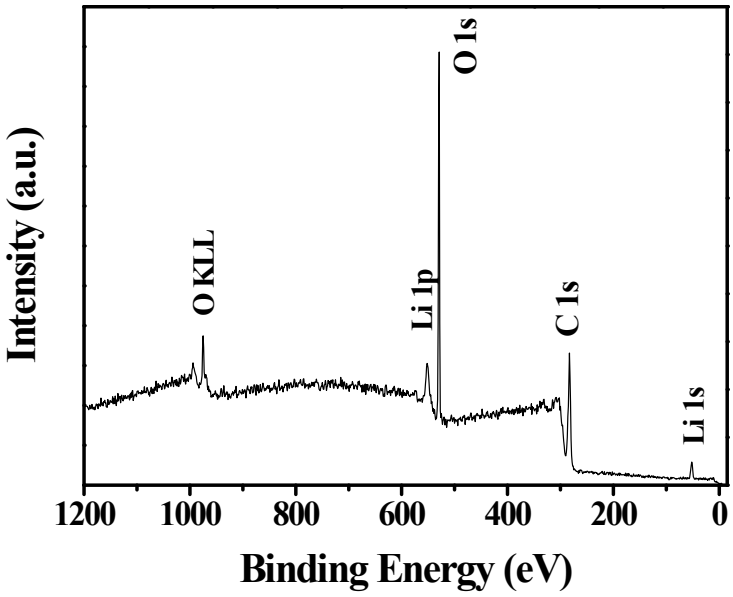

Figure 1S XPS survey spectrum of the as-deposited Li–Co–Ni–Mn oxide thin film.

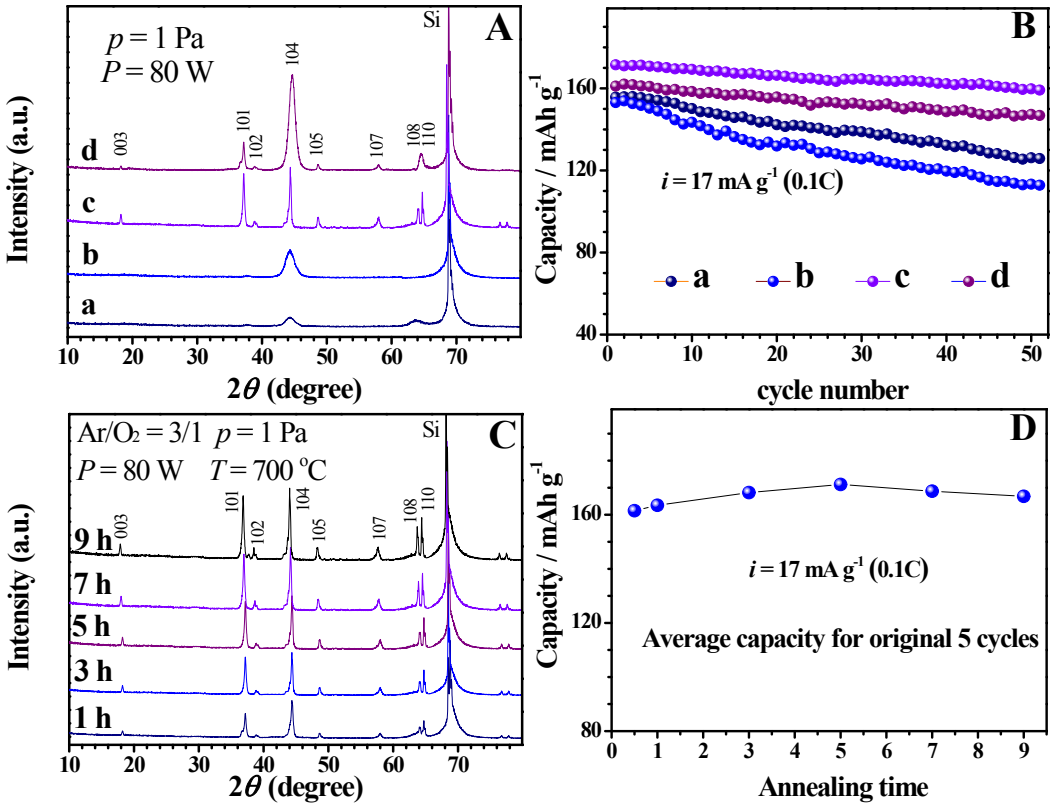

Figure 2S (A) XRD patterns and (B) Cycle performance of thin films, which are deposited in Ar/O_2 (3:1) at (a) room temperature, (c) annealed at 700 °C, and deposited in pure Ar at (b) room temperature and (d) annealed at 700 °C; (C) XRD patterns and (D) discharge capacity versus annealing time of 700 °C annealed films. Cycling curves (B, D) are recorded using coin-type half-cell between 2.8 and 4.5 V at a current density of 17 mA g^{-1}.

Figure 2 shows crystalline structural properties and electrochemical performances of thin films prepared under different conditions. In Figure 2A, the film (b) deposited in pure Ar only shows a broad peak at 44° refer to the (104) reflection. The 700 °C annealed film (d) also shows a strong broad (104) peak, but with a depressed (108/110) peak. This indicates that the pure Ar deposited condition is beneficial to the (104) plane growth, but not to the (110) plane. Thus, in Figure 2B, the specific capacity and cycle performance of thin films (b,d) prepared in pure Ar are inferior to those of films (a,c) prepared in Ar/O_2 (3:1). In Figure 2C, the crystallinty of 700 °C annealed films is gradually improved upon the increased annealing time, it is important to note that the diffraction peaks shift slightly to the lower 2θ angle while the annealing time is elongated to 7 h or 9 h, indicating a lithium deficient layered structure. This is caused by the loss of lithium ions

during annealing for long times. In Figure 2D, the annealed film for 5 h shows the highest average capacity for original 5 cycles. On the basis of these results, the optimal crystalline growth condition is determined to be the thin film deposited in Ar/O_2 (3:1 in volume) and in situ annealed at 700 °C for 5 h.