

SUPPLEMENTARY INFORMATION

**In situ studies of lithium-ions diffusion in Lithium-rich thin film cathode by
Scanning Probe Microscopy Techniques**

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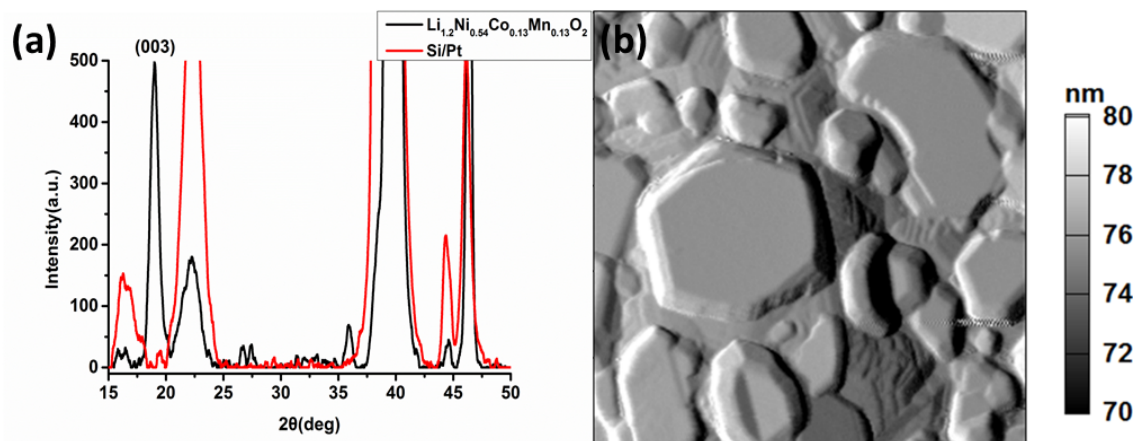


Figure S1. (a) XRD pattern of $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$ thin film and the Si/Pt substrate. Comparing with the XRD pattern of the $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$ powder,¹ the cathode film shows the relative higher intensity of the (003) plane reflection whereas the reflection of (101) and (104) cannot be found. Hence, this XRD pattern confirms that the majority of the grains in the $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$ thin film are with (003) orientation. The X-ray diffraction (XRD) experiments were conducted with commercial equipment (Model XRD-7000, Shimadzu Corporation, Kyoto, Japan) with Cu $K\alpha$ radiation ($\lambda=1.5418\text{\AA}$). (b) AFM deflection image showing grains have well-defined hexagon-like geometry.

1. Yan, B., Liu, J., Song, B., Xiao, P., Lu, L., *Scientific Reports*, 2013, 3, 3332.

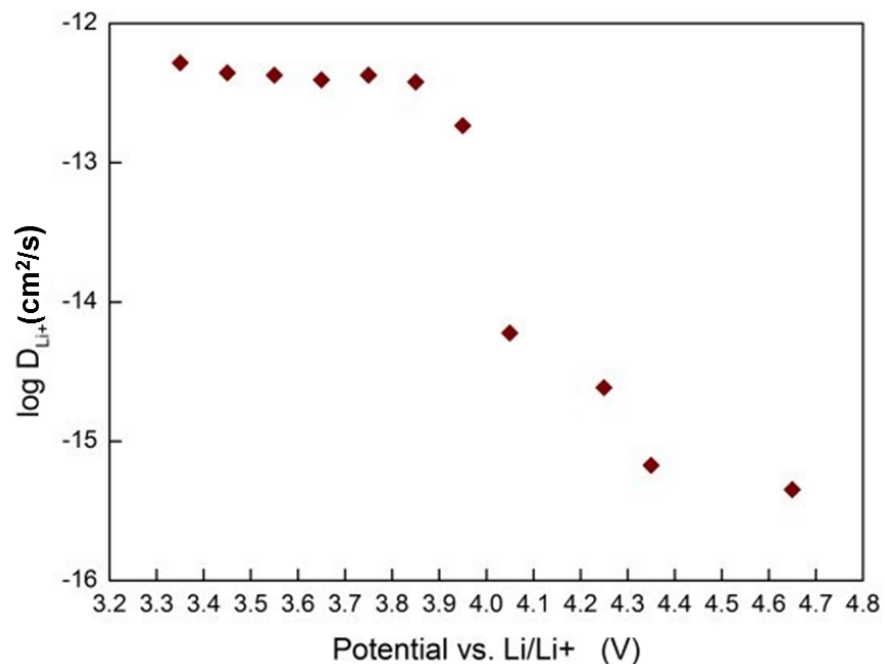


Figure S2. Chemical diffusion coefficient (log scale) of Li vs. potential during charge state in Li-rich cathode film. The diffusion coefficients are measured using the PITT (potentiostatic intermittent titration technique, 1400 CellTest System, Solartron Analytical, UK). During the measurement, a potential with step increment of 10mV was applied while a current change with time was measured until an equilibration was reached. Then a subsequent step was applied. The residual currents were less than 1% of the maximum current at the beginning of the potential step.

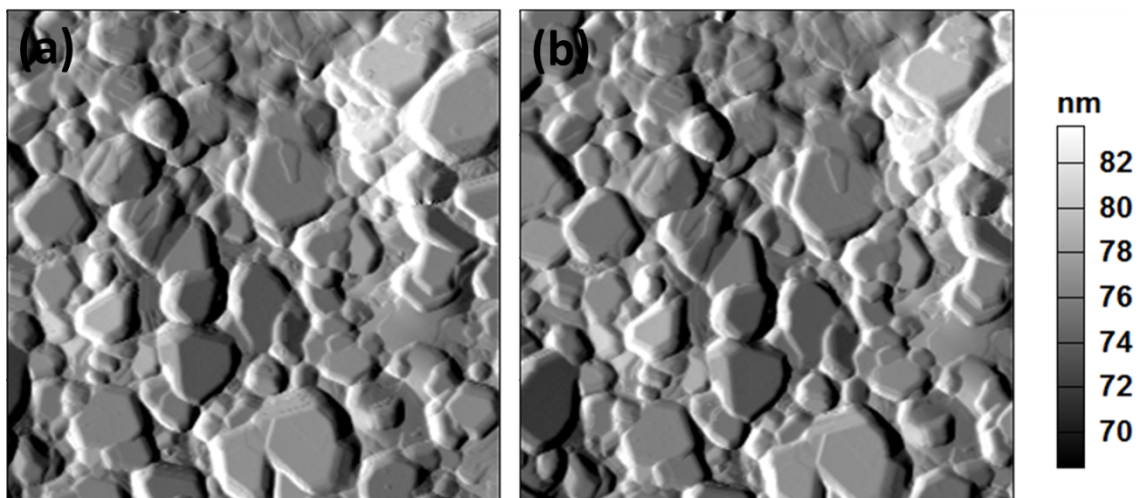


Figure S3. The AFM topographic images of a same region by (a) a tip after BE-ESM images; and (b) a new tip (both are AC240TM, Olympus, Japan). These images show the same features in the scanned area by the two tips, hence, they prove the observed changes in the thin film cathode are caused by electrical field and not due to the possible changes of the tip geometry.