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Supporting Information

Interface Engineering on a Li Metal Anode for an Electro-Chemo-Mechanically Stable Anodic Interface in All-Solid-State Batteries

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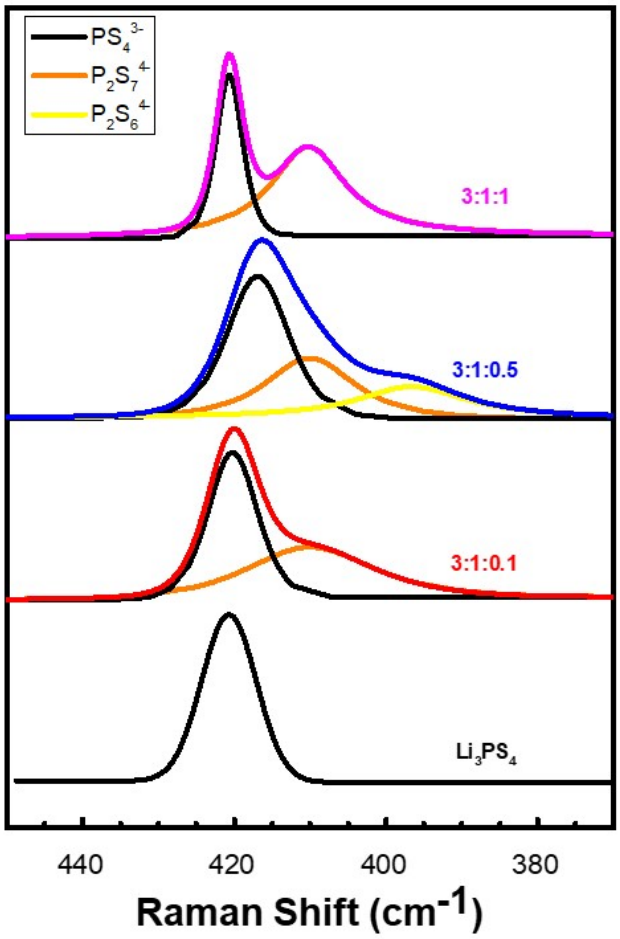
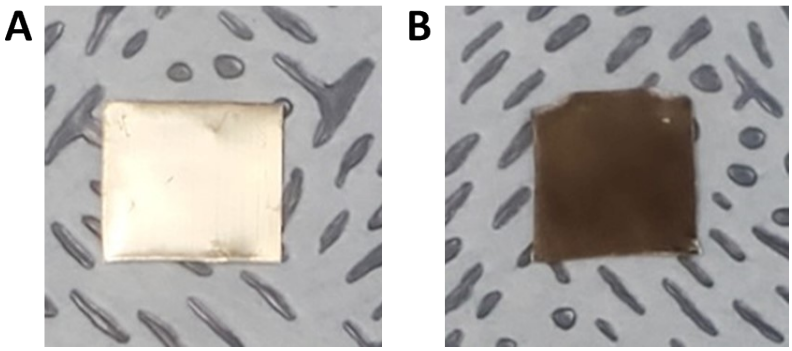


Fig. S1 Spectra decomposition of the Raman spectrum. The black, orange, and yellow lines are fitted deconvoluted curves for PS_4^{3-}, $P_2S_7^{4-}$, and $P_2S_6^{4-}$, respectively.



$$3Li_2S + P_2S_5 + 0.5InI_3 + Li\ metal \rightarrow xLi_4PS_4I + \ yLi_3PS_4 + z0.5LiI + wLi - In\ alloy$$

Fig. S2 Photograph of A) bare Li, B) IILPS-coated Li, and possible chemical reaction.

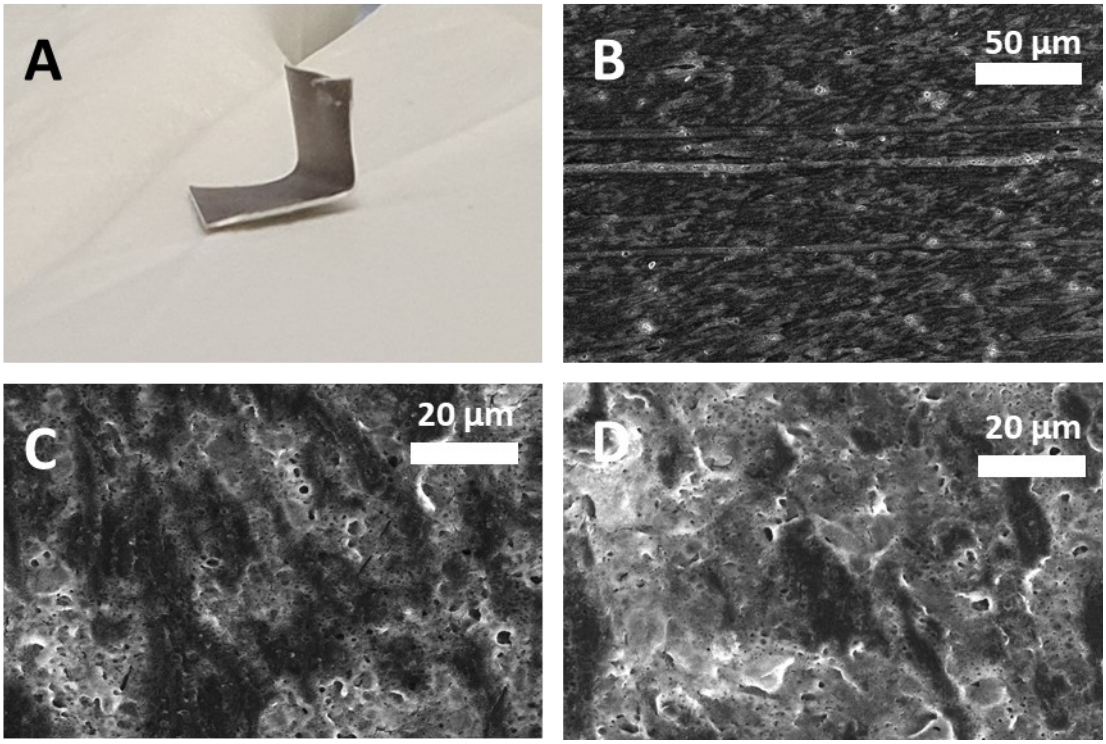


Fig. S3 A) Photograph of bending IILPS coated Li to 90° and B) SEM image IILPS coating layer on Li after bending and unfolding to 90°. SEM image of IILPS coating layer on Li C) before peel-off test and D) after peel-off test.

In SEM images of IILPS coating layer on Li metal after peel-off and bending tests, no noticeable surface crack is observed. This result suggests the physical stability of IILPS-coating layer on Li metal under harsh mechanical stress.

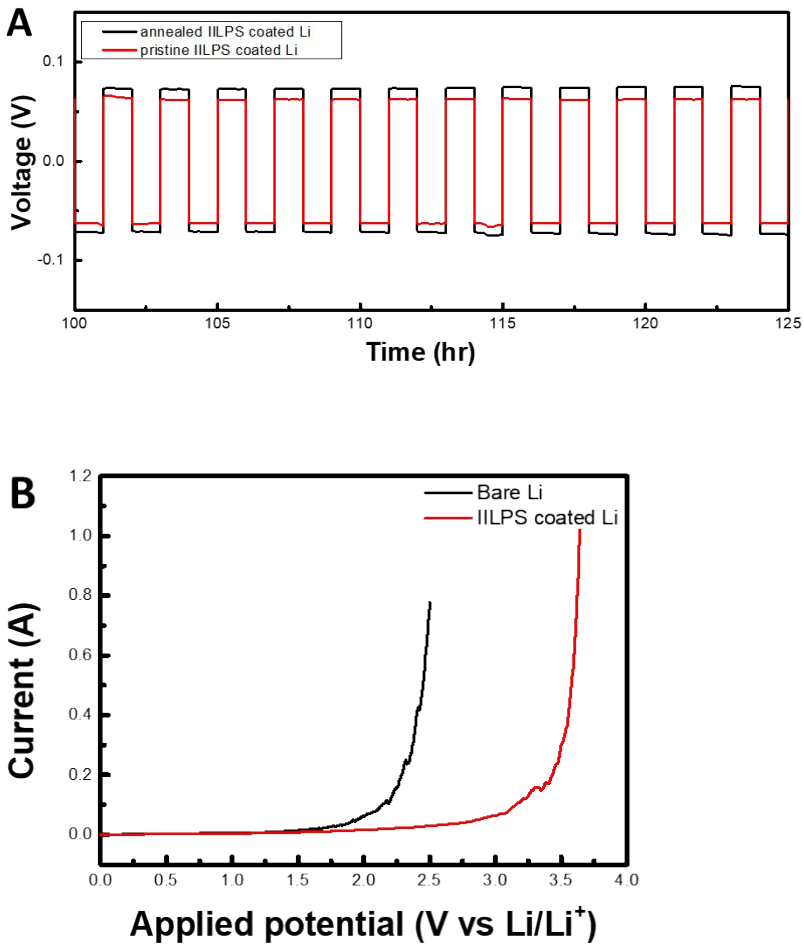


Fig. S4 A) Galvanostatic cycling test of Li/Li symmetric cells with pristine IILPS-coated Li and annealed (at 140 °C) IILPS-coated Li anodes. The symmetric cells were stacked as Li/IILPS//Li_3PS_4 SE//IILPS//Li. The cell was charged and discharged to a cut off capacity of 0.5 mAh cm^{-2} at a temperature of 60 °C and a pressure of 5 MPa. B) Linear sweep voltammetry tests of Li/IILPS//Li_3PS_4 SE//IILPS/Li and Li//Li_3PS_4 SE//Li at 60°C, 5 MPa.

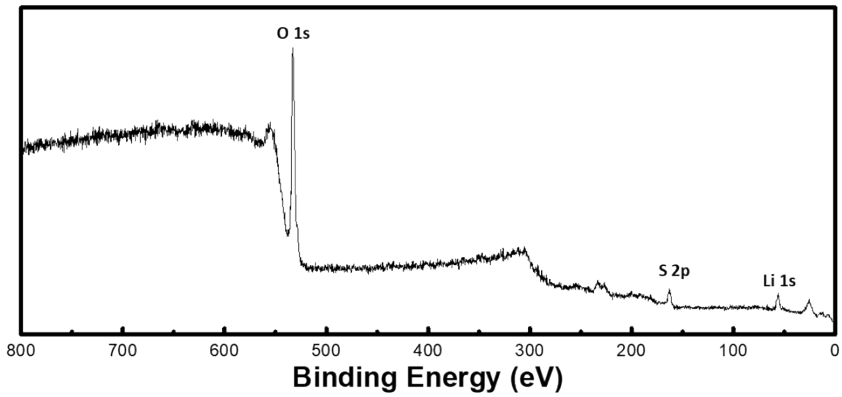


Fig. S5 XPS survey scan of the bare Li metal electrode after galvanostatic cycling.

The O 1s signal in this XPS survey scan of the cycled bare Li electrode was due to the exposure of the XPS sample to air during the sample loading process. Although the exposure time was very short, the sample was significantly oxidized. However, no other elements were detected in the XPS survey scans.

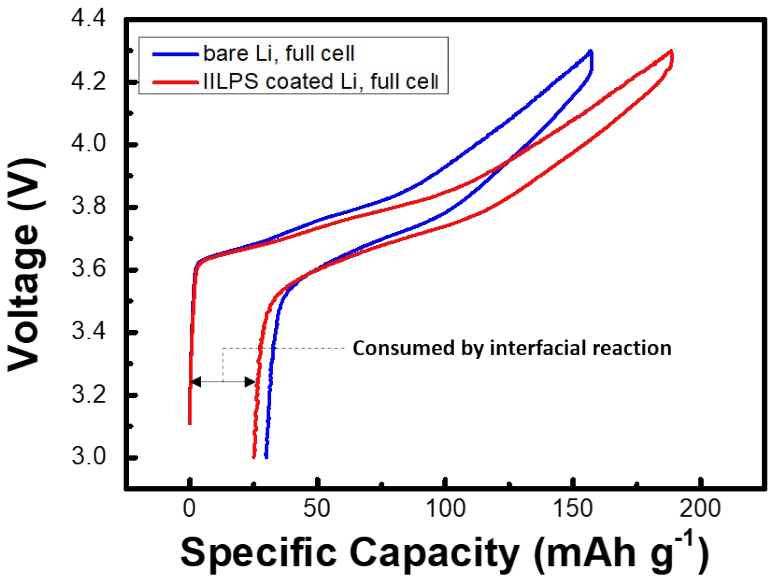


Fig. S6 First cycle voltage profiles of NCM811 full cells with a bare Li anode and an IILPS-coated Li anode. The full cells were stacked as NCM811//Li_3PS_4 SE //Li and NCM811/ Li_3PS_4 SE//IILPS/Li. The cells were tested at a current density of 0.1C (1C = 202 mA $g^{-1}_{cathode}$), a temperature of 60 ℃, and a pressure of 50 torque (~30 MPa). The voltage range was 3.0 to 4.3 V.

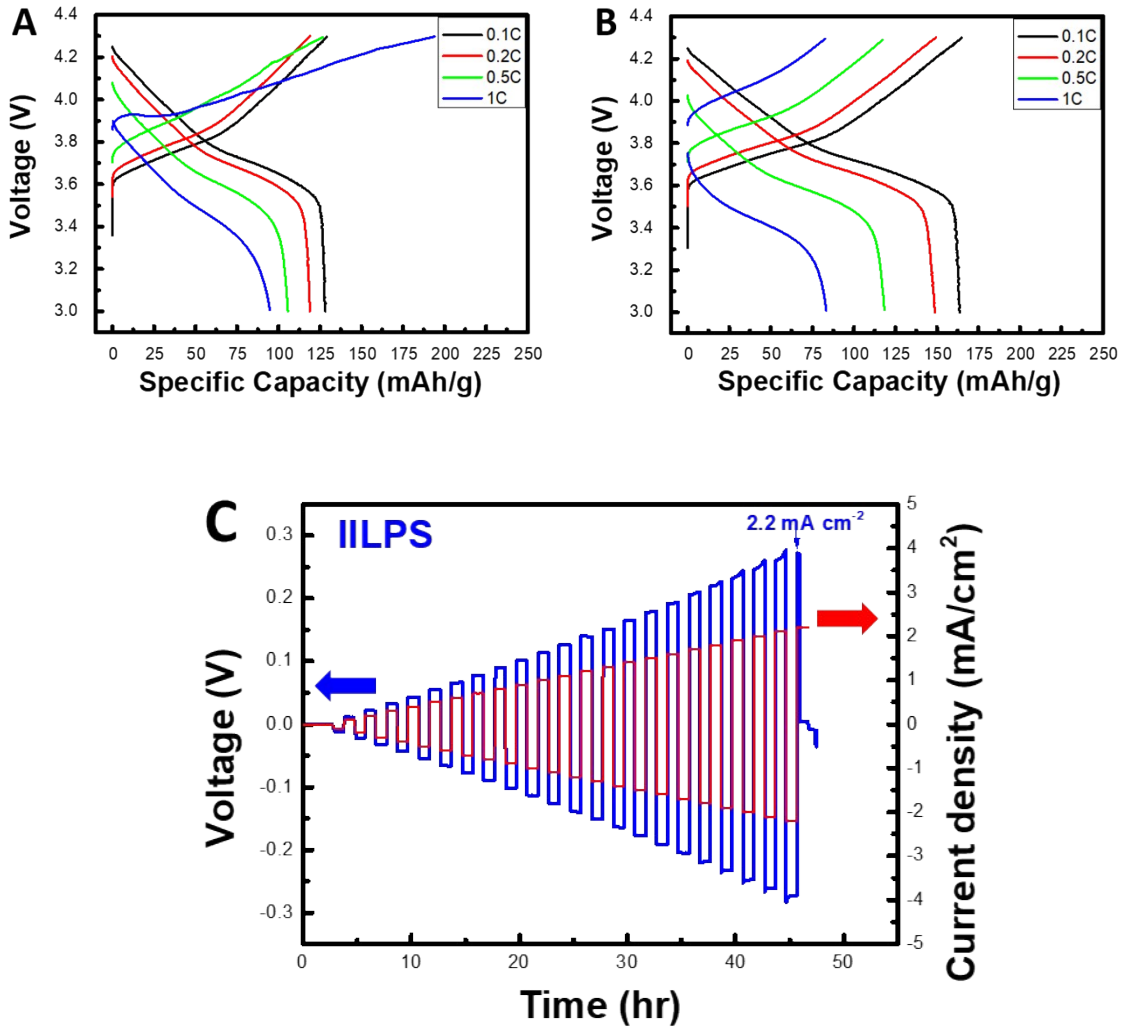


Fig. S7 Charge-discharge profiles of NMC 811 full cells with A) a bare Li anode and B) an IILPS-coated Li anode during the rate capability test of Fig. 8C. C) Galvanostatic cycling of the Li/IILPS//Li_3PS_4 SE//IILPS/Li cells for 1 hr plating/stripping at current densities from 0.1 to 2.2 mA cm^{-2} with 0.1 mA cm^{-2} step increase at 60 °C.