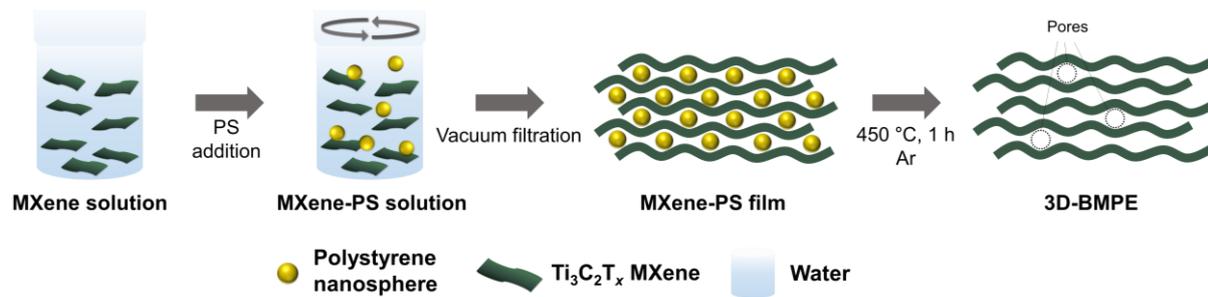


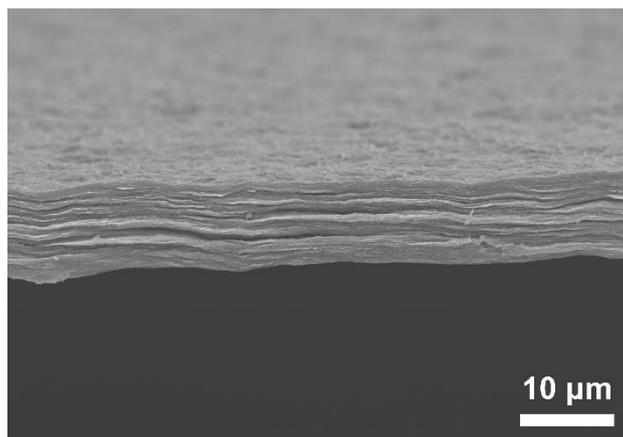
## Supporting information

3D-structured bifunctional MXene paper electrodes for protection and activation of Al metal anodes

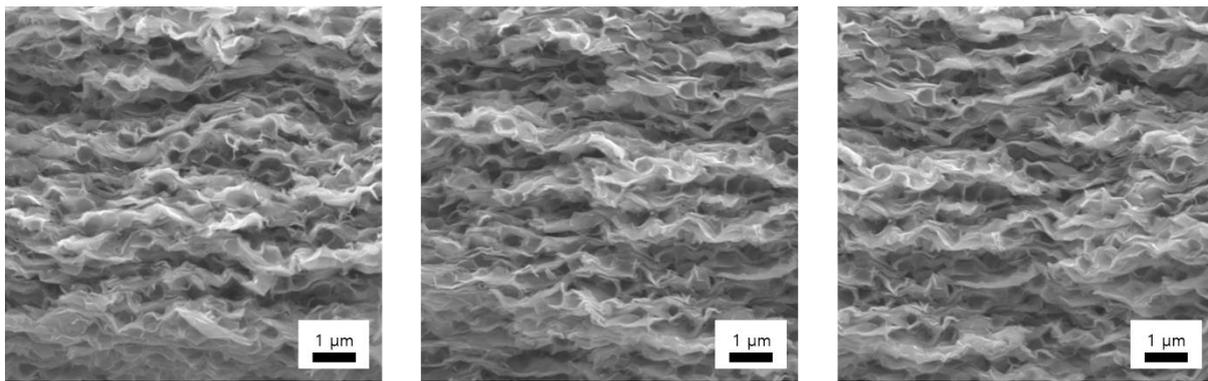
*Yeong Hoon Heo<sup>†</sup>, Juyun Lee<sup>†</sup>, Son Ha, Jong Chan Hyun, Dong Hyuk Kang, Juhee Yoon, Hyun Soo Kim, Yeonhwa Choi, Yun Chan Kang, Hyoung-Joon Jin, Seon Joon Kim<sup>\*</sup>, and Young Soo Yun<sup>\*</sup>*



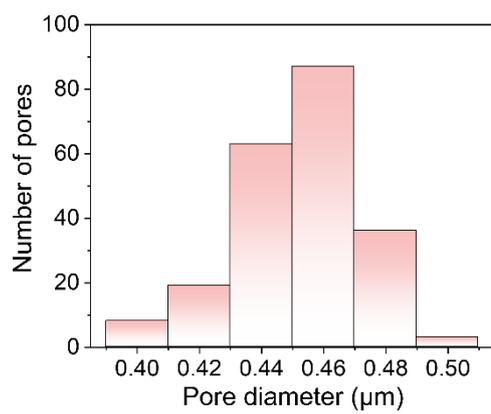
**Figure S1.** Schematic illustration of the synthesis process of 3D-BMPE.



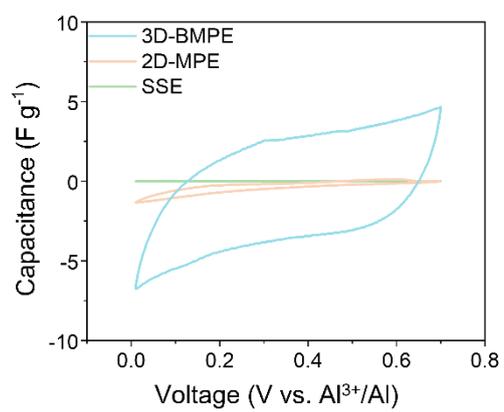
**Figure S2.** Cross-sectional FE-SEM image of 2D-MPE.



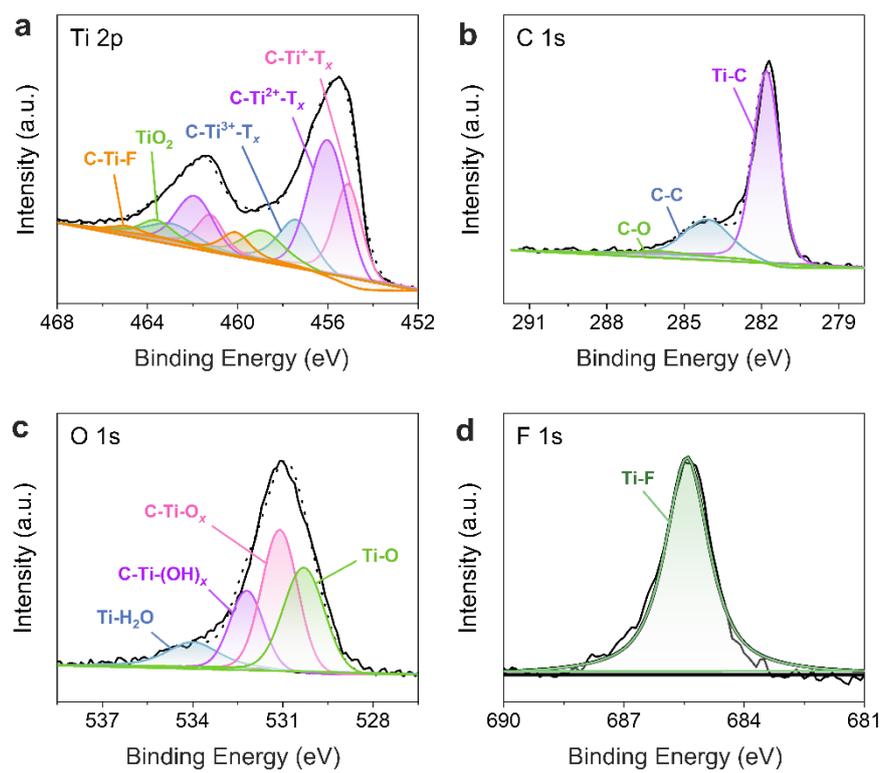
**Figure S3.** Cross-sectional images of 3D-BMPE across various regions in the film.



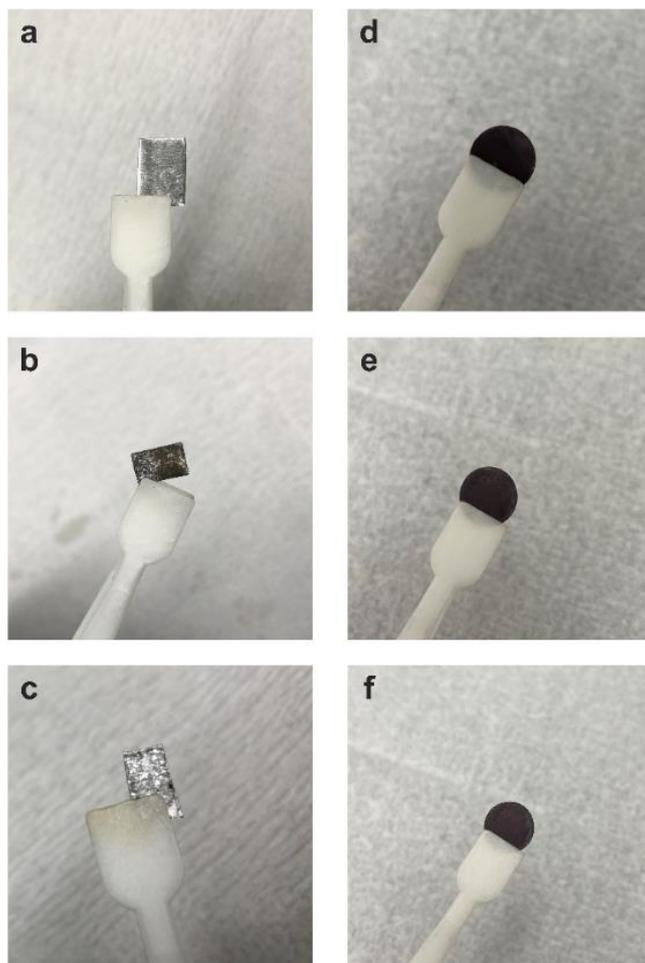
**Figure S4.** Macropore size distribution data of 3D-BMPE characterized from several FE-SEM images.



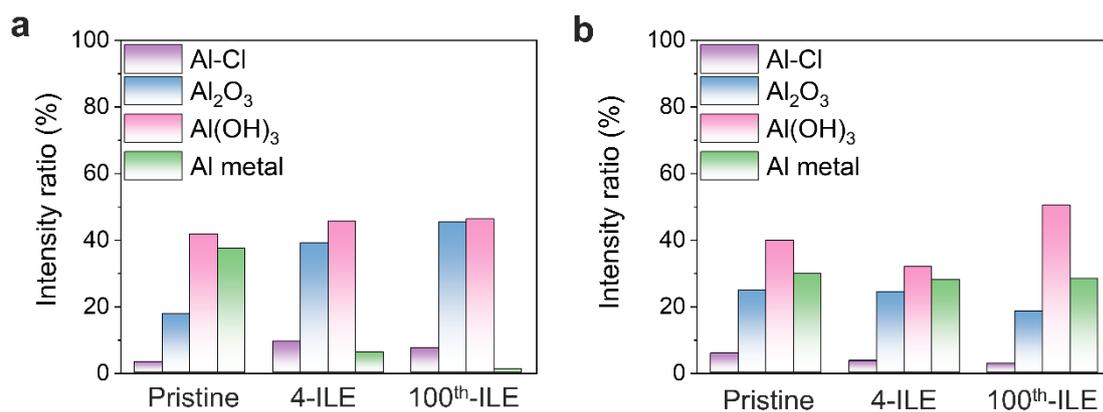
**Figure S5.** Cyclic voltammograms of SSE, 2D-MPE, and 3D-BMPE at a sweep rate of 1 mV s<sup>-1</sup> in ILE over a voltage window of 0.01–0.7 V vs. Al<sup>3+</sup>/Al.



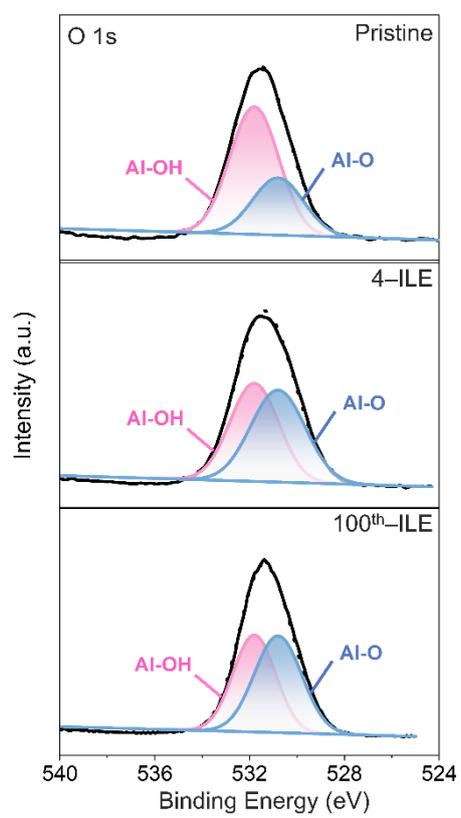
**Figure S6.** Deconvoluted XPS spectra of 2D-MPE: (a) Ti 2p, (b) C 1s, (c) O 1s, and (d) F 1s.



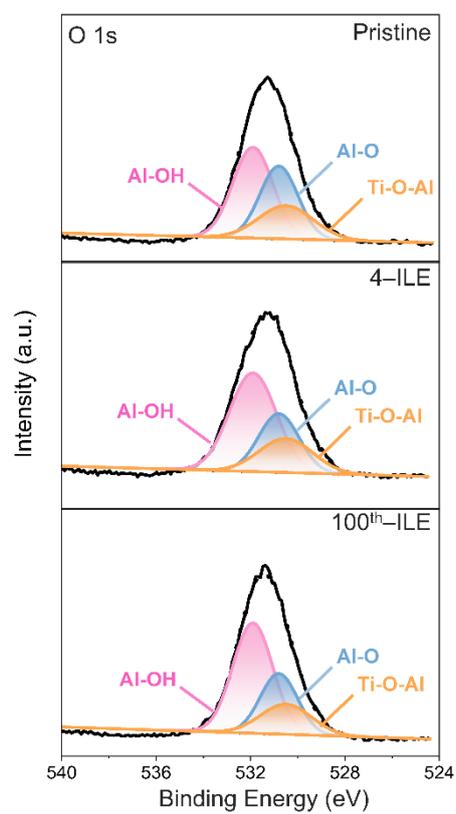
**Figure S7.** Optical images of Al foil (a) at initial state, (b) after immersed in the ILE for four weeks, and (c) after 100<sup>th</sup> galvanostatic aluminum deposition/dissolution cycle. Optical images of 3D-BMPE including Al metal of 3 mA h cm<sup>-2</sup> (d) at initial state, (e) after immersed in ILE for four weeks, and (f) after 100<sup>th</sup> galvanostatic aluminum deposition/dissolution cycle.



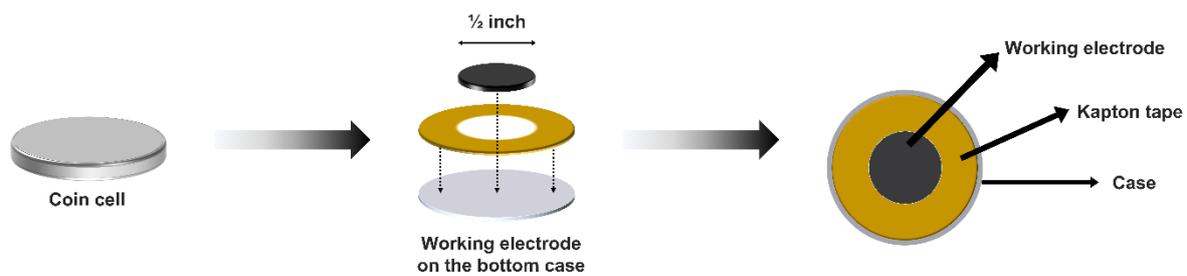
**Figure S8.** Bar graphs of deconvoluted XPS Al 2p spectra of (a) Al foil and (b) 3D-BMPE including Al metal of 3 mA h cm<sup>-2</sup> at different experimental conditions.



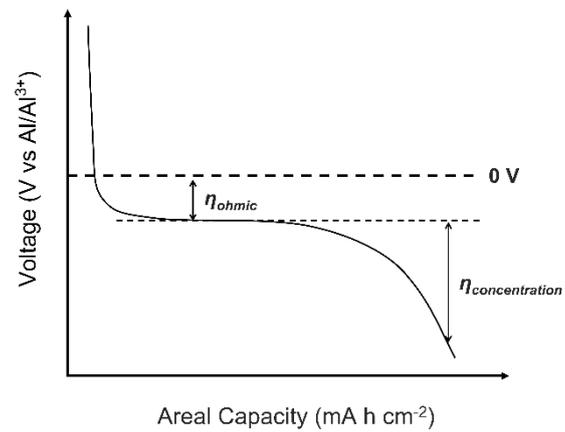
**Figure S9.** Deconvoluted XPS O 1s spectra of Al foil at different experimental conditions.



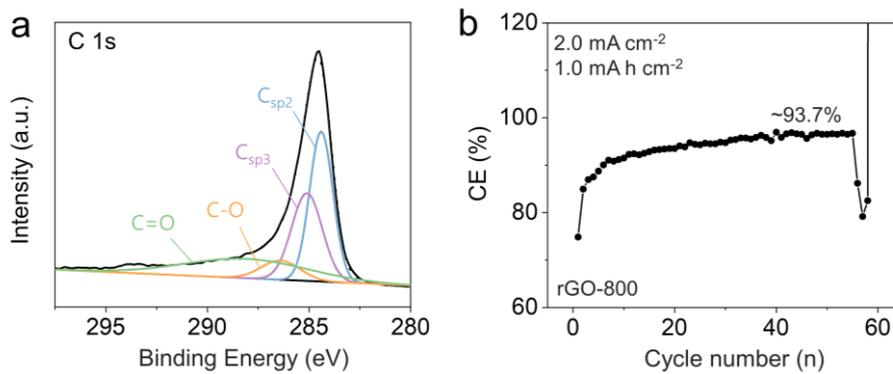
**Figure S10.** Deconvoluted XPS O 1s spectra of 3D-BMPE at different experimental conditions.



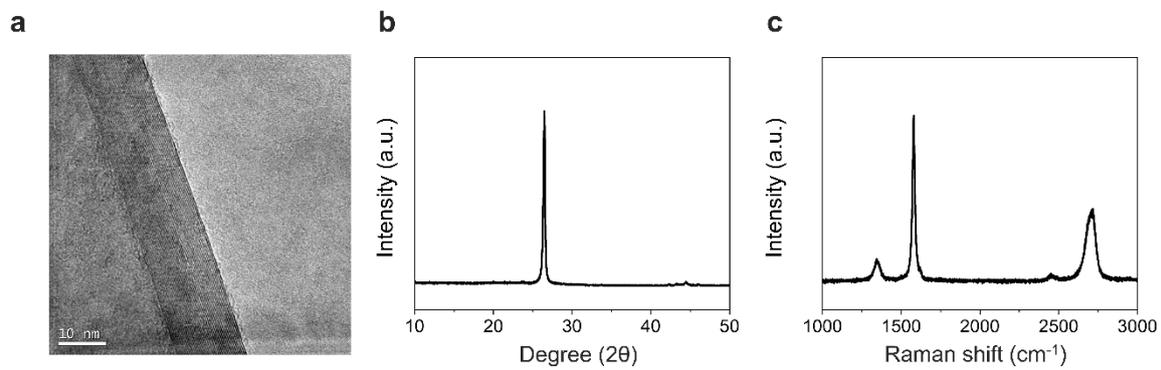
**Figure S11.** Schematic diagram of handmade coin cell coated with a polymer tape, except for the center hole of half-inch in diameter.



**Figure S12.** Schematic image showing ohmic and concentration overpotentials.



**Figure S13.** (a) Deconvoluted XPS C 1s spectrum and (b) cycling performance data of reduced graphene oxide-based AMA.



**Figure S14.** Material characterization of commercial graphite cathode: (a) TEM image, (b) XRD pattern, and (c) Raman spectrum.

**Table S1.** Comparison for electrochemical performances of the previously reported results.

| Electrode material | Current density (mA cm <sup>-2</sup> ) | $\eta_t$ (mV@mA cm <sup>-2</sup> ) | Cycle number (n) | C.E. (%) | Reference                                       |
|--------------------|--|------------------------------------|------------------|----------|---|
| 3D-BMPE            | 0.5 – 2.0                              | 180@2                              | 2000             | 99.95    | This work                                       |
| Carbon             | 1.6 – 4                                | 200@1.6                            | 350              | 99.8     | <i>Nat. Energy</i> , 2021, 6, 398               |
| Carbon             | 0.25 – 1                               | 120@0.5                            | 35               | 99.7     | <i>Carbon Energy</i> , 2022, 4, 155             |
| Carbon             | 1                                      | 200@1                              | 50               | 99.2     | <i>ChemElectroChem</i> , 2017, 4, 2345          |
| Al foil            | 1 – 5                                  | 300@3                              | 50               | -        | <i>Energy Stor. Mater.</i> 2021, 34, 194        |
| Cu-Al              | 0.5 – 3                                | 50@1.5                             | 500              | -        | <i>J. Electrochem. Soc.</i> 2019, 166, 15, 3539 |
| Au-SS              | 1 – 3                                  | 500@3                              | 150              | ~98      | <i>J. Mater. Chem. A</i> , 2020, 8, 23231       |