Electronic Supplementary Material (ESI) for Dalton Transactions.

Supporting Information

Design of Thiol-Lithium Ion Interaction in Metal-Organic Framework for High-Performance Quasi-Solid Lithium Metal Batteries

Qi Zhang, [†a,b] *Yingbo Xiao,* [†a] *Qi Li,*[a] *Jia Wang,*[a] *Sijia Guo,*[a] *Xin Li,*[a] *Yuan Ouyang,*[a] *Qinghan Zeng,*[a] *Wenchao He*[a] *and Shaoming Huang*[a,b,*]

[a]Guangzhou Key Laboratory of Low-Dimensional Materials and Energy Storage Devices, School of Materials and Energy, Guangdong University of Technology, Guangzhou, 510006, China
[b]Synergy Innovation Institute of GDUT, Heyuan, 517000, China.
[†] These authors contributed equally.
*Corresponding author: E-mail: smhuang@gdut.edu.cn

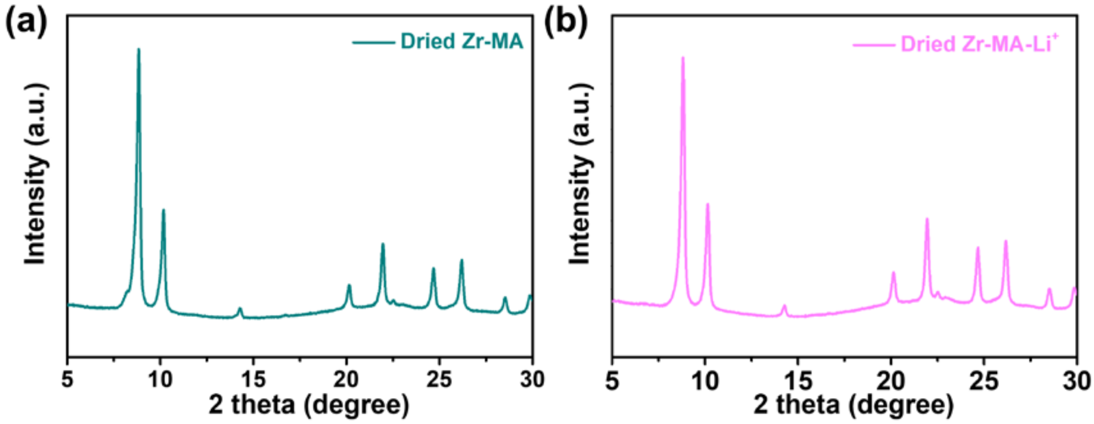

Fig. S1 The XRD patterns of dried Zr-MA (a) and dried Zr-MA-Li^+ (b).

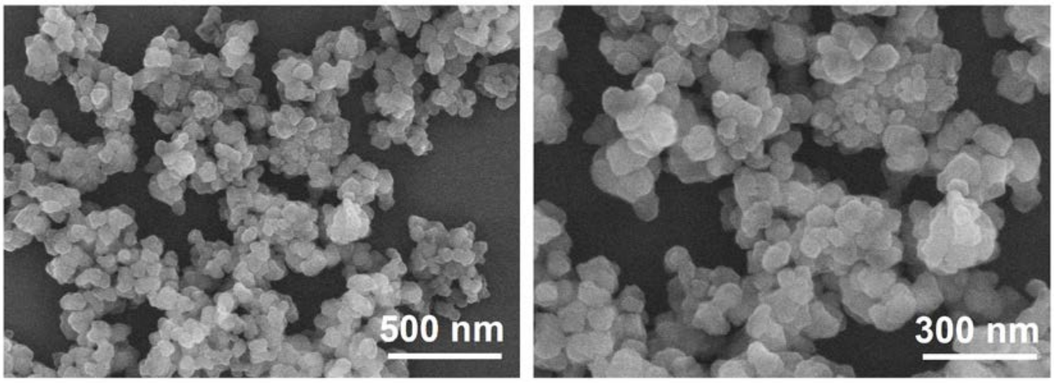

Fig. S2 SEM images for Zr-MA-Li^+.

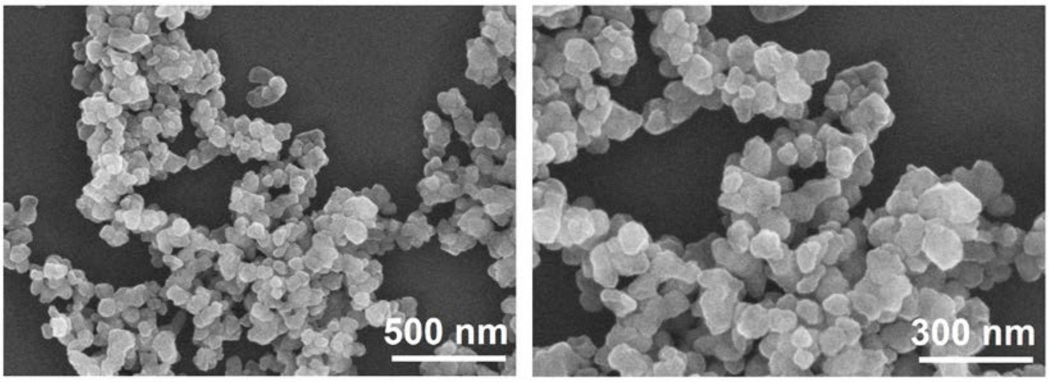

Fig. S3 SEM images for Zr-MA-Na^+.

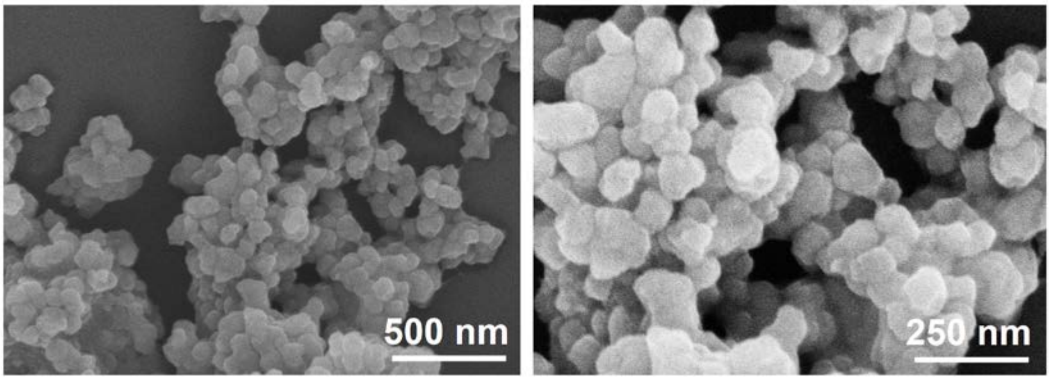

Fig. S4 SEM images for Zr-MA-K^{+}.

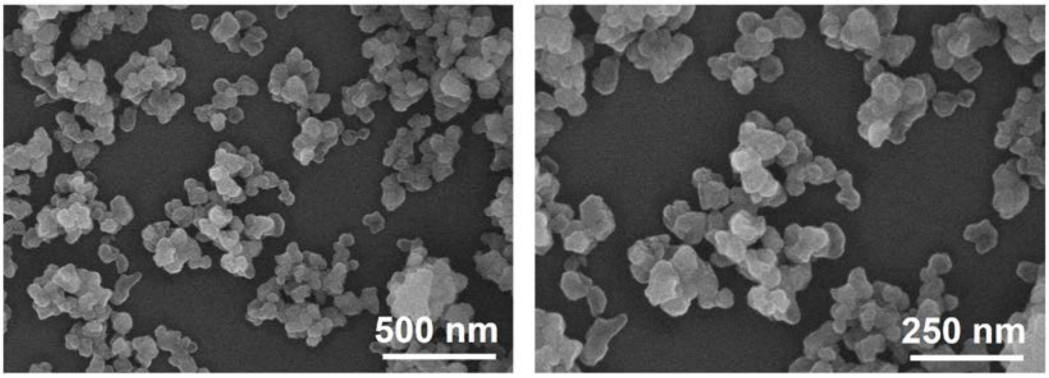

Fig. S5 SEM images for Zr-MA-Zn^{2+}.

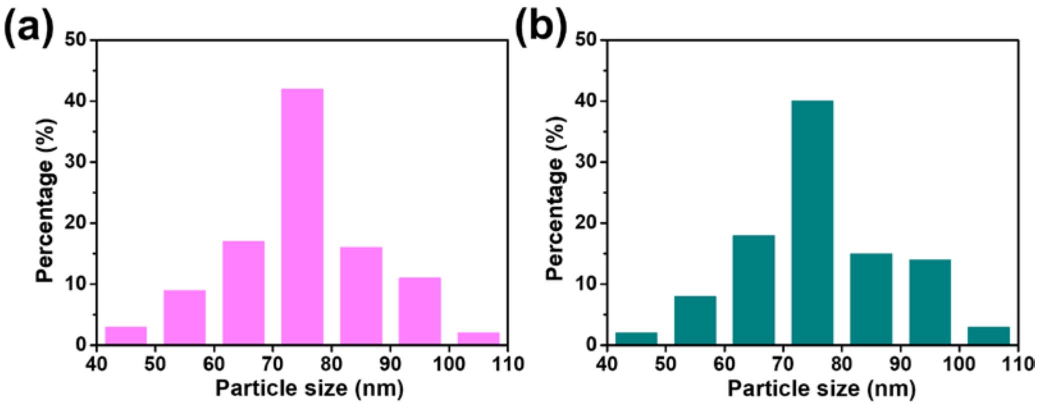

Fig. S6 Particle size distribution of Zr-MA (a) and Zr-MA-Li^{+} (b).

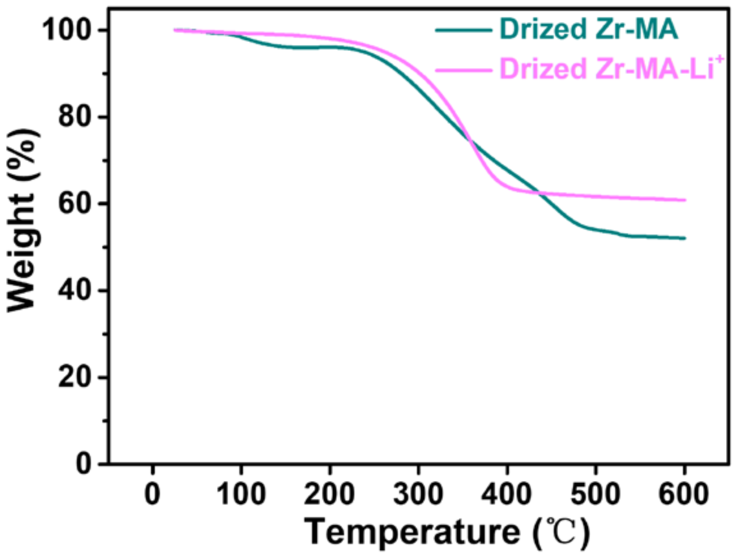

Fig. S7 The TGA curves of dried Zr-MA and dried Zr-MA-Li^+.

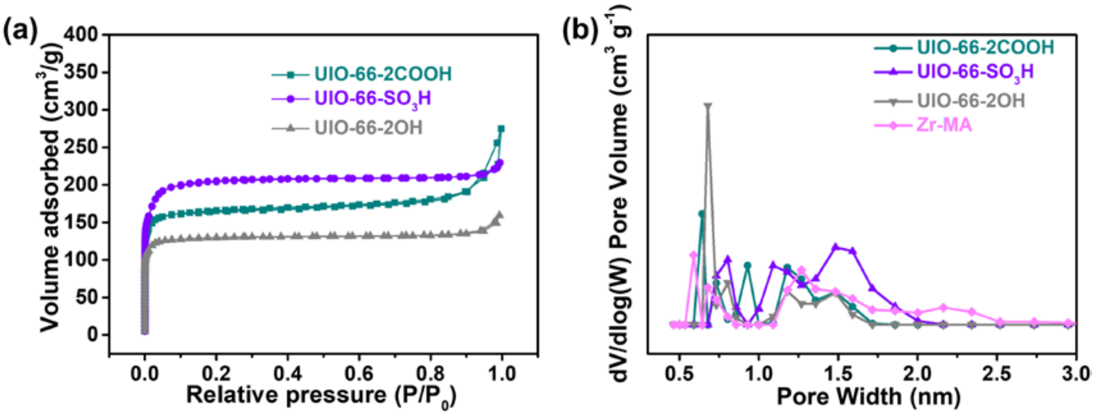

Fig. S8 (a) Nitrogen adsorption-desorption isotherms, and (b) corresponding pore size distribution of UIO-66-2COOH, UIO-66-SO_3H, UIO-66-2OH and Zr-MA.

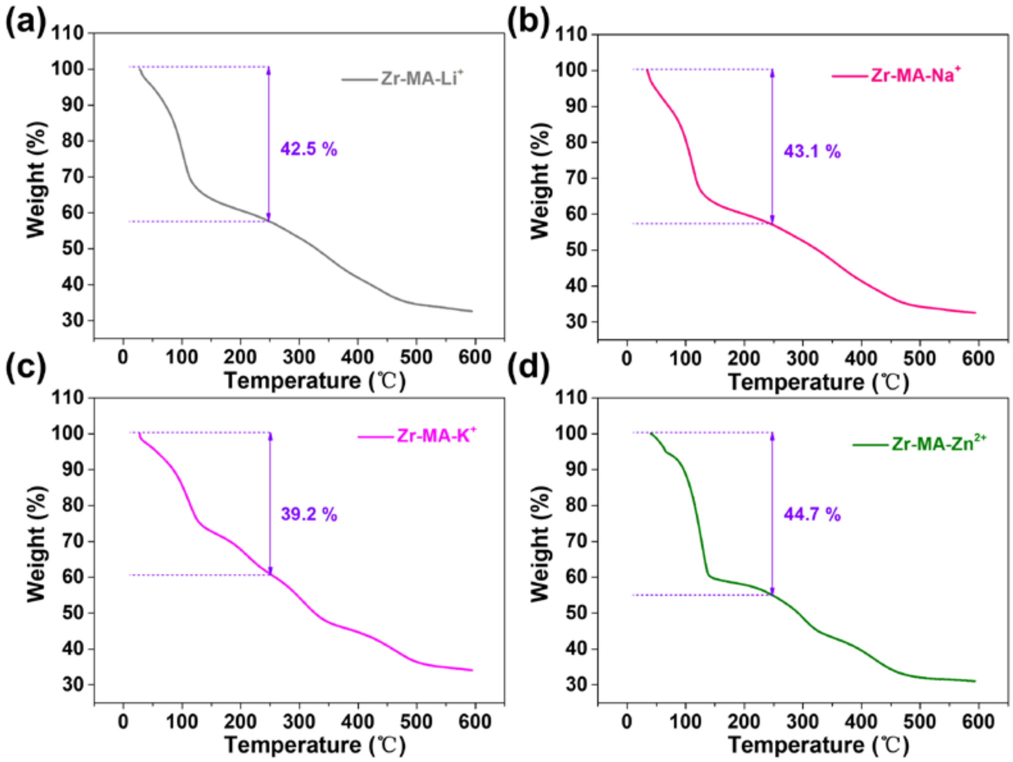

Fig. S9 The TGA curves of Zr-MA-Li^+ (a), Zr-MA-Na^+ (b), Zr-MA-K^+ (c) and Zr-MA-Zn^{2+} (d).

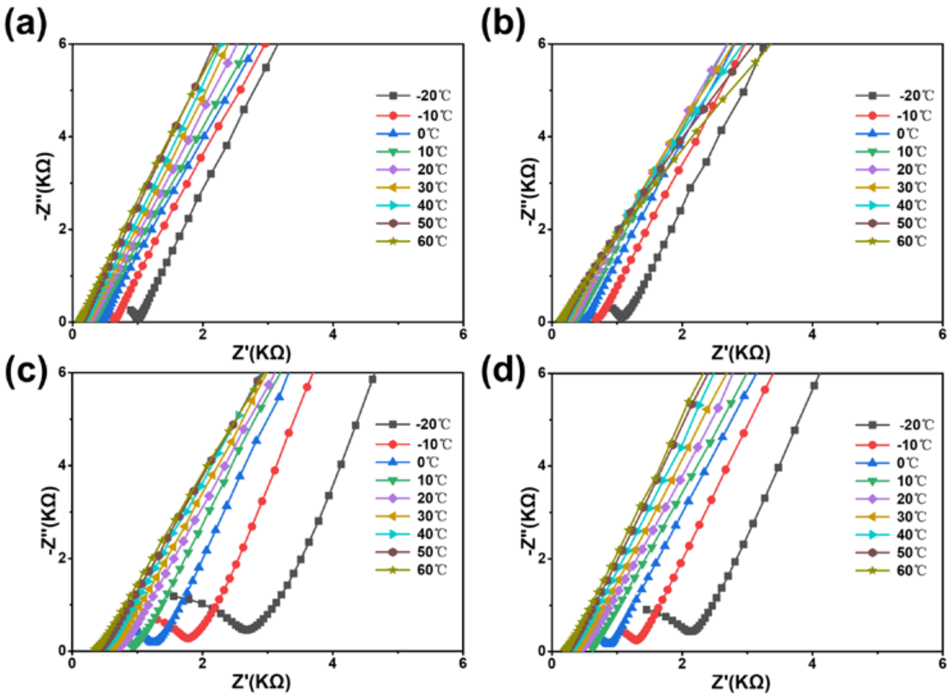

Fig. S10 EIS within frequency of 1×10^{-5} Hz to 10 Hz of (a) Zr-MA-Li^+, (b) Zr-MA-Na^+, (c) Zr-MA-K^+, and (d) Zr-MA-Zn^{2+} at temperatures from -20 ℃ to 60 ℃.

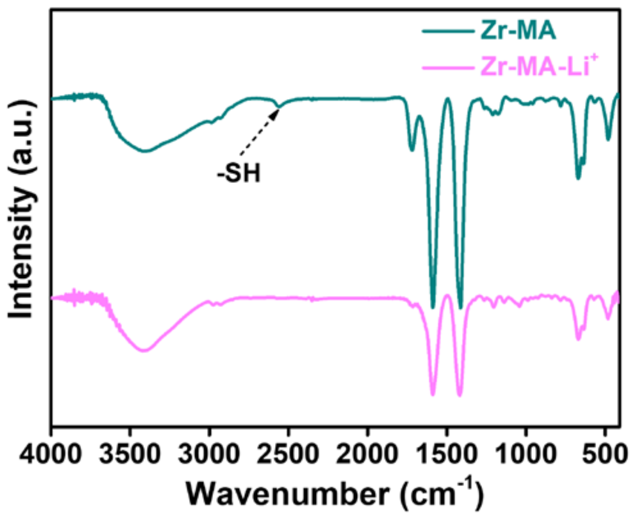

Fig. S11 FT-IR spectra of Zr-MA and Zr-MA-Li$^+$.

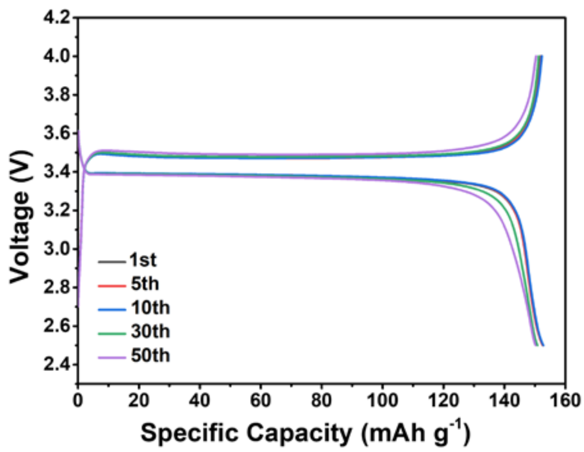

Fig. S12 The charge-discharge profiles of Li|SE-Zr-MA-Li$^+$|$LiFePO_4$ batteries under 0.2 C.

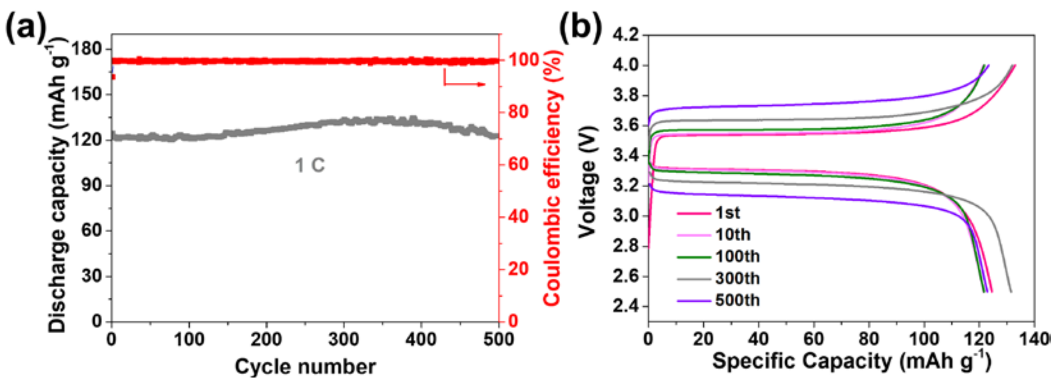

Fig. S13 (a) The cycle performance with Coulombic efficiency and (b) charge-discharge profiles of Li|SE-Zr-MA-Li$^+$|$LiFePO_4$ batteries at 1 C.

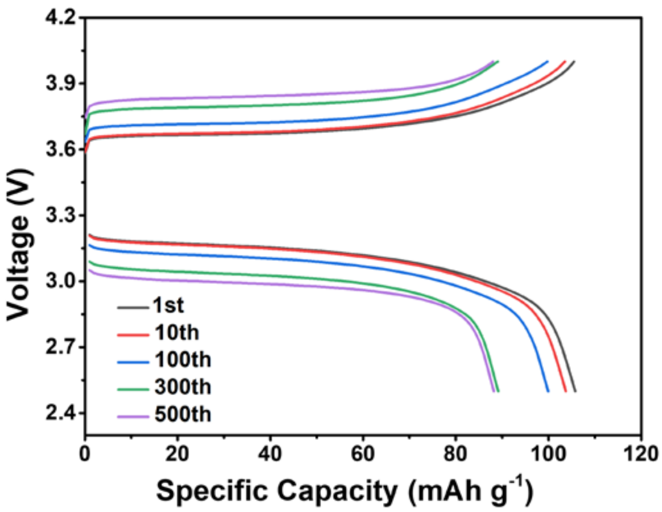

Fig. S14 The charge-discharge profiles of Li|SE-Zr-MA-Li$^+$|$LiFePO_4$ batteries under 2 C.

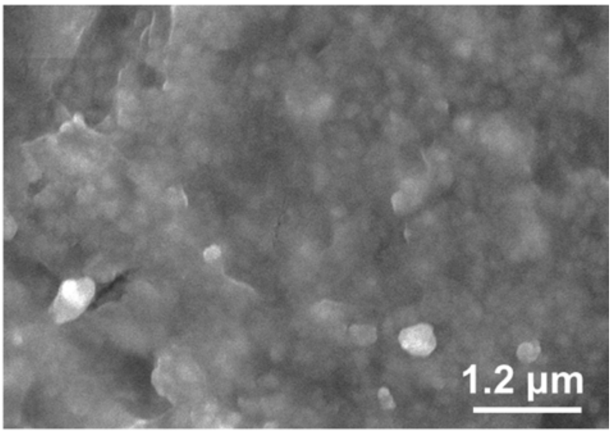

Fig. S15 SEM of SE-Zr-MA-Li$^+$ after long-cycle cycling.

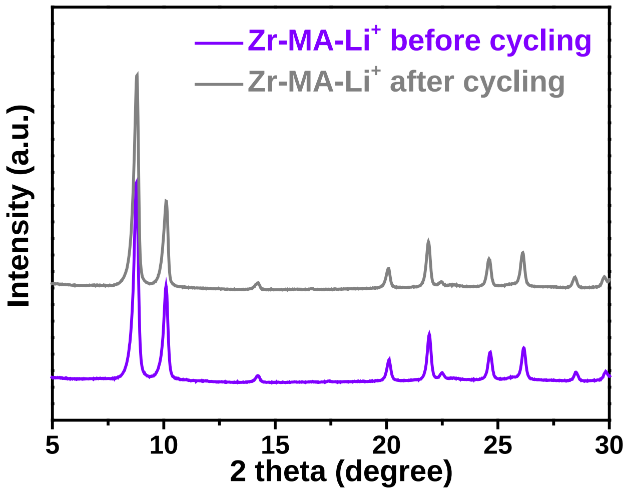

Fig. S16 PXRD of Zr-MA-Li$^+$ before and after long-cycle cycling.

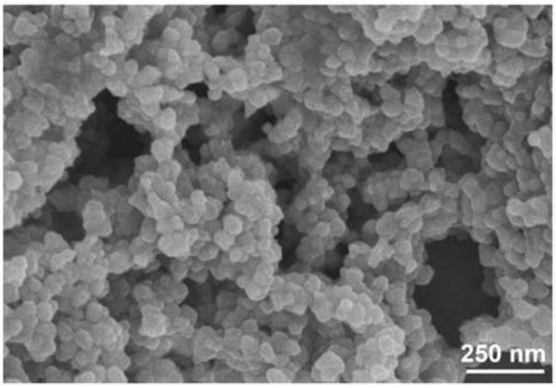

Fig. S17 SEM of Zr-MA-Li^+ after long-cycle cycling.

Tab. S1 Elemental analysis results of Zr-MA.

	Calculated					Found			
	Zr%	**C%**	**H%**	**O%**	**S%**	**C%**	**H%**	**O%**	**S%**
Zr-MA	34.8	18.4	1.8	32.7	12.3	20.3	2.79	28.5	12.2

Tab. S2 The comparison of pKa for thiophenol, phenol, benzenesulfonic acid, and benzoic acid.

	Thiophenol	**Phenol**	**Benzenesulfonic acid**	**Benzoic acid**
pKa	10.2	9.95	4.19	4.21

Tab. S3 The ion conductivities of Zr-MA-Li^+, Zr-MA-Na^+, Zr-MA-K^+, Zr-MA-Zn^{2+} at different temperatures.

Samples	**Ionic conductivity (×10^{-4} S cm^{-1})**								
	-20℃	**-10℃**	**0℃**	**10℃**	**20℃**	**30℃**	**40℃**	**50℃**	**60℃**
Zr-MA-Li^+	1.78	2.78	3.97	5.29	6.92	8.66	10.8	13.7	17.4
Zr-MA-Na^+	1.52	2.36	3.36	4.57	6.01	7.69	9.75	12.1	14.3
Zr-MA-K^+	0.65	1.00	1.44	1.89	2.42	2.89	3.43	4.08	4.98
Zr-MA-Zn^{2+}	0.84	1.13	2.03	2.83	3.82	4.90	6.25	7.75	9.28

Tab. S4 Comparison of electrochemical performances of $LiFePO_4$|Li cells with MOFs based on solid state electrolytes.

Electrolyte	Cycle performance (mAh g^{-1})	Rate performance (mAh g^{-1})	Ref.
LPC@UM	about 94 (1 C, 500 cycles, 25 °C)	146 (0.2 C, 25 °C) 106 (2 C, 25 °C)	1
Li-IL@MOF	132 (1 C, 100 cycles, 25 °C) ($LiFePO_4$ loading of ≈25 mg cm^{-2})	68 (0.05 C, -20 °C) 93 (0.5 C, 25 °C) 143 (0.5 C, 80 °C)	2
ILE@MOF	151 (0.1 C, 60 cycles, 60 °C)	-	3
P@CMOF	about 91.5 (1 C, 60 cycles, 60 °C)	126 (0.5 C, 60 °C) 106 (1 C, 60 °C) 67.4 (5 C, 60 °C)	4
MOF-688	120 (~0.2 C, 200 cycles)	-	5
PL/UiOLiTFSI	147.4 (0.2 C, 100 cycles) 132 (1 C, 500 cycles)	145 (0.2 C, RT) 136 (1 C, RT) 109 (3 C, RT)	6
Li-IL@UIO-66-$2CO_2H$	148.5 (0.1 C, 100 cycles) 115 (2 C, 500 cycles)	150 (0.1 C, RT) 145 (0.5 C, RT) 135 (1 C, RT)	7
SE-PMOF	145 (0.2 C, 100 cycles) 127 (1 C, 500 cycles)	153 (0.2 C, RT) 150 (0.5 C, RT) 144 (1 C, RT)	8
UiO-66-2COOH/PVDF-HFP	-	131 (0.2 C, RT) 112 (0.5 C, RT) 90 (2 C, RT)	7
UiO-66-2OH/PVDF-HFP	-	137 (0.2 C, RT) 132 (0.5 C, RT) 70 (2 C, RT)	8
Zr-MA-Li^+	120 (1 C, 500 cycles) 90 (2 C, 700 cycles)	153 (0.2 C) 135 (0.5 C, RT) 115 (2 C, RT)	This work

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