Supporting Information

2D MXene-Containing Polymer Electrolytes for All-Solid-State Lithium Metal Batteries

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1.   Wide-angle X-ray diffraction (WAXD)

WAXD was performed on a Rigaku S-MAX 3000 SAXS system, with a microfocus sealed

tube X-ray source providing monochromatic beam at wavelength of 1.54 Å. WAXD patterns
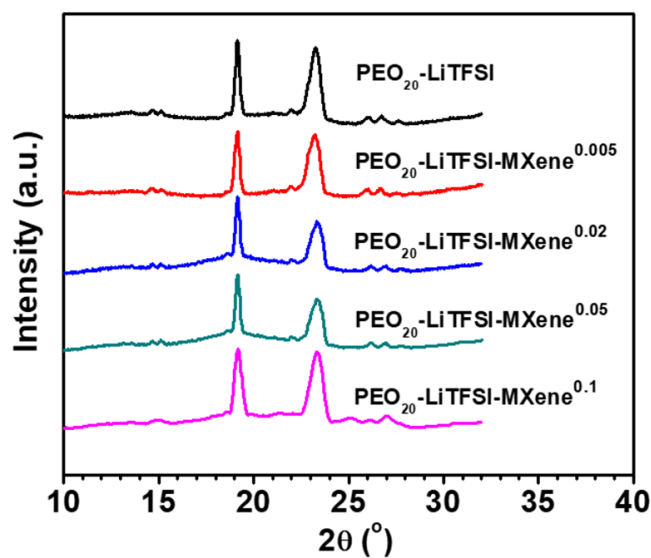
were collected with an image plate.

Fig. S1. WAXD patterns for PEO_{20}-LiTFSI and CPEs with different MXene content.

2. Electronic conductivity measurement

The electronic conductivity of the CPE was measured with a DC method. The CPE membrane was placed in-between two stainless steel electrodes. Polarization with a $\Delta V = 1$ V was performed.
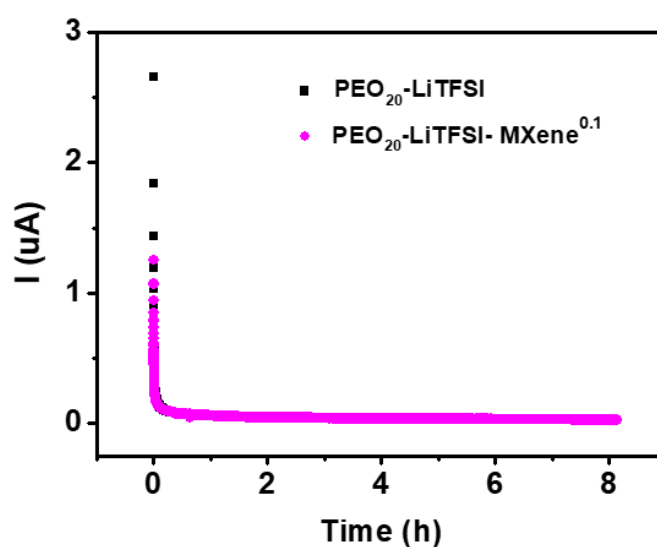

Fig. S2. DC polarization curves for PEO_{20}-LiTFSI and PEO_{20}-LiTFSI-MXene$^{0.1}$ at 60 °C.

3. Vogel-Tammann-Fulcher fitting of the temperature dependent ionic conductivity.

The temperature-dependent conductivity data were fitted using the modified Vogel-Tammann-Fulcher (VTF) equation (Equation 1):

$$\sigma = AT^{-1/2}e^{-E_a/[R(T-T_0)]}$$

where, T_0 is the Vogel temperature which is ~ (T_g-50). The pre-factor A is related to the charge carrier concentration of dissociated ions and E_a is related to the activation energy of ion motion.
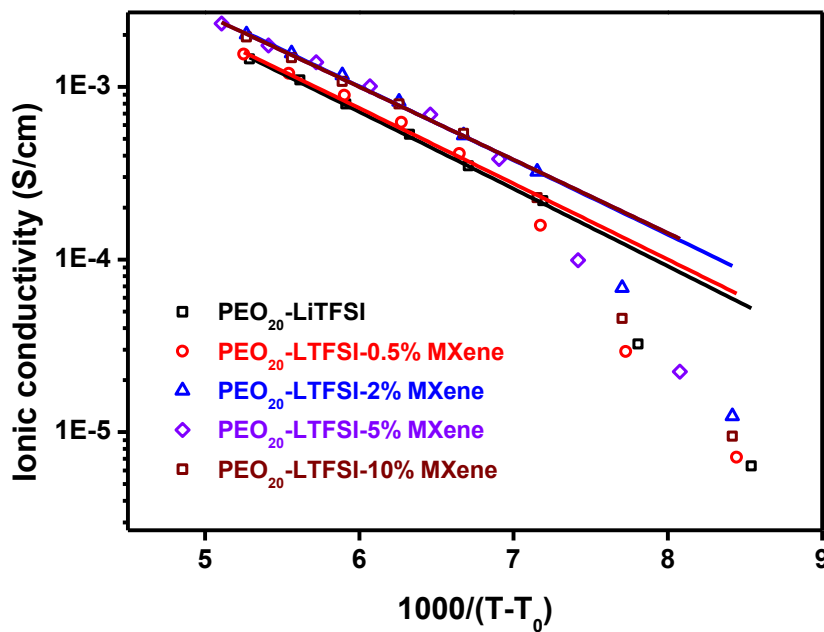

Fig. S3. VTF fitting of the temperature dependent ionic conductivity.

Table S1. VFT fitting parameters of the solid polymer electrolytes.

Samples	A (S cm^{-1} K$^{1/2}$)	E$_a$ (kJ mol^{-1})
PEO$_{20}$-LiTFSI	7.83 ± 2.19	8.83 ± 0.42
PEO$_{20}$-LiTFSI-MXene$^{0.005}$	7.28 ± 2.91	8.66 ± 0.61
PEO$_{20}$-LiTFSI-MXene$^{0.02}$	8.57 ± 2.55	8.48 ± 0.45
PEO$_{20}$-LiTFSI-MXene$^{0.05}$	7.85 ± 2.69	8.38 ± 0.53
PEO$_{20}$-LiTFSI-MXene$^{0.1}$	7.88 ± 2.70	8.37 ± 0.53

The fitting results show that at high temperature, the ion transport follows a VTF mechanism. The diverge from VTF behavior at low temperature is due to PEO crystallization. It seems that with adding MXenes, the activation energy slight decreases, although we acknowledge that the change is relatively small.

4. Galvanostatic cycling of the Li|MCPE|Li symmetric cell

Galvanostatic cycling measurement was performed on a Li|MCPE|Li symmetric coin cell (2032 type) with an Arbin battery tester at 60 °C. The current density is 0.3 mA cm^{-2}. The diameter of the lithium foil was 6 mm. The thickness of the composite polymer electrolytes PEO$_{20}$-LiTFSI-MXene$^{0.02}$ was around 150 μm. Coin cell was assembled in an argon-filled MBraun glove box (H$_2$O< 1 ppm, O$_2$ < 1 ppm). Repeated one-hour charge and one-hour discharge cycles were performed for the galvanostatic cycling measurements.
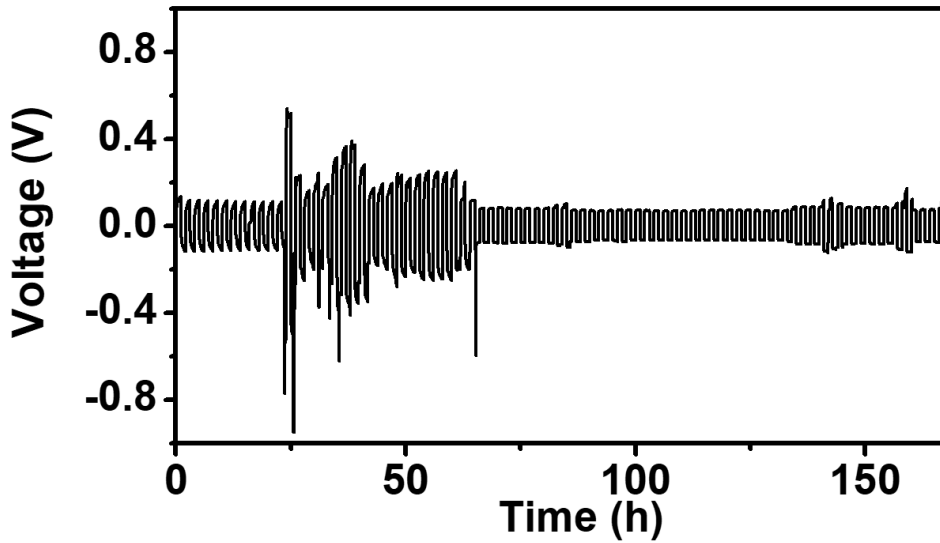
Fig. S4. Galvanostatic cycling curve of the Li symmetric cell with PEO$_{20}$-LiTFSI-MXene$^{0.02}$ as

separator at a current density of 0.3 mA cm^{-2} at 60 °C.

5. Electrochemical impedance spectroscopy (EIS) of the LiFePO$_4$|PEO$_{20}$-LiTFSI-MXene$^{0.02}$|Li battery.
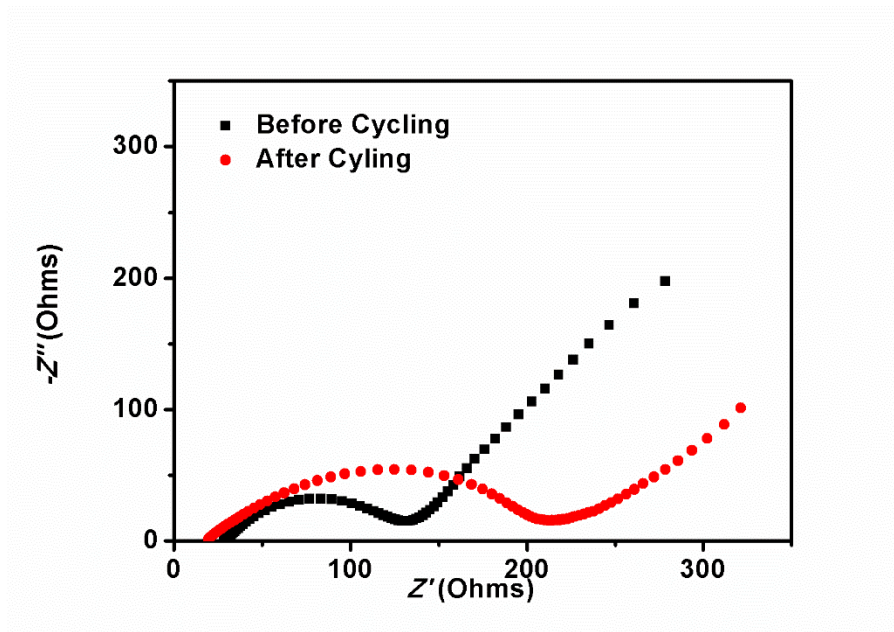

Fig. S5. Nyquist plots of the LiFePO$_4$|PEO$_{20}$-LiTFSI-MXene$^{0.02}$|Li battery before and after 100

galvanostatic cycles at a C/3 rate and 60 °C