

## Supplementary Materials

# Dry Approach Production of Garnet Solid Electrolyte Membrane for Lithium Batteries

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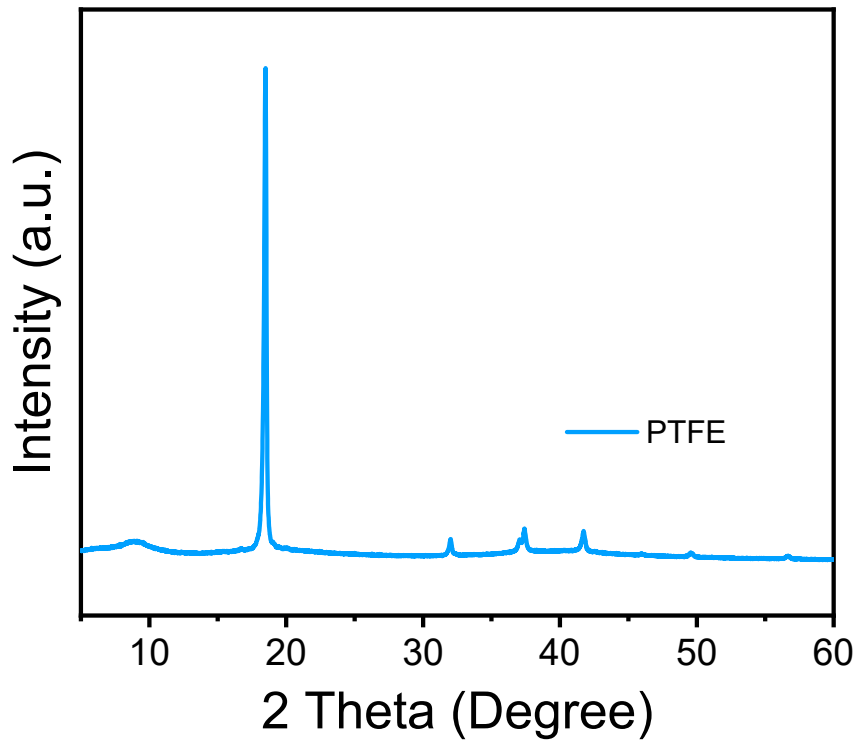


Fig. S1 The XRD of PTFE powder

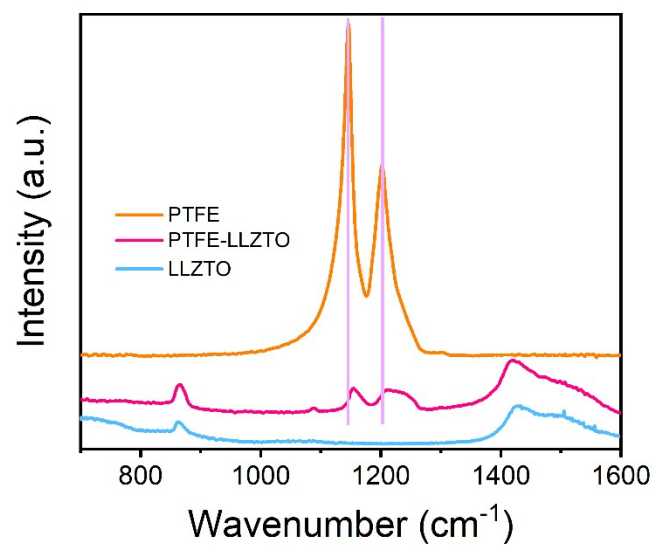


Fig. S2 Infrared spectroscopy of PTFE, LLZTO and PTFE-LLZTO membrane.

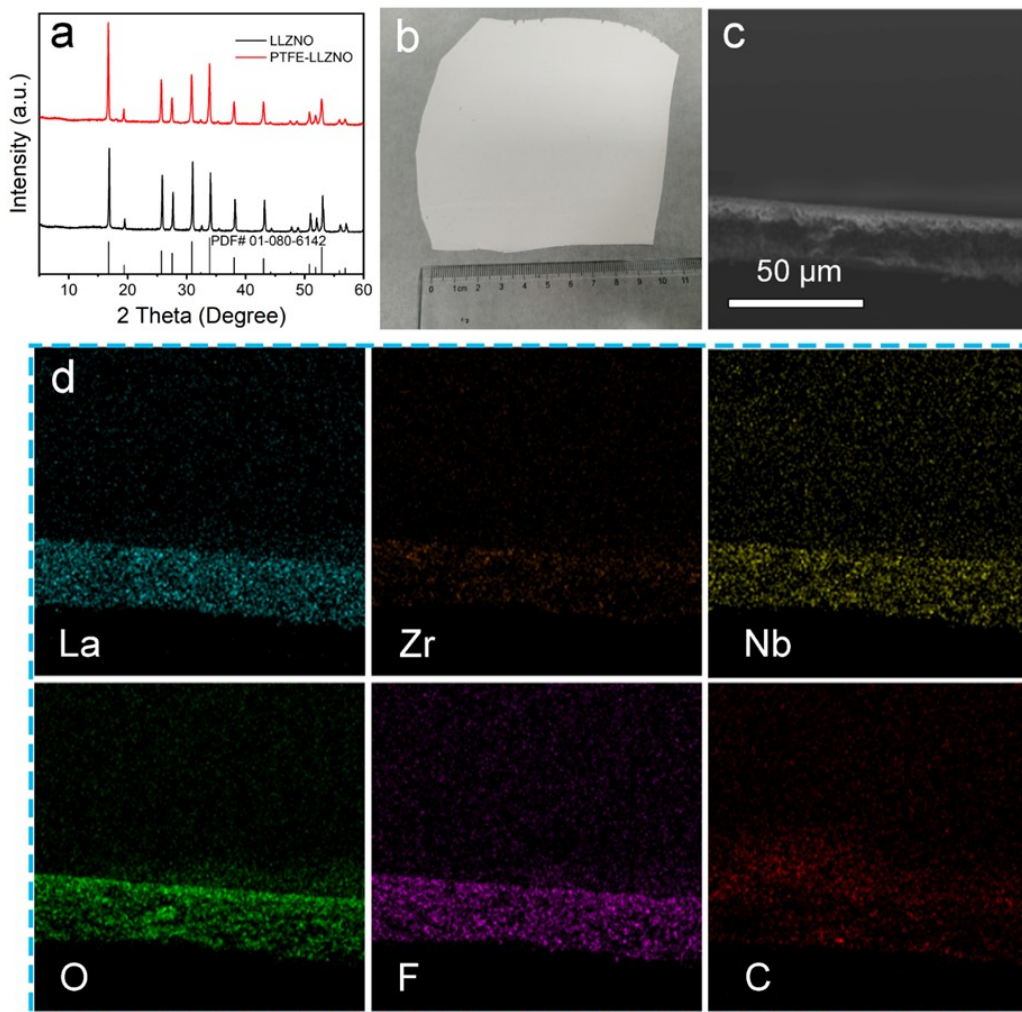


Fig. S3 a) The XRD of LLZNO and PTFE-LLZNO electrolyte membrane; b) The picture of LLZNO; c) The SEM of the cross-sectional of PTFE-LLZNO electrolyte membrane; d)The EDS mapping of Fig. c.

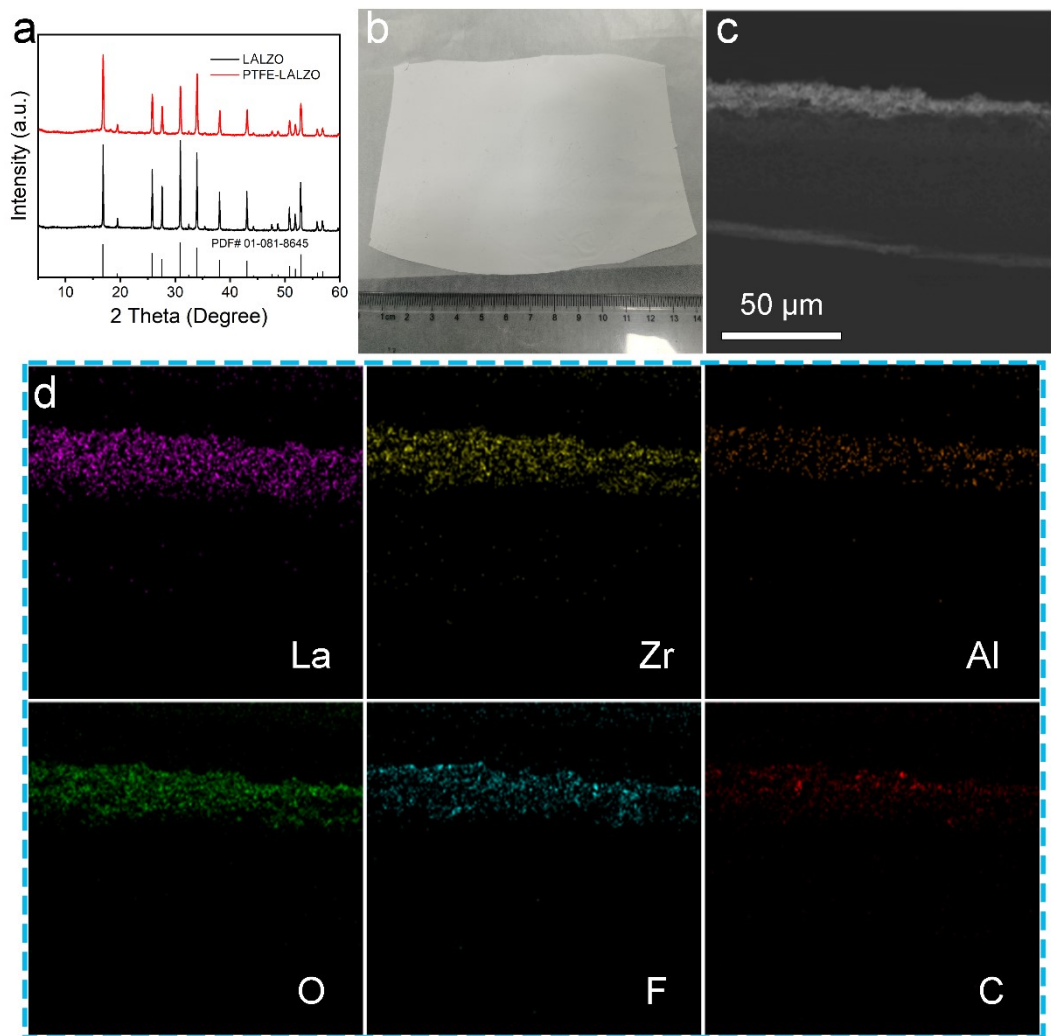


Fig. S4 a) The XRD of LALZO and PTFE-LALZO electrolyte membrane; b) The picture of LALZO; c) The SEM of the cross-sectional of PTFE- LALZO electrolyte membrane; d)The EDS mapping of Fig. c.

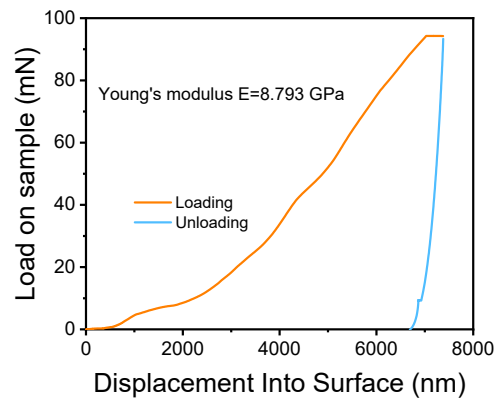


Fig. S5 Load Displacement Curve of PTFE-LLZTO Membrane

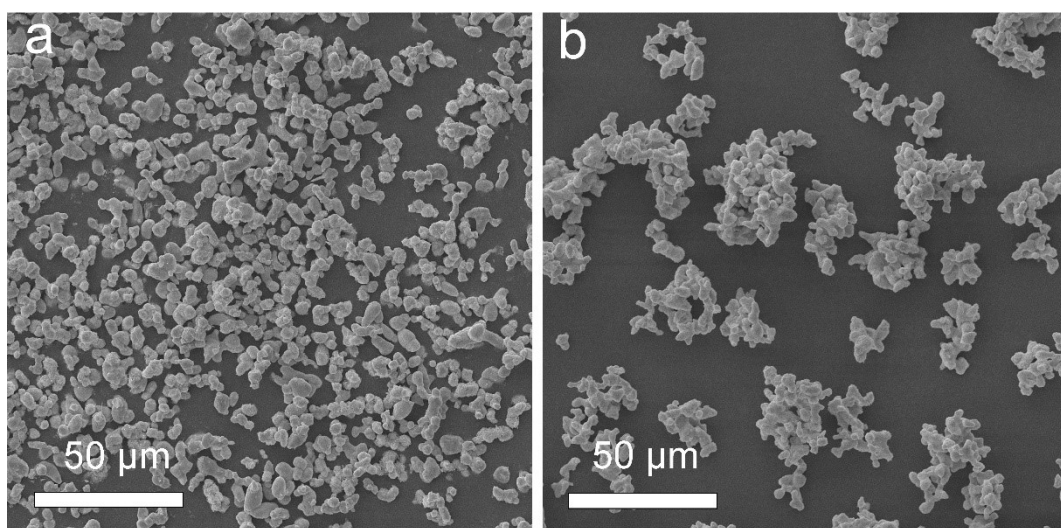


Fig. S6 The SEM of PTFE@LLZTO powder

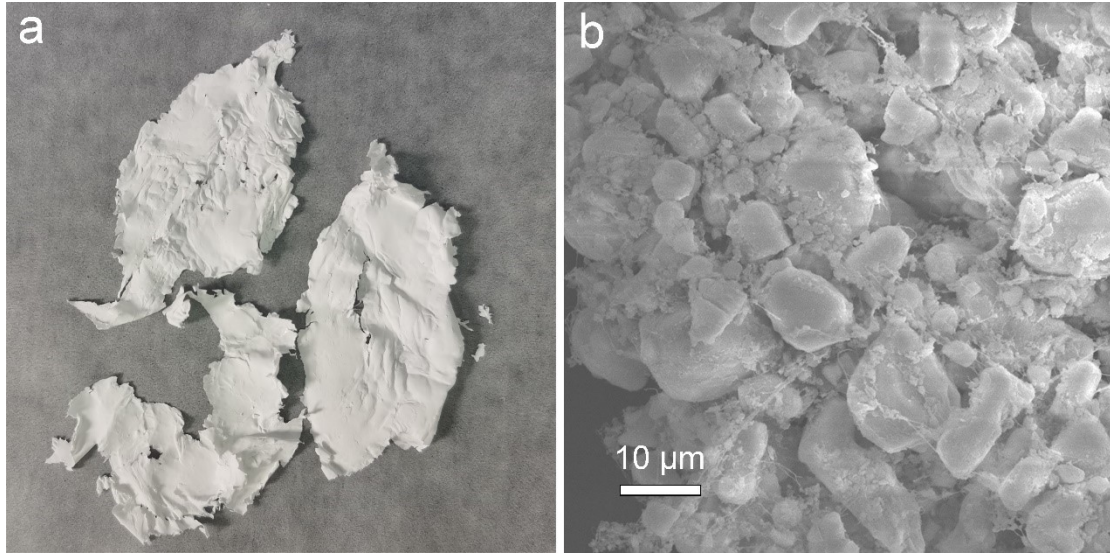


Fig. S7 a) The picture of PTFE-LLZTO electrolyte membrane by ball mill to obtain the PTFE-LLZTO matrix; b) the SEM of PTFE-LLZTO matrix by a ball mill



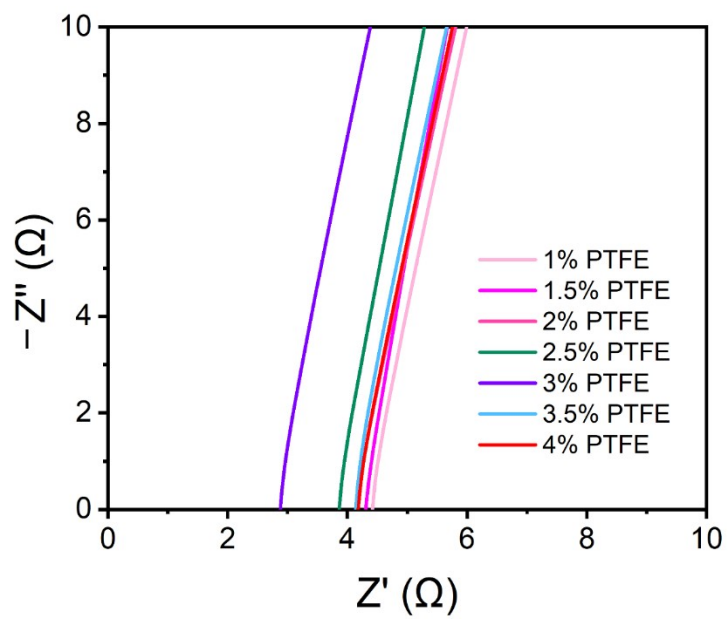


Fig. S8 The EIS of PTFE-LLZTO electrolyte membrane with different content PTFE.

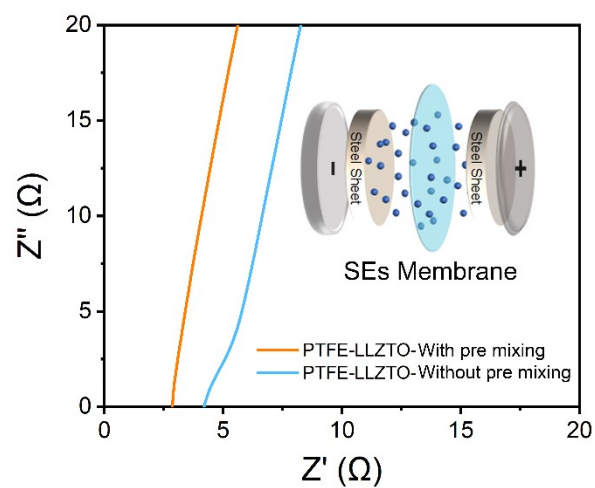


Fig. S9 The EIS of PTFE-LLZTO membrane with (a) and without (b) pre mixing at different locations on the same membrane.

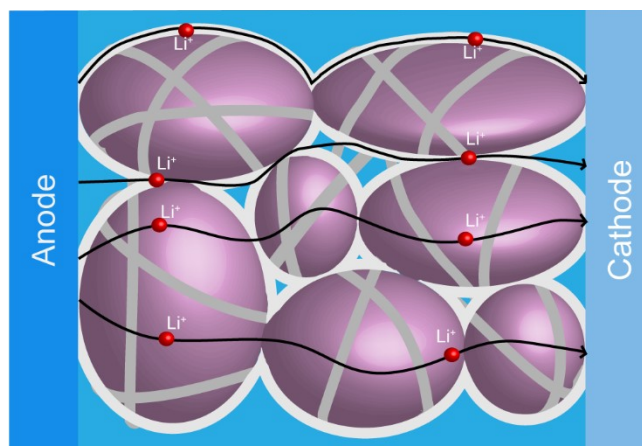


Fig. S10 Schematic diagram of  $\text{Li}^+$  transport channel for PTFE-LLZTO membrane at discharge.  $\text{Li}^+$  is expected to have three transmission channels, LLZTO, Interface, LLZTO-Interface-LLZTO (“Interface” is the interface of liquid electrolytes (LEs) and LLZTO).

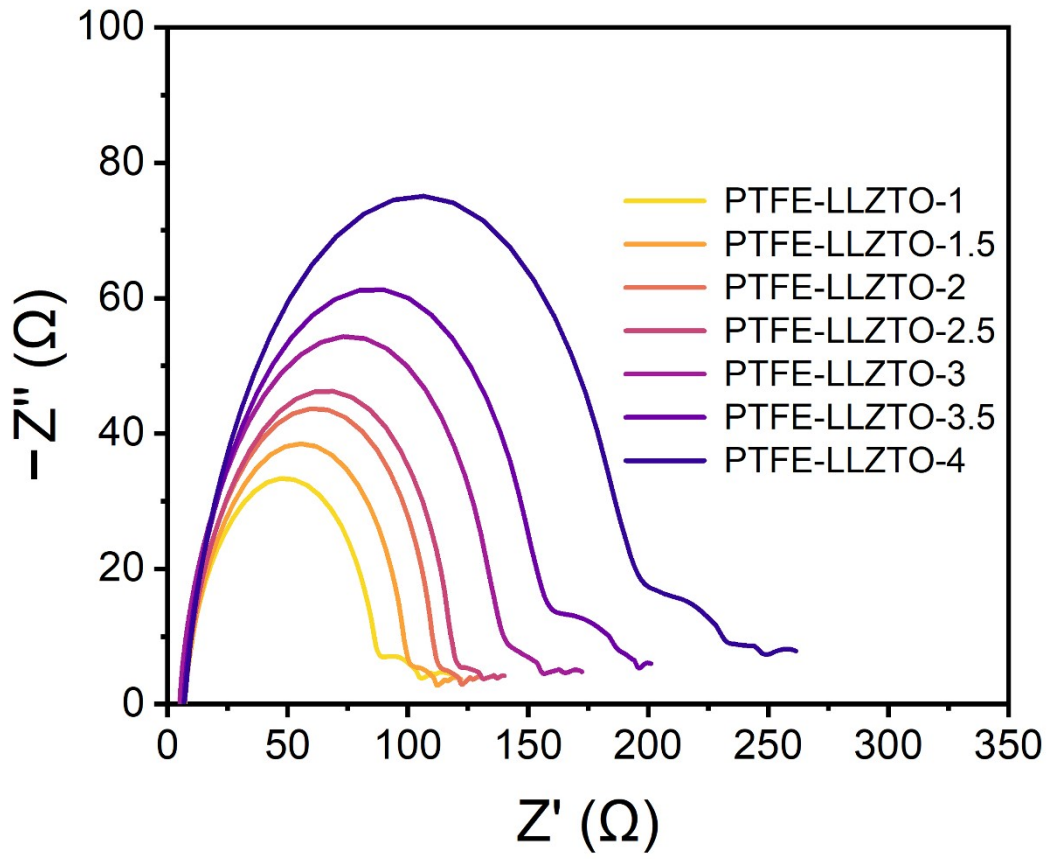


Fig. 11 The interfacial EIS of Li/PTFE-LLZTO with different content PTFE

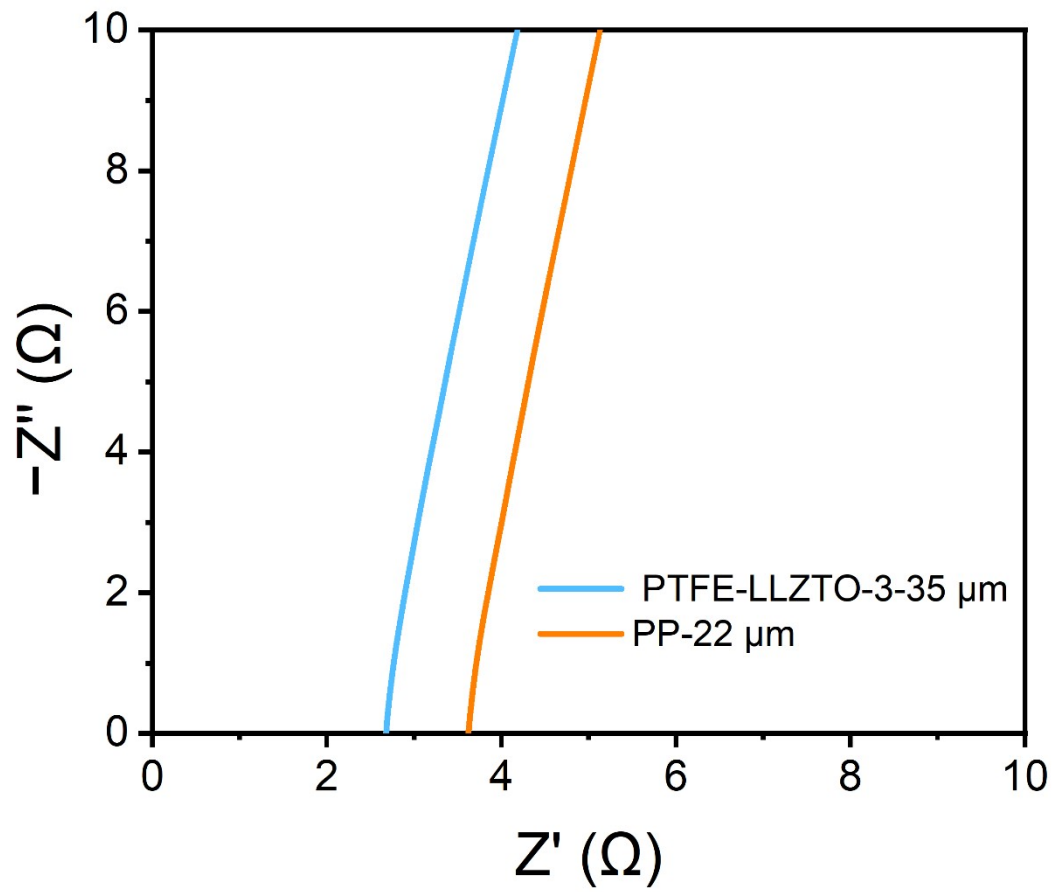


Fig. S12 The EIS of PTFE-LLZTO-3-35 μm and PP

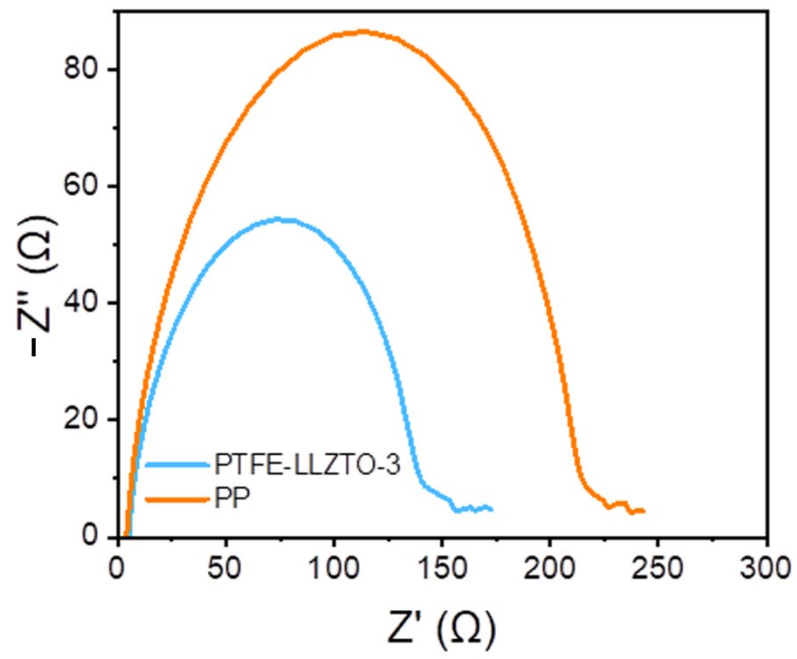


Fig. S12 The interfacial EIS of PTFE-LLZTO-3 and PP with Li

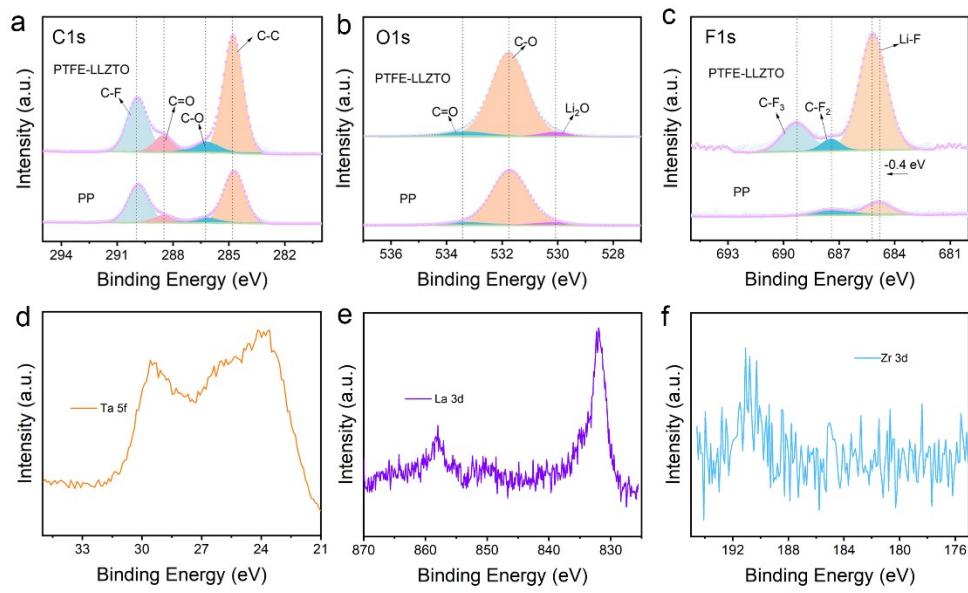


Fig. 13 XPS spectroscopy of a) C1s, b) O1s and c) F1s on Li anode of Li/PTFE-LLZTO/Li and Li/PP/Li after 20 hours of cycling, XPS spectroscopy of a) Ta5f, b) La3d and c) Zr3d on Li anode of Li/PTFE-LLZTO/Li after 20 hours cycling.

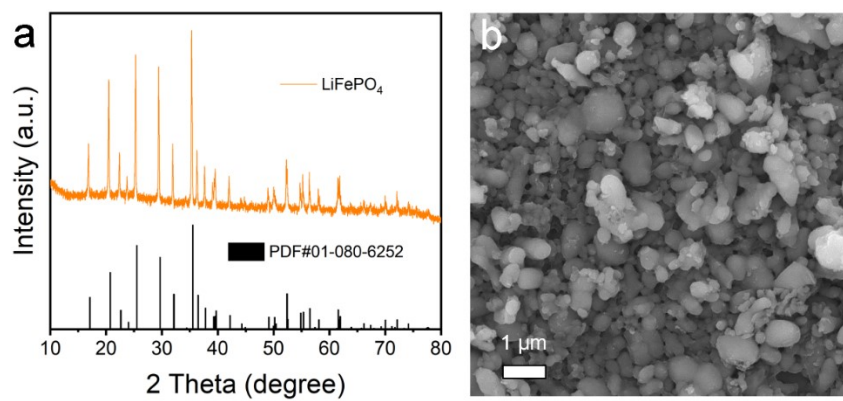


Fig. S14 a) The XRD pattern of LiFePO<sub>4</sub>; b) The SEM image of LiFePO<sub>4</sub>