

Supplementary Information:

Impact of 2D Layered VS_2 on the Electrochemical Properties of Gel Polymer Electrolytes for Sodium-Ion Battery Applications

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1. Preparation route of gel polymer electrolytes:

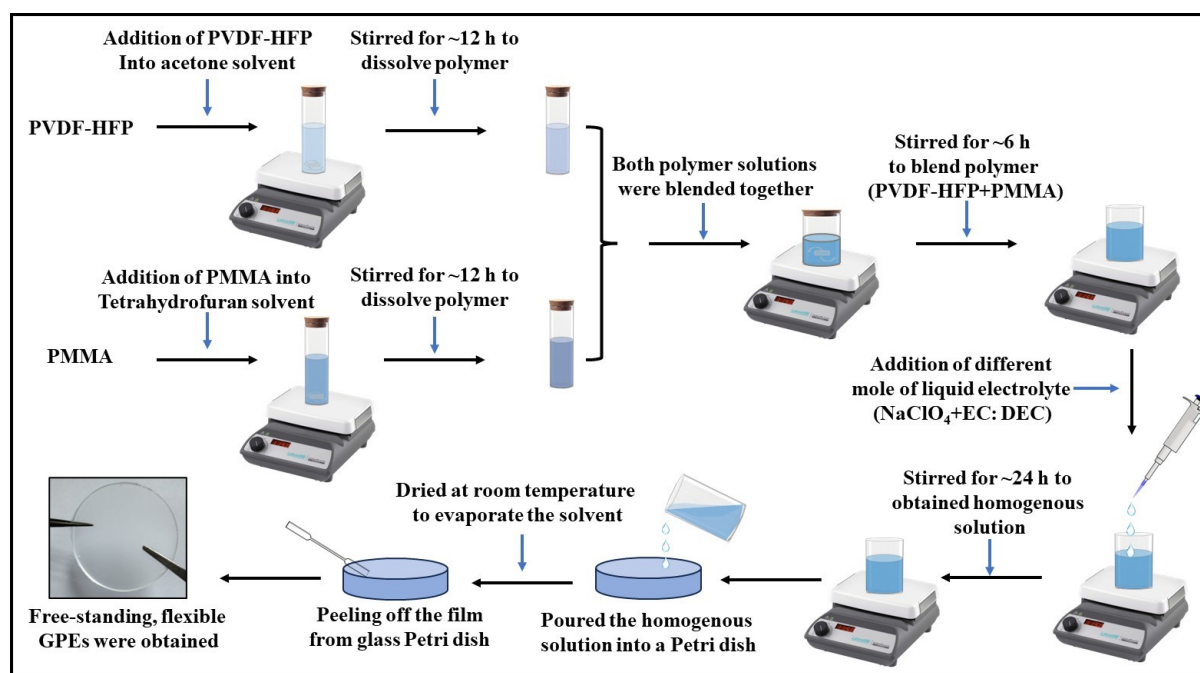


Fig. S1 Schematic illustration of the preparation of gel polymer electrolytes.

2. Preparation route of composite gel polymer electrolytes:

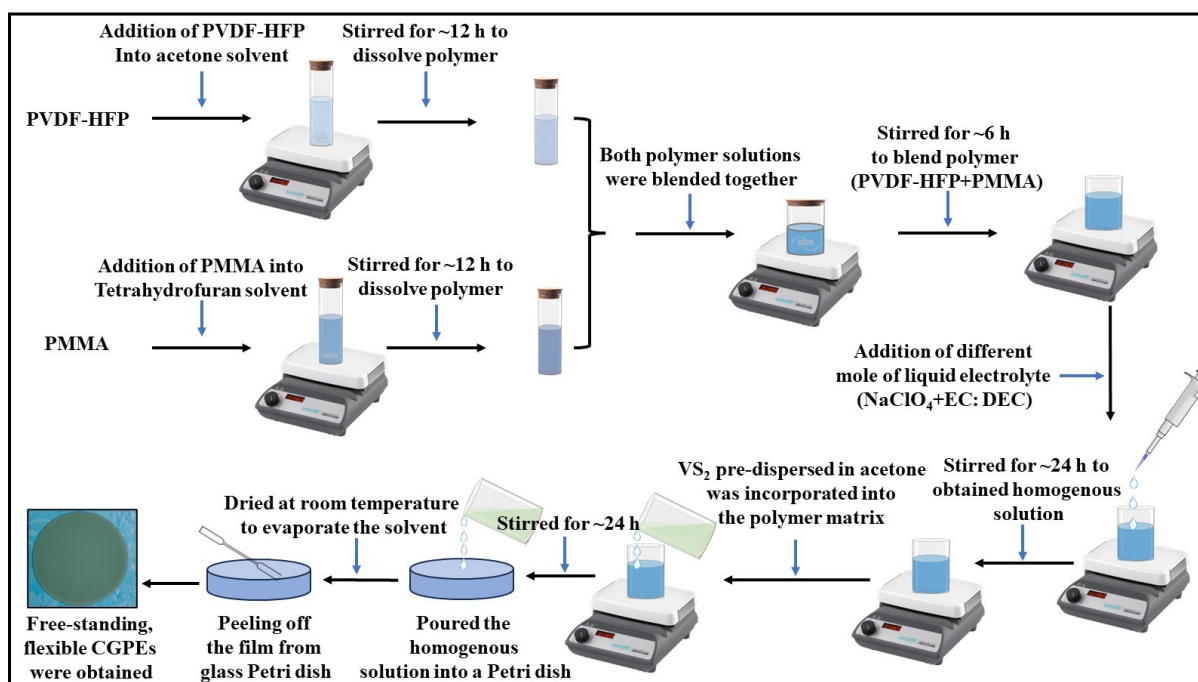


Fig. S2 Schematic illustration of the preparation of composite gel polymer electrolytes.

3. **Fabrication of electrodes and Na ion coin cell:** The figure S3 shows the schematic of hard carbon anode preparation and coin-cell assembly. Hard carbon, PVDF, and carbon black were mixed in NMP to form a homogeneous slurry, coated onto copper foil, and vacuum-dried at 80 °C for 12 h. The electrode was punched into 14 mm discs and assembled into CR2032 coin cells with sodium metal and a polymer electrolyte inside an argon-filled glovebox.

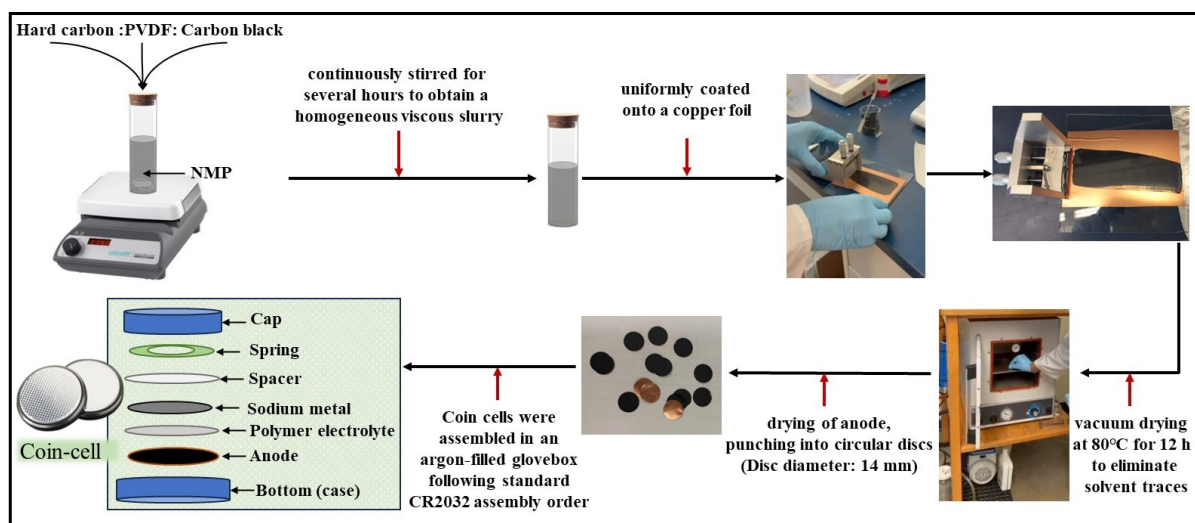


Fig. S3 Schematic of anode and coin cell fabrication

Table S1: FTIR deconvolution of GPEs in the 1860–1700 cm⁻¹ region with peak positions and intensities.

	Peak position	Intensity	Peak position	Intensity	Peak position	Intensity	Peak position	Intensity
Polymer electrolytes	Free EC (C=O stretching)		Na ⁺ -EC coordinated carbonyl		Na ⁺ -DEC coordinated carbonyl			PMMA C=O (free / Na ⁺ -coordinated)
GPE-1	1814	0.0011	1793	0.0051	1763	0.0058	1734	0.0029
GPE-2	1813	0.0037	1794	0.0094	1769	0.0097	1743	0.0029
GPE-3	-----	-----	1798	0.023	1765	0.0316	1734	0.0090
GPE4	-----	-----	1803	0.372	1769	0.592	1735	0.0623
GPE-5	-----	-----	1802	0.3500	1768	0.5760	1731	0.1016
GPE-6	-----	-----	1803	0.2300	1769	0.3981	1735	0.0587
CGPE-3			1801	0.3716	1767	0.6690	1737	0.0719

Table S2: EIS fitting parameters of the HC|CGPE-3|Na cell.

The chi-squared value of the fitting is **0.0023**.

Element	Value	Unit	Error %
R _b	13.85	Ω	5.9
CPE-T	5.28E-5	F.s ^(P-1) 0.5 ≤ P < 0.8	30.1
CPE-P	0.553	– (dimensionless)	5.0
R _{ct}	94.3	Ω	5.6
W _s -R	0.232	Ω	43305
W _s -T	0.015	s	109.4
W _s -P	0.999	– (dimensionless)	32.19

4. The detailed crystallinity calculations derived from both XRD and DSC analyses have been provided below for clarity.

XRD Analysis:

The crystallinity % calculated using the following equation

$$\chi_c = \frac{A_c}{A_c + A_A} \times 100\% \quad (1)$$

Here, A_c corresponds to the area associated with the crystalline peak, whereas A_A refers to the area attributed to the amorphous peaks. The peak area, total area and crystallinity % given in the table.

S. No.	Sample	Integral crystalline peak area	Integral total area (crystalline + amorphous)	Crystallinity %
1.	PVDF-HFP	5.31218	14.48221	36.68
2.	PMMA	2.49755	16.19688	15.42

3.	PVDF-HFP: PMMA	3.76892	20.42777	18.45
4.	GPE-1	3.05661	14.39764	21.23
5.	GPE-2	3.01797	18.30187	16.49
6.	GPE-3	2.24950	17.91099	12.56
7.	GPE-4	1.67806	18.04373	9.30
8.	GPE-5	1.75699	20.79286	8.45
9.	GPE-6	4.15718	13.85729	30.00
10.	CGPE-1	1.75083	18.16223	9.64
11.	CGPE-2	1.61869	18.90997	8.56
12.	CGPE-3	1.87672	22.47578	8.35
13.	CGPE-4	1.64951	17.62307	9.36

DSC Analysis:

The crystallinity from DSC was determined using:

$$X_c(\%) = \frac{\Delta H_m}{\Delta H_m^o \times \omega_p} \times 100 \quad (2)$$

Where, ΔH_m is the melting enthalpy (J/g), ΔH_m^o is the heat of fusion of 100% crystalline Poly(vinylidene fluoride-co-hexafluoropropylene) = 104.5 J/g, ω_p is weight fraction of the crystalline polymer.

The melting enthalpy was obtained from:

$$\Delta H_m = \frac{\text{melting peak area (mJ)}}{\text{sample mass (mg)}}$$

The melting peak areas of GPE-5 and CGPE-3 were calculated using Origin software. The sample mass was already included during the DSC measurement, then the reported peak area is already normalized. From the DSC thermograms, the melting peak area was determined to be 11.42 for GPE-5 and 7.42 for CGPE-3, corresponding to melting enthalpy (ΔH_m) values of 11.42 J/g and 7.42 J/g, respectively. For the preparation of the GPE, the polymer blend consisted of 90 wt% PVDF-HFP and 10 wt% PMMA. Since PMMA is an amorphous polymer and does not exhibit melting enthalpy, only PVDF-HFP contributes to the heat of fusion. Therefore, a weight fraction (ω_p) of 0.9 was used in the crystallinity calculation. Accordingly, the calculated crystallinity values for GPE-5 and CGPE-3 are 12.14% and 7.80%, respectively. These values are in good agreement with the crystallinity results obtained from XRD analysis.

5. LSV curve of GPE-5, and specific capacity versus cycle number curve:

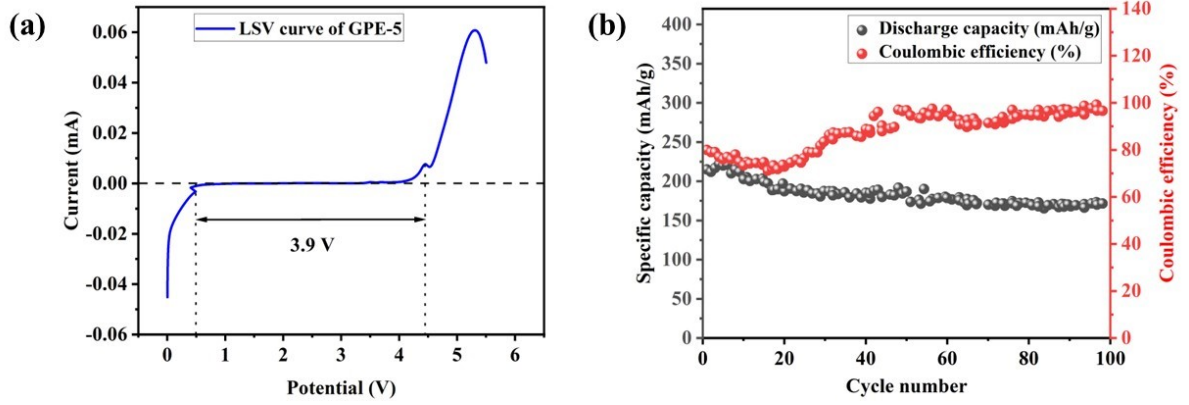


Fig. S4 (a) LSV curve of GPE-5, (b) specific capacity versus cycle number derived from the GCD measurements.

Table S3. Bulk resistance (R_b), ionic conductivity (σ), and thickness values of the prepared samples.

Sample	R_b (Ω)	σ (S/cm)	Thickness (mm)
GPE-1	5.1×10^5	1.0×10^{-7}	0.510 mm
GPE-2	118	4.2×10^{-4}	0.496 mm
GPE-3	79	9.6×10^{-4}	0.758 mm
GPE-4	49	1.4×10^{-3}	0.686 mm
GPE-5	25	2.5×10^{-3}	0.625 mm
GPE-6	200	2.5×10^{-4}	0.500 mm
CGPE-1	35	2.9×10^{-3}	1.015 mm
CGPE-2	29	3.1×10^{-3}	0.899 mm
CGPE-3	22	3.6×10^{-3}	0.792 mm
CGPE-4	37	3.3×10^{-3}	1.221 mm

6. EIS equivalent circuit discussion: Fig. 13(a) in main manuscript, the first element, R_b , represents the bulk resistance of the electrolyte. This bulk resistance arises from the intrinsic resistance to ion migration through the electrolyte matrix and corresponds to the high-frequency intercept of the Nyquist plot on the real axis. The bulk resistance (R_b) is connected in series with a parallel combination of a constant phase element (CPE) and the charge transfer resistance (R_{ct}). The parallel R_{ct} , CPE combination corresponds to the semicircle observed in the intermediate-frequency region of the Nyquist plot. The depressed semicircle, rather than an ideal semicircle, indicates non-ideal capacitive behaviour at the electrode–electrolyte interface. This deviation arises from factors such as surface roughness, electrode inhomogeneity, porosity, grain boundaries, and a distribution of relaxation times. Therefore, an ideal capacitor cannot accurately describe the interfacial double-layer capacitance. Instead, a CPE is used to represent the non-ideal capacitive behaviour of the interface caused by uneven charge distribution and structural heterogeneity at the electrode–electrolyte boundary. The charge transfer resistance (R_{ct}) represents the resistance associated with the electrochemical reaction, specifically the transfer of ions across the electrode–electrolyte interface during intercalation and deintercalation processes. Finally, the Warburg diffusion element (W_s) appears in the low-frequency region as a straight line inclined at approximately 45° . This element accounts for the diffusion-controlled transport of ions within the electrode and electrolyte. At low frequencies, ions have sufficient time to penetrate deeper into the electrode material, and the impedance becomes dominated by diffusion processes rather than charge transfer kinetics.

