

# Supplementary Information

## Ice-Templated Free-Standing Reduced Graphene Oxide for Dendrite-Free Lithium Metal Batteries

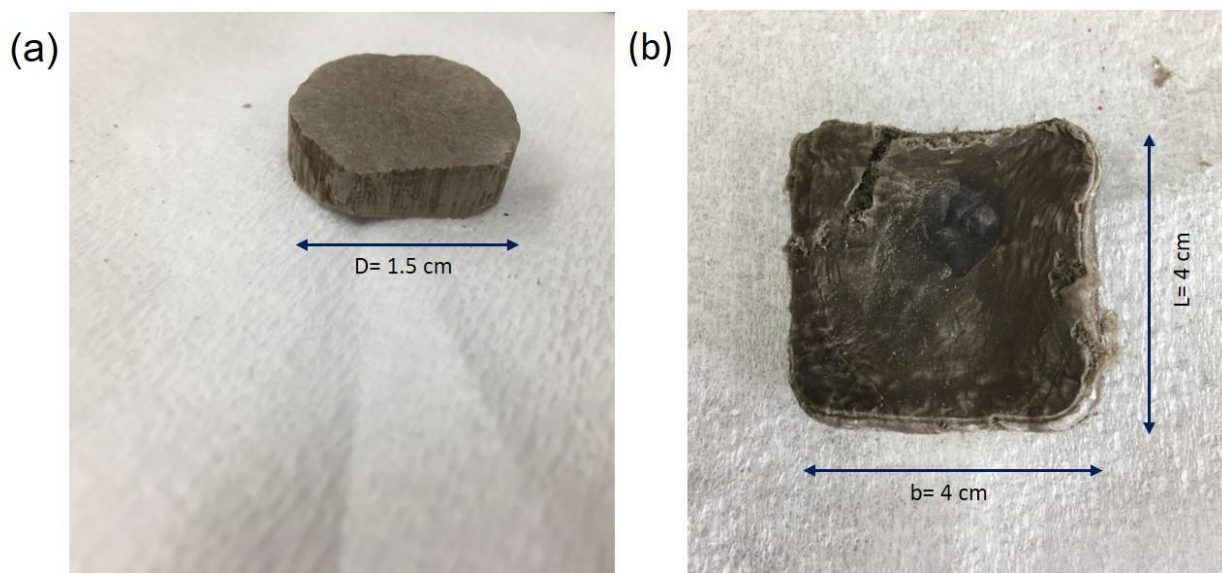
*Brindha Moorthy<sup>a</sup>, Rubha Ponraj<sup>a</sup>, Jong Hyuk Yun<sup>a</sup>, Ji Eun Wang<sup>b</sup>, Dong Jun Kim<sup>b\*</sup> and Do Kyung Kim<sup>a\*</sup>*

<sup>a</sup>Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Republic of Korea

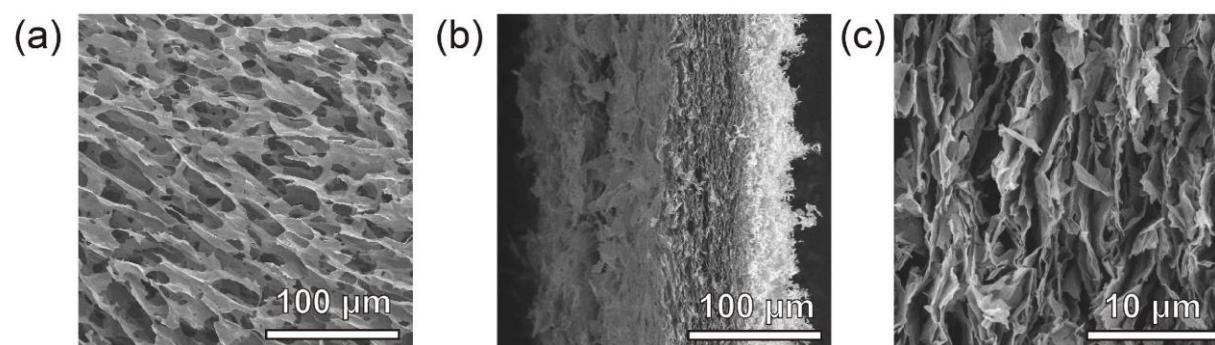
<sup>b</sup>School of Chemistry, University of New South Wales, Sydney, NSW 2052, Australia

KEYWORDS Lithium metal anode, ice-templating method, dendrite-free, free-standing rGO foam, lithium metal batteries

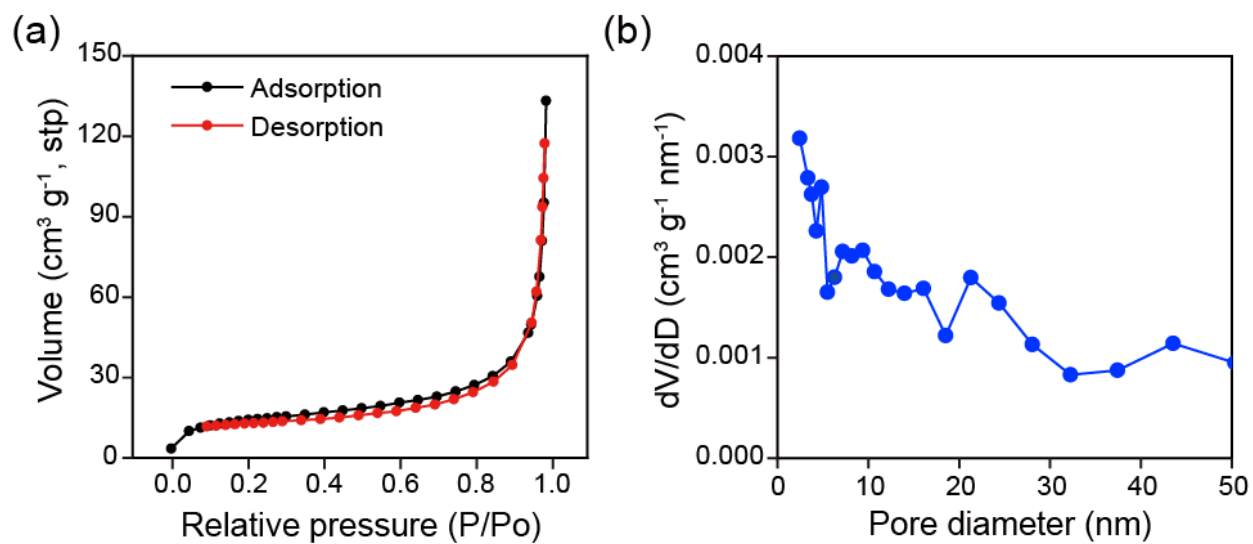
\*E-mail: *dongjun.kim@unsw.edu.au; dkkim@kaist.ac.kr*



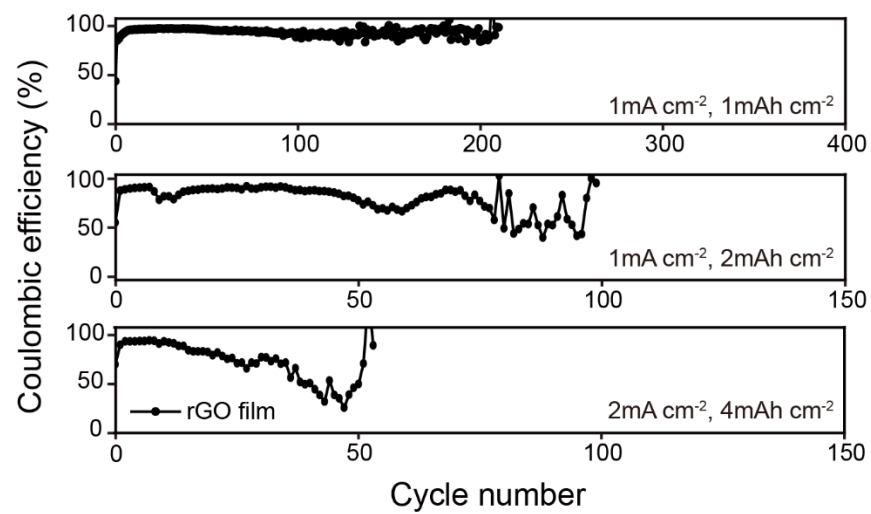
**Figure S1.** The optical images of two different sized free-standing Ice-rGF for (a) coin cell and (b) pouch cell format, whose dimension is controlled by the size and the shape of mold.



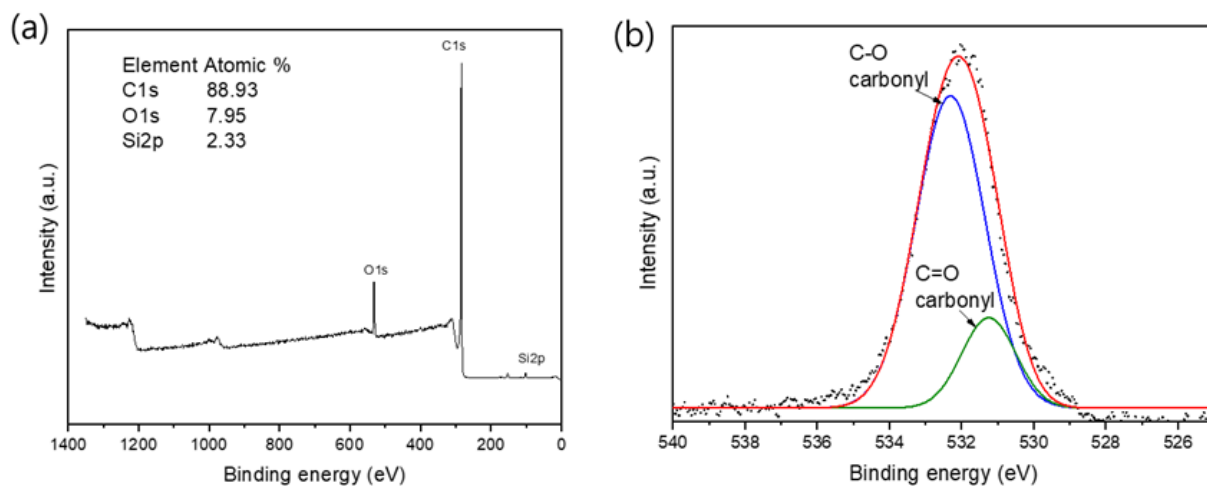
**Figure S2.** (a) Low magnification of the surface and (b-c) the cross-sectional SEM image of Ice-rGF.



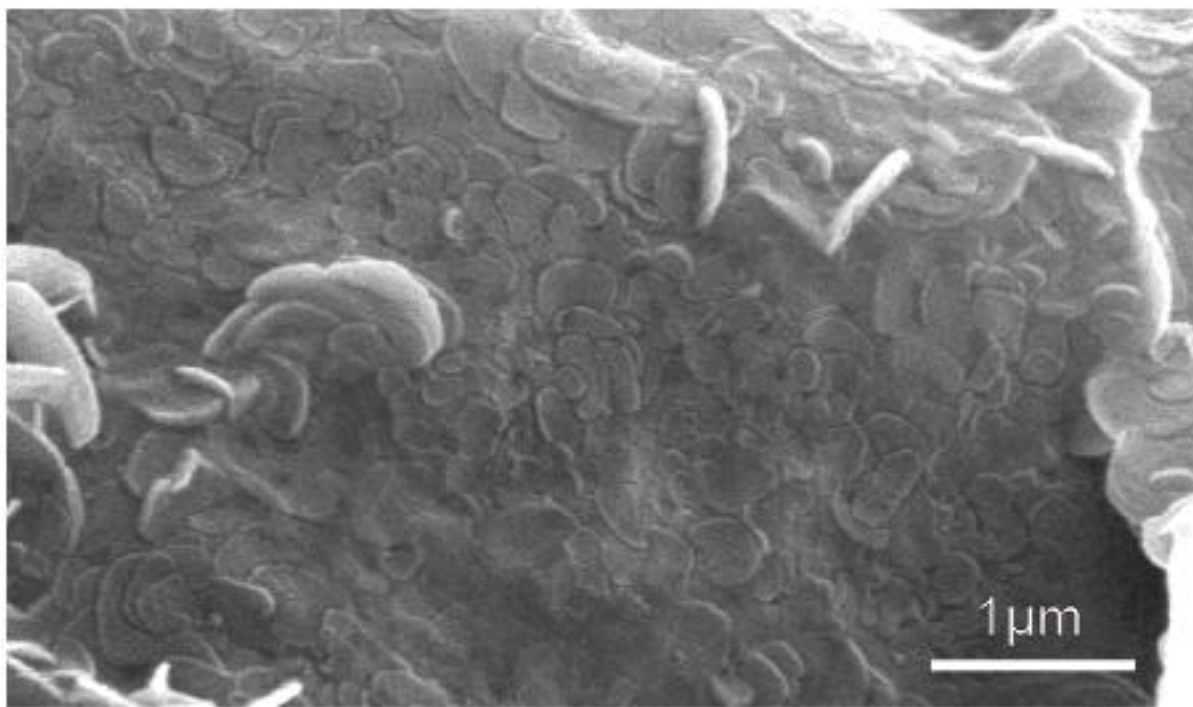
**Figure S3.** (a) Nitrogen adsorption-desorption isotherm result and (b) corresponding pore size distribution of pristine Ice-rGF.



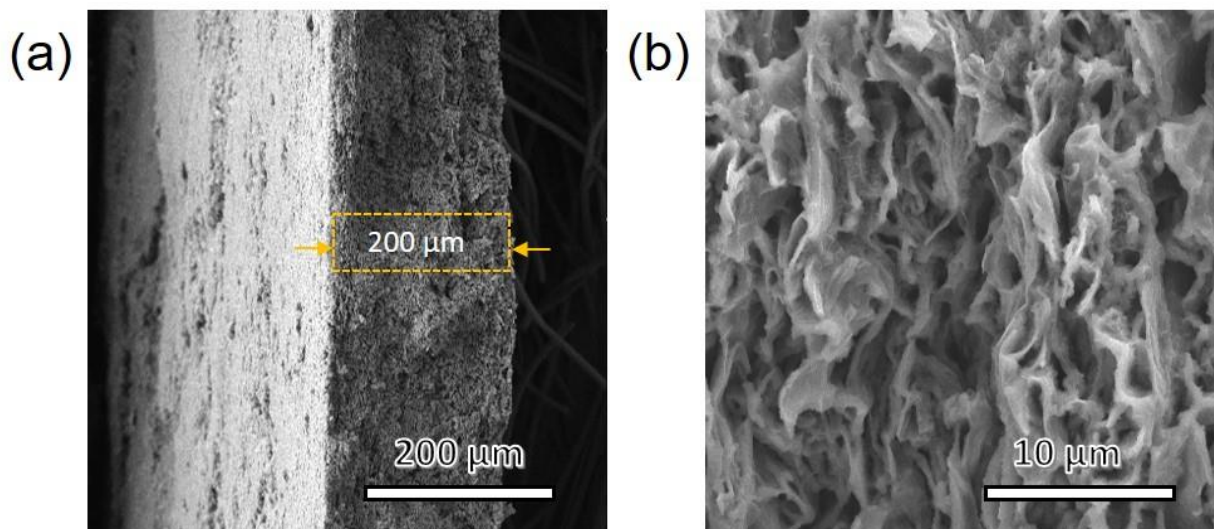
**Figure S4.** Coulombic efficiency of rGO film with various current densities and areal capacities.



**Figure S5.** XPS analysis of Ice-rGF (a) survey spectrum with the inset showing elemental analysis of Ice-rGF, and (b) De-convoluted O 1s XPS spectra of Ice-rGF. (A small impurity of Si 2p is from the substrate for XPS analysis)

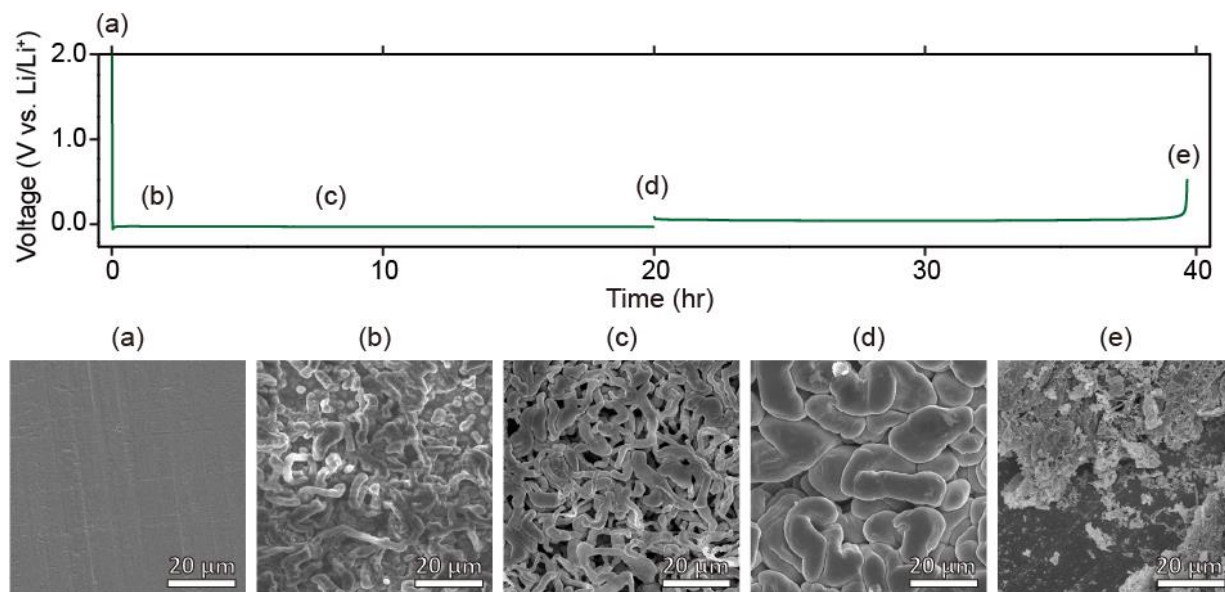


**Figure S6.** Higher magnification SEM image of Ice-rGF electrode after 4 mAh cm<sup>-2</sup>, corresponding to **Figure 4c**.

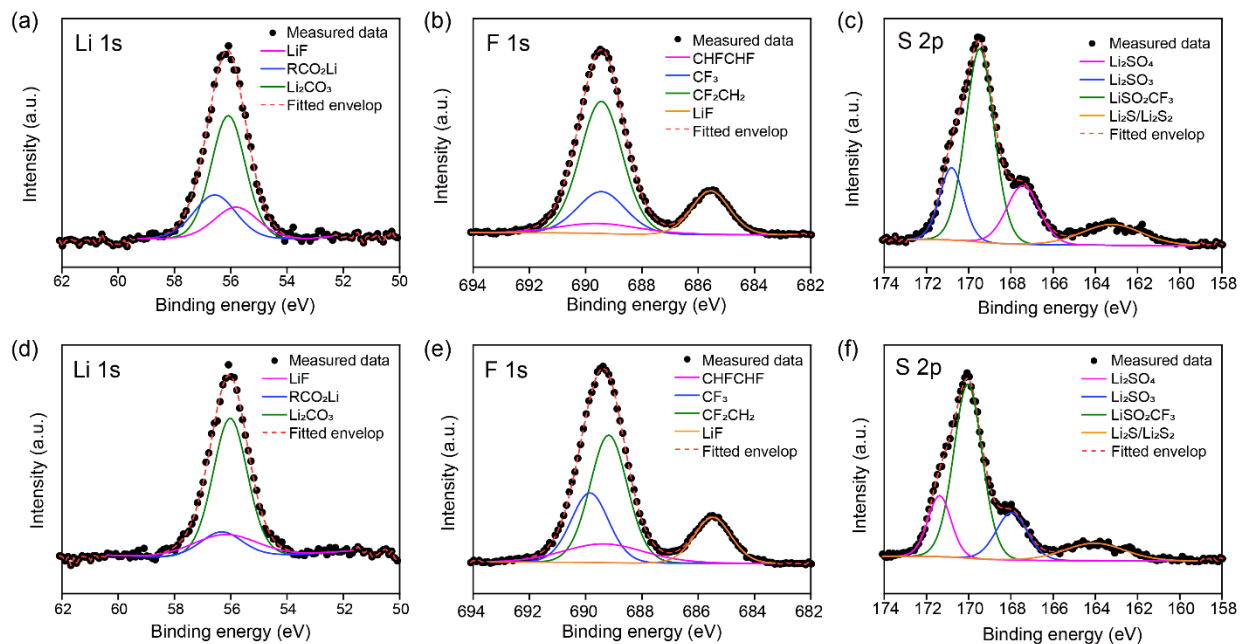


**Figure S7.** (a) SEM images of Ice-rGF at  $1\text{mA cm}^{-2}$  to  $1\text{mAh cm}^{-2}$  after 200 cycles (cross-sectional view), and (b) the high-magnification image.





**Figure S8.** (a) Voltage profile during first cycle of planar Cu at  $0.5 \text{ mA cm}^{-2}$  current density for  $10 \text{ mAh cm}^{-2}$ . SEM images of planar Cu electrode (a) at pristine condition, after deposition of Li for (b)  $1 \text{ mAh cm}^{-2}$ , (c)  $4 \text{ mAh cm}^{-2}$ , (d)  $10 \text{ mAh cm}^{-2}$ , and (e) after stripping of Li for  $9.7 \text{ mAh cm}^{-2}$ .



**Figure S9.** XPS chemical analysis of SEI composition of the electrode a) b) and c) Li 1s, F 1s and S 2p spectra respectively after 1st cycle and d), e), and f) Li 1s, F 1s and S 2p after 50th cycles respectively.