Support Information

π-d conjugation regulates the cathode/electrolyte interface in all-solid-state lithium-ion batteries

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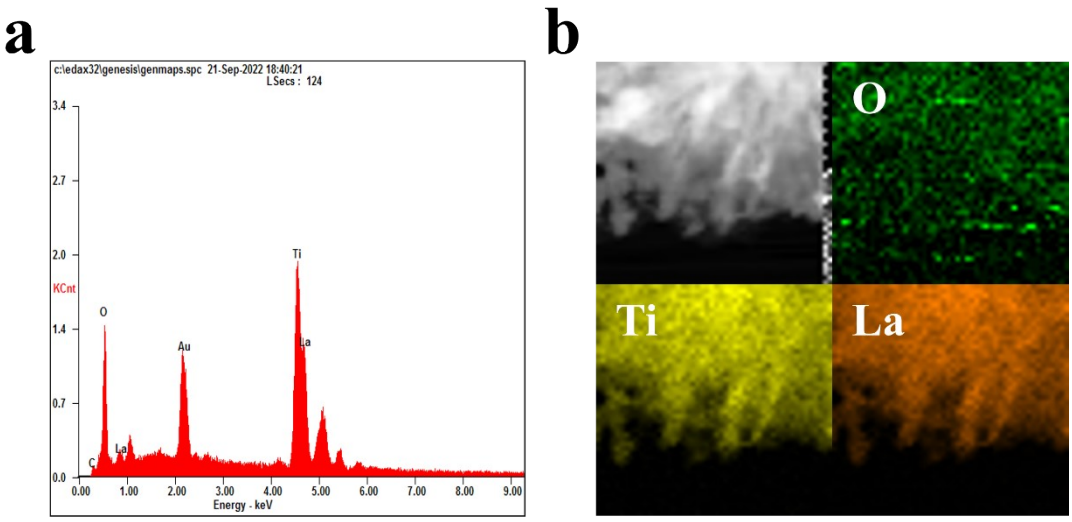

Figure S1. The spectrum of energy dispersive spectroscopy (a) and elemental map (b) of LLTO.

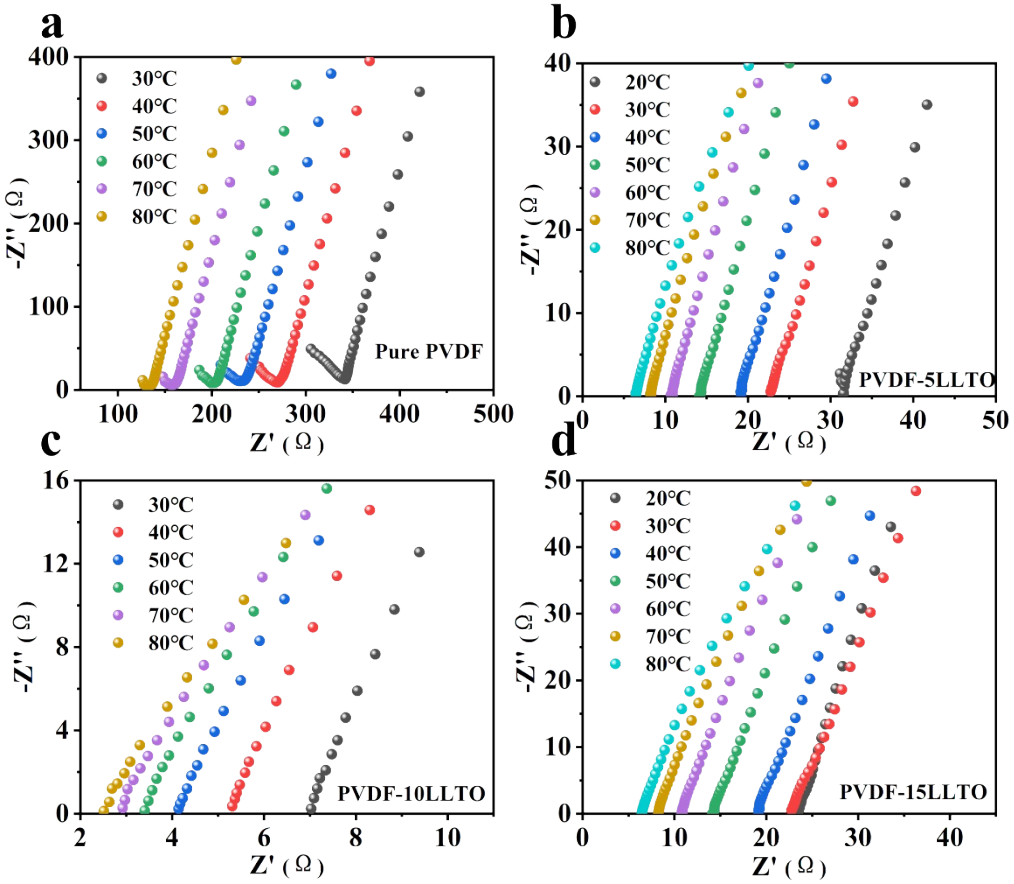

Figure S2. EIS curves of (a) Pure PVDF, (b) PVDF-5LLTO, (c) PVDF-10LLTO and (d) PVDF-15LLTO at different temperatures.

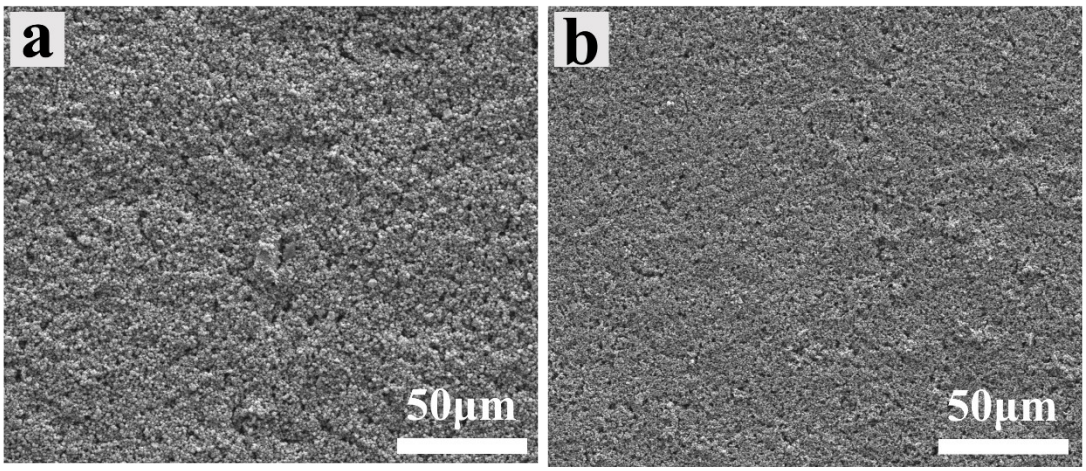

Figure S3. SEM micrographs of (a) PVDF-5LLTO and (b) PVDF-15LLTO electrolytes.

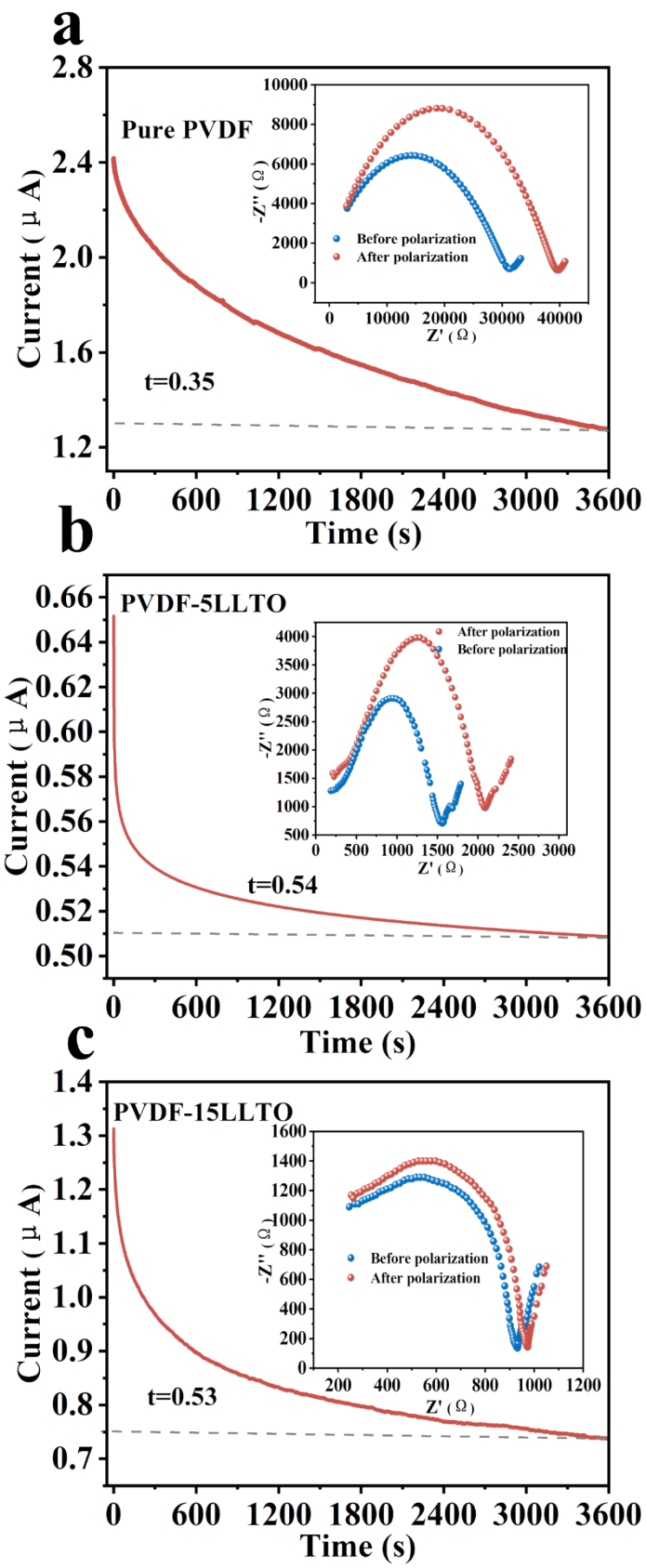

Figure S4. Polarization curves, and initial and steady-state impendence diagrams of (a) Pure PVDF, (b) PVDF-5LLTO and (c) PVDF-15LLTO CPEs.

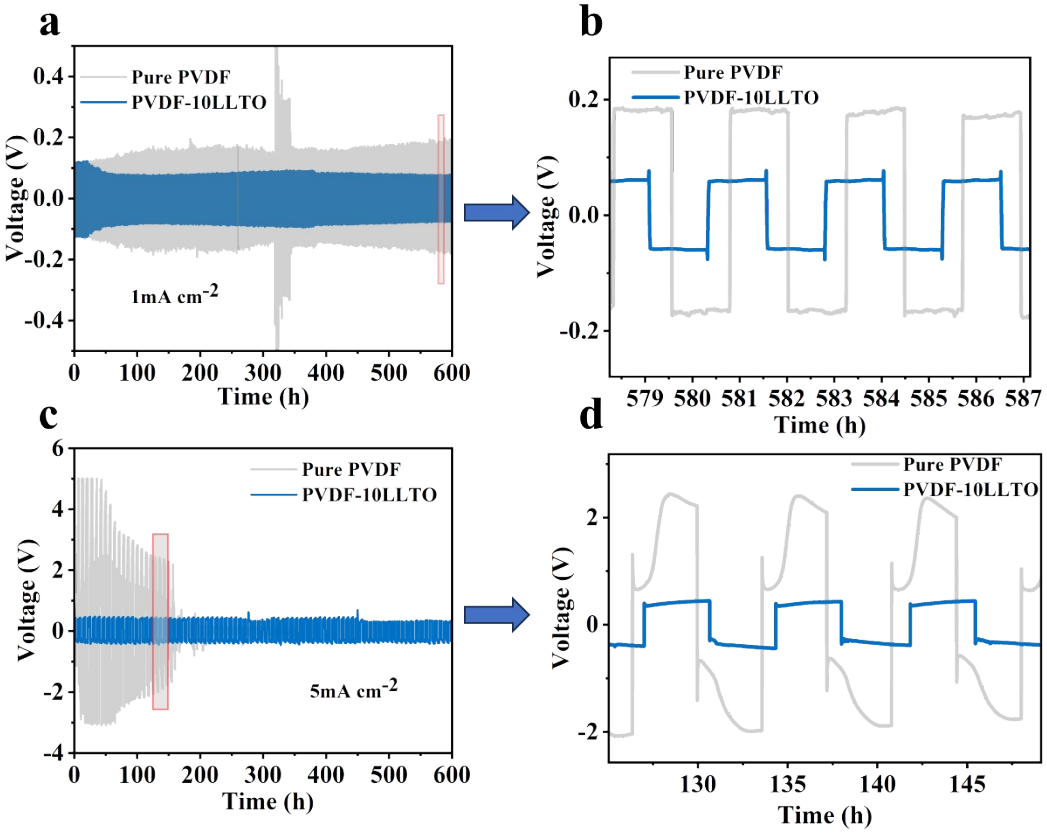

Figure S5. The charge-discharge performance diagram and its partial enlarged view with Pure PVDF and PVDF-10LLTO electrolytes at stripping/plating capacity of 1 mAh cm^{-2} and current density as (a, b) 1 mA cm^{-2} and (c, d) 5 mA cm^{-2}.

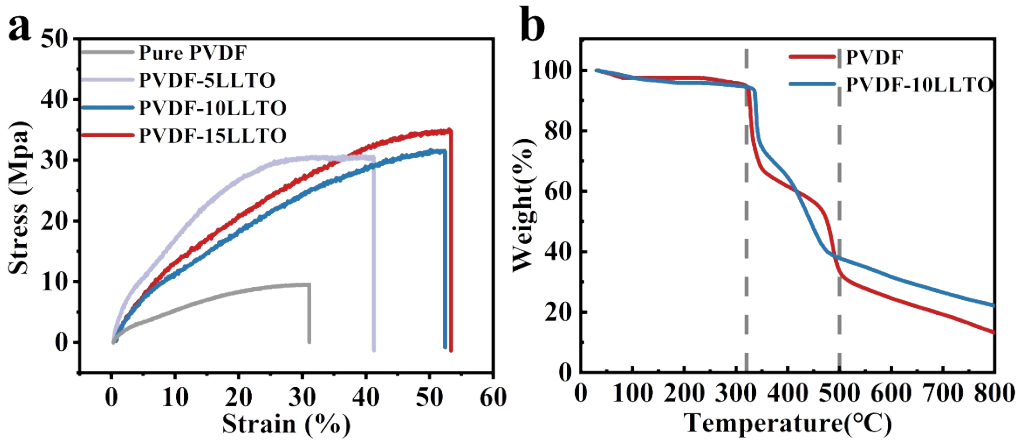

Figure S6. (a)Stress-Strain curve of PVDF-LLTO solid state electrolyte; (b)TG curve of Pure PVDF and PVDF-10LLTO.

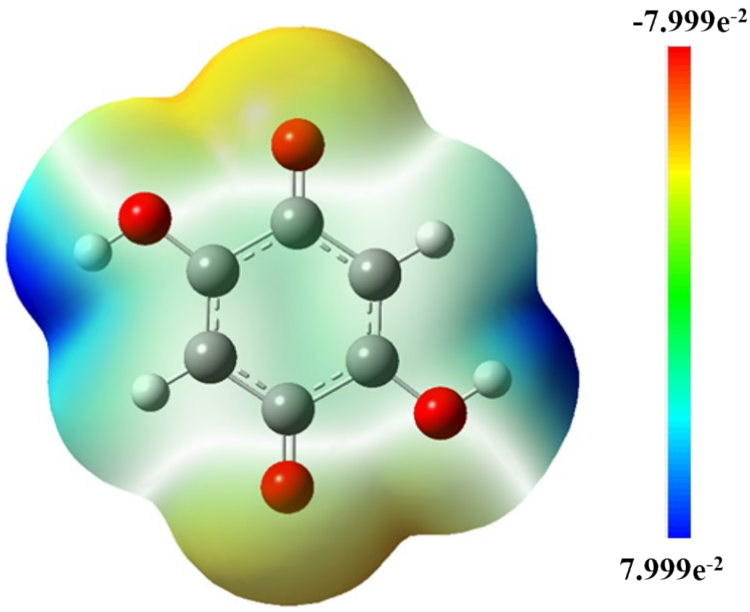

Figure S7. Static potential diagram of DHBQ; The calculations were carried out using Gaussian 09 program package. In order to better describe the bond structure, b3lyp functional combined with 6–31g(d) basis set was selected for geometry optimizations.[1]

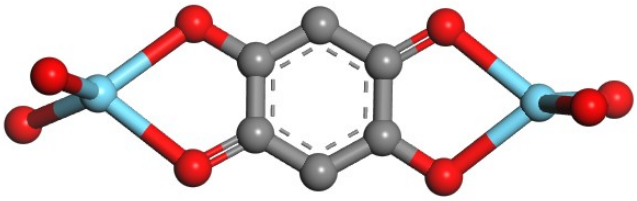

Figure S8. The π-d coordination structure.

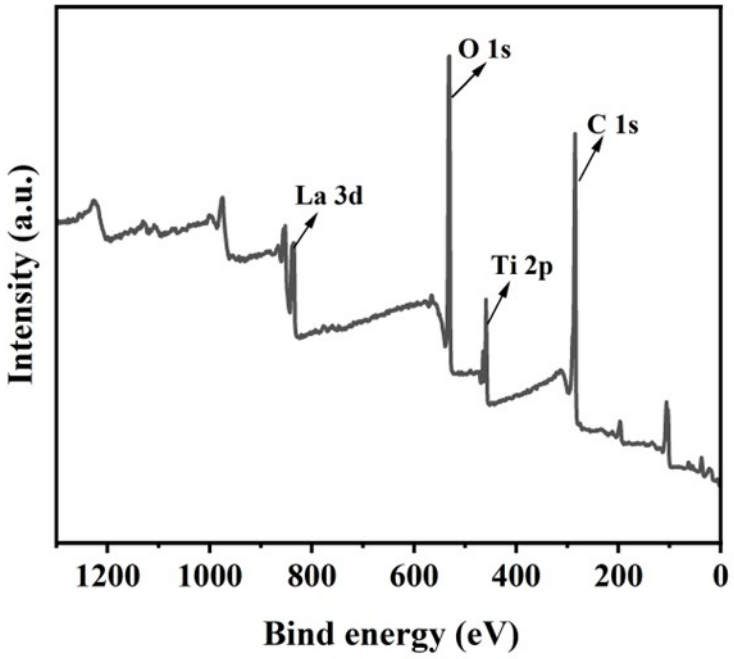

Figure S9. Full XPS spectra of DHBQ-LLTO.

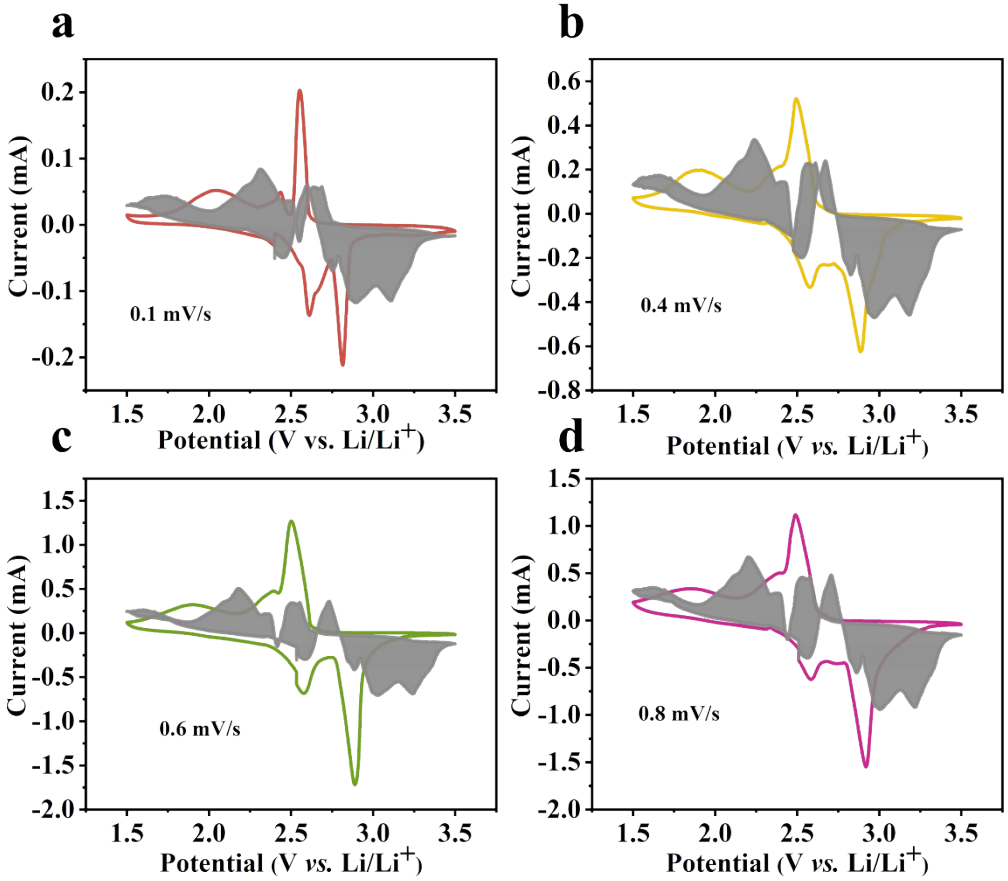

Figure S10. Capacitive contribution at (a) 0.1, (b) 0.4, (c) 0.6 and (d) 0.8 mV s^{-1}.

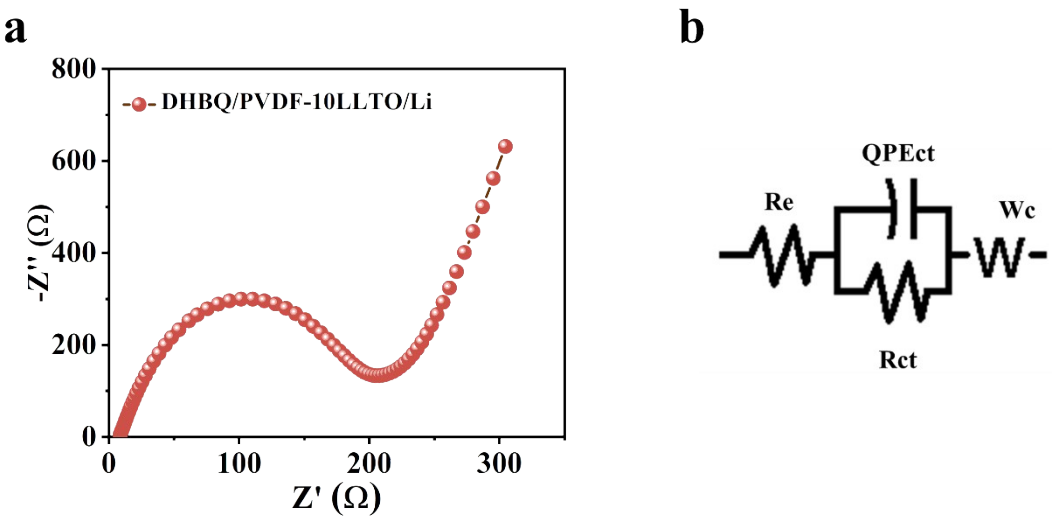

Figure S11. (a) AC impedance spectrum of DHBQ/PVDF-LLTO/Li ASSLB and (b) equivalent circuit diagram corresponding to AC impedance spectra.

Table S1. The element content of LLTO

Element	Wt%	At%
OK	24.66	65.69
AuM	15.32	03.32
TiK	21.58	19.20
LaL	38.44	11.79
Matrix	Correction	ZAF

Table S2. Ionic conductivity of Pure PVDF, PVDF-5LLTO, PVDF-10LLTO, PVDF-15LLTO CPEs at different temperatures.

Temperature	Pure PVDF	PVDF-5LLTO	PVDF-10LLTO	PVDF-15LLTO
30℃ ($S\ cm^{-1}$)	3.17×10^{-5}	2.57×10^{-4}	1.69×10^{-3}	4.03×10^{-4}
40℃ ($S\ cm^{-1}$)	9.12×10^{-5}	3.24×10^{-4}	2.24×10^{-3}	4.67×10^{-4}
50℃ ($S\ cm^{-1}$)	1.07×10^{-4}	4.30×10^{-4}	2.88×10^{-3}	6.34×10^{-4}
60℃ ($S\ cm^{-1}$)	1.23×10^{-4}	5.12×10^{-4}	3.50×10^{-3}	8.45×10^{-4}
70℃ ($S\ cm^{-1}$)	1.56×10^{-4}	7.23×10^{-4}	4.06×10^{-3}	1.10×10^{-3}
80℃ ($S\ cm^{-1}$)	1.86×10^{-4}	9.38×10^{-4}	4.74×10^{-3}	1.41×10^{-3}

Table S3. Activation energy of Pure PVDF, PVDF-5LLTO, PVDF-10LLTO, PVDF-15LLTO CPEs.

	Pure PVDF	PVDF-5LLTO	PVDF-10LLTO	PVDF-15LLTO
Ea（eV）	0.287	0.241	0.191	0.242

Reference:

[1] M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb J. R. Cheeseman, G. Scalmani, V. Barone, B. Mennucci, G. A. Petersson, et al, Gaussian 09, Revision D.01, Gaussian, Inc., Wallingford CT, 2013.