

**Supporting Information**  
**of**  
**“Natto” binder of poly- $\gamma$ -glutamate enabling to enhance**  
**silicon/graphite composite electrode performance**  
**for lithium-ion batteries**

Takahiro Mochizuki,<sup>1</sup> Shoko Aoki,<sup>1</sup> Tatsuo Horiba,<sup>1</sup> Martin Schulz-Dobrick,<sup>2</sup> Zhen-Ji Han,<sup>2</sup> Sayuri Fukuyama,<sup>2</sup> Hiroshi Oji,<sup>3</sup> Satoshi Yasuno,<sup>3</sup> and Shinichi Komaba<sup>1,\*</sup>

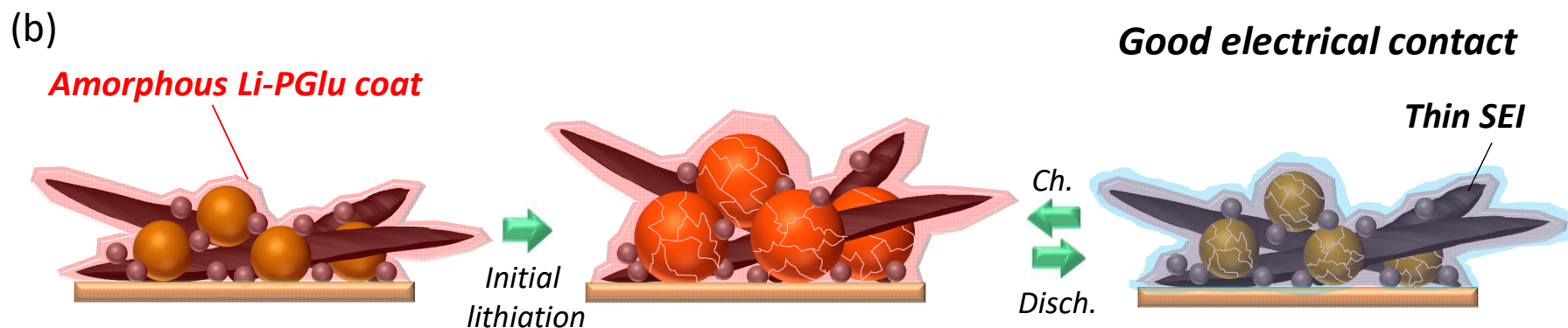
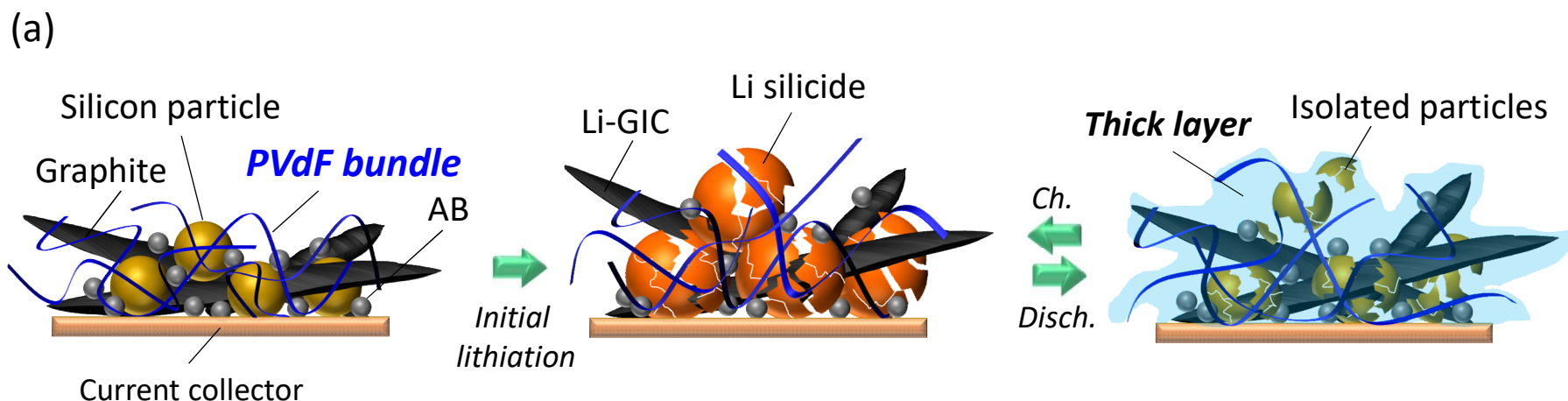
<sup>1</sup> *Department of Applied Chemistry, Tokyo University of Science, Tokyo 162-8601, Japan*

<sup>2</sup> *BASF Japan Ltd., Amagasaki, Hyogo 660-0083, Japan*

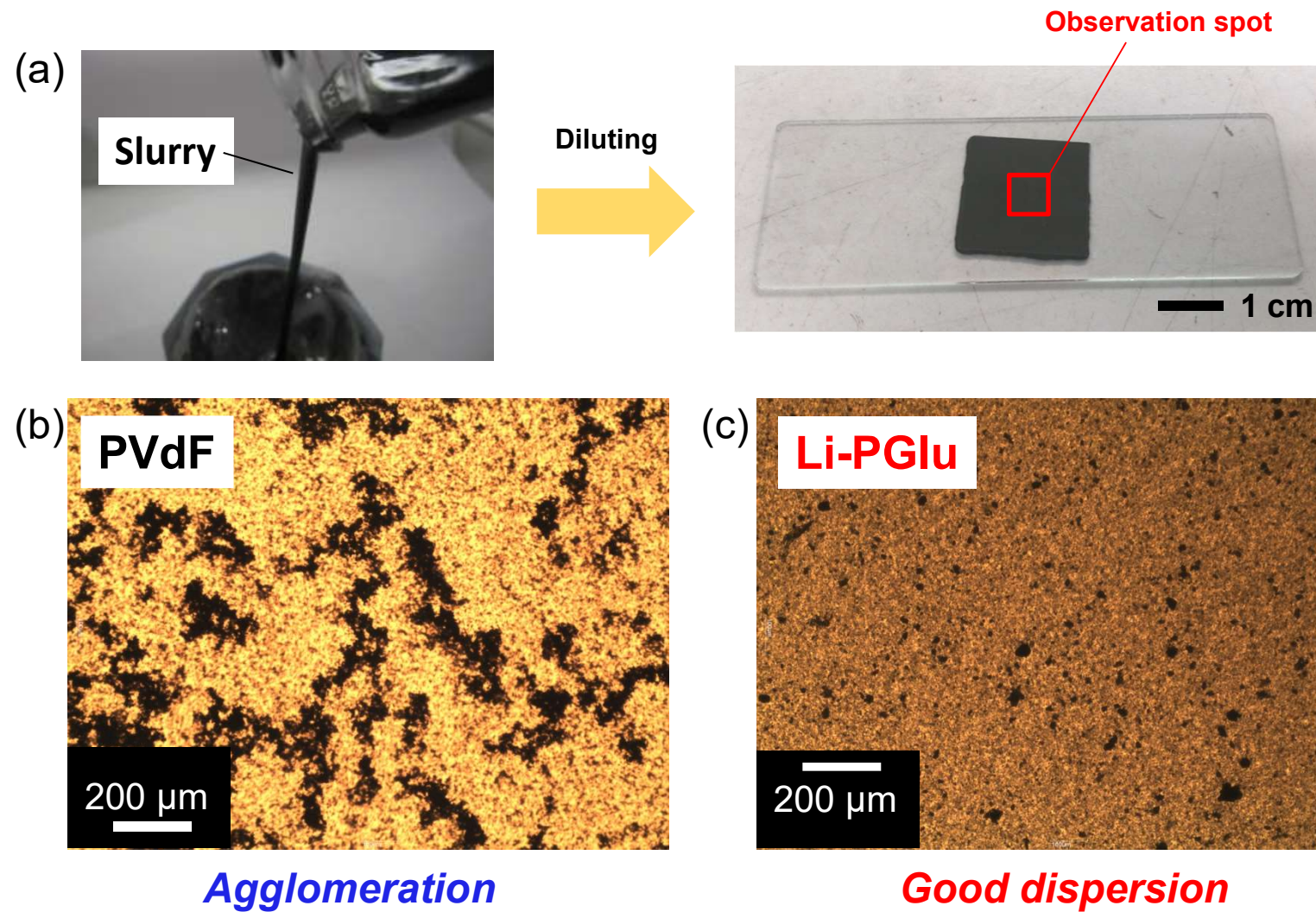
<sup>3</sup> *Japan Synchrotron Radiation Research Institute (JASRI), 1-1-1 Kouto, Sayo-gun, Hyogo, 679-5198 Japan*

*\*Correspondence to komaba@rs.kagu.tus.ac.jp*

The numbers of pages, figures, and tables are 6, 5, and 0, respectively.

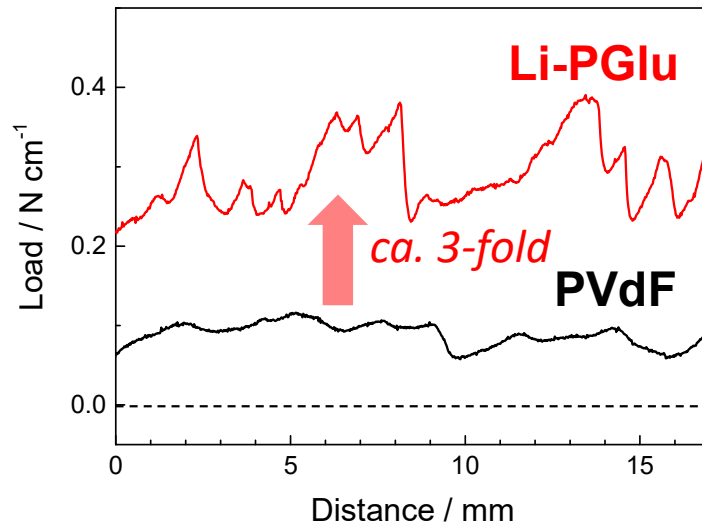
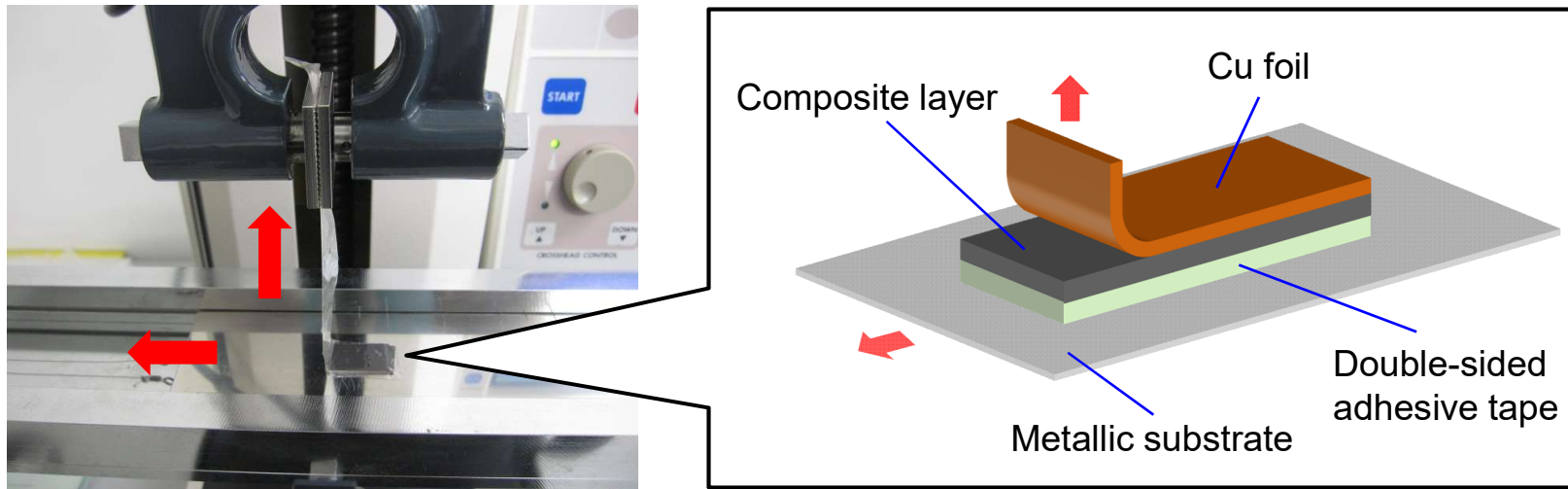


**Figure S1** Illustration of morphology change of Si/graphite composite electrode with (a) PVdF and (b) Li-PGlu binders during lithiation and delithiation cycle, accompanied with large volume change by lithium silicide formation and annumulation of electrolyte decomposition products.



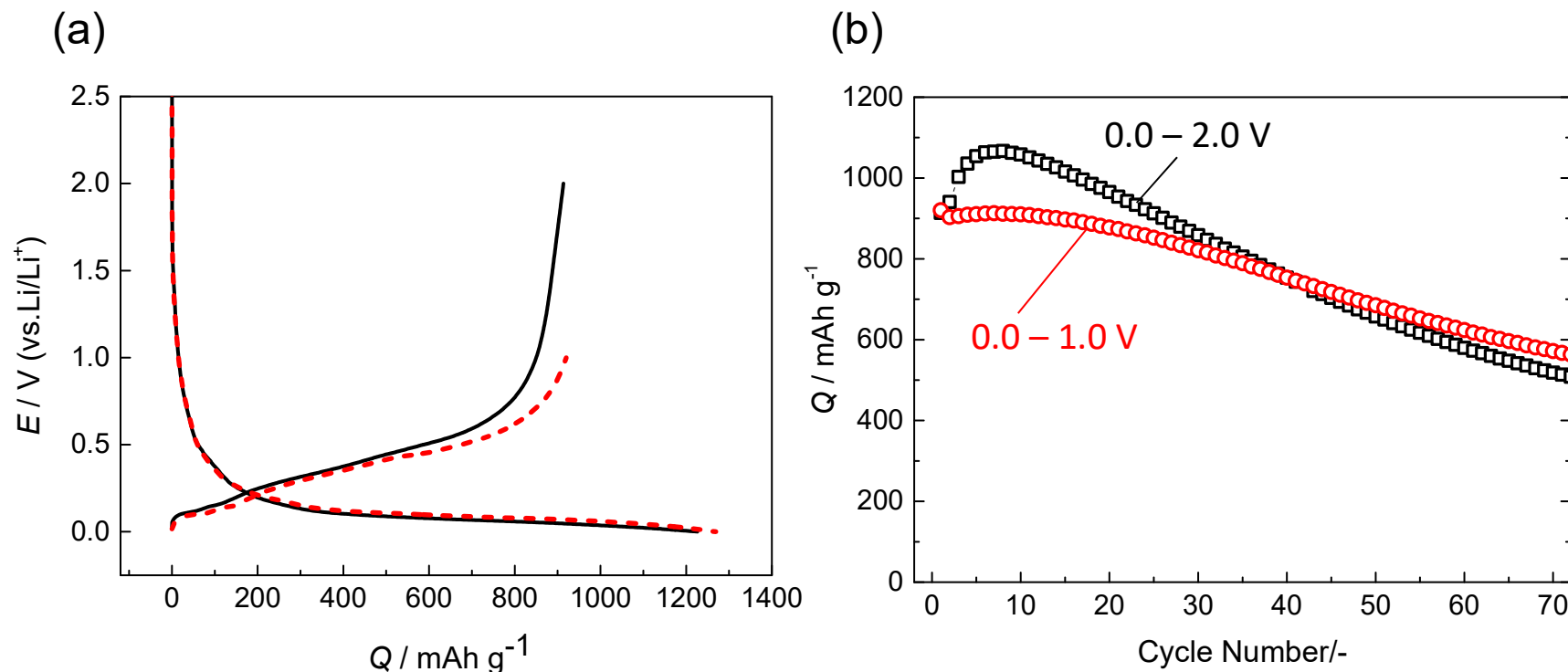
**Figure S2** Optical microscopic observation; (a) set-up of slurry sample and images of slurries (electrode mixture dispersion) with (a) PVdF in NMP and (b) Li-PGlu in water.

### Photo of setup of 90 °-peel test

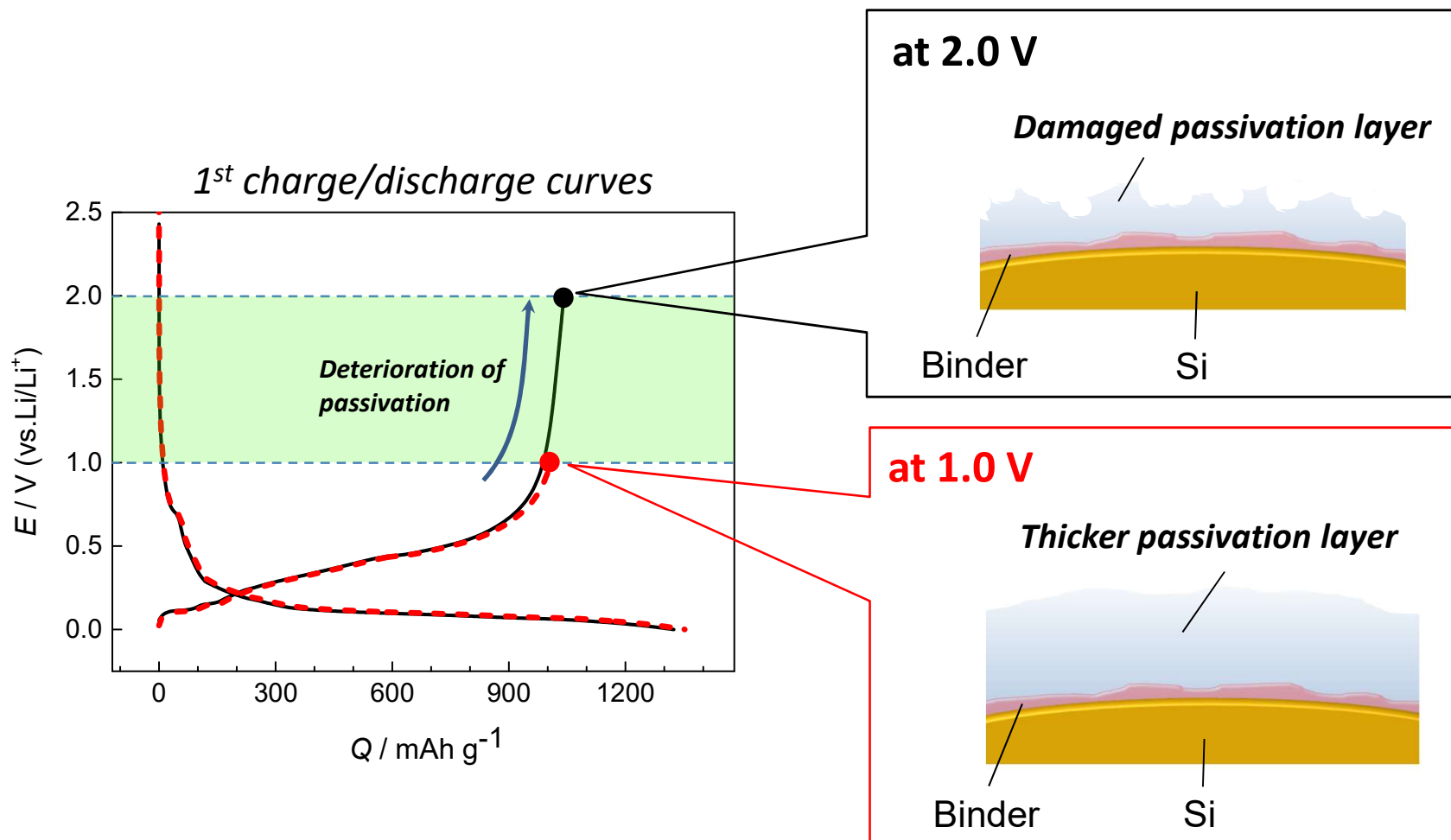


| Tention / N cm <sup>-1</sup> |      |
|------------------------------|------|
| PVdF                         | 0.09 |
| Li-PGlu                      | 0.29 |

**Figure S3** Adhesive strength test of the PVdF and Li-PGlu composite electrode onto Cu foil.



**Figure S4** Charge-discharge performance of the electrodes of Si:graphite:AB:Li-PGlu = 3:5:1:3. To modulate emphasis of the binder dependency in the spectra, the binder content in electrode samples for HAXPES measurement was increased from 3:5:1:1 to 3:5:1:3; (a) charge/discharge curves at the first cycle and (b) discharge capacity versus cycle number plots.



**Figure S5** Schematic illustration of anodic damage of SEI surface layer on Si tested under 2.0 V and 1.0 V cut-off conditions for the HAXPES analysis of Figure 11.