

Supporting Information for

**Flexible and Stable PEO-Based Polymer Composite Solid Electrolyte  
Membranes Incorporating NASICON-Type  $\text{Li}_{1.3}\text{Al}_{0.3}\text{Ti}_{1.7}(\text{PO}_4)_3$  for High -  
Performance All-Solid-State Lithium Batteries**

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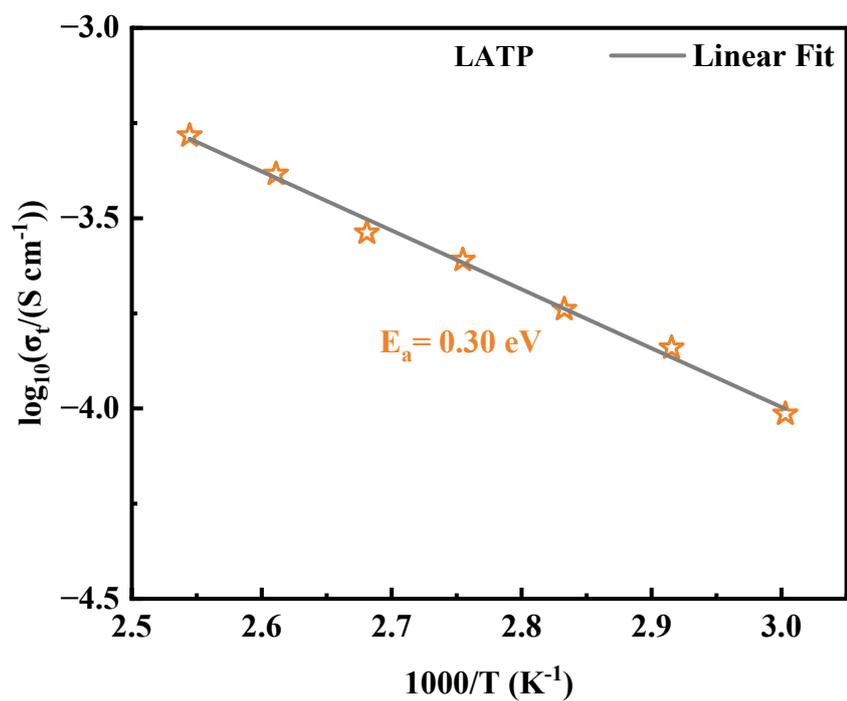
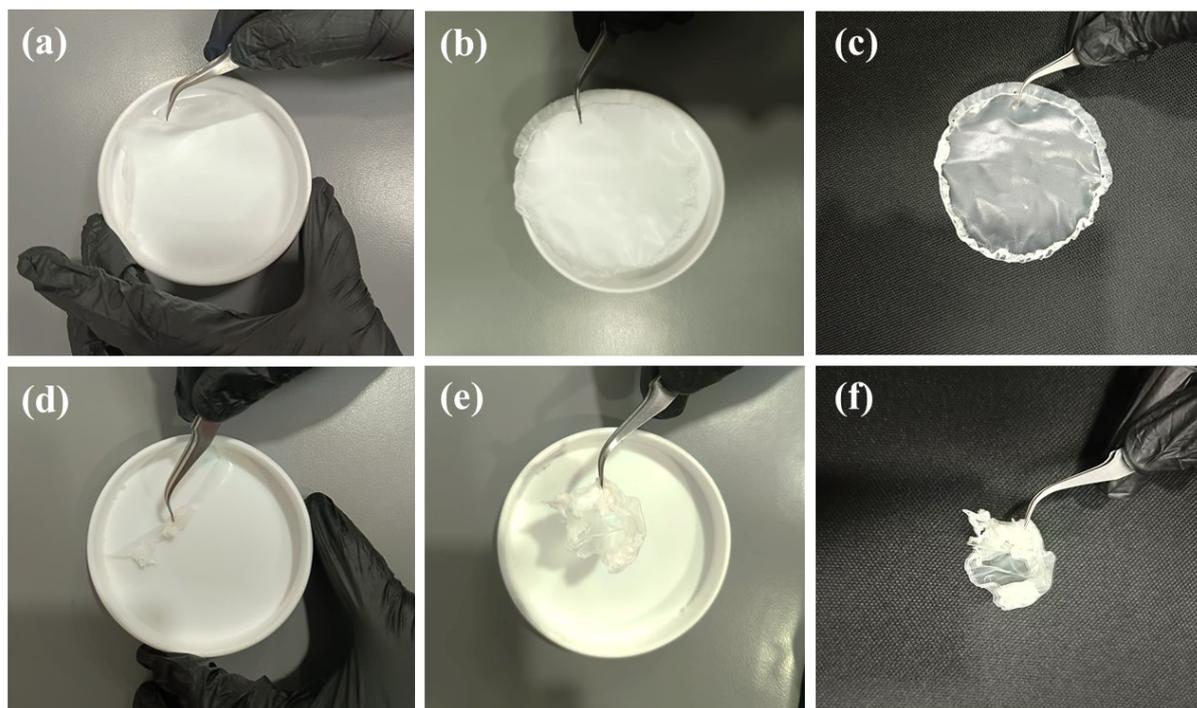
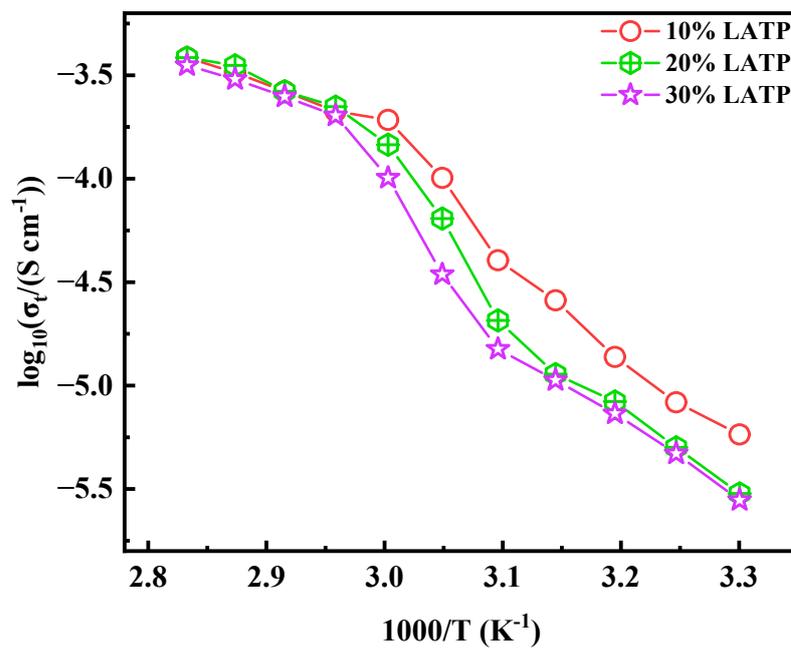


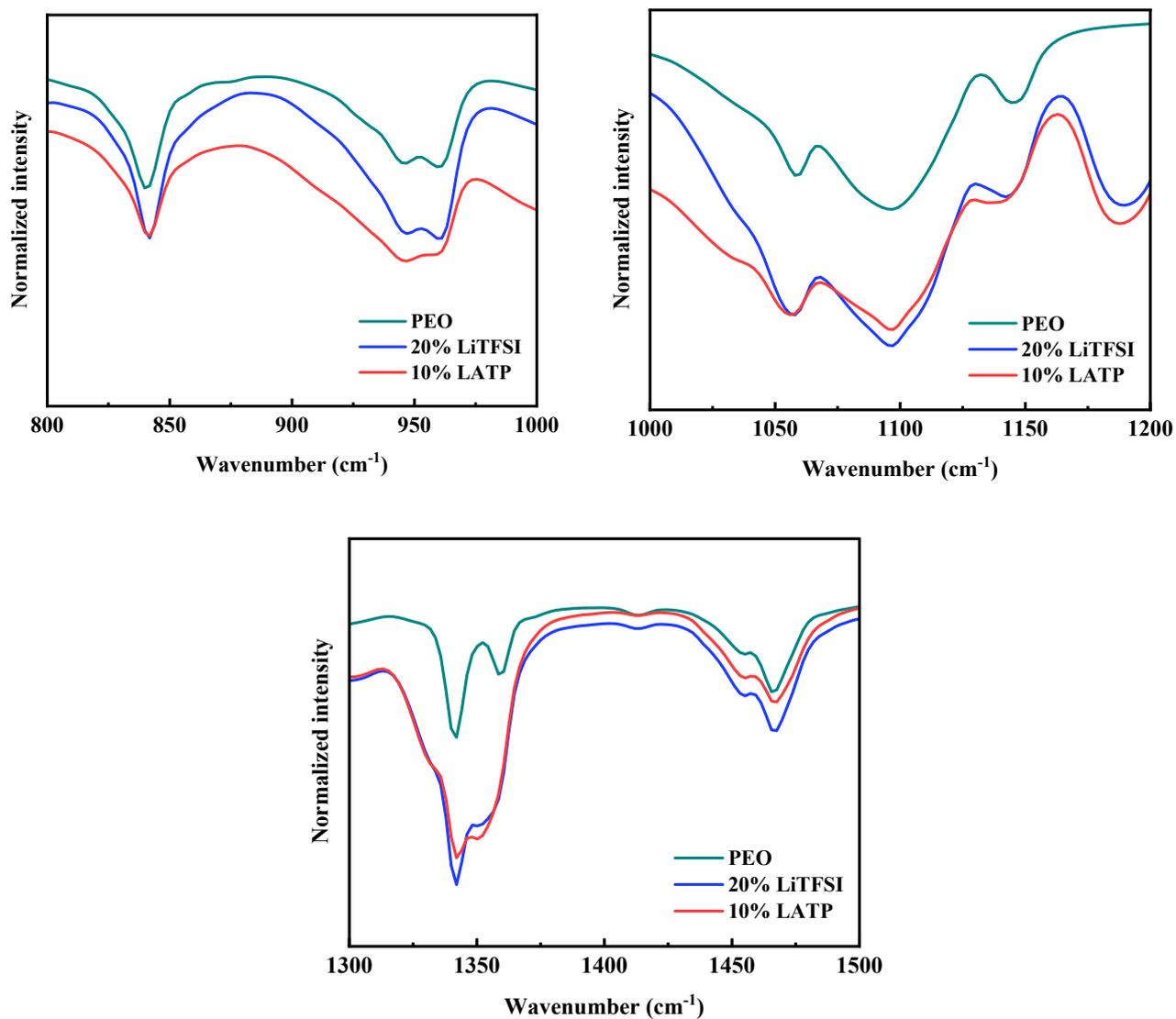
Fig. S1. Temperature dependent conductivity plot of LATP pellet.



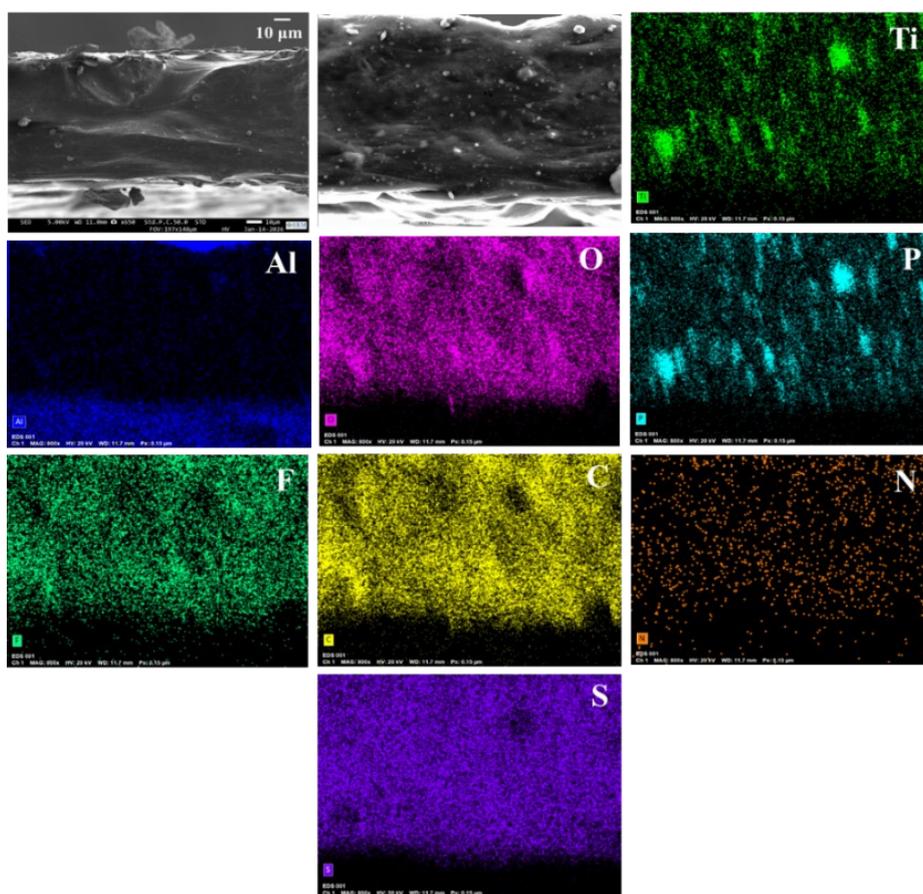
**Fig. S2 (a-c).** Photographs of the 20% LiTFSI and **(d-f).** 25% LiTFSI polymer membranes at RT during their development.



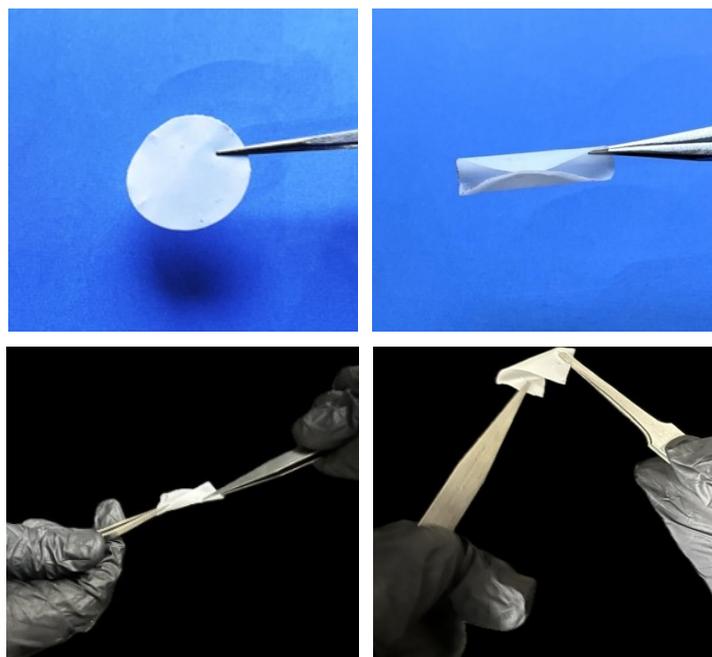
**Fig S3.** Temperature-dependent ionic conductivity (30-80°C) of PCSE membranes with varying LATP ceramic filler concentrations.



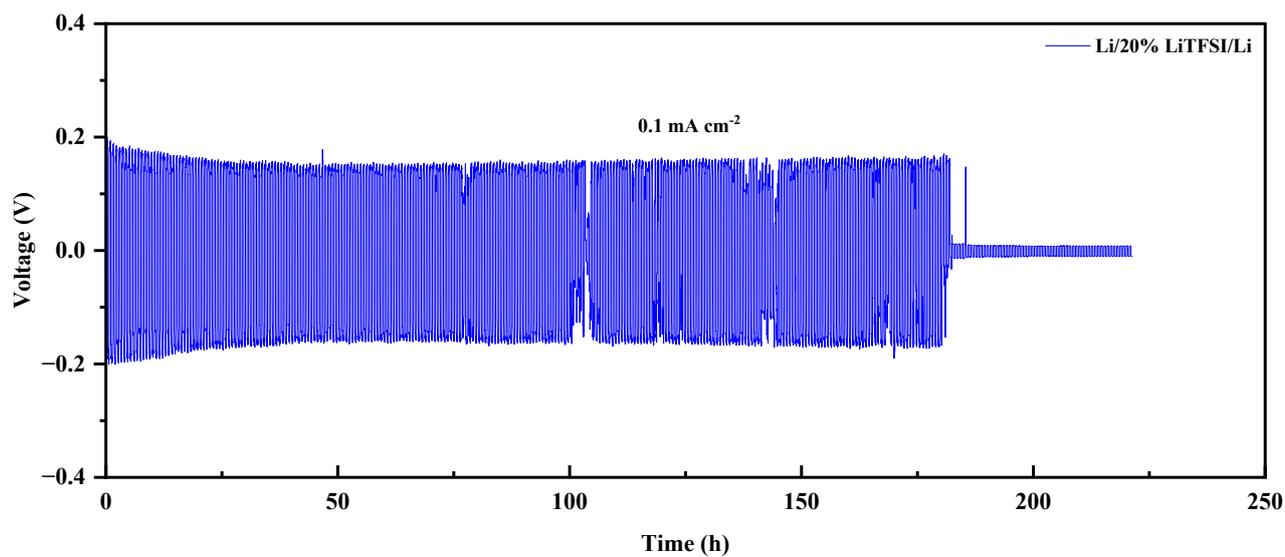
**Fig. S4.** Magnified regions of the FTIR spectra at specific wavenumber ranges.



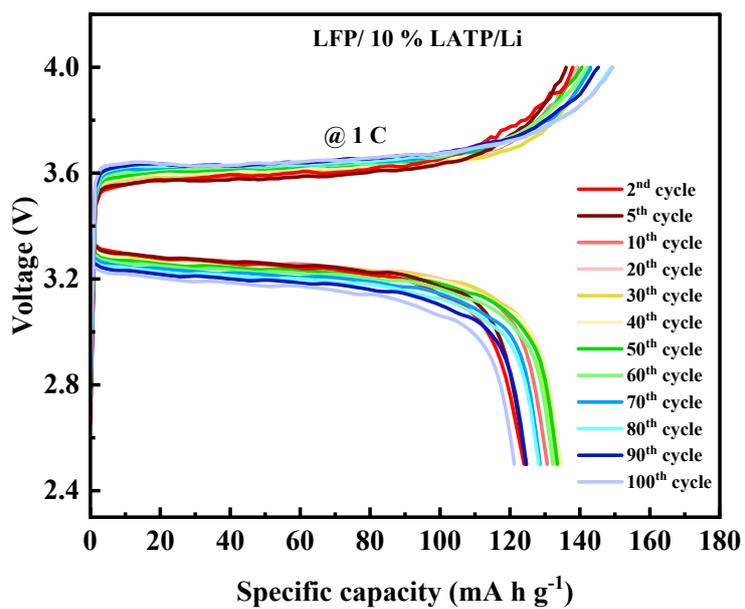
**Fig S5.** Cross-sectional SEM image with corresponding elemental mapping of the 10% LATP PCSE membrane.



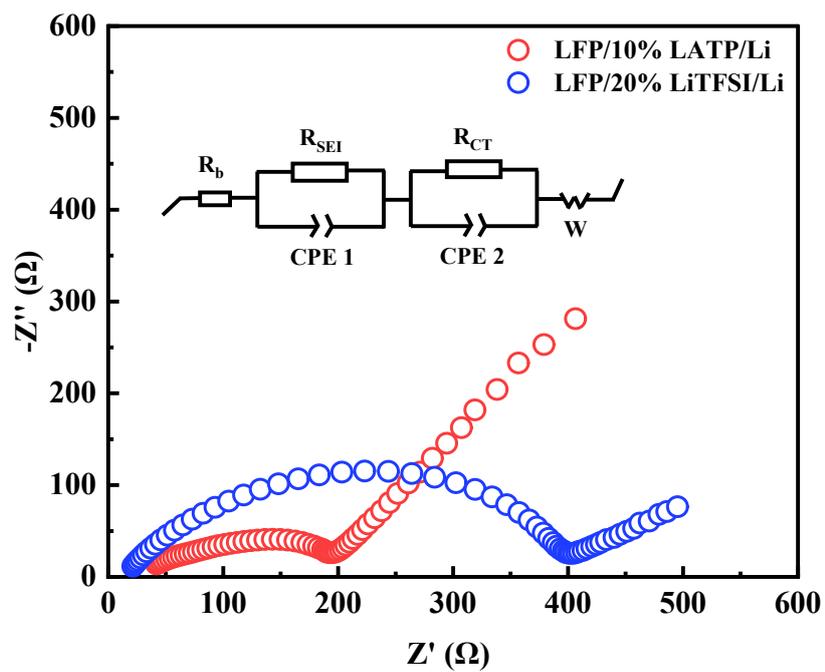
**Fig S6.** Digital photographs showing the flexibility of the 10% LATP PCSE membrane under bending and twisting tests.



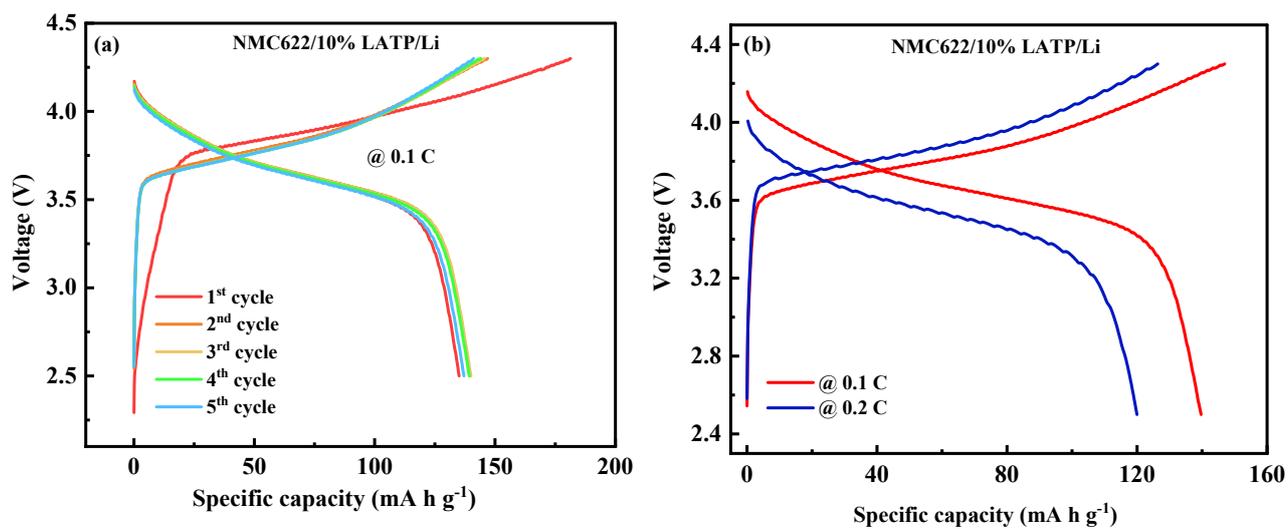
**Fig. S7.** Cycling stability of the Li/20% LiTFSI/ Li symmetric cell during lithium stripping-plating at a constant current density of 0.1 mA cm<sup>-2</sup>.



**Fig. S8.** Galvanostatic charge-discharge profiles showing the cycling performance of LFP/10% LATP/Li cell at 1 C.



**Fig. S9.** Nyquist plots of the LFP/PCSE/Li cell before cycling at 60°C.



**Fig. S10(a)** Charge-discharge profiles of NMC622/10% LATP/Li cell at 0.1 C. **(b)** Charge-discharge curves of NMC622/10% LATP/Li cell at two different current rates.

**Table S1:** Conductivity values of different PEO-based polymer membranes.

<b>Sample name</b>	<b>Conductivity (RT)</b>	<b>Conductivity (60°C)</b>
	<b>(S cm<sup>-1</sup>)</b>	<b>(S cm<sup>-1</sup>)</b>
15% LiTFSI	$0.13 \times 10^{-5}$	$0.30 \times 10^{-4}$
20% LiTFSI	$0.28 \times 10^{-5}$	$0.22 \times 10^{-3}$
10% LATP	$0.34 \times 10^{-5}$	$0.19 \times 10^{-3}$
20% LATP	$0.26 \times 10^{-5}$	$0.81 \times 10^{-4}$
30% LATP	$0.22 \times 10^{-5}$	$0.49 \times 10^{-4}$

**Table S2.** Comparison of the ionic conductivity and electrochemical performance of 20% LiTFSI and 10% LATP polymer membranes with previously reported values for polymer and composite polymer electrolytes.

Polymer Electrolyte	Ionic conductivity (S cm <sup>-1</sup> )	Discharge capacity (mA h g <sup>-1</sup> ) LiFePO <sub>4</sub> //Li	Current rate	Reference
PEO-LiTFSI	2.95 × 10 <sup>-4</sup> at 55°C	151.6 at 55°C	0.2 C	<b>1</b>
PEO-LiTFSI	2.86 × 10 <sup>-4</sup> at 60°C	152.02 at 60°C	0.2 C	<b>2</b>
PVDF–HFP- LiTFSI	7.1 × 10 <sup>-5</sup> at RT	112 at RT	0.2 C	<b>3</b>
PVDF–HFP-LiTFSI	2.5 × 10 <sup>-4</sup> at 25°C	127.4 at 25°C	0.1 C	<b>4</b>
PEO-LiTFSI (20 % LiTFSI)	2.2 × 10 <sup>-4</sup> at 60°C	143.4 at 60°C	0.1 C	This work
PEO-LiClO <sub>4</sub> -LATP	1.6 × 10 <sup>-3</sup> at 80°C	130.2 at 80°C	0.2 C	<b>5</b>
PEO-LiTFSI-LATP- FEC	1.99 × 10 <sup>-4</sup> at 30°C	147 at RT	0.1 C	<b>6</b>
PEO-LiTFSI-LAGP	1.6 × 10 <sup>-5</sup> at 20°C	166 at 80°C	0.1 C	<b>7</b>
PEO-LiTFSI- LLZTO	2.12 × 10 <sup>-4</sup> at 60°C	153.8 at 60°C	0.3 C	<b>8</b>

PVDF-LiTFSI-	$2.44 \times 10^{-4}$	155	0.2 C	<b>9</b>
LATP	at 25°C	at 30°C		
PEO-LiTFSI-LATP (10% LATP)	$1.9 \times 10^{-4}$ at 60°C	151.6 at 60°C	0.1 C	This work

## References

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