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Energy Materials

Supplementary Material: Ionic liquids and their derivatives for lithium batteries: role, design strategy, and perspectives

Supplementary Table 1. Comparison of the use of ILs in liquid electrolytes for LMBs.

Electrolyte a)	Cathode b)	Capacity (mAh/g)	C-Rate	No. of cycles	Capacity retention (%)	Ref.
IL + Li-salt system						
[EMI][FSI] + 5M Li[FSI] + 0.16M Na[TFSI]	LCO	~135	0.7C c)	1200	81	[1]
	NMC811	181	0.5C	200	94	
0.8[$Pyr_{1,4}$][FSI]-0.2Li[TFSI]	LRNM	~155	1C c)	2000	56.0	[2]
	LTO	163	0.5C d)	2000	99.6	
	NM88	~200	0.3C c)	1000	88	[3]
0.8[$P_{4,4,4,4}$][IM_{14}]-0.2Li[TFSI]	LRNM	250	0.1C c)	100	84.4	[4]
0.8[$N_{2,2,1,2O1}$][FTFSI]-0.2Li[FTFSI]	LRNM	153	0.5C	500	65.5	[5]
1.2M Li[FSI] in [$Pyr_{1,3}$][FSI]	NMC811	189	0.2C c)	150	95	[6]
4.2M Li[FSI] in [$Pyr_{1,3}$][FSI]	NMC811	~180	1C c)	1000	~77	[7]
0.8[EMI][FSI]-0.2Li[TFSI]	LRNM	~210	0.1C	50	~70	[8]
OLEs + ILs						
3M Li[TFSI] in 25% [$Pyr_{1,3}$][TFSI] +	LNMO	~110	1C	300	90	[9]

75% EC/DEC (1:1 v/v)						
1M Li[TFSI] in DOL/DME (1:1 v/v) + 25 vol% [$Pyr_{1,2O1}$][TFSI]	NMC111	~125	0.3C	200	64.8	[10]
		~125	1C	200	57.6	
1M Li[FSI] in [$Pyr_{1,2O2}$][TFSI]/DOL (4:1 v/v)	LFP	160	0.2C	200	93	[11]
1M Li[FSI] in DOL/DME (1:1 v/v) + 20 wt% 0.9[BzMI][TFSI]-0.1[EMI][TFSI]	LFP	~125	1C	500	76	[12]
1M [$Pyr_{6,6}$][FSI] + 1M Li[TFSI] in DOL/DME (1:1 v/v)	LFP	~160	1C [c)]	800	76.4	[13]
	NMC622	~160	1C [c)]	600	80	
	NMC622	~150	0.5C [c)]	250	78.2	
3.2mol kg^{-1} Li[FSI] in [$Pyr_{1,3}$][FSI]/DME (4:1 wt/wt)	NMC811	~200	0.5C [c)]	300	80	[14]
	NMC622	~175	0.14C [c)]	200	97	
1M $LiPF_6$ in EC/DMC/DEC (1:1:1 v/v/v) + 0.3M [$Pip_{1,2O1}$][BOB]	LNMO	120	1C	100	99.2	[15]
1M $LiPF_6$ in EC/DMC/DEC (1:1:1 v/v/v) + 0.3M [$Pip_{1,2O1}$][DFOB]	LNMO	120	1C	100	98.1	
0.3M Li[TFSI] in [EC/DMC (1:1 wt/wt) + [$Pyr_{1,3}$][TFSI] + FEC (45:45:10)]	LRNMC	153	1C [c)]	1200	~80	[16]
1M $LiPF_6$ in EC:DMC (1:1 v/v)	LNMO	110	0.5C	200	~90	[17]

+ 30 or 50 wt% [$Pyr_{1,4}$]PF_6						
Li[FSI]/[$Pyr_{1,4}$][FSI]/BTFE (3:4:4 in mol)	LFP	150	1C	400	94.6	[18]
	NMC532	~135	1C c)	150	93.9	
0.1M Li[DFOB] in [$Pyr_{1,2O1}$][TFSI]/TTE (1:2 v/v)	LFP	~150	0.5C	500	86	[19]
	LCO	~145	0.2C	180	96	
	NMC622	~200	0.2C	100	96	
Li[FSI]/[EMI][FSI]/BTFE (1:2:2 in mol)	NMC811	185	1C c)	200	96	[20]
Li[FSI] /[EMI][FSI]/dFBn (1:2:2 in mol)	NMC811	192	1C c)	500	93	[21]
Li[FSI]/[Pip_{13}][FSI]/TTE (1:2:4 in mol)	LFP	~110	5C	1000	87	[22]
Li[TFSI] /[$Pyr_{1,3}$][FSI]/TTE (1:2:2 in mol)	LCO	~155	0.5C	400	80	[23]
0.8[Pyr_{H4}]TFSI-0.2Li[TFSI] + 10 wt% VC	LFP	170	0.05C e)	50	~75	[24]
0.8[Pyr_{H4}]FSI-0.2Li[FSI] + 10 wt% VC	NMC622	155	0.1C f)	60	95	

a) EC: ethylene carbonate, DEC: diethyl carbonate, DOL: 1,3-dioxolane, DME: dimethoxyethane, DMC: dimethyl carbonate, FEC: fluoroethylene carbonate, VC: vinylene carbonate, BTFE: bis(2,2,2-trifluoroethyl)ether, TTE: 1,1,2,2-tetrafluoroethyl 2,2,3,3-tetrafluoropropyl ether, dFBn: 1,2-difluorobenzene, b) LCO: $LiCoO_2$, NMC811: $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, LRNM: $Li_{1.2}Ni_{0.2}Mn_{0.6}O_2$, LTO: $Li_4Ti_5O_{12}$, NM88: $LiNi_{0.88}Mn_{0.03}C_{0.09}O_2$, NMC111: $LiNi_{0.33}Mn_{0.33}Co_{0.33}O_2$, LFP: $LiFePO_4$, NMC622: $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$, LNMO: $LiNi_{0.5}Mn_{1.5}O_2$, LRNMC: $Li_{1.2}Mn_{0.56}Ni_{0.16}Co_{0.08}O_2$, c) the battery was first activated by cycling at a lower C-rate, d) the battery was charged/discharged first at 0.1C, 0.2C, 0.5C, 1C, 2C. e) the battery was charged/discharged first at 0.05C, 0.1C. f) the battery was charged/discharged first at 0.05C, 0.1C, 0.2C, 0.5C, 1C.

Supplementary Table 2. Summary of PIL compositions and their performances as the electrolyte for LMBs (σ: conductivity, Q: specific capacity in mAh/g).

Cation		Additive (IL, salt, etc…)	Composition in weight [b)]	σ (mS/cm) [c)]	Cell performances			Ref.
Position	Type [a)]				Cathode [d)]	Q [c)]	C-rate	
Linear								
Side	Py+N	[C$_2$mim][TFSI]/Li[TFSI]	PIL:IL:salt = 100:45:10	0.126	LFP	145	0.1C	[25]
Side	Py+N	[C$_{2O2}$mmim][TFSI]/ Li[TFSI]	PIL:IL:salt = 100:100:30	0.046	LFP	120	0.1C	[26]
Side	Im	[C$_{2O2}$mmim][TFSI]/ Li[TFSI]	PIL:IL:salt = 100:65:20	0.0189	LFP	~160	1C (60 °C)	[27]
Backbone	Pyr	[Pyr$_{1,4}$][TFSI]/ Li[TFSI] (9:1)	ILE uptake = 587%	~2	LFP	150	0.5C	[28]
						162	1C (55 °C)	
						145	5C (120 ° C)	
Backbone	Pyr	1M Li[FSI] in [Pyr$_{1,3}$][FSI]	PIL:additive = 4:6 [e)]	0.8	NMC811	162	0.1 mA cm^{-2}	[29]
					LNMO	132	0.1 mA cm^{-2}	
Backbone	Pyr	[Pyr$_{14}$][TFSI]/ Li[TFSI]	PIL:IL:salt = 28:12:60	0.16	LFP	140	0.2C (40°C)	[30]
Backbone	Pyr	1 M Li[TFSI] in [C$_2$mim][TFSI]	PIL:additive = 2:8	3.4	LFP	169	0.1C	[31]
						127	1C	

Backbone	Pyr	[Li(G4)][TFSI]	PIL:additive = 33:67	~0.3	LFP	150	0.1C	[32]
Backbone	Pyr	$[Pyr_{13}][FSI]/Li[FSI]$	PIL:IL:salt = 1:1:1 f)	0.22 (50 °C)	NMC622	~170	0.1C (40 °C) g)	[33]
						~160	0.1C g)	
Copolymer or Polymer blend								
Side chain	Im	Li[TFSI]	co-PIL:salt = 10:3 e)	0.246	LFP	142	0.2C	[34]
Side chain	Im	$[C_2mim][TFSI]$ /Li[TFSI]	co-PIL:IL:salt = 20:5:8	0.19	LFP	135	0.1C	[35]
Backbone	Pyr	$[Pyr_{1,2O1}][TFSI]$ /Li[TFSI] (9:1) f)	PIL:PTFEMA: additive = x:2:8 h)	0.82	LFP	138	1C	[36]
Side chain	Im	Li[TFSI]	co-PIL:PEO:salt = 22.5:67.5:10	0.011	LFP	163	0.2C (60 °C)	[37]
Cross-linked PIL or its copolymer or polymer blend								
Side chain	Im	Li[TFSI]	xl-PIL:PMIA:salt = 75:x:25 i)	0.197	LFP	134	0.5C	[38]
Side chain	Im	Li[TFSI]	xl-PIL:Li[TFSI] = 33:12	0.14	LFP	140	0.2C	[39]
Side chain	Im	Li[TFSI]	Li/O = 1:10 f)	0.0318	LFP	154	0.1C	[40]
						138	0.5C	
Side chain	Im	$[C_2mim][TFSI]$ /Li[TFSI]	xl-PIL:PVdF-HFP:IL:salt = 2:0.0011:1.3:1 f)	1.06	LFP	154	0.1C	[41]
Side	Im	$[C_2mim][TFSI]$	xl-PIL:IL:salt	0.055	LFP	153	0.1C	[42]

chain		/Li[TFSI]	= 10:2:2				(60 °C)	
Bridge	Im	[C$_2$mim][TFSI] /Li[TFSI]	xl-PIL:IL:salt = 3:1.2:0.5	1.03	LFP	166	0.1C j)	[43]
						94	5C j)	
					NMC622	188	0.1C j)	
Bridge	Im	[C$_2$mim][TFSI] /Li[TFSI]	xl-PIL:PVdF-HFP:IL:salt = 1:1:5:1.2	1.8	LFP	150	0.5C j)	[44]
						116	4C j)	
					NCM622	189	0.1C j)	
Bridge, side	Im	[C$_{2O2}$mmim][TFSI] /Li[TFSI]	xl-PIL:IL:salt = 6:21:4.2	1.41	LFP	148	0.2C	[45]
Bridge	Im	Li[TFSI] /succinonitrile	crPIL:salt:SN 4:4:5 k)	1.07	LFP	153	0.2C	[46]
						133	1C	
Bridge	Im	Li[TFSI]	xl-PIL:Li[TFSI] 3:0.5	0.27	LFP	146	0.5C	[47]
						113	2.5C	

a) Py+N: dication of pyridinium and ammonium, Im: imidazolium, Pyr: pyrrolidinium, b) values are calculated according to the information in the literatures and may be rounded, c) values at ambient temperature (25±5 °C) or at room temperature unless specific values are reported, d) LFP: $LiFePO_4$, NMC811: $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, LNMO: $LiMn_{1.5}Ni_{0.5}O_4$, NMC622: $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$, e) integrated with glass fiber separator, f) molar ratio, PVdF-HFP: poly(vinylidene fluoride-co-hexafluoropropylene), g) the battery was first activated by cycling at a lower C-rate, h) PTFEMA: cross-linked poly(2,2,2-trifluoroethyl methacrylate), ratio between PIL: PTFEMA is not specified, i) PMIA: poly-m-phenylene isophthalamide, ratio between xl-PIL and PMIA is not specified, j) temperature was not specified, k) SN: succinonitrile.

Supplementary Table 3. Summary of the use of DESs in the electrolyte for LMBs (Q: capacity).

Electrolyte [a)]	**Cathode** [b)]	**C-Rate**	**Q (mAh/g)**	**No. of cycles**	**Capacity retention (%)**	**Ref.**
$LiPF_6$:TFA (1:4 in mol) + 10 wt% EC + 5 wt% FEC	LFP	0.1C	~150	70	~67	[48]
Li[TFSI]:Li[DFOB]:SN (0.8:0.2:10 in mol)	LCO	0.5C	~170	200	94	[49]
		1C	~165	200	70	
		2C	~155	500	72	
Li[TFSI]:Li[DFOB]:SN (0.17:0.03:0.8 in mol)	LCO	1C	~180	500	70	[50]
1M $LiPF_6$ in EC/EMC/DMC (1:1:1 v/v/v) + 1 wt% Li[TFSI]:Urea (1:3 in mol)	LFP	1C	149	1000	92.1	[51]
$LiClO_4$:MSM:H_2O (1:1.8:1 in mol)	LTO	20C	~130	1000	85.2	[52]
	LMO	4.5C	~45	1000	91	
Li[TFSI]:NMA (1:4 in mol) + 2 wt% $LiNO_3$	NMC811	0.5C	~175	600	74	[53]
	NMC622	0.5C	~160	600	84	
85 vol% Li[TFSI]:NMA (1:4 in mol) + 15 vol% DMPA/EGDMA/AcMo (1:2:18 in mol)	LFP	0.5C	110	250	95	[54]
Li[TFSI]:NMA (1:4 in mol) + 10 wt% FEC + 3 wt% UPyMA + 1.5 wt% PETEA + 0.1 wt% AIBN	LMO	0.1C	117	200	86.1	[55]

70 wt% Li[TFSI]:NMA (1:4 in mol) in PVDF-HFP	LFP	0.1C	164	100	81.4	[56]
Li[TFSI]:PDOL (1:3 in mol)	LFP	0.2C	132	200	90	[57]
Li[TFSI]:NMA (1:4 in mol) + LiFSI + PEO + 10 wt% UiO66-NH_2 [c)]	LFP	0.2C	159	350	93.3	[58]
Li[TFSI]:NMA:AIBN:SSH:PETEA (10:40:0.01:3:3 in mol)	LFP	0.1C	156	100	86.1	[59]
Li[TFSI]:Li[DFOB]:SN:Cyanoethyl cellulose (0.8:0.02:1:0.364 wt/wt/wt/wt)	LCO	1C	165	200	85	[60]
Li[TFSI]:NML (1:6 in mol) + UpyMA/PEGDA (1:100 wt/wt) + 0.2 wt% AIBN [d)]	LCO	0.5C	189	1000	80	[61]

[a)] TFA: 2,2,2-trifluoroacetamide, EC: Ethylene carbonate, FEC: Fluoroethylene carbonate, SN: succinonitrile, EMC: Ethyl methyl carbonate, DMC: Dimethyl carbonate, MSM: methylsulfonylmethane, NMA: *N*-methylacetamide, DMPA: dimethoxy-2-phenylacetophenone, EGDMA: ethylene glycol dimethacrylate, AcMo: 4-acryloylmorpholine, UpyMA: 2-(3-(6-methyl-4-oxo-1,4-dihydropyrimidin-2-yl)ureido) ethyl methacrylate, PETEA: pentaerythritol tetraacrylate, PDOL: poly (1,3-dioxolane) , AIBN: 2, 2-azodiisobutyronitrile, NML: *N*-methylurea, PEGDA: polyethylene glycol diacrylate. [b)] LFP: $LiFePO_4$, LCO: $LiCoO_2$, LTO: $Li_4Ti_5O_{12}$, LMO: $LiMn_2O_4$, NMC811: $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$, NMC622: $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$. [c)] The molar ratio EO:Li^+ was kept at 18:1. [d)] The molar ratio EO:Li^+ was kept at 20:1.

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