

## Supporting Information

### Grain boundary effects on Li-ion diffusion in $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$ thin film cathode studied by scanning probe microscopy techniques

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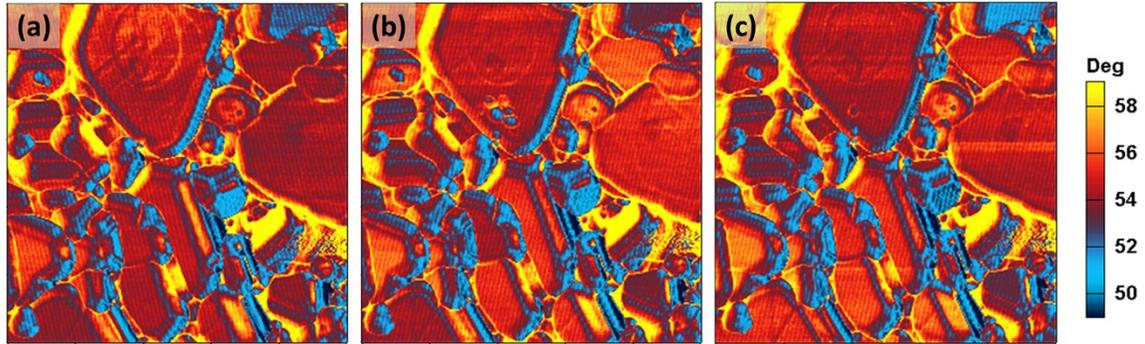


Figure S1. Biased-AFM phase images of the  $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$  cathode thin film. The bias is applied in a single point at the grain interior, and the scanning size is  $1 \times 1 \mu\text{m}^2$ . The phase images of the same area: (a) before bias, (b) under +7 V bias and (c) 30 minutes after bias of +7 V. The corresponding phase images show the protuberances are clearly different from the surrounding materials.

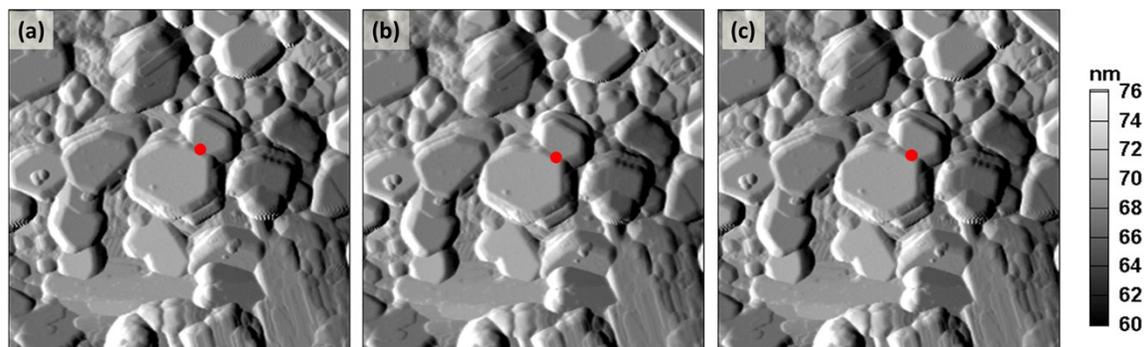


Figure S2. Biased-AFM of  $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$  cathode thin film in synthetic air (with 21% oxygen and 79% nitrogen, water content < 5ppm). The scanned area is  $1.5 \times 1.5 \mu\text{m}$ . (a) Before the bias; (b) after +7 V bias; and (c) after -7 V bias. The red dot is where the bias applied. There are basically no observable surface deformation in the images.

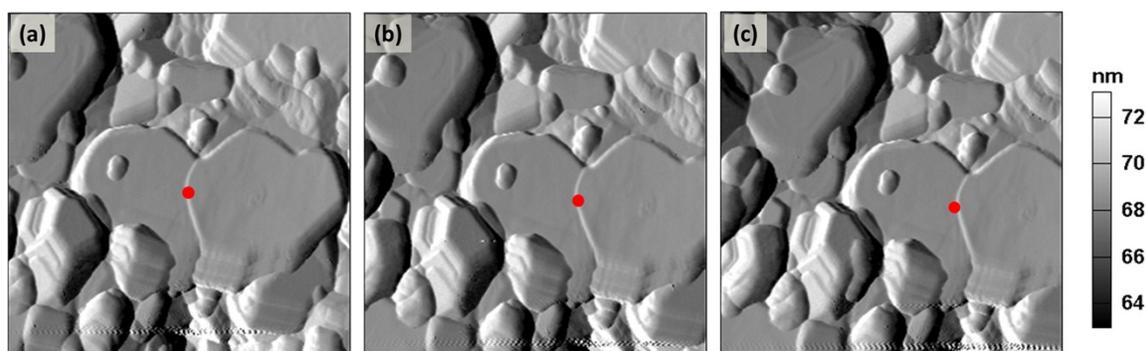


Figure S3. Biased-AFM of  $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$  cathode thin film in Argon gas (purity of 99.9997%, water content < 5ppm, and oxygen content <5 ppm). The scanned area is  $1.0 \times 1.0 \mu\text{m}$ . (a) Before the bias; (b) after +7 V bias; and (c) after -7 V bias. The red dot is where the bias applied. There are basically no observable surface deformation in the images.

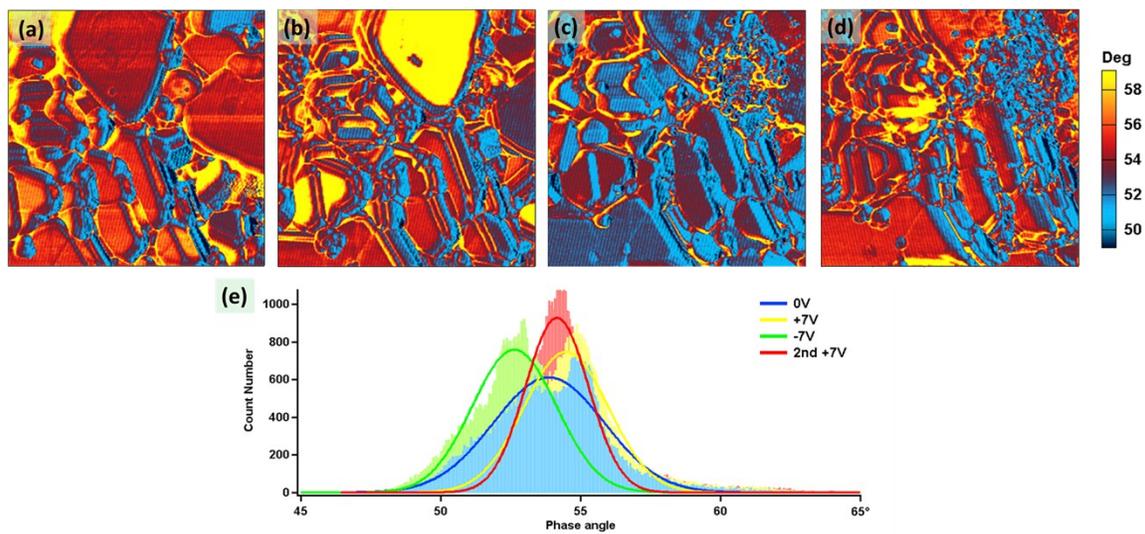


Figure S4. Biased-AFM phase images of the  $\text{Li}_{1.2}\text{Co}_{0.13}\text{Ni}_{0.13}\text{Mn}_{0.54}\text{O}_2$  cathode thin film. The bias is applied in a single point at the grain boundary, and the scanning size is  $1 \times 1 \mu\text{m}^2$ . The phase images of the same area: (a) before bias, (b) under + 7 V bias, (c) under the bias of -7 V, and (d) under the bias of +7 V again; (e) is the distribution histogram of phase angles. Some obvious extrusions are formed in image (b) and (d). The phases of the grains show clear changes with the applied bias. The “intercalated” grains show positive shift of the phase angle, whereas the “de-intercalated” grains show negative shift of the phase angle, both by a few degrees. This suggests the possible relationship between the phase angle and Li-ions movements in the grains.