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

Supporting Information

Interface Engineering in Solid State Li Metal Batteries by Quasi-2D Hybrid Perovskites

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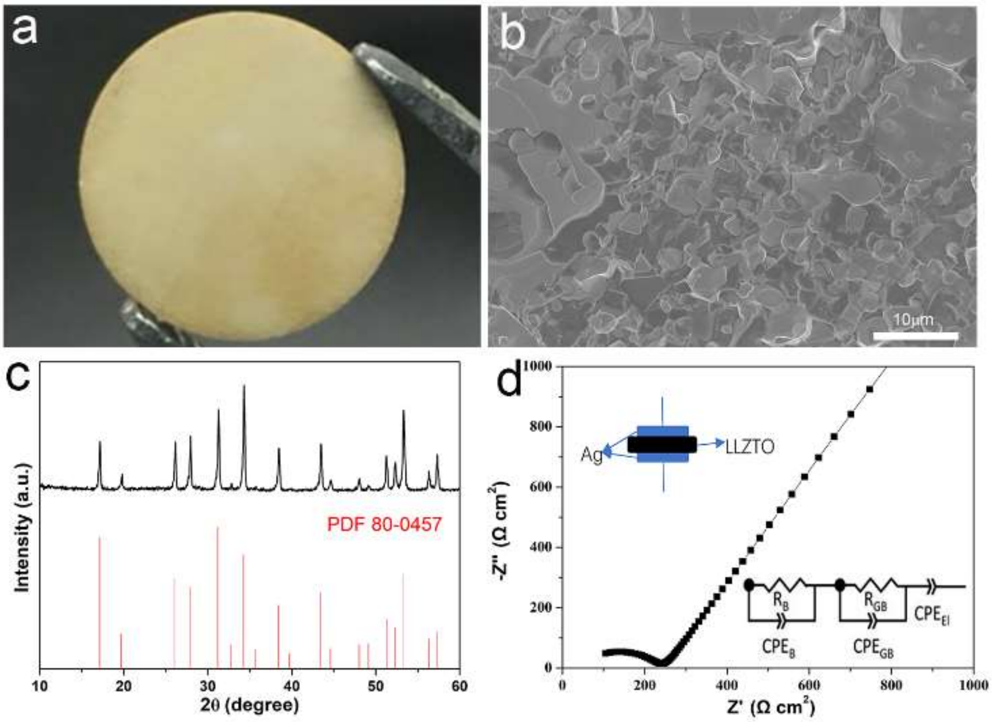

Figure S1. Digital photo of a polished yellowish LLZTO pellet (a), SEM image of cross-section (b) and XRD pattern (c) of LLZTO, EIS of Ag/LLZTO/Ag blocking symmetric cell at room temperature.

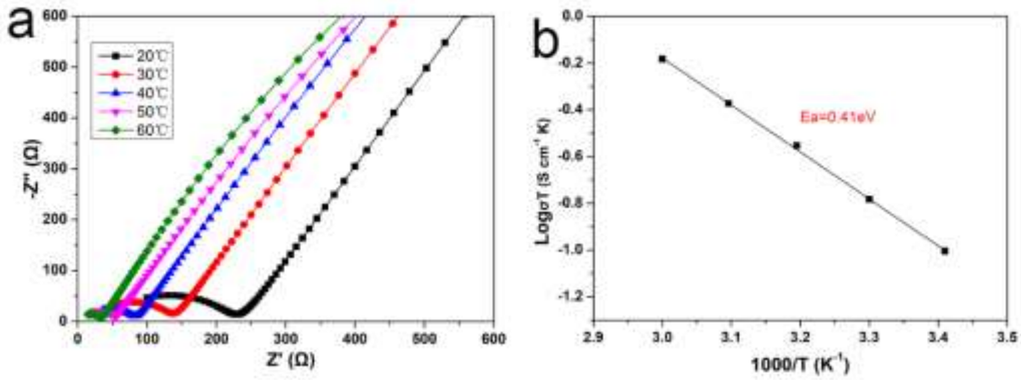

Figure S2. (a) EIS of LLZTO pellets under temperature range from 20–60℃, Arrhenius plot of the LLZTO ionic conductivity

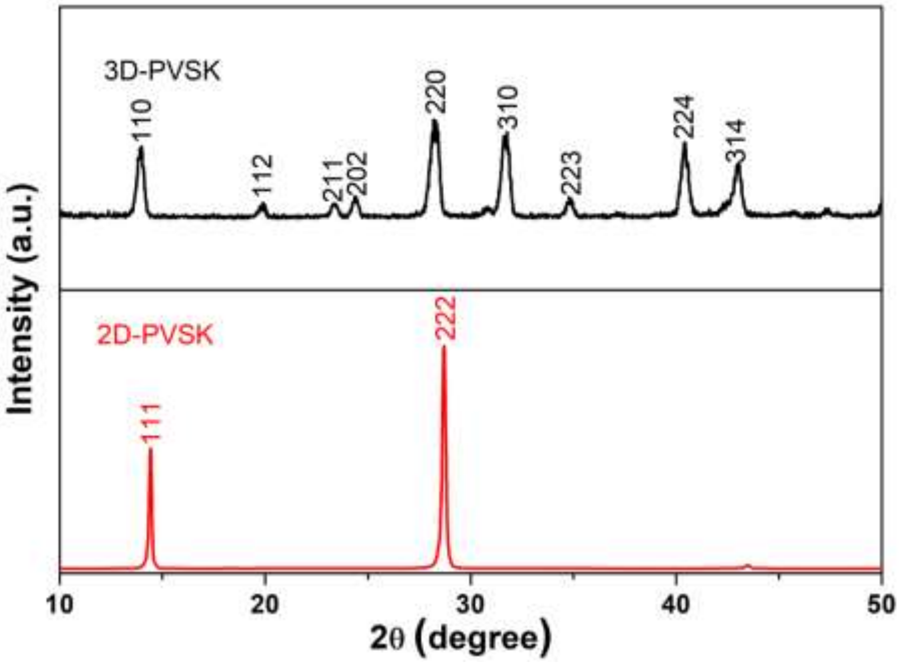

Figure S3. XRD patterns of as-prepared 3D-PVSK (black line) and 2D-PVSK (red line).

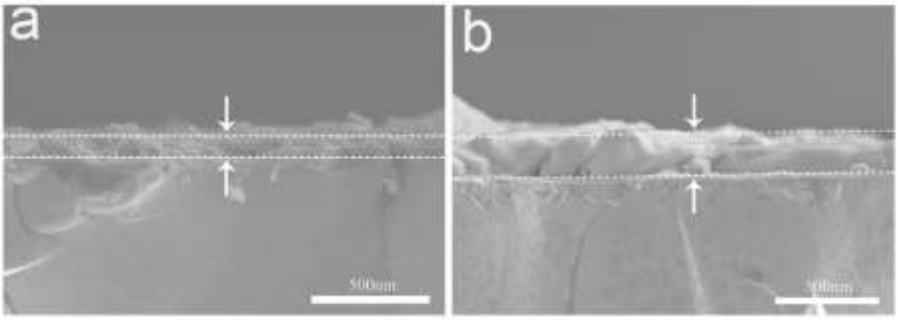

Figure S4. SEM images of cross-section for 3D-PVSK/LLZTO (a) and 2D-PVSK/LLZTO (b).

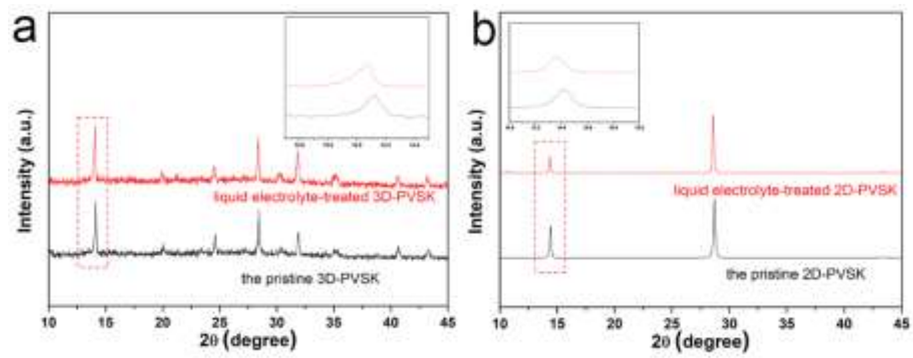

Figure S5. XRD patterns of the pristine 3D-PVSK and liquid electrolyte-treated 3D-PVSK (a), the pristine 2D-PVSK and liquid electrolyte-treated 2D-PVSK (b).

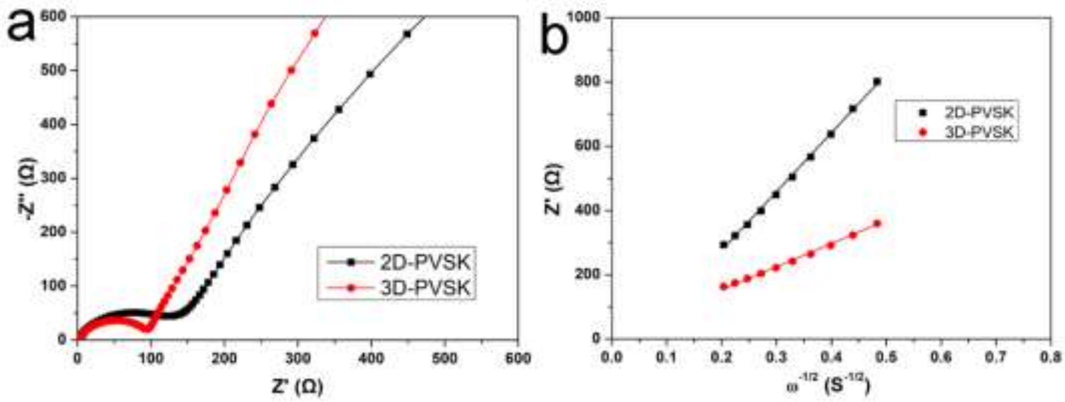

Figure S6. Nyquist plots (a) and plots of $\omega^{1/2}$ versus Z' (b) of the 3D-PVDK and 2D-PVSK electrodes.

The electrochemical reaction mechanism of Li^+ with PVSK and lithium diffusion coefficient can be described as follows:[1,2]

$$CH_3NH_3PbI_3 + nLi^+ + ne^- \leftrightarrow Li_nCH_3NH_3PbI_3 \quad (1)$$

$$(C_4H_9NH_3)_2(CH_3NH_3)_3Pb_4I_{13} + nLi^+ + ne^- \leftrightarrow Li_n(C_4H_9NH_3)_2(CH_3NH_3)_3Pb_4I_{13} \quad (2)$$

$$Zreal = R + \sigma\omega^{-1/2} \quad (3)$$

$$D = R^2T^2/2A^2n^4F^4C^2\sigma^2 \quad (4)$$

According to the previous report,[3] n is the number of electrons transferred in the lithiation process, which is as high as n = 3. The Warburg factor (σ) can be determined to be 1814 and 703 by fitting of Z' with $\omega^{-1/2}$ in Figure S6b. R is the gas constant (8.314 J mol^{-1} K^{-1}), T is the Kelvin temperature (298 K), A is the area of the electrode (1.1 cm^2), F is the Faraday constant (96486 C mol^{-1}), C is the concentration of Li^+ in the lattice (6.596×10^{-3} mol cm^{-3}), D is the diffusion coefficient (cm^2 s^{-1}), the Li^+ diffusion coefficients were calculated to be 2.52×10^{-18} and 1.68×10^{-17} cm^2 s^{-1} for the 2D-PVSK and 3D-PVSK, respectively. These results indicated that the 3D-PVSK presents lower charge transfer resistance and hence higher Li^+ diffusion coefficient, which is favorable for rapid charge and discharge process.

References

1. Ge P.; Hou H.; Li S.; Yang L.; Ji X., *Adv. Funct. Mater.* 2018, **28**, 1801765.
2. Wu X.; Yuan X.; Yu J.; Liu J.; Wang F.; Fu L.; Zhou W.; Zhu Y.; Zhou Q.; Wu Y., *Nanoscale* 2017, **9**, 11004-11011.
3. Vicente N.; Garcia-Belmonte G., *J. Phys. Chem. Lett.* 2017, **8**, 1371-1374.