Supporting Information

Quasi-Solid Polymer Electrolyte Initiated by Two-Dimensional Functional Nanosheets for

Stable Lithium Metal Batteries

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Fig. S1. X-ray powder diffraction (XRD) patterns of K₃Sb₃P₂O_{14.}



Fig. S2. AFM imaging of $Sb_3P_2O_{14}^{3-}$ nanosheets with a corresponding height profile (inset).



Fig. S3. The thermogravimetric curves of a) GPE and b) LE.



Fig. S4. X-ray powder diffraction (XRD) patterns of poly-DOL.



Fig. S5. Gel Permeation Chromatography (GPC) of GPE.



Fig. S6. The ionic conductivity of a) GPE; b) 0.1 wt% $H_3Sb_3P_2O_{14}$ &1 M LiTFSI in DOL; c) 0.3 wt% $H_3Sb_3P_2O_{14}$ &1 M LiTFSI in DOL&EC&FEC d) 0.3 M LiPF₆+0.7 M LiTFSI in DOL&EC&FEC.



Fig. S7. Electrochemical impedance spectroscopy (EIS) curves of GPE-Li and LE-Li symmetric cells.



Fig. S8. CEs for the 0.3 M LiPF₆+0.7 M LiTFSI in DOL&EC&FEC and 0.3 wt% $H_3Sb_3P_2O_{14}$ &1 M LiTFSI in DOL&EC&FEC at a 1 mA cm⁻² and 1 mA h cm⁻².



Fig. S9. Long-term cycling of symmetrical Li cells for the 0.3 M LiPF₆+0.7 M LiTFSI in DOL&EC&FEC and 0.3 wt% $H_3Sb_3P_2O_{14}$ &1 M LiTFSI in DOL&EC&FEC at a 1 mA cm⁻² and 1 mA h cm⁻².



Fig. S10. a) Electrochemical impedance spectroscopy and b) corresponding magnified view for Li||LE||Li cell after different plating/stripping cycles.



Fig. S11. a) Electrochemical impedance spectroscopy and b) corresponding magnified view for Li||GPE||Li cell after different plating/stripping cycles.



Fig. S12. a) SEM image and b) magnified SEM image of LiFePO₄ powder.



Fig. S13. The rate performance of $Li \| LiFePO_4$ full cells.



Fig. S14. Electrochemical impedance spectroscopy (EIS) of Li||LiFePO₄ full cells a) before and b) after 100 cycles at 1 C.



Fig. S15. The surface SEM images and corresponding mapping images for a-c) LiFePO₄ cathode and d-f) Li anode in the GPE system after 100 cycles at 1 C.



Fig. S16. The surface SEM images of a) LiFePO₄ cathode and b) Li anode in the LE system after 100 cycles at 1 C.

Initiator	Electrolyte	Current density (mA cm ⁻²)	Capacity (mAh cm ⁻²)	Coulombic efficiency (%)	Cycle number	Reference
LiPF ₆	2 M LiPF ₆ + 1 M LiTFSI / PDOL +DME (1:1 , v/v)	1	1	~90	100	1
LiPF ₆	1 M LiPF ₆ +EC/DOL/	1	\sim	/	100	2
LiPF ₆	1 M LiTFSI+ LiPF ₆ in DOL/FEC/MP	0.1	0.2	\sim 94	200	3
TB	2 M LiTFSI+ 3wt%TB in DOL	0.5	0.5	~86.2	200	4
Al(OTf) ₃	0.5 mM Al(OTf) ₃ + 2 m LiTFSI/DOL	1	1	\sim 98	300	5
Sn(OTf) ₂	2 mM Sn(OTf) ₂ +2M in LiTFSI/DOL	1	1	~98	120	6
H ₃ Sb ₃ P ₂ O ₁₄	1 M LiTFSI+0.1wt%	0.5	0.5	~94.2	200	This
	H ₃ Sb ₃ P ₂ O ₁₄ in DOL/EC/FEC	1	1	~94.3	100	- work

 Table S1. The comparison of coulombic efficiency of various poly-DOL electrolytes.

Initiator	Electrolyte	Current density (mA cm ⁻²)	Capacity (mAh cm ⁻²)	Voltage hysteresis (mV)	Life (h)	Reference
LiPF ₆	2 M LiPF ₆ + 1 M LiTFSI + DOL + DME (1:1 , v/v)	1	1	\sim 50	400	1
LiPF ₆	2 M LiTFSI + 0.3 M LiDFOB/DOL+SN	0.5	0.5	~85	700	7
LiPF ₆	1 M LiTFSI +LiPF ₆ (1 wt %) +DADS (1 wt %)/DOL	1	1	/	500	8
LiPF ₆	1 M LiTFSI+1 M LiPF ₆ in DME/DOL	0.5	1	$\sim \! 100$	1200	9
LiPF ₆	1 M LiTFSI+ LiPF ₆ in DOL/FEC/MP	1	1	/	400	3
LiPF ₆	2.5 M LiPF6 + 1 M LiTFSI in DOL	1	1	/	450	10
nano Al ₂ O ₃	1 M LiTFSI+4%wt% Al ₂ O ₃ in DME/DOL	1	1	~44	1000	11
LiBF_4	2.0 wt% TTE+1.0 M LiTFSI+ 0.2 M LiBF ₄ in DOL	1	1	\sim 70	1000	12
ZnCl ₂	1.0 M LiTFSI in DOL and DME $(1:1 \text{ v/v}) + 5 \text{ wt\% LiNO}_3$	1	1	/	660	13
Al(OTf) ₃	0.5 mM Al(OTf) ₃ + 2 m LiTFSI/DOL	1	1	/	200	5
Al(OTf) ₃	0.4 mM Al(OTf) ₃ + 2 m LiTFSI/DOL	0.25	0.5	/	800	14
Sc(OTf) ₃	2 M LiTFSI + 7 mM Sc(OTf) ₃ in DOL/EC/EMC	0.5	0.5	~47	500	15
Sn(OTf) ₂	2 mM Sn(OTf) ₂ +2M in LiTFSI/DOL	1	1	/	200	6
H ₃ Sb ₃ P ₂ O ₁₄	1 M LiTFSI+0.1wt% H ₃ Sb ₃ P ₂ O ₁₄ in DOL/EC/FEC	1	1	50	500	This work

Table S2. The performance comparison of various poly-DOL electrolytes in symmetric Li || Li cell.

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