Supporting Information for
Constructing Dynamic Na^+ Transport Channels in RT Na-S Battery Separators to Suppress Polysulfides Shuttling and Accelerate Reaction Kinetics

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Experimental instruments and methods

1. Characterizations

The morphologies and structures of the MIL-121 and MIL-121/Na were observed by FESEM (JEOL JSM-7800F) and X-ray diffractometer (Bruker). The powder of MIL-121 and MIL-121/Na was determined by a Thermogravimetric analyzer (TGA, Q50, USA). Raman characterization was performed using a LabRAM HR Evolution (Horiba) Raman microscope with an excitation laser of 532 nm. The analyses for the specific surface area and the pore-size distribution were performed on Micromeritics under N_2 atmosphere by JW-BK-400.

2. Electrochemical measurements

Tafel curve (Tafel), Cyclic voltammetry (CV), Linear sweep voltammetry (LSV), Electrochemical impedance spectroscopy (EIS) and i-t curve were measured on a CHI 760E electrochemical workstation. A battery test system obtained galvanostatic charge-discharge (GCD) curves (LAND CT2001A, China).

3. Ionic conductivity

Ionic conductivity was measured by electrochemical AC impedance spectroscopy (EIS) in the frequency range from 1 kHz to 0.1 Hz with an AC amplitude of 100 mV via a CHI760E electrochemical workstation. The following equation calculated the ionic conductivity:

$$\sigma = \frac{L}{(R * A)}$$

L is the measured thickness, S is the facing area of the two stainless steel foils, and R is the horizontal intercept obtained from electrochemical impedance spectroscopy.

4. Na ion transference number

Na ion transference number was measured utilizing an amperometric technique with an applied DC voltage of 20 mV and EIS measurement in a Na symmetric cell based on MIL-121/Na, MIL-121 and GF. The amperometric technique tested EIS before and after the DC polarisation. The t_{Na^+} value was calculated according to Bruce's equation:

$$t_{Na^+} = \frac{I_S(\Delta V - I_0 R_0)}{I_0(\Delta V - I_S R_S)}$$

where ΔV is the polarization voltage (10 mV), I_0 is the initial current, which is the steady state current, R_0 is the initial resistance, and R_s is the steady state total resistance.

5. Activation energy

The activation energy was calculated based on the Arrhenius relation with a linear fitting coefficient of over 0.99 after collecting electrochemical impedance at different temperatures. The Arrhenius relation:

$$\frac{1}{R_a} = Aexp(-\frac{E_a}{RT})$$

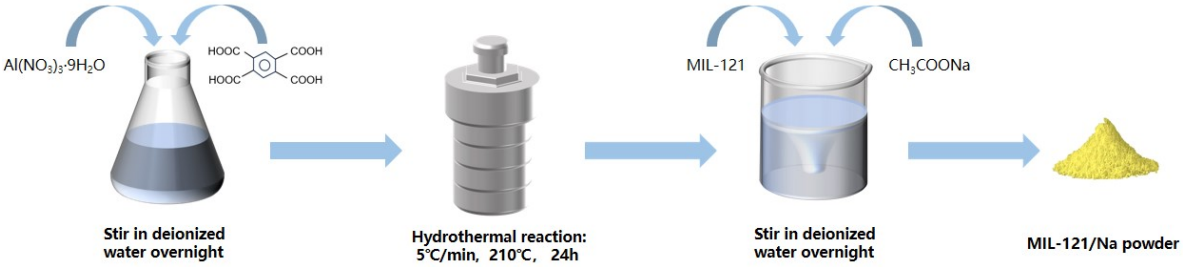

Figure S1. Schematic diagram of MIL-121 and MIL-121/Na synthesis process.

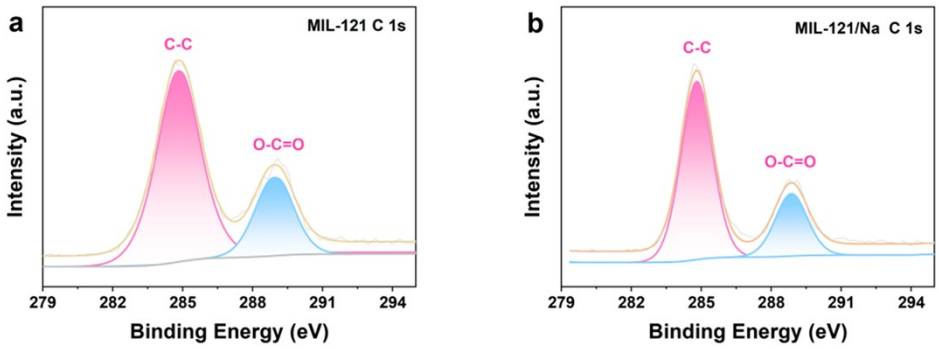

Figure S2. XPS spectra of C 1s of MIL-121 and MIL-121/Na.

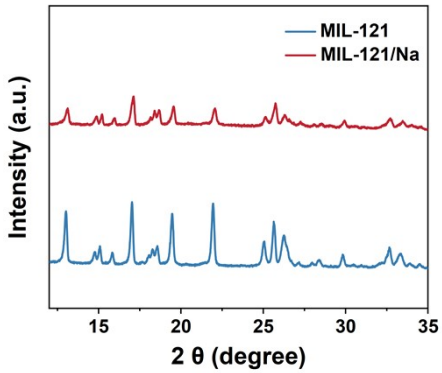

Figure S3. XRD of MIL-121 and MIL-121/Na.

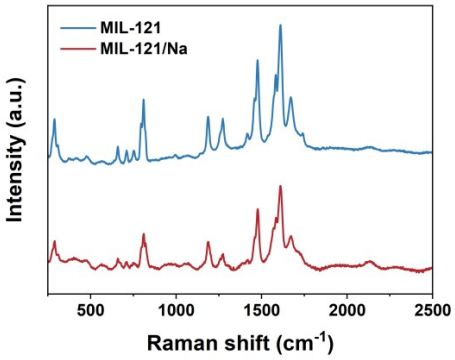

Figure S4. Raman of MIL-121 and MIL-121/Na.

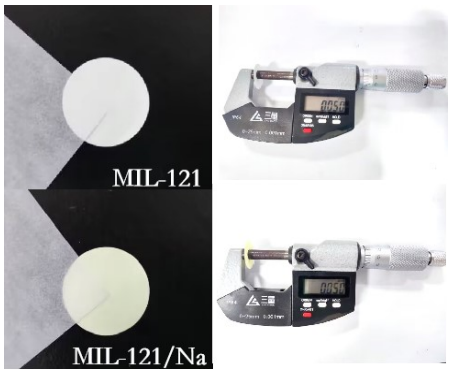

Figure S5. Thickness and color of MIL-121 and MIL-121/Na.

Figure S6. Image of MIL-121/Na separator flexibility.

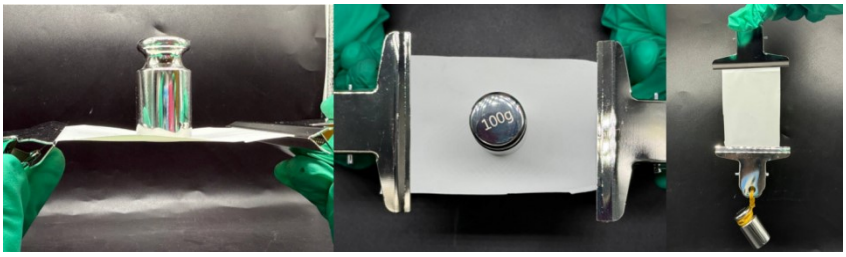

Figure S7. Tensile test of MIL-121.

Figure S8. Tensile test of MIL-121/Na.

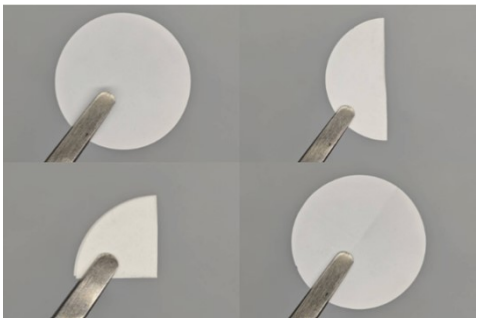

Figure S9. Digital images of the MIL-121 separator, single, double folded, and recovered.

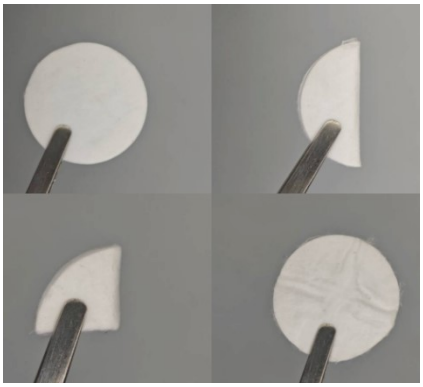

Figure S10. Digital images of the GF separator, single, double folded, and recovered.

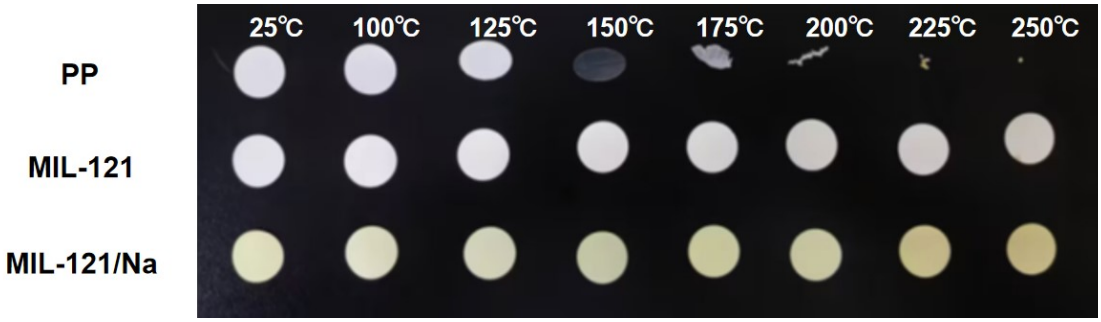

Figure S11. The heat shrinkage performance of PP, MIL-121, and MIL-121/Na.

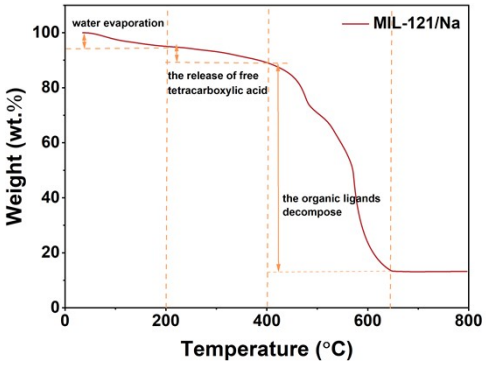

Figure S12. TGA of MIL-121/Na.

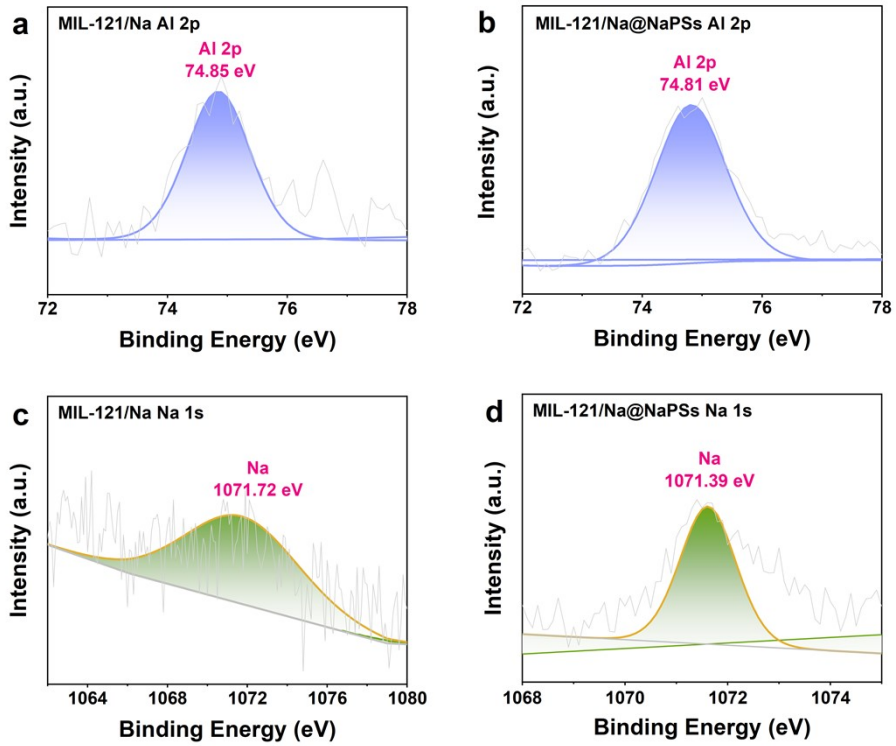

Figure S13. XPS spectra of a) Al 2p of MIL-121/Na, b) Al 2p of MIL-121/Na@NaPSs, c) Na 1s of MIL-121/Na and d) Na 1s of MIL-121/Na@NaPSs.

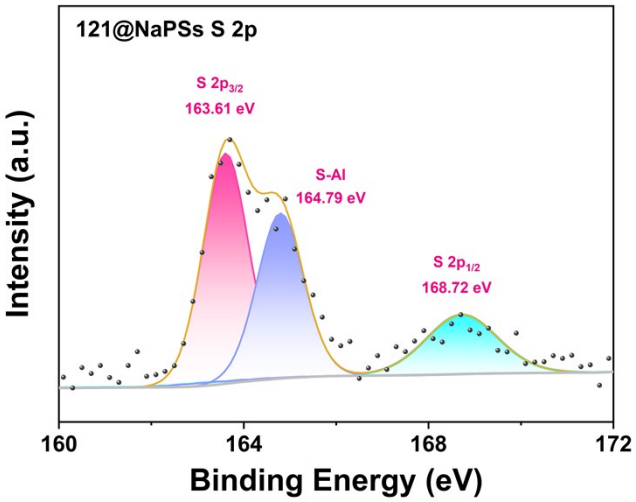

Figure S14. XPS spectra of S 2p of MIL-121 separator after the shuttle experiment test.

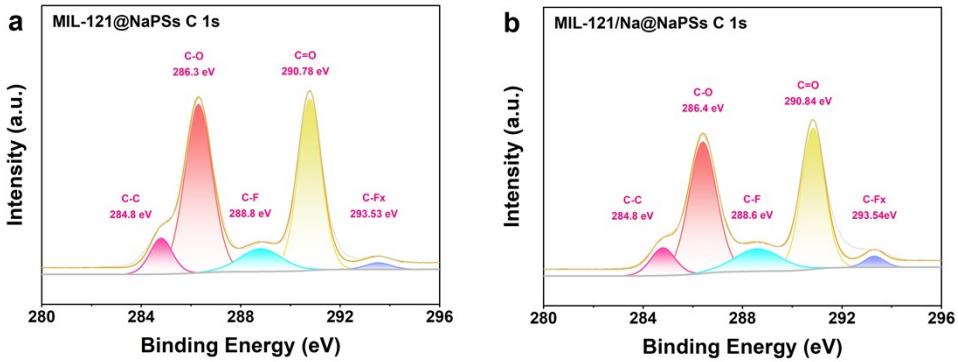

Figure S15. XPS spectra of C 1s of a) MIL-121@NaPSs and b) MIL-121/Na@NaPSs.

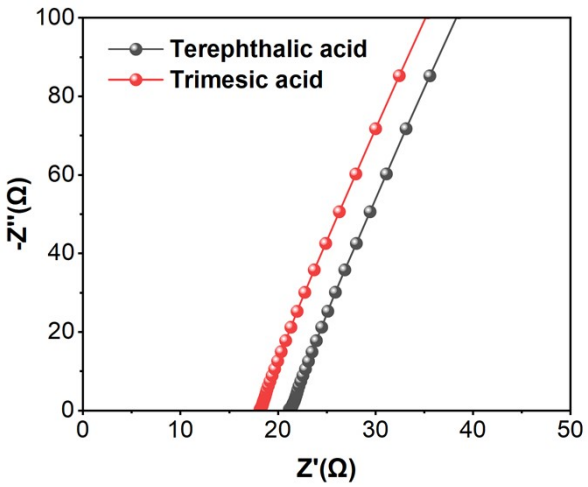

Figure S16. Ion conductivity of separators prepared with terephthalic acid and trimesic acid as constituents.

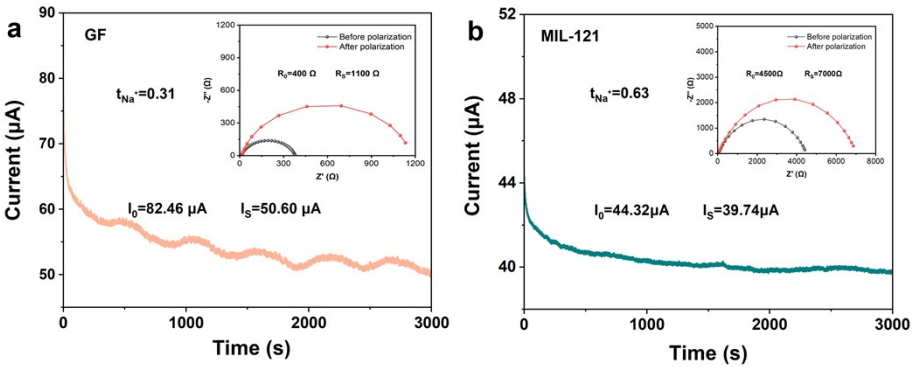

Figure S17. Electrochemical impedance spectra and direct-current polarisation curves of symmetric cells based on a) GF and b) MIL-121 separator.

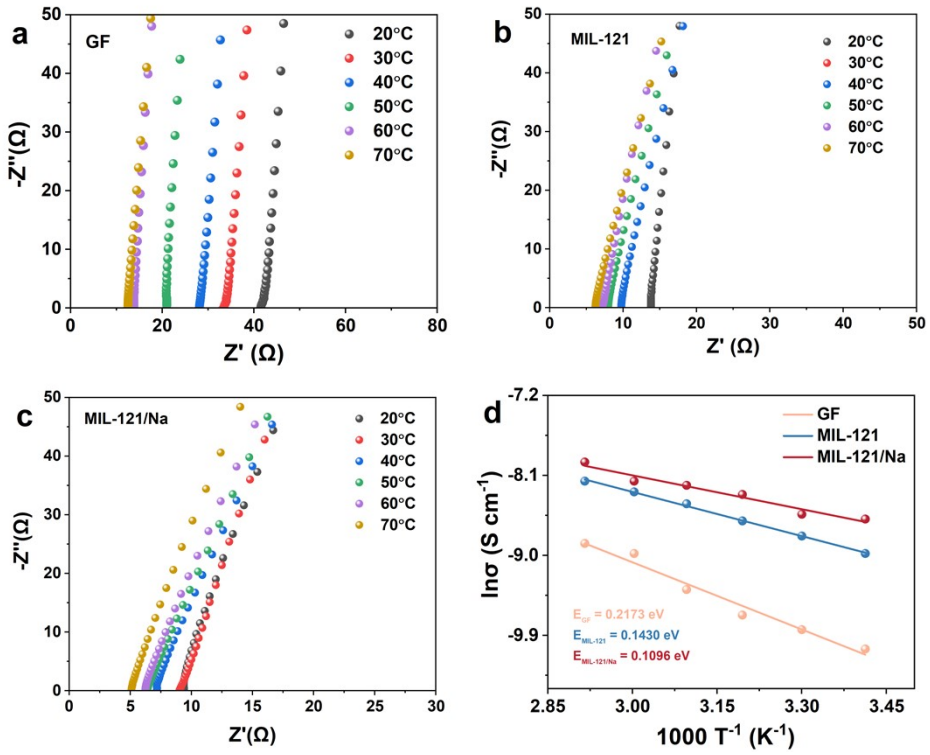

Figure S18. Electrochemical impedance spectra of a) GF b) MIL-121 and c) MIL-121/Na obtained at different temperatures. d) Arrhenius curves used to deduce the activation energy values for three membranes.

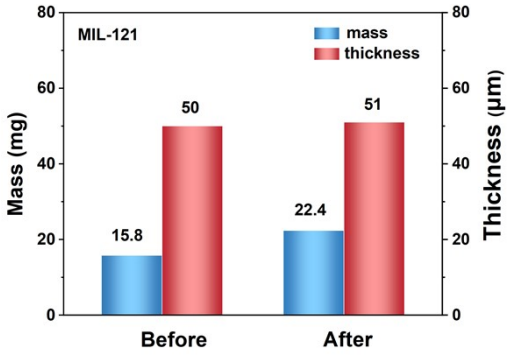

Figure S19. Variation of MIL-121 separator in mass and thickness after immersion in electrolyte.

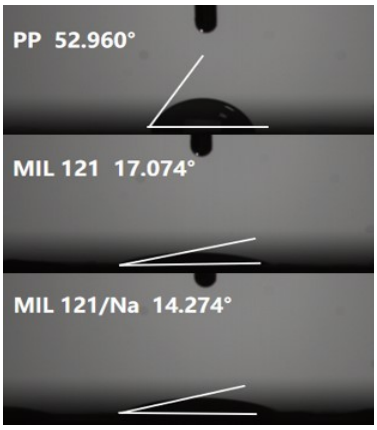

Figure S20. Contact angle test of PP, MIL-121, and MIL-121/Na separator.

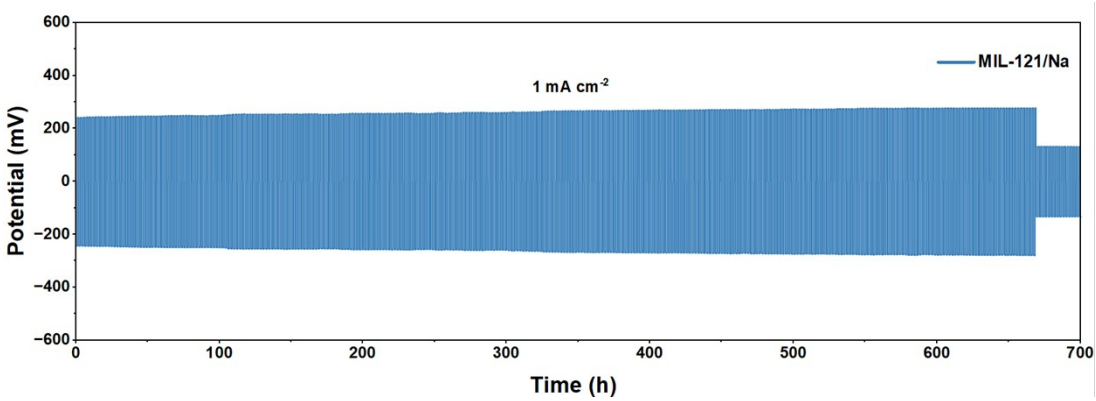

Figure S21. The long cycle of symmetrical cell with MIL-121/Na separator at 1 mA cm^{-2} current density.

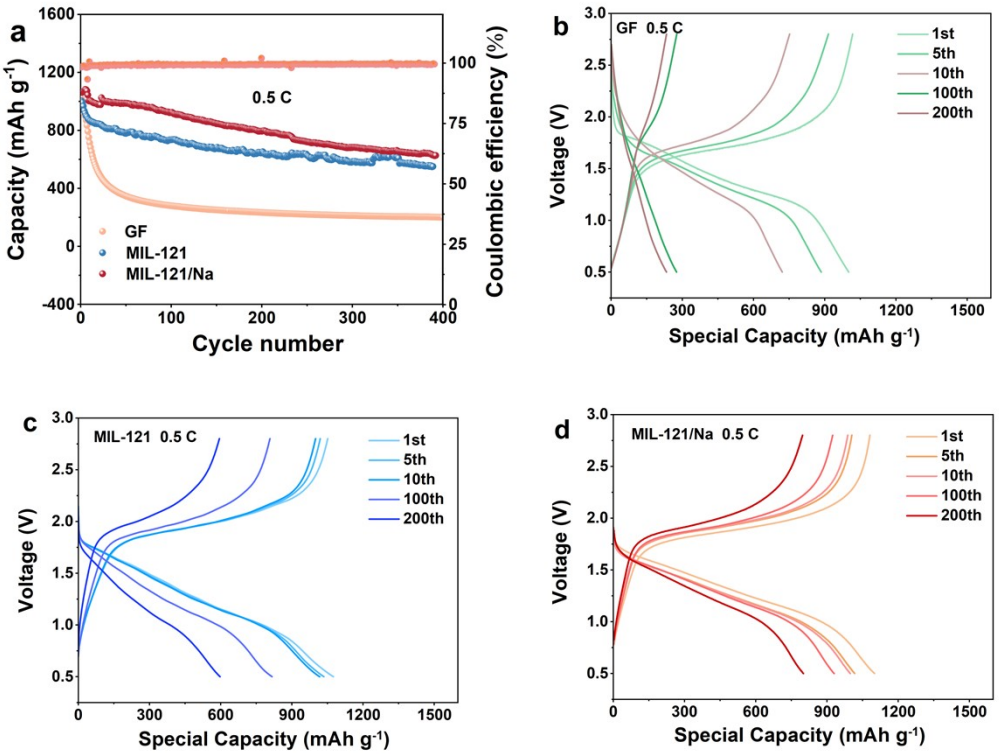

Figure S22. a) GF, MIL-121, and MIL-121/Na long cycle curves at 0.5C, b-d) and the specific capacity-voltage curves of the three at the 1st, 5th, 10th, 100th, and 200th cycles.

Figure S23. A pouch cell with a MIL-121/Na separator.

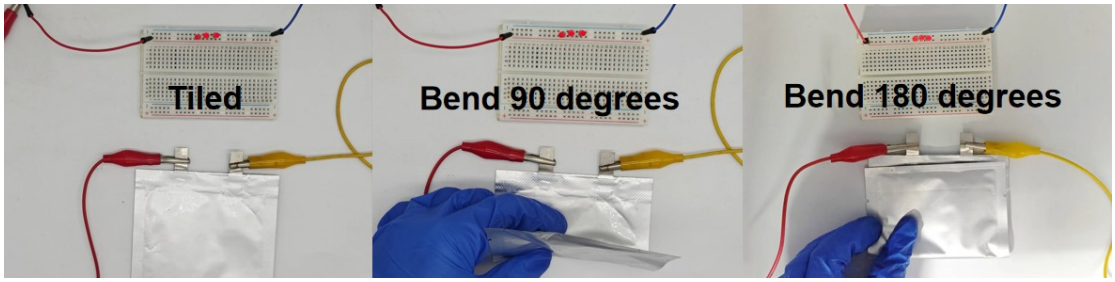

Figure S24. The pouch cell lights up the small bulb under tiling, bending 90 degrees and bending 180 degrees respectively.

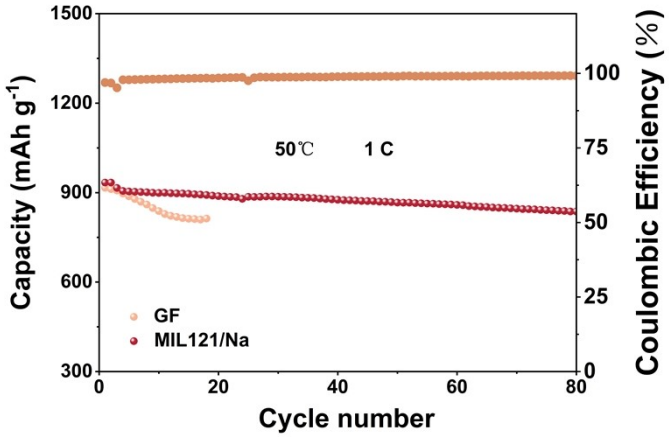

Figure S25. Long cycle curves of sodium-sulfur batteries with GF and MIL-121/Na separators at 1 C rate and 50 ℃.