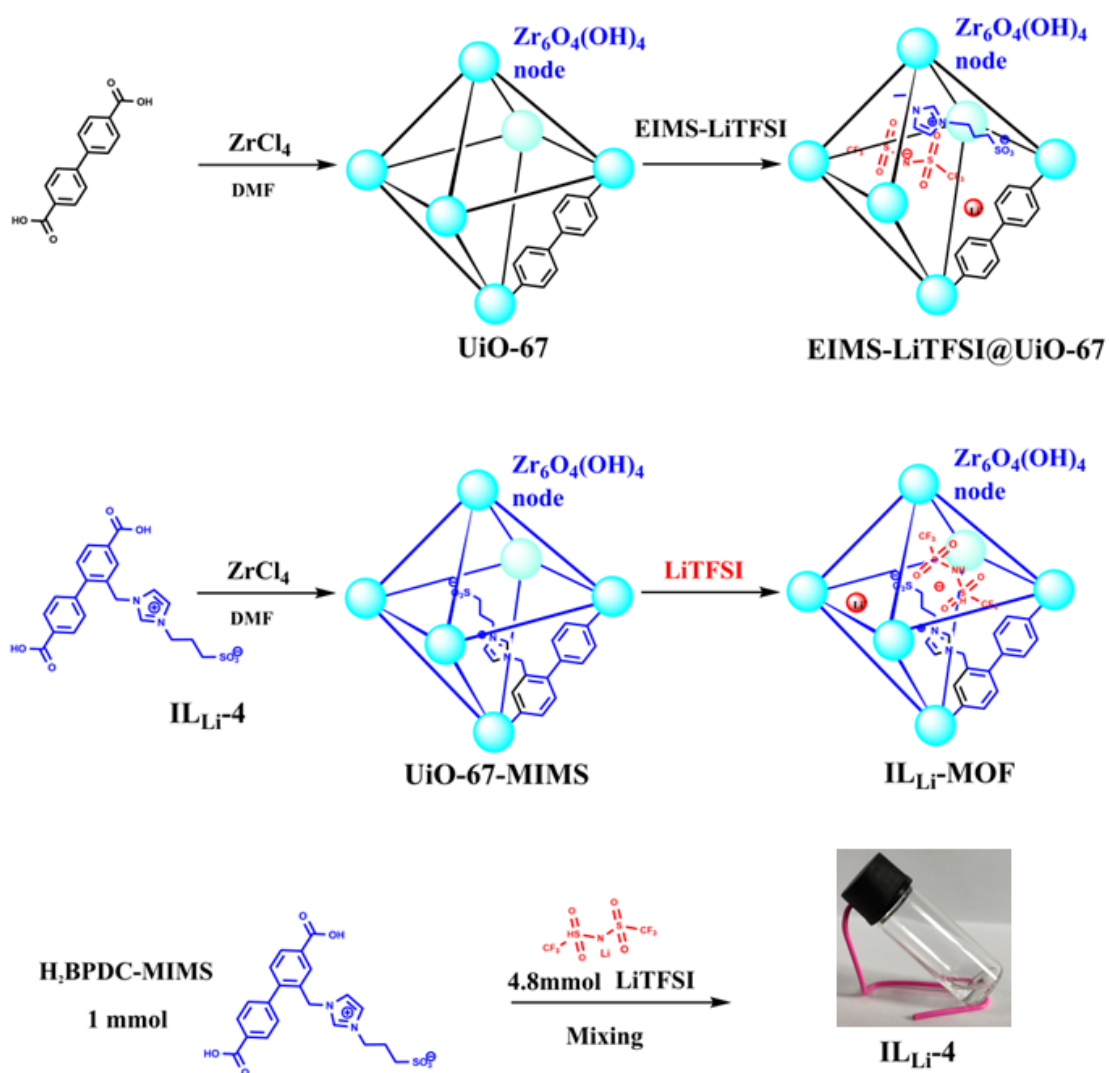


Ionic-liquid Functionalized Metal-organic Framework and Its High Performance Solid Electrolyte for Lithium-ion Conduction



Scheme S1. Synthesis diagram for UiO-67, EIMS-LiTFSI@UiO-67, UiO-67-MIMS, IL_{Li}-MOF and IL_{Li}-4.

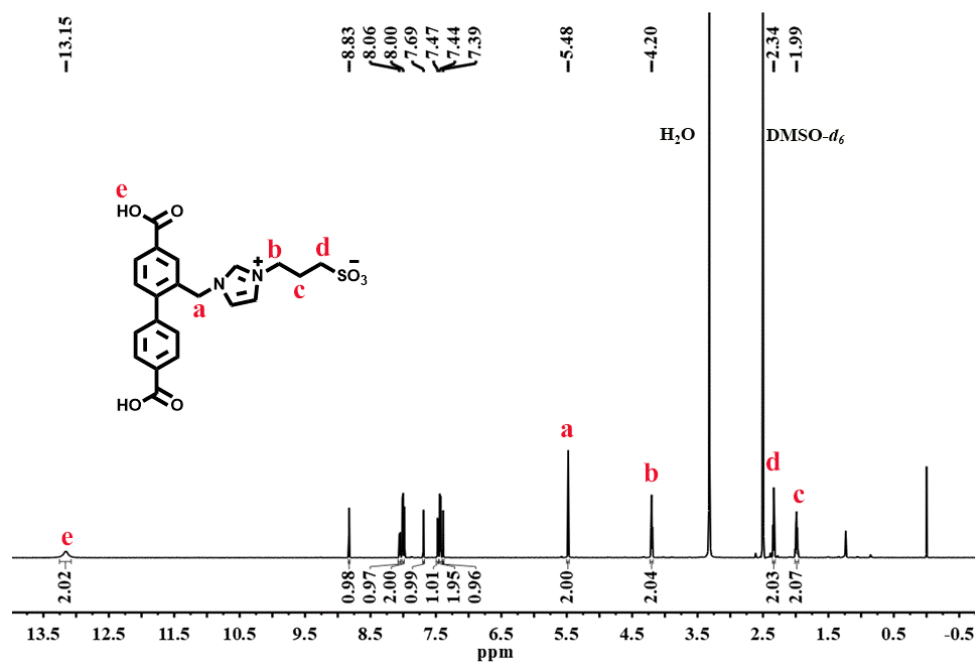


Figure S1. ¹H-NMR spectrum of H₂BPDC-MIMS in DMSO-*d*₆.

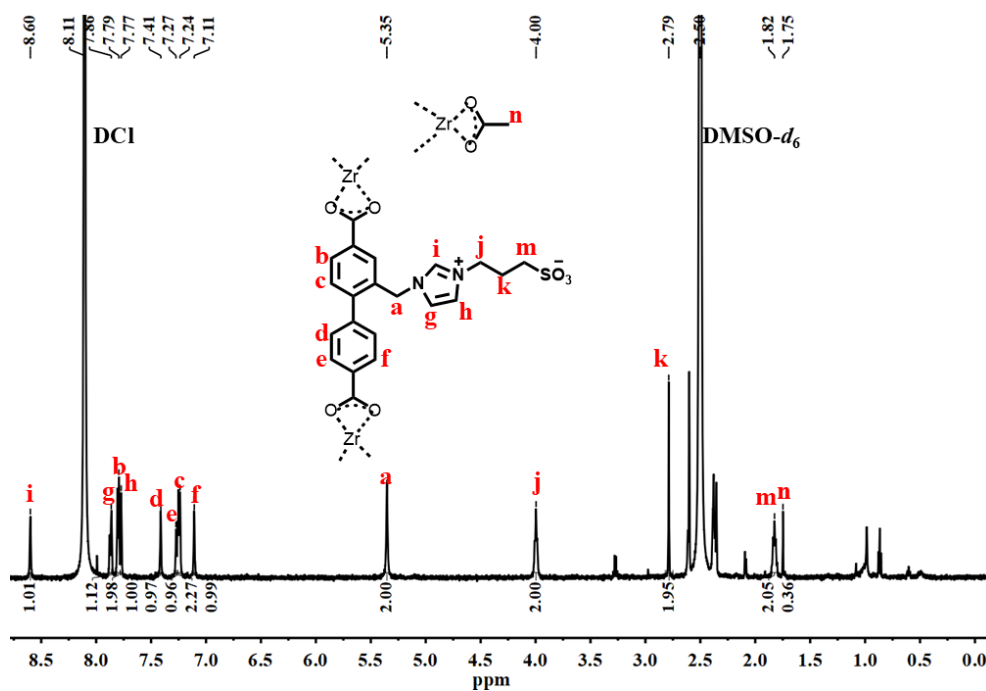


Figure S2. ¹H NMR spectrum of UiO-67-MIMS in DMSO-*d*₆ and DCl.

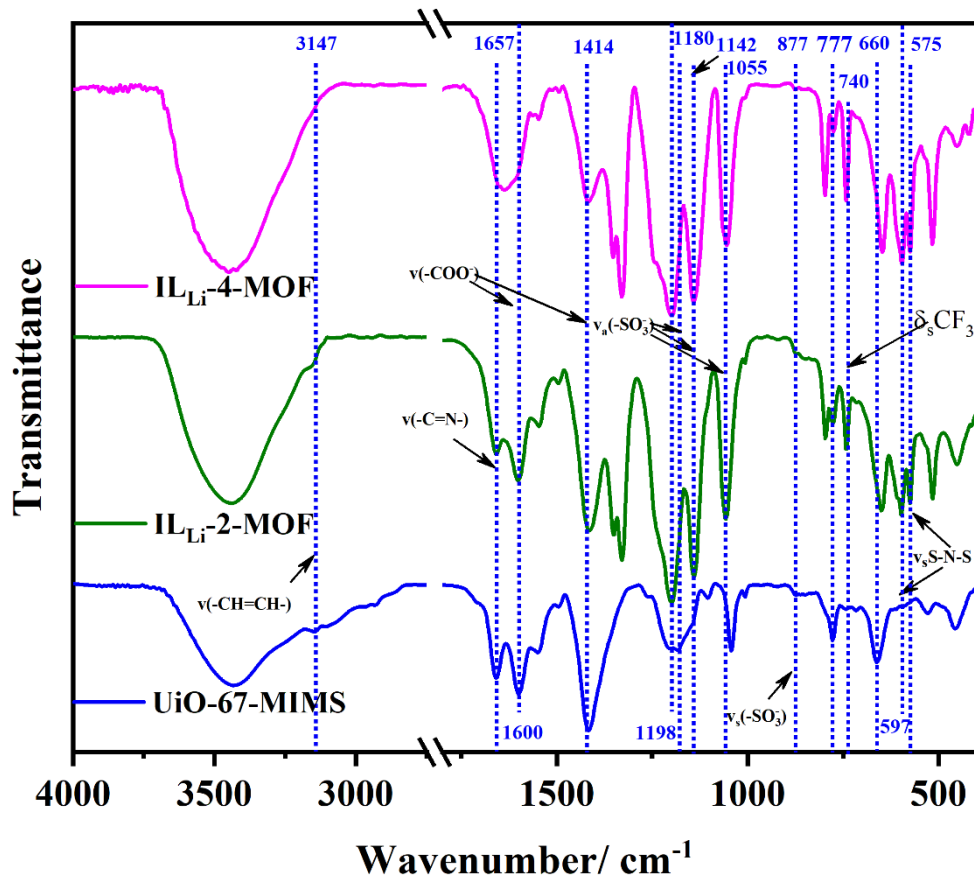


Figure S3. FT-IR curves of $\text{IL}_{\text{Li}}\text{-4-MOF}$, $\text{IL}_{\text{Li}}\text{-2-MOF}$ and UiO-67-MIMS .

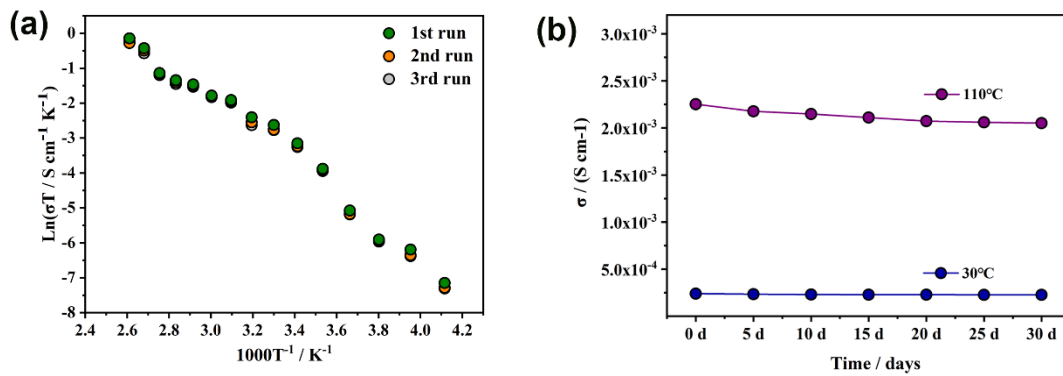


Figure S4. Recycle test (from -30 to 110°C) and long-time stability of $\text{IL}_{\text{Li}}\text{-4-MOF}$ test.

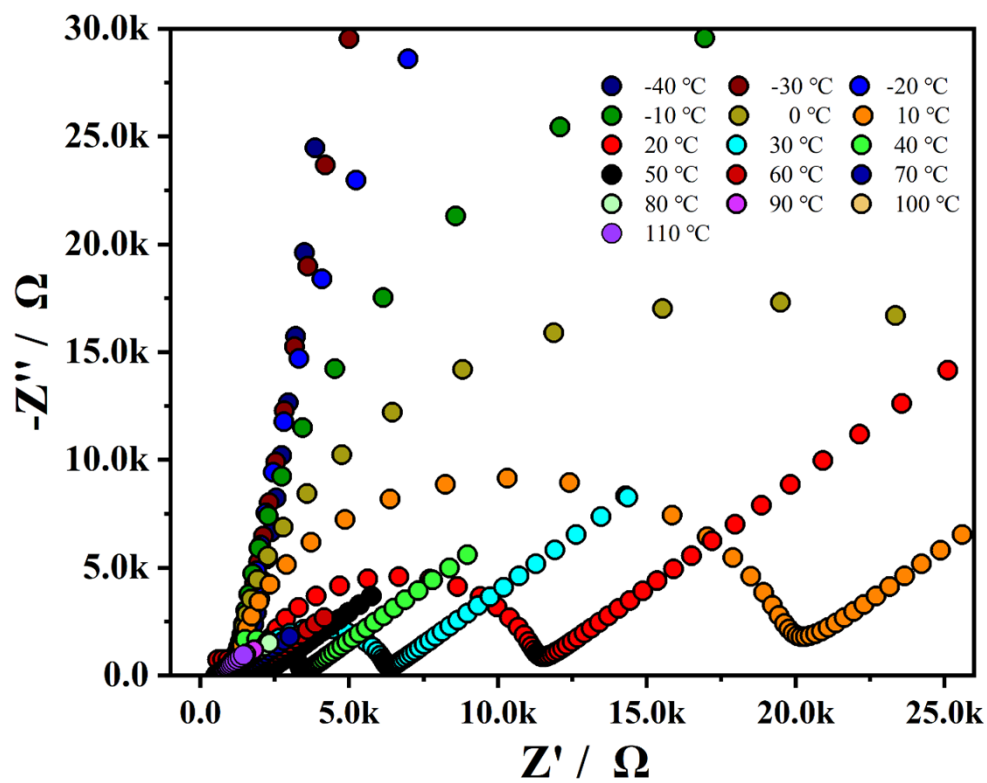


Figure S5. Nyquist plots of IL_{Li-2} -MOF from $-40\text{ }^{\circ}\text{C}$ to $110\text{ }^{\circ}\text{C}$.

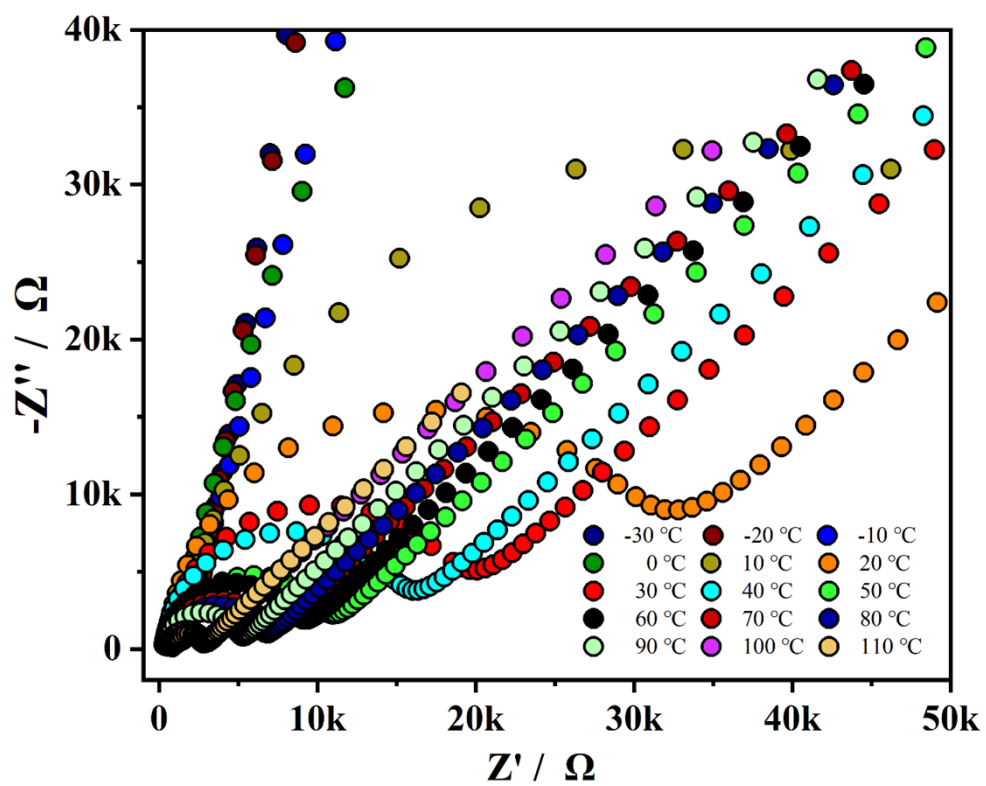


Figure S6. Nyquist plots of IL_{Li-4} -MOF from $-30\text{ }^{\circ}\text{C}$ to $110\text{ }^{\circ}\text{C}$.

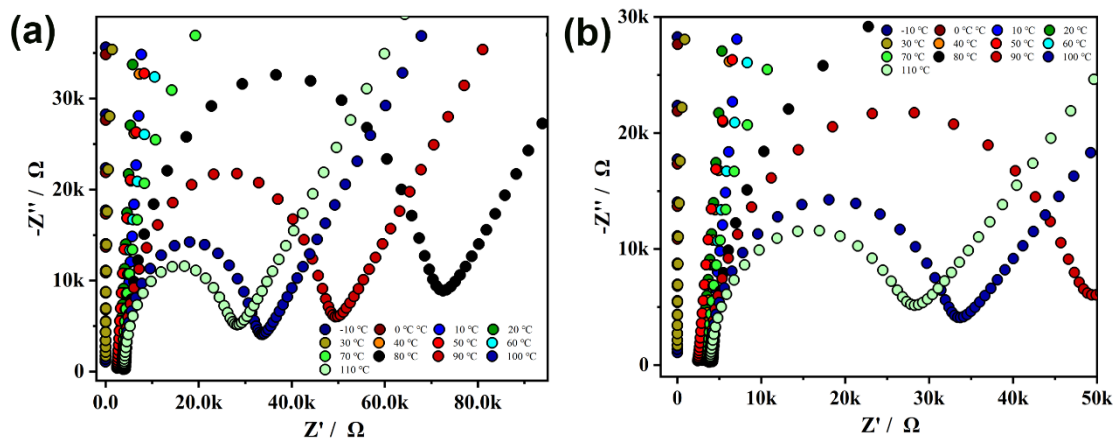


Figure S7. Nyquist plots of EIMS-LiTFSI@UiO-67 from -10 °C to 110 °C.

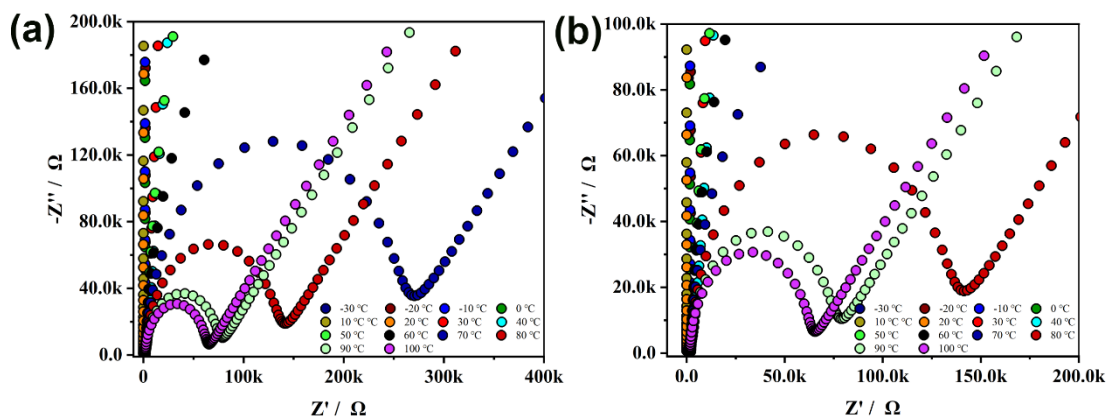


Figure S8. Nyquist plots of IL_{Li}-4 from -30 °C to 110 °C.

Table S1. Comparison of components in UiO-67-MIMS, EIMS-LiTFSI, **IL_{Li}-2-MOF** and **IL_{Li}-4-MOF**

Sample	EIMS (mg)	UiO-67-MIMS (mg)	LiTFSI (mg)	V_{MOF} : V_{LiTFSI}	Stoichiometry Formulaa (L = BPDC-MIMS) ^a	Formula weight
UiO-67-MIMS	/	/	/	/	Zr ₆ O ₄ (OH) ₄ (L) _{5.3} (O ₂ CCH ₃) _{1.4}	~3106.9
EIMS-LiTFSI	218	/	1378	/	EIMS (LiTFSI) _{4.8}	~1366.5
IL_{Li}-2-MOF	/	50	56.8	1:1	Zr ₆ O ₄ (OH) ₄ (L) _{5.3} (O ₂ CCH ₃) _{1.4} (LiTFSI) _{12.5}	~6752.8
IL_{Li}-4-MOF	/	50	113.6	1:2	Zr ₆ O ₄ (OH) ₄ (L) _{5.3} (O ₂ CCH ₃) _{1.4} (LiTFSI) ₂₅	~10398.7

V_{MOF} represents the pore volume of MOFs, V_{LiTFSI} represents the calculated total van der Waals' volume of lithium salt ¹, ^aStoichiometry formula is based on UiO-67-MIMS which was determined through ¹H NMR (Supplementary Figure. 2)

Table S2. BET, pore volume, pore size of UiO-67, UiO-67-MIMS, **IL_{Li}-2-MOF** and **IL_{Li}-4-MOF**.

Sample	BET (m ² /g)	Pore volume cm ³ /g	Pore size (Å)
UiO-67	2222	1.067	~12
UiO-67-MIMS	320.22	0.423	~6 / ~12
IL_{Li}-2-MOF	8.83	0.016	ignorable
IL_{Li}-4-MOF	5.32	0.011	ignorable

Table S3. Conductivities of **IL_{Li}-2-MOF**, **IL_{Li}-4-MOF**, **IL_{Li}-4** and **EIMS-LiTFSI@UiO-67** at selected temperature.

Sample Ionic conduction (S cm ⁻¹) Temperature (°C)	IL_{Li}-2-MOF	IL_{Li}-4-MOF	IL_{Li}-4	EIMS- LiTFSI@UiO- 67
	110	1.35×10^{-2}	2.25×10^{-3}	1.31×10^{-4}
50	3.37×10^{-3}	4.57×10^{-4}	4.92×10^{-6}	3.80×10^{-5}
30	1.62×10^{-3}	2.39×10^{-4}	6.08×10^{-7}	9.70×10^{-6}
-30	1.25×10^{-5}	3.23×10^{-6}	/	/

Table S4. Comparison of ionic conductivities of **IL_{Li}-2-MOF** and **IL_{Li}-4-MOF** with representative conductors based on MOF matrix blended with IL or salt of Li⁺.

Composites (IL/Salt@MOF)	T/°C	σ / S cm ⁻¹	Reference
UN-LiM-DEME	25	3.43×10^{-4}	2
UN-LiM-EMIM	25	4.75×10^{-4}	2
Li-MOF@NWF/PEO	30	1.0×10^{-4}	3
PSZ20	25	2.97×10^{-5}	4
COF-SS-Li	40	1.28×10^{-4}	5
CSIL	30	2.40×10^{-3}	6
PI-PEO-TMP-2	30	4.70×10^{-5}	7
UiO-66-LiSS-50 wt % EC/PC	25	7.80×10^{-4}	8
MIT-20-LiCl	25	1.30×10^{-5}	9
MIT-20-LiBr	25	4.40×10^{-5}	9

MIT-20-LiBF ₄	25	4.80 × 10 ⁻⁴	9
LC-PP	30	5.00 × 10 ⁻⁴	10
LP-PP	30	7.00 × 10 ⁻⁴	10
PEO-ZIF-8	25	4.70 × 10 ⁻⁴	11
Cu ₂ (BPY) ₂ (NDIDS)	28	2.30 × 10 ⁻⁴	12
IL_{Li}-4	30	6.08 × 10 ⁻⁷	
IL_{Li}-2-MOF	30	1.62 × 10 ⁻³	this work
IL_{Li}-4-MOF	30	2.39 × 10 ⁻⁴	this work

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