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Supporting information

Fluoroalkyl Iodide Additive for Li–O_2 Battery Electrolytes Enables Stable Cycle Life and High Reversibility

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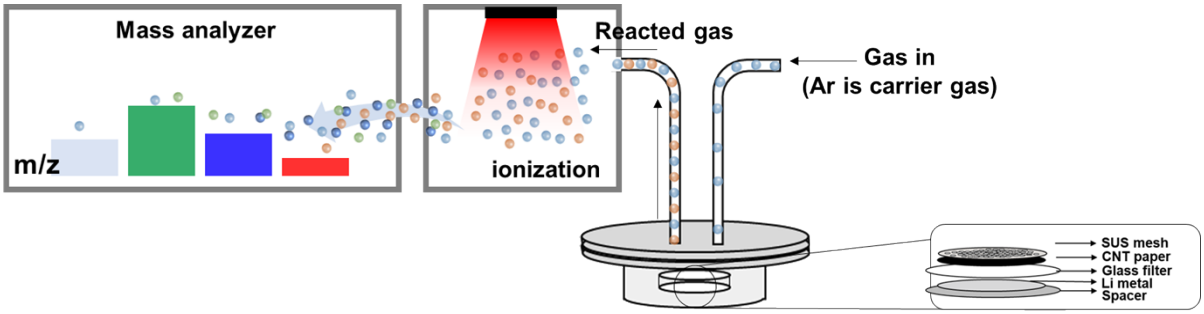

Fig. S1. A schematic illustration representing the setup of the DEMS and the structure of a custom designed HS air type cell.

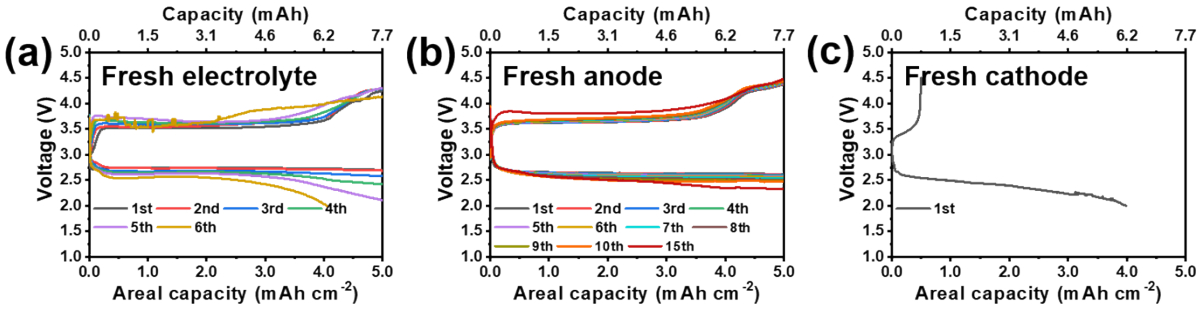

Fig. S2. Discharge–charge curves of rebuild Li–O_2 cells using (a) fresh electrolyte, (b) fresh anode, and (c) fresh cathode.

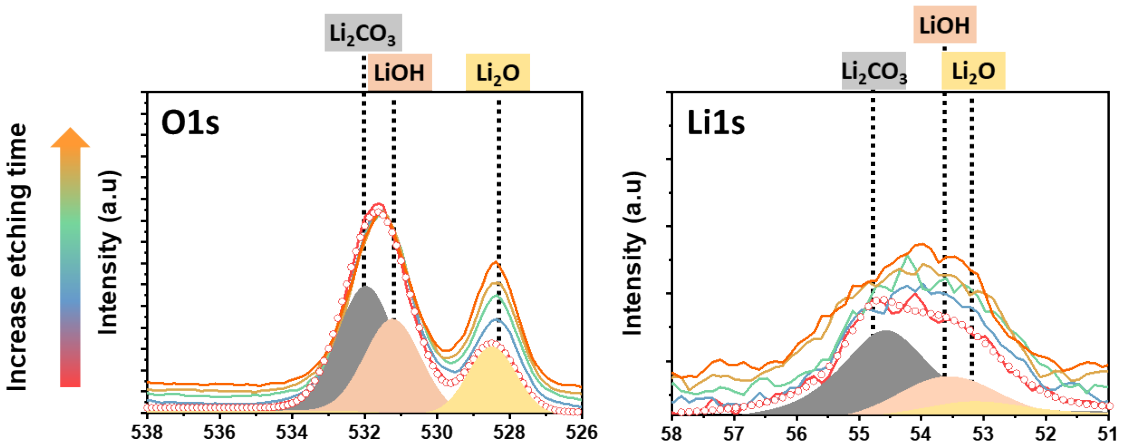

Fig. S3 XPS depth profiles of Li metal after 25 cycles in an Li–O_2 cell using 1 M $LiNO_3$/N,N-dimethylacetamide electrolyte.

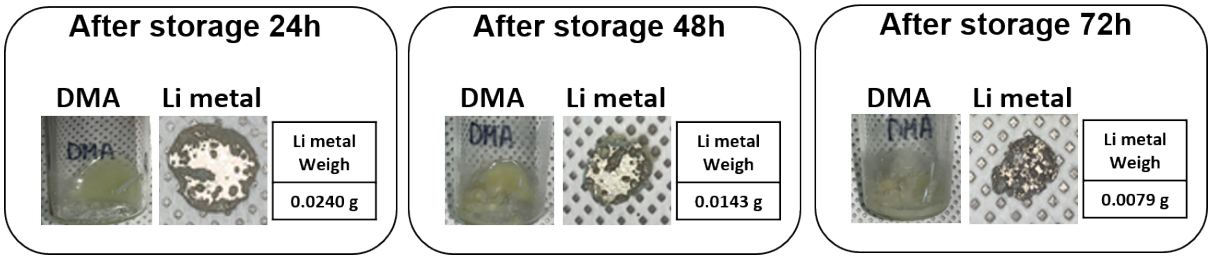

Fig. S4 Digital photos of N,N-dimethylacetamide (DMA) and Li and the obtained Li metal weight after storage of Li metal in DMA for different time.

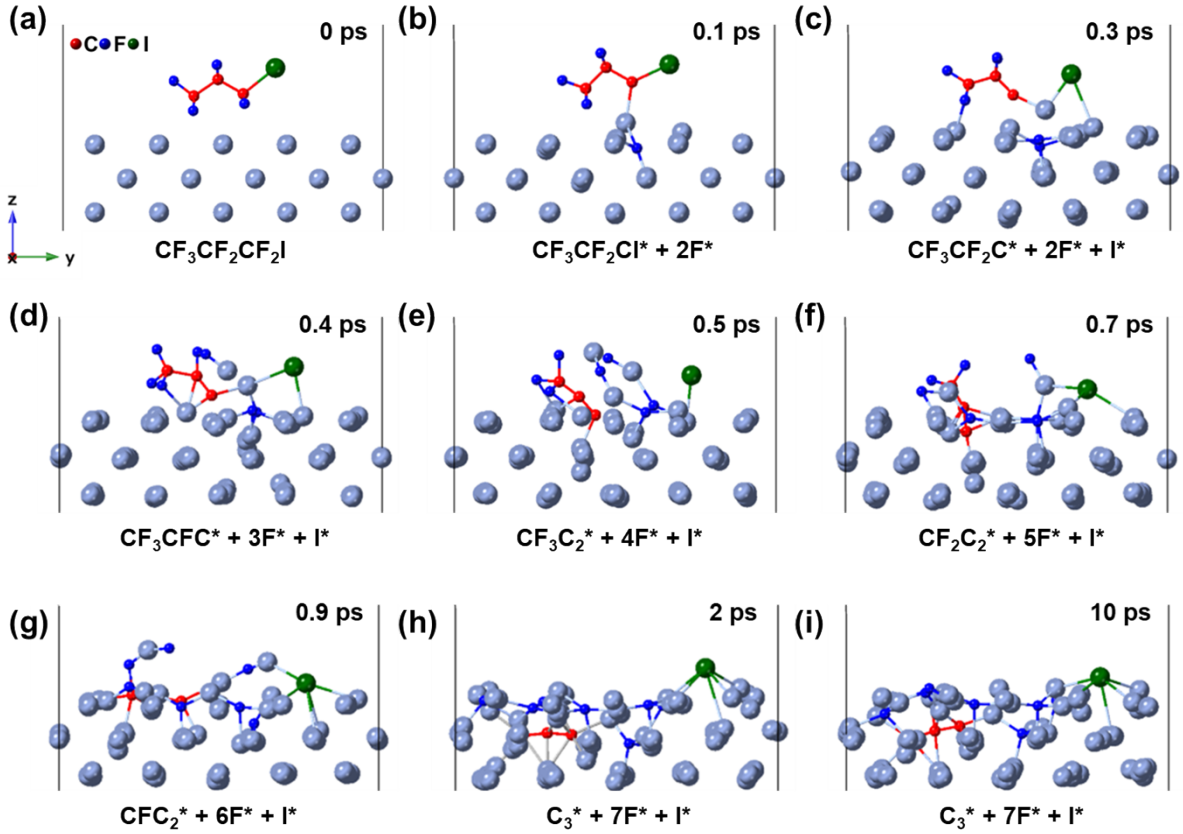

Fig. S5 *Ab initio* molecular dynamics calculations at 300 K of the dissociation of $CF_3(CF_2)_2I$ on the Li(100) surface.

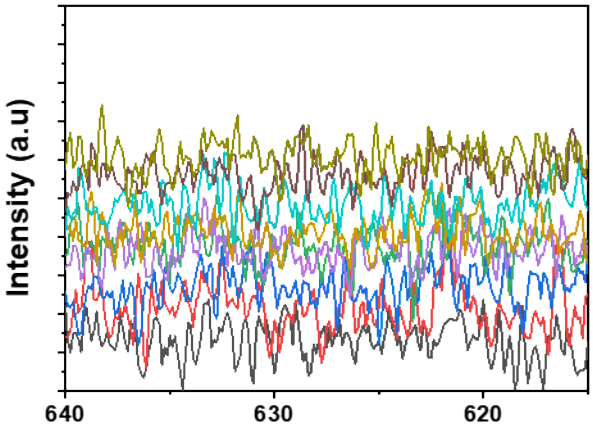

Fig. S6 XPS depth profiles of I 3d spectra of Li metal after storage in $CF_3(CF_2)_2I$-containing electrolyte.

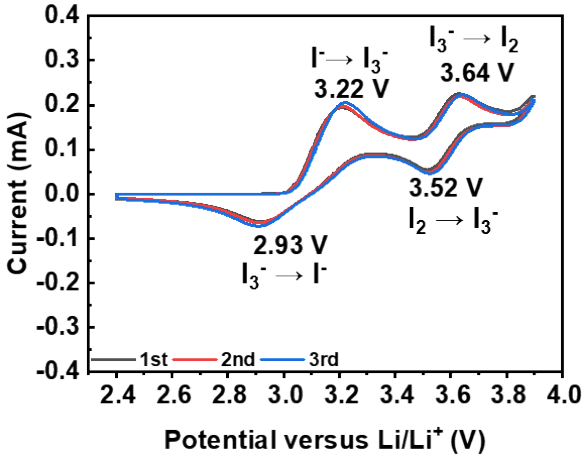

Fig. S7 Three-electrode CV curves for an electrolyte composed of 0.2 M LiI + 1 M $LiNO_3$/N,N-dimethylacetamide.

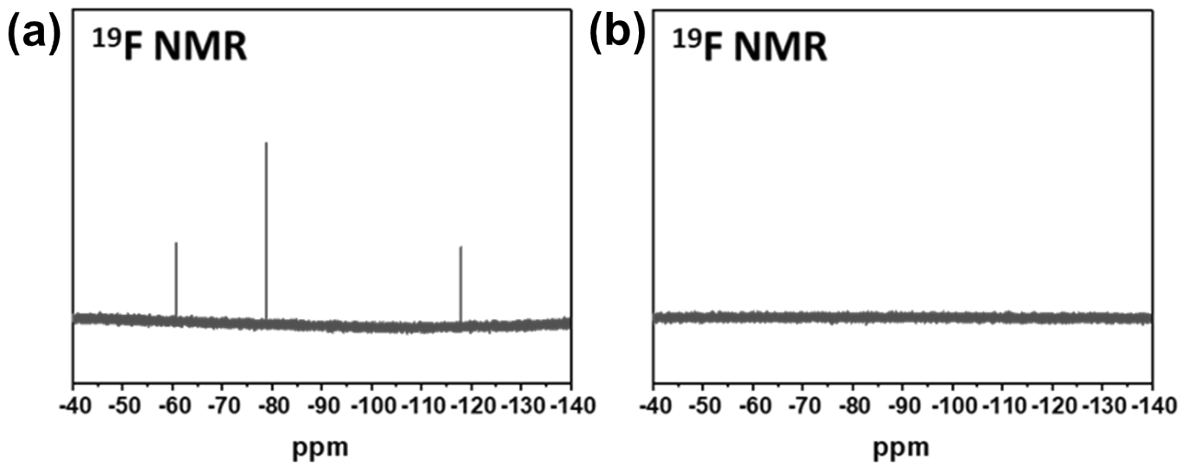

Fig. S8. The ^{19}F NMR spectrum of 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/DMA (a) before and (b) after reaction with Li metal.

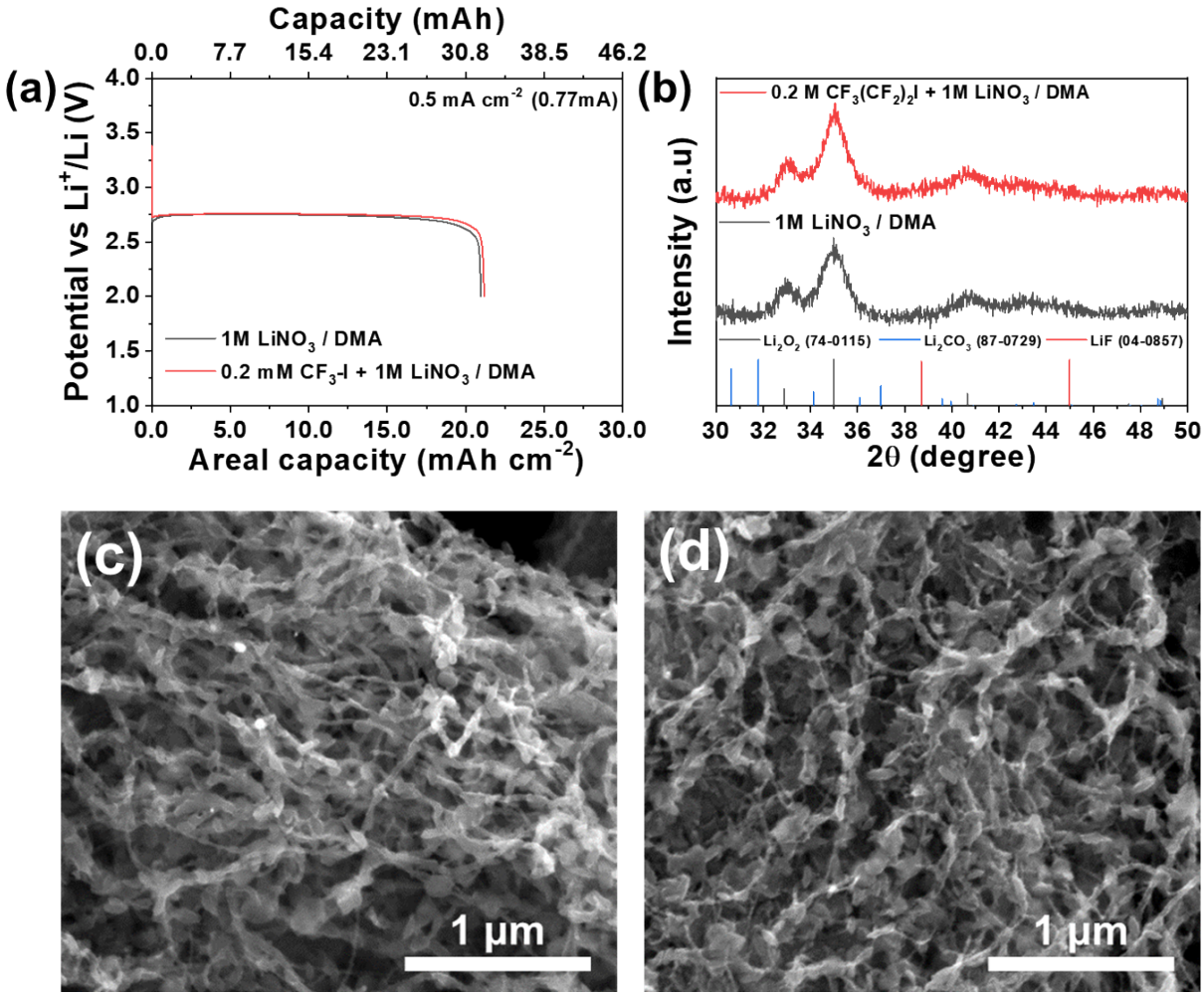

Fig. S9. (a) Discharge–charge curves of Li–O_2 cell using 1 M $LiNO_3$/N,N-dimethylacetamide (DMA) (black) and 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$ in DMA (red) at a current density of 0.5 mA cm^{-2} until 2.0 V. (b) XRD patterns of the discharged cathode in 1 M $LiNO_3$/DMA and 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/DMA and (c), (d) the respective SEM images.

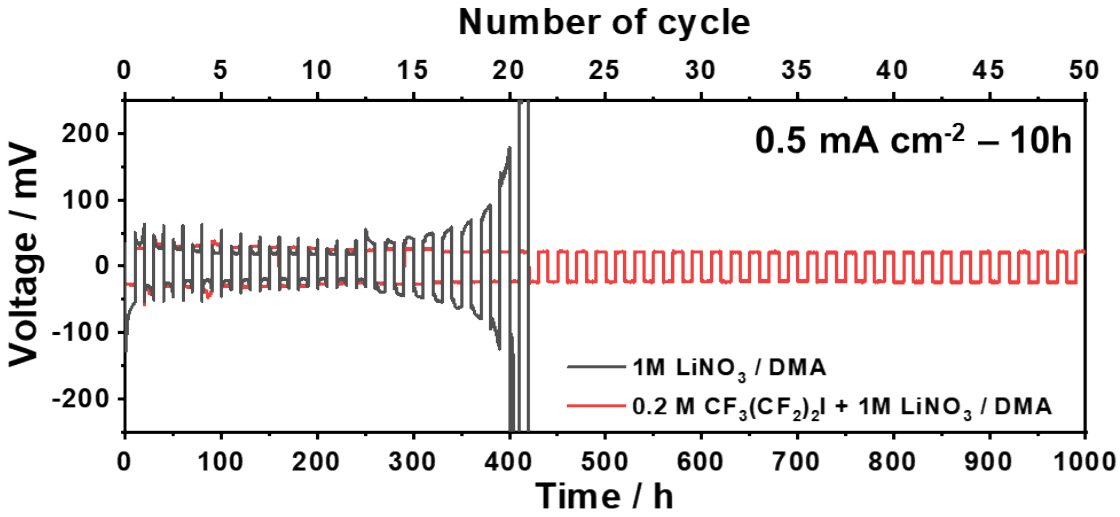

Fig. S10. Voltage profiles of a symmetric Li/Li cell cycled in 1 M $LiNO_3$/N,N-dimethylacetamide (DMA) (black) and 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/DMA (red) at a current density of 0.5 mA cm^{-2} under O_2.

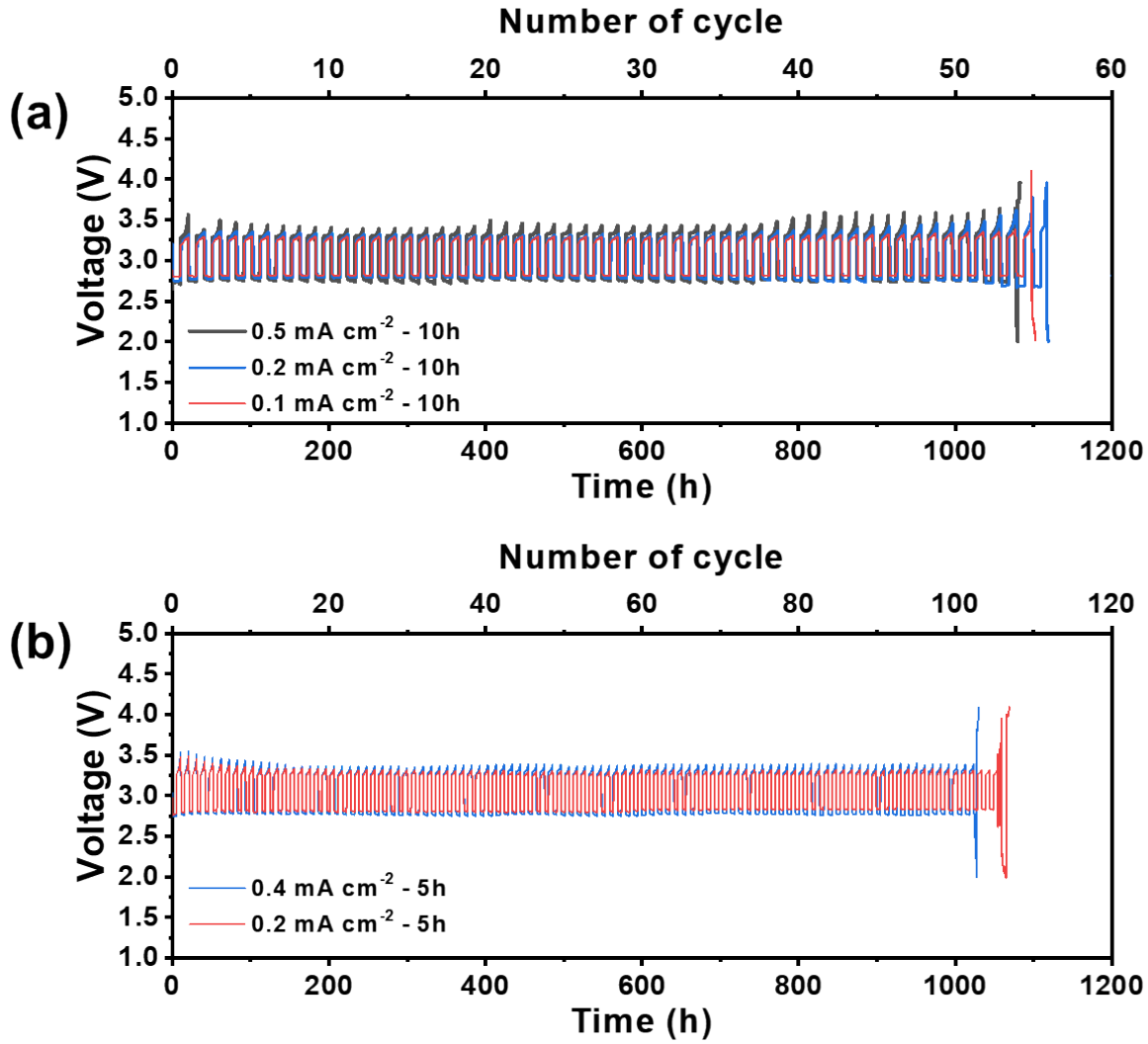

Fig. S11. Time–voltage profile of Li–O_2 cell using 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/N,N-dimethylacetamide under various current densities with a time limit of (a) 10 h and (b) 5 h.

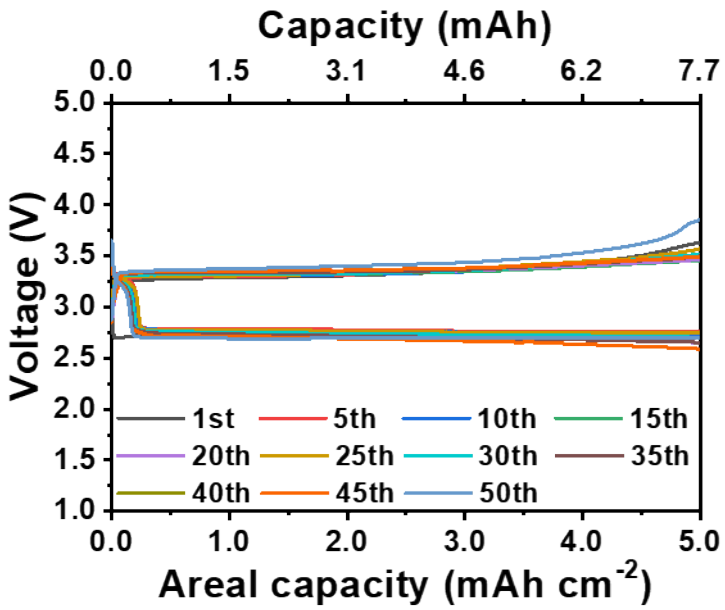

Fig. S12. Discharge–charge curves of Li–O_2 cell rebuild using fresh 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/N,N-dimethylacetamide electrolyte.

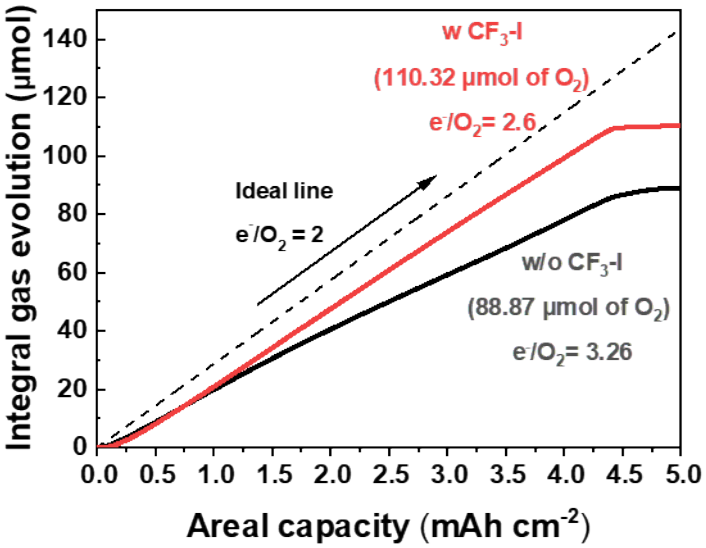

Fig. S13. Integral gas evolution for O_2 determined by in situ differential electrochemical mass spectroscopy measurements using 1 M $LiNO_3$/N,N-dimethylacetamide (DMA) (black) and 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/DMA (red) during the charging process. The dashed line indicates the ideal oxidation of Li_2O_2 with an e^-/O^2 value of 2.

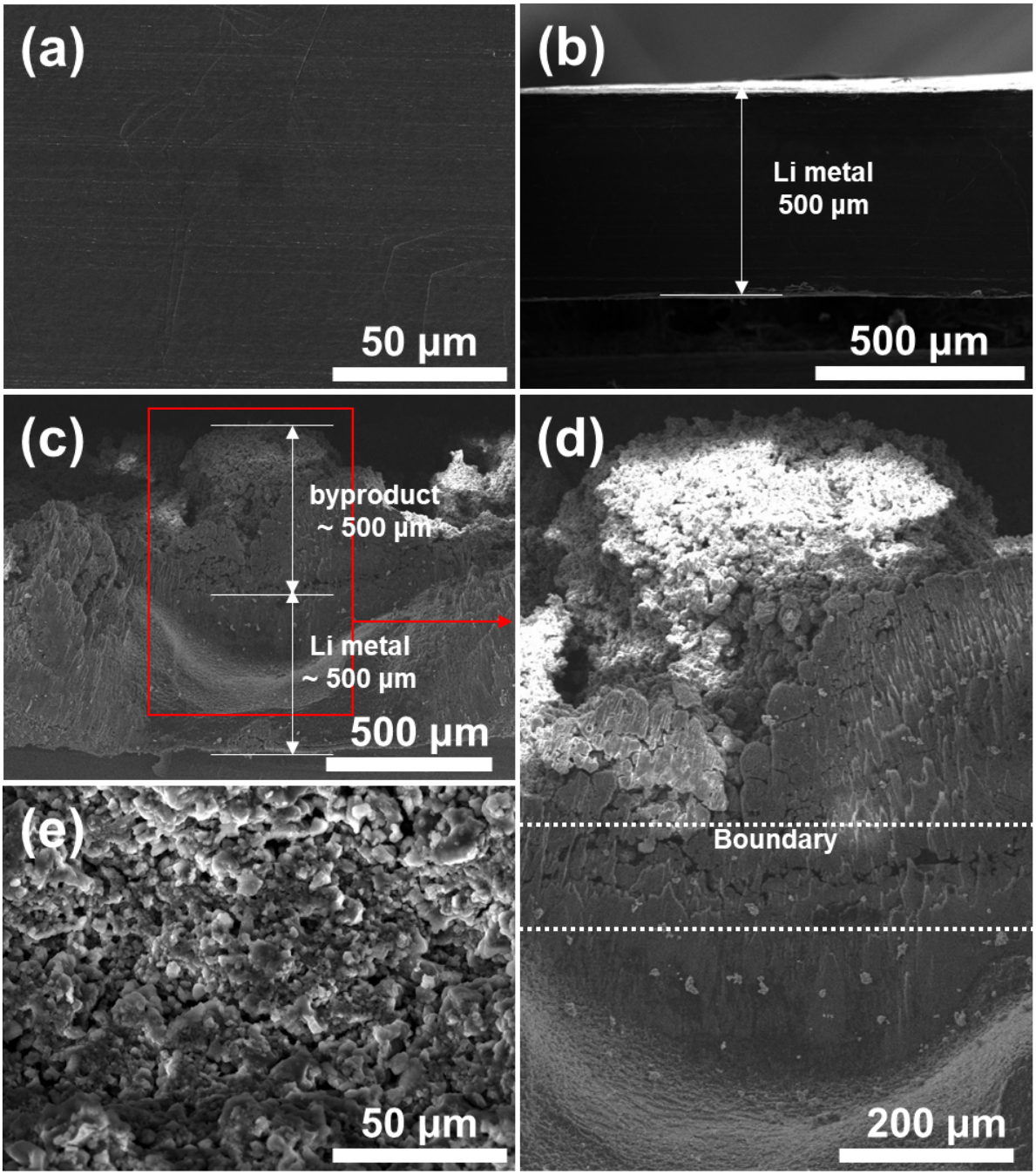

Fig. S14. SEM images of (a) surface and (b) a cross-section of pristine Li metal. SEM images of (c) a cross-section and (e) surface of the Li metal after 25 cycles in an Li–O_2 cell using 1 M $LiNO_3$/N,N-dimethylacetamide electrolyte. (d) High-magnification SEM image of (c).

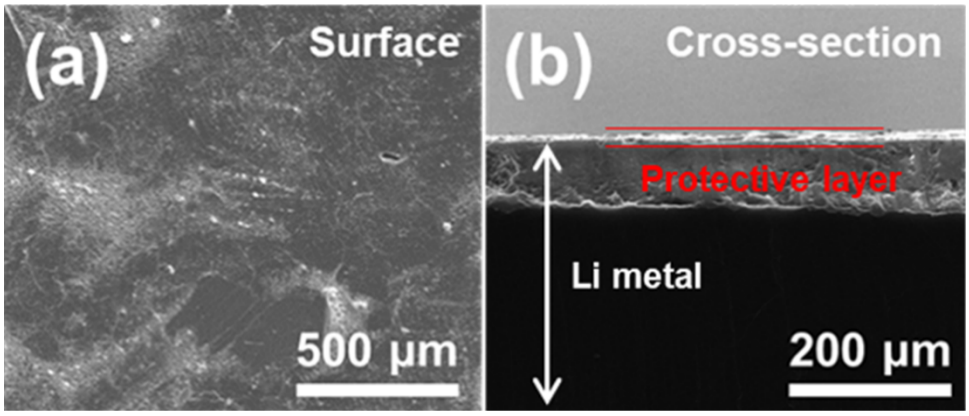

Fig. S15. SEM images of (a) surface and (b) a cross-section of Li metal after discharging to a capacity of 5 mA h cm^{-2} using 0.2 M $CF_3(CF_2)_2I$ + 1 M $LiNO_3$/N,N-dimethylacetamide electrolyte.