

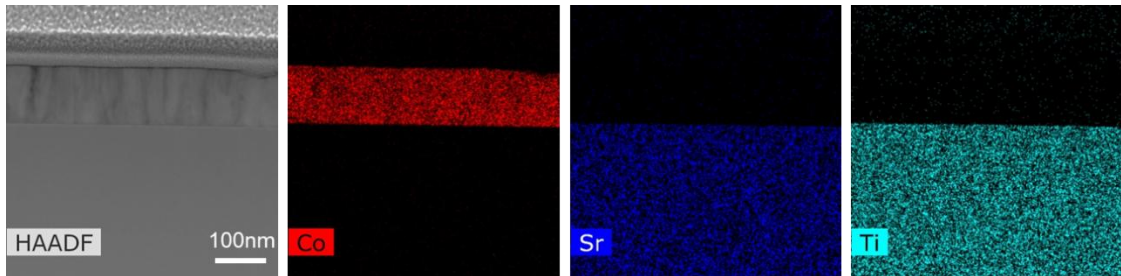
# Supporting Information for “LiCoO<sub>2</sub> Epitaxial Film Enabling Precise analysis of Interfacial Degradations”

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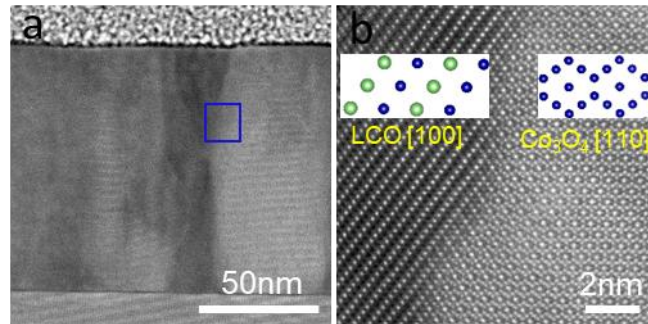
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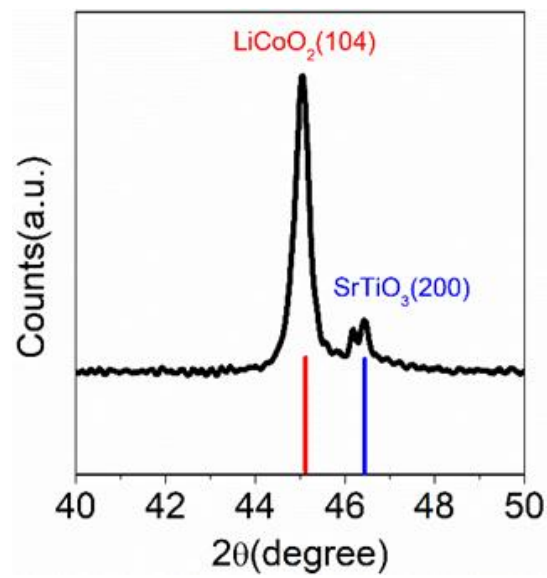
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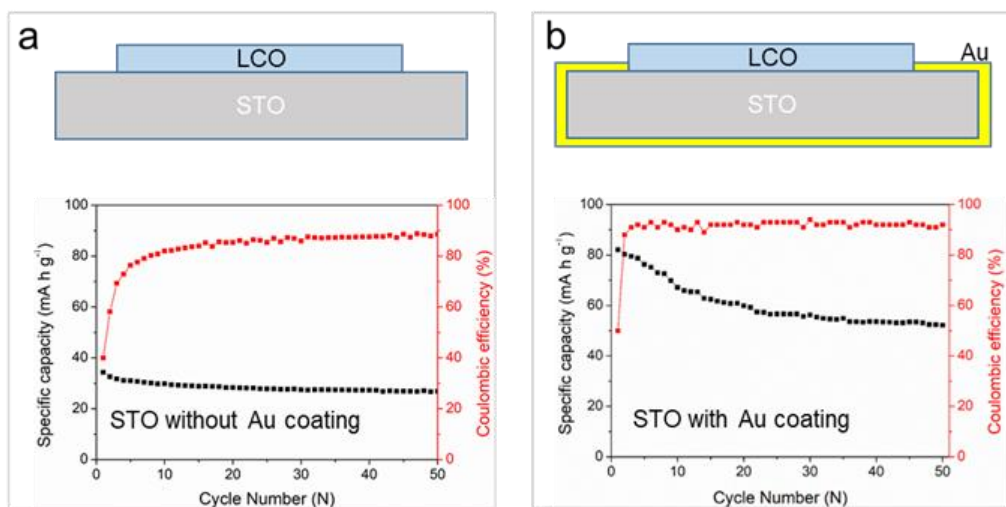
**Fig. S1.** STEM-HAADF image of the pristine LCO film and corresponding  
STEM-EDS mapping to show the uniform distribution of Co, Sr and Ti in pristine  
LCO film and STO substrate.



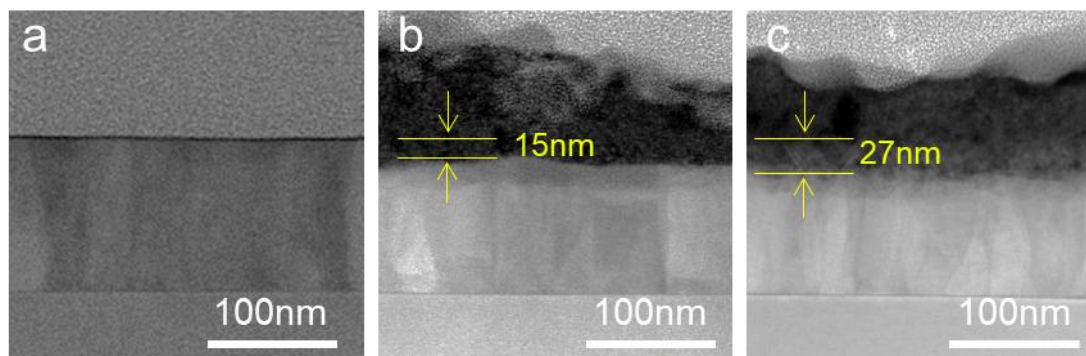
**Fig. S2.** STEM-HAADF images of LCO and Co<sub>3</sub>O<sub>4</sub> impurity.



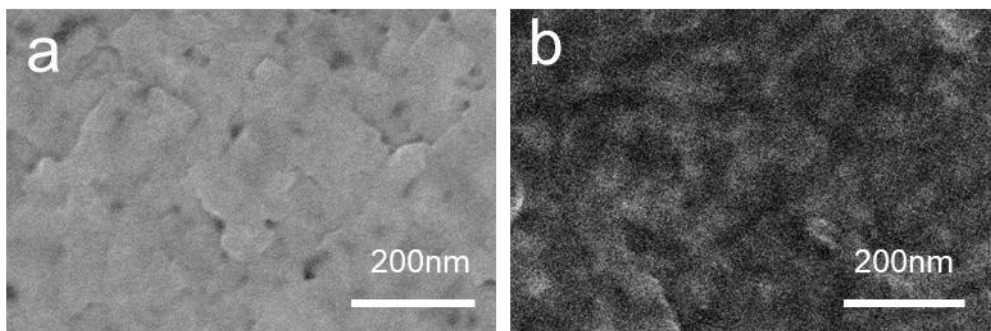
**Fig. S3.** XRD pattern of the pristine LCO epitaxial film.



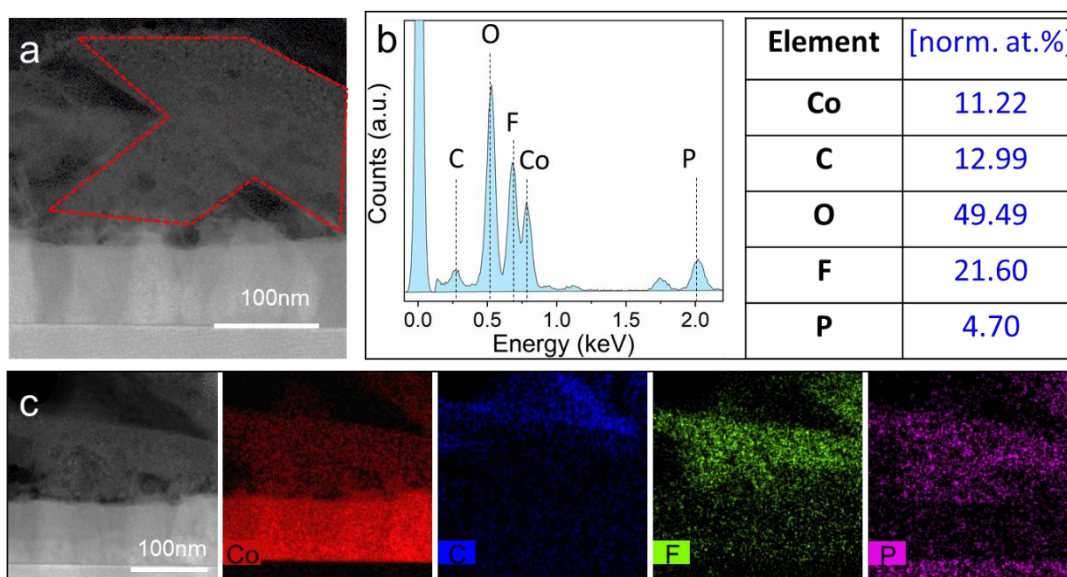
**Fig. S4.** Capacity retentions and corresponding coulombic efficiencies of the LCO film cycled at 0.2C rate (1C=140 mAh g<sup>-1</sup>) with 2.7 V-4.5 V voltage window, (a) without and (b) with Au coating on STO.



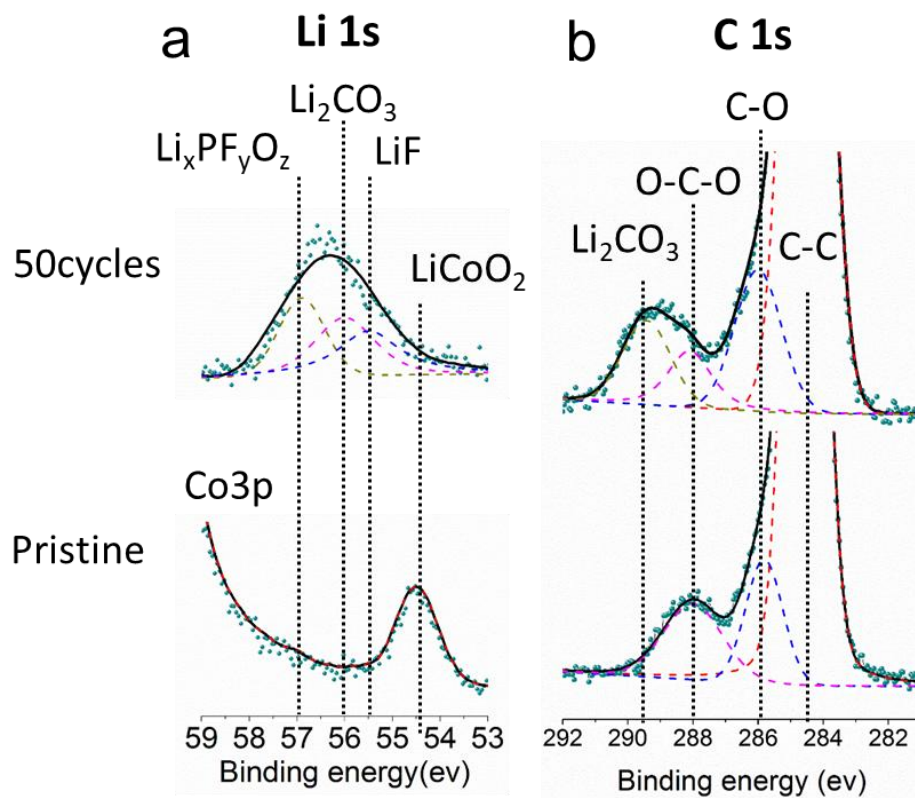
**Fig. S5.** STEM-HAADF images of the pristine LCO film (a) and the film after 50 cycles at 2.7 V-4.5 V (b, c), demonstrating the corrosion layer thickness.



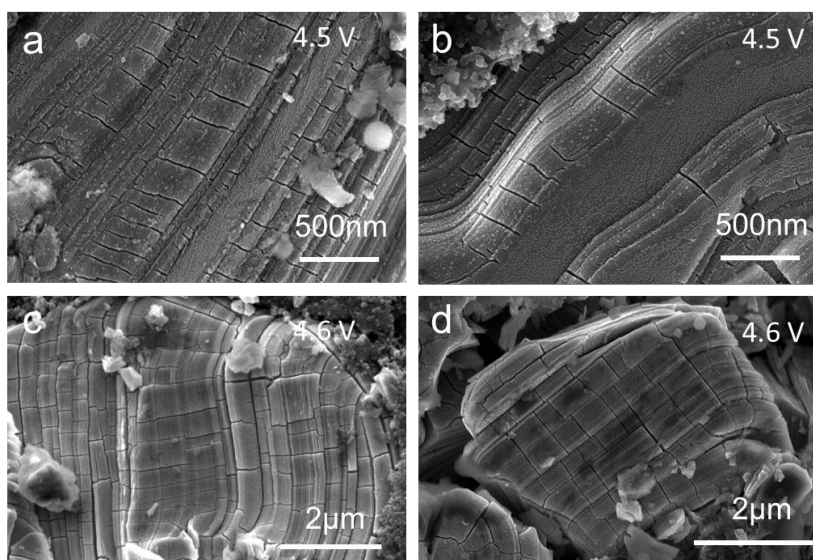
**Fig. S6.** (a) SEM images of the pristine LCO film surface. (b) SEM images of the LCO film surface after 50 cycles at 2.7-4.5 V.



**Fig. S7.** (a) A STEM-HAADF image of the LCO film and CEI layer. (b) The EDS spectrum and quantification results acquired from the red area in (a). (c) EDS mapping of LCO film and CEI layer.

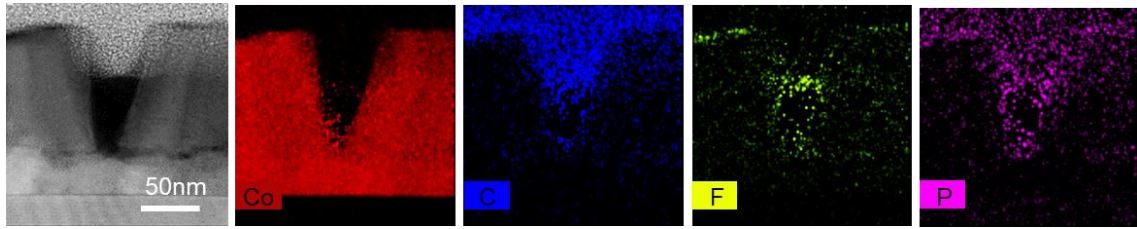


**Fig. S8** (a) Li1s and (b) C1s XPS spectra of the LCO film at pristine state and after 50 cycles at 2.7 V-4.5 V.



**Fig. S9.** (a, b) SEM images of the LCO particles after 100 cycles at 2.7 V-4.5 V. (c, d) SEM images of the LCO particles after 200 cycles at 2.7 V-4.6 V. High density of surface cracks are observed in both samples.





**Fig. S10.** EDS mapping of LCO film at the position with a crack to show that a large amount of carbon, fluorine and phosphorus accumulated inside the crack.