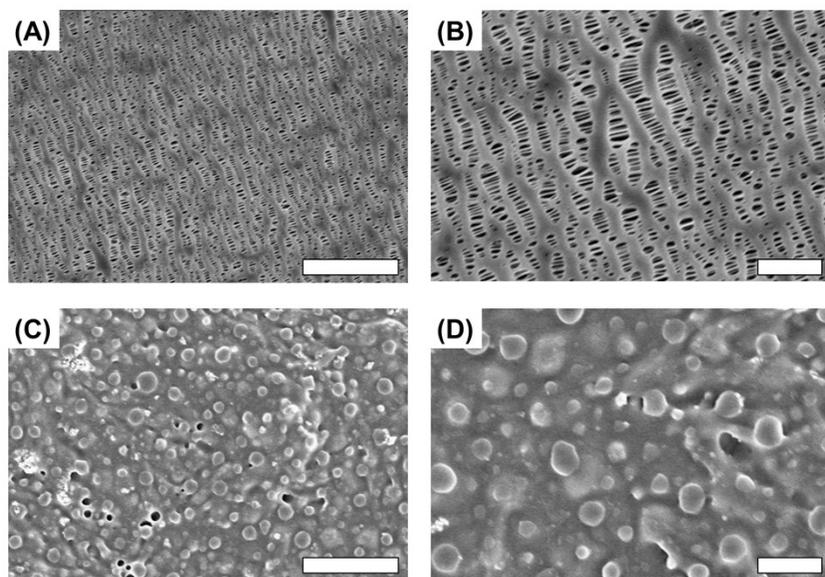


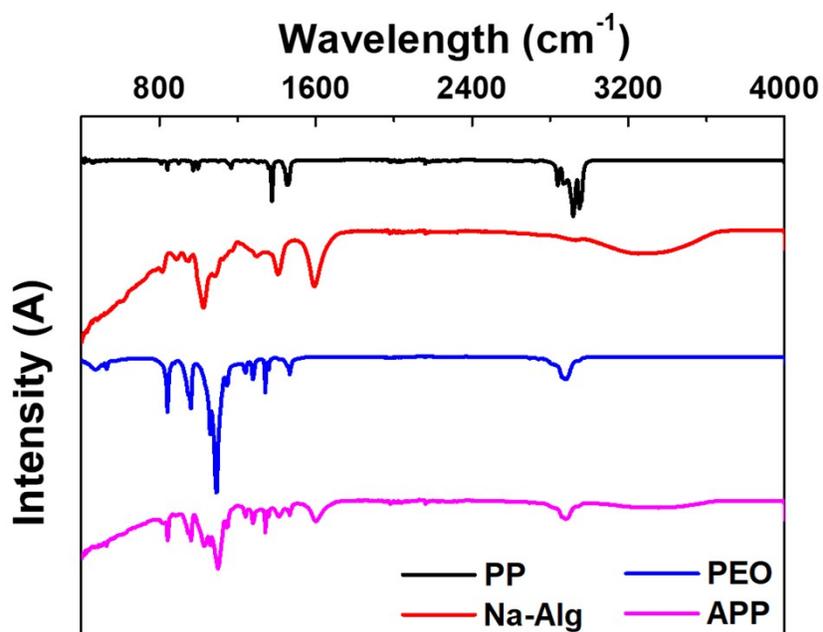
Supporting information

**Biopolymer-based Functional Separator for Stable Li Metal Battery with  
Additive-Free Commercial Electrolyte**

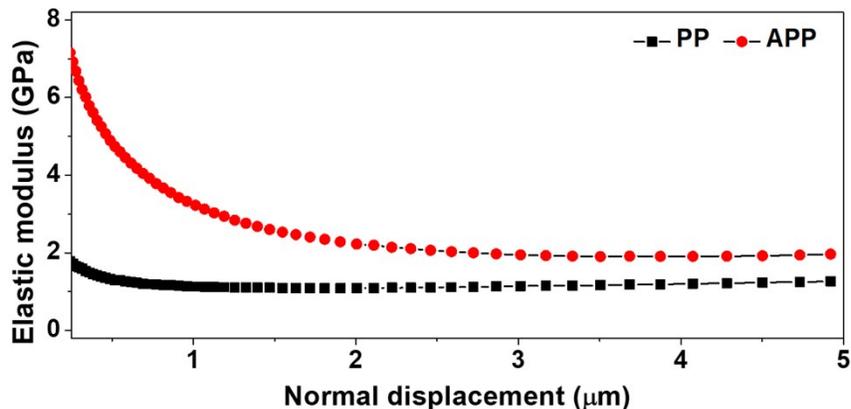
Jooyoung Jeong,<sup>a,b</sup> Jia Lee,<sup>b</sup> Jinuk Kim,<sup>a</sup> Jinyoung Chun,<sup>c</sup> DongGyu Kang,<sup>d</sup> Seung Min Han,<sup>d</sup>  
Changshin Jo\*<sup>e</sup> and Jinwoo Lee\*<sup>a</sup>



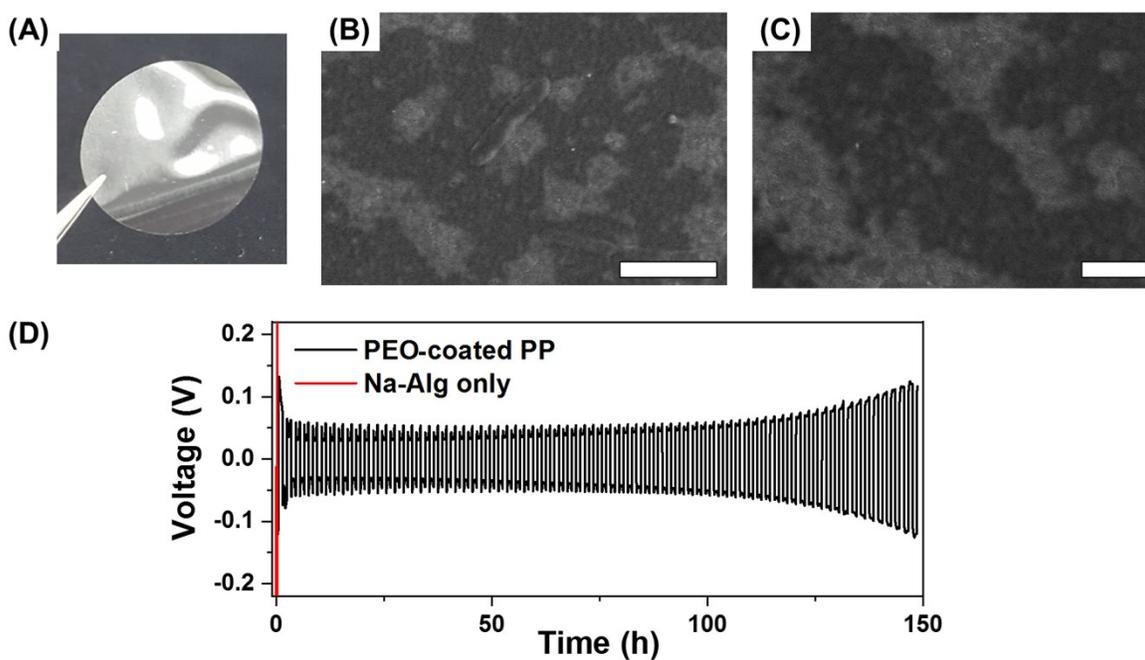
**Fig. S1** SEM images of (A), (B) PP separator and (C), (D) APP film (A), (C) at low magnitude (scale bar = 3 μm) and (B), (D) at high magnitude (scale bar = 1 μm)



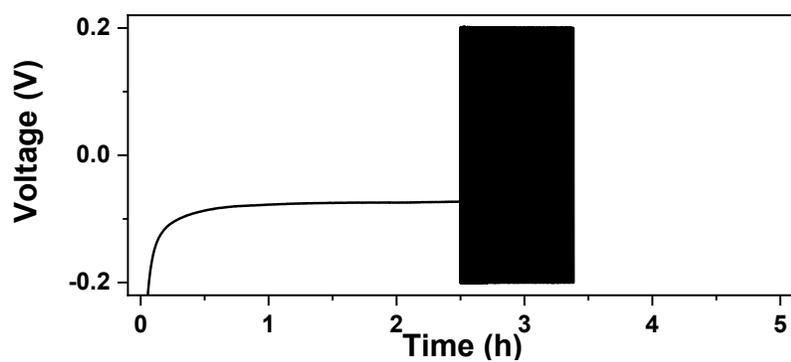
**Fig. S2** Fourier-transformed infra red spectra of PP (black), APP (Magenta), Na-Alg (Red), and PEO (Blue).



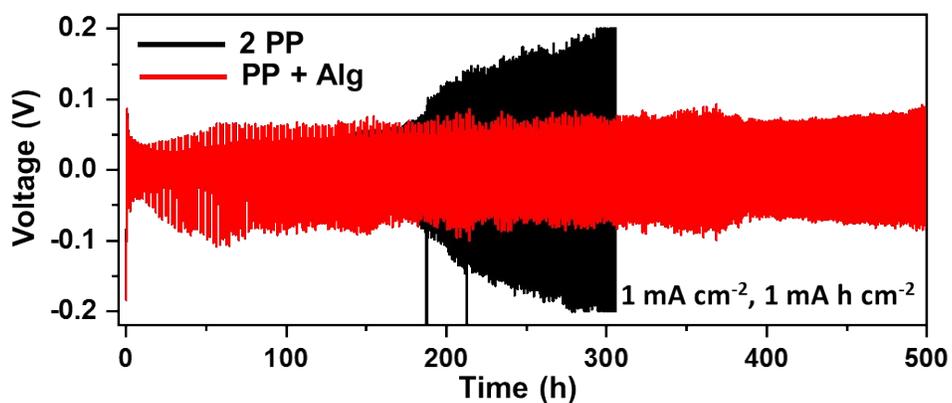
**Fig. S3** Nano-indentation analysis result of PP separator (black) and APP film (red)



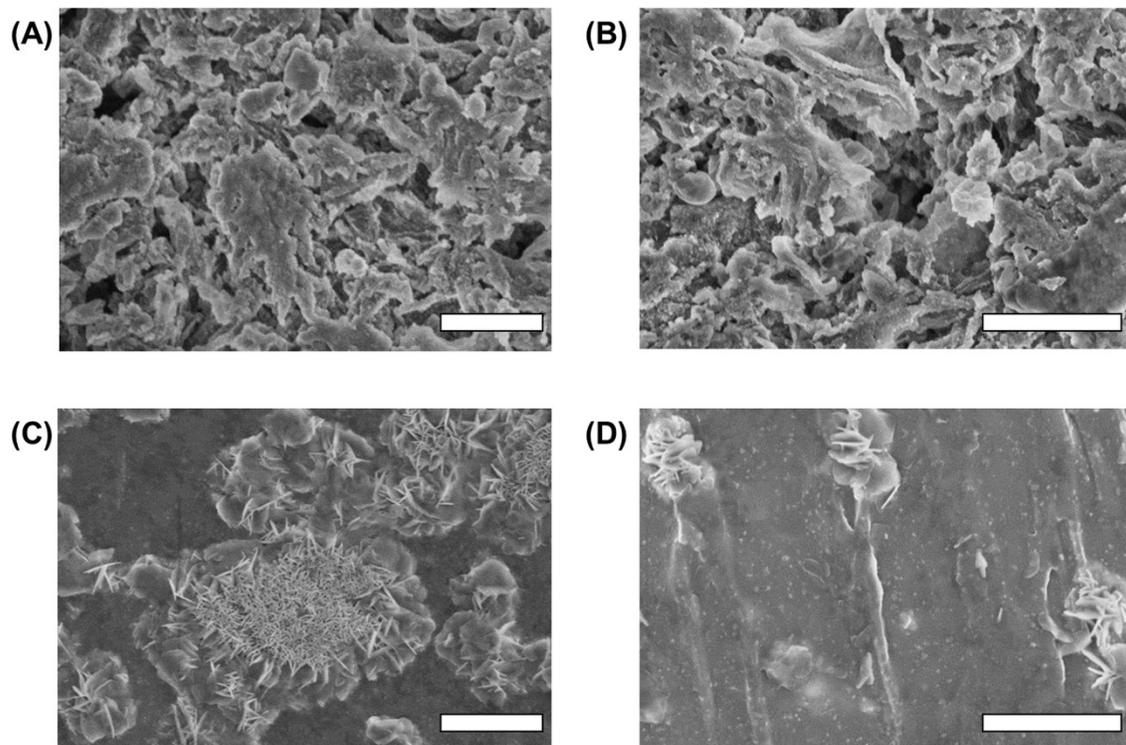
**Fig. S4** Polymer film composed of only Na-Alg. (A) In photograph and SEM images at (B) low magnification (scale bar = 3 μm) and (C) high magnification (scale bar = 1 μm) and (D) voltage profile of symmetric cell test with PEO-coated PP separator and Na-Alg only film in 1 M LiPF<sub>6</sub> in EC:DMC=1:1 electrolyte at rate = 1 mA cm<sup>-2</sup> and capacity cut = 1 mA h cm<sup>-2</sup>.



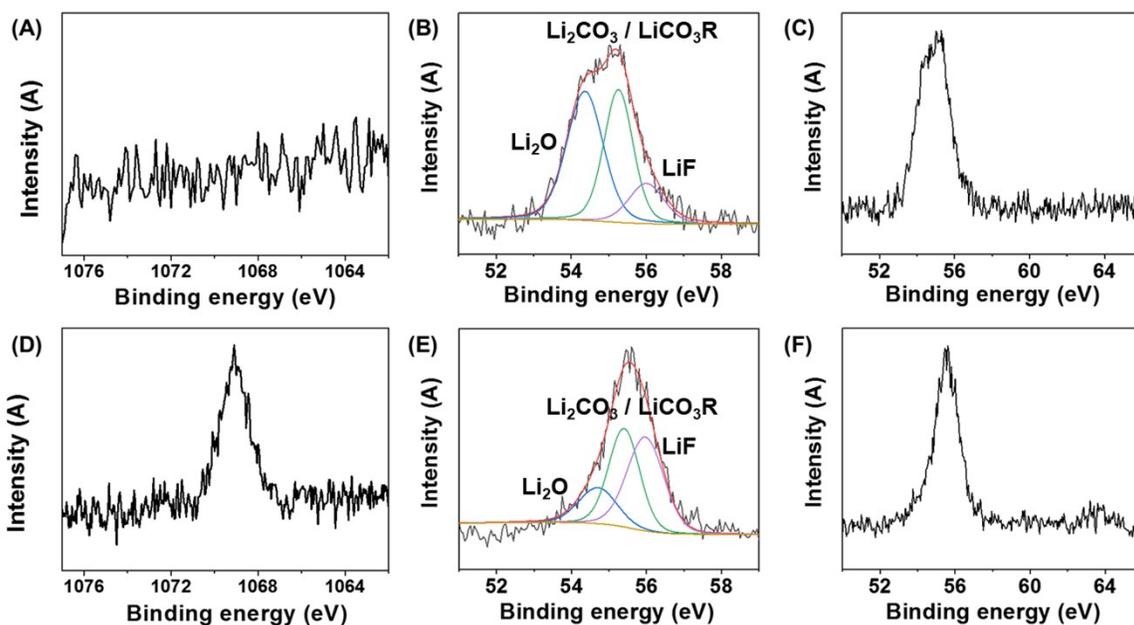
**Fig. S5** Cycle life of symmetric cell using PP separator and 1 M LiPF<sub>6</sub> in EC:DMC=1:1 electrolyte at rate = 5 mA cm<sup>-2</sup> and capacity cut = 5 mA h cm<sup>-2</sup>.



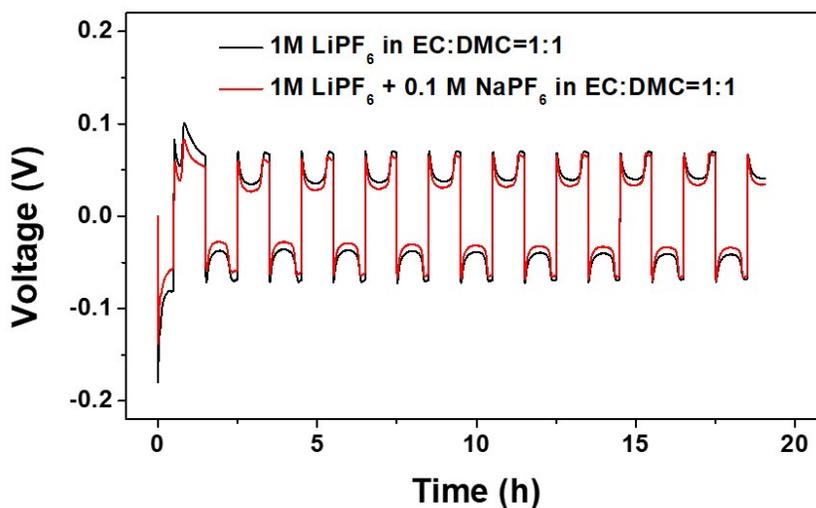
**Fig. S6** Voltage profile of symmetric cell test with two PP separator and PP + APP film in 1 M LiPF<sub>6</sub> in EC:DMC=1:1 electrolyte at rate = 1 mA cm<sup>-2</sup> and capacity cut = 1 mA h cm<sup>-2</sup>.



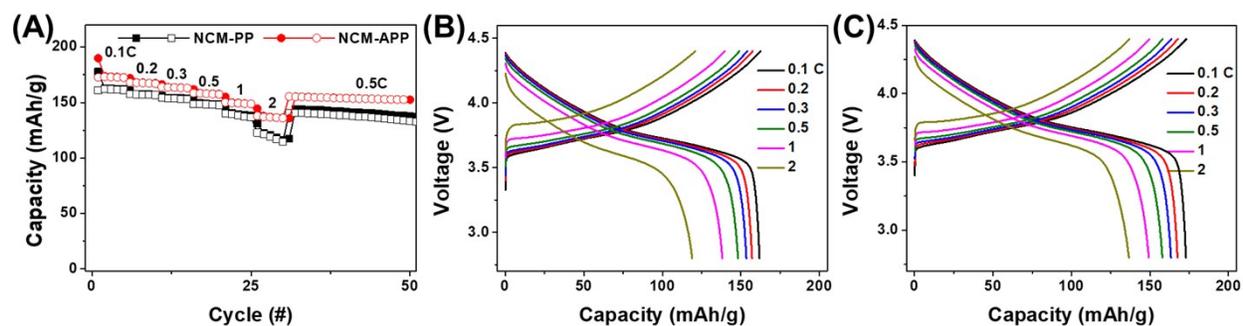
**Fig. S7** SEM images of (A), (B) PP-Li and (C), (D) at different part of Li metal. (all scale bars = 2 μm)



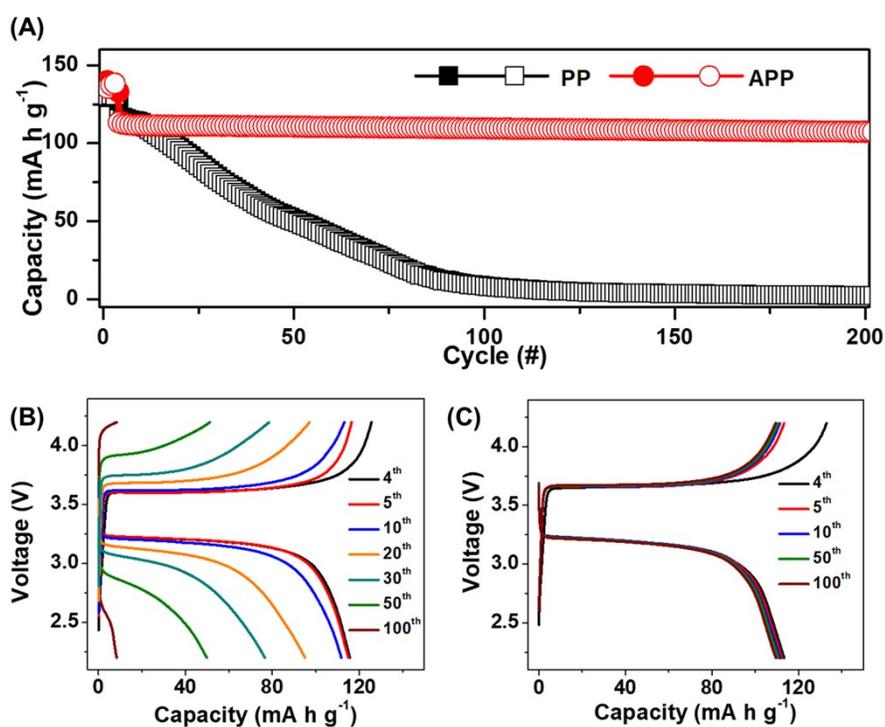
**Fig. S8** XPS spectra of (A)-(C) PP-Li and (D)-(F) APP-Li. (A),(D) Na 1s, (B),(E) Li 1s, and (C),(F) Li 1s with Na 2s binding energy range.



**Fig. S9** Voltage profile symmetric cell with and without  $\text{NaPF}_6$  in electrolyte with PP as separator.



**Fig. S10** (A) Rate performance of NCM622 full-cell with PP or APP as separator and its voltage profile of (B) PP full-cell and (C) APP full-cell



**Fig. S11** LiFePO<sub>4</sub> (LFP) full-cell performance with 1 M LiPF<sub>6</sub> in EC:DMC=1:1 electrolyte. (A) Cycle life, and voltage profile of (B) PP cell and (C) APP cell (4<sup>th</sup>, 5<sup>th</sup>, 10<sup>th</sup>, 20<sup>th</sup>, 30<sup>th</sup>, 50<sup>th</sup>, and 100<sup>th</sup> cycle).

**Table S1** Performance of results in various LMB studies ordered by their experimental details.  
 Note that additives are components for electrolytes in most of other works.

Modified part	Material used	Cell test	Fabrication method	Test condition		Cycle life of sym. cell (h / cycle) (at rate/capacity cut)	Overpotential (mV)	Electrolyte
				Rate (mA cm <sup>-2</sup> )	Capacity cut (mA h cm <sup>-2</sup> )			
Separator	Na-Alg + PEO	Symmetric	Polymer solution (Na-Alg+PEO dissolved in water) was casted on Al dish and then water was evaporated	0.5 ~ 10	1 ~ 8	1000 / 500 (at 1 / 1)	< 10	1 M LiPF <sub>6</sub> in EC/DMC=1:1
Electrode	Sn foil	Symmetric	Roll-pressing of Li foil & Sn foil by repetitive calendaring and folding under Ar	5 ~ 30	5 ~ 30	400 / 200 (at 5 / 5)	< 10	1 M LiPF <sub>6</sub> in EC/PC/DEC=1:1:1 + 10% FEC or 1% VC
Electrode	C <sub>60</sub> & Mg	Symmetric	Vacuum thermal evaporation deposition of C <sub>60</sub> and Mg	1 ~ 7	1	550 / 275 (at 1 / 1)	50	1 M LiPF <sub>6</sub> in EC/DEC/EMC=1:1:1
Electrode	P(SF-DOL)-GO nanosheet	Symmetric (electrodeposited)	Li-electrodeposited 3-D Cu was immersed with solution of graphene oxide (GO) and P(SF-DOL) with THF as solvent under Ar	2	4	320 / 80 (at 2 / 4) (99.1% retention)	-	1 M LiPF <sub>6</sub> in EC/EMC/FEC=3:7:1 or EC/DMC + 2% LiBOB
Electrode	1,3-dioxolane	Symmetric	Li foil was treated with 1,3-dioxolane under Ar	1	1	167 / 84 (at 1 / 1)	~10	1 M LiTFSI in TEGDME
Electrode	g-C <sub>3</sub> N <sub>4</sub> coated Ni foam	Symmetric (electrodeposited)	Ni foam was immersed into the solution with exfoliated g-C <sub>3</sub> N <sub>4</sub> by ultrasonication in water and then Li was electrodeposited with amount of 9 mA h cm <sup>-2</sup>	1 ~ 2	1 ~ 8	1000 / 500 (at 1 / 1)	10	1 M LiTFSI in DOL/DME=1:1 + 1% LiNO <sub>3</sub>
Separator	Egg shell membrane	Half & Symmetric	Egg shell membrane collected after acetic acid etching was solvothermally-treated with 90% trifluoroethanol solution	1 ~ 10	1 ~ 5	2000 / 1000 (at 1 / 1)	12	(Hal0) 1 M LiTFSI in DOL/DME=1:1 + 1% LiNO <sub>3</sub> (Sym) 1 M LiPF <sub>6</sub> in EC/DEC/EMC=1:1:1 + 1% FEC
Separator	MOF (HKUST-1) & PVdF-HFP	1-8 Symmetric	Solution of synthesized HKUST-1 nanoparticles and PVdF-HFP with acetone as solvent was poured on PP separator at vacuum state	0.5 ~ 20	0.25 ~ 10	1000 / 1000 (at 10 / 5)	185	1 M LiTFSI in DOL/DME=1:1 + 0.2% LiNO <sub>3</sub>
Separator	Mesoporous SiO <sub>2</sub> film	Symmetric	Synthesized mesoporous SiO <sub>2</sub> film (GO templated) was coated on commercial PP separator by vacuum filtration	1 ~ 2	1 ~ 2	230 / 115 (at 1 / 1)	30	1 M LiTFSI in DOL/DMC=1:1 + 1% LiNO <sub>3</sub>

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