

Supplementary Material

Evaluation of asymmetric poly(vinylidene fluoride)-coated polyimide separator with three-dimensionally homogeneous microporous structure for high-safety lithium-ion battery

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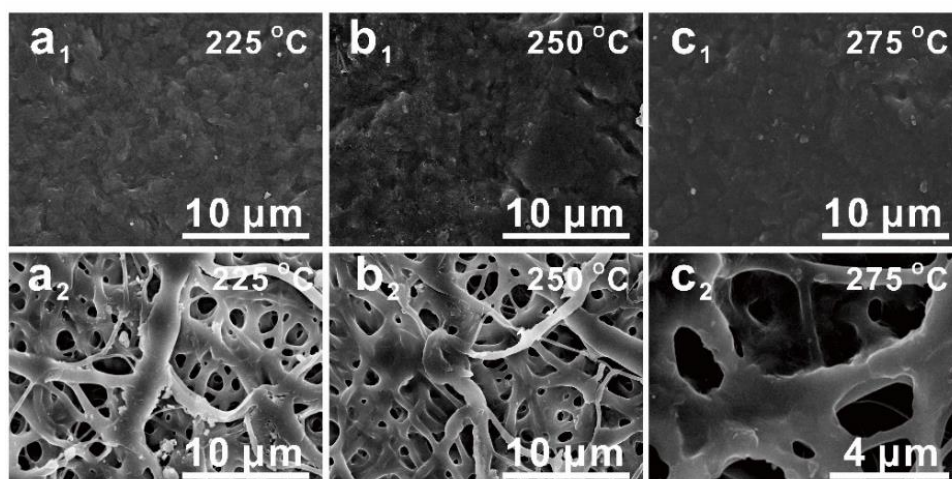


Figure S1. The modified side and unmodified side SEM images of 3DHM API/PVDF separators before and after the thermal treatments for 0.5 hour.

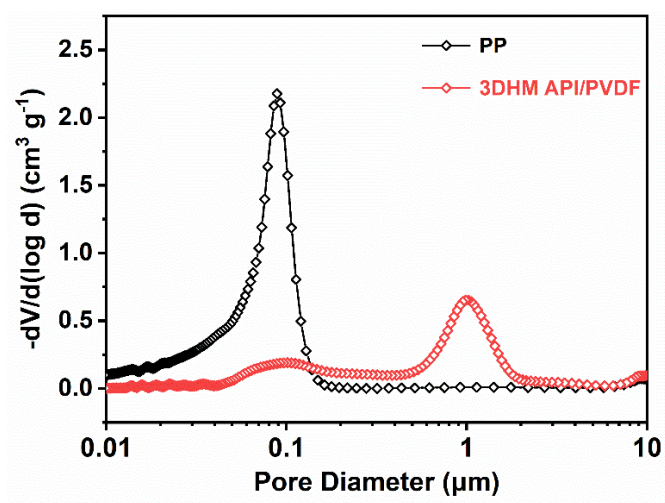


Figure S2. Pore size distributions of PP and 3DHM API/PVDF.

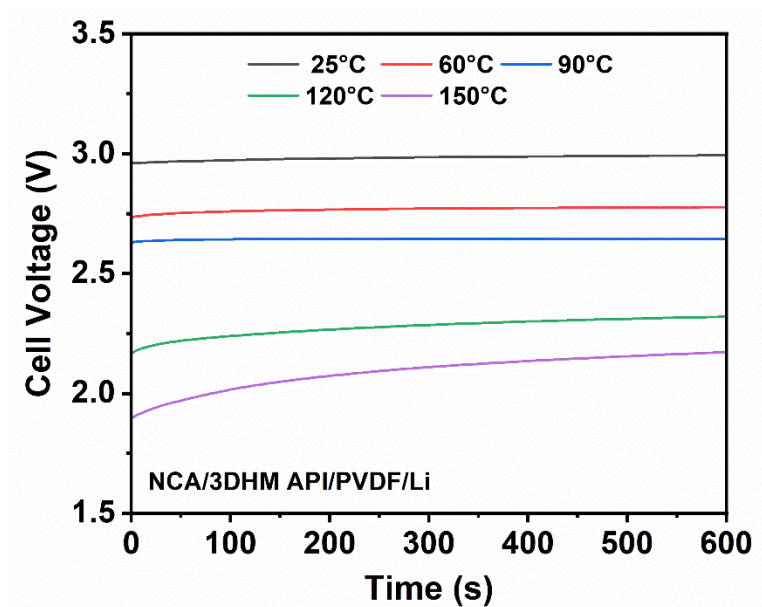


Figure S3. The profiles of the open circuit voltage for the NCA/3DHM API/PVDF/Li after being treated for 1 hour at different temperatures.

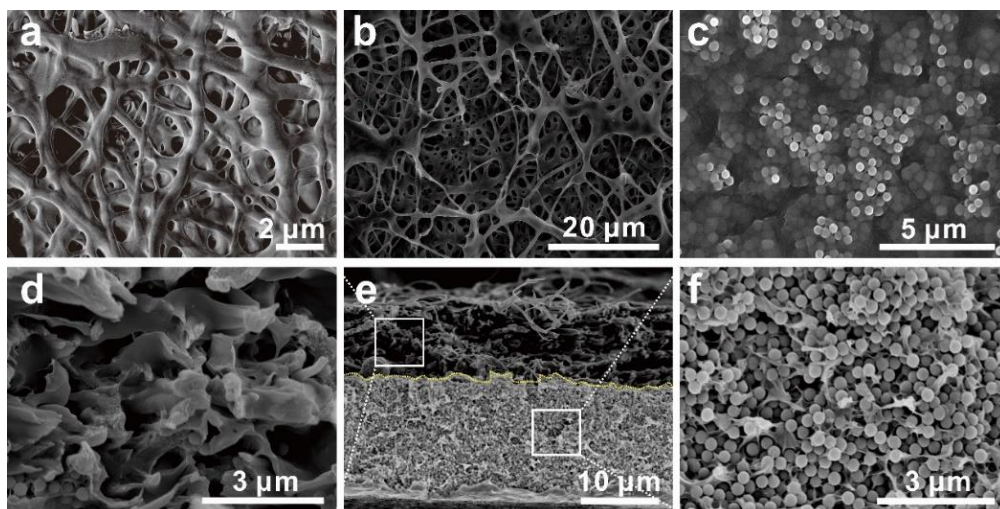


Figure S4. SEM images of (a) the pristine PI film and (b, c, d, e and f) the surface and cross-sectional SEM images of API/PVDF-HFP-SiO₂ (b) the unmodified side, (c) the modified side, (d) the upper section, (e) the overall cross-section and (f) the lower section

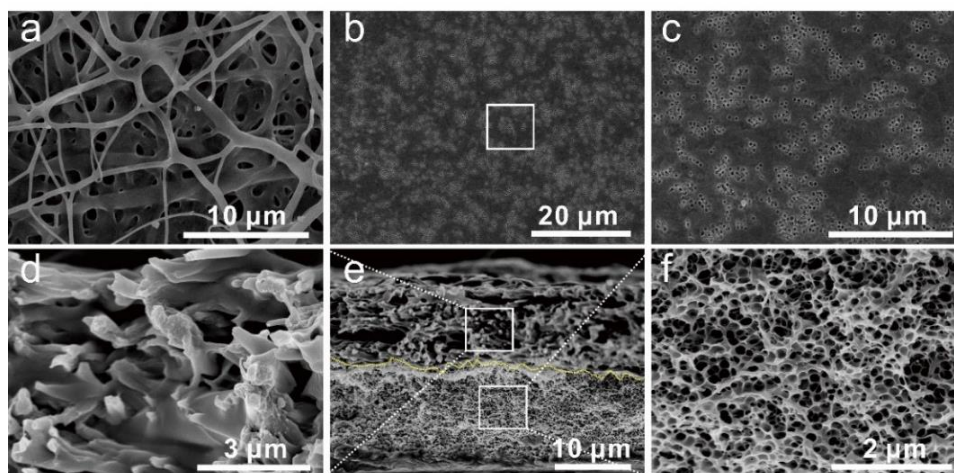


Figure S5. Surface and cross-sectional SEM images of 3DHM API/PVDF-HFP separator (a) the unmodified side, (b and c) the modified side, (d) the upper section, (e) the overall cross-section and (f) the lower section.

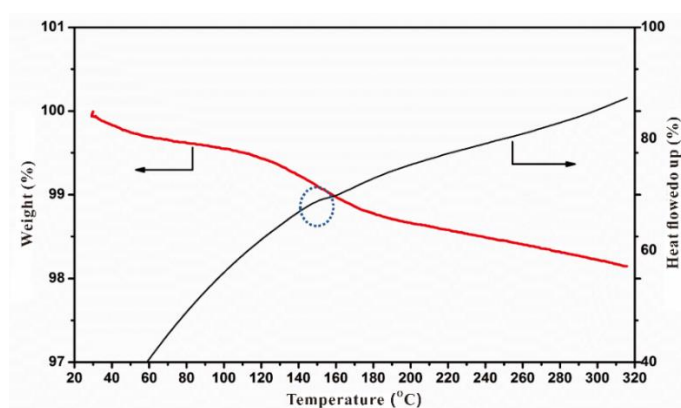


Figure S6. TGA-DSC of 3DHM API/PVDF-HFP separator.

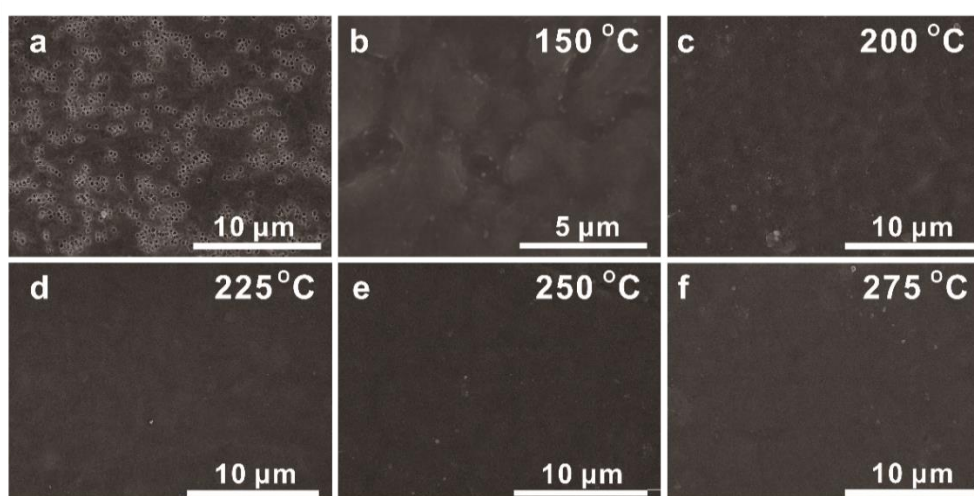


Figure S7. The modified side SEM images of 3DHM API/PVDF-HFP separators before and after the thermal treatments for 30 minutes.

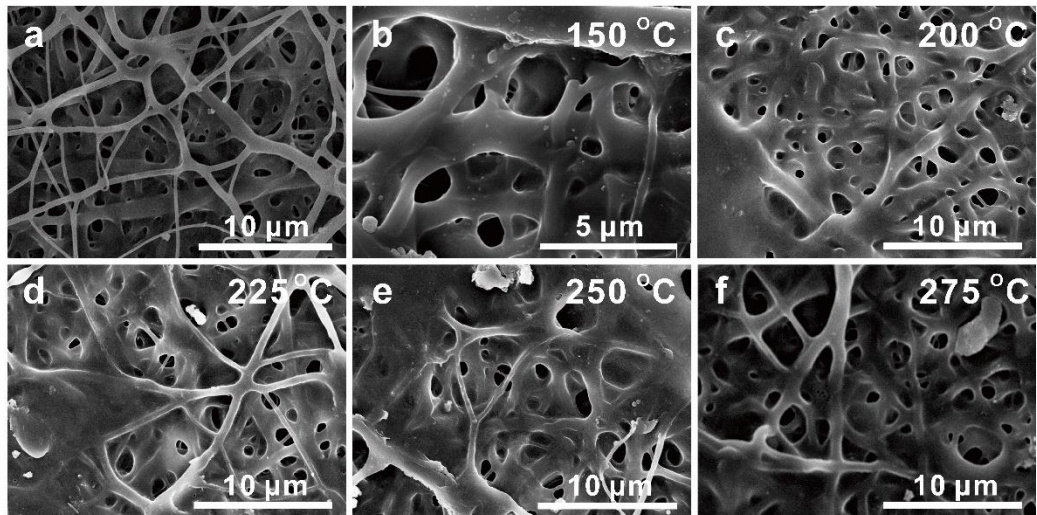


Figure S8. The unmodified side SEM images of 3DHM API/PVDF-HFP separators before and after the thermal treatments for 30 mins.



Figure S9. Optical photos of modified layer of 3DHM API/PVDF-HFP separator before and after the thermal treatments.

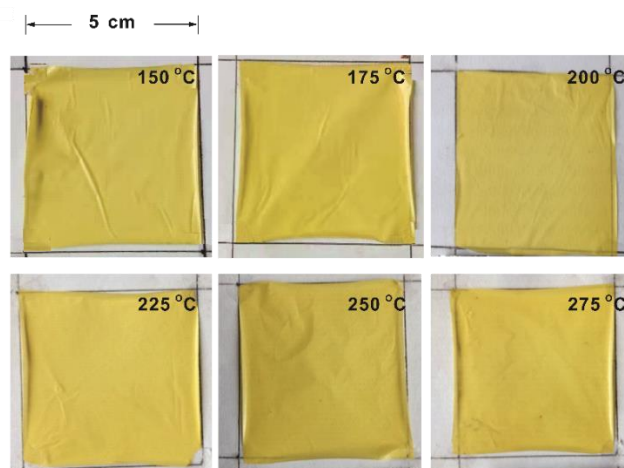


Figure S10. Optical photos of modified layers of 3DHM API/PVDF-HFP separators after the thermal treatments for 30 mins at different temperature.

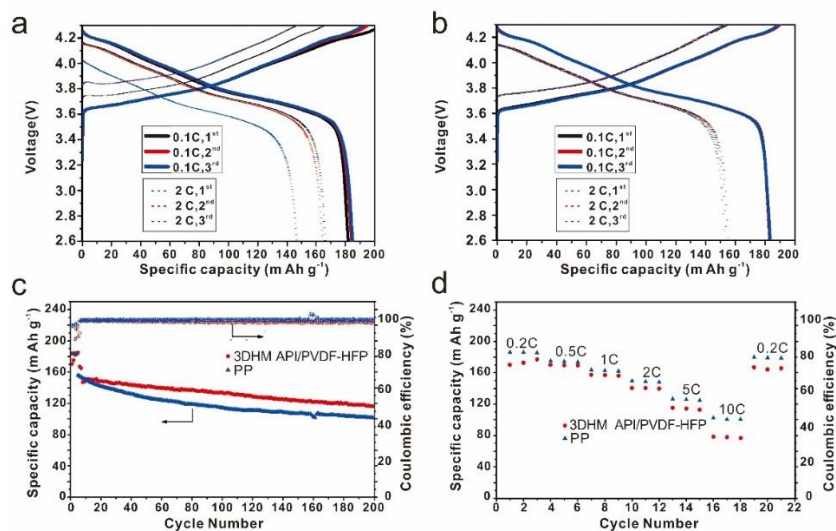


Figure S11. The electrochemical performance of NCA/separator/Li battery with a 3DHM API/PVDF-HFP separator and PP membrane. The charge-discharge curves of batteries with (a) 3DHM API/PVDF-HFP separator and (b) PP membrane at rates of 0.1 C and 2 C. (c) The cycle performance and (d) the rate performance of the battery.

Table S1. Comparison of 3DHM API/PVDF and 3DHM API/PVDF-HFP separators with other reported separators with shutdown function.

Separator	Ionic conductivity (25°C)	Shutdown Temperature	Cathode	Anode	Current	Battery Performance	Ref
BM220/PAN membrane	4.46 mS cm ⁻¹	170°C	LiFePO ₄	Li	1C	100% capacity retention after 500 cycles.	ACS Appl. Mater. Interfaces 2023, 15, 1, 2112-2123
BM220/PAN membrane	4.46 mS cm ⁻¹	170°C	NCM523	Li	8C	79% capacity retention after 500 cycles.	ACS Appl. Mater. Interfaces 2023, 15, 1, 2112-2123
EVOLi-PVA	1.37 mS cm ⁻¹	155°C	LiCoO ₂	Li	0.5C	89.6% capacity retention after 100 cycles.	Journal of The Electrochemical Society, 2021 168 110510

Poly(p-phenylene terephthalamide) modified PE separators	7.46 mS cm ⁻¹	180°C	LiCoO ₂	Li	0.2C	82.7 mAh g ⁻¹ after 50 cycles. (mass loading LiCoO ₂ mass loading:12.8 mg cm ⁻²)	Journal of Membrane Science, 2019, 581, 355-361
AA-BM/polyacrylonitrile	4.7 mS cm ⁻¹	150°C	LiFePO ₄	Li	1C	120.0 mAh g ⁻¹ after 200 cycles	Energy Technol.2022,10, 2200183
AA-BM/polyacrylonitrile	4.7 mS cm ⁻¹	150°C	LiFePO ₄	Li	5C	98.6 mAh g ⁻¹ after 100 cycles	Energy Technol.2022,10, 2200183
Separator	Ionic conductivity (25°C)	Shutdown Temperature	Cathode	Anode	Current	Battery Performance	Ref
Polyethylene-based separator (ASPESA)	0.39 mS cm ⁻¹	120°C	LiFePO ₄	Li	1C	144.5 mAh g ⁻¹ after 900 cycles	Particuology, 2024, 91, 29-37

Polyacrylonitrile/Polyethylene Oxide/Polyacrylonitrile Membrane	1.54 mS cm ⁻¹	80°C	LiFePO ₄	Li	0.1C	initial discharge specific capacity of 153.8 mAh g ⁻¹	Journal of The Electrochemical Society, 2020, 167, 020509
EVOLi-PVA	1.37 mS cm ⁻¹	155°C	LiCoO ₂	Li	0.5C	89.6% capacity retention after 100 cycles.	Journal of The Electrochemical Society, 2021 168 110510
PI/PVDF/PI	3.46 mS cm ⁻¹	170°C	LiMnO ₂	Li	0.5C	97.1% capacity retention after 100 cycles.	Electrochimica Acta, 2015, 176, 727-734
3DHM API/PVDF	0.68 mS cm ⁻¹	175°C	LiNi _{0.8} Co _{0.15} Al _{0.05} O ₂	Li	1C	120 mA hg ⁻¹ after 200 cycles	here
