

Supporting information for:

Langevin behavior of the dielectric decrement in ionic liquid water mixtures

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1 Ionic liquid force fields

In the following, we give the complete file of topology and parameters readable by the program package CHARMM [1].

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READ RTF CARD
=====
*.  T O P O L O G Y   O F   I O N I C   L I Q U I D S
*.
*.  Parametrization: GAAMP, scaling of epsilon_LJ
=====
*
  99      1
MASS      1 H1      1.008000
MASS      2 H4      1.008000
MASS      3 H5      1.008000
MASS      4 HC      1.008000
MASS      6 C3      12.010000
MASS      7 CC      12.010000
MASS      8 CD      12.010000
MASS      9 CG      12.010000
MASS     10 O       16.000000
MASS     13 N1      14.010000
MASS     14 NA      14.010000
MASS     15 NE      14.010000
MASS     16 F       19.000000
MASS     18 S6      32.060000
MASS     60 LP      0.000000    ! general lone pair
MASS     61 DRUD    0.000000    ! drude particle

AUTOGENERATE DRUDE ! use of DRUDE
!=====
!EMIM
!=====
RESI EMIM 1.000
GROUP
ATOM C      C3      0.045  ALPHA -2.025  THOLE 1.135
ATOM H      H1      0.053
ATOM H1     H1      0.053
ATOM H2     H1      0.053
ATOM N      NA      0.102  ALPHA -0.901  THOLE 1.138
ATOM C1     CC      0.003  ALPHA -1.533  THOLE 1.409
ATOM N1     NA      0.102  ALPHA -0.901  THOLE 1.138
ATOM C2     CC     -0.076  ALPHA -1.563  THOLE 1.324
ATOM C3     CD     -0.076  ALPHA -1.563  THOLE 1.324
ATOM H3     H5      0.183

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ATOM	H4	H4	0.168			
ATOM	H5	H4	0.168			
ATOM	C4	C3	-0.005	ALPHA	-1.675	THOLE 1.109
ATOM	H6	H1	0.075			
ATOM	H7	H1	0.075			
ATOM	C5	C3	0.008	ALPHA	-2.049	THOLE 1.125
ATOM	H8	HC	0.023			
ATOM	H9	HC	0.023			
ATOM	H10	HC	0.023			
BOND	C	H		!	DIST	1.0796
BOND	C	H1		!	DIST	1.0785
BOND	C	H2		!	DIST	1.0794
BOND	C	N1		!	DIST	1.4658
BOND	N	C1		!	DIST	1.3138
BOND	N	C3		!	DIST	1.3781
BOND	N	C4		!	DIST	1.4777
BOND	C1	N1		!	DIST	1.3157
BOND	C1	H3		!	DIST	1.0692
BOND	N1	C2		!	DIST	1.3785
BOND	C2	C3		!	DIST	1.3416
BOND	C2	H4		!	DIST	1.0679
BOND	C3	H5		!	DIST	1.0688
BOND	C4	H6		!	DIST	1.0814
BOND	C4	H7		!	DIST	1.0798
BOND	C4	C5		!	DIST	1.5218
BOND	C5	H8		!	DIST	1.0835
BOND	C5	H9		!	DIST	1.0834
BOND	C5	H10		!	DIST	1.0837
ANGL	C	N1	C1	!	ANGLE	126.4111
ANGL	C	N1	C2	!	ANGLE	125.6300
ANGL	H	C	H1	!	ANGLE	109.5510
ANGL	H	C	H2	!	ANGLE	110.2559
ANGL	H	C	N1	!	ANGLE	109.2611
ANGL	H1	C	H2	!	ANGLE	109.5172
ANGL	H1	C	N1	!	ANGLE	108.8792
ANGL	H2	C	N1	!	ANGLE	109.3538
ANGL	N	C1	N1	!	ANGLE	109.8941
ANGL	N	C1	H3	!	ANGLE	125.1030
ANGL	N	C3	C2	!	ANGLE	107.1959
ANGL	N	C3	H5	!	ANGLE	122.1025
ANGL	N	C4	H6	!	ANGLE	106.7043
ANGL	N	C4	H7	!	ANGLE	106.9082
ANGL	N	C4	C5	!	ANGLE	112.0836
ANGL	C1	N	C3	!	ANGLE	107.9285
ANGL	C1	N	C4	!	ANGLE	126.1921
ANGL	C1	N1	C2	!	ANGLE	107.9577
ANGL	N1	C1	H3	!	ANGLE	125.0025
ANGL	N1	C2	C3	!	ANGLE	107.0236
ANGL	N1	C2	H4	!	ANGLE	122.0406
ANGL	C2	C3	H5	!	ANGLE	130.7003
ANGL	C3	N	C4	!	ANGLE	125.8669
ANGL	C3	C2	H4	!	ANGLE	130.9352
ANGL	C4	C5	H8	!	ANGLE	111.2780
ANGL	C4	C5	H9	!	ANGLE	111.5813
ANGL	C4	C5	H10	!	ANGLE	109.0336
ANGL	H6	C4	H7	!	ANGLE	107.7086
ANGL	H6	C4	C5	!	ANGLE	111.3594
ANGL	H7	C4	C5	!	ANGLE	111.7864
ANGL	H8	C5	H9	!	ANGLE	108.5717
ANGL	H8	C5	H10	!	ANGLE	108.3168
ANGL	H9	C5	H10	!	ANGLE	107.9497
DIHE	H	C	N1	C1	!	DIHE -120.0991
DIHE	H1	C	N1	C1	!	DIHE -0.4861
DIHE	H2	C	N1	C1	!	DIHE 119.1407
DIHE	H	C	N1	C2	!	DIHE 60.3500
DIHE	H1	C	N1	C2	!	DIHE 179.9630
DIHE	H2	C	N1	C2	!	DIHE -60.4102
DIHE	C3	N	C1	N1	!	DIHE -0.0723

DIHE	C4	N	C1	N1	!	DIHE	-178.8479
DIHE	C3	N	C1	H3	!	DIHE	-179.8641
DIHE	C4	N	C1	H3	!	DIHE	1.3603
DIHE	C1	N	C3	C2	!	DIHE	0.1214
DIHE	C4	N	C3	C2	!	DIHE	178.9021
DIHE	C1	N	C3	H5	!	DIHE	179.7449
DIHE	C4	N	C3	H5	!	DIHE	-1.4744
DIHE	C1	N	C4	H6	!	DIHE	-17.3257
DIHE	C3	N	C4	H6	!	DIHE	164.1119
DIHE	C1	N	C4	H7	!	DIHE	-132.3592
DIHE	C3	N	C4	H7	!	DIHE	49.0783
DIHE	C1	N	C4	C5	!	DIHE	104.8236
DIHE	C3	N	C4	C5	!	DIHE	-73.7389
DIHE	N	C1	N1	C	!	DIHE	-179.6202
DIHE	H3	C1	N1	C	!	DIHE	0.1719
DIHE	N	C1	N1	C2	!	DIHE	-0.0039
DIHE	H3	C1	N1	C2	!	DIHE	179.7882
DIHE	C	N1	C2	C3	!	DIHE	179.7003
DIHE	C1	N1	C2	C3	!	DIHE	0.0802
DIHE	C	N1	C2	H4	!	DIHE	-0.0517
DIHE	C1	N1	C2	H4	!	DIHE	-179.6718
DIHE	N1	C2	C3	N	!	DIHE	-0.1217
DIHE	H4	C2	C3	N	!	DIHE	179.6001
DIHE	N1	C2	C3	H5	!	DIHE	-179.7009
DIHE	H4	C2	C3	H5	!	DIHE	0.0208
DIHE	N	C4	C5	H8	!	DIHE	-60.5075
DIHE	H6	C4	C5	H8	!	DIHE	58.9472
DIHE	H7	C4	C5	H8	!	DIHE	179.4809
DIHE	N	C4	C5	H9	!	DIHE	60.9322
DIHE	H6	C4	C5	H9	!	DIHE	-179.6130
DIHE	H7	C4	C5	H9	!	DIHE	-59.0794
DIHE	N	C4	C5	H10	!	DIHE	-179.9216
DIHE	H6	C4	C5	H10	!	DIHE	-60.4668
DIHE	H7	C4	C5	H10	!	DIHE	60.0669

IMPH	C4	C1	N	C3
IMPH	H3	N1	C1	N
IMPH	C	C1	N1	C2
IMPH	C3	H4	C2	N1
IMPH	C2	H5	C3	N

! =====
! TRIF
! =====

RESI TRIF -1.000

GROUP

ATOM	C	C3	0.366	ALPHA	-1.227	THOLE	1.068
ATOM	F	F	-0.190	ALPHA	-0.028	THOLE	1.450
ATOM	F1	F	-0.190	ALPHA	-0.028	THOLE	1.450
ATOM	F2	F	-0.190	ALPHA	-0.028	THOLE	1.450
ATOM	S	S6	0.998	ALPHA	-2.658	THOLE	1.128
ATOM	O	O	-0.353	ALPHA	-0.271	THOLE	1.471
ATOM	LPA0	LP	-0.145				
ATOM	LPB0	LP	-0.100				
ATOM	O1	O	-0.353	ALPHA	-0.271	THOLE	1.471
ATOM	LPA01	LP	-0.145				
ATOM	LPB01	LP	-0.100				
ATOM	O2	O	-0.353	ALPHA	-0.271	THOLE	1.471
ATOM	LPA02	LP	-0.145				
ATOM	LPB02	LP	-0.100				

BOND	C	F		!	DIST	1.3239
BOND	C	F1		!	DIST	1.3237
BOND	C	F2		!	DIST	1.3237
BOND	C	S		!	DIST	1.8184
BOND	S	O		!	DIST	1.4431
BOND	S	O1		!	DIST	1.4425
BOND	S	O2		!	DIST	1.4425
BOND		O	LPA0		O	LPB0
BOND		O1	LPA01		O1	LPB01
BOND		O2	LPA02		O2	LPB02

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LONEPAIR relative LPA0 0 S C distance 0.35 angle 110.00 dihe 0.00
LONEPAIR relative LPB0 0 S C distance 0.35 angle 110.00 dihe 180.00
ANISOTROPY 0 S LPA0 LPB0 A11 0.56750 A22 0.37401
LONEPAIR relative LPA01 01 S C distance 0.35 angle 110.00 dihe 0.00
LONEPAIR relative LPB01 01 S C distance 0.35 angle 110.00 dihe 180.00
ANISOTROPY 01 S LPA01 LPB01 A11 0.56750 A22 0.37401
LONEPAIR relative LPA02 02 S C distance 0.35 angle 110.00 dihe 0.00
LONEPAIR relative LPB02 02 S C distance 0.35 angle 110.00 dihe 180.00
ANISOTROPY 02 S LPA02 LPB02 A11 0.56750 A22 0.37401

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ANGL C S 0 ! ANGLE 102.6274
ANGL C S 01 ! ANGLE 102.6487
ANGL C S 02 ! ANGLE 102.6487
ANGL F C F1 ! ANGLE 107.1013
ANGL F C F2 ! ANGLE 107.1013
ANGL F C S ! ANGLE 111.7239
ANGL F1 C F2 ! ANGLE 107.1326
ANGL F1 C S ! ANGLE 111.7459
ANGL F2 C S ! ANGLE 111.7459
ANGL O S 01 ! ANGLE 115.3458
ANGL O S 02 ! ANGLE 115.3458
ANGL O1 S 02 ! ANGLE 115.3600

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DIHE F C S 0 ! DIHE -180.0000
DIHE F1 C S 0 ! DIHE -60.0184
DIHE F2 C S 0 ! DIHE 60.0184
DIHE F C S 01 ! DIHE -60.0079
DIHE F1 C S 01 ! DIHE 59.9737
DIHE F2 C S 01 ! DIHE -179.9895
DIHE F C S 02 ! DIHE 60.0079
DIHE F1 C S 02 ! DIHE 179.9895
DIHE F2 C S 02 ! DIHE -59.9737

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!=====
!DCA
!=====
RESI DCA -1.000
GROUP
ATOM N N1 -0.611 ALPHA -0.405 THOLE 1.211
ATOM C CG 0.419 ALPHA -1.042 THOLE 1.298
ATOM N1 NE -0.616 ALPHA -0.983 THOLE 1.337
ATOM C1 CG 0.419 ALPHA -1.042 THOLE 1.298
ATOM N2 N1 -0.611 ALPHA -0.405 THOLE 1.211

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BOND N C ! DIST 1.1508
BOND C N1 ! DIST 1.3125
BOND N1 C1 ! DIST 1.3125
BOND C1 N2 ! DIST 1.1508

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ANGL N C N1 ! ANGLE 175.1727
ANGL C N1 C1 ! ANGLE 118.5067
ANGL N1 C1 N2 ! ANGLE 175.1727

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DIHE N C N1 C1 ! DIHE -180.0000
DIHE C N1 C1 N2 ! DIHE -179.9999

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END

READ PARA CARD

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*=====
*. P A R A M E T E R S O F I O N I C L I Q U I D S
*.
*. Parametrization: GAAMP, scaling of epsilon_LJ
*=====
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!=====
ATOMS
!=====
MASS 1 H1 1.008000

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MASS	2	H4	1.008000	
MASS	3	H5	1.008000	
MASS	4	HC	1.008000	
MASS	6	C3	12.010000	
MASS	7	CC	12.010000	
MASS	8	CD	12.010000	
MASS	9	CG	12.010000	
MASS	10	O	16.000000	
MASS	13	N1	14.010000	
MASS	14	NA	14.010000	
MASS	15	NE	14.010000	
MASS	16	F	19.000000	
MASS	18	S6	32.060000	
MASS	60	LP	0.000000	! general lone pair
MASS	61	DRUD	0.000000	! drude particle

!=====

BONDS

!=====

!

! EMIM

!

C3	H1	335.90	1.093
C3	NA	334.70	1.456
CC	NA	438.80	1.371
CD	NA	438.80	1.371
CC	H5	356.00	1.079
CC	CD	504.00	1.371
CC	H4	350.10	1.083
CD	H4	350.10	1.083
C3	C3	303.10	1.535
C3	HC	337.30	1.092

!

! TRIF

!

C3	F	363.80	1.344
C3	S6	254.00	1.774
O	S6	541.10	1.436
O	LP	0.00	0.000

!

! DCA

!

CG	NE	509.50	1.326
CG	N1	994.70	1.143

!

! Drudes

!

C3	DRUD	500.00	0.000
CC	DRUD	500.00	0.000
CD	DRUD	500.00	0.000
CG	DRUD	500.00	0.000
O	DRUD	500.00	0.000
NA	DRUD	500.00	0.000
N1	DRUD	500.00	0.000
NE	DRUD	500.00	0.000
F	DRUD	500.00	0.000
S6	DRUD	500.00	0.000

!=====

ANGLES

!=====

!

! EMIM

!

C3	NA	CC	62.560	125.090
H1	C3	H1	39.180	109.550
H1	C3	NA	49.900	109.450
NA	CC	NA	73.650	109.330
NA	CC	H5	49.760	122.100
NA	CD	CC	72.910	109.420
NA	CD	H4	50.220	119.660

! SAME AS NA C2 NA

NA	C3	C3	65.730	112.810
CC	NA	CD	63.880	128.010
CC	NA	CC	68.940	109.900
NA	CC	CD	72.910	109.420
NA	CC	H4	50.220	119.660
CC	CD	H4	47.190	129.110
CD	NA	C3	62.560	125.090
CD	CC	H4	47.190	129.110
C3	C3	HC	46.370	110.050
H1	C3	C3	46.360	110.070
HC	C3	HC	39.430	108.350

!

! TRIF

!

C3	S6	O	41.660	108.320
F	C3	F	71.260	107.160
F	C3	S6	81.220	109.670
O	S6	O	46.660	119.730

!

! DCA

!

CG	NE	CG	66.000	140.000
NE	CG	N1	65.040	174.070

!=====

DIHEDRALS

!=====

!

! EMIM

!

X	C3	NA	X	0.000	2	0.0
X	CC	NA	X	1.700	2	180.0
X	CD	NA	X	1.700	2	180.0
X	CC	CD	X	4.000	2	180.0
X	C3	C3	X	0.156	3	0.0
CC	NA	C3	C3	0.027	1	0.0
CC	NA	C3	C3	0.232	2	180.0
CC	NA	C3	C3	0.314	3	180.0
CC	NA	C3	C3	0.033	4	180.0
CC	NA	C	C3	0.0255	6	0.0

!

! TRIF

!

F	C3	S6	O	0.144	3	0.0
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!

! DCA

!

X	CG	NE	X	0.000	2	180.0
CG	NE	CG	N1	0.000	2	180.0 ! SAME AS X C1 NE X

!=====

IMPROPERS

!=====

!

! EMIM

!

C3	CC	NA	CD	1.100	2	180.0	! USING DEFAULT VALUE
H5	NA	CC	NA	1.100	2	180.0	! USING DEFAULT VALUE
C3	CC	NA	CC	1.100	2	180.0	! USING DEFAULT VALUE
CD	H4	CC	NA	1.100	2	180.0	! USING DEFAULT VALUE
CC	H4	CD	NA	1.100	2	180.0	! USING DEFAULT VALUE

!=====

NONBONDED

!=====

!	EMIN	RMIN/2	EMIN/2	RMIN	(FOR 1-4'S)
!	(KCAL/MOL)	(A)			

!=====

!

! EMIM

!

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C3      0.00   -0.0741   1.9080      0.00   -0.0371   1.9080
NA      0.00   -0.1427   1.8240      0.00   -0.0713   1.8240
CC      0.00   -0.0602   1.9080      0.00   -0.0301   1.9080
CD      0.00   -0.0600   1.9080      0.00   -0.0300   1.9080
H1      0.00   -0.0157   1.3870      0.00   -0.00775   1.3870
H5      0.00   -0.0150   1.3590      0.00   -0.0075   1.3590
H4      0.00   -0.0150   1.4090      0.00   -0.0075   1.4090
HC      0.00   -0.0157   1.4870      0.00   -0.00775   1.4870
!
! TRIF
!
S6      0.00   -0.1000   2.0000      0.00   -0.0500   2.00
F       0.00   -0.0607   1.7500      0.00   -0.0304   1.7500
O       0.00   -0.2005   1.6612      0.00   -0.1003   1.6612
!
! dca
!
N1      0.00   -0.1584   1.8240      0.00   -0.0792   1.8240
NE      0.00   -0.1399   1.8240      0.00   -0.0699   1.8240
CG      0.00   -0.1703   1.9080      0.00   -0.0851   1.9080
!
! Wildcard for Drudes and dummy atom
!
DRUD    0.0   -0.0000   0.0000      0.00   -0.00   0.00
LP      0.0   -0.0000   0.0000      0.00   -0.00   0.00

END
RETURN

```

2 Fit parameters

The autocorrelation functions of the aqueous and ionic contributions to the collective rotational dipole moment are fitted bi- or triexponentially for each contribution x with $x = \{\text{water, cation, anion}\}$,

$$\langle \vec{M}_D(0) \cdot \vec{M}_D^x(t) \rangle \simeq \sum_{k=1}^3 a_k e^{-t/\tau_k}. \quad (1)$$

The autocorrelation function $\langle \vec{J}(0) \cdot \vec{J}(t) \rangle$ of the electric current $\vec{J}(t)$ can be modelled by damped oscillators,

$$\langle \vec{J}(0) \cdot \vec{J}(t) \rangle \simeq \sum_{k=1}^4 a_k e^{-t/\tau_k} \cos(\omega_k t + \delta_k) \quad (2)$$

where we restrict the third and fourth component to an exponential, *i.e.* $\omega_3 = \omega_4 = 0$ and $\delta_3 = \delta_4 = 0$. The components $k = 3$ and $k = 4$ are responsible for the slow relaxation components and almost invisible in $\langle \vec{J}(0) \cdot \vec{J}(t) \rangle$. Thus, they are fitted instead via the mean-squared displacement of $\mathbf{M}_J(t)$ via

$$\langle \Delta \mathbf{M}_J^2(t) \rangle \simeq mt + \sum_{k=3}^4 a_k^* \cdot (1 - e^{-t/\tau_k}) \quad (3)$$

with

$$a_k^* = \frac{a_k}{2\tau_k^2}. \quad (4)$$

A detailed description of the methods can be found in Ref. [2, 3, 4]. The respective parameters are given in the Table S1.

	c_{IL}	$\langle \vec{M}_D(0) \cdot \vec{M}_D^W(t) \rangle$						$\langle \vec{M}_D(0) \cdot \vec{M}_D^C(t) \rangle$						$\langle \vec{M}_D(0) \cdot \vec{M}_D^A(t) \rangle$					
		a_1	τ_1	a_2	τ_2	a_3	τ_3	a_1	τ_1	a_2	τ_2	a_3	τ_3	a_1	τ_1	a_2	τ_2	a_3	τ_3
C ₂ mim OTf	0.25	296	0.13	9495	10.0			3.75	0.28	17.3	33.1			3.99	0.69	138	24.8		
	0.50	445	0.58	8677	11.5			6.69	0.36	24.5	15.4			10.5	0.68	277	28.1		
	1.00	317	0.23	2490	7.2	4887	20.8	12.6	0.28	20.5	10.8	34.4	70.4	27.0	0.66	532	34.8		
	2.01	212	0.22	5211	25.4	245	357	20.7	0.33	91.8	35.3	23.5	92.1	62.5	0.91	806	41.3	115	253
	3.02	206	0.5	2505	46.6	668	160	21.5	0.23	139	63.6			88.2	0.83	635	57.7	407	173
	4.01	82	0.32	151	13.7	1243	211	27.2	0.33	206	137			91.7	0.43	144	14.8	811	246
	5.24							37.4	0.64	49.5	20.7	239	636	175	0.97	370	29.3	303	359
C ₂ mim N(CN) ₂	0.25	342	0.26	9856	10.1			3.81	0.31	8.45	16.2			5.13	0.62	1.18	10.3		
	0.50	361	0.27	8950	10.8			6.23	0.22	20.2	13.8	12.6	69.0	7.91	0.40	5.65	6.33		
	0.99	489	0.48	7878	13.5			9.84	0.17	21.3	5.89	29.9	38.0	11.5	0.26	13.9	4.15		
	1.95	443	0.64	5629	19.0			14.3	0.13	13.1	1.68	83.8	39.5	18.5	0.21	30.0	4.37	12.1	52.4
	2.90	286	0.51	3884	27.0			20.4	0.16	22.2	4.6	104	75.1	24.1	0.19	41.2	6.33	17.1	52.3
	3.83	156	0.26	182	5.7	2072	44.4	19.2	0.11	22.4	2.7	118	68.4	27.8	0.17	29.9	4.99	48.3	48.8
	4.91	72	0.26	119	9.8	652	72.1	23.6	0.22	23.4	9.02	137	190	26.3	0.14	26.3	6.10	49.4	88.9
	6.11							26.9	0.44	37.4	21.4	126	491	29.4	0.18	10.8	2.90	7.66	76.8
	c_{IL}	$\langle \Delta \mathbf{M}_J^2(t) \rangle$					$\langle \vec{J}(0) \cdot \vec{J}(t) \rangle$												
		m	a_1^*	τ_1	a_2^*	τ_2	a_1	τ_1	ω_1	δ_1	a_2	τ_2	ω_2	δ_2					
C ₂ mim OTf	0.25	45.9	40.6	0.53	289	48.0	783	0.071	17.6	-1.04	1783	0.28	3.26	0.68					
	0.50	76.9	79.5	0.48	643	54.4	1257	0.051	20.4	-1.59	6074	0.22	2.52	0.95					
	1.00	110	169	0.46	882	43.4	4120	0.049	15.0	-1.52	20197	0.22	1.46	1.23					
	2.01	118	333	0.41	1771	52.7	5269	0.069	21.0	-1.30	18461	0.24	3.74	0.80					
	3.02	78.5	441	0.39	907	13.6	17939	0.068	15.3	-1.00	17389	0.27	4.23	0.85					
	4.01	34.7	502	0.36	897	12.1	17901	0.075	19.5	-1.33	52718	0.17	3.57	1.08					
	5.24	14.6	452	0.36	761	12.3	28265	0.116	20.2	-0.89	51469	0.13	4.13	1.23					
C ₂ mim N(CN) ₂	0.25	53.2	40.6	0.47	456	79.8	1129	0.043	30.8	-1.15	3797	0.20	2.92	0.94					
	0.50	84.9	83.6	0.46	1060	68.4	3143	0.041	25.4	-1.10	5478	0.21	3.99	0.76					
	0.99	143	141	0.38	863	11.7	2858	0.044	37.9	-1.54	21399	0.17	2.87	1.04					
	1.95	179	319	0.40	1740	30.2	23022	0.048	19.8	-0.94	13230	0.26	4.01	0.90					
	2.90	206	463	0.39	789	17.9	34714	0.050	20.3	-0.91	21226	0.24	3.88	1.01					
	3.83	154	594	0.39	1359	22.8	43712	0.061	21.3	-0.81	39956	0.20	2.79	1.23					
	4.91	90.7	625	0.44	1142	18.1	48745	0.075	21.3	-0.63	29366	0.21	3.46	1.23					
	6.11	39.0	472	0.33	942	16.0	50408	0.110	23.0	-0.47	31071	0.16	4.32	1.23					

Table S1: Fit parameters to obtain the dielectric spectra of [C₂mim][OTf] and [C₂mim][N(CN)₂] mixtures with water.

References

- [1] B. R. Brooks, C. L. Brooks III, A. D. MacKerell Jr., L. Nilsson, R. J. Petrella, B. Roux, Y. Won, G. Archontis, C. Bartels, S. Boresch, A. Caffisch, L. Caves, Q. Cui, A. R. Dinner, M. Feig, S. Fischer, J. Gao, M. Hodoscek, W. Im, K. Kuczera, T. Lazaridis, J. Ma, V. Ovchinnikov, E. Paci, R. W. Pastor, C. B. Post, J. Z. Pu, M. Schaefer, B. Tidor, R. M. Venable, H. L. Woodcock, X. Wu, W. Yang, D. M. York, and M. Karplus. “CHARMM: The biomolecular simulation program.” *J. Comput. Chem.*, **30**(2009), 1545.
- [2] C. Schröder and O. Steinhauser. “On the dielectric conductivity of molecular ionic liquids.” *J. Chem. Phys.*, **131**(2009), 114504.
- [3] C. Schröder and O. Steinhauser. “Using fit functions in computational dielectric spectroscopy.” *J. Chem. Phys.*, **132**(2010), 244109.
- [4] C. Schröder. “Collective translational motions and cage relaxations in molecular ionic liquids.” *J. Chem. Phys.*, **135**(2011), 024502.