

## ***In-situ* investigation of Li permeation through grain boundaries in garnet-based solid electrolyte**

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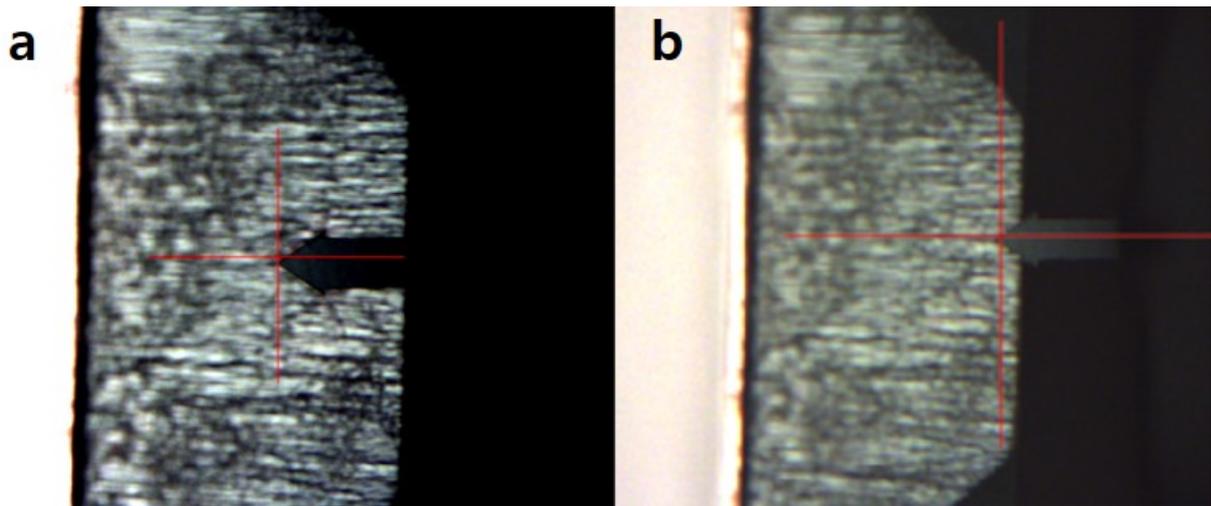


Figure S1. Optical microscope images of the AFM tip. a, The tip is located in the middle of the LLZTO. b, The tip is located near the In electrode.

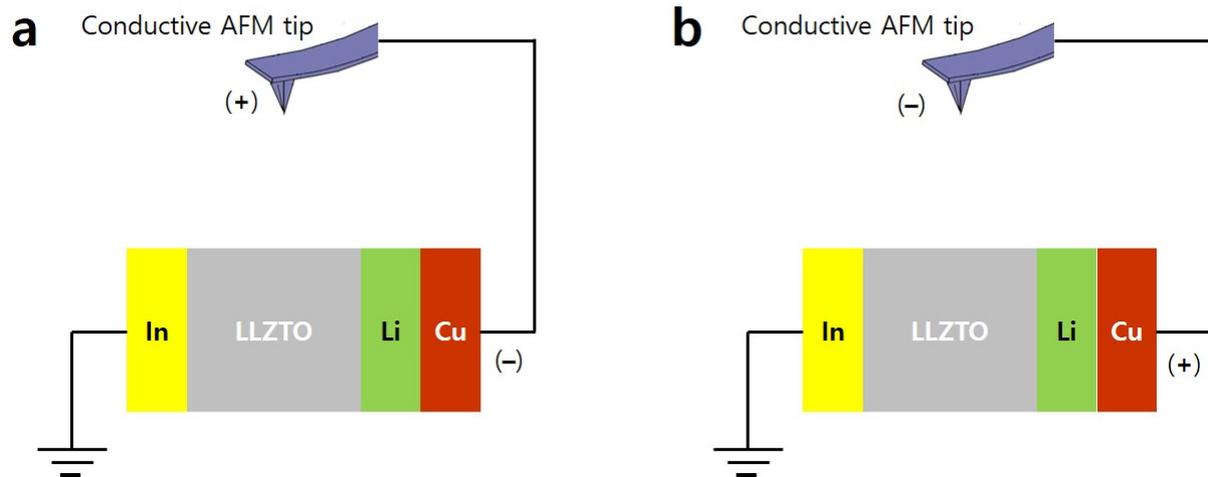


Figure S2. The C-AFM setup. a, The sample is negatively biased relative to a conductive AFM tip scanning the cross-section. The current flows from the sample to the tip. b, The sample is positively biased relative to a conductive AFM tip scanning the cross-section. The Li ions ( $\text{Li}^+$ ) flow from the sample toward the tip.

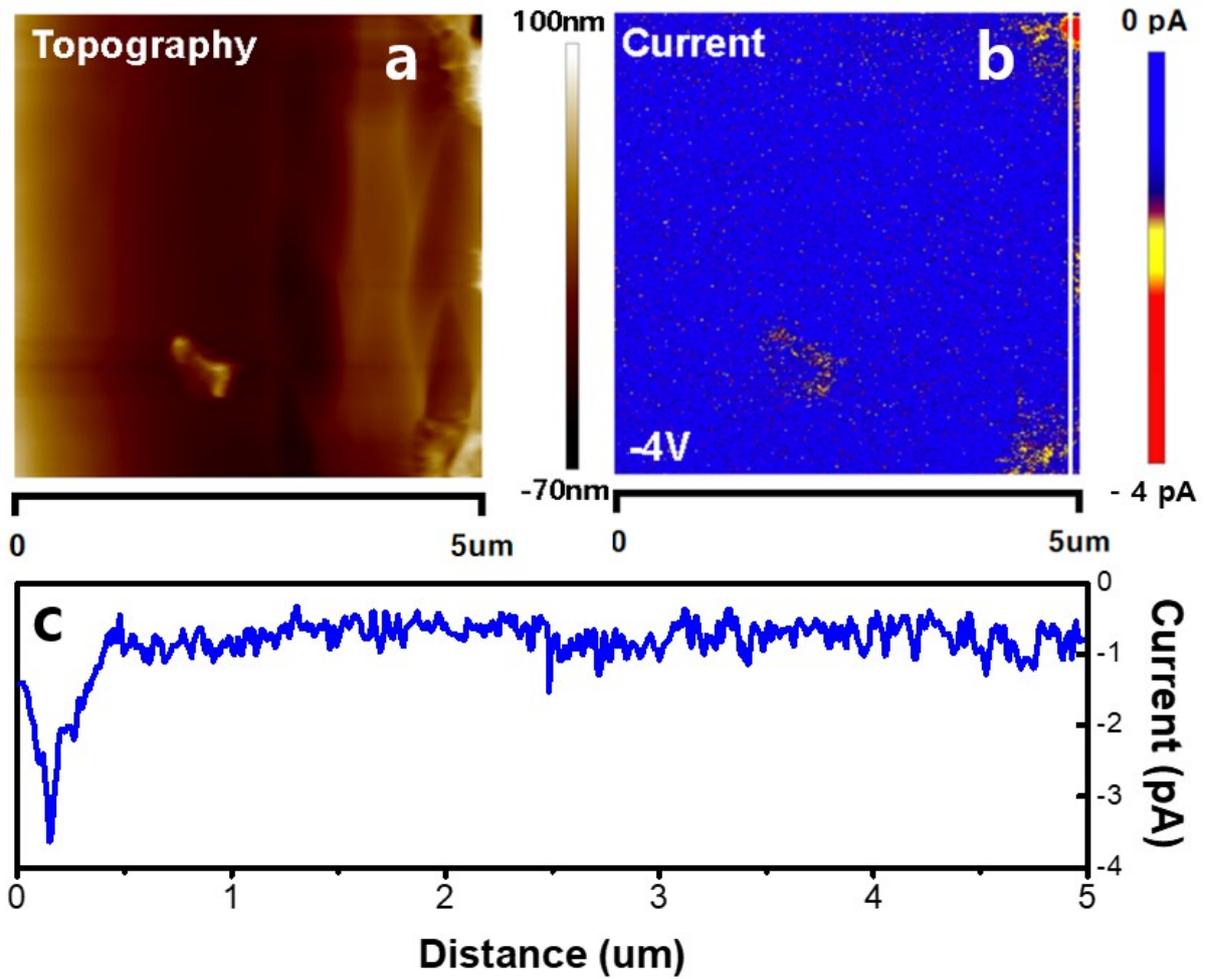


Figure S3. Conductive AFM images of the cell under negative bias voltage. a, Topography image of the cell. The region of interest (ROI) is  $25 \mu\text{m}^2$ . b, Current map in conductive mode. c, The current profile along the white line in Fig. S3b.

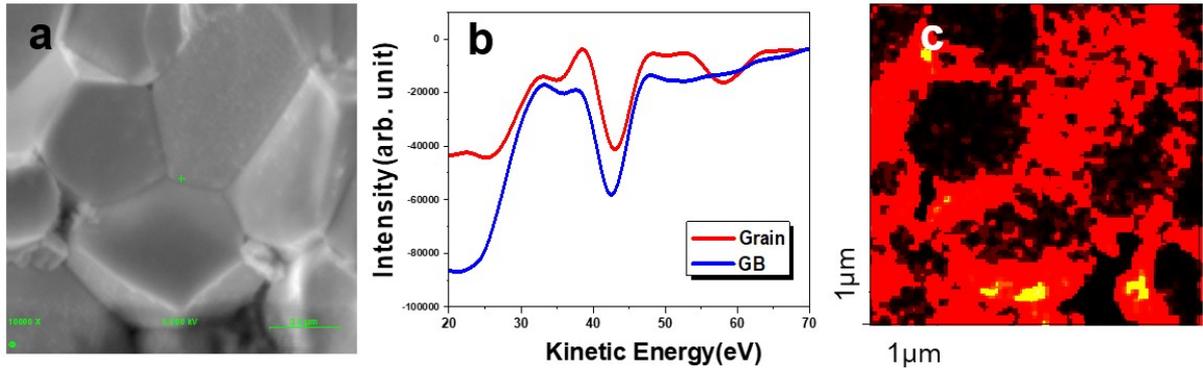


Figure S4. Grain and GB analysis by AES and C-AFM for cycled LLZTO samples. a, SEM image. b, Li Auger peak. c, Corresponding Li Auger map.

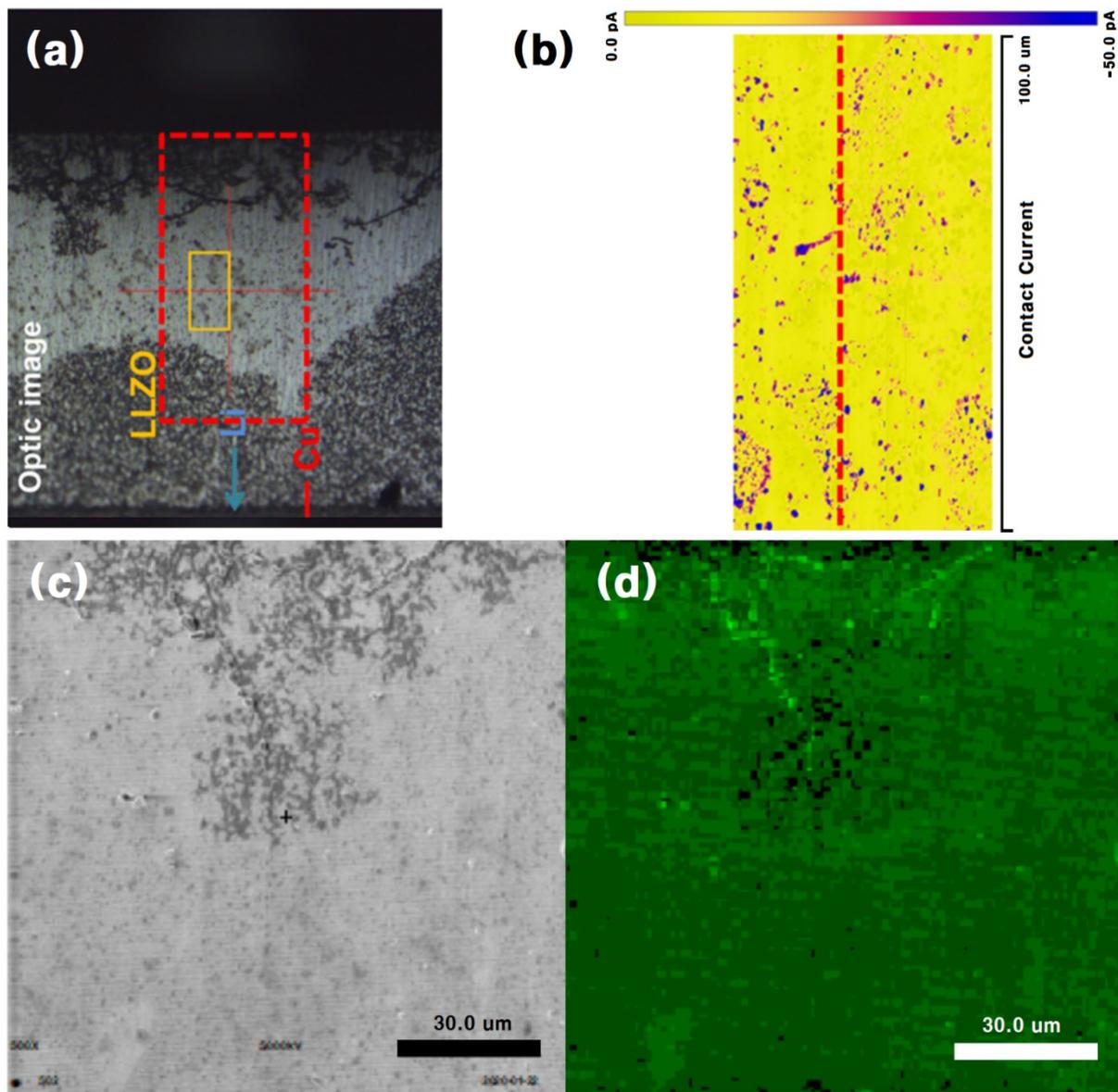


Figure S5. Conductive AFM images of the electrically shorted cell. a, Cross-sectional optical image of the cell. b, Current map in conductive mode. c, SEM image of the region in Fig. S4b. d, Li Auger map.