Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information

Dopamine Modified $Li_{6.4}La_3Zr_{1.4}Ta_{0.6}O_{12}$/PEO Solid-State Electrolyte: Enhanced Thermal and Electrochemical Properties

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Part I: Characterizations of LLZTO bulk electrolyte, powder and composite electrolytes

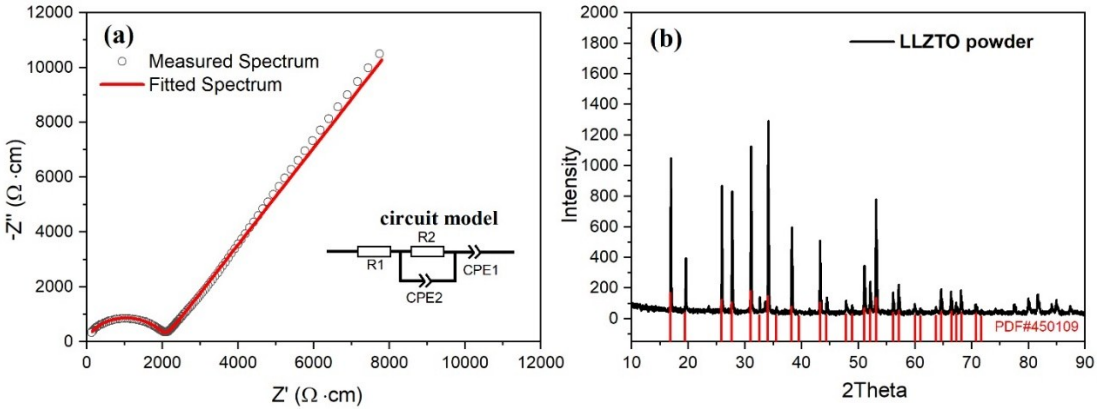

Fig. S1 (a) Electrochemical Impedance Spectrum of the synthesized bulk LLZTO; (b)XRD of LLZTO powder

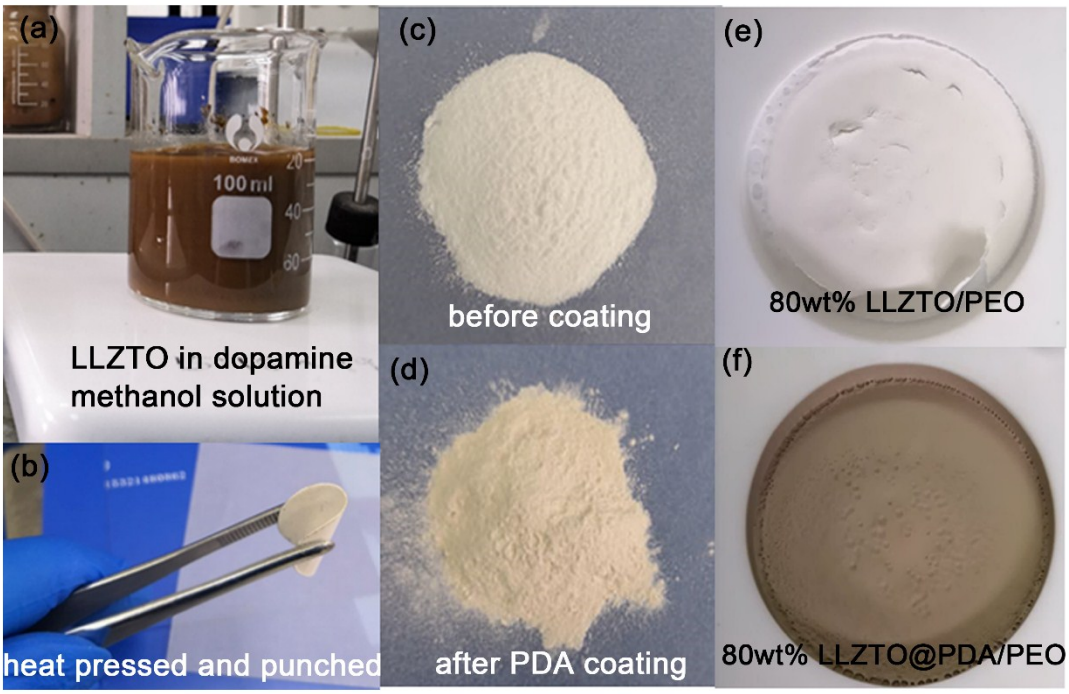

Fig. S2. (a) LLZTO powder dispersed in methanol solution dissolved dopamine and after a while the color of solution changed to brown. (b) The composite solid-state electrolyte after heat pressed

and cut. (c)The color of pristine LLZTO is white; (d) after polydopamine coating, the color of LLZTO change to light brown. (e) 80 wt% LLZTO@PDA/PEO and (f) 80 wt% LLZTO/PEO after dried in PTFE mold;

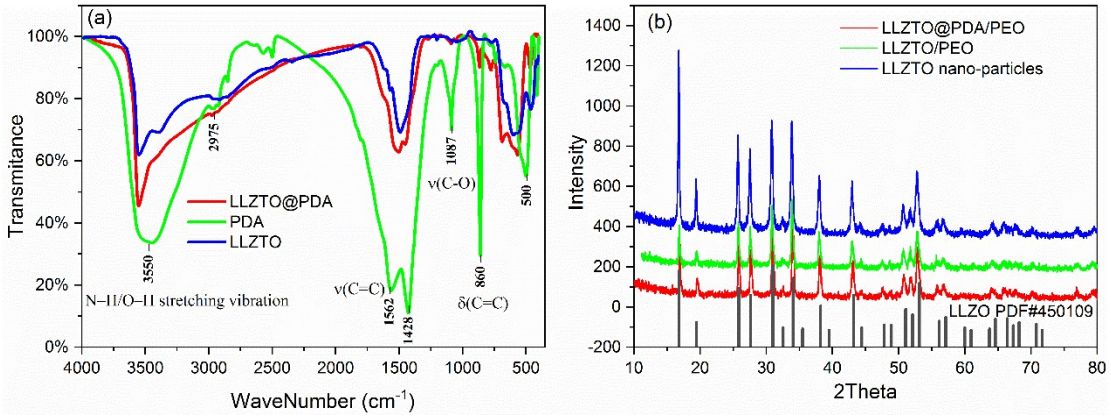

Fig. S3. (a) Infrared spectra of LLZTO, LLZTO@PDA and the pure PDA. The IR peaks near 3550 cm^{-1}, 1562 cm^{-1}, 1428 cm^{-1} indicate that polydopamine was coated on the LLZTO surface; (b) XRD of LLZTO nano-particles, LLZTO@PDA/PEO and LLZTO/PEO composite solid-state electrolyte.

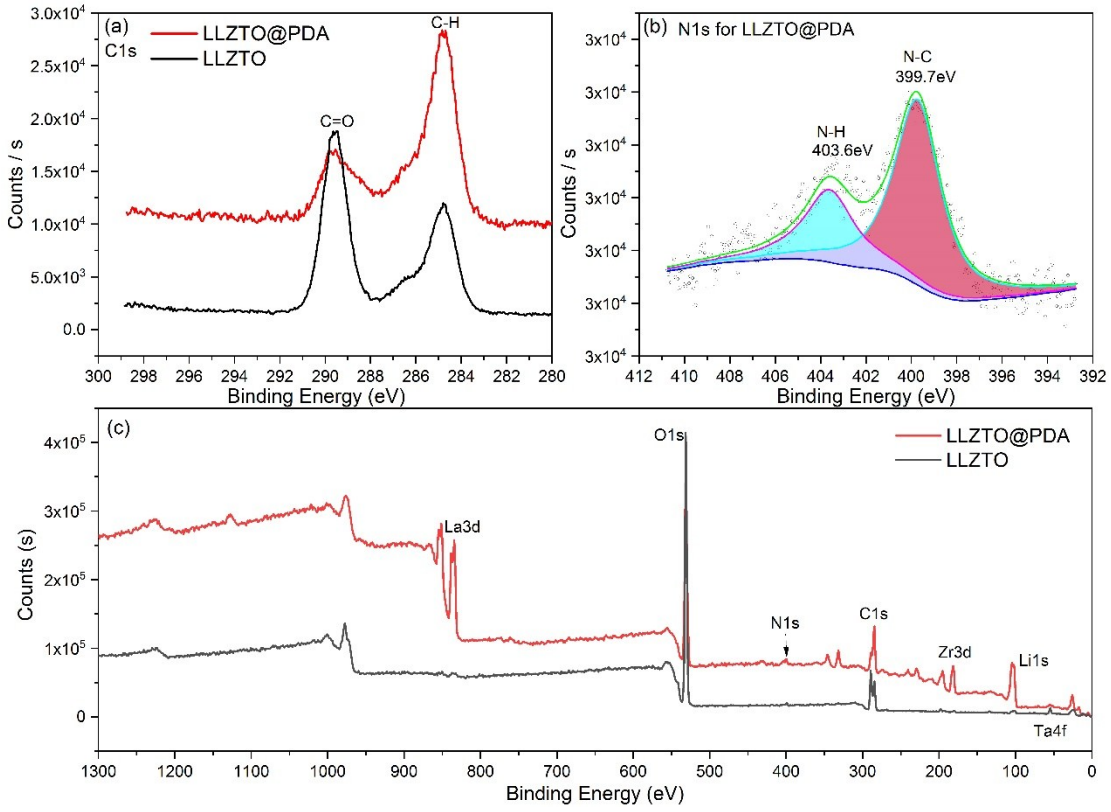

Fig. S4. XPS spectra of LLZTO and LLZTO@PDA powders. (a) High- resolution XPS spectra of C1s region; (b) High-resolution XPS spectra of N1s region of LLZTO@PDA (c) The full XPS spectra of LLZTO and LLZTO@PDA

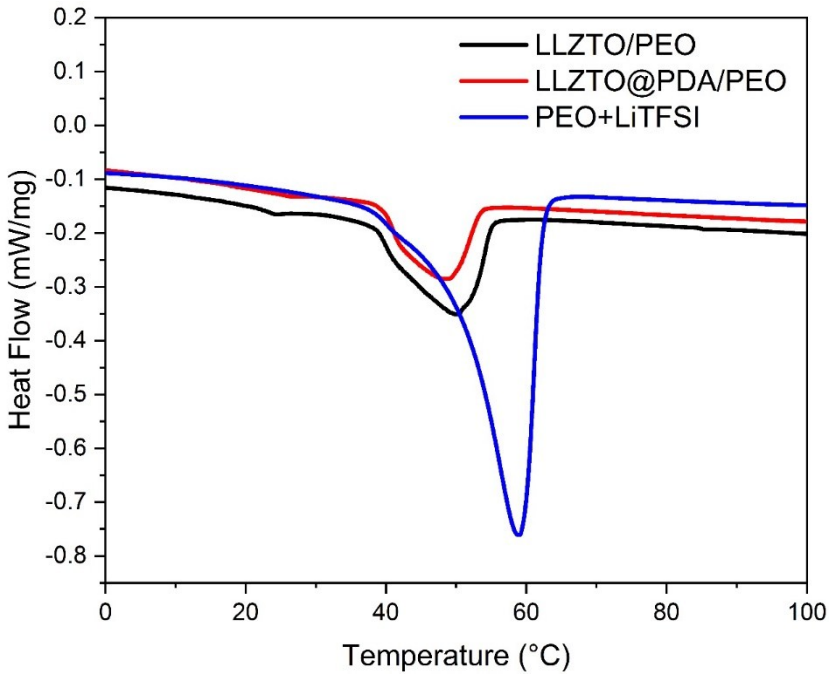

Fig. S5. DSC curves of PEO/LiTFSI, LLZTO in PEO and LLZTO@PDA in PEO

Part II: Characterizations of polydopamine coated nano-ZrO_2 particles and composite electrolyte based on ZrO_2 and PEO

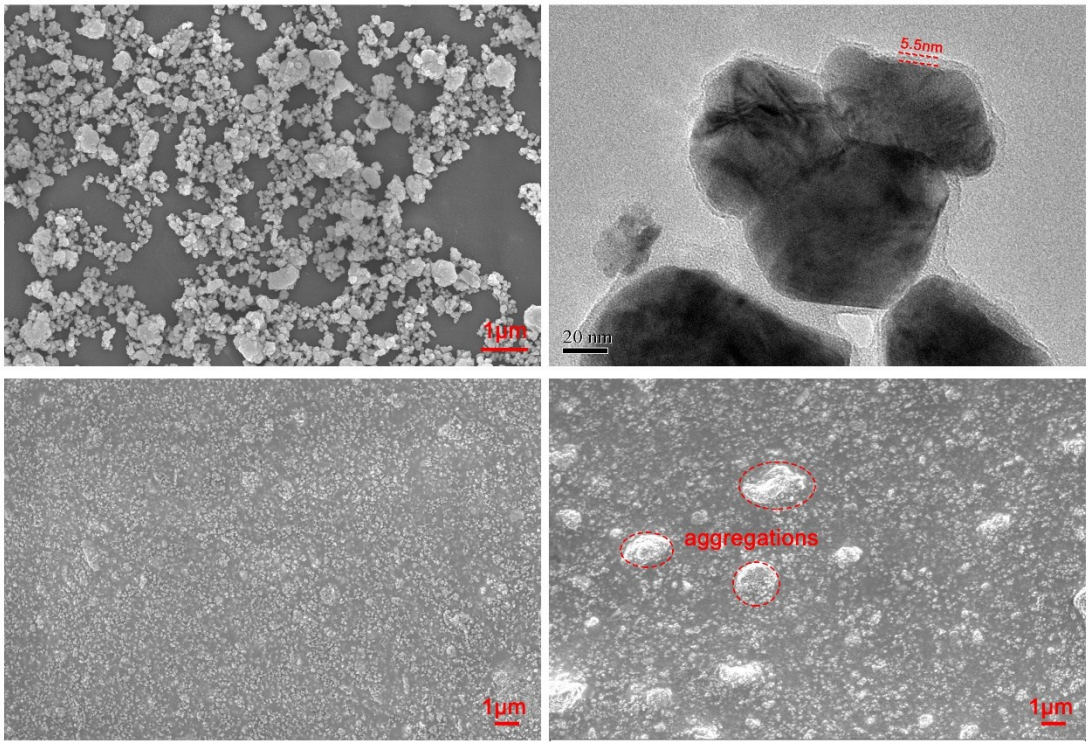

Fig. S6 (a) SEM image of nano-sized ZrO_2 powder (b)TEM photograph of PDA coated nano-ZrO_2, which indicates that the nano-ZrO_2 was coated with 5~6nm polydopamine. (c) Cross-section image of ZrO_2@PDA/PEO composite electrolyte, where 80wt% ZrO_2@PDA dispersed well in the PEO matrix. (d) Cross-section image of ZrO_2/PEO composite electrolyte, where 80wt% ZrO_2 dispersed in PEO and aggregations formed in the matrix.

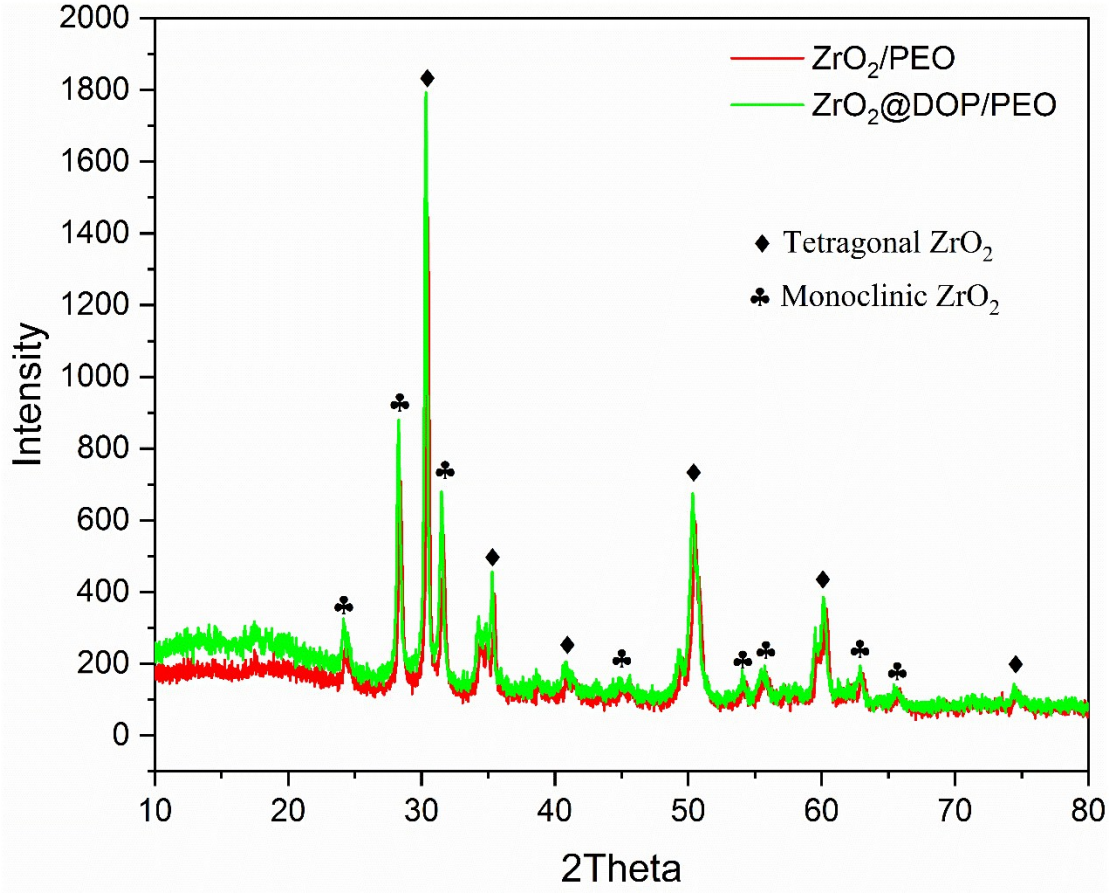

Fig. S7 XRD spectrums of ZrO_2@PDA/PEO and ZrO_2/PEO composite electrolyte. The spectrum indicates that ZrO_2 powders combined tetragonal and monoclinic phases. No peeks assigned to crystalline PEO are found.

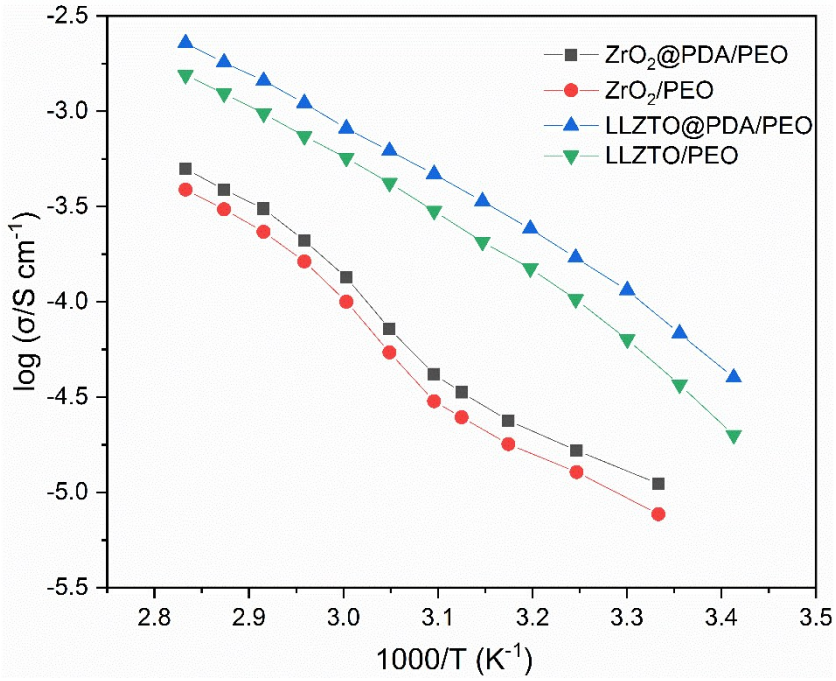

Fig. S8 The logσ~1000/T plots of ZrO_2@PDA/PEO and ZrO_2/PEO electrolyte in the range from room temperature to 80 °C. The conductivities of LLZTO@PDA/PEO and LLZTO/PEO electrolyte are also plotted in the graph as comparison.

Part III: Polydopamine coated LLZTO tablets and Li^+ transporting through the interface

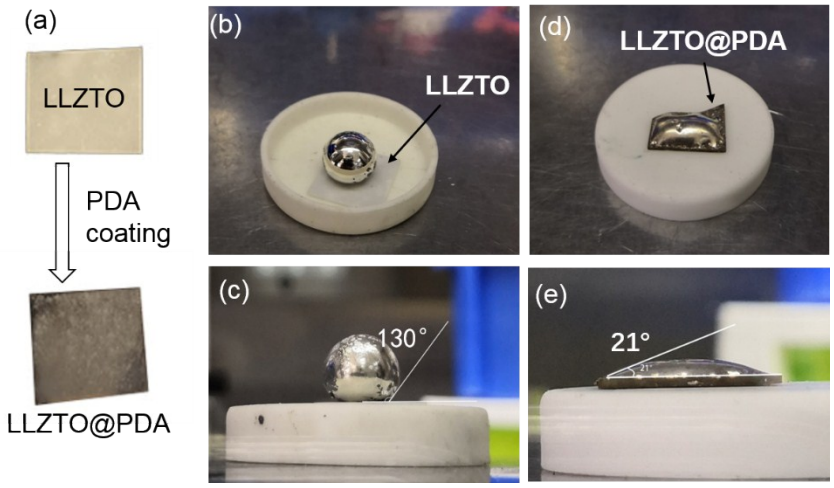

Fig.S9 Demonstrations of contact angles of LLZTO tablet or polydopamine coated LLZTO tablet with molten lithium droplet. (a) The color change of LLZTO after polydopamine coating; (b) and (c) the contact angle of molten lithium droplet on pristine LLZTO tablet; (d) and (e) the contact angle of molten lithium droplet on LLZTO@PDA tablet; The reduced contact angle from 130° to 21° indicates that molten lithium has good wetting ability with polydopamine modified LLZTO surface.

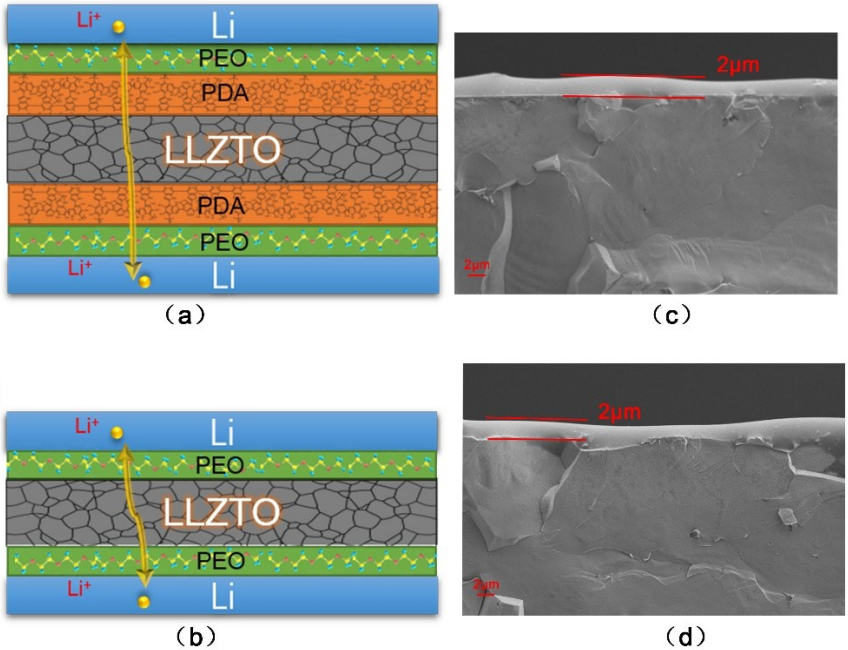

Fig.S10 schematic diagrams of Li-Li symmetrical cells and the interface between Li electrode and LLZTO was buffered with PEO/LiTFSI electrolyte. The PEO buffer layer was spin-coated onto the LLZTO surface. (a) The LLZTO tablet was first coated with polydopamine layer before PEO spin-coating, and the cell was notated as Li|PEO|PDA|LLZTO|PDA|PEO|Li. (b) No polydopamine layer was formed between LLZTO and PEO, and the cell was notated as Li|PEO |LLZTO |PEO|Li. The

height ratio of structural layers in the schematic diagrams do not describe the true thickness ratio. (c) and (d), the cross-section images show the thickness of PEO buffer layer are both about 2μm in those Li-Li symmetrical cells.

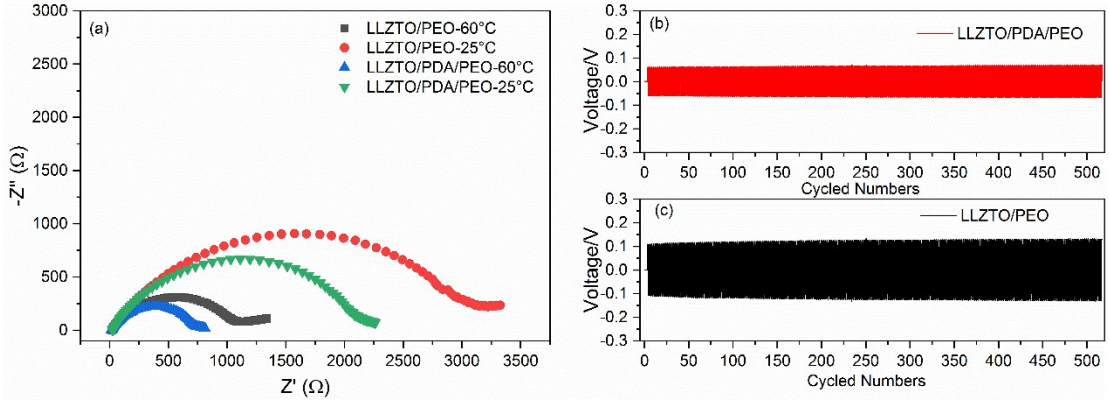

Fig.S11 (a) The total electrochemical impedance spectrums of Li|PEO|LLZTO|PEO|Li and Li|PEO|PDA|LLZTO|PDA|PEO|Li cells at 25 °C and 60 °C. (b) The symmetrical cells were cycled at 60 °C with a current density of 25μA cm^{-2} to demonstrate the Li^{+} transporting ability through polydopamine layer. Each cycle cost 20 minute. The cell cycled over 500 times without evident polarization increase, which indicates a good stability of interface.