

## Supporting Information

### Functional-Oriented Design of Gradient Composite Fluoride Interphase for Enhanced Silicon Anode Performance

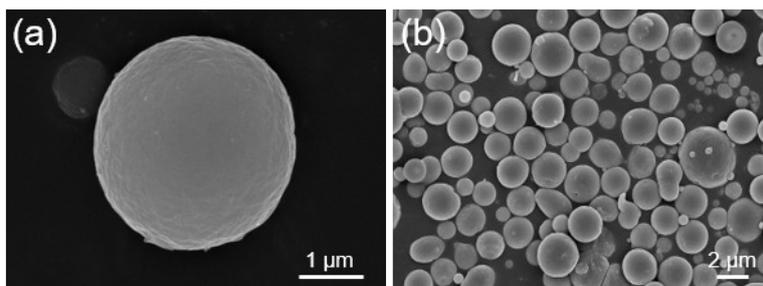
*Yu Jing<sup>a</sup>, Zhixing Wang<sup>a, b, c</sup>, Huajun Guo<sup>a, b, c</sup>, Xinhai Li<sup>a, b, c</sup>, Hui Duan<sup>a, b, c</sup>, Wenjie Peng<sup>a, b, c</sup>, Guochun Yan<sup>a, b, c</sup>, Guangchao Li<sup>a, b, c\*</sup>, Jiexi Wang<sup>a, b, c</sup>*

<sup>a</sup> School of Metallurgy and Environment, Central South University, 410083 Changsha, China

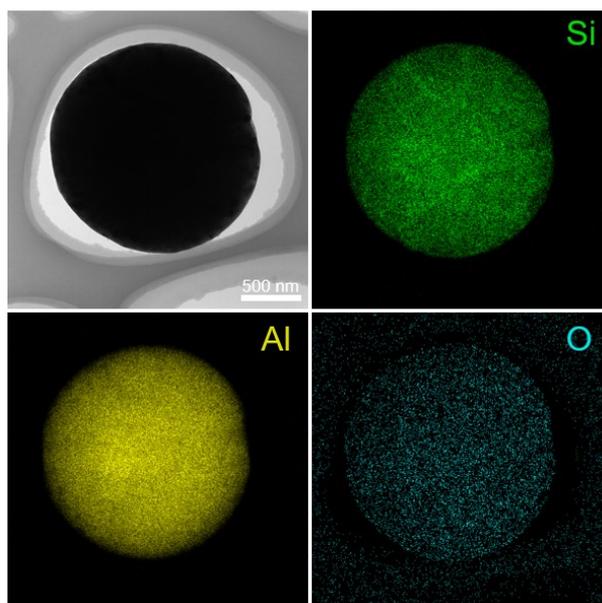
<sup>b</sup> Engineering Research Center of the Ministry of Education for Advanced Battery Materials, Central South University, Changsha, 410083, China

<sup>c</sup> Hunan Provincial Key Laboratory of Nonferrous Value-Added Metallurgy, Central South University, Changsha, 410083, China

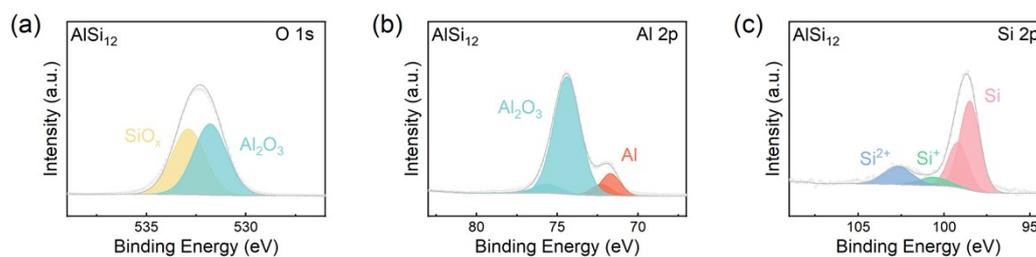
Corresponding author e-mail address: [guangchao\\_li@csu.edu.cn](mailto:guangchao_li@csu.edu.cn);



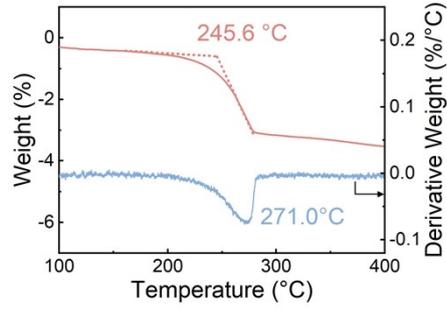
**Figure S1.** (a, b) SEM images of pristine  $\text{AlSi}_{12}$  alloy



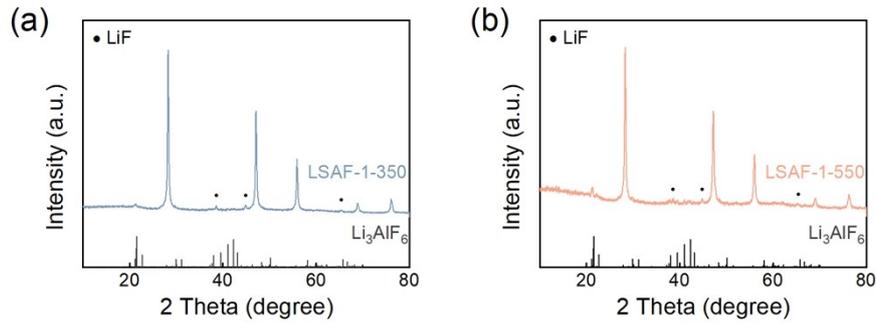
**Figure S2.** TEM and elemental mapping images of  $\text{AlSi}_{12}$  alloy



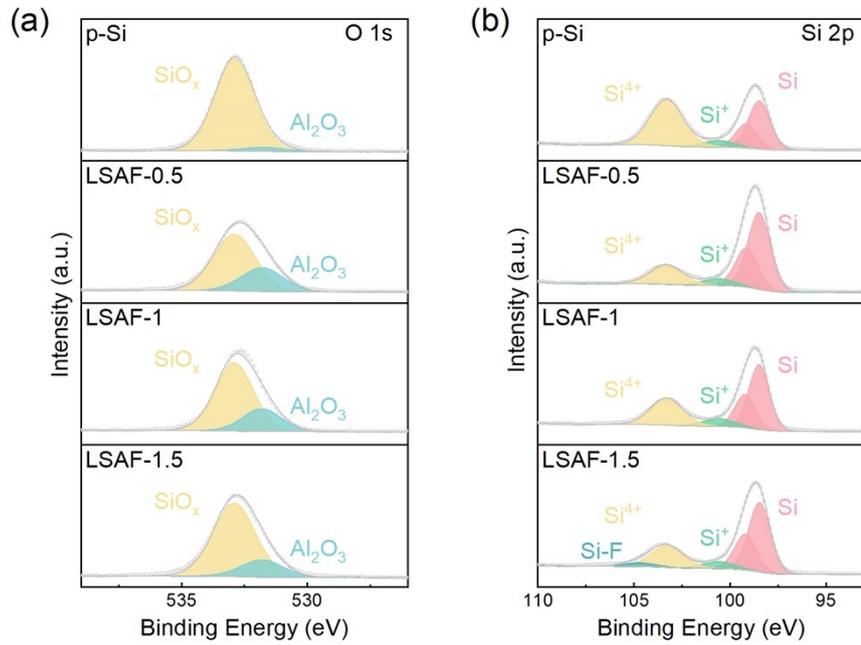
**Figure S3.** The XPS spectra of  $\text{AlSi}_{12}$  alloy. (a) O 1s, (b) Al 2p, (c) Si 2p



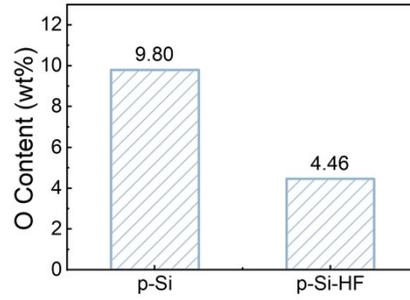
**Figure S4** DTG curves of LSAF-1



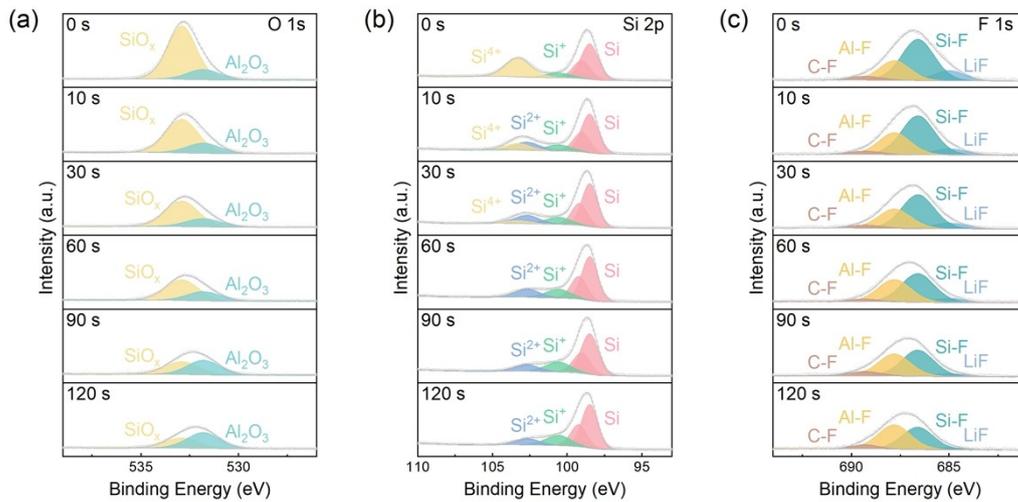
**Figure S5.** XRD pattern of LSAF-1 after heat treatment at (a) 350 and (b) 550 °C



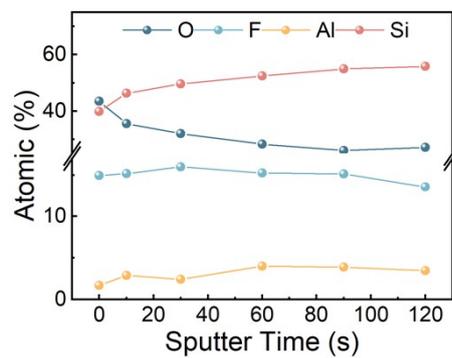
**Figure S6.** (a) O 1s and (b) Si 2p XPS spectra of materials



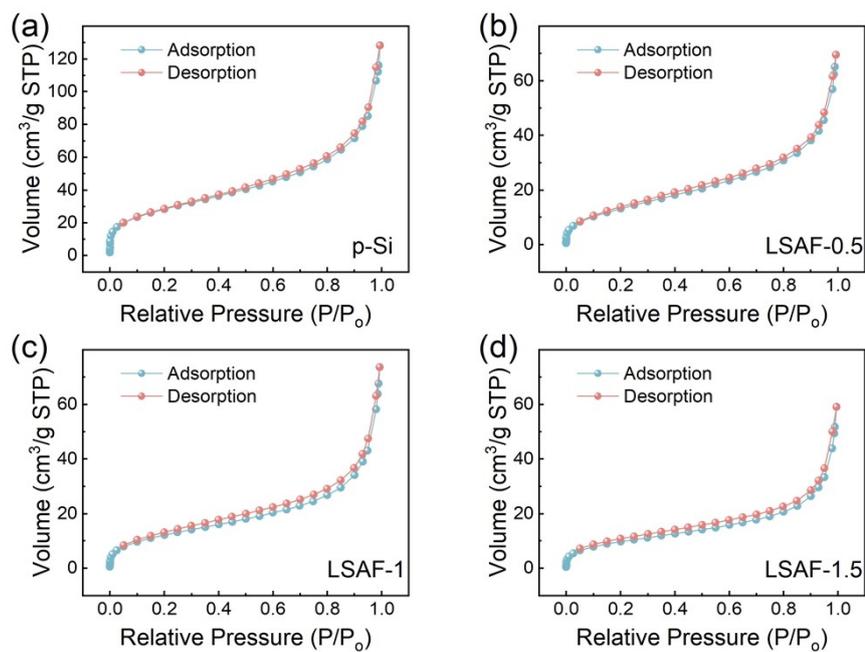
**Figure S7.** Comparison of the oxygen content in p-Si before and after HF etching



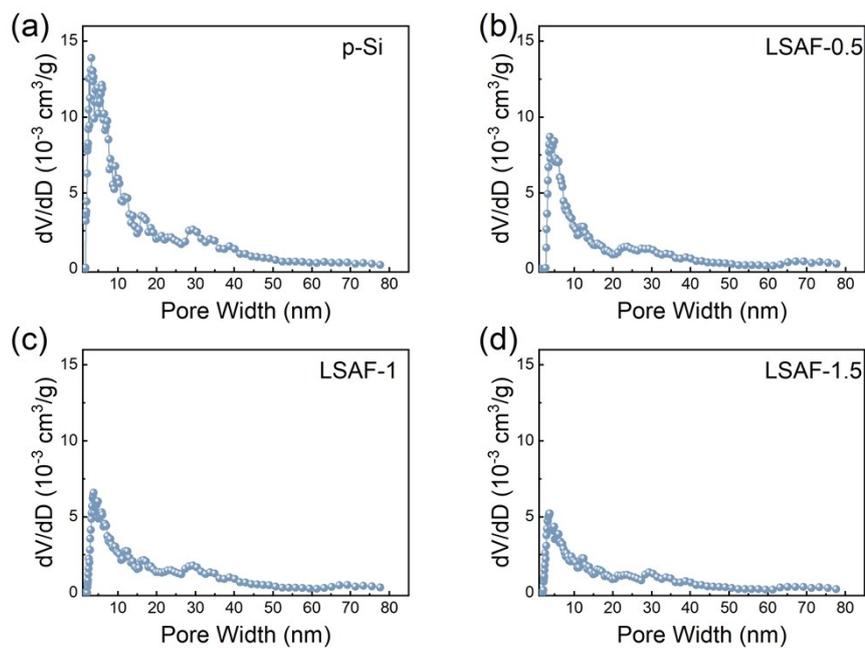
**Figure S8.** (a) O 1s, (b) Si 2p and (c) F 1s spectra of LSAF-1 after different  $\text{Ar}^+$  etching times



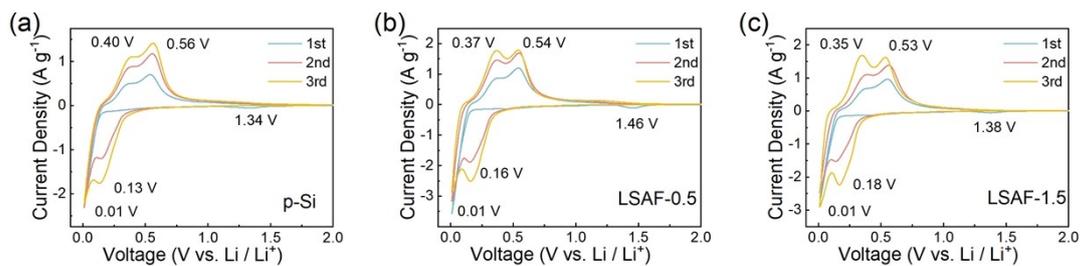
**Figure S9.** The atomic percentage of each element in the LSAF-1 sample with sputter time



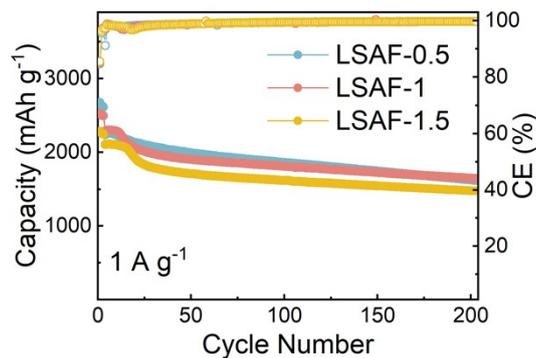
**Figure S10.**  $N_2$  isothermal adsorption/desorption curves of (a) p-Si, (b) LSAF-0.5, (c) LSAF-1, (d) LSAF-1.5



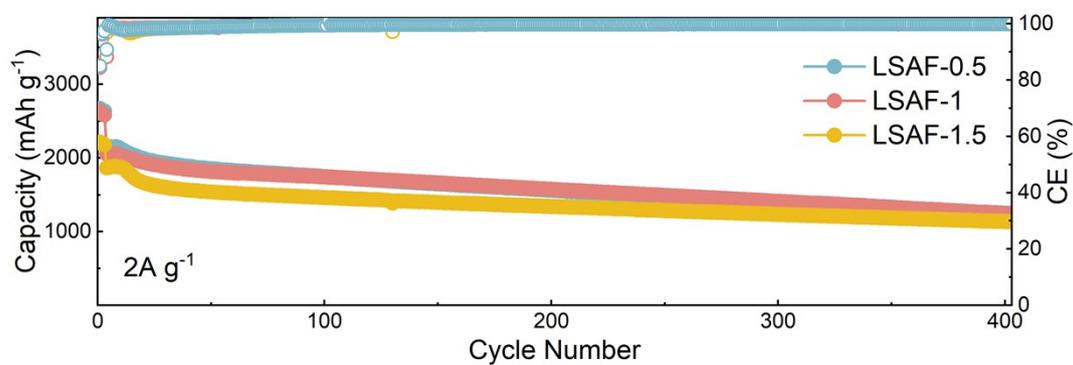
**Figure S11.** Pore size distribution of (a) p-Si, (b) LSAF-0.5, (c) LSAF-1, (d) LSAF-1.5



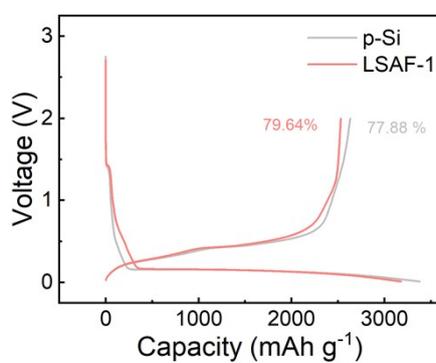
**Figure S12.** CV curves of (a) p-Si, (b) LSAF-0.5, (c) LSAF-1.5



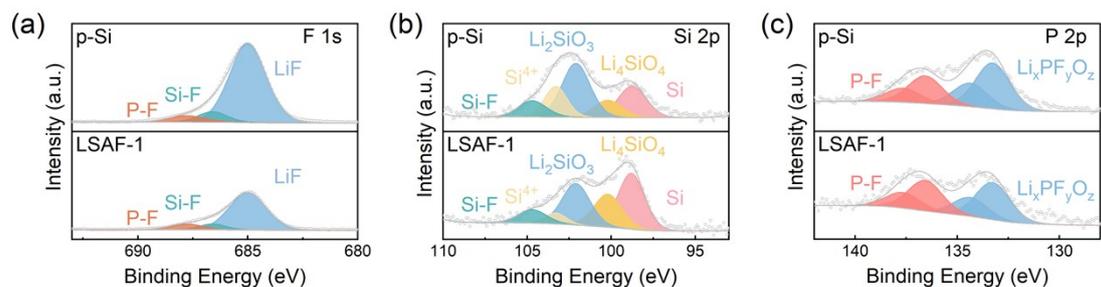
**Figure S13.** Cycle performance of materials with different coating contents at  $1 \text{ A g}^{-1}$



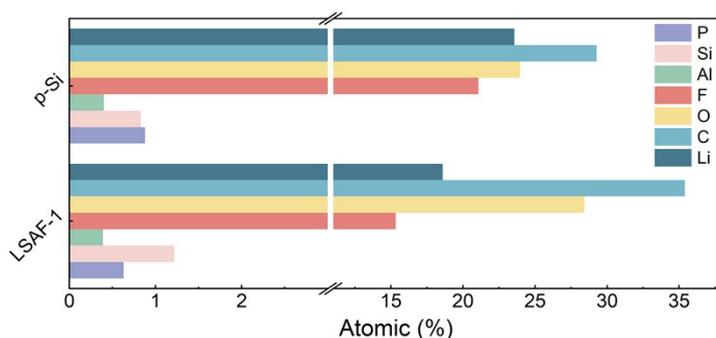
**Figure S14.** Cycle performance of materials with different coating contents at  $2 \text{ A g}^{-1}$



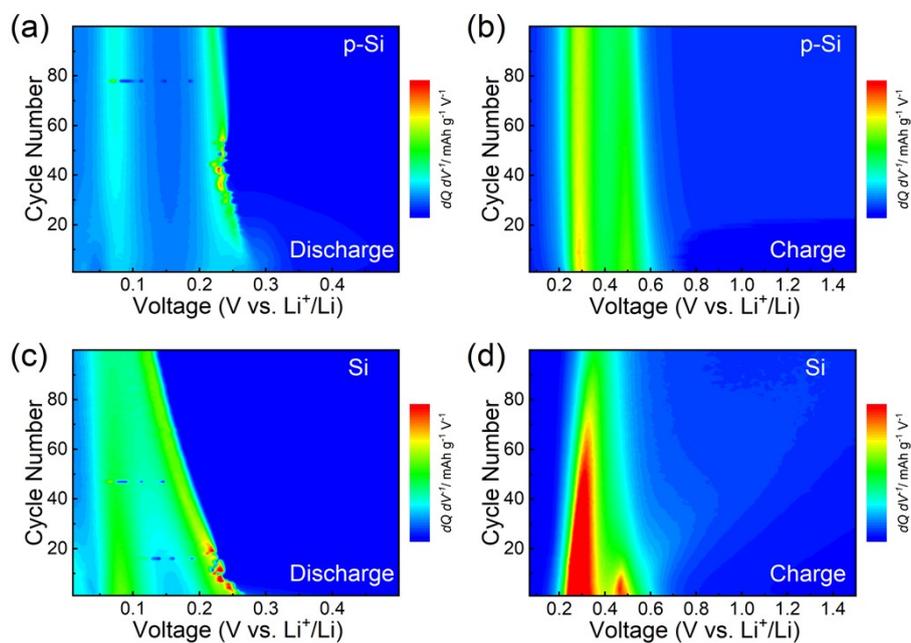
**Figure S15.** charge-discharge curves under  $45 \text{ }^\circ\text{C}$



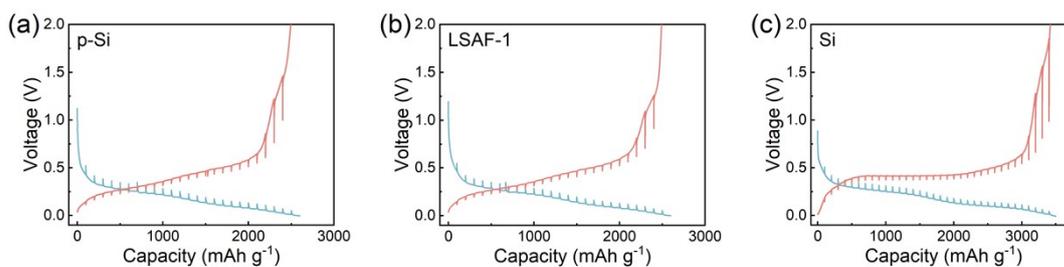
**Figure S16.** (a) F 1s, (c) Si 2p and (d) P 2p XPS spectra of electrodes after 3 cycles at  $0.2 \text{ A g}^{-1}$



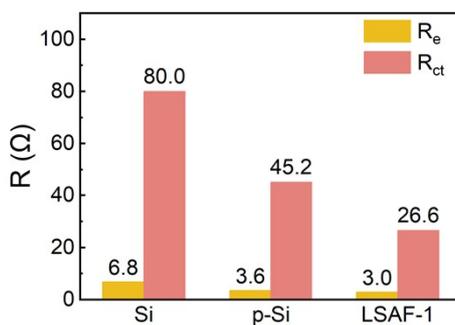
**Figure S17.** The atomic percentage of each element in the SEI film



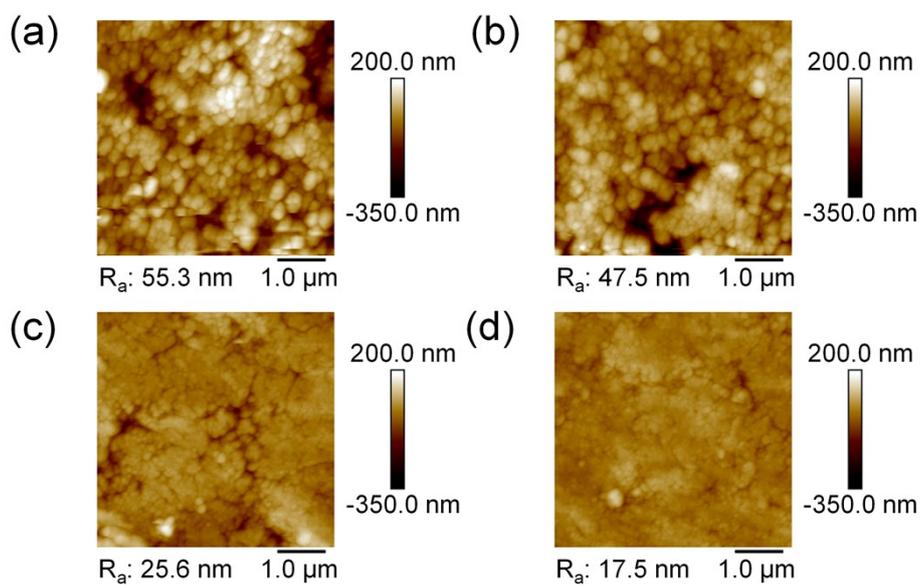
**Figure S18.**  $dQ/dV$  mappings for the (a) discharging and (b) charging processes of p-Si.  $dQ/dV$  mappings for the (c) discharging and (d) charging processes of Si



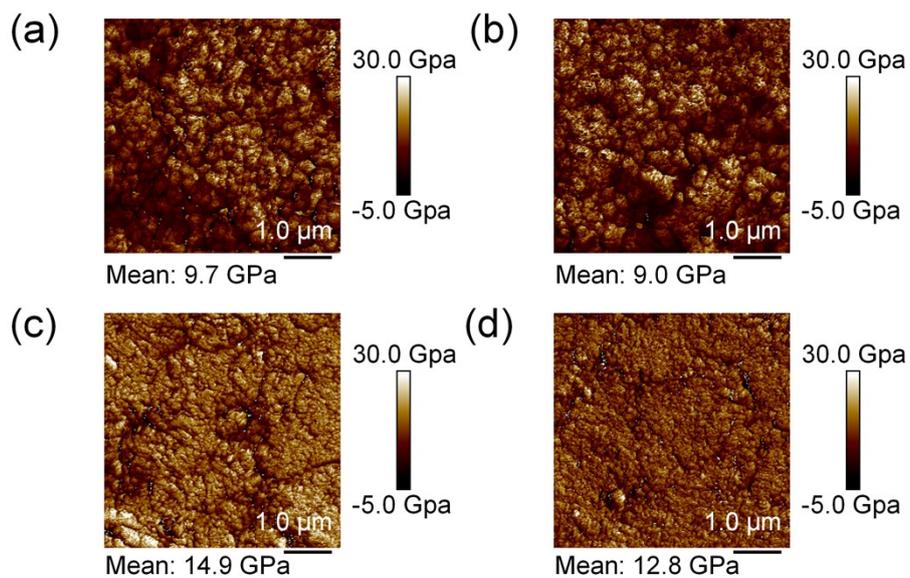
**Figure S19.** GITT curves of (a) p-Si, (b) LSAF-1, (c) Si



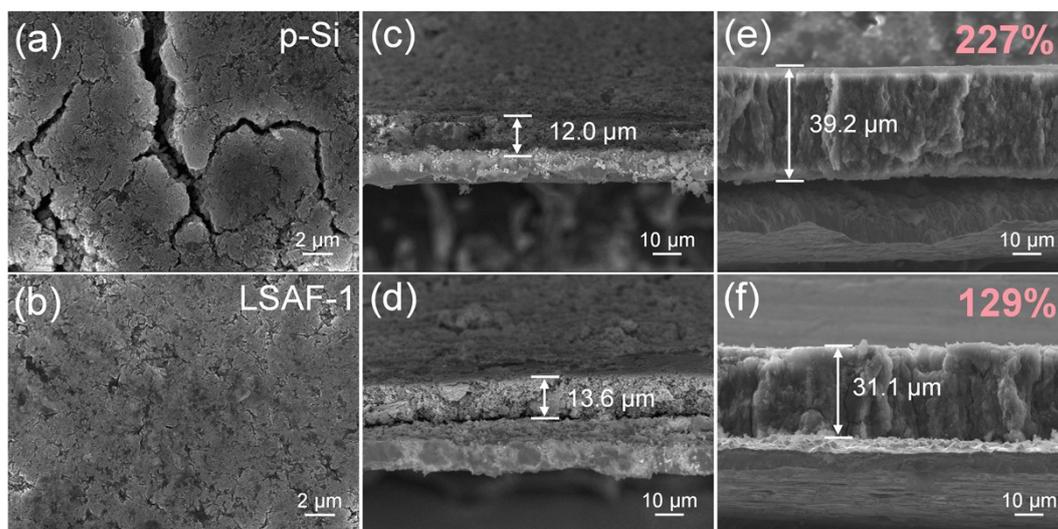
**Figure S20.** EIS fitting parameters of electrodes before cycling



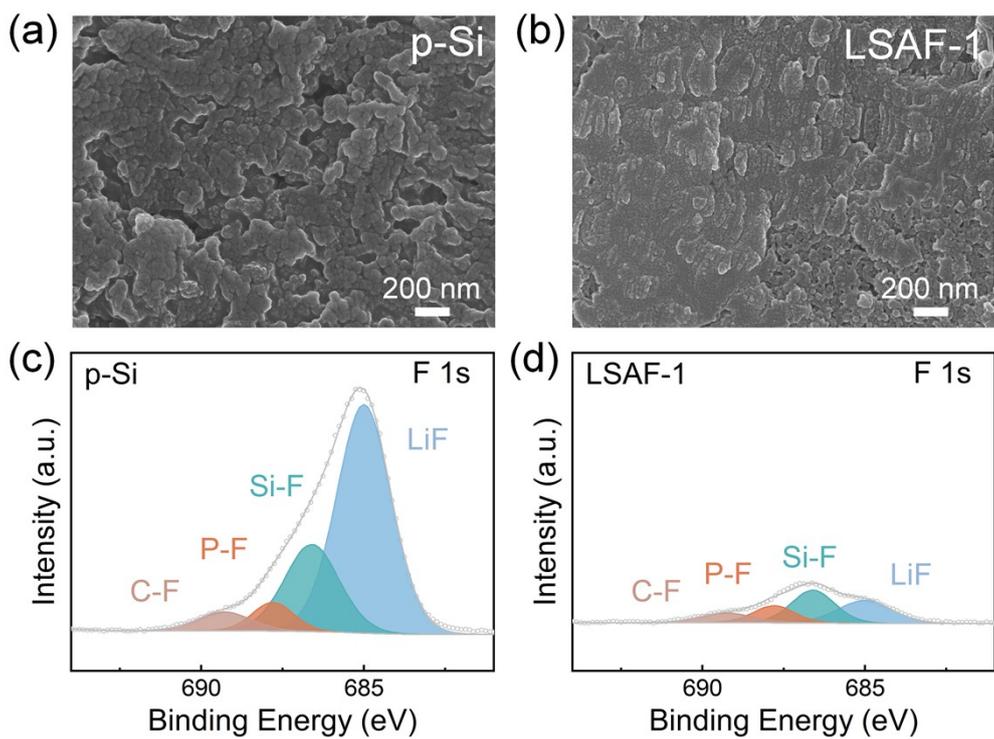
**Figure S21.** Surface roughness of electrodes in different regions. (a-b) p-Si; (c-d) LSAF-1.



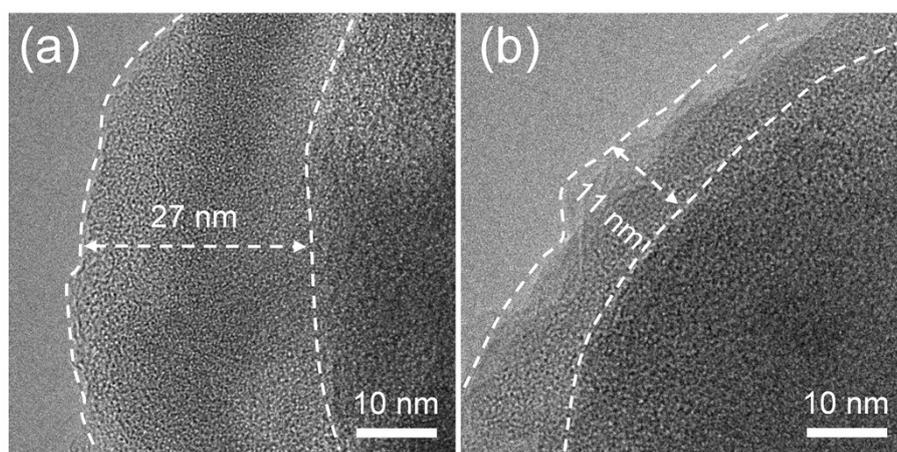
**Figure S22.** 2D plots of DMT modulus of electrodes in different regions. (a-b) p-Si; (c-d) LSAF-1.



**Figure S23.** SEM images of p-Si and LSAF-1 anodes. (a-b) Top view after cycling; the cross-section view (c-d) before and (e-f) after cycling.



**Figure S24.** Surface morphology and chemical state analysis of electrodes after high-temperature cycling. SEM images of (a) p-Si and (b) LSAF-1 anodes. F 1s XPS spectra of (c) p-Si and (d) LSAF-1 anodes.



**Figure S25.** HRTEM images of (a) p-Si and (b) LSAF-1 anodes after cycling.