ELECTRONIC SUPPLEMENTARY INFORMATION

Functional SrF_2 Coated Separator Enabling Robust and Dendrite-Free Solid Electrolyte Interphase on Lithium Metal Anode

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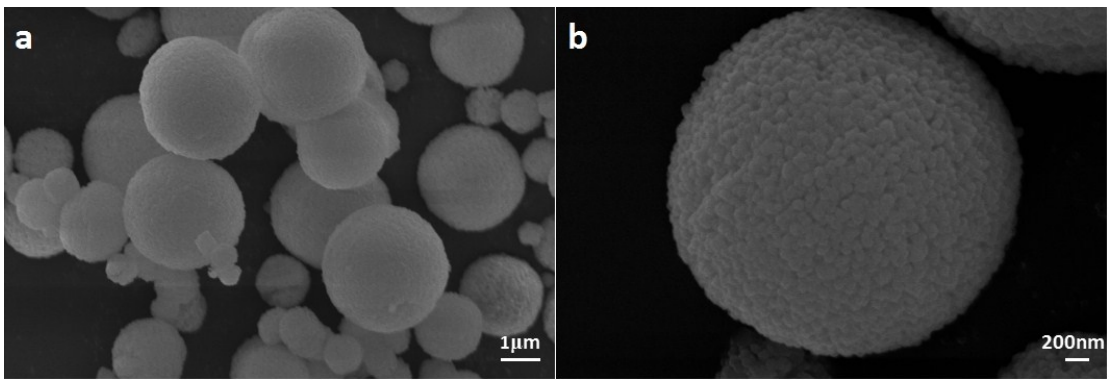


Figure S1. (a), (b) SEM images of the as-synthesized SrF_2 microspheres, taken at different magnifications (scale bars on bottom right).

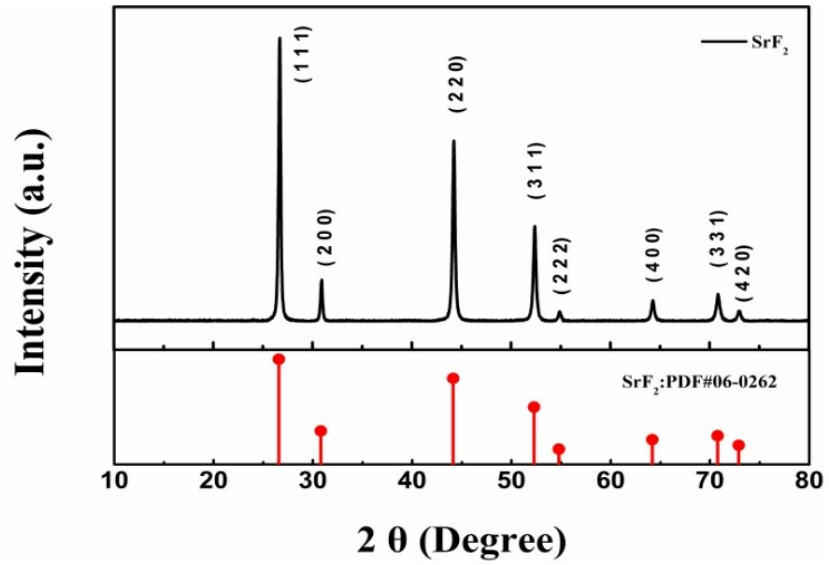


Figure S1 cont. (c) XRD pattern of the as-synthesized SrF_2 microspheres.

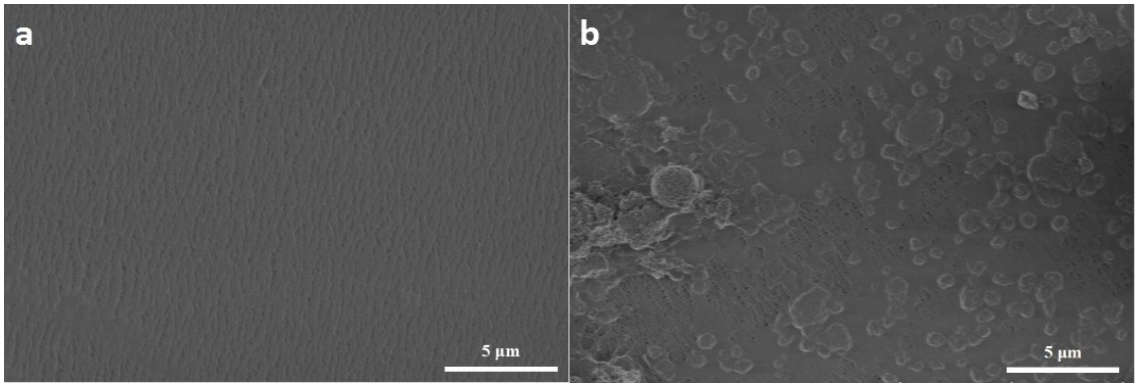


Figure S2. SEM images for **(a)** the pristine PP separator and **(b)** the SrF_2 microspheres coated PP separator.

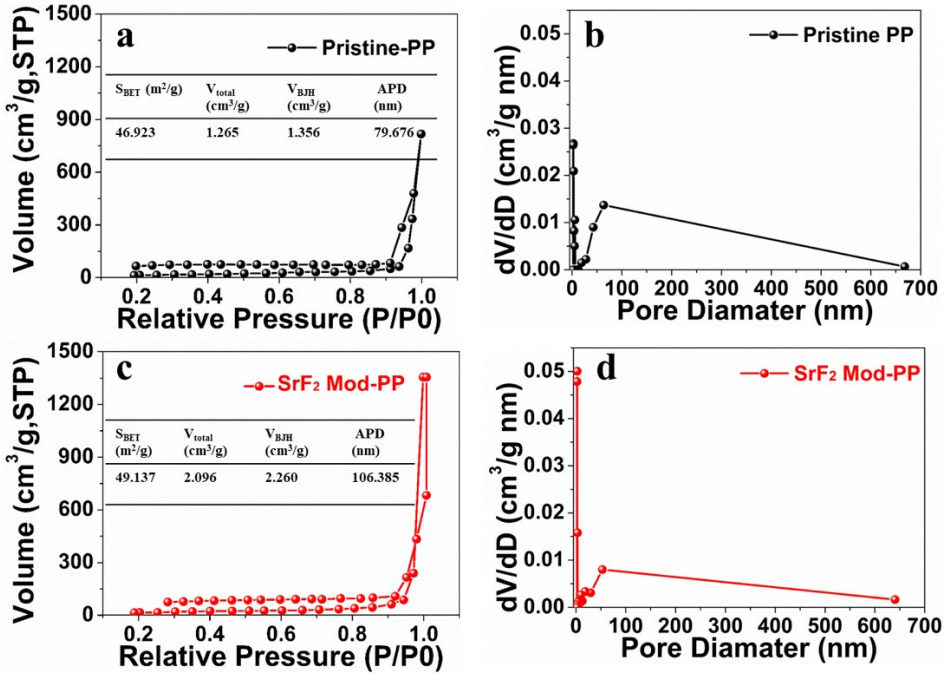


Figure S3. (a), **(c)** N_2 isotherm absorption-desorption curves of pristine and SrF_2 coated separator. **(b)**, **(d)** Pore size distribution of pristine and SrF_2 coated separator.

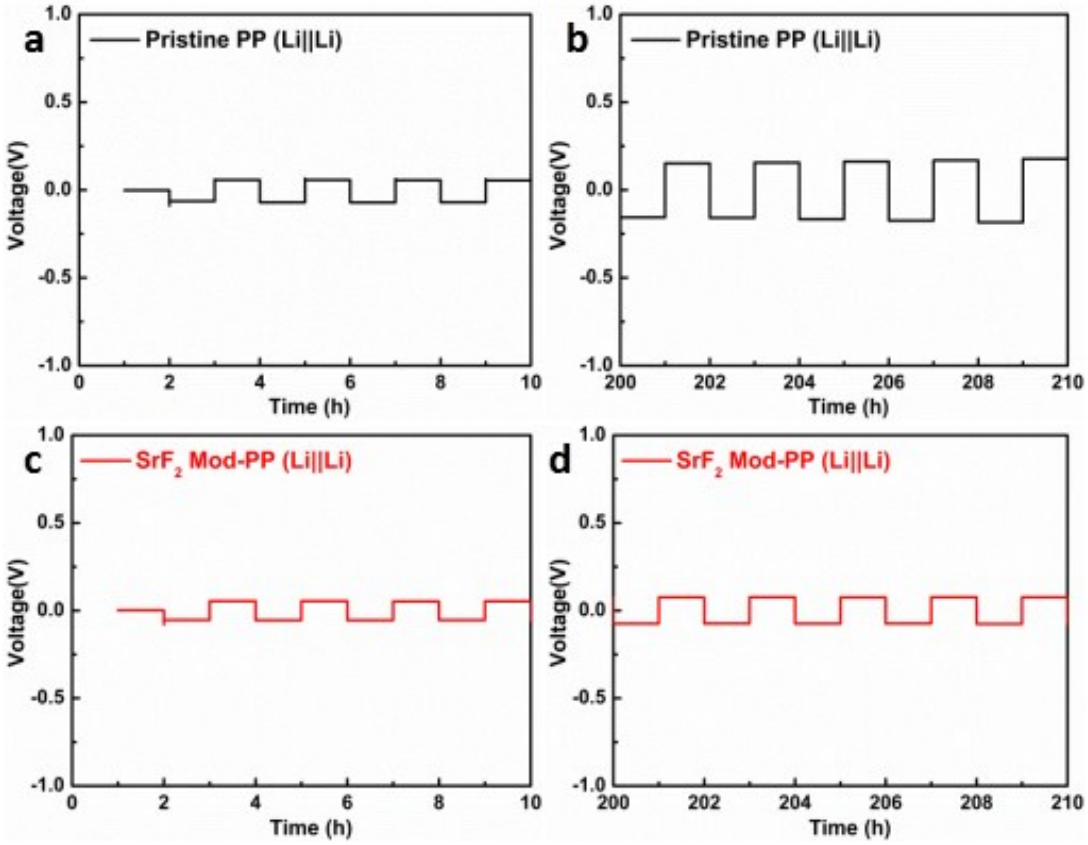


Figure S4. (a) Expanded potential-time plots of the Li||Li cells using, **(b)** pristine separator, and **(c)**, **(d)** SrF_2 coated separator.

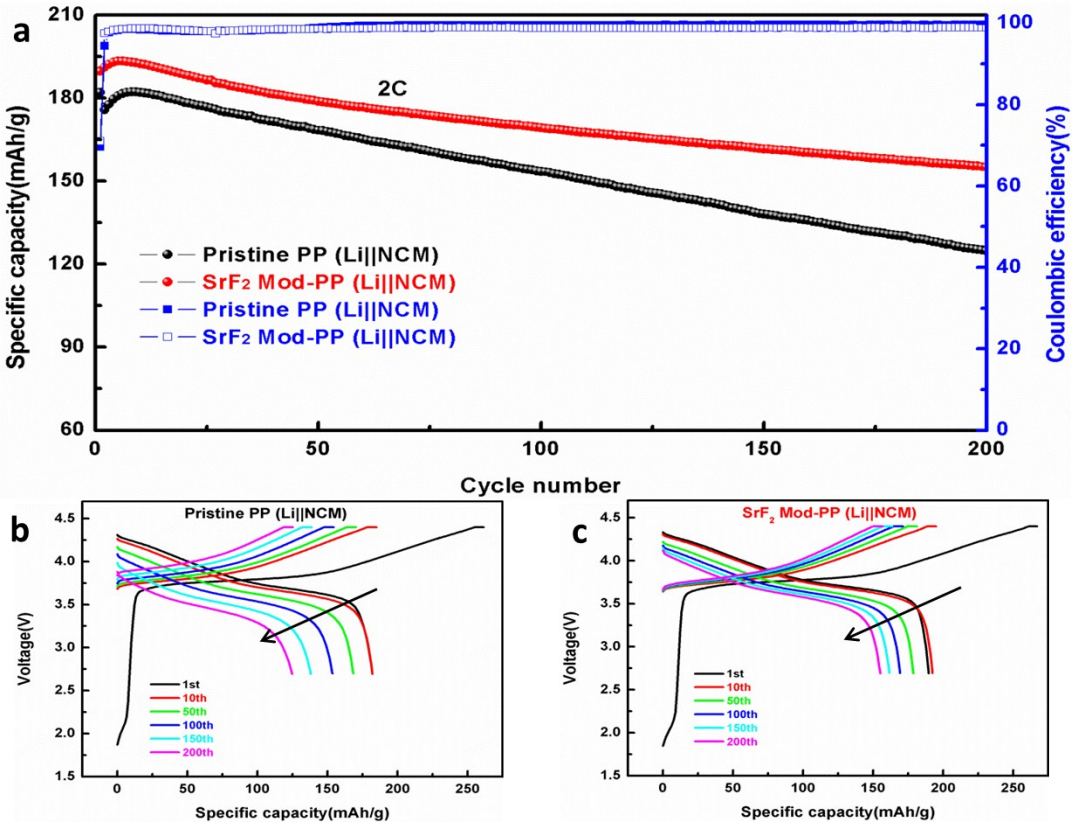


Figure S5. (a) Cycling performance of Li||NCM cells with pristine and SrF_2 modified separators. Tested at a current density of 400 mA/g (2C) at an elevated temperature of 60 ℃. **(a)** and **(c)** The galvanostatic charge-discharge data from which (a) was derived.

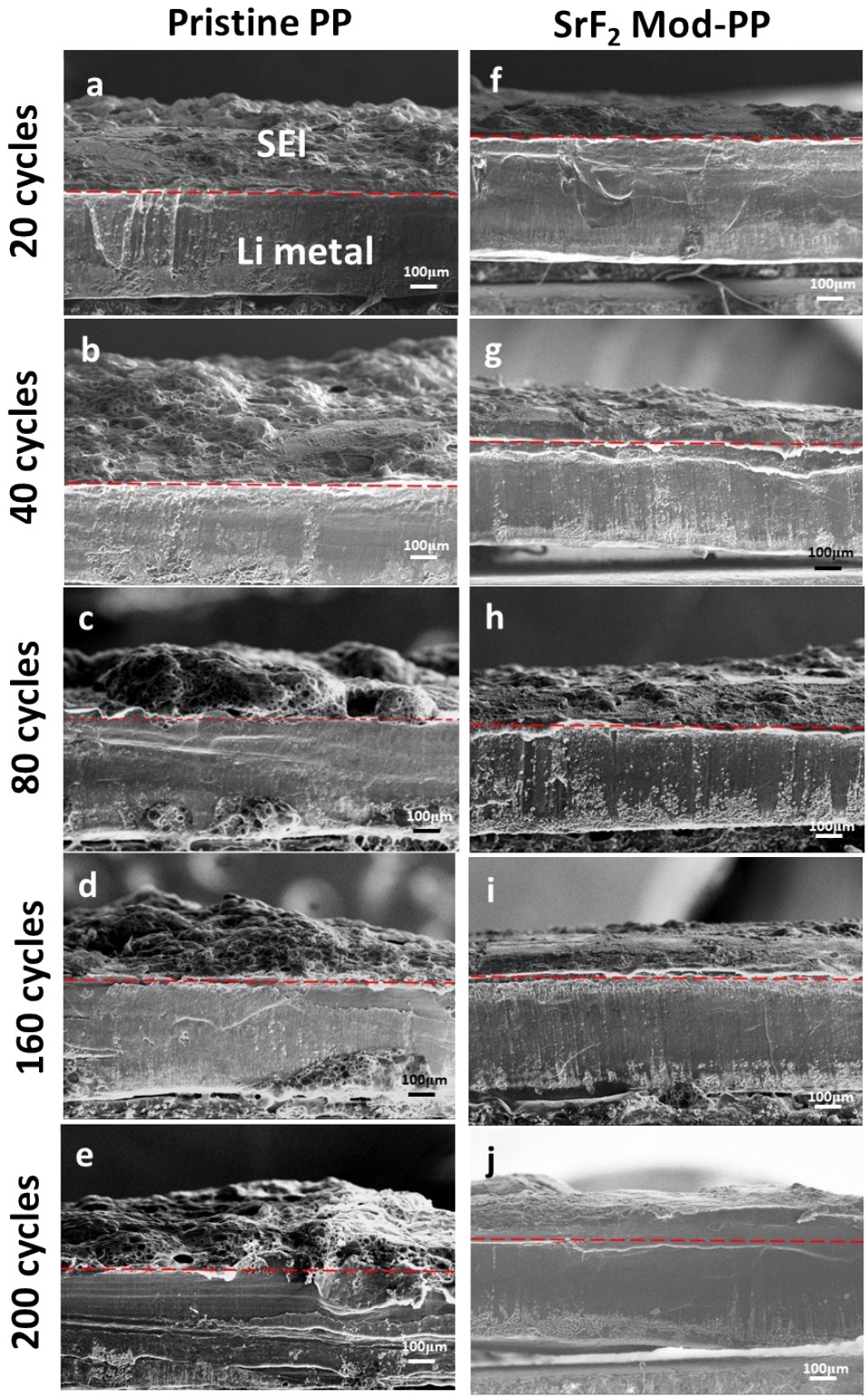


Figure S6. SEM images of the cross-sections of the post cycled Li metal anodes from Li||NCM cells **(a)** - **(e)** pristine separator, **(f)** - **(j)** SrF_2 coated separator. Samples are shown at 20^{th}, 40^{th}, 80^{th}, 160^{th} and 200^{th} cycles, tested at 200 mA/g for 2 hrs. per cycle.

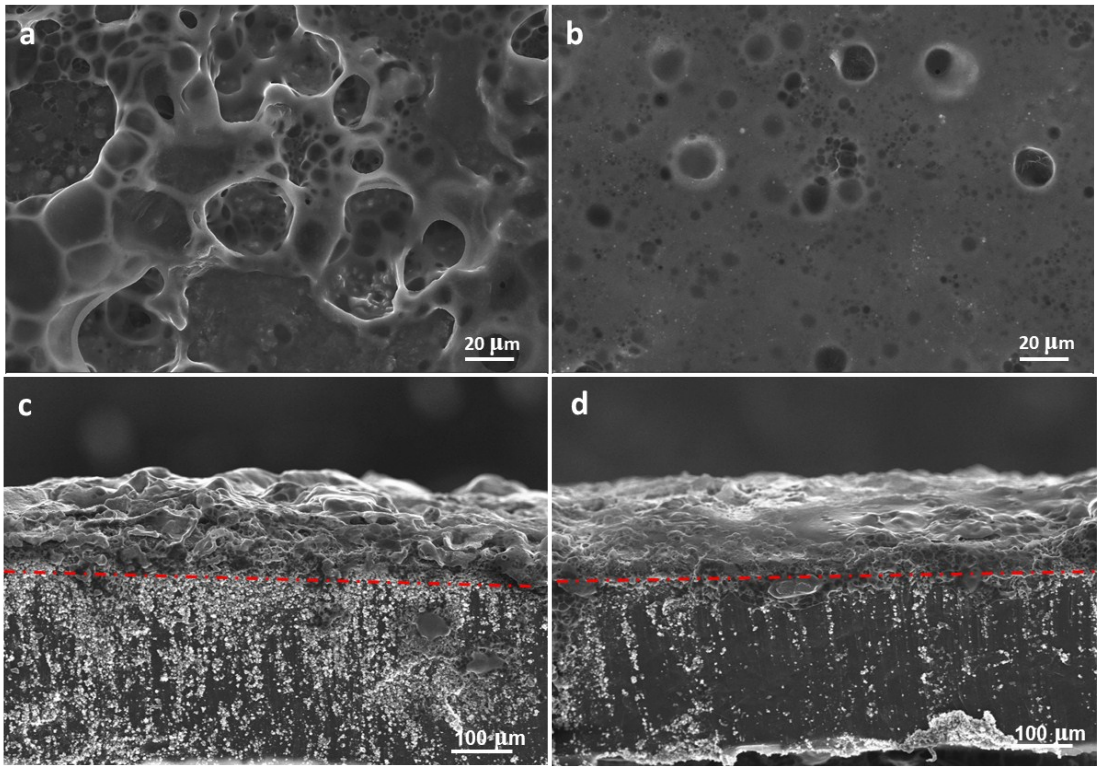


Figure S7. Top-down **(a) - (b)** and cross-section **(c) - (d)** SEM images of the formed SEI morphologies of Li metal anodes from Li||NCM cells using the (a), (c) of pristine separator and (b), (d) of SrF_2 coated separator after 200 cycles at 400 mA/g (2C) and 60 ℃.

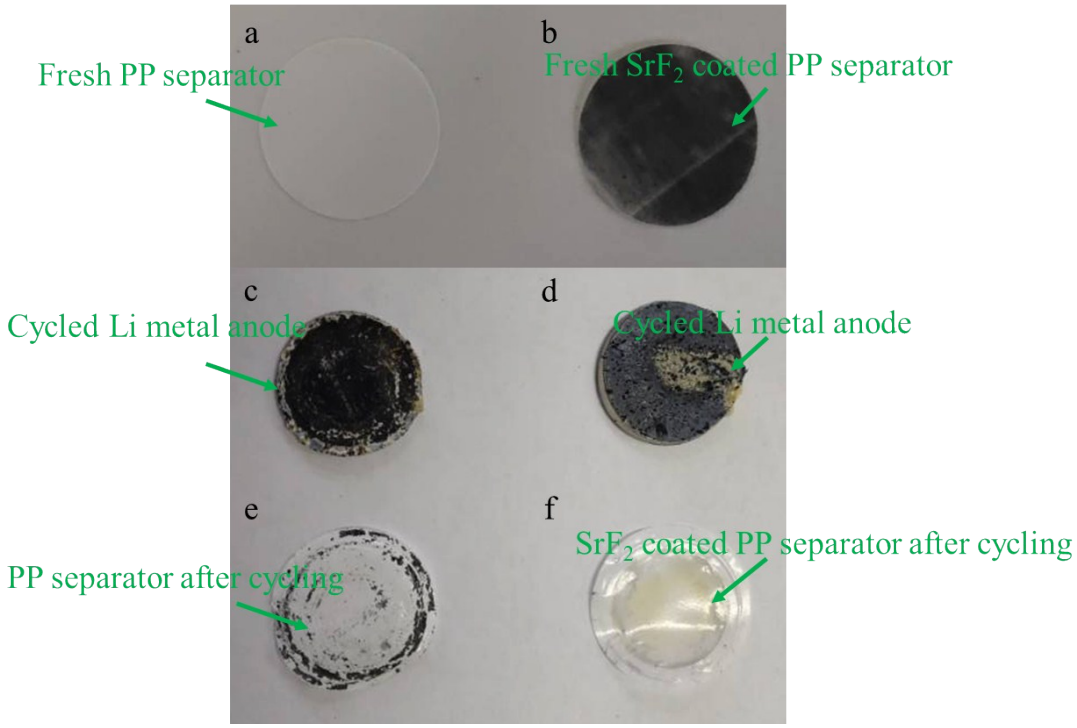


Figure S8. (a) Pristine PP separator uncycled. **(b)** SrF_2 coated separator, uncycled. **(c)** Post-cycled Li metal anode with pristine. **(d)** Post-cycled Li anode with SrF_2 coated. **(e)** Post-cycled pristine separator. **(f)** Post-cycled SrF_2 coated separator.

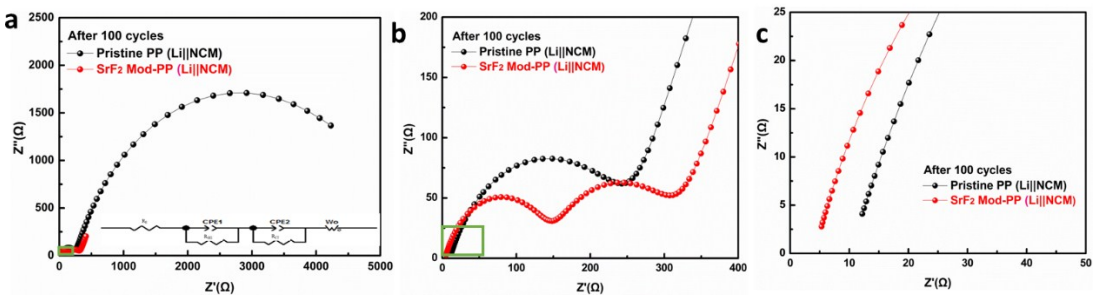


Figure S9. (a) Nyquist plots of Li||NMC coin cells, comparing pristine and SrF_2 coated separators at the 100th cycle, tested at 400 mA/g for 1 h per cycle, and 60°C. The impedance spectra were collected at charged state of 4.1 V. **(b)** and **(c)** Enlarged view of the high-frequency region as labeled by the green rectangles.

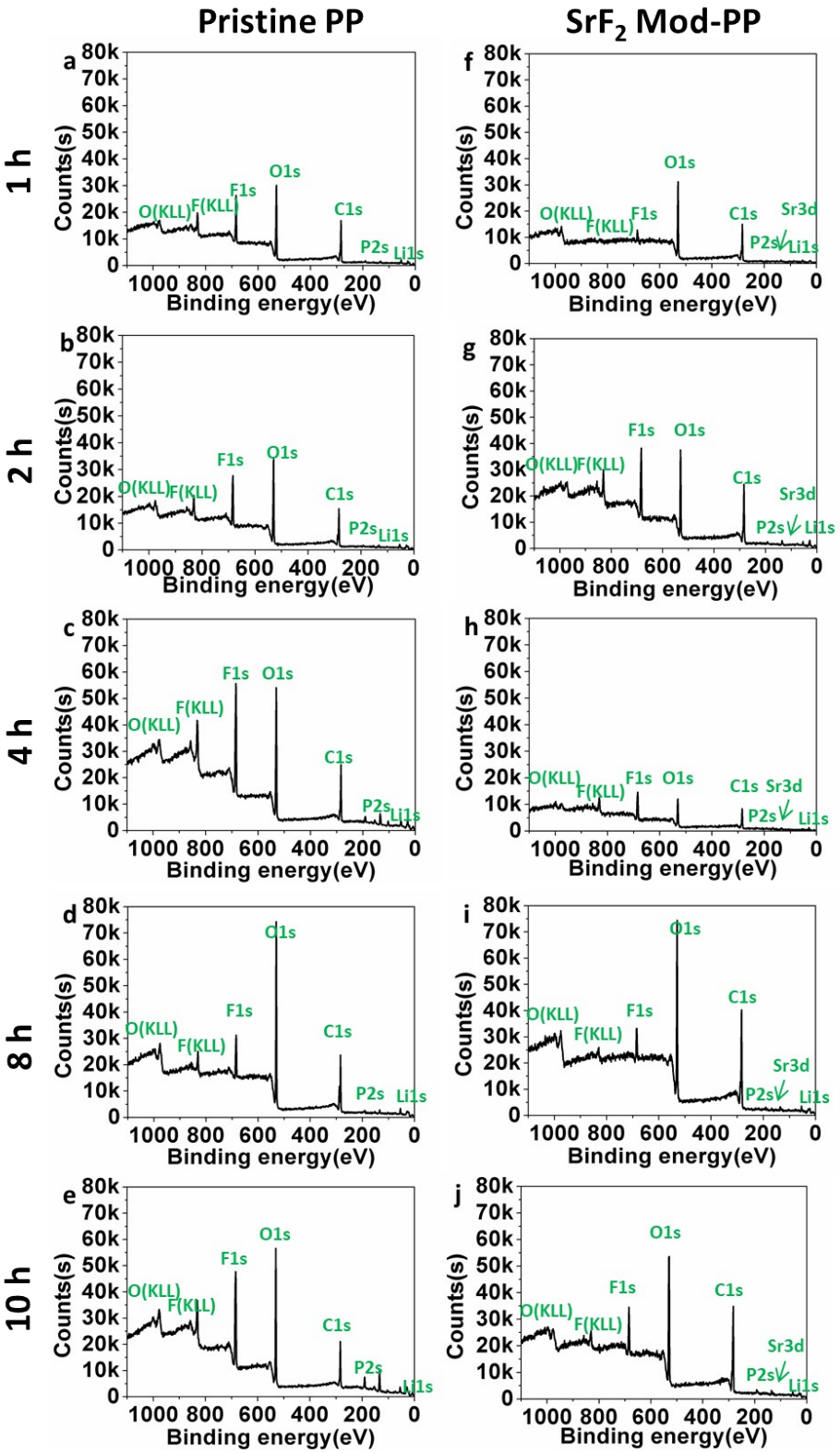
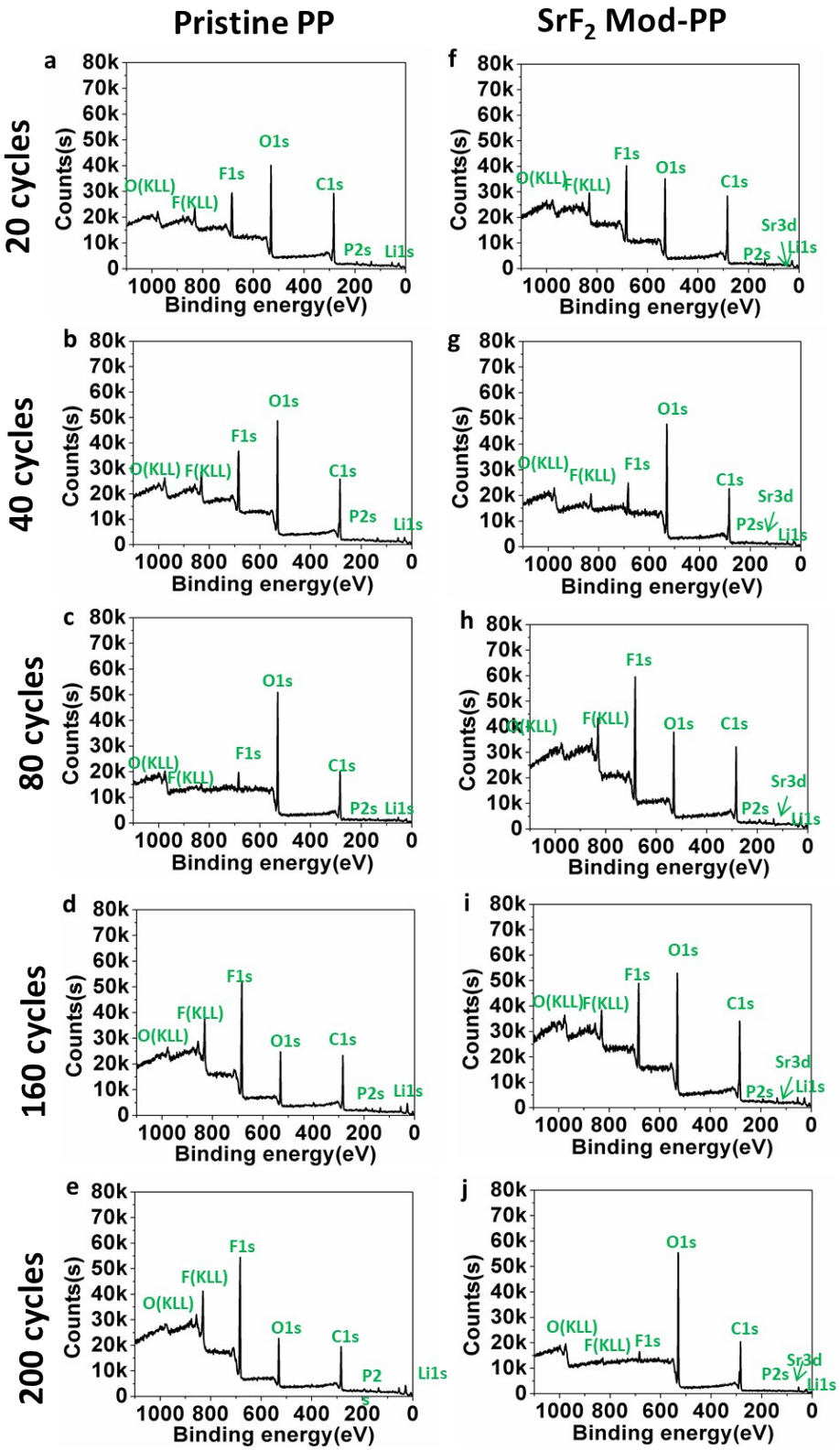


Figure S10. XPS survey spectra for the Li metal anodes under different Li deposition times at 20 mA/g (C/10) for 1h, 2h, 4h, 8h and 10h, corresponding to total 20, 40, 80, 160 and 200 mAh/g, respectively using **(a)** - **(e)** the pristine separator, and **(f)** - **(j)** the SrF_2 coated separator.



Supplementary Figure S11. XPS survey spectra for the Li metal anodes under different electrochemical cycles of 20, 40, 80, 160 and 200 at 200 mA/g (1C) using **(a)** – **(e)** the pristine separator, and **(f)** – **(j)** the SrF_2 coated separator.

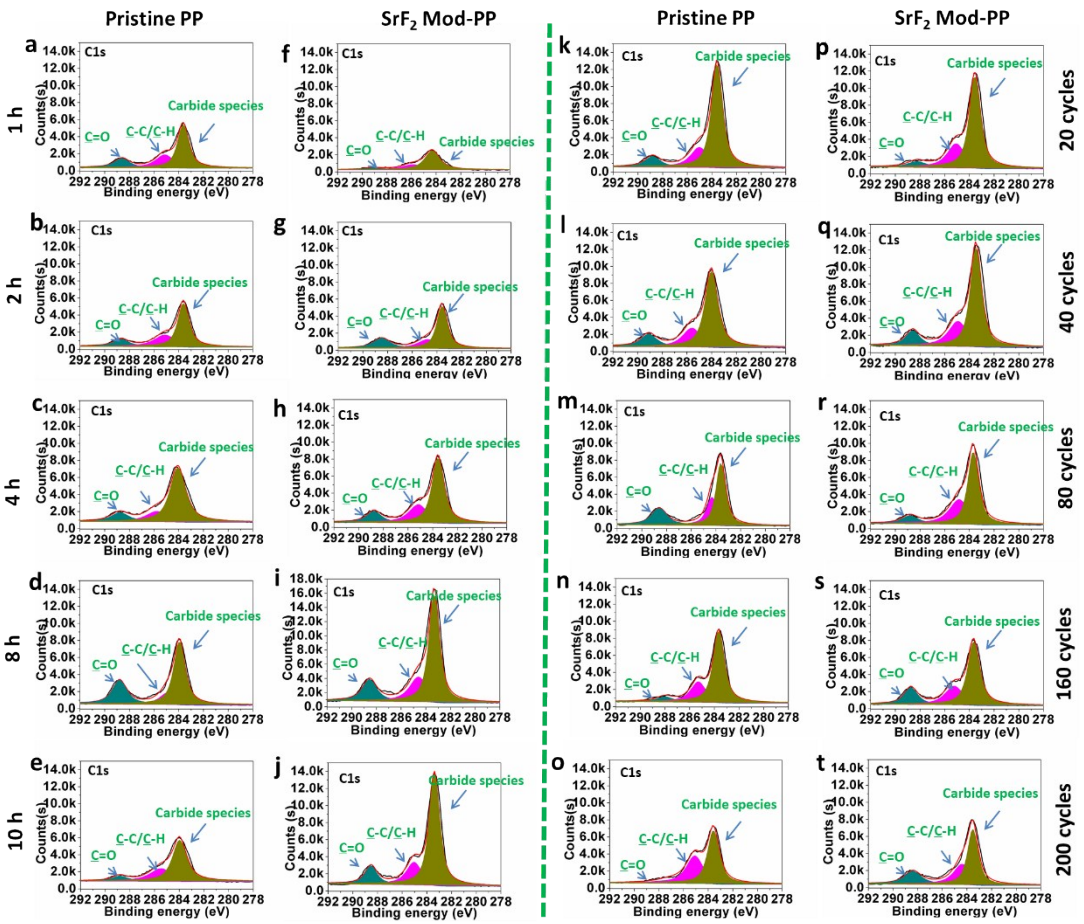


Figure S12. High resolution C1s spectra for the Li metal anodes under different Li deposition times of 1 h, 2 h, 4 h, 8 h and 10 h using **(a)** – **(e)** the pristine separator, and **(f)** – **(j)** SrF_2 coated separator. High resolution C1s spectra under different cycles of 20, 40, 80, 160 and 200 for using **(k)** – **(o)** the pristine separator, and **(p)** – **(t)** the SrF_2 coated separator.

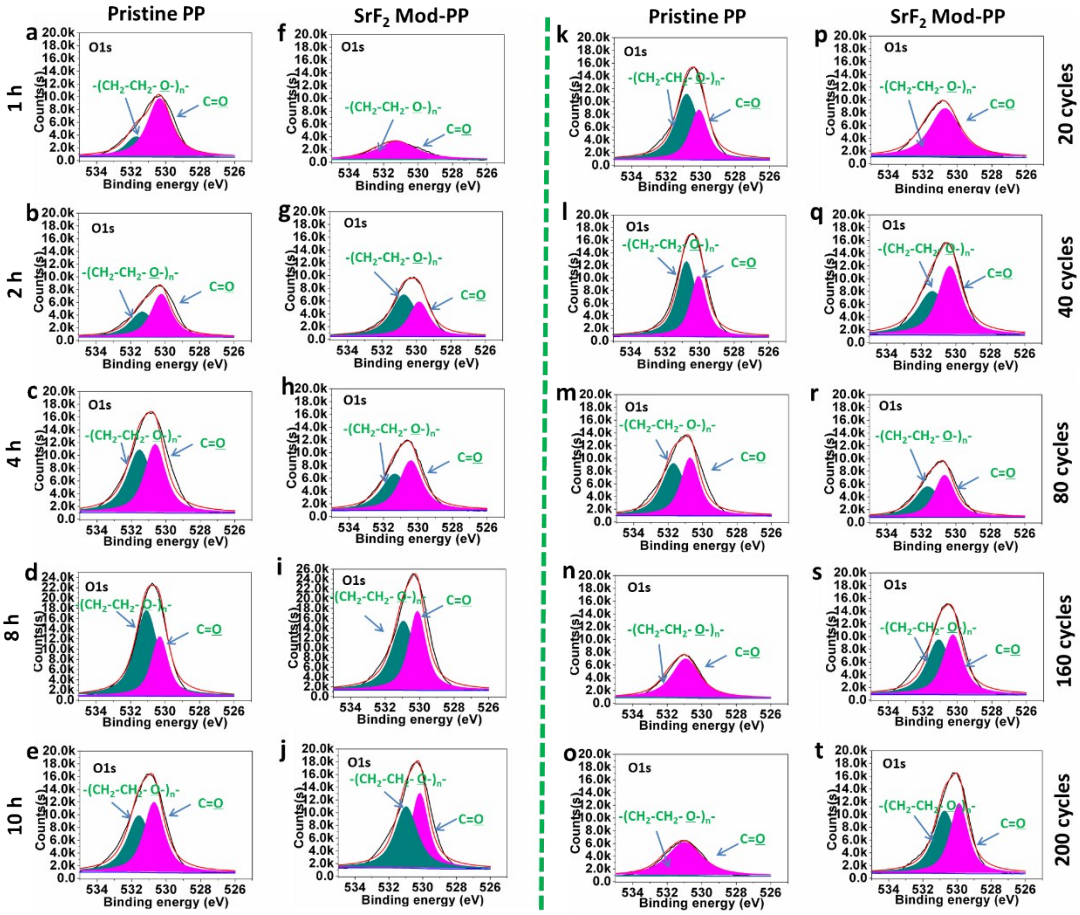


Figure S13. High resolution O1s spectra for the Li metal anodes under different Li deposition times of 1 h, 2 h, 4 h, 8 h and 10 h using **(a)** – **(e)** the pristine PP separator, and **(f)** – **(j)** the SrF_2 coated PP separator. High resolution O1s spectra under different cycles of 20, 40, 80, 160 and 200 for using **(k)** – **(o)** the pristine separator, and **(p)** – **(t)** SrF_2 coated separator.

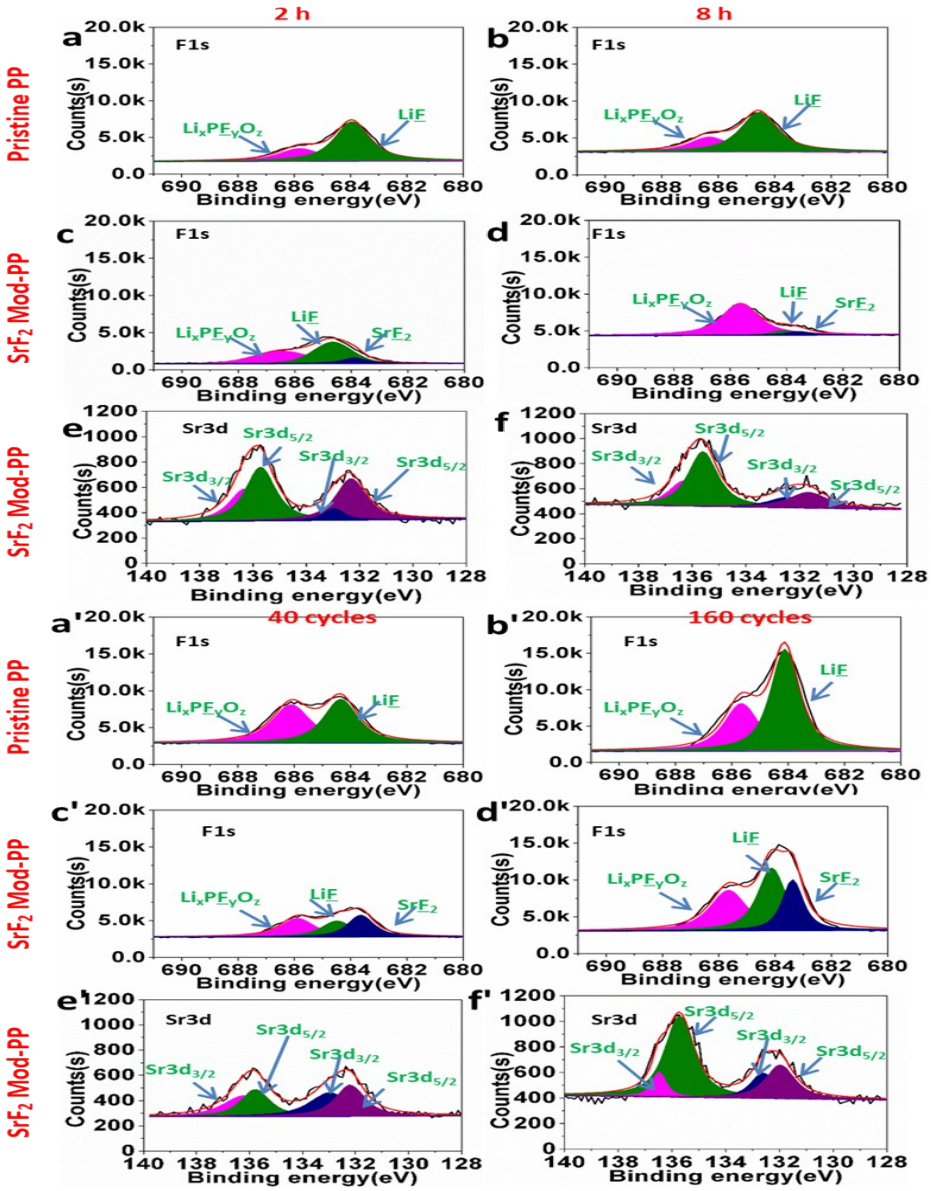


Figure S14. (a) - (f) and **(a') - (f')** High – resolution XPS spectra for post-cycled Li metal anodes with pristine vs. SrF_2 modified separator. **(a) - (i)** Testing conditions are the same as in **Fig. 3**, 20 mA g^{-1} for 2h and 10h. **(a') – (i')** Tested for 40 and 160 cycles at 200 mA g^{-1} for 2 hrs. **(a, a') – (b, b')** and **(c, c') – (d, d')** F1s spectra for baseline and SrF_2 coated, **(e, e') – (f, f')** Sr3d spectra for SrF_2 coated.

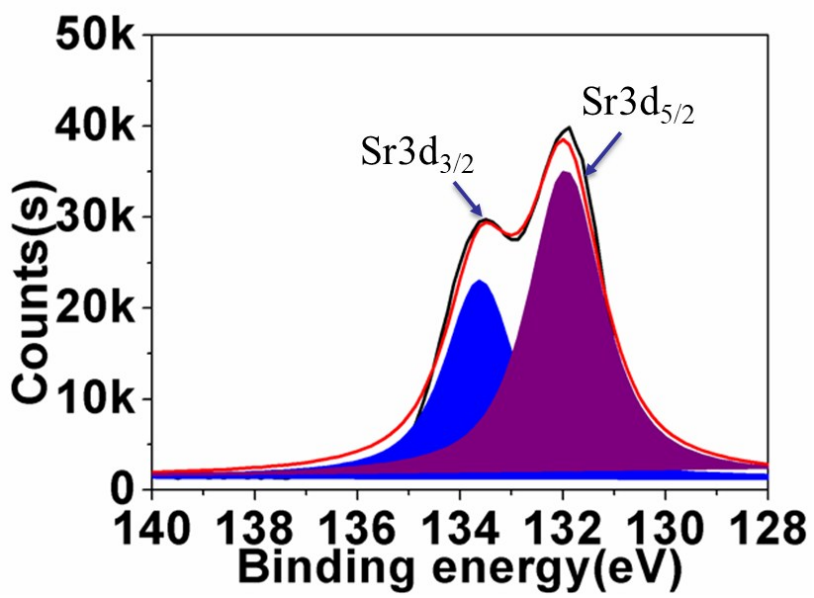


Figure S15. High resolution Sr3d spectrum for the as-prepared SrF_2 sample.