

Supporting Information.

Hierarchical Chitin Fibers with Aligned Nanofibrillar Architectures: A Nonwoven-Mat Separator for Lithium Metal Batteries

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1. Supporting Figures

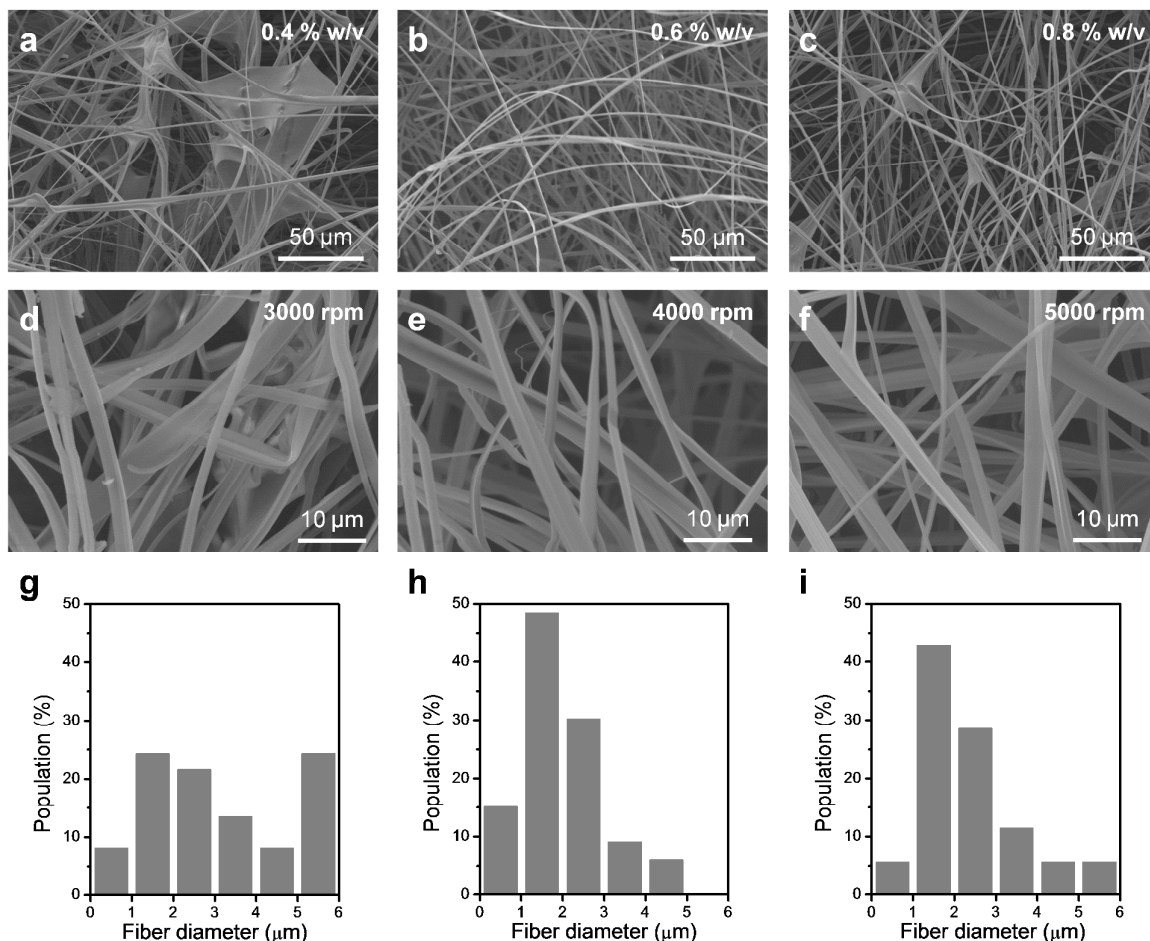


Figure S1. (a-c) SEM images of CJS-processed Chiber as a function of the concentration of spinning dope [(a) 0.4 w/v %, (b) 0.6 w/v % and (c) 0.8 w/v %] with a fixed rotation speed of 5000 rpm. (d-f) SEM images of CJS-processed Chiber as a function of rotation speed [(d) 3000 rpm, (e) 4000 rpm, and (f) 5000 rpm] with a fixed spinning dope concentration of 0.6 % w/v. (g-i) Histogram plots representing the diameter distribution of Chiber from 0.6 % w/v spinning dope according to rotation speed [(g) 3000 rpm, (h) 4000, and (i) 5000 rpm].

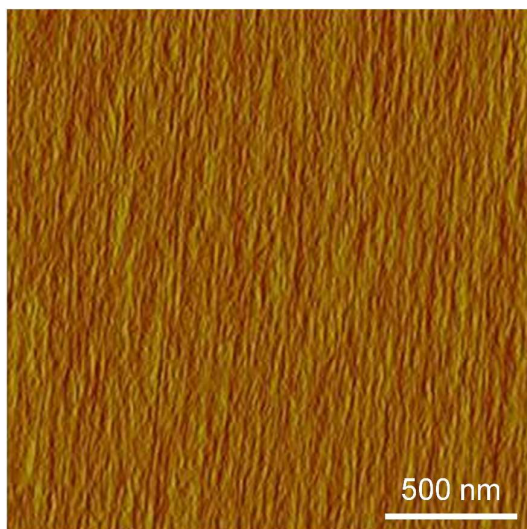


Figure S2. AFM phase image of single Chiber from manuscript Figure 1(f).

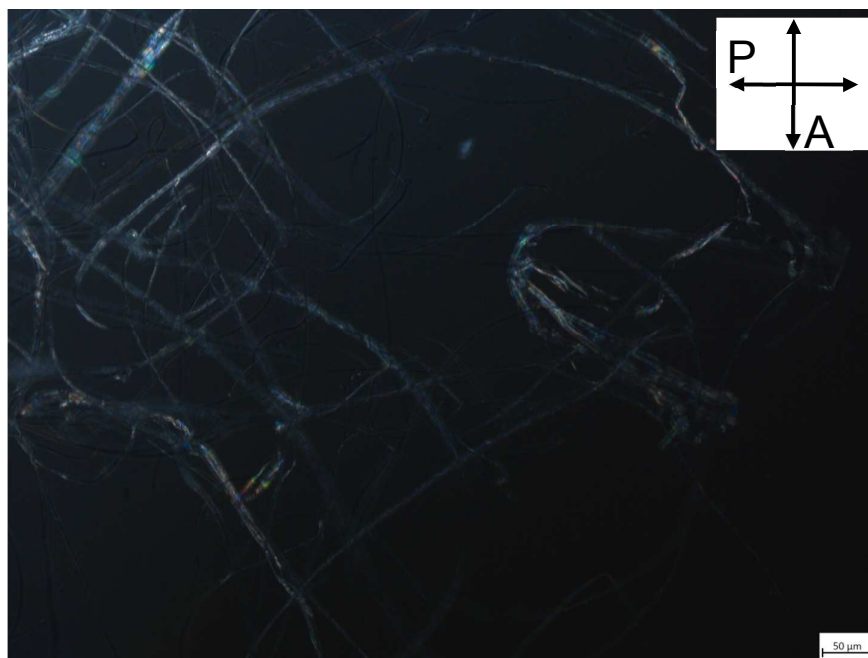


Figure S3. A polarization optical microscopy (POM) image of Chiber, showing birefringence clearly.

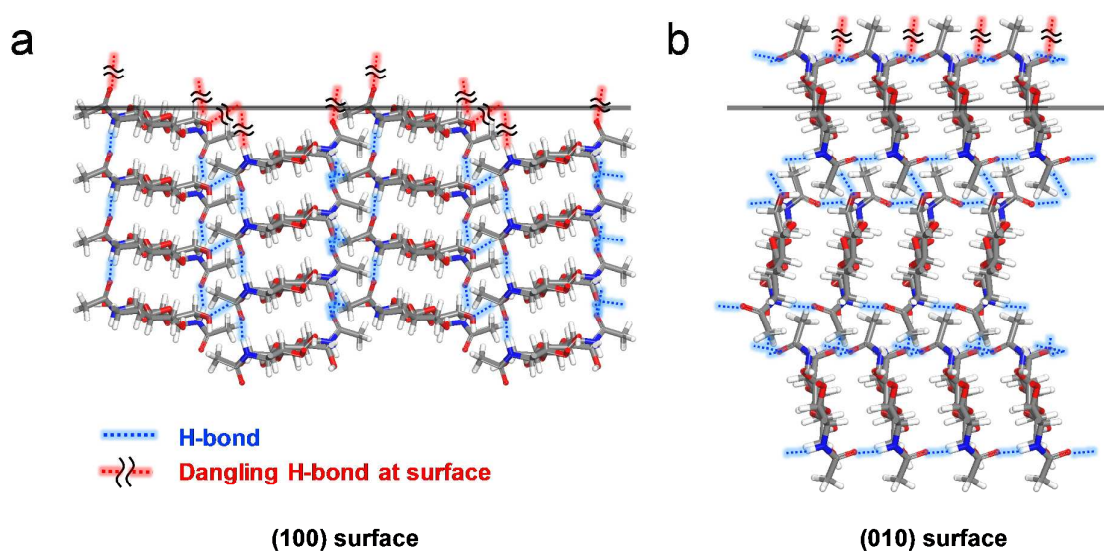


Figure S4. The atomic configuration of (a) (100) and (b) (010) planes of α -chitin fibers. The blue dotted lines represent the hydrogen bonds inside the alpha chitin fibers and the red dotted lines represent the dangling hydrogen bonds of the polar amid and hydroxyl groups exposed on the chitin fiber surface. Carbon, hydrogen, nitrogen, and oxygen are gray, white, blue, and red colors, respectively.

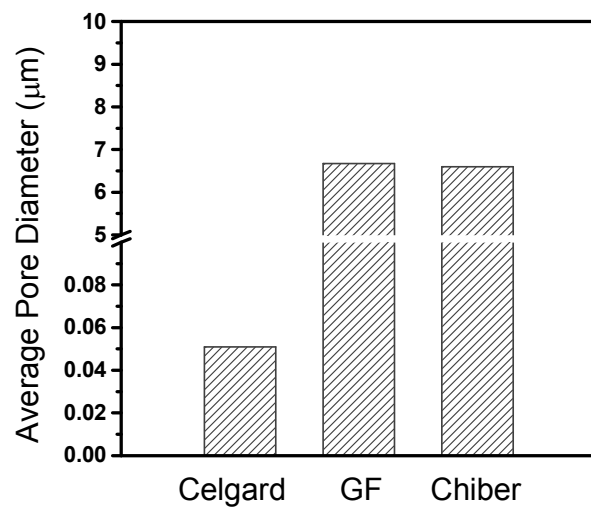


Figure S5. Plot of average pore diameter of Celgard, GF and Chiber separator obtained by mercury porosimetry.

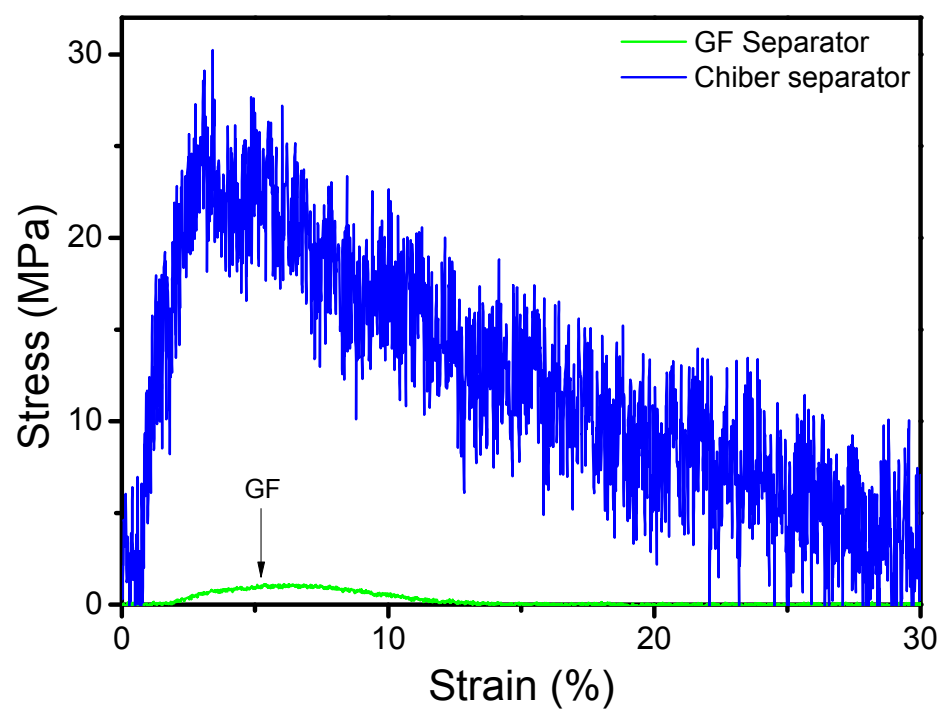


Figure S6. Stress-strain curves of regular GF (green) and Chiber (blue) separator. The extension rate is 0.5 mm/min.

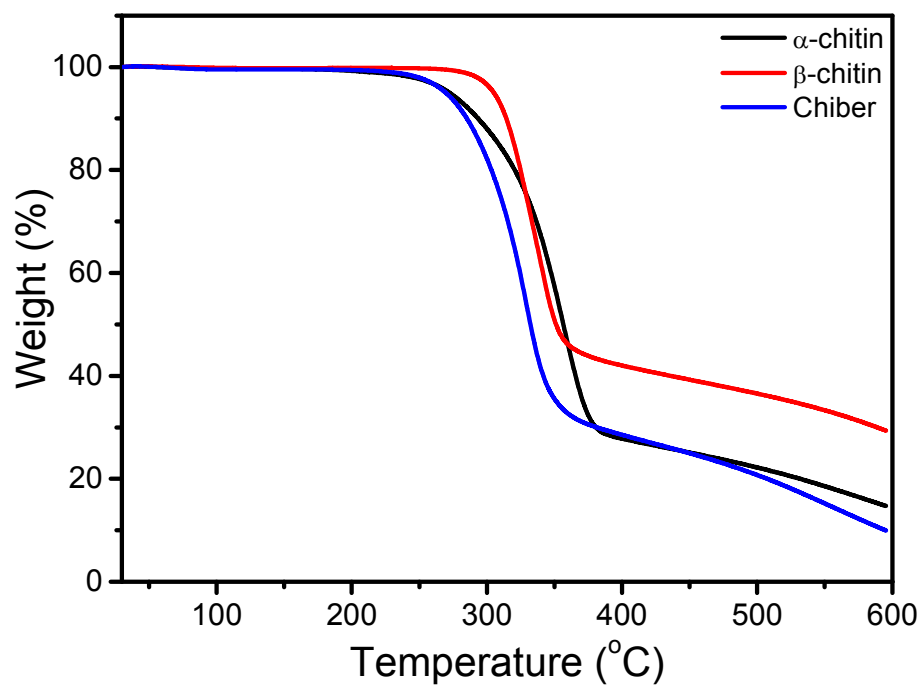


Figure S7. TGA thermographs of α -chitin (black line), β -chitin (red line) and Chiber (blue line) (5 °C/min ramp speed, N₂ atmosphere).

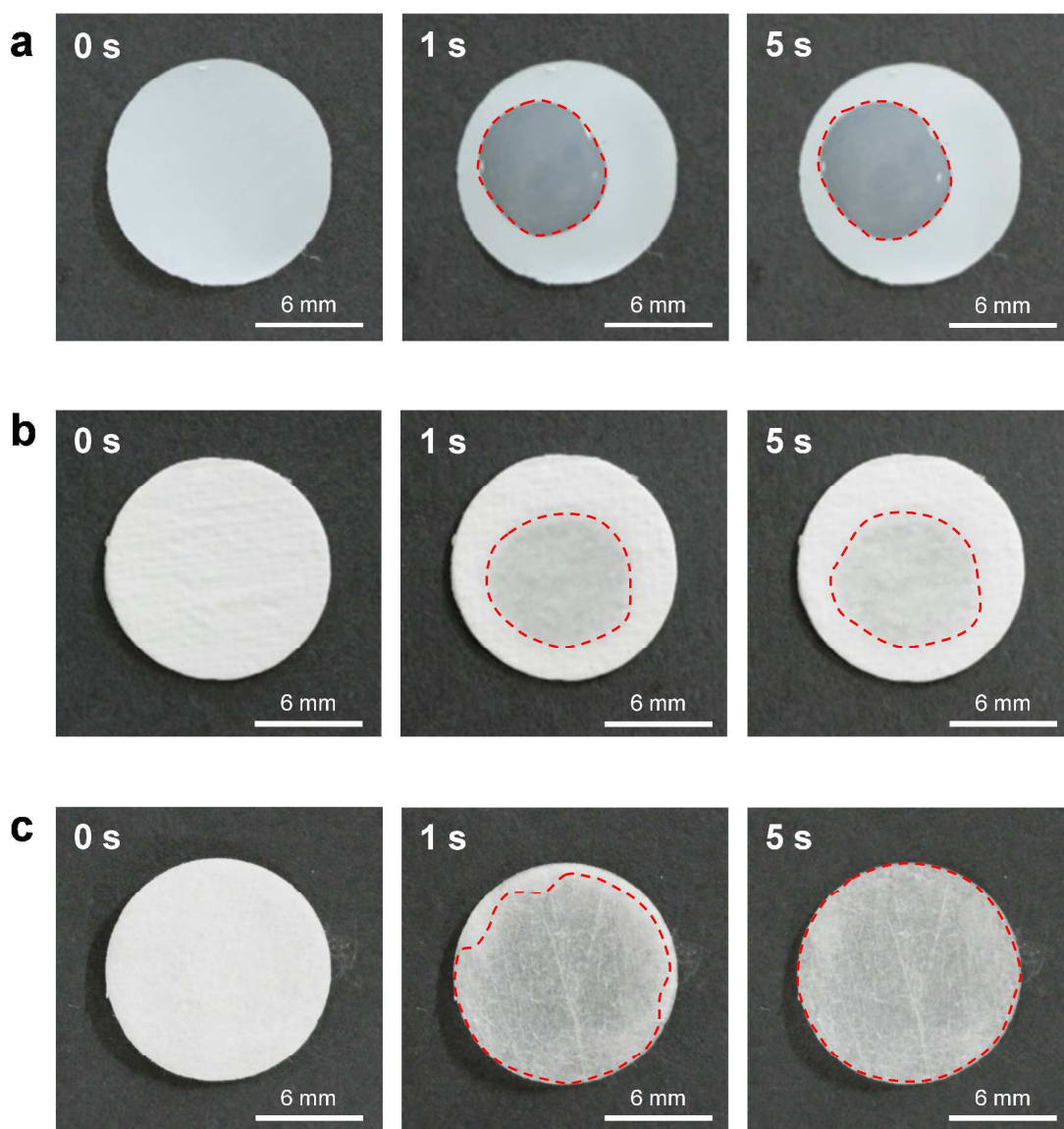


Figure S8. Time lapse photographs of Electrolyte uptaking test for 12 mm (a) Celgard, (b) GF and (c) Chiber separators after dropping 10 μ l of 1M LiTFSI-DME electrolyte.

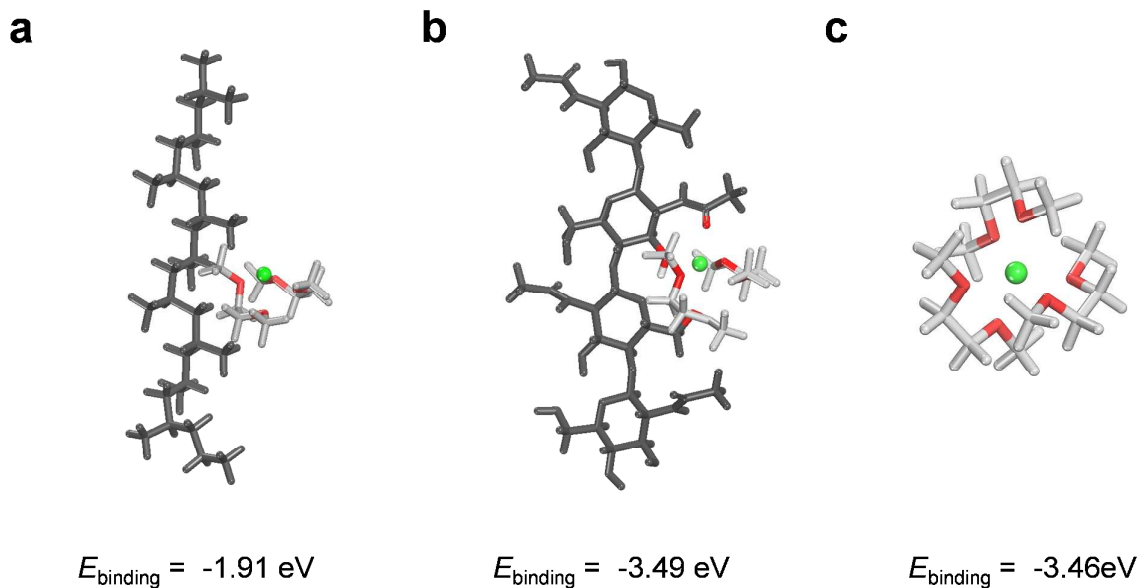


Figure S9. The optimized geometries of (a) $[\text{Li}(\text{DME})_2(\text{PP})]^+$, (b) $[\text{Li}(\text{DME})_2(\text{chitin})]^+$, and (c) $[\text{Li}(\text{DME})_3]^+$ complexes with the calculated binding energies of Li^+ to surrounding molecules. The polymer, DME molecule, and oxygen atom are dark gray, light gray and red colors, respectively.

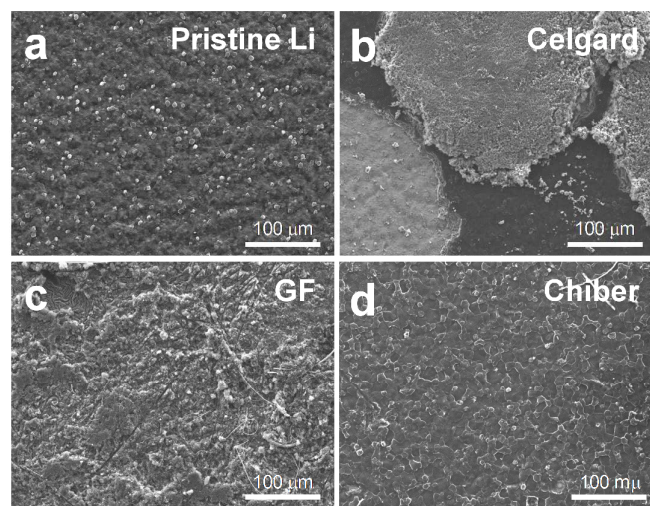


Figure S10. SEM plane-view images of 5 μm thick Li metal surface of (a) pristine Li and after 10 times plating/stripping cycled (b) Celgard, (c) GF and (d) Chiber separators.

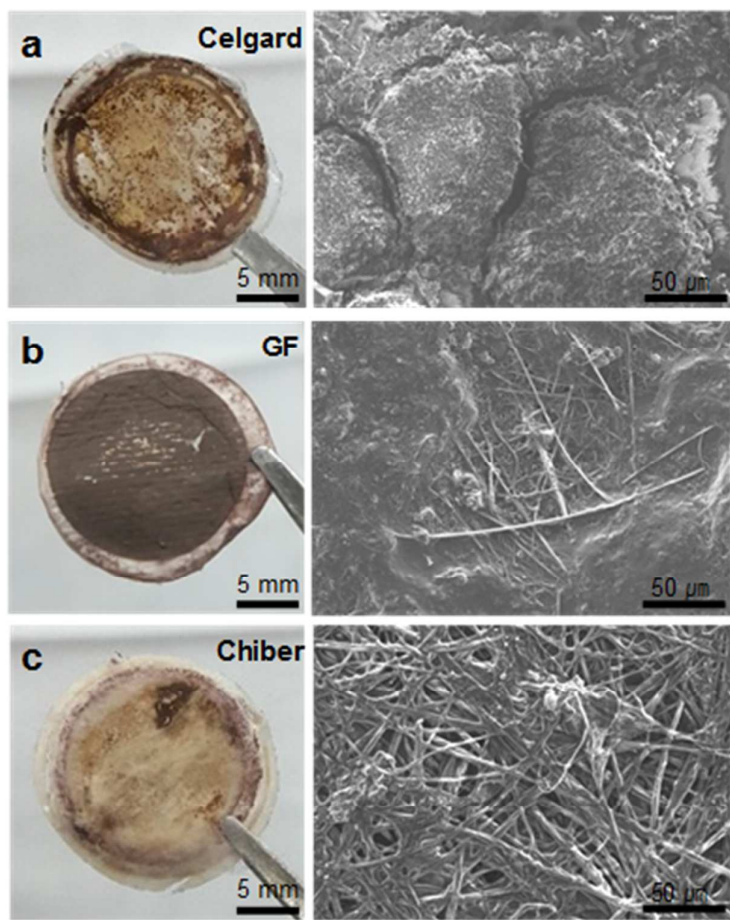


Figure S11. Photographs and SEM images of (a) Celgard, (b) GF, and (c) Chiber separator after cell failure in Li/Li symmetry cells.

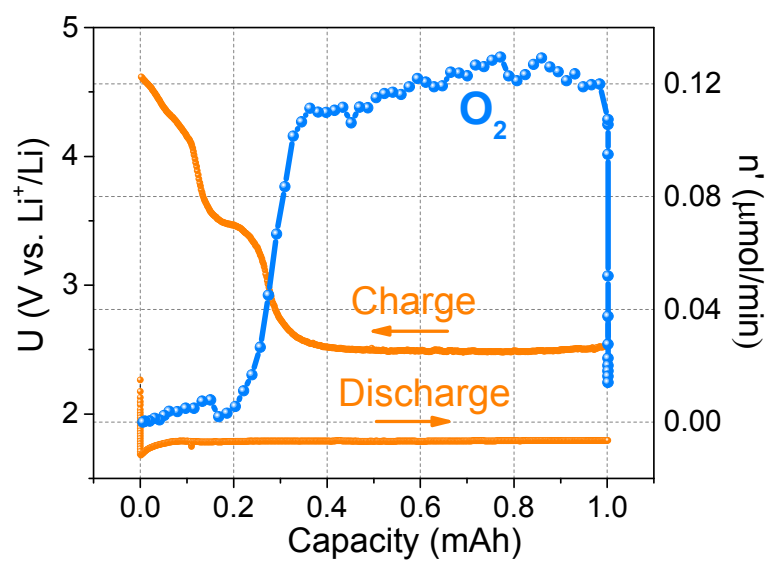


Figure S12. Galvanostatic discharge/charge plot and in situ DEMS result of Chiber contained Na-O₂ battery. 1 M NaOTf-DME electrolyte was used for all electrochemical measurement.

Table S1. The information of the model systems. N_{chains} , N_{DME} , N_{Li^+} and N_{TFSI^-} are the number of chains constituting the nanofiber, DME molecules, Li ions, and TFSI ions contained in each model system, respectively.

System	Box size $X \times Y \times Z$ (nm)	N_{chains}	N_{DME}	N_{Li^+}	N_{TFSI^-}
PP	$7.0 \times 4.0 \times 7.1$	44	736	76	76
α -chitin	$7.0 \times 4.2 \times 7.0$	37	772	80	80
β -chitin	$7.0 \times 4.2 \times 7.0$	37	779	81	81