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Supplementary information

Fabrication of liquid cell for in situ transmission electron microscopy of electrochemical processes

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for

Fabrication of Liquid Cell for In-Situ Transmission Electron Microscopy of Electrochemical Processes

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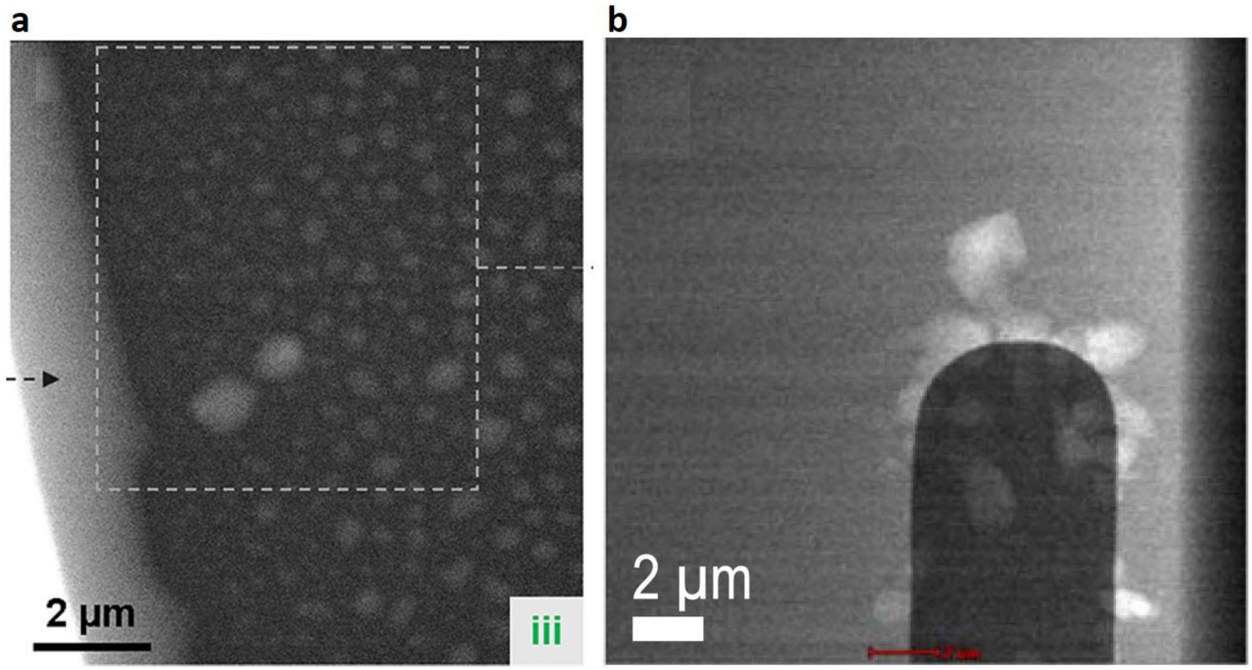
Supplementary Table

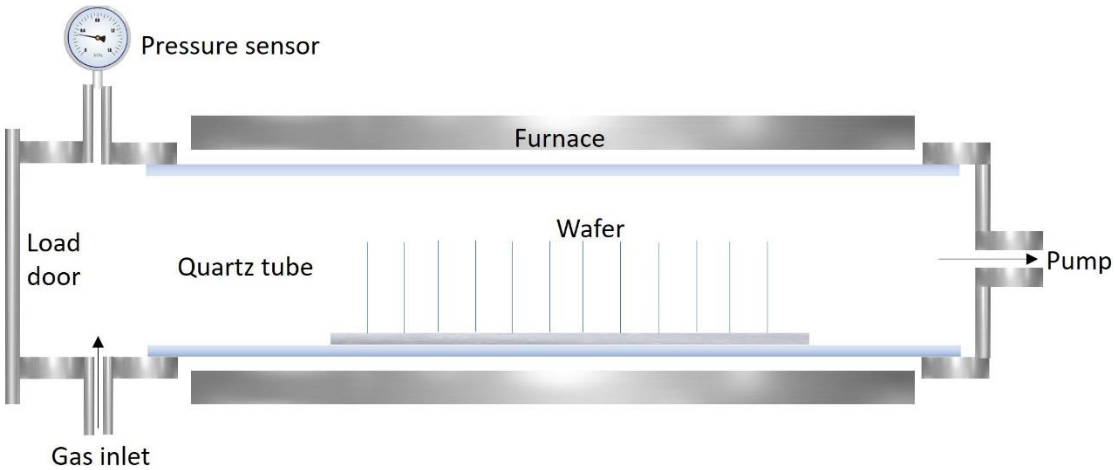
Supplementary Table 1. Comparisons between commercially developed electrochemical liquid cell and our design.

Key parameters	Hummingbird Scientific https://hummingbirdscientific.com/	Protochips https://www.protochips.com/	This protocol
Thickness of SiN_x window (nm)	~50	~50	~35
Thickness of liquid layer (nm)	~1000	~1000 or ~500	~150
TEM spatial resolution (nm)	~100	~100	~15
Size of SiN_x window (μm ×μm)	~ 50 × 50	~ 400 × 50	~ 25 × 6
Size of reservoir (μm ×μm)	-	-	~ 700 × 500
Mechanical strength	Weaker	Weaker	Stronger
Acid resistance	Stronger	Stronger	Weaker
Sealing material	*O-ring*	*O-ring*	Indium metal and epoxy
Reusable	Yes	Yes	No

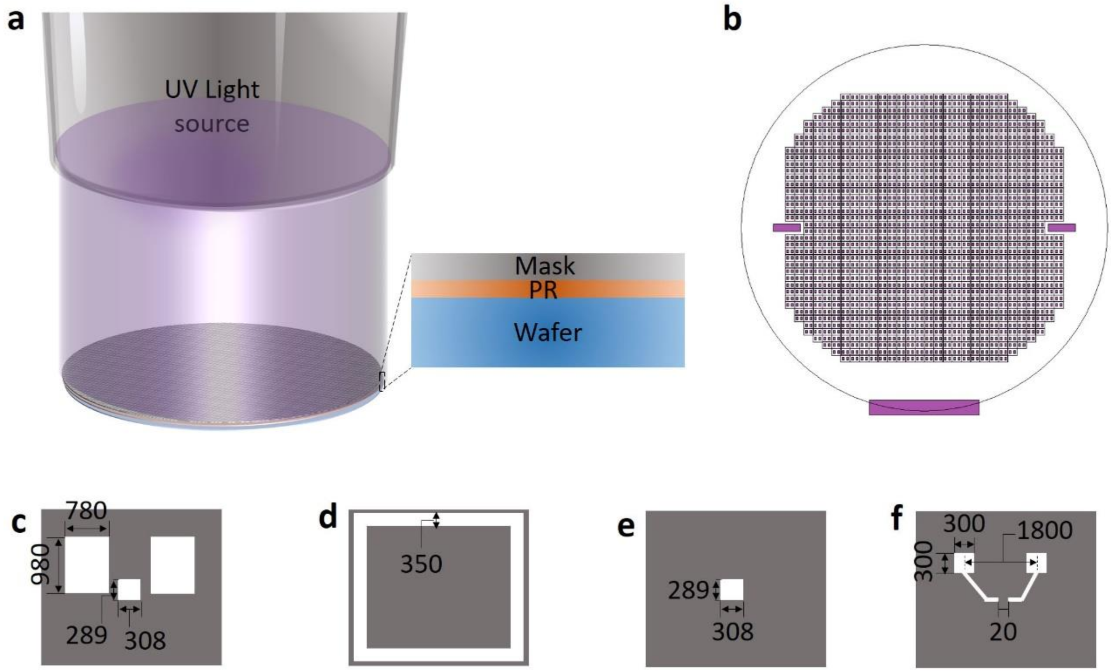
Supplementary Figures



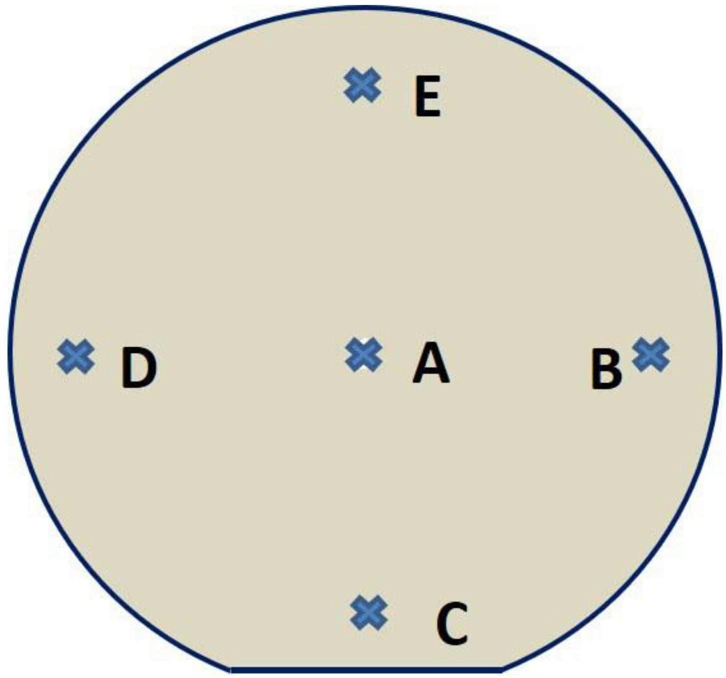
Supplementary Figure 1 | **Comparison of spatial resolution. a |** TEM image observed using electrochemical liquid cell of Hummingbird company. **b |** TEM image observed using electrochemical liquid cell of Protochips company. Part **a** is adapted with permission from ref.[1]. American Chemistry Society. Part **b** is adapted with permission from ref.[2]. Springer Nature Limited.



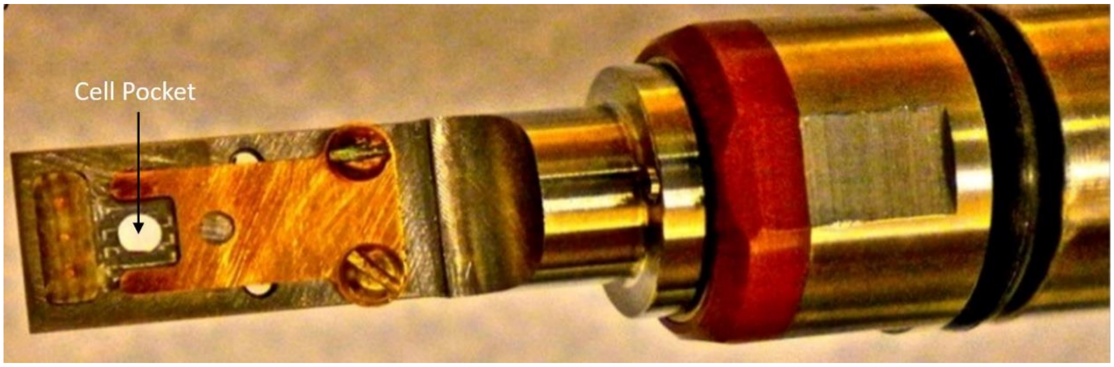
Supplementary Figure 2 | Schematic illustration of the low-pressure chemical vapor deposition (LPCVD) setup. LPCVD furnace. For the detailed information, the reader is referred to the Chapter 5.17 of the following link: (https://nanolab.berkeley.edu/public/manuals/equipment_manual.shtml).



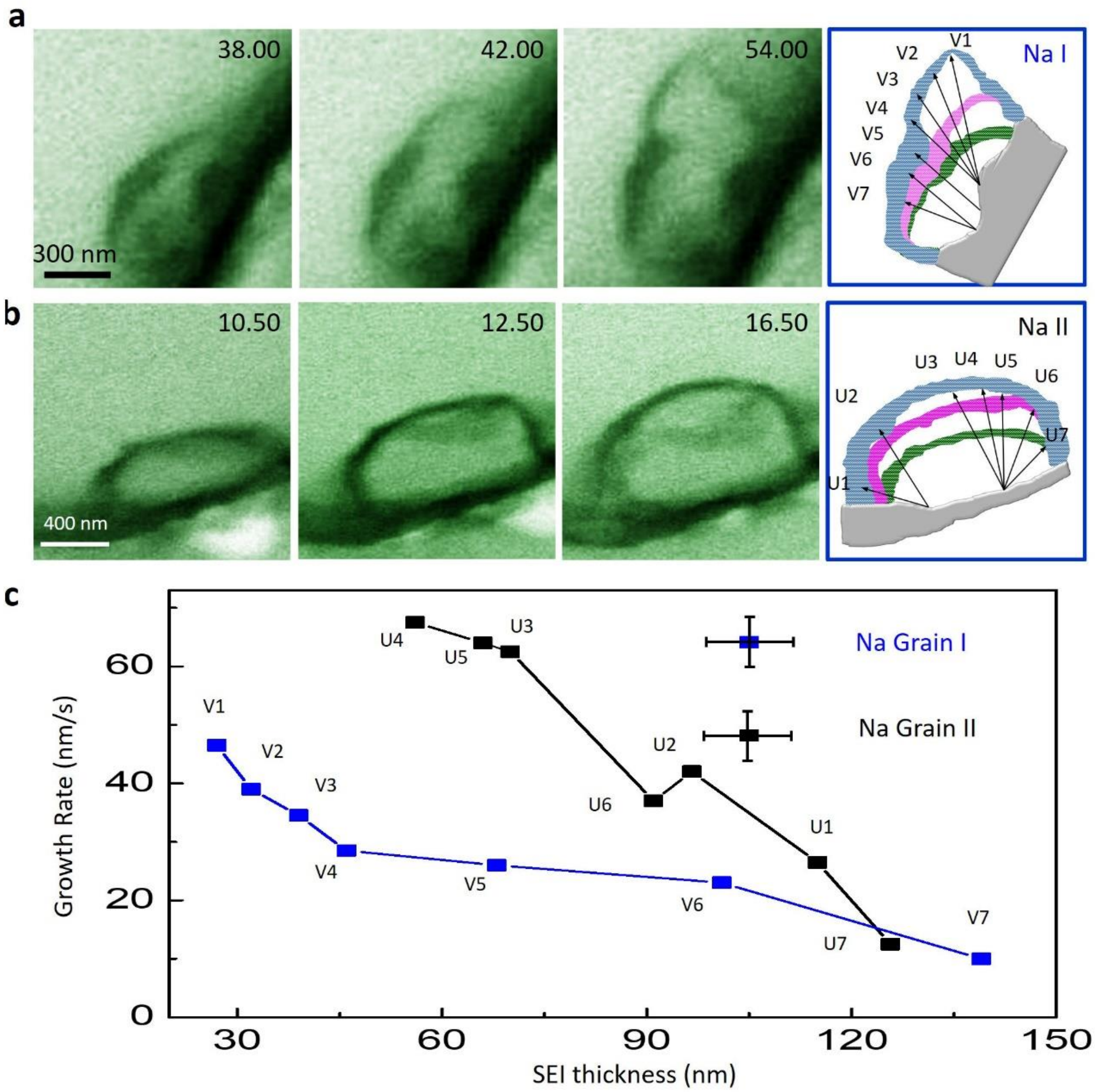
Supplementary Figure 3 | Schematic illustrations of the lithography setup and mask. a | Lithography setup. **b |** Top view of mask with 1096 cell units. **c |** Top view of one cell unit in the mask used for the 1st lithography patterning of top chips. **d |** Top view of one cell unit in the mask used for the 2nd lithography patterning of top chips. **e |** Top view of one cell unit in the mask used for the 1st lithography patterning of bottom chips. **f |** Top view of one cell unit in the mask used for the 2nd lithography patterning of bottom chips. Values in the figure are in microns. Of note, ensure that the window (289 × 308 μm, in panel **c** and **e**) is in the middle of the mask. Ensure that the paired patterns (in panel **c** and **f**) are symmetrical to the left and right along the center line. For the detailed information about lithography setup, the reader is referred to the Chapter 4.3 of the following link: (https://nanolab.berkeley.edu/public/manuals/equipment_manual.shtml).



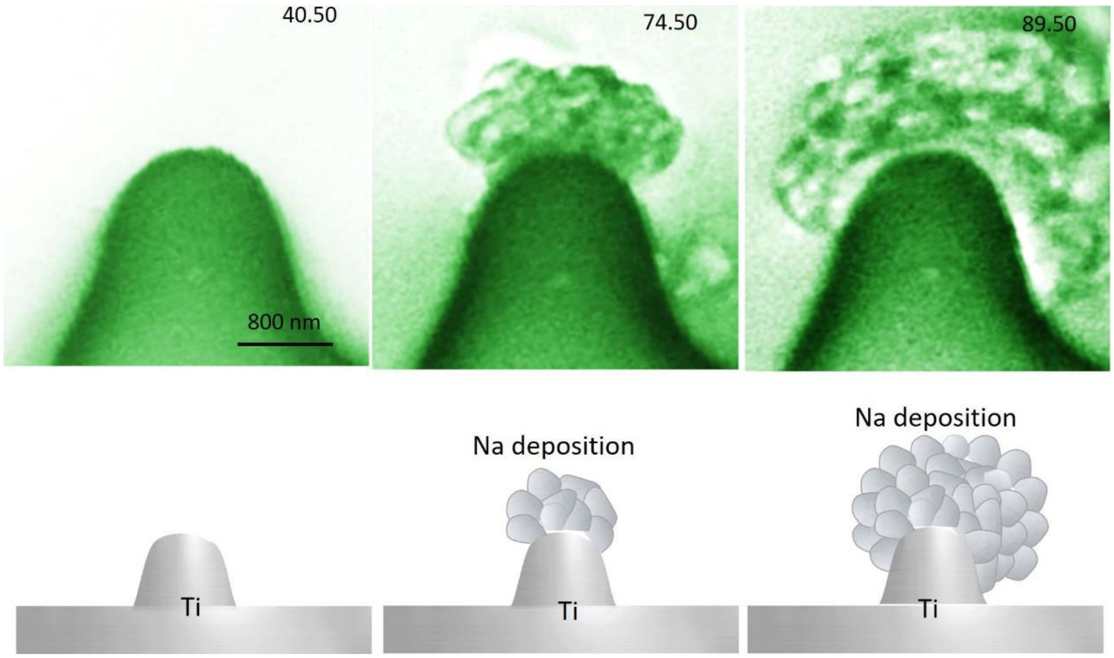
Supplementary Figure 4 | Thickness measurement of SiN_x windows. The thickness of the grown SiN_x is critical, and needs to be kept at about 35 nm or less. We used the Nanospec film thickness measurement system to check the thickness of SiN_x window (as described in step 5). Taking one silicon wafer in the process of our protocol as an example, we selected five points at different positions (A-E) on it for testing. The experimental data (A: 33.1 nm, B: 32.6 nm, C: 33.0 nm, D: 32.8 nm, E: 32.3 nm) show that, the SiN_x thicknesses at different positions on the silicon wafer are all less than 35 nm, which is thinner than the commercial SiN_x windows thickness (50 nm).



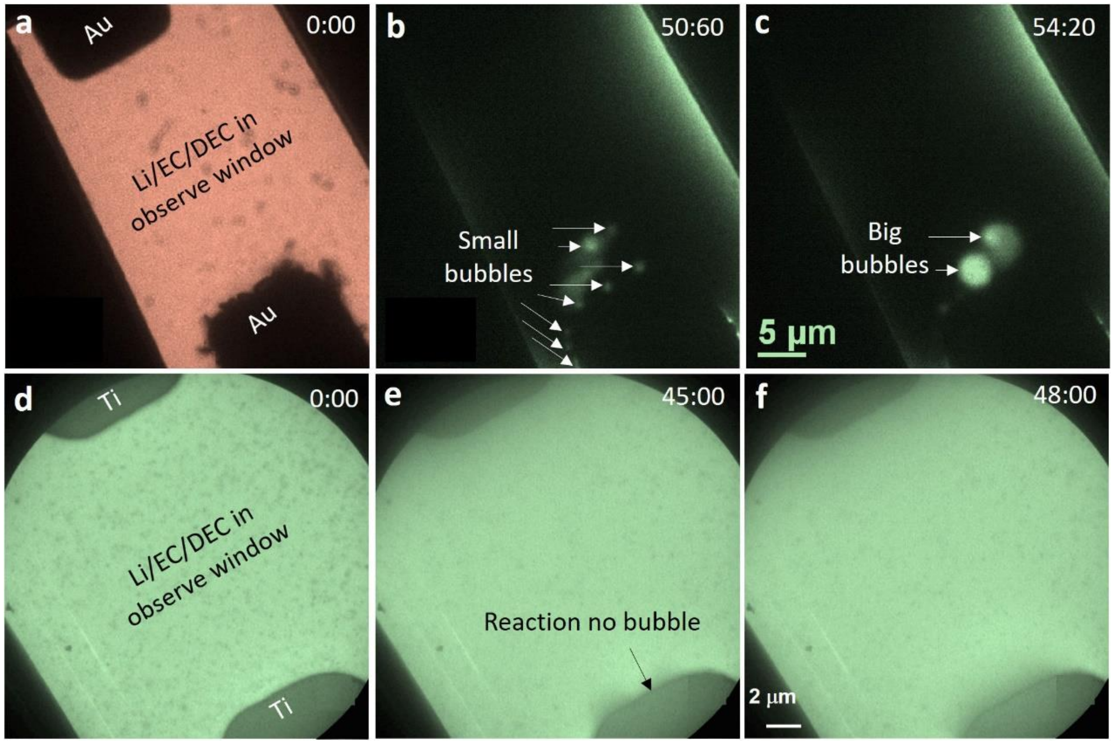
Supplementary Figure 5 | Photo of the custom TEM holder. The custom TEM holder provides a platform for fixing liquid cell and connecting the TEM equipment. There is a cell pocket at the TEM holder tip for placing liquid cell. Adapted with permission from ref.[3]. Royal Society of Chemistry.



Supplementary Figure 6 | The SEI thickness vs electrodeposition rate. a, b | Sequential images from **Supplementary Video 7** (left) show the growth of a Na grain and the corresponding SEI layer (right). The SEI layer is defined by the dark layer on the Na grain surface. Variations of SEI thickness are observed. The local SEI thickness is obtained by drawing a line through the SEI layer at different locations and measuring the distance between two edges of SEI with half decrease of the contrast value. **c |** The growth rate of two Na grains as a function of SEI layer thickness. Adapted with permission from ref.[4]. Elsevier.



Supplementary Figure 7 | In-situ TEM observation of Na electrodeposition on undulant Ti surface using electrochemical liquid cell III (Ti electrode, commercial $NaPF_6$/PC electrolyte). Top: sequential TEM images from **Supplementary Video 8**. Bottom: schematic illustration of Na electrodeposition on undulant Ti surface. The TEM images are adapted with permission from ref.[4]. Elsevier.



Supplementary Figure 8 | Sequential images show the electrode-electrolyte interface reaction on both sides of the electrodes in two electrochemical cells. a-c | Au electrodes. **d-f |** Ti electrodes. Note that Au electrode has bubbles and the Ti doesn't. Adapted with permission from ref.[3]. Royal Society of Chemistry.

Supplementary References

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