SUPPORTING INFORMATION

Sodium diffusion in ionic liquid-based electrolytes for Na-ion batteries: the effect of polarizable force fields

Arianna Massaro,[1,2] Jocasta Avila,[2] Kateryna Goloviznina,[2] Ivan Rivalta,[2,3] Claudio Gerbaldi,[4] Michele Pavone,[1] Margarida F. Costa Gomes,[2] and Agilio A. H. Pádua[2,a)]

[1)] *Department of Chemical Sciences, Università di Napoli "Federico II", via Cintia 26, 80126 Naples, Italy*

[2)] *Univ Lyon, Ens de Lyon, CNRS UMR 5182, Université Claude Bernard Lyon 1, Laboratoire de Chimie, F69342, Lyon, France*

[3)] *Dipartimento di Chimica Industriale "Toso Montanari", Università degli Studi di Bologna, Viale del Risorgimento 4, I-40136 Bologna, Italy*

[4)] *GAME Lab, Department of Applied Science and Technology (DISAT), Politecnico di Torino, 10129 Torino, Italy*

[a)] Electronic mail: agilio.padua@ens-lyon.fr

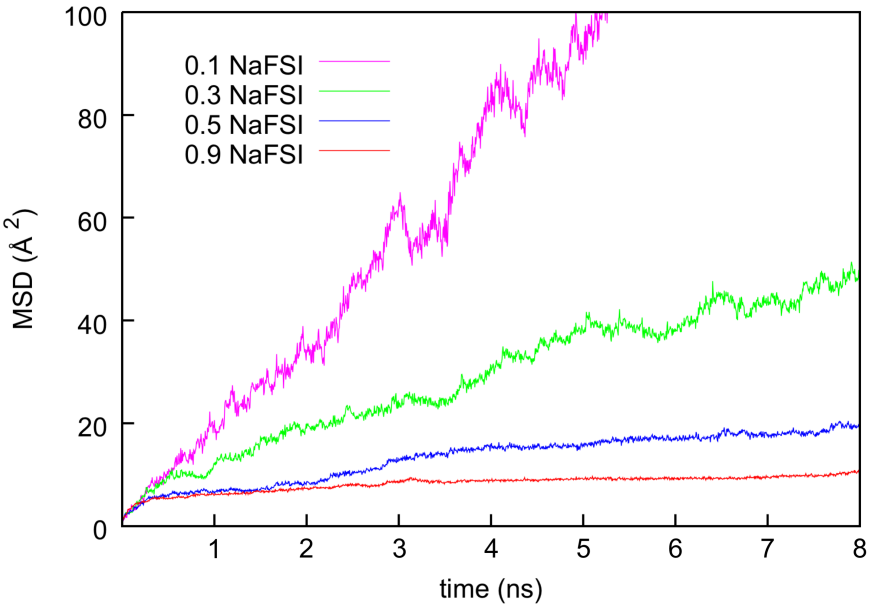

Figure S1. Plot of mean-squared displacement (MSD) of Na^+ as function of time for each salt content.

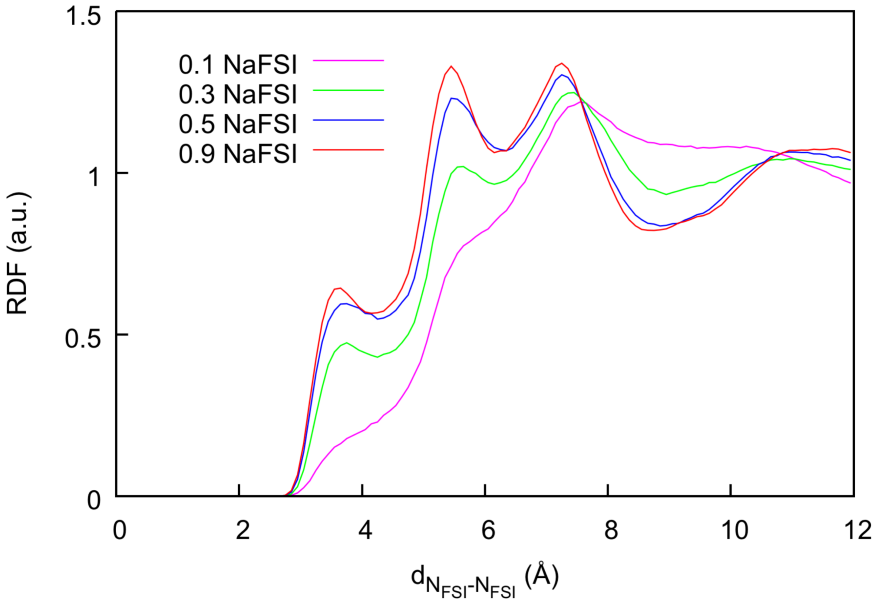

Figure S2. Radial distribution function (RDF) of N_{FSI}–N_{FSI} distances.

Table S1. $\mathcal{A}$ values and viscosity with corresponding deviations, η, computed from non-equilibrium MD trajectories for the 0.1 Na[FSI] system with Drude model.

$\mathcal{A}(10^{-5}$ Å fs$^{-2})$	η (mPa s)
0.01	21.1 ± 1.6
0.02	23.6 ± 1.0
0.04	17.4 ± 0.3

Table S2. Density, ρ, self-diffusion coefficients, D_+, D_- and D_{Na^+}, respectively for $C_3C_1pyr^+$, FSI^- and Na^+ calculated with both scaled-charges (ScaleQ) and polarizable model (Drude) at 353 K. Experimental densities at 348 K are marked with *. Viscosity, η, and corresponding deviations computed with Drude model via non-equilibrium MD simulations at 353 K are also reported.

	ρ (g cm^{-3})	D_+ (m^2 s^{-1})	D_- (m^2 s^{-1})	D_{Na^+} (m^2 s^{-1})	η (mPa s)
[C_3C_1pyr][FSI]					
Exp [1]	1.295	14.0×10^{-11}	17.0×10^{-11}	–	–
ScaleQ	1.254	2.71×10^{-11}	3.20×10^{-11}	–	–
Drude	1.275	7.08×10^{-11}	7.84×10^{-11}	–	14.3 ± 0.4
Exp (this work)	1.296	–	–	–	10.58
0.1 Na[FSI] : 0.9 [C_3C_1pyr][FSI]					
Exp [2]	1.328*	–	–	–	–
ScaleQ	1.309	2.57×10^{-11}	2.92×10^{-11}	2.38×10^{-11}	–
Drude	1.312	4.85×10^{-11}	4.99×10^{-11}	3.27×10^{-11}	23.6 ± 1.0
Exp (this work)	1.329	–	–	–	12.73
0.3 Na[FSI] : 0.7 [C_3C_1pyr][FSI]					
Exp [2]	1.419*	–	–	–	–
ScaleQ	1.391	1.60×10^{-11}	1.58×10^{-11}	9.37×10^{-12}	–
Drude	1.403	2.27×10^{-11}	2.03×10^{-11}	1.09×10^{-11}	74.8 ± 5.8
Exp (this work)	1.407	–	–	–	18.76
0.5 Na[FSI] : 0.5 [C_3C_1pyr][FSI]					
Exp [2]	1.532*	–	–	–	–
ScaleQ	1.497	1.08×10^{-11}	1.01×10^{-11}	7.76×10^{-12}	–
Drude	1.526	9.27×10^{-12}	6.39×10^{-12}	4.57×10^{-12}	330 ± 57
Exp (this work)	1.512	–	–	–	38.73
0.9 Na[FSI] : 0.1 [C_3C_1pyr][FSI]					
ScaleQ	1.855	5.41×10^{-12}	4.50×10^{-12}	3.96×10^{-12}	–
Drude	1.945	3.46×10^{-12}	2.23×10^{-12}	2.45×10^{-12}	583 ± 151

Table S3. Salt and IL masses, m, and molar fractions, x, of Na[FSI]+[C_3C_1pyr][FSI] solutions with corresponding uncertainties.

$m_{\mathrm{Na[FSI]}}$ (g)	$m_{[C_3C_1\mathrm{pyr}][\mathrm{FSI}]}$ (g)	x
0.24625 ± 0.00008	3.34916 ± 0.00007	0.10034 ± 0.00003
0.94658 ± 0.00002	3.35114 ± 0.00008	0.29994 ± 0.00001
2.19761 ± 0.00002	3.39323 ± 0.00003	0.49555 ± 0.00001

Table S4. Parameters, A_0 and A_1, from linear functions used to fit experimental density as a function of temperature, $\rho = A_0 + A_1 T$, and corresponding absolute average deviation (AAD).

Sample	A_0 ($\mathrm{g\,cm^{-3}}$)	A_1 ($\mathrm{g\,cm^{-3}\,K^{-1}}$)	AAD%
pure IL	1.5716	-7.8061×10^{-4}	0.007
0.1 Na[FSI]	1.6123	-8.0364×10^{-4}	0.008
0.3 Na[FSI]	1.7083	-8.5271×10^{-4}	0.008
0.5 Na[FSI]	1.8366	-9.2016×10^{-4}	0.010

Table S5. Parameters, A, B and T_0, from VTFH functions used to fit experimental viscosity as a function of temperature, $\log \eta = A + B/(T - T_0)$, and corresponding absolute average deviation (AAD).

Sample	A	B (K)	T_0 (K)	AAD%
pure IL	−0.6894	370.6	136.8	0.6
0.1 Na[FSI]	−0.6066	352.5	147.6	0.4
0.3 Na[FSI]	−0.5080	319.9	173.5	0.1
0.5 Na[FSI]	−2.9417	1335.2	57.4	4.2

Table S6: Experimental density, ρ, and viscosity, η, of the samples listed in Table S3 measured in the temperature range of 293 K to 353 K. The percent deviations, δ, refer to the fitting functions with coefficients listed in Table S4 and S5.

T (K)	ρ (g cm^{-3})	δ_ρ (%)	η (mPa s)	δ_η (%)
[C_3C_1pyr][FSI]				
293.153	1.342929	−0.01	47.86	0.4
303.154	1.334965	−0.0002	34.76	−0.4
313.154	1.327060	0.007	26.17	−1.2
323.154	1.319211	0.01	19.69	1.3
333.154	1.311455	0.007	15.73	0.4
343.154	1.303733	0.0003	12.86	−0.5
353.154	1.296094	−0.01	10.58	−0.1
0.1 Na[FSI]				
293.149	1.376948	−0.01	65.49	−0.3
303.154	1.368701	0.001	45.39	0.5
313.154	1.360558	0.01	33.18	0.3
323.154	1.352486	0.01	25.16	0.05
333.154	1.344511	0.007	19.70	−0.4
343.155	1.336571	−0.0004	15.81	−0.8
353.155	1.328705	−0.01	12.73	0.7
0.3 Na[FSI]				
293.149	1.458528	−0.02	146.6	0.06
303.154	1.449747	0.002	91.3	−0.1
313.154	1.441109	0.01	60.82	−0.2
323.154	1.432560	0.01	42.58	0.2
333.154	1.424109	0.006	31.32	0.07
343.154	1.415677	−0.001	23.89	−0.06
353.154	1.407318	−0.01	18.76	−0.06
0.5 Na[FSI]				

(*continued on next page*)

Table S6 – (*continued from previous page)*

T (K)	ρ (g cm^{-3})	d (%)	η (mPa s)	d (%)
293.153	1.567144	−0.02	502.3	4.9
303.154	1.557688	−0.001	340.1	−9.5
313.154	1.548301	0.01	–	–
323.155	1.539041	0.02	118.8	1.9
333.154	1.529946	0.008	76.6	3.8
343.154	1.520889	−0.001	53.09	1.5
353.154	1.511913	−0.02	38.73	−3.4

* The viscosity value measured at 313.154 K was considered an outlier and excluded from the fitted data.

Table S7. NMR chemical shifts values, δ (ppm), of ^{1}H and ^{19}F nuclei.

Nuclei	Peak	pure IL	0.1 Na[FSI]	0.3 Na[FSI]	0.5 Na[FSI]
^{1}H	1	3.488	3.472	3.443	3.409
^{1}H	2	3.295	3.279	3.245	3.205
^{1}H	3	3.281	3.258	3.232	3.185
^{1}H	4	3.275	3.237	3.214	3.165
^{1}H	5	3.263	–	–	–
^{1}H	6	3.255	–	–	–
^{1}H	7	3.024	3.009	2.983	2.954
^{1}H	8	2.226	2.216	2.202	2.182
^{1}H	9	–	1.860	1.838	–
^{1}H	10	1.853	1.841	1.820	–
^{1}H	11	1.833	1.822	1.801	–
^{1}H	12	1.820	1.812	1.790	1.776
^{1}H	13	1.813	1.801	1.780	1.767
^{1}H	14	1.794	1.782	1.760	1.757
^{1}H	15	1.776	1.764	1.743	–
^{1}H	16	1.014	1.005	0.990	0.972
^{1}H	17	0.996	0.987	0.972	0.955
^{1}H	17	0.978	0.969	0.974	–
^{19}F	1	52.546	52.621	52.701	52.626

Table S8. Self-diffusion coefficients of $C_3C_1pyr^+$, D_+, and FSI^-, D_-, determined from DOSY-NMR experiments at 298 K and extrapolated to 353 K using the present viscosity data.

	298 K		353 K	
Sample	$D_+(m^2\,s^{-1})$	$D_-(m^2\,s^{-1})$	$D_+(m^2\,s^{-1})$	$D_-(m^2\,s^{-1})$
pure IL	3.27×10^{-11}	3.44×10^{-11}	1.78×10^{-10}	1.87×10^{-10}
0.1 Na[FSI]	2.35×10^{-11}	2.57×10^{-11}	1.46×10^{-10}	1.59×10^{-10}
0.3 Na[FSI]	1.07×10^{-11}	1.10×10^{-11}	1.01×10^{-10}	1.04×10^{-10}
0.5 Na[FSI]	4.06×10^{-12}	2.69×10^{-12}	6.34×10^{-11}	4.20×10^{-11}

REFERENCES

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