Supplementary Information (SI) for Nanoscale.

Supporting Information

Challenges, opportunities, and roadmap for ionic liquids-based electrolytes in advancing energy storage devices

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Table S1 Properties of some bulk Ionic liquid-based electrolytes

Ionic Liquids (ILs) electrolyte	Viscosity (mPa.s)	Conductivity ($mScm^{-1}$)	Electrochemical stability window (V)	Ref.
1-ethyl-3-methylimidazolium bis(fluorosulfonyl)imide (EMIM)(FSI)	19	17.74	3.5	[1]
Ethyl-3-methylimidazolium bis-trifluoromethylsulfonylimide (EMIM)(TFSI)	34	10.8	4.0	[2]
1-ethyl-3-methyl imidazolium tetrafluoroborate (EMIM)(BF_4)	32	16.01	4.0	[3]
1-ethyl-3-methylimidazolium trifluoromethanesulfonate (EMIM)(Otf)	45.7	8.5	4.1	[4]
1-ethyl-3-methylimidazolium chloride (EMIM)(Cl)	-	1.85	2.8	[5]
1-ethyl-3-methylimidazolium hexafluorophosphate (EMIM)(PF_6)	-	1.93	3.2	[6]
1-butyl-3-methylimidazolium hexafluorophosphate (BMIM)(PF_6)	-	2.1	3.0	[7]
N-propyl-*N*-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide (PYR_{13}TFSI)	58.7	4.92	5.0	[8]
1-butyl-1-methylpyrrolidinium bis(trifluoromethylsulfonyl)imide (PYR_{14}TFSI)	85	2.2	>4.8	[9]
N-methyl-N-propylpiperidinium bis(trifluoromethylsulfonyl)imide (Pip_{13}TFSI)	-	10	4.35	[10]

Table S2 Ionic conductivity, cation/ion transference number, electrochemical voltage stability window, and thermal stability of ionic liquid-based gel polymer electrolytes.

ILs based electrolytes	Ionic conductivity (S/cm)	Transference number (ion/cation)	Electrochemical stability window vs Li/Li^+(V)	Thermal Stability (°C)	Ref.
PEO based polymer electrolytes					
PEO+ LiDFOB+[EMIm][TFSI]	~1.85×10^{-4}	-	2.5 to 4.0	-	[11]
PEO + LiTFSI + [BMP][TFSI]	2.5×10^{-5} at 20 °C	0.41	5.2	340	[12]
PEO+LiTFSI+Pyr_{13}[TFSI]	~ 10^{-4} to 10^{-3} at 30° C	< 0.31	-	-	[13]
PEO+LiTFSI+[PP_{13}][TFSI]	2.06×10^{-4} at RT	0.339	~4.5 to 4.7	220	[14]
PEO+ LiDFOB+[EMIM][TFSI]	2.98×10^{-4} at 20 °C	0.39	4.6	310	[15]
PEO+LiTFSI+Pyr13[TFSI]	2.8×10^{-4} at 20 °C	-	~6	-	[16]
PEO+LiTFSI+[EMIM][TFSI]	2.67×10^{-4}	0.108		-	[17]
PEO+LiTFSI+[PP_{13}][TFSI]	8.93×10^{-5} at 40 °C	0.0934	~ 5.2 for both		
PEO+$LiClO_4$+[BMIM][PF_6]	~10^{-4} at 40°C	-	-	-	[18]
PEO+LiTFSI+[BMIM][TFSI]	1.5×10^{-4} at 30 °C	0.27	4.0	350	[19]
PEO+LiTFSI+[$PYRA_{1201}$][TFSI]	3.4×10^{-3}	0.23	~4.2	-	[20]
PEO+LiTFSI+[$PYRA_{12(02)201}$][TFSI]	6.2×10^{-3}	0.17	-	-	
PVdF-HFP based polymer electrolytes					
PVdF-HFP+ LiTFSI+[BMIM][TFSI]	2×10^{-3} at RT	0.22	-	300	[21]
PVdF-HFP +LiTFSI+[EMIM][TFSI]	3.8×10^{-4} at 25 °C	0.4	4.7	200	[22]
PVdF-HFP +LiTFSI+[Py_{24}][TFSI]	2.7×10^{-4} at RT	-	5.75	200	[23]
PVdF-HFP +LiTFSI+[B_4MePy] [TFSI]	2.8×10^{-4} at RT	0.7	5.5 at 20 °C	-	[24]
PVdF-HFP +LiTFSI+[PMIM][TFSI]	9.5×10^{-3} at 30 °C	0.22	-	200	[25]
PVdF-HFP +$LiClO_4$+[EMIM][DCA]	6.0×10^{-4} at RT	-	-	300	[26]

Table S3 IL-based liquid electrolytes in supercapacitor.

ILs electrolyte	Electrode	C_{sp} (Fg^{-1})	ESW (V)	Energy Density ($WhKg^{-1}$)	Ref.
[EMI][TFSI]	3D-organically modified carbon	146-178	3.5	-	[27]
[EMI][TFSI]	N-doped GNs	104	3.6	-	[28]
[EMI][TFSI]	Carbon nanotubes (CNTs)	135	3	-	[29]
[EMI][TFSI]	Mesoporous graphene	-	4	135.6	[30]
[EMI][TFSI]	Activated carbon	5.57	-	-	[31]
[EMI][TFSI]	Mesoporous carbon	26.86	-	-	
[EMI][TFSI]	MWCNT	6.35	-	-	
[EMI][TFSI]	RGO	8.18	-	-	
[EMI][TFSI]	Graphene	332	-	156	[32]
[EMI][TFSI]	CNT	201	-	171	[33]
[BMI][BF_4]	GO-CMK-5	144.4	3.5	-	[34]
[BMI][BF_4]	SWCNT & RGO	222	3.5	-	[35]
[BMI][BF_4]	AC	111	3.5	-	[36]
[BMI][BF_4]	KOH treated carbon-xerogel	-	1.1	-	[37]
[EMI][BF_4]	Glucose derived AC	158	3	-	[38]
[EMI][BF_4]	CNT spaced Graphene aerogels	245.5	1	-	[39]
[EMI][BF_4]	Soybean root derived 3D-hierachical porous carbon	276	1	-	[40]
[EMI][BF_4]	N/O-doped rod-like microporous carbons	-	-	89.5	[41]
[EMI][BF_4]	Mesoporous graphene	250	-	85.6	[42]
[BMI][PF_6]	Reduced graphene oxide (RGO)	158	1	-	[43]
[BMI][PF_6]/DMF	MnO_2	523.3	3	-	[44]
[BMI][PF_6]	RGO	74	4	-	[45]
[BMI] [BF_4]		45	3	-	
[EMI][BF_4]-[EMI]Br	Activated carbon fibre cloths	59	1.5	-	[46]
[EMI][TFSI]-[EMI]I	porous carbon	200.6	3.5	175.6	[47]
[PYR][TFSI]+[DIPEA][TFSI]	AC	120	2	-	[48]

$[PYR_{13}][TFSI]$	Carbon Films	102	2.5	-	[49]
$[Et_4N][BF_4]$	K10/MWCNT/MnO_2	100	3.5	-	[50]
$[PYR_{1,4}][DCA]$	Graphene nanosheets (GNs)	330	3.3	-	[51]
[BMP][DCA]	Graphene nanosheets (GNs)	235	3.3	-	[52]
$[Pyr_{1,4}][C(CN)_3]$	Activated carbon	27.3	-	4.5	[53]
$[Pip_{1,4}][C(CN)_3]$	Activated carbon	17.7	-	0.9	
$[Pyr_{1,4}][B(CN)_4]$	Activated carbon	20	-	6	[54]
$[Pip_{1,4}][B(CN)_4]$	Activated carbon	14.8	-	1	
$[N_{2224}][N(CN)_2]$	Graphene	42	-	55	[55]
$[N_{222},Propargyl][N(CN)_2]$	Graphene	55	-	49	

Table S4 IL-based gel/solid electrolytes in supercapacitor.

IL based GPE electrolytes	Electrode	Csp (Fg^{-1})	ESW (V)	Energy Density and Power Density	Ref.
EMI TFSI/TMOS/DMDMS	AC	177 $mFcm^{-2}$	0–3		[56]
PVDF-HFP/EMI TFSI	Carbon electrode	118-115	0–2.5 V	21.9 $Whkg^{-1}$, 6.25 $kWkg^{-1}$	[57]
PAM/1-vinyl-3-methylimidazolium bis(trifluoromethylsulfonyl) imide	PEDOT/ carbon cloth	157.8		14.22 $Whkg^{-1}$	[58]
PHEMA-co-PEGDMA/EMI BF_4	AC	193.33	0–3 V	49.55 $Whkg^{-1}$ and 1.23 $kWkg^{-1}$	[59]
$EMIMBF_4$/PVDF-HFP	Graphite	17.4	3.5 V	56 $mWhcm^{-3}$	[60]
PEGDA/EMIMTFSI	MWCNT	5.3	0–2 V	0.17 $mWhcm^{-3}$	[61]
Fumed SiO_2/EMI TFSI	Porous carbon		2 V	26 Wcm^{-3}	[62]
PVDF-HFP-[BMI][TFSI]-NaI	Activated carbon	334	1.5	26.1 Wh kg^{-1}	[63]
PVA/PVP-[EMI][HSO_4]-HQ	Activated carbon	485	1.2	24.3Wh kg^{-1}	[64]

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