Supporting Information for

Correlating Nanoscale Structure with Electrochemical Property of Solid Electrolyte Interphases in Solid-State Battery Electrodes

Jimin Oh†, Gun Park†, Hongjun Kim†, Sujung Kim‡, Dong Ok Shin†, Kwang Man Kim†, Hye Ryung Byon#, Young-Gi Lee‡, and Seungbum Hong†,*

†Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejon 34141, Republic of Korea

‡ICT Creative Research Laboratory, Electronics and Telecommunications Research Institute (ETRI), Daejon 34129, Republic of Korea

#Department of Chemistry, Korea Advanced Institute of Science and Technology (KAIST), Daejon 34141, Republic of Korea

*Corresponding Author: seungbum@kaist.ac.kr
Figure S1. Lithiation profiles of NG electrode with different DOLs.
Figure S2. Voltage and specific capacity, as function of LSTP contents.
Figure S3. Current-voltage profile in Ref. (LSTP0), LSTP12, and LSTP30 electrodes.
Figure S4. Height maps of pristine and 1 cycled electrode in LSTP50 and LSTP70.
Figure S5. Height, amplitude at different 10 positions, and correlation maps at selected 5 positions from height, amplitude images (colored images) of 1 cycled electrodes in a) Ref. (LSTP0), b) LSTP12, c) LSTP24, and d) LSTP30.
Figure S6. EIS results of pristine and 1 cycle electrodes in a) Ref. (LSTP0) and b) LSTP30.
Figure S7. Typical force-distance curves of 1 cycled electrode in a) Ref. (LSTP0) and b) LSTP30 (spring constant $k = 0.164$ N m$^{-1}$).
Figure S8. SEM/EDS results of pristine and cycled electrodes in (a) Ref. (LSTP0), (b) LSTP12, (c) LSTP24, and (d) LSTP30 samples.

Figure S9. SEM images of pristine [(a)-(d), (i)-(l)] and after 1 cycled electrodes [(e)-(h), (m)-(p)] of LSTP0 [(a), (e), (i), (m)], LSTP12 [(b), (f), (j), (n)], LSTP24 [(c), (g), (k), (o)], and LSTP30 [(d), (h), (l), (p)] samples at two different scales (see scale bars). SEM images of LSTP12 sample [(q), (r), (s)] and corresponding EDS images for (s) mapping C, O and Ti elemental distributions [Reproduced with permission from H. Kim, PhD thesis, KAIST (2020)].
Figure S10. Photoelectron signals during argon etching in C 1s, F 1s, O 1s and Li 1s signals in 1 cycled a) LSTP24 and b) LSTP30 electrodes.
Table S1. Depth distributions in the lithium and fluorine

|       | max([Li])-min([Li]) | a) [Li]_{surface} | max([Li])-min([Li]) / [Li]_{surface} |
|-------|---------------------|-------------------|--------------------------------------|
| LSTP24 | 4.11                | 38.73             | 0.106                                |
| LSTP30 | 2.25                | 38.28             | 0.059                                |

|       | max([F])-min([F])  | a) [F]_{surface}  | max([F])-min([F]) / [F]_{surface}   |
|-------|---------------------|-------------------|--------------------------------------|
| LSTP24 | 8.79                | 14.28             | 0.615                                |
| LSTP30 | 10.03               | 20.14             | 0.498                                |

a) [X]_{surface} means the atomic percentage of X element at etching time 0 s