Supporting information for: Ion Transport in Polymerized Ionic Liquid - Ionic Liquid Blends

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Table S1: Simulation details of polyIL-IL blend electrolytes with varying polyIL wt%.

polyIL wt%	#polyIL Chains	#Atoms	#$BMIM^+$	#PF_6^-	T_g
0 (pure IL)	0	8996	256	256	201 [a]
25	2	8266	192	256	250
50	4	8340	128	256	293
75	6	8414	64	256	362
100 (pure polyIL)	8	8488	0	256	453 [b]

[a] Reference.[S4]
[b] Reference.[S4]

1 Methods

1.1 Interaction Potential

We used the following interaction potential to describe the interactions in polyIL-IL blend electrolytes:

$$U(\boldsymbol{r}) = k_r^{\text{bonds}}\left(r - r_0\right)^2 + k_\theta^{\text{angles}}\left(\theta - \theta_0\right)^2 + \frac{1}{2}\sum_{n=1}^{4} K_n^{\text{dihedrals}}\left[1 + (-1)^{n+1}\cos\left(n\phi\right)\right] + K^{\text{impropers}}\left[1 + d\cos\left(n\phi\right)\right] + U^{\text{nb}}(\boldsymbol{r}). \quad (1)$$

The values of non-bonded and improper parameters for pure IL electrolytes are adopted from the optimized potential for liquid simulations - all atoms (OPLS-AA) force field set.[S1,S2] Additionally, refined interaction parameters based on quantum molecular dynamics simulations and charge scaling approach were taken from the work of Bhargava and Balasubramanian.[S3] For polyILs, the intramolecular parameters required to describe the polymerization of cations were taken from our previous work.[S4]

1.2 System Setup

The $BMIM^+$ cations were covalently linked to vinyl backbone at the head of imidazolium ring, as show in Figure S2, to eventually construct a polyIL chain containing 32 monomers, equivalent to a molecular weight of 4.87 kDa. The polyIL chain was equilibrated at classical level which includes a minimization, slow heating and a 5 ns long NPT simulation at an elevated temperature of 1000 K. Equilibrated geometry of a polyIL chain at 1000 K excluding counterions was randomly distributed in a simulation box. Number of polyIL chains required to generate a system of polyIL wt% of 100, 75, 50, 25 and 0 is 8, 6, 4, 2 and 0 chains, respectively. The systems with different polyIL wt% were prepared to be charge neutral by inserting fixed number of PF_6^- (256 ions) and appropriate number of non-polymerized $BMIM^+$ ions at random locations in simulation box. For instance, since a system with 8 polyIL chains contain 8×32 units of net positive electrostatic charge, we inserted 8×32=256 PF_6^- and 0 non-polymerized $BMIM^+$ ions to neutralize the excess charge on polymer backbone. Different systems prepared and details of the ionic species count were summarized in Table S1.

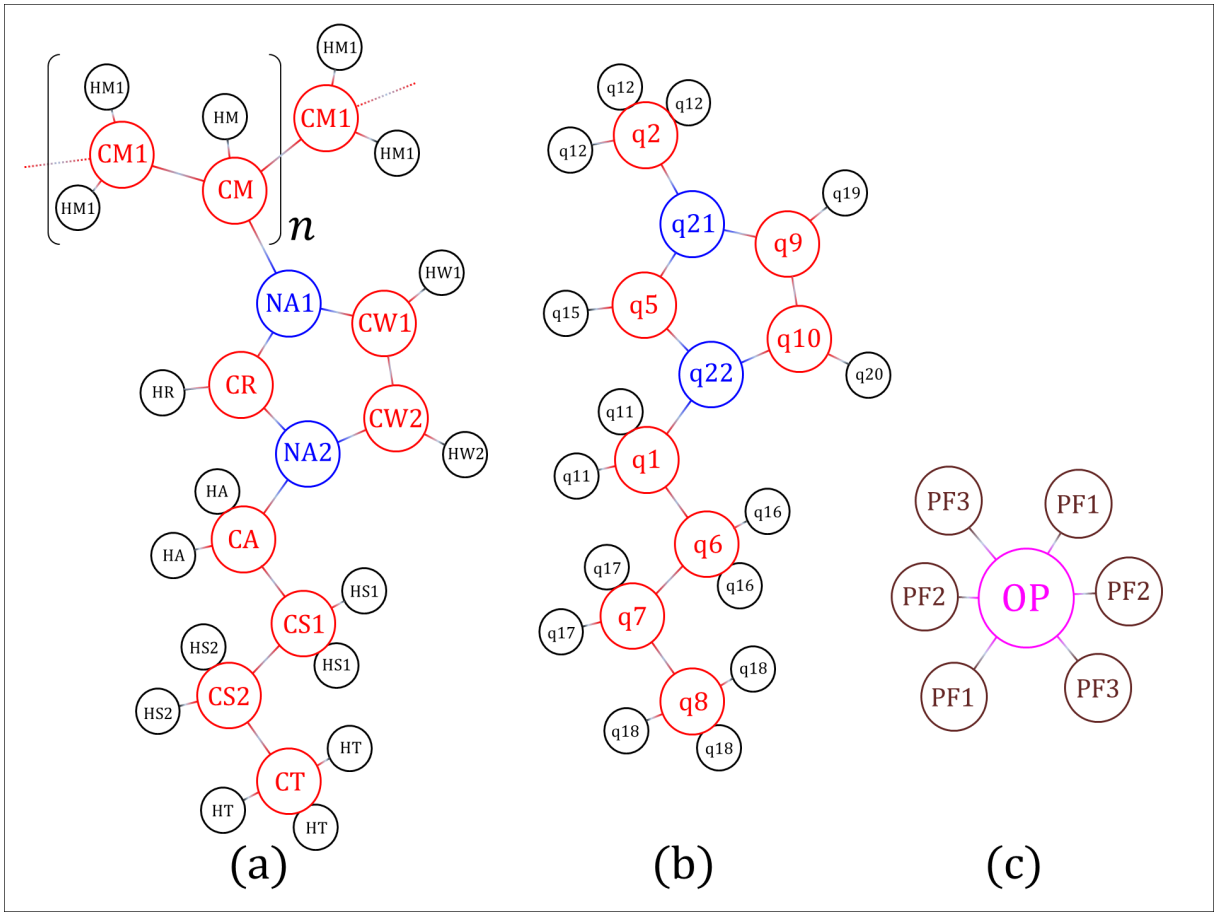


Figure S1: Atom type nomenclature used for (a) polyIL backbone and its polymerized $BMIM^+$ cation, (b) non-polymerized $BMIM^+$ cation and (c) PF_6^- anion, aiding the description of force field parameters provided in Tables S2-S7.

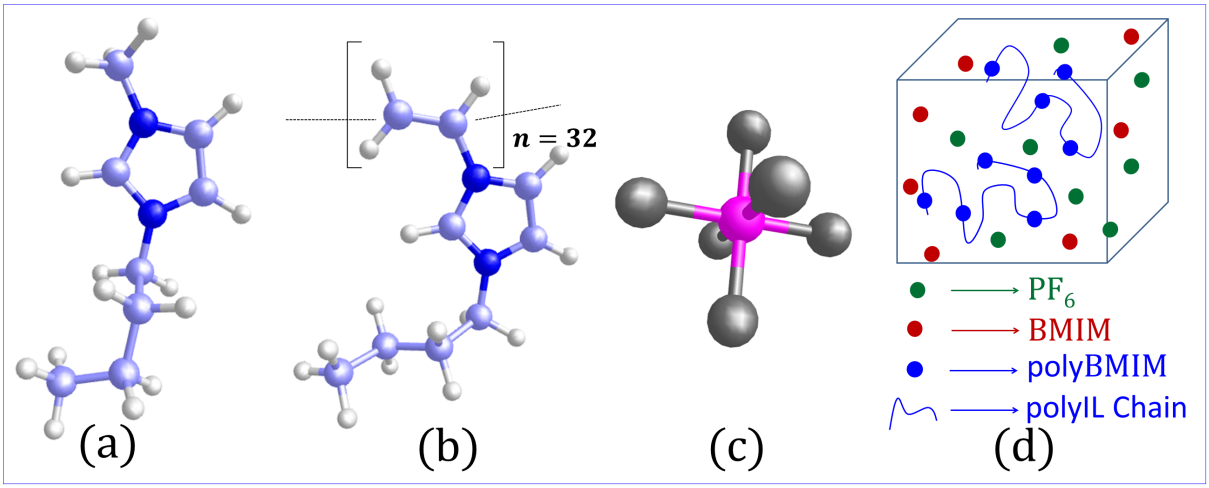


Figure S2: Chemical structure of (a) non-polymerized 1-butyl-3-methylimidazolium ($BMIM^+$) cation and (b) polymerized $BMIM^+$ ion and (c) hexafluorophosphate (PF_6^-) anion. The methyl group on $BMIM^+$ cation is replaced with vinyl linker to build polyIL chain. (d) Schematic of typical simulation box representing a polyIL-IL blend electrolyte containing polyIL chains, non-polymerized $BMIM^+$ and PF_6^- ions.

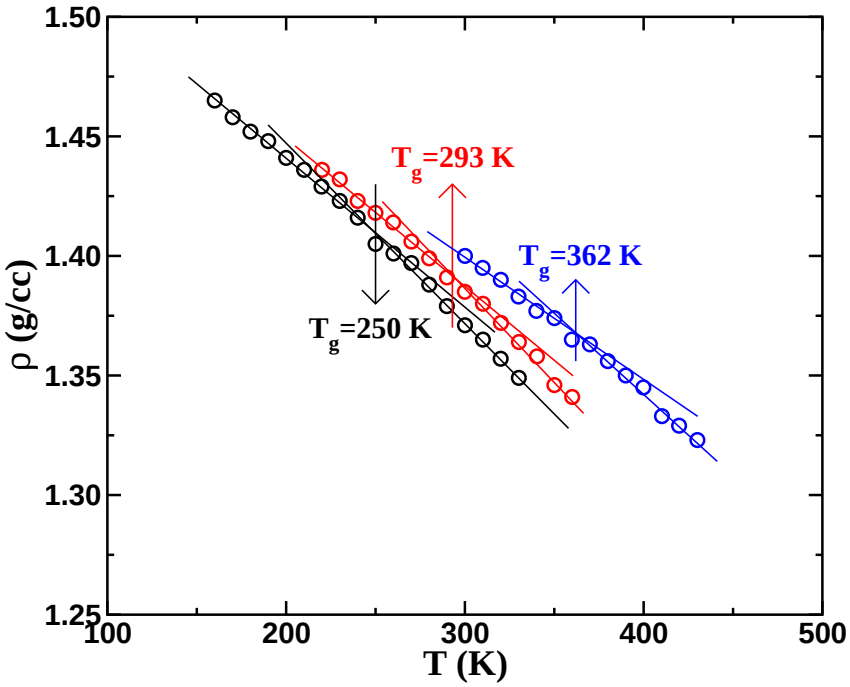


Figure S3: Density variation with temperature for polyIL-IL blend electrolytes to estimate their glass transition temperatures. Legend: Black-wt% 25, red-wt% 50 and blue-wt% 75.

1.3 System Size Dependency

The box size dependency is a standard issue to be carefully considered in MD simulations of finite box when discussing the absolute value of diffusivities. Unlike the structural features

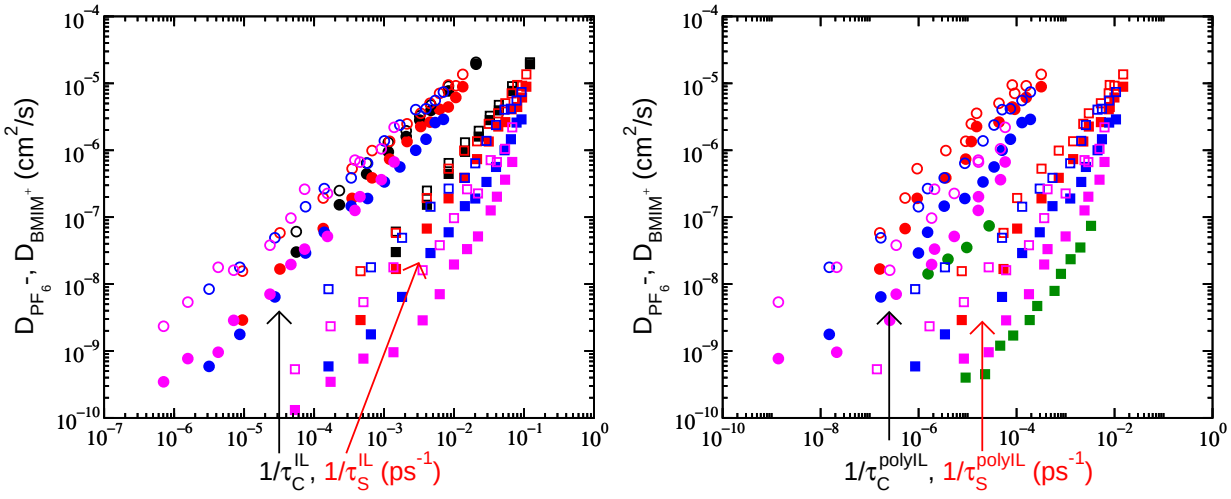


Figure S4: Diffusion coefficient of ions compared against inverse of different timescales corresponding to (a) non-polymerized IL pairs and (b) polyIL pairs in polyIL-IL blend electrolytes. Circles represent τ_C and squares represent τ_S. The closed and open symbols represent D_{PF6} and D_{BMIM}, respectively. The correlations shown in Figure 12 of the main article are replicated here for a comparison. Color legend is the same as Figure 12 of the main article.

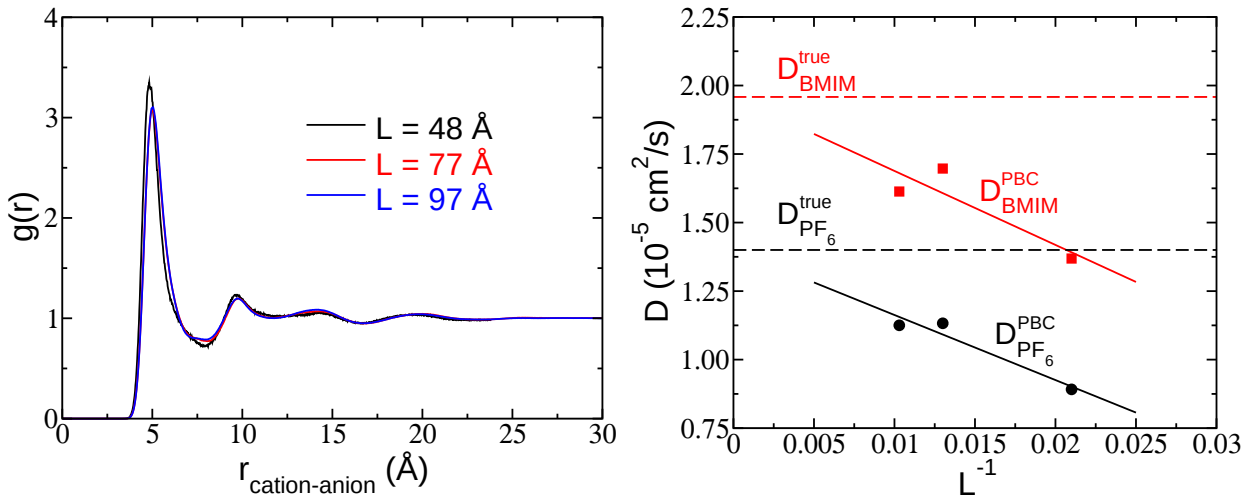
which do not depend (significantly) on box size, it is generally accepted in simulations of finite box that diffusivity changes with box.

To address the finite size issues, we undertook additional simulations (which were computationally expensive) for two larger box lengths (cubic boxes of sides 77 Å (1024 pairs of ions) and 97 Å (2048 pairs of ions) in addition to 48 Å (256 pairs of ions) of the main paper) and found only marginal changes to the structural features with box size (results displayed Figure S5(a)). In all three cases (including the results presented in the main manuscript), the RDFs do asymptote to unity, indicating recovery of bulk behavior at large distance.

The diffusivities shown in Figure S5(b) were found to exhibit a dependence on inverse box size similar to the relation $D_{\mathrm{PBC}} = D_{\mathrm{true}} - 2.837\frac{k_B T}{6\pi\eta \mathrm{L}}$ suggested by (i) Dunweg & Kremer[S5] and (ii) Yeh & Hummer.[S6] Such a result provides a simple means to extract (if needed) the asymptotic values of diffusivities which are free of box size dependencies.

Despite the above system size effects, our findings regarding the dependencies of diffusivities with respect to the different relaxation times in the system were found to be still applicable. Hence, none of the conclusions reported in the paper were influenced by the quantitative changes in diffusivities with increasing system size.

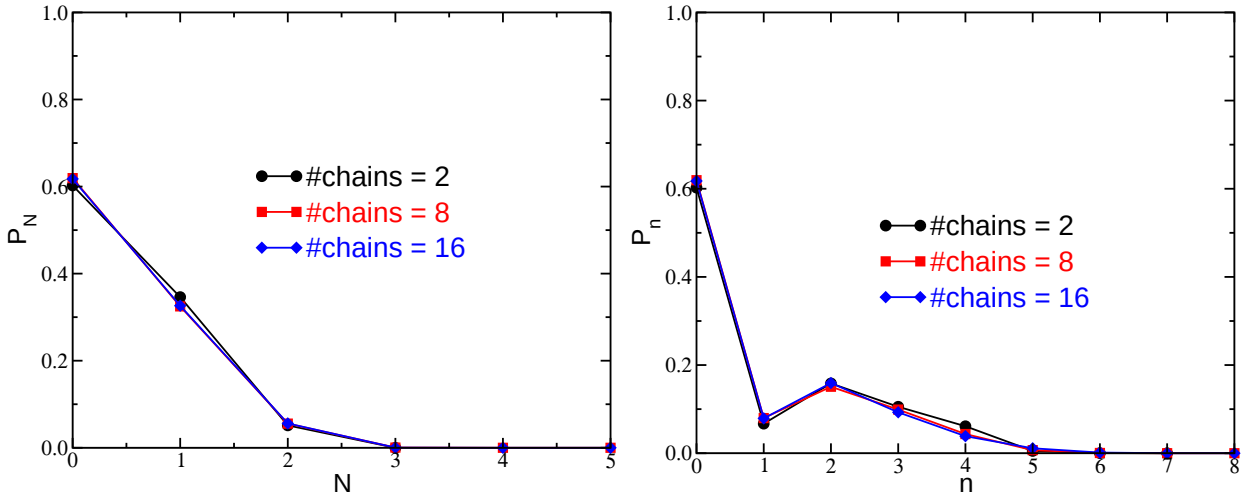
A closely related issue is the possibility of system size dependency on anion association statistics with the number of available chains for association. This is more relevant for the case of polyIL wt% 25 since there are only two chains in the simulation box for anion coordination and hopping. To examine the validity of main findings presented in the paper, we performed additional simulations with large number of chains at 25 and 50 wt% loadings of polyILs. The results presented in Figures S6 and S7 indicate that there are no significant



(a) (b)

Figure S5: System size dependency of the (a) radial distribution function between polymerized cations and anions and (b) diffusion coefficient of ions at wt% 25 and 600 K. The structure depends only marginally on the simulation box. The diffusion coefficient of anions and cations change with box size such that $D_{\text{PBC}} = D_{\text{true}} - 2.837\frac{k_B T}{6\pi\eta L}$, where D_{PBC} and D_{true} are diffusion coefficients calculated from simulations and the true value. The D_{true} shown in the Figure was calculated by extrapolating to infinite box length as an intercept on y-axis.

changes to the anion association behavior at any loading. While we were somewhat surprised at the lack of dependence on the number of chains, the outcomes do serve to validate the results and conclusions presented in the original manuscript.



(a) (b)

Figure S6: System size dependency of anion association statistics at wt% 25 and 600 K: (a) Probability of anion associating with N number of chains and (b) Probability of anion associating with n number of polymerized cations.

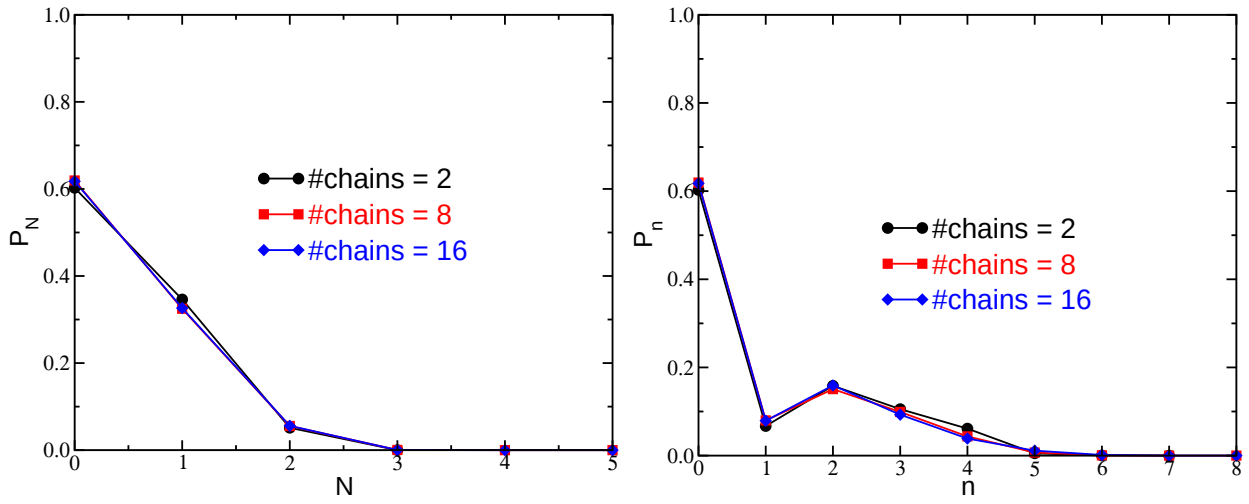


Figure S7: System size dependency of anion association statistics at wt% 50 and 600 K: (a) Probability of anion associating with N number of chains and (b) Probability of anion associating with n number of polymerized cations.

2 Complete List of Force Field Parameters for the polyIL-IL Blend Electrolytes

type i	ϵ_{ii}(kcal/mol)	σ_{ii}(Å)	Remarks
CA	0.066	3.50	OPLS-AA[S1,S2]
CM	0.066	3.50	OPLS-AA[S1,S2]
CM1	0.066	3.50	OPLS-AA[S1,S2]
$CM2_{terminus}$	0.066	3.50	OPLS-AA[S1,S2]
CR	0.070	3.55	OPLS-AA[S1,S2]
CS1	0.066	3.50	OPLS-AA[S1,S2]
CS2	0.066	3.50	OPLS-AA[S1,S2]
CT	0.066	3.50	OPLS-AA[S1,S2]
CW1	0.070	3.55	OPLS-AA[S1,S2]
CW2	0.070	3.55	OPLS-AA[S1,S2]
HA	0.030	1.92	BHARGAVA[S3]
HM	0.030	1.92	BHARGAVA[S3]
HM1	0.030	2.50	OPLS-AA[S1,S2]
HM2	0.030	2.50	OPLS-AA[S1,S2]
HR	0.030	1.72	BHARGAVA[S3]
HS1	0.030	2.50	OPLS-AA[S1,S2]
HS2	0.030	2.50	OPLS-AA[S1,S2]
HT	0.030	2.50	OPLS-AA[S1,S2]
HW1	0.030	1.72	BHARGAVA[S3]
HW2	0.030	1.72	BHARGAVA[S3]
NA1	0.170	3.25	OPLS-AA[S1,S2]
NA2	0.170	3.25	OPLS-AA[S1,S2]
OP	0.200	3.74	BHARGAVA[S3]
PF1	0.061	3.1181	OPLS-AA[S1,S2]
PF2	0.061	3.1181	OPLS-AA[S1,S2]

PF3	0.061	3.1181	OPLS-AA[S1,S2]
q1	0.066	3.50	OPLS-AA[S1,S2]
q2	0.066	3.50	OPLS-AA[S1,S2]
q5	0.070	3.55	OPLS-AA[S1,S2]
q6	0.066	3.50	OPLS-AA[S1,S2]
q7	0.066	3.50	OPLS-AA[S1,S2]
q8	0.066	3.50	OPLS-AA[S1,S2]
q9	0.070	3.55	OPLS-AA[S1,S2]
q10	0.070	3.55	OPLS-AA[S1,S2]
q11	0.030	1.92	BHARGAVA[S3]
q12	0.030	1.92	BHARGAVA[S3]
q15	0.030	1.72	BHARGAVA[S3]
q16	0.030	2.50	OPLS-AA[S1,S2]
q17	0.030	2.50	OPLS-AA[S1,S2]
q18	0.030	2.50	OPLS-AA[S1,S2]
q19	0.030	1.72	BHARGAVA[S3]
q20	0.030	1.72	BHARGAVA[S3]
q21	0.170	3.25	OPLS-AA[S1,S2]
q22	0.170	3.25	OPLS-AA[S1,S2]

Table S2: Force field parameters for the Lennard-Jones interactions.

type i	Mass (a.m.u)	$q_i(e)$	Remarks
CA	12.01070	-0.071916	Ref. S4
HA	01.00794	0.103288	Ref. S4
CS1	12.01070	-0.112314	Ref. S4
HS1	01.00794	0.063370	Ref. S4
CS2	12.01070	0.058799	Ref. S4
HS2	01.00794	0.032434	Ref. S4
CT	12.01070	-0.168031	Ref. S4
HT	01.00794	0.056042	Ref. S4
NA2	14.00670	0.083055	Ref. S4
CR	12.01070	-0.081640	Ref. S4
HR	01.00794	0.215218	Ref. S4
NA1	14.00670	0.276722	Ref. S4
CW1	12.01070	-0.203382	Ref. S4
HW1	01.00794	0.238040	Ref. S4
CW2	12.01070	-0.165764	Ref. S4
HW2	01.00794	0.244756	Ref. S4
CM	12.01070	-0.191281	Ref. S4
HM	01.00794	0.200553	Ref. S4
CM1	12.01070	-0.286559	Ref. S4
HM1	01.00794	0.198717	Ref. S4
CM2	12.01070	-0.1432795	Ref. S4
HM2	01.00794	0.066239	Ref. S4
OP	30.973761	1.340000	OPLS-AA[S1,S2]
PF1	18.997999	-0.390000	OPLS-AA[S1,S2]
PF2	18.997999	-0.390000	OPLS-AA[S1,S2]
PF3	18.997999	-0.390000	OPLS-AA[S1,S2]
q1	12.010700	-0.2941	OPLS[S2]
q10	12.010700	-0.2417	OPLS[S2]
q11	01.007940	0.1463	OPLS[S2]
q12	01.007940	0.1567	OPLS[S2]
q15	01.007940	0.2095	OPLS[S2]
q16	01.007940	0.0245	OPLS[S2]
q17	01.007940	-0.0036	OPLS[S2]
q18	01.007940	0.0964	OPLS[S2]
q19	01.007940	0.2665	OPLS[S2]
q2	12.010700	-0.3599	OPLS[S2]
q20	01.007940	0.2703	OPLS[S2]
q21	14.006700	0.2444	OPLS[S2]
q22	14.006700	0.2677	OPLS[S2]
q5	12.010700	-0.1151	OPLS[S2]
q6	12.010700	0.1013	OPLS[S2]
q7	12.010700	0.1360	OPLS[S2]
q8	12.010700	-0.3220	OPLS[S2]
q9	12.010700	-0.2566	OPLS[S2]

Table S3: Force field parameters for the Coulomb interactions. The charges were scaled by a factor of 0.8 uniformly on all atoms of anion and cation.

type i	type j	k_r (kcal/mol/Å^2)	r_0 (Å)	Remarks
CA	CS1	280.159	1.534	Ref. S4
CA	HA	357.723	1.092	Ref. S4
CA	NA2	287.313	1.478	Ref. S4
CS1	HS1	347.494	1.091	Ref. S4
CS1	CS2	281.515	1.536	Ref. S4
CS2	HS2	349.100	1.095	Ref. S4
CS2	CT	287.616	1.532	Ref. S4
CT	HT	351.780	1.093	Ref. S4
NA2	CR	552.388	1.335	Ref. S4
NA2	CW2	408.436	1.384	Ref. S4
CR	HR	401.204	1.074	Ref. S4
CR	NA1	464.417	1.385	Ref. S4
NA1	CW1	410.333	1.386	Ref. S4
NA1	CM	274.189	1.486	Ref. S4
CW1	CW2	531.524	1.352	Ref. S4
CW1	HW1	396.395	1.081	Ref. S4
CW2	HW2	395.457	1.079	Ref. S4
CM	HM	363.651	1.083	Ref. S4
CM	CM1	276.101	1.532	Ref. S4
CM	CM2	276.101	1.532	Ref. S4
CM1	HM1	348.875	1.095	Ref. S4
CM2	HM2	348.875	1.095	Ref. S4
OP	PF1	500.000	1.596	OPLS-AA[S1,S2]
OP	PF2	500.000	1.596	OPLS-AA[S1,S2]
OP	PF3	500.000	1.596	OPLS-AA[S1,S2]
q8	q18	340.000	1.084	OPLS-AA[S1,S2]
q5	q21	477.000	1.315	OPLS-AA[S1,S2]
q5	q22	477.000	1.315	OPLS-AA[S1,S2]
q9	q10	520.000	1.336	OPLS-AA[S1,S2]
q1	q11	340.000	1.080	OPLS-AA[S1,S2]
q2	q21	337.000	1.465	OPLS-AA[S1,S2]
q6	q16	340.000	1.087	OPLS-AA[S1,S2]
q7	q17	340.000	1.087	OPLS-AA[S1,S2]
q1	q6	268.000	1.526	OPLS-AA[S1,S2]
q1	q22	337.000	1.476	OPLS-AA[S1,S2]
q6	q7	268.000	1.531	OPLS-AA[S1,S2]
q2	q12	340.000	1.080	OPLS-AA[S1,S2]
q7	q8	268.000	1.528	OPLS-AA[S1,S2]
q5	q15	367.000	1.069	OPLS-AA[S1,S2]
q9	q19	367.000	1.068	OPLS-AA[S1,S2]
q10	q20	367.000	1.068	OPLS-AA[S1,S2]
q10	q22	427.000	1.378	OPLS-AA[S1,S2]
q9	q21	427.000	1.378	OPLS-AA[S1,S2]

Table S4: Force field parameters for the bonded interactions.

type i	type j	type k	k_θ (kcal/mol/rad^2)	θ_0 (degrees)	Remarks
HA	CA	CS1	63.034	111.966	Ref. S4
CS1	CA	NA2	78.829	112.124	Ref. S4
HA	CA	NA2	71.809	106.307	Ref. S4
HA	CA	HA	63.013	107.853	Ref. S4
CA	CS1	HS1	67.208	109.146	Ref. S4
CA	CS1	CS2	80.939	112.443	Ref. S4
HS1	CS1	CS2	55.219	108.941	Ref. S4
HS1	CS1	HS1	56.563	107.458	Ref. S4
CS1	CS2	HS2	59.461	107.908	Ref. S4
CS1	CS2	CT	80.400	114.212	Ref. S4
HS2	CS2	HS2	82.095	106.005	Ref. S4
HS2	CS2	CT	58.884	109.477	Ref. S4
CS2	CT	HT	63.976	111.352	Ref. S4
HT	CT	HT	68.335	108.358	Ref. S4
CA	NA2	CR	84.594	126.178	Ref. S4
CA	NA2	CW2	88.387	126.334	Ref. S4
CR	NA2	CW2	97.245	108.165	Ref. S4
NA2	CR	HR	66.082	125.132	Ref. S4
NA2	CR	NA1	252.744	109.164	Ref. S4
HR	CR	NA1	64.617	125.669	Ref. S4
CR	NA1	CW1	266.725	107.813	Ref. S4
CR	NA1	CM	90.685	126.384	Ref. S4
CW1	NA1	CM	97.374	125.886	Ref. S4
NA1	CW1	CW2	255.171	106.709	Ref. S4
NA1	CW1	HW1	68.306	122.790	Ref. S4
HW1	CW1	CW2	58.627	130.162	Ref. S4
NA2	CW2	CW1	268.303	107.291	Ref. S4
NA2	CW2	HW2	61.818	122.991	Ref. S4
CW1	CW2	HW2	64.721	130.027	Ref. S4
NA1	CM	HM	81.775	104.182	Ref. S4
NA1	CM	CM1	100.742	109.948	Ref. S4
NA1	CM	CM2	100.742	109.948	Ref. S4
CM1	CM	HM	62.360	109.018	Ref. S4
CM2	CM	HM	62.360	109.018	Ref. S4
CM1	CM	CM1	86.605	113.354	Ref. S4
CM1	CM	CM2	86.605	113.354	Ref. S4
CM	CM1	CM	93.377	116.755	Ref. S4
CM	CM1	HM1	65.863	109.352	Ref. S4
CM	CM2	HM2	65.863	109.352	Ref. S4
HM1	CM1	HM1	54.233	108.068	Ref. S4
HM2	CM2	HM2	54.233	108.068	Ref. S4
PF1	OP	PF1	75.000	180.000	OPLS-AA[S1,S2]
PF2	OP	PF2	75.000	180.000	OPLS-AA[S1,S2]
PF3	OP	PF3	75.000	180.000	OPLS-AA[S1,S2]
PF1	OP	PF2	75.000	90.000	OPLS-AA[S1,S2]
PF1	OP	PF3	75.000	90.000	OPLS-AA[S1,S2]
PF2	OP	PF3	75.000	90.000	OPLS-AA[S1,S2]
q15	q5	q21	35.00	125.10	OPLS-AA[S1,S2]
q15	q5	q22	35.00	125.10	OPLS-AA[S1,S2]
q11	q1	q11	33.00	108.90	OPLS-AA[S1,S2]
q18	q8	q18	33.00	107.90	OPLS-AA[S1,S2]
q19	q9	q21	35.00	122.00	OPLS-AA[S1,S2]
q20	q10	q22	35.00	122.00	OPLS-AA[S1,S2]
q1	q22	q5	70.00	126.80	OPLS-AA[S1,S2]
q2	q21	q9	70.00	125.60	OPLS-AA[S1,S2]
q7	q8	q18	37.50	111.10	OPLS-AA[S1,S2]
q12	q2	q12	33.00	109.80	OPLS-AA[S1,S2]

q9	q10	q20	35.00	130.90	OPLS-AA[S1,S2]
q10	q9	q19	35.00	130.90	OPLS-AA[S1,S2]
q9	q10	q22	70.00	107.10	OPLS-AA[S1,S2]
q10	q9	q21	70.00	107.10	OPLS-AA[S1,S2]
q21	q5	q22	70.00	109.80	OPLS-AA[S1,S2]
q6	q1	q11	37.50	111.10	OPLS-AA[S1,S2]
q11	q1	q22	37.50	107.50	OPLS-AA[S1,S2]
q16	q6	q16	33.00	106.70	OPLS-AA[S1,S2]
q17	q7	q17	33.00	106.70	OPLS-AA[S1,S2]
q1	q22	q10	70.00	125.30	OPLS-AA[S1,S2]
q1	q6	q16	37.50	108.60	OPLS-AA[S1,S2]
q5	q21	q9	70.00	107.90	OPLS-AA[S1,S2]
q5	q22	q10	70.00	107.90	OPLS-AA[S1,S2]
q6	q7	q17	37.50	109.60	OPLS-AA[S1,S2]
q7	q6	q16	37.50	109.60	OPLS-AA[S1,S2]
q1	q6	q7	58.35	113.30	OPLS-AA[S1,S2]
q8	q7	q17	37.50	109.70	OPLS-AA[S1,S2]
q6	q1	q22	58.35	113.00	OPLS-AA[S1,S2]
q6	q7	q8	58.35	112.30	OPLS-AA[S1,S2]
q12	q2	q21	37.50	109.20	OPLS-AA[S1,S2]
q2	q21	q5	70.00	126.40	OPLS-AA[S1,S2]

Table S5: Force field parameters for the angle interactions.

type i	type j	type k	type l	K_1	K_2	K_3	K_4	Remarks
NA2	CR	NA1	CW1	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
HM	CM	NA1	CR	0.000	0.000	0.000	0.0	OPLS-AA[S1,S2]
HS1	CS1	CS2	CT	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
CM2	CM	CM1	HM1	0.000	0.000	2.67777	0.0	Ref. S4
CM1	CM	CM1	HM1	0.000	0.000	2.67777	0.0	Ref. S4
CM1	CM	CM2	HM2	0.000	0.000	2.67777	0.0	Ref. S4
HM	CM	CM1	CM	0.000	0.000	2.67777	0.0	Ref. S4
HA	CA	NA2	CW2	-1.400	-2.650	0.175	0.0	OPLS-AA[S1,S2]
HA	CA	NA2	CR	0.000	0.000	0.000	0.0	OPLS-AA[S1,S2]
CW1	CW2	NA2	CA	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
HA	CA	CS1	CS2	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
NA2	CA	CS1	CS2	-1.788	0.756	-0.288	0.0	OPLS-AA[S1,S2]
CW1	CW2	NA2	CR	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
CM1	CM	NA1	CW1	0.000	0.000	2.37904	0.0	Ref. S4
CM2	CM	NA1	CW1	0.000	0.000	2.37904	0.0	Ref. S4
CM1	CM	NA1	CR	2.45324	-0.559275	-0.813571	1.52831	Ref. S4
CM2	CM	NA1	CR	2.45324	-0.559275	-0.813571	1.52831	Ref. S4
CW2	CW1	NA1	CM	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
HW1	CW1	CW2	HW2	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
HM	CM	CM2	HM2	0.000	0.000	2.78174	0.0	Ref. S4
HM	CM	CM1	HM1	0.000	0.000	2.78174	0.0	Ref. S4
HW1	CW1	CW2	NA2	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
NA1	CW1	CW2	HW2	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
NA1	CM	CM1	HM1	-0.839253	-0.310509	4.68038	0.846371	Ref. S4
NA1	CM	CM2	HM2	-0.839253	-0.310509	4.68038	0.846371	Ref. S4
NA1	CW1	CW2	NA2	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
HM	CM	NA1	CW1	0.000	0.000	0.124	0.0	OPLS-AA[S1,S2]
HS1	CS1	CS2	HS2	0.000	0.000	0.318	0.0	OPLS-AA[S1,S2]
HA	CA	CS1	HS1	0.000	0.000	0.318	0.0	OPLS-AA[S1,S2]
NA2	CA	CS1	HS1	0.000	0.000	0.000	0.0	OPLS-AA[S1,S2]
CS1	CA	NA2	CW2	-1.910	-1.500	0.290	0.0	OPLS-AA[S1,S2]
CS1	CA	NA2	CR	-1.659	-0.555	-0.375	0.0	OPLS-AA[S1,S2]
HR	CR	NA1	CW1	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
NA1	CR	NA2	CW2	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
NA1	CR	NA2	CA	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
NA1	CM	CM1	CM	0.000	0.000	4.44212	0.0	Ref. S4
HW2	CW2	NA2	CA	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
CS1	CS2	CT	HT	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
CA	CS1	CS2	CT	1.300	-0.050	0.200	0.0	OPLS-AA[S1,S2]
CM1	CM	CM1	CM	-0.775803	0.313249	4.69238	0.31007	Ref. S4
CM2	CM	CM1	CM	-0.775803	0.313249	4.69238	0.31007	Ref. S4
HW1	CW1	NA1	CM	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
HW2	CW2	NA2	CR	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
CW2	CW1	NA1	CR	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
HR	CR	NA2	CW2	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
HR	CR	NA2	CA	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
NA2	CR	NA1	CM	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
HR	CR	NA1	CM	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
HW1	CW1	NA1	CR	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
HS2	CS2	CT	HT	0.000	0.000	0.318	0.0	OPLS-AA[S1,S2]
CA	CS1	CS2	HS2	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
q11	q1	q22	q5	0.000	0.000	0.000	0.0	OPLS-AA[S1,S2]
q10	q9	q21	q2	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q22	q5	q21	q2	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]

q6	q1	q22	q10	-1.910	-1.50	0.29	0.0	OPLS-AA[S1,S2]
q1	q6	q7	q17	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
q17	q7	q8	q18	0.000	0.000	0.318	0.0	OPLS-AA[S1,S2]
q15	q5	q21	q2	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
q11	q1	q22	q10	-1.400	-2.65	0.175	0.0	OPLS-AA[S1,S2]
q11	q1	q6	q16	0.000	0.000	0.318	0.0	OPLS-AA[S1,S2]
q21	q5	q22	q1	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
q9	q10	q22	q1	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q1	q6	q7	q8	1.300	-0.050	0.200	0.0	OPLS-AA[S1,S2]
q19	q9	q21	q2	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q10	q9	q21	q5	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q9	q10	q22	q5	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q12	q2	q21	q5	0.000	0.000	0.000	0.0	OPLS-AA[S1,S2]
q11	q1	q6	q7	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
q19	q9	q10	q20	0.000	10.750	0.000	0.0	OPLS-AA[S1,S2]
q19	q9	q10	q22	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
q20	q10	q9	q21	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
q21	q9	q10	q22	0.000	10.75	0.000	0.0	OPLS-AA[S1,S2]
q15	q5	q22	q1	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
q16	q6	q7	q17	0.000	0.000	0.318	0.0	OPLS-AA[S1,S2]
q20	q10	q22	q1	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q22	q1	q6	q16	0.000	0.000	0.000	0.0	OPLS-AA[S1,S2]
q21	q5	q22	q10	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
q22	q5	q21	q9	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
q19	q9	q21	q5	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q20	q10	q22	q5	0.000	3.000	0.000	0.0	OPLS-AA[S1,S2]
q12	q2	q21	q9	0.000	0.000	0.124	0.0	OPLS-AA[S1,S2]
q6	q1	q22	q5	-1.659	-0.555	-0.375	0.0	OPLS-AA[S1,S2]
q8	q7	q6	q16	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
q6	q7	q8	q18	0.000	0.000	0.366	0.0	OPLS-AA[S1,S2]
q22	q1	q6	q7	-1.788	0.756	-0.288	0.0	OPLS-AA[S1,S2]
q15	q5	q22	q10	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]
q15	q5	q21	q9	0.000	4.651	0.000	0.0	OPLS-AA[S1,S2]

Table S6: Force field parameters for the dihedral interactions; Units of $K_{1,2,3,4}^{\text{dihedrals}}$ are kcal/mol.

type i	type j	type k	type l	$K^{\text{impropers}}$ (kcal/mol)	d	n	Remarks
X	X	NA1	X	1.0	-1	2	OPLS-AA [S1,S2]
X	X	NA2	X	1.0	-1	2	OPLS-AA [S1,S2]
X	X	CW1	X	1.1	-1	2	OPLS-AA [S1,S2]
X	X	CW2	X	1.1	-1	2	OPLS-AA [S1,S2]
X	X	CR	X	1.1	-1	2	OPLS-AA [S1,S2]
X	X	q21	X	1.0	-1	2	OPLS-AA [S1,S2]
X	X	q22	X	1.0	-1	2	OPLS-AA [S1,S2]
X	X	q9	X	1.1	-1	2	OPLS-AA [S1,S2]
X	X	q10	X	1.1	-1	2	OPLS-AA [S1,S2]
X	X	q5	X	1.1	-1	2	OPLS-AA [S1,S2]

Table S7: Force field parameters for improper interactions. Note that X is any possible atom type that is bonded to atom of type k.

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