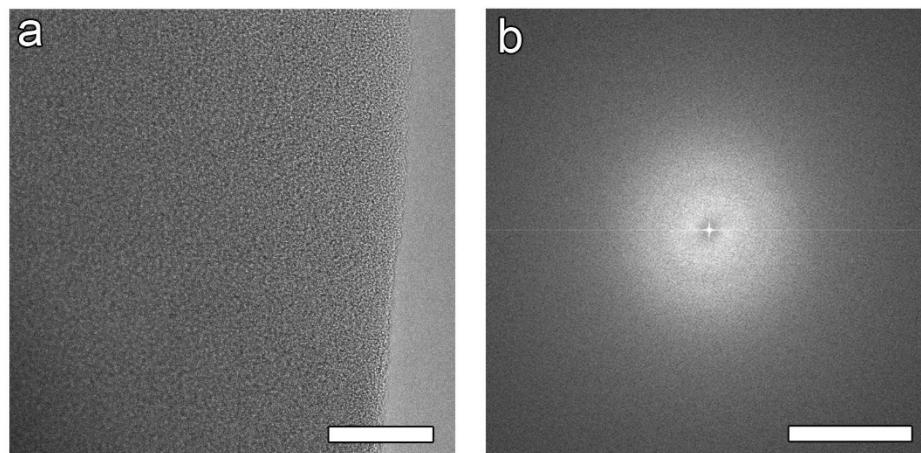


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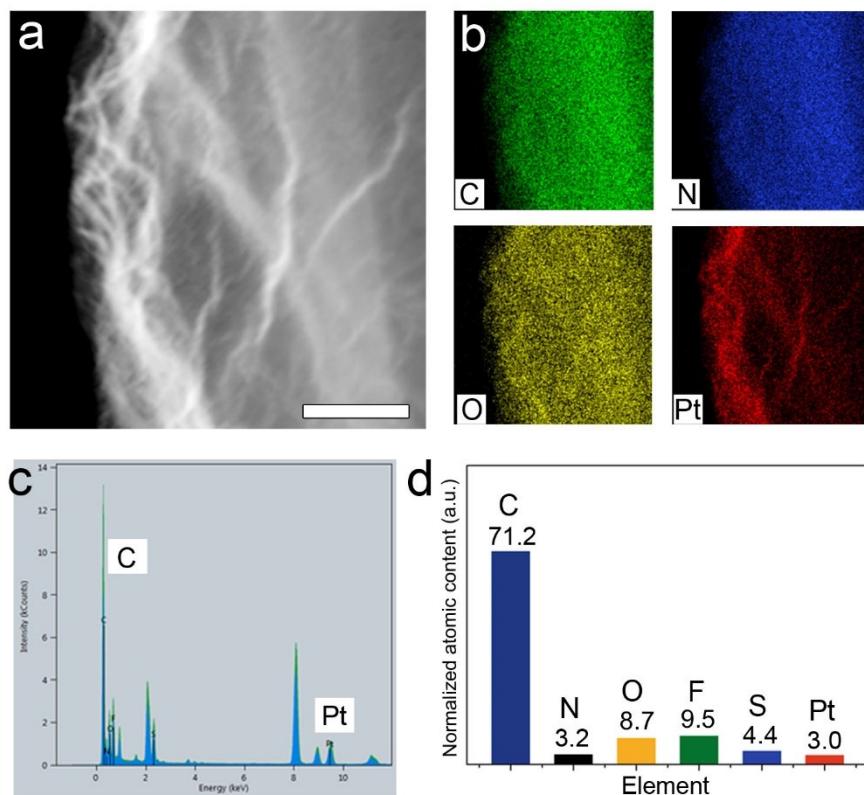
**Electronic supplementary information**

**Platinum Nano-Interlayer Enhanced Interface for Stable All-Solid-State Batteries Observed via Cryo-Transmission Electron Microscopy**

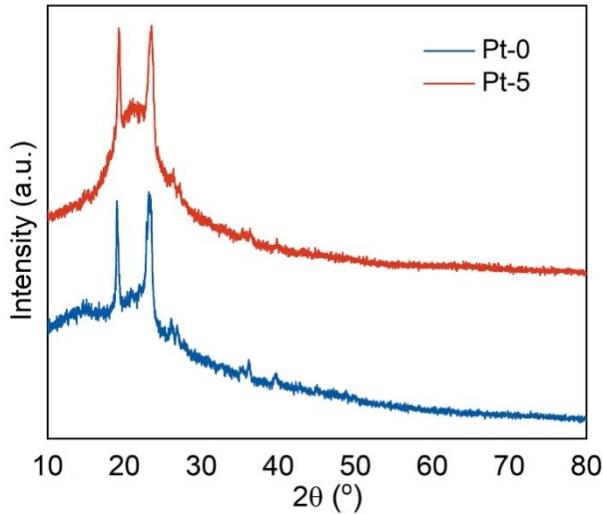
Ouwei Sheng,<sup>a</sup> Chengbin Jin,<sup>a</sup> Mei Chen,<sup>a</sup> Zhijin Ju,<sup>a</sup> Yujing Liu,<sup>a</sup> Yao Wang,<sup>a</sup> Jianwei Nai,<sup>a</sup> Tiefeng Liu,<sup>a</sup> Wenkui Zhang,<sup>a</sup> and Xinyong Tao<sup>a,\*</sup>



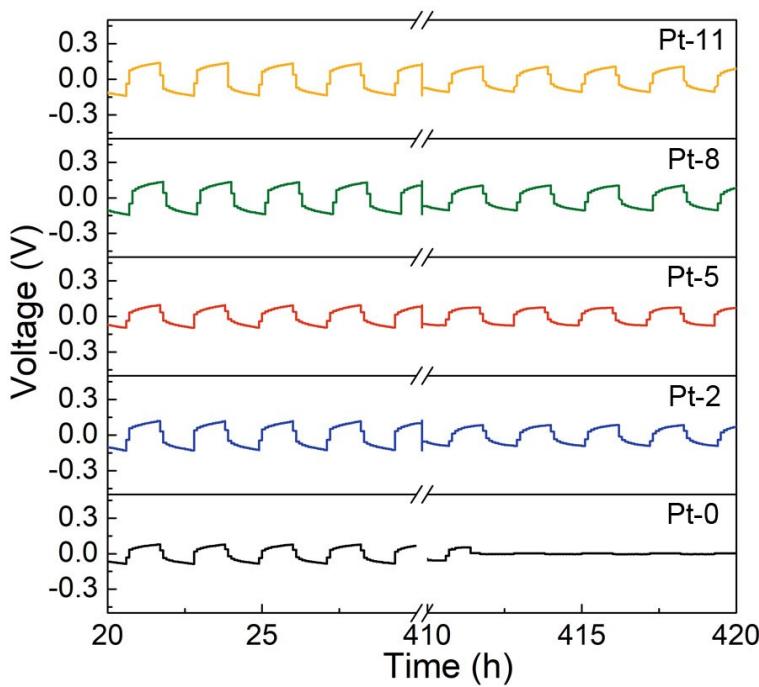
**Fig. S1** (a) The HRTEM image of pure PEO based electrolyte. (b) The corresponding FFT of (a). Scale bar: (a) 20 nm, (b) 5 nm<sup>-1</sup>.



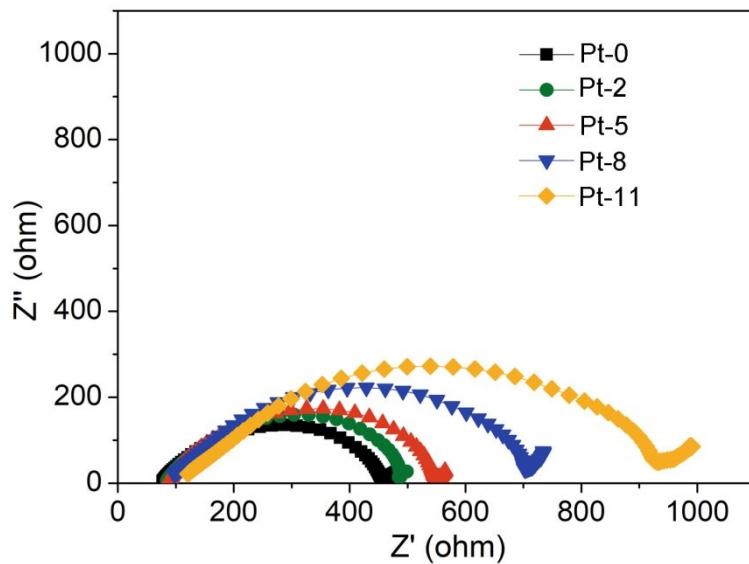
**Fig. S2** (a) The STEM image of Pt modified electrolyte. (b) The corresponding distribution of C, O, N and Pt elements. (c) The intensity spectrum of C, N, O, F, S, Cu and Pt elements. (d) The corresponding atomic content. Scale bar: (a) 50 nm.



**Fig. S3** XRD pattern of PEO electrolyte surface after circulation.

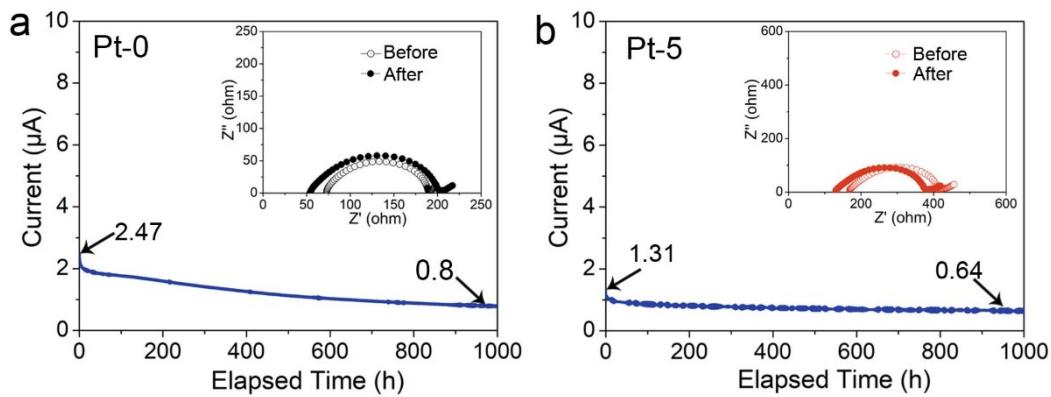


**Fig. S4** Local enlarged voltage profiles of the lithium (Li)-Li cells with Pt-0, Pt-2, Pt-5, Pt-8 and Pt-11 electrolytes at  $0.1 \text{ mA cm}^{-2}$  under  $50^\circ\text{C}$ .

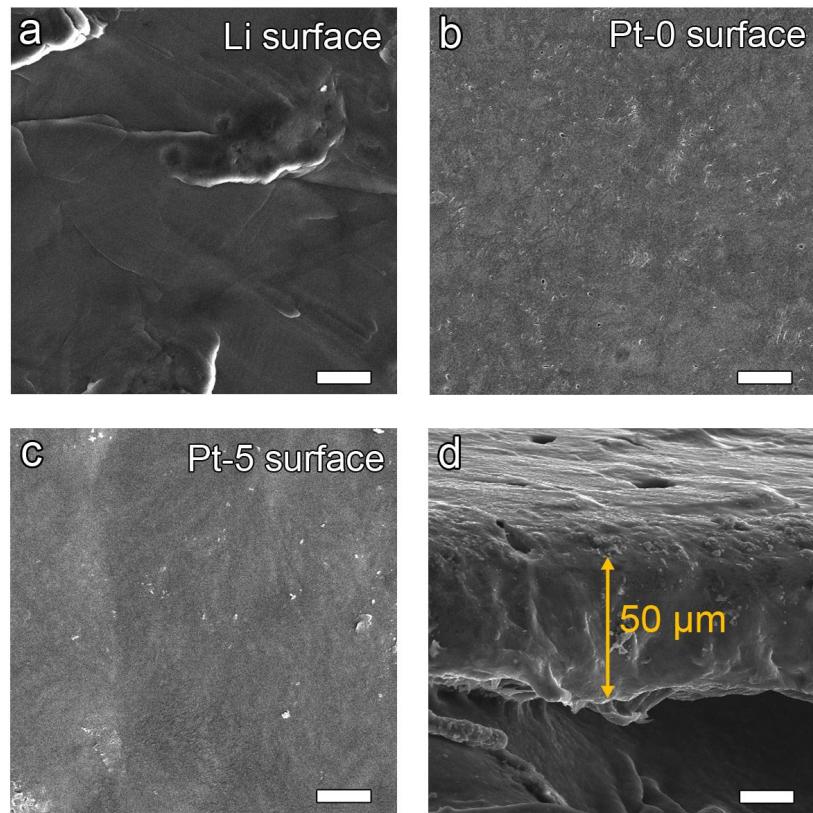


**Fig. S5** The impedance variation of Li-Li cell using different electrolyte before cycle.

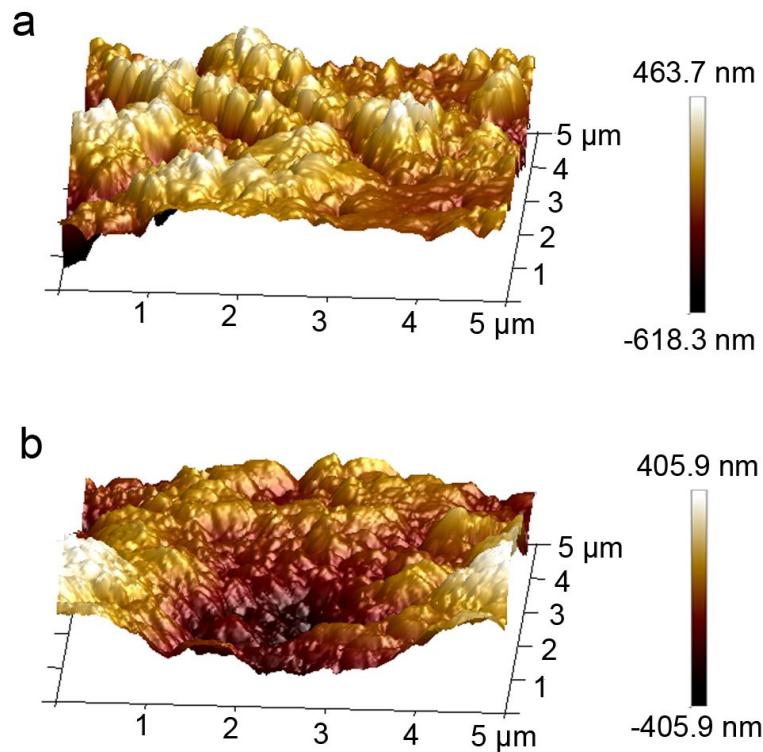
When sputtering 2, 5, 8, and 11 nm Pt on SPE surface, the impedance of Li/PEO interface is 400, 470, 600, and 800  $\Omega$ , respectively.



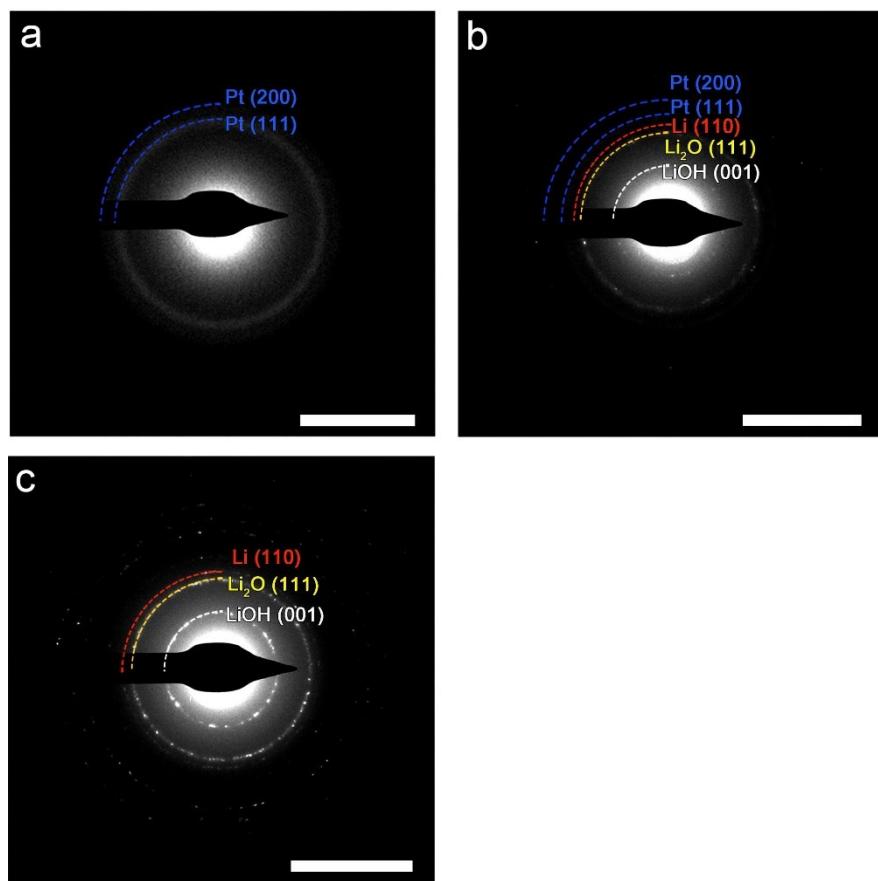
**Fig. S6** (a, b) Chronoamperometry of the (a) Li/Pt-0/Li and (b) Li/Pt-5/Li cells. Insets: the impedance spectra of the same cell before and after polarization.



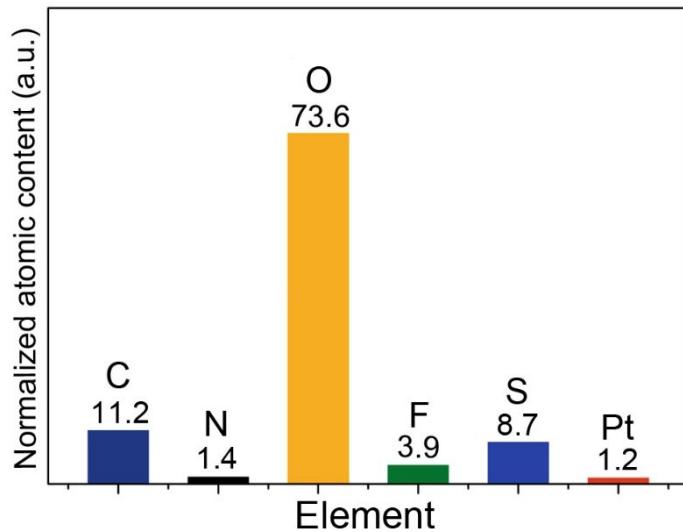
**Fig. S7** (a) SEM image of fresh Li metal before cycling. (b, c) SEM images of (b) Pt-0 electrolyte and (c) Pt-5 electrolyte before cycling. (d) Cross-sectional SEM image of the Pt-5 electrolyte where the thickness of SPE was indexed. Scale bar: (a-d) 20  $\mu\text{m}$ .



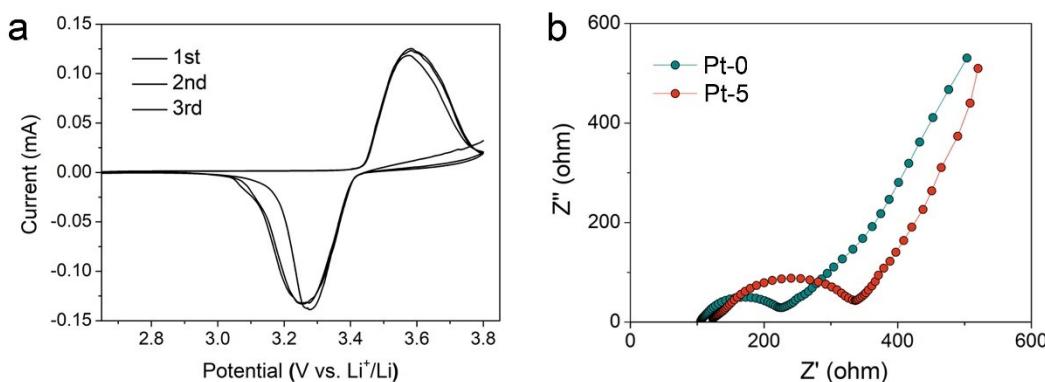
**Fig. S8** AFM height images of Li metal surface with (a) Pt-0 electrolyte and (b) Pt-5 electrolyte after 100 cycles.



**Fig. S9** Selective area electron diffraction (SAED) patterns of (a) Pt-5 electrolyte before Li deposition, (b, c) Pt-5 electrolyte after Li deposition. Scale bars, 5 nm<sup>-1</sup>. Green circle: Pt, 1.98, 2.29 Å, Red circle: Li, 2.48 Å, yellow circle: Li<sub>2</sub>O, 2.66 Å, white circle: LiOH, 4.35 Å.



**Fig. S10** The atomic content of C, N, O, F, S and Pt elements after Li deposition using Pt-5 electrolyte.



**Fig. S11** (a) CV curves of the LiFePO<sub>4</sub> full cell using the Pt-5 electrolyte in the voltage of 2.6–3.8 V at 50 °C. (b) EIS spectra of different ASSLMBs before cycling.

**Table S1** Electrochemical properties for Li-Li half-cells and Li-LiFePO<sub>4</sub> full-cells using different electrolyte in the recent years.

Electrolyte	Working Temp.	Li-Li cell	Li-LiFePO <sub>4</sub> cell	Year	Ref.
PI/PEO/LiTFSI	60 °C	1000 h (0.1 mA cm <sup>-2</sup> )	138 mAh g <sup>-1</sup> (0.5 C) 300 th	2019	<sup>1</sup>
PEO/CQDs-Li	60 °C	120 h (0.05, 0.1 mA cm <sup>-2</sup> )	120 mAh g <sup>-1</sup> (4 C) 97.1% (200 th)	2018	<sup>2</sup>
IPN9-10PPC	90 °C	300 h (1.5 mA cm <sup>-2</sup> )	141.5 mAh g <sup>-1</sup> (0.2 C) 92.7% (200 th)	2020	<sup>3</sup>
PEO-TEGDME-TEGDMA	25 °C	400 h (0.05 mA cm <sup>-2</sup> )	140 mAh g <sup>-1</sup> (0.1 C) 98.8% (100 th)	2019	<sup>4</sup>
LiEFA/PEO	70 °C	250 h (0.1 mA cm <sup>-2</sup> )	130 mAh g <sup>-1</sup> (C/3) 25 th	2019	<sup>5</sup>
LiFSI/FPE+LiFSI/PEO	70 °C	1000 h (0.1 mA cm <sup>-2</sup> )	130 mAh g <sup>-1</sup> (0.2 C) 200 th	2019	<sup>6</sup>
Pt-5	50 °C	2000 h (0.1 mA cm <sup>-2</sup> )	150 mAh g <sup>-1</sup> (0.5 C) 98% (270 th)	In this work	

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