

Patterned Interlayer Enables a Highly-stable and Reversible Sodium Metal Anode for Sodium-metal Batteries

Chhail Bihari Soni¹, Sungjemmenla¹, Vineeth S.K.², C. Sanjay Kumar¹ and Vipin Kumar^{1,2*}

¹Department of Energy Science and Engineering, Indian Institute of Technology Delhi, Hauz Khas, New Delhi, 110016, India

²University of Queensland–IIT Delhi Academy of Research (UQIDAR), Indian Institute of Technology Delhi, Hauz Khas, New Delhi 110016, India

*Email- vkumar@dese.iitd.ac.in

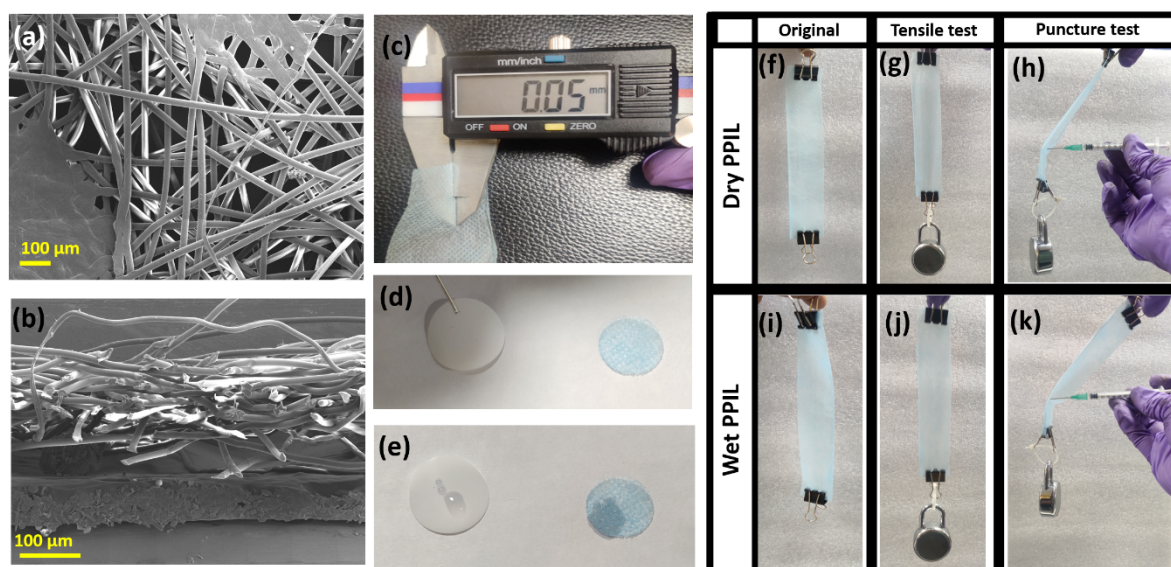


Fig. S1 (a, b) Micro-structure of PPIL fibre, cross-sectional image of as received PPIL after treatment (c) Thickness of PPIL fibre after applying a maximum pressure to compress (d) Celgard (PP-PE-PP) and PPIL fibre in dry condition (e) Celgard and PPIL fibre after putting 20 μ l of electrolyte (f) original PPIL fibre without loading (g) PPIL fibre in tensile test with a mass of 70 gm (h) puncture test on PPIL fibre. All these tests are in dry condition of PPIL fibre (i) original PPIL fibre without loading (j) PPIL fibre in tensile test with a mass of 70 gm (k) puncture test on PPIL fibre. These all tests are in a wet condition of PPIL fibre

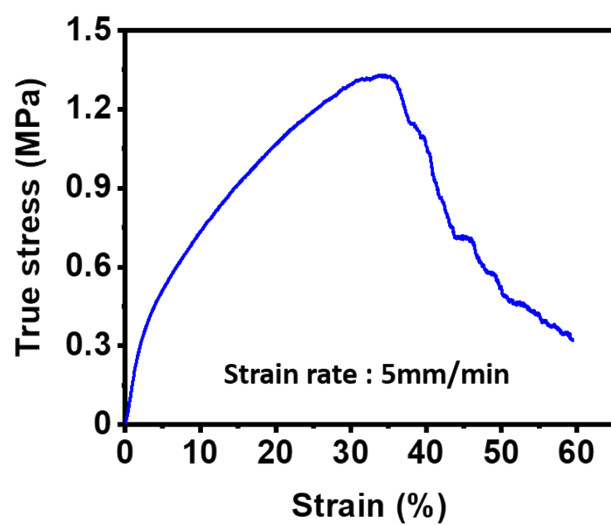


Fig. S2 Static tensile test of PPIL interlayer at 5 mm/min strain rate

Table-S1 Results of tensile test

E_{modulus} (MPa)	σ_{max} (MPa)	% ϵ at F_{max}	σ_{break} (MPa)	% ϵ at break
9.4706	1.329979	33.7436	0.3189	59.4766

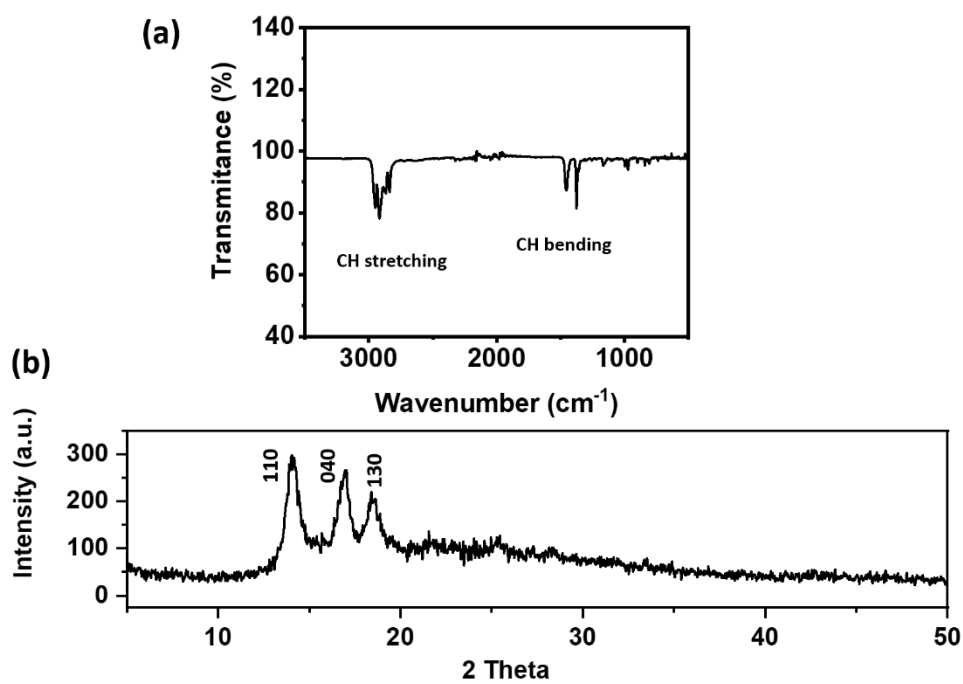


Fig. S3 (a) FTIR spectrum of PPIL fibre (b) XRD spectra of PPIL fibre

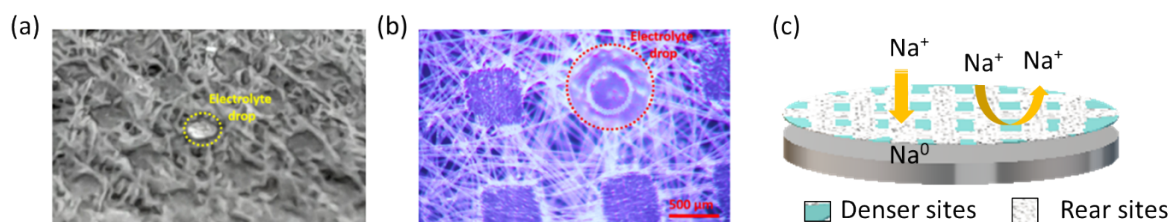


Fig. S4 (a) Digital image of the electrolyte droplet over the PPIL patch, and (b) Optical image of the electrolyte droplet over the PPIL patch (c) schematic of Na deposition through rear

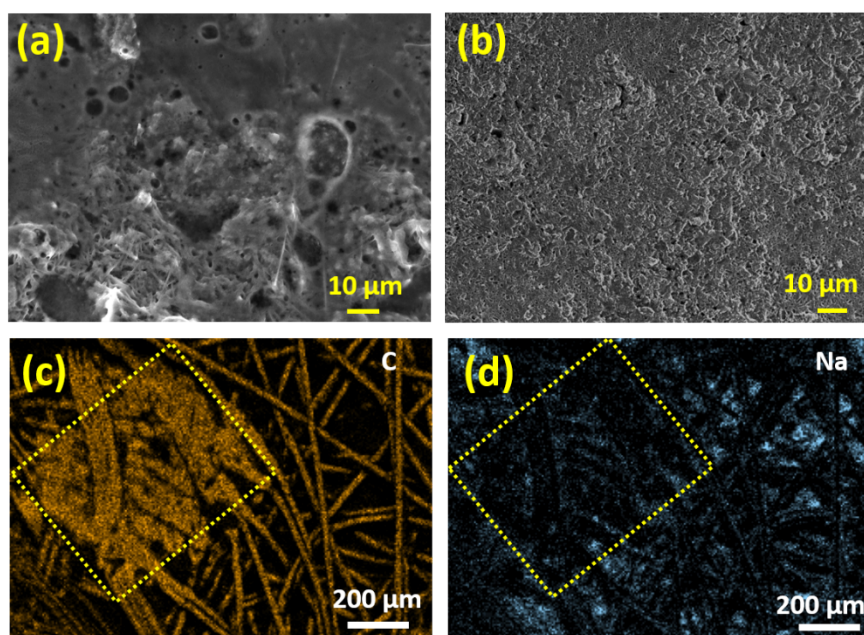


Fig. S5 Morphological analysis of Na metal surface after 10 cycles at 10 mA cm^{-2} current density and 1 mA h cm^{-2} capacity (a) Na metal surface without PPIL (b) Na metal surface after removing the PPIL interlayer, showing smooth and uniform morphology (c, d) Na metal surface with PPIL showing guided deposition through. All these tests are in ether-based electrolyte.

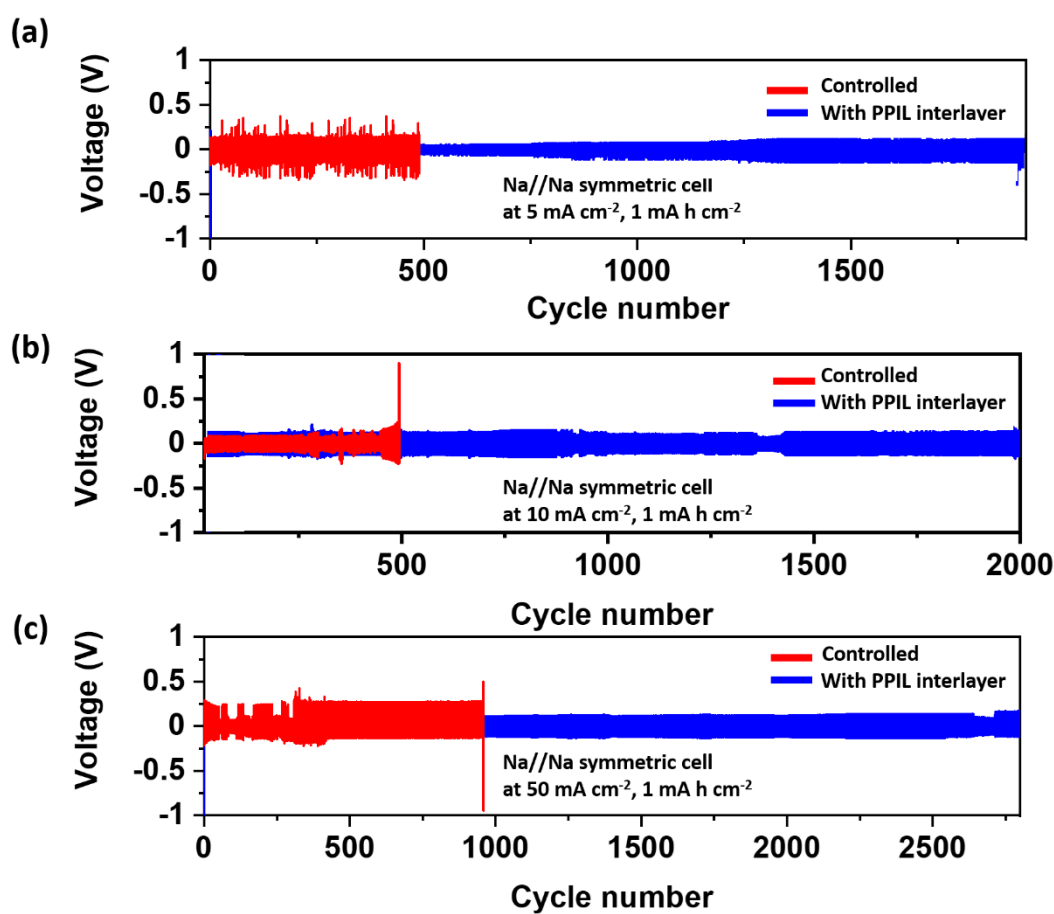


Fig. S6 Galvanostatic cyclic performance of controlled and PPIL protected Na//Na symmetric cell, at (a) 5 mA cm^{-2} (b) 10 mA cm^{-2} and (c) 50 mA cm^{-2} in ether-based electrolyte

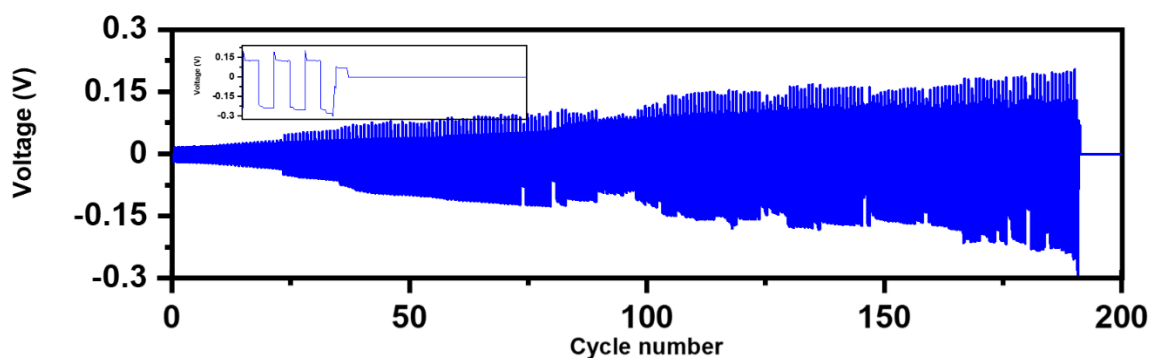


Fig. S7 Stripping/plating sodium in sodium symmetric cell configuration (Na//Na) with PPIL (2 layers) as a diaphragm at a current density of 10 mA cm^{-2} and a capacity of 1 mAh cm^{-2} . Inset shows an enlarged view of the voltage profile.

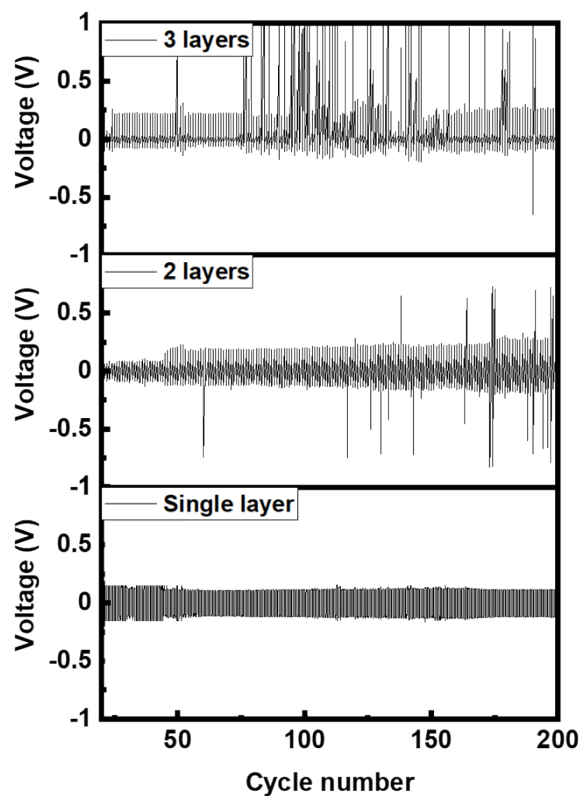


Fig. S8 Galvanostatic cycling of Na//Na symmetric cell with multiple layers of PPIL protective interlayer at 10 mA cm^{-2} current density and 1 mA h cm^{-2} capacity. It was noticed that the single layer of PPIL was optimum with uniform deposition and comparatively less overpotential.

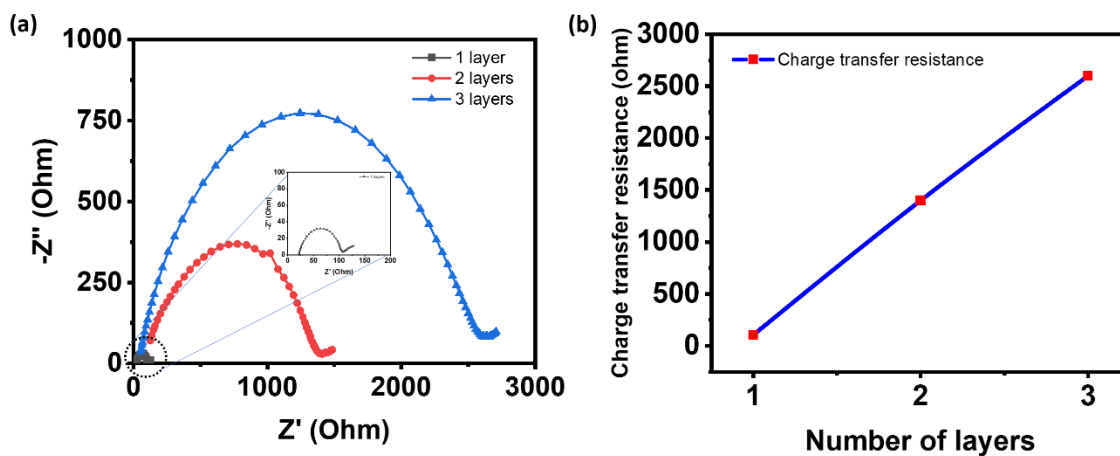


Fig. S9 (a) EIS of Na//Na symmetric cell in PPIL protected with multiple layers, in carbonate based electrolyte. (b) Charge transfer resistance with respect to multiple layers of PPIL

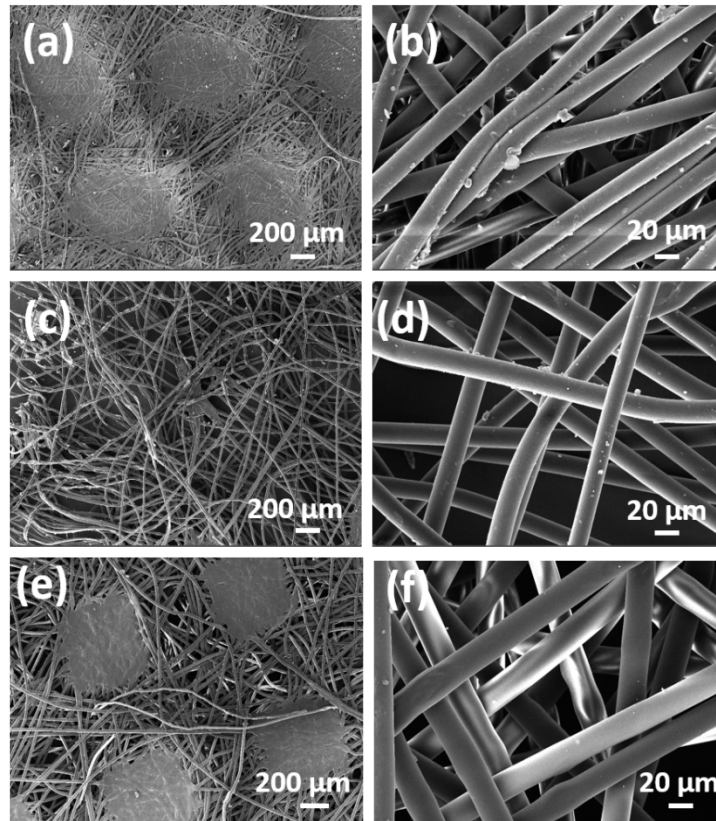


Fig. S10 FESEM images of PPIL fibre (a, b) dense patches (c, d) without patches (e, f) optimum patches

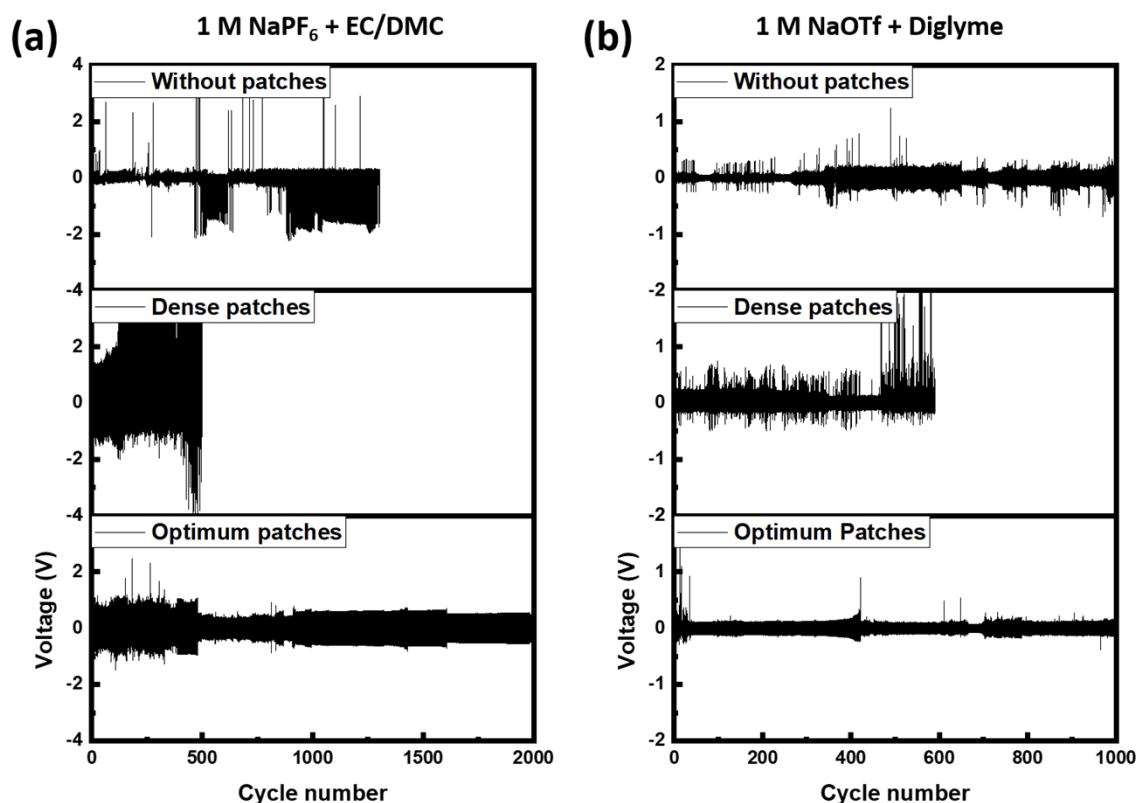


Fig. S11 Galvanostatic cycling performance of Na//Na symmetric cell at 10 mA cm⁻² current density and 1 mA h cm⁻² capacity for three different types of fibres, i.e., without patches, dense patches and optimum patches in (a) Carbonate and (b) Ether-based electrolyte.

In the Randles-Sevcik equation, the Na-ion apparent diffusion coefficient (D) can be computed¹.

$$I_p = 0.4463nFAC\left(\frac{nFvD}{RT}\right)^{1/2}$$

Where, F is Faraday's constant, 96455 C mol⁻¹L, n is the number of mol participating in the reaction (for 1 redox peak, n is generally 1), A is the working electrode area (1 cm²), C is concentration of sodium ions in the working electrode (mol cm⁻³), v is scan rate, R is gas constant (8.314 J mol⁻¹ K⁻¹), T is temperature (298.15 K) and I_p is the peak current.

Table S(T1) Parameters used to calculate the diffusion coefficient

Scan rate (v) (mV s ⁻¹)	square root of scan rate	Peak current (mA) (With PPIL)	Peak current (mA) (Without PPIL)
0.5	0.7071	0.8913	0.6346
1.5	1.2247	1.3591	1.0910

2	1.4142	1.6056	1.2419
3	1.7320	2.0275	1.6911

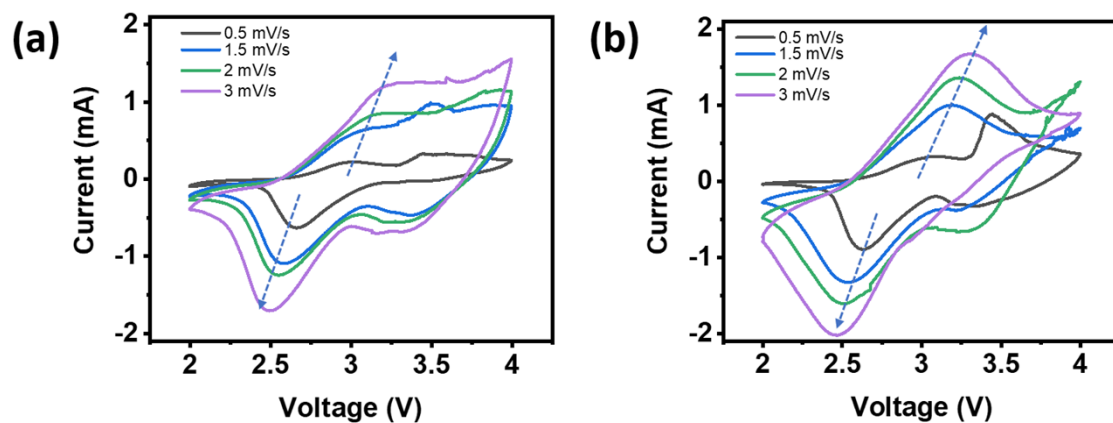


Fig. S12 Cyclic voltammetry of Na//PB full cells at different scan rates (a) without PPIL (b) with PPIL

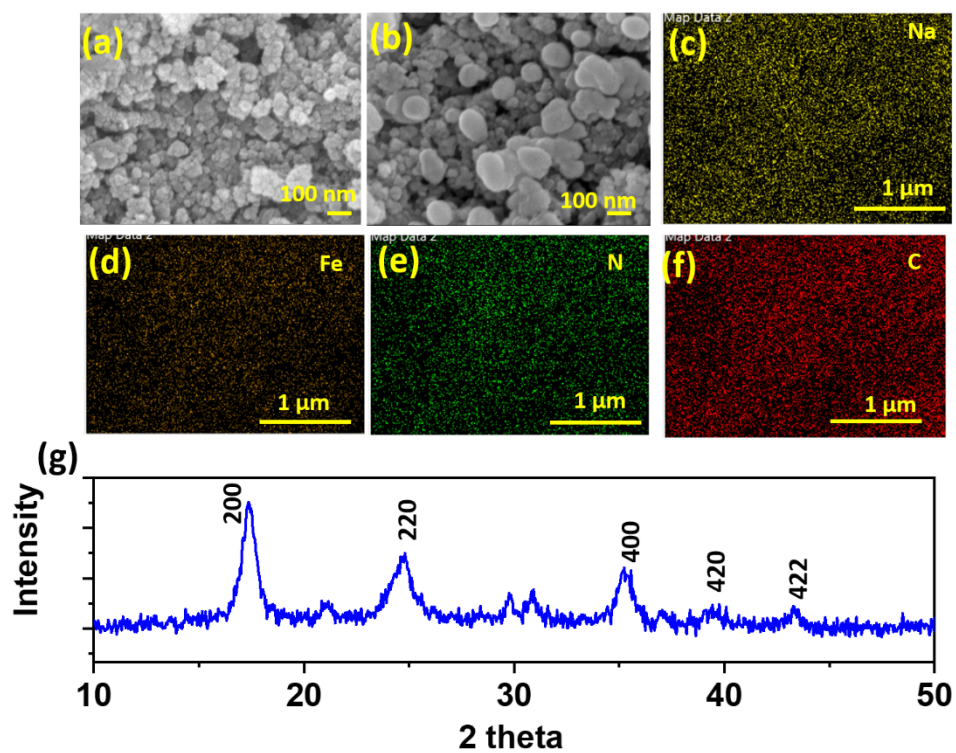


Fig. S13 (a, b) FESEM images of the as-synthesized PB material (c-f) EDX mapping of the elements present in the PB (g) XRD spectrum of as-synthesized PB powder

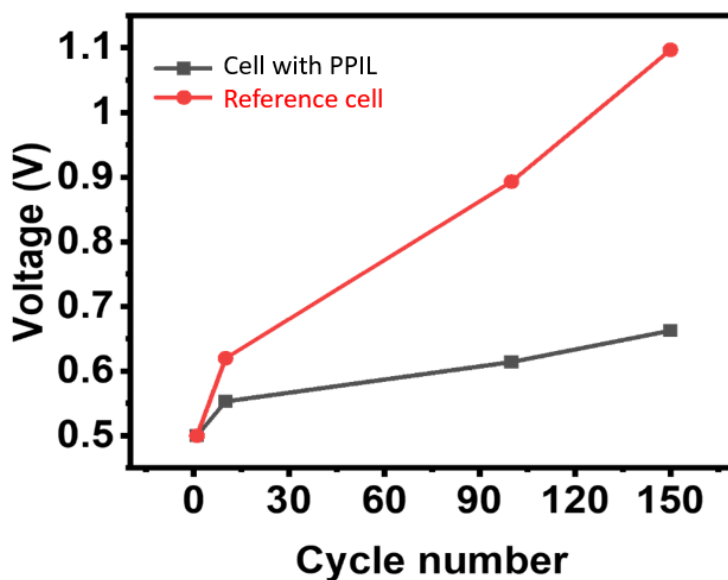


Fig. S14 Polarization of controlled and PPIL protected cell for different cycle number in ascending order

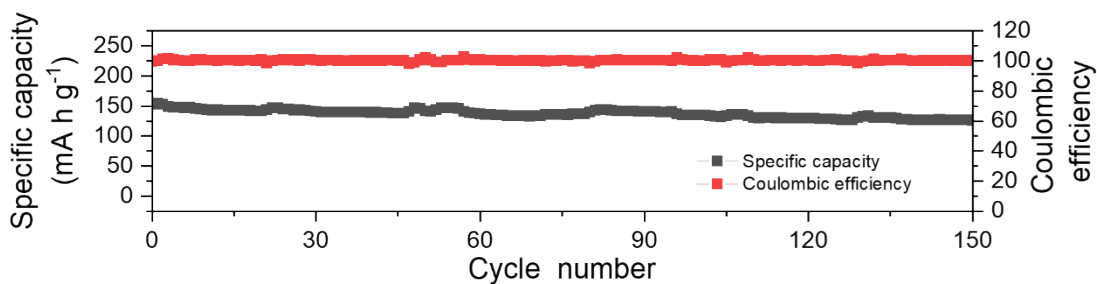


Fig. S15 Cyclic performance of Na//PB full cell with PPIL protective layer at 500 mA g⁻¹ in carbonate-based electrolyte

References:

- 1 H. Li, Y. Wang, J. Jiang, Y. Zhang, Y. Peng and J. Zhao, *Electrochim. Acta*, 2017, **247**, 851–859.