SUPPORTING INFORMATION FOR

Structural, Thermodynamic and Transport Properties of Aqueous Reline and Ethaline Solutions from Molecular Dynamics Simulations

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The Generalized Amber Force Field parameters and the partial charges for aqueous reline and ethaline solutions are reported. The number of molecules and the box lengths for all simulated system are presented. Raw data for the calculated densities, thermal expansivities, viscosities, self-diffusivities, and ionic conductivities of reline and ethaline solutions are presented as a function of the mass fraction of water and the temperature. Viscosities, diffusivities and conductivities for neat reline and ethaline are shown, where MD simulations are compared with available experimental results. All water-related radial distribution functions of reline and ethaline at 303.15 K are shown. The Supporting Information also provides radial distribution functions of water-reline and water-ethaline mixtures of a mass

fraction of water of 40% at various temperatures. The fraction of hydrogen bonds for both reline and ethaline are provided.

1.Force field parameters for reline-water mixtures

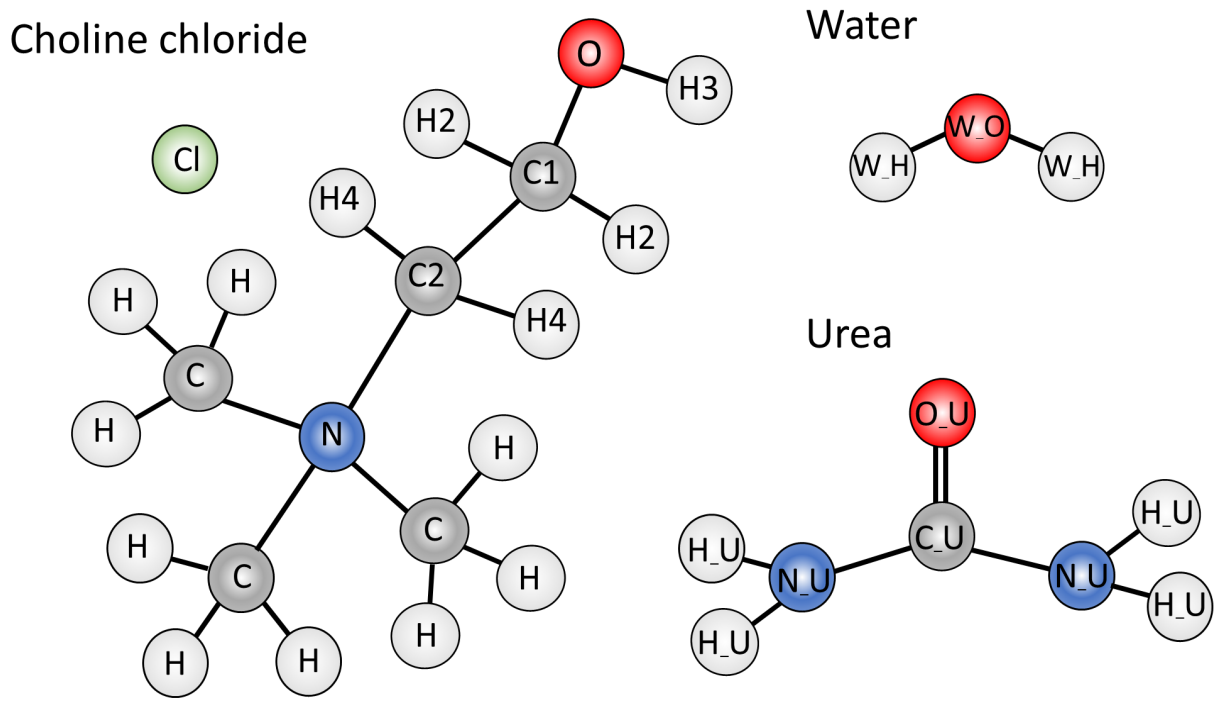

Figure S1: Molecular structure of choline chloirde, urea and water, and atom labels.

Table S1: Partial charges and masses for all atomic species. Choline chloride and urea parameters are taken by Perkins et al.[1] Water molecules are modeled using the the SPC/E water model.[2] See Figure S1 for the atom labels.

No	Name	$q/[e]$	$m/$[u]
1	C	-0.10736	12.0107
2	C1	0.12008	12.0107
3	C2	-0.02576	12.0107
4	C_U	1.0401	12.0107
5	Cl	-0.8	35.453
6	H	0.09544	1.00794
7	H2	0.0408	1.00794
8	H3	0.3636	1.00794
9	H4	0.08928	1.00794
10	H_U	0.4167	1.00794
11	N	0.04016	14.0067
12	N_U	-1.0246	14.0067
13	O	-0.49512	15.9994
14	O_U	-0.6577	15.9994
15	W_H	0.424	1.00794
16	W_O	-0.848	15.9994

Table S2: Lennard-Jones parameters of choline chloride and urea molecules using GAFF.[3] Lennard-Jones parameters for water molecules using the the SPC/E model.[2] See Figure S1 for the atom labels.

No	Name	ϵ/k_{B}/[K]	σ/[Å]
1	C	55.052	3.3996
2	C1	55.052	3.3996
3	C2	55.052	3.3996
4	C_U	43.277	3.3996
5	Cl	50.322	4.401
6	H	7.901	1.9599
7	H2	7.901	2.4713
8	H3	0.503	0.1
9	H4	7.901	1.9599
10	H_U	7.901	1.069
11	N	85.547	3.2499
12	N_U	85.547	3.25
13	O	105.877	3.0664
14	O_U	105.676	2.96
15	W_H	0	0
16	W_O	78.200	3.1656

Table S3: Bond-stretching parameters for water-reline mixtures. Choline choride and urea molecules are modelled using GAFF.[3] Water molecules are modelled using the the SPC/E model.[2] See Figure S1 for the atom labels. The bond energy is calculated as: $E_{\mathrm{Bond}}(r)=K_{\mathrm{r}}(r\text{-}r_0)^2$.

No	Name	$K_{\mathrm{r}}/k_{\mathrm{B}}$/[K Å^{-2}]	r_0/[Å]
1	C-H	170440.5	1.09
2	C2-N	147745.3	1.5
3	C1-O	158061.3	1.43
4	C1-H2	169031.5	1.09
5	H_U-N_U	206420.7	1.01
6	C-N	147745.3	1.5
7	C1-C2	152525.9	1.54
8	W_H-W_O	226448.9	1
9	C2-H4	170440.5	1.09
10	C_U-N_U	240639.7	1.35
11	H3-O	185990.0	0.97
12	C_U-O_U	326086.4	1.21

Table S4: Angle bending parameters for water-reline mixtures. Choline choride and urea molecules are modelled using GAFF.[3] Water molecules are modelled using the SPC/E model.[2] See Figure S1 for the atom labels. The bond energy is calculated as: $E_{\mathrm{Angle}}(\theta)=K_\theta(\theta\text{-}\theta_0)^2$.

No	Name	K_θ/k_B/[K rad^{-2}]	θ_0
1	C-N-C2	31622.3	110.6
2	H2-C1-H2	19716.1	109.6
3	C2-C1-O	34078.0	109.4
4	H-C-N	24667.8	107.9
5	C2-C1-H2	23329.3	110.1
6	W_H-W_O-W_H	27677.1	109.47
7	C_U-N_U-H_U	24763.4	118.5
8	H2-C1-O	25649.1	109.9
9	C1-O-H3	23696.6	108.2
10	H4-C2-N	24667.8	107.9
11	C1-C2-N	32432.5	114.3
12	H_U-N_U-H_U	19992.9	117.9
13	N_U-C_U-N_U	37640.8	113.4
14	H-C-H	19645.7	110.7
15	N_U-C_U-O_U	38159.1	122
16	C-N-C	31622.3	110.6
17	H4-C2-H4	19645.7	110.7
18	C1-C2-H4	23158.2	111.7

Table S5: Dihedral torsion parameters for water-reline mixtures. Choline choride and urea molecules are modelled using GAFF.[3] See Figure S1 for the atom labels. The torsion energy for charmm style is calculated as: $E_{\mathrm{Dihedral}}(\phi)=\frac{V_n}{2}[1+\cos(n\phi\text{-}\gamma)]$. The torsion energy for opls style is calculated as: $E_{\mathrm{Dihedral}}(\phi)=\frac{K_1}{2}[1+\cos(\phi)]+\frac{K_2}{2}[1\text{-}\cos(2\phi)]+\frac{K_3}{2}[1+\cos(3\phi)]$.

No	Name	$V_{\mathrm{n}}/2/k_{\mathrm{B}}$/[K]	n	γ	Style
1	H4-C2-N-C	78.50	3	0	charmm
2	C1-C2-N-C	78.50	3	0	charmm
3	O-C1-C2-H4	78.50	3	0	charmm
4	H-C-N-C2	78.50	3	0	charmm
5	H2-C1-C2-H4	78.50	3	0	charmm
6	N_U-C_U-N_U-H_U	1258.05	2	180	charmm
7	H2-C1-O-H3	84.04	3	0	charmm
8	H2-C1-C2-N	78.50	3	0	charmm
9	H-C-N-C	78.50	3	0	charmm
10	O-C1-C2-N	78.50	3	0	charmm
No	Name	K_1/k_{B}/[K]	K_2/k_{B}/[K]	K_3/k_{B}/[K]	Style
11	O_U-C_U-N_U-H_U	2012.88	2516.10	0	opls
12	C2-C1-O-H3	251.61	0	161.03	opls

Table S6: Improper torsion parameters for water-reline mixtures. Choline choride and urea molecules are modelled using GAFF.[3] See Figure S1 for the atom labels. The torsion energy is calculated as: $E_{\mathrm{Dihedral}}(\phi)=\frac{V_n}{2}[1+\cos(n\phi\text{-}\gamma)]$.

No	Name	$V_{\mathrm{n}}/2/k_{\mathrm{B}}/[\mathrm{K}]$	n	γ
1	C_U-H_U-N_U-H_U	553.54	180	2
2	N_U-N_U-C_U-O_U	5283.81	180	2

2. Force field parameters for ethaline-water mixtures

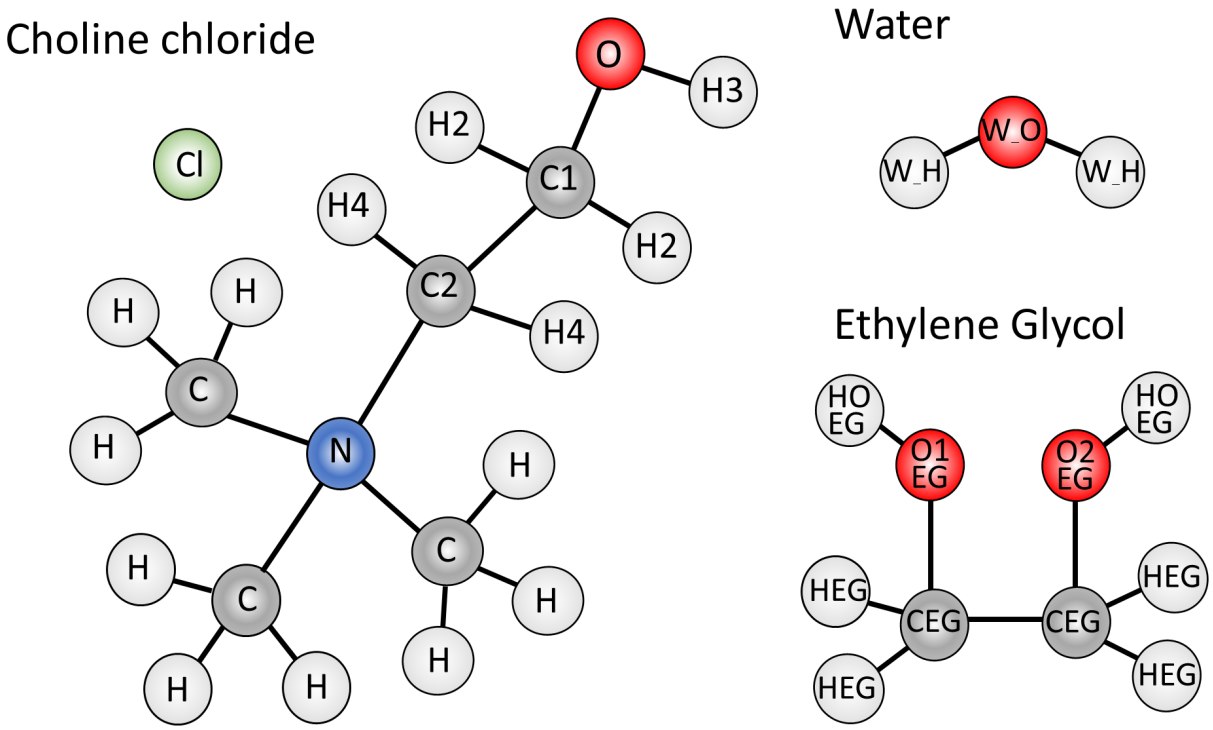

Figure S2: Molecular structure of choline chloirde, ethylene glycol and water, and atom labels.

Table S7: Partial charges and masses for all atomic species. Choline chloride and ethylene glycol parameters are taken by Perkins et al.[1] Water molecules are modeled using the the SPC/E water model.[2] See Figure S2 for the atom labels.

No	Name	$q/[e]$	$m/$[u]
1	C	0.1094	3.3996
2	C1	0.1094	3.3996
3	C2	0.1094	3.3996
4	CEG	0.1094	3.3996
5	Cl	0.1	4.401
6	H	0.0157	1.9599
7	H2	0.0157	2.4713
8	H3	0.001	0.1
9	H4	0.0157	1.9599
10	HEG	0.0157	2.4713
11	HOEG	0.001	0.1
12	N	0.17	3.2499
13	O	0.2104	3.0664
14	O1EG	0.2104	3.0664
15	O2EG	0.2104	3.0664
16	W_H	0	0
17	W_O	0.1554	3.1656

Table S8: Lennard-Jones parameters of choline chloride and ethylene glycol molecules using GAFF.[3] Lennard-Jones parameters for water molecules using the the SPC/E model.[2] See Figure S2 for the atom labels.

No	Name	ϵ/k_B/[K]	σ/[Å]
1	C	55.052	3.3996
2	C1	55.052	3.3996
3	C2	55.052	3.3996
4	CEG	55.052	3.3996
5	Cl	50.322	4.401
6	H	7.901	1.9599
7	H2	7.901	2.4713
8	H3	0.503	0.1
9	H4	7.901	1.9599
10	HEG	7.901	2.4713
11	HOEG	0.503	0.1
12	N	85.547	3.2499
13	O	105.877	3.0664
14	O1EG	105.877	3.0664
15	O2EG	105.877	3.0664
16	W_H	0	0
17	W_O	78.200	3.1656

Table S9: Bond-stretching parameters for ethaline-reline mixtures. Choline choride and ethylene glycol molecules are modelled using GAFF.[3] Water molecules are modelled using the the SPC/E model.[2] See Figure S2 for the atom labels. The bond energy is calculated as: $E_{\mathrm{Bond}}(r)=K_{\mathrm{r}}(r\text{-}r_0)^2$.

No	Name	$K_{\mathrm{r}}/k_{\mathrm{B}}$/[K Å^{-2}]	r_0/[Å]
1	C-H	170440.5	1.091
2	C2-N	147745.3	1.499
3	C1-O	158061.3	1.426
4	C1-H2	169031.5	1.093
5	CEG-CEG	152525.9	1.535
6	C1-C2	152525.9	1.535
7	C-N	147745.3	1.499
8	W_H-W_O	226448.9	1
9	CEG-HEG	169031.5	1.093
10	CEG-O1EG	158061.3	1.426
11	CEG-O2EG	158061.3	1.426
12	HOEG-O1EG	185990.0	0.974
13	HOEG-O2EG	185990.0	0.974
14	C2-H4	170440.5	1.091
15	H3-O	185990.0	0.974

Table S10: Angle bending parameters for water-ethaline mixtures. Choline choride and ethylene glycol molecules are modelled using GAFF.[3] Water molecules are modelled using the the SPC/E model.[2] See Figure S2 for the atom labels. The angle energy is calculated as: $E_{\mathrm{Angle}}(\theta)=K_\theta(\theta\text{-}\theta_0)^2$.

No	Name	K_θ/k_B/[K rad^{-2}]	θ_0
1	CEG-O1EG-HOEG	23701.6	108.16
2	C-N-C2	31602.2	110.64
3	HEG-CEG-O1EG	25664.2	109.88
4	HEG-CEG-O2EG	25664.2	109.88
5	H2-C1-H2	19726.2	109.55
6	C2-C1-O	34068.0	109.43
7	H-C-N	24657.8	107.91
8	CEG-CEG-O1EG	34068.0	109.43
9	CEG-CEG-HEG	23349.4	110.07
10	CEG-CEG-O2EG	34068.0	109.43
11	C2-C1-H2	23349.4	110.07
12	W_H-W_O-W_H	27677.1	109.47
13	H2-C1-O	25664.2	109.88
14	C1-O-H3	23701.6	108.16
15	CEG-O2EG-HOEG	23701.6	108.16
16	HEG-CEG-HEG	19726.2	109.55
17	C1-C2-N	33212.5	108.93
18	H4-C2-N	24657.8	107.91
19	H-C-H	19625.6	110.74
20	C-N-C	31602.2	110.64
21	C1-C2-H4	23148.1	111.74
22	H4-C2-H4	19625.6	110.74

Table S11: Dihedral torsion parameters for water-ethaline mixtures. Choline choride and ethylene glycol molecules are modelled using GAFF.[3] See Figure S2 for the atom labels. The torsion energy for charmm style is calculated as: $E_{\mathrm{Dihedral}}(\phi)=\frac{V_n}{2}[1+\cos(n\phi-\gamma)]$. The torsion energy for opls style is calculated as: $E_{\mathrm{Dihedral}}(\phi)=\frac{K_1}{2}[1+\cos(\phi)]+\frac{K_2}{2}[1-\cos(2\phi)]+\frac{K_3}{2}[1+\cos(3\phi)]$. The torsion energy for multi/harmonic style is calculated as: $E_{\mathrm{Dihedral}}(\phi)=\sum K_n\cos^{n-1}(\phi)$.

No	Name	$V_n/2/k_B$/[K]	n	γ	Style
1	HEG-CEG-O1EG-HOEG	84.04	3	0	charmm
2	C1-C2-N-C	78.50	3	0	charmm
3	H4-C2-N-C	78.50	3	0	charmm
4	O-C1-C2-H4	78.50	3	0	charmm
5	HEG-CEG-CEG-HEG	78.50	3	0	charmm
6	H-C-N-C2	78.50	3	0	charmm
7	H2-C1-C2-H4	78.50	3	0	charmm
8	H2-C1-O-H3	84.04	3	0	charmm
9	H2-C1-C2-N	78.50	3	0	charmm
10	HEG-CEG-O2EG-HOEG	84.04	3	0	charmm
11	H-C-N-C	78.50	3	0	charmm
12	HEG-CEG-CEG-O1EG	125.80	1	0	charmm
13	O-C1-C2-N	78.50	3	0	charmm
14	HEG-CEG-CEG-O2EG	125.80	1	0	charmm
No	Name	K_1/k_B/[K]	K_2/k_B/[K]	K_3/k_B/[K]	Style
15	CEG-CEG-O2EG-HOEG	251.61	0	161.03	opls
16	C2-C1-O-H3	251.61	0	161.03	opls
17	CEG-CEG-O1EG-HOEG	251.61	0	161.03	opls
No	Name	K_1/k_B/[K]	K_2/k_B/[K]	K_3/k_B/[K]	Style
18	O1EG-CEG-CEG-O2EG	72.46	-217.39	1182.57	multi/harmonic
		K_4/k_B/[K]	K_5/k_B/[K]		
		289.85	0		

Results

Table S12: The number of molecules of each constituting component in the simulations of aqueous reline and ethaline solutions.

Mass Fraction of Water/[%]	**Reline Mixtures**			**Ethaline Mixtures**		
	Number of Molecules			**Number of Molecules**		
	Urea	**ChCl**	**Water**	**EG**	**ChCl**	**Water**
0	100	50	0	100	50	0
2	100	50	15	100	50	15
5	100	50	38	100	50	38
20	100	50	180	100	50	183
40	100	50	481	100	50	488
60	100	50	1081	100	50	1098
80	100	50	2884	100	50	2930

Table S13: Average box lengths of aqueous reline and ethaline solutions as a function of temperature and water content at 1 atm.

Box Length of Reline/[Å]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	26.10	26.31	26.61	28.44	31.74	36.81	47.02	28.88
313.15	26.18	26.36	26.68	28.51	31.79	36.89	47.12	28.94
333.15	26.20	26.45	26.77	28.61	31.93	37.06	47.32	29.07
343.15	26.28	26.51	26.82	28.68	32.03	37.14	47.45	29.14
363.15	26.34	26.59	26.92	28.77	32.16	37.30	47.69	29.30
Box Length of Ethylene/[Å]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	26.97	27.18	27.47	29.16	32.32	37.33	47.47	28.88
313.15	27.01	27.22	27.47	29.22	32.40	37.40	47.55	28.94
333.15	27.16	27.31	27.62	29.33	32.54	37.58	47.78	29.07
343.15	27.25	27.40	27.67	29.38	32.63	37.67	47.90	29.14
363.15	27.34	27.51	27.81	29.54	32.76	37.87	48.16	29.30

Table S14: Densities of aqueous reline solutions as a function of the mass fraction of water and temperature at 1 atm. MD results are compared with experiments by Yadav et al.[4]

Computed Densities/[kg m^{-3}]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	1211.3	1206.9	1201.1	1171.0	1125.3	1079.9	1036.8	993.3
313.15	1206.9	1201.6	1195.1	1164.2	1118.4	1073.1	1030.8	987.7
333.15	1195.2	1189.4	1183.8	1151.2	1104.6	1059.1	1016.8	974.2
343.15	1188.0	1182.0	1177.1	1143.9	1096.9	1051.9	1009.6	967.9
363.15	1175.4	1170.5	1163.5	1130.4	1082.3	1036.1	993.7	951.3
Experimental Densities[4]/[kg m^{-3}]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	1194.5	1190.0	1183.3	1151.0	1109.9	1070.7	1032.8	995.4
313.15	1188.7	1184.3	1177.8	1145.9	1105.0	1066.0	1028.4	992.2
333.15	1177.3	1173.0	1166.6	1135.0	1094.4	1055.6	1018.5	983.3
343.15	1170.8	1166.5	1160.1	1128.7	1088.3	1049.7	1012.9	977.8
363.15	1155.4	1151.2	1144.9	1113.9	1074.1	1035.9	999.6	965.0

Table S15: Densities of aqueous ethaline solutions as a function of the mass fraction of water and temperature at 1 atm. MD results are compared with experiments by Yadav et al.[5]

Computed Densities/[kg m^{-3}]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	1116.9	1114.7	1116.2	1105.3	1080.6	1053.0	1024.5	993.3
313.15	1108.6	1108.2	1107.8	1098.6	1073.4	1046.2	1018.3	987.7
333.15	1096.2	1094.8	1094.4	1084.1	1058.9	1032.0	1004.4	974.2
343.15	1089.3	1088.1	1087.1	1076.3	1051.8	1024.4	997.1	967.9
363.15	1074.9	1074.6	1073.7	1061.8	1035.7	1008.5	981.1	951.3
Experimental Densities[5]/[kg m^{-3}]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	1111.4	1109.8	1107.3	1092.9	1069.7	1044.3	1018.8	995.7
313.15	1105.7	1104.1	1101.6	1087.3	1064.4	1039.5	1014.7	992.2
333.15	1094.7	1093.1	1090.6	1076.3	1053.9	1029.4	1005.2	983.2
343.15	1088.7	1087.1	1084.6	1070.4	1048.0	1023.7	999.6	977.8
363.15	1074.4	1072.8	1070.4	1056.4	1034.3	1010.2	986.4	965.0

Table S16: Thermal expansion coefficients of reline-water mixtures at various mass fractions of water at 1 atm.. Experimental data by Yadav et al.[4]

Thermal Expansivity/[K^{-1}]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
MD Simulations	6.2	6.00	6.29	6.60	6.88	7.02	7.05	6.50
Experiments[4]	5.5	5.48	5.49	5.50	5.46	5.37	5.25	5.08

Table S17: Thermal expansion coefficients of ethaline-water mixtures at various mass fractions of water at 1 atm. Experimental data by Yadav et al.[5]

Thermal Expansivity/[K^{-1}]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
MD Simulations	6.2	6.00	6.29	6.60	6.88	7.02	7.05	6.50
Experiments[5]	5.5	5.48	5.49	5.50	5.46	5.37	5.25	5.08

Table S18: Viscosities of aqueous reline solutions as a function of the mass fraction of water and temperature at 1 atm. MD results are compared with experiments by Yadav et al.[4]

Computed Viscosities/[mPa s]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	511±25	226±12	87±3	9±2	2.6±0.3	1.5±0.1	1±0.1	0.65±0.02
313.15	243±14	105±4	55±2	6±1	2.3±0.3	1.2±0.2	0.8±0.1	0.53±0.03
333.15	62±1	39±2	23±2	4.1±0.7	1.6±0.2	0.8±0.1	0.71±0.07	0.42±0.02
343.15	37±2	25±2	14±2	3.6±0.3	1.5±0.2	0.77±0.09	0.6±0.1	0.34±0.02
363.15	20±1	13±1	8±1	2.4±0.2	1.1±0.1	0.67±0.07	0.46±0.05	0.30±0.01
Experimental Viscosities[4]/[mPa s]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	527.28	216.72	88.04	9.15	2.76	1.46	1.05	0.80
313.15	238.08	112.60	52.06	6.92	2.24	1.21	0.87	0.67
333.15	68.65	40.33	22.41	4.20	1.57	0.89	0.63	0.48
343.15	41.96	27.51	16.12	3.43	1.35	0.77	0.56	0.42
363.15	19.95	14.36	9.20	2.44	1.03	0.63	0.47	0.38

Table S19: Viscosities of aqueous ethaline solutions as a function of the mass fraction of water and temperature at 1 atm.

Computed Viscosities/[mPa s]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	39±4	32±2	26±2	12±2	4.9±0.3	1.9±0.2	0.92±0.06	0.65±0.02
313.15	30±1	27±2	22±2	9±1	2.9±0.3	1.42±0.07	0.74±0.08	0.53±0.03
333.15	13±1	12±1	10±1	5.1±0.7	1.9±0.2	1.06±0.07	0.65±0.05	0.42±0.02
343.15	10.8±0.7	10.2±0.9	9±1	3.92±0.04	1.6±0.1	0.89±0.09	0.6±0.1	0.34±0.02
363.15	7.7±0.4	6.6±0.6	6.0±0.8	2.5±0.3	1.2±0.1	0.67±0.09	0.45±0.08	0.30±0.01

Table S20: Finite size-corrected self-diffusion coefficients of urea, choline cation and chloride anion in reline-water mixtures as a function of the mass fraction of water and temperature at 1 atm.

Size-corrected Self-diffusion Coefficients/[10^{-11} m^2 s^{-1}]						
Mass Fraction of Water/[%]						
	0			**2**		
T/[K]	**Urea**	**Ch**	**Cl**	**Urea**	**Ch**	**Cl**
303.15	0.63±0.04	0.37±0.02	0.47±0.03	1.3±0.2	0.8±0.1	1.0±0.1
313.15	1.10±0.07	0.62±0.04	0.82±0.06	2.3±0.1	1.4±0.1	1.9±0.1
333.15	4.2±0.3	2.5±0.1	3.2±0.2	7.3±0.3	4.4±0.2	6.2±0.3
343.15	7.2±0.4	4.2±0.2	5.6±0.4	9.8±0.3	5.9±0.3	8.4±0.2
363.15	13.6±0.7	8.2±0.5	10.8±0.7	18.9±0.5	11.5±0.2	15.7±0.5
	5			**20**		
T/[K]	**Urea**	**Ch**	**Cl**	**Urea**	**Ch**	**Cl**
303.15	2.8±0.2	1.7±0.1	2.6±0.2	17.8±0.5	12.3±0.5	22±1
313.15	4.34±0.2	2.55±0.1	4.03±0.3	26±1	18.4±0.3	31±1
333.15	10.72±0.3	6.60±0.2	10±0.2	46±2	31±2	52±2
343.15	15.7±0.3	10.1±0.3	14.8±0.2	55±2	38±1	64±2
363.15	28.1±0.7	18.1±0.6	26.6±0.6	83±2	57±1	96±2
	40			**60**		
T/[K]	**Urea**	**Ch**	**Cl**	**Urea**	**Ch**	**Cl**
303.15	55±2	41±2	75±2	103±3	76±2	140±6
313.15	71±2	52±2	93±2	128±1	97±2	172±3
333.15	109±3	81±2	139±3	179±5	139±5	235±11
343.15	127±3	94±2	163±6	209±4	159±2	280±9
363.15	173±5	125±4	217±4	285±6	212±7	363±14

	80			**100**
T/[K]	**Urea**	**Ch**	**Cl**	**Water**
303.15	154±3	117±4	207±7	314±5
313.15	187±3	144±4	251±5	380±8
333.15	252±2	190±3	343±4	521±4
343.15	304±3	230±7	397±13	608±9
363.15	389±6	294±7	537±13	778±9

Table S21: Finite size-corrected self-diffusion coefficients of ethylene glycol, choline cation and chloride anion in ethaline-water mixtures as a function of the mass fraction of water and temperature at 1 atm.

Size-corrected Self-diffusion Coefficients/[10^{-11} m^2 s^{-1}]						
Mass Fraction of Water/[%]						
	0			**2**		
T/[K]	**EG**	**Ch**	**Cl**	**EG**	**Ch**	**Cl**
303.15	9.2±0.3	2.5±0.1	3.5±0.1	12±2	3.4±0.9	5±1
313.15	13.8±0.8	3.8±0.2	5.2±0.2	16.3±0.4	4.5±0.1	6.8±0.2
333.15	32±1	9.1±0.2	12.5±0.5	34±2	9.6±0.4	14.1±0.6
343.15	36±1	10.4±0.3	13.9±0.5	38±2	11.2±0.5	16.1±0.8
363.15	61±2	17.6±0.6	23.3±0.8	65±2	19.4±0.9	28±1
	5			**20**		
T/[K]	**EG**	**Ch**	**Cl**	**EG**	**Ch**	**Cl**
303.15	13.5±0.5	3.9±0.2	6.0±0.3	24±2	8.3±0.5	13.4±0.4
313.15	18±1	5.2±0.4	7.7±0.5	36.9±1	11.9±0.4	19.7±0.5
333.15	42±1	12.1±0.3	18.4±0.5	66±2	22.7±0.8	36±1
343.15	45±1	13.4±0.5	20.0±0.7	84±4	29.8±0.8	47±1
363.15	77±3	23.4±0.8	34.0±0.9	137±6	47±2	73±3
	40			**60**		
T/[K]	**EG**	**Ch**	**Cl**	**EG**	**Ch**	**Cl**
303.15	71±3	25.3±0.7	42±1	145±3	57±1	97±4
313.15	97±5	36±1	59±4	173±6	70±3	118±5
333.15	149±3	58±2	90±2	262±8	106±3	172±4
343.15	184±5	71±2	115±4	317±7	127±5	202±4
363.15	268±5	98±2	161±4	437±9	176±4	280±7

	80			**100**
T/[K]	**EG**	**Ch**	**Cl**	**Water**
303.15	228±5	93±2	152±3	314±5
313.15	288±11	119±4	191±4	380±8
333.15	403±8	162±3	263±7	521±4
343.15	496±7	189±5	308±17	608±9
363.15	644±13	264±7	401±10	778±9

Table S22: Ionic conductivities of aqueous reline solutions as a function of the mass fraction of water and temperature at 1 atm.

Computed Ionic Conductivities/[S m^{-1}]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	0.09	0.19	0.45	2.92	7.10	8.50	6.12	0
313.15	0.15	0.35	0.66	4.09	8.54	10.20	7.18	0
333.15	0.56	1.12	1.51	6.38	12.06	13.12	8.98	0
343.15	0.94	1.51	2.24	7.50	13.54	14.89	10.17	0
363.15	1.70	2.84	3.76	10.55	16.85	18.14	12.54	0

Table S23: Ionic conductivities of aqueous ethaline solutions as a function of the mass fraction of water and temperature at 1 atm.

Computed Ionic Conductivities/[S m^{-1}]								
T/[K]	**Mass Fraction of Water/[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
303.15	0.61	0.82	0.94	1.72	3.91	5.79	4.50	0
313.15	0.86	1.07	1.18	2.41	5.31	6.84	5.49	0
333.15	1.92	2.29	2.58	4.14	7.66	9.36	6.98	0
343.15	2.09	2.61	2.73	5.23	9.25	10.67	7.85	0
363.15	3.28	4.43	4.38	7.61	12.05	13.74	9.75	0

Table 24: Coordination number of aqueous reline and ethaline solutions as a function of mass fraction of water.

Coordination Number of Aquoues Reline Solutions								
T/[K]	**Water Mass Concentration /[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
Urea-Urea	3.35	3.42	3.43	3.55	3.89	4.32	4.89	N/A
Urea-Choline	3.45	3.46	3.56	3.85	3.99	4.21	4.66	N/A
Urea-Chloride	2.83	2.72	2.66	2.56	2.86	2.98	3.00	N/A
Urea-Water	N/A	2.62	2.39	2.39	2.57	2.68	2.87	N/A
Choline-Choline	0.86	0.90	0.96	1.18	1.51	2.70	4.85	N/A
Choline-Chloride	4.41	4.44	4.46	4.58	4.61	4.62	5.11	N/A
Choline-Water	N/A	3.93	3.92	3.91	3.53	3.18	2.97	N/A
Chloiride-Chloride	1.50	1.60	1.74	2.20	1.99	1.83	1.70	N/A
Chloride-Water	N/A	3.50	3.29	2.28	1.78	1.49	1.34	N/A
Water-Water	N/A	2.87	3.05	2.45	1.86	1.57	1.36	1.28
Coordination Number of Aquoues Ethaline Solutions								
T/[K]	**Water Mass Concentration /[%]**							
	0	**2**	**5**	**20**	**40**	**60**	**80**	**100**
EG-EG	3.42	3.53	3.65	3.97	3.90	3.83	3.80	N/A
EG-Choline	3.43	3.44	3.45	3.58	3.60	3.32	3.31	N/A
EG-Chloride	2.53	2.46	2.27	1.80	1.59	1.41	1.26	N/A
EG-Water	N/A	1.76	1.79	1.83	1.79	1.67	1.57	N/A
Choline-Choline	1.20	1.21	1.36	1.62	1.75	2.96	4.88	N/A
Choline-Chloride	4.71	4.76	4.80	4.93	4.81	4.79	4.39	N/A
Choline-Water	N/A	4.36	4.33	4.12	3.72	3.41	3.13	N/A
Chloiride-Chloride	0.86	1.11	1.36	1.68	1.67	1.55	1.23	N/A
Chloride-Water	N/A	4.37	3.89	2.99	2.26	1.86	1.60	N/A
Water-Water	N/A	2.87	3.05	2.45	1.86	1.57	1.36	1.28

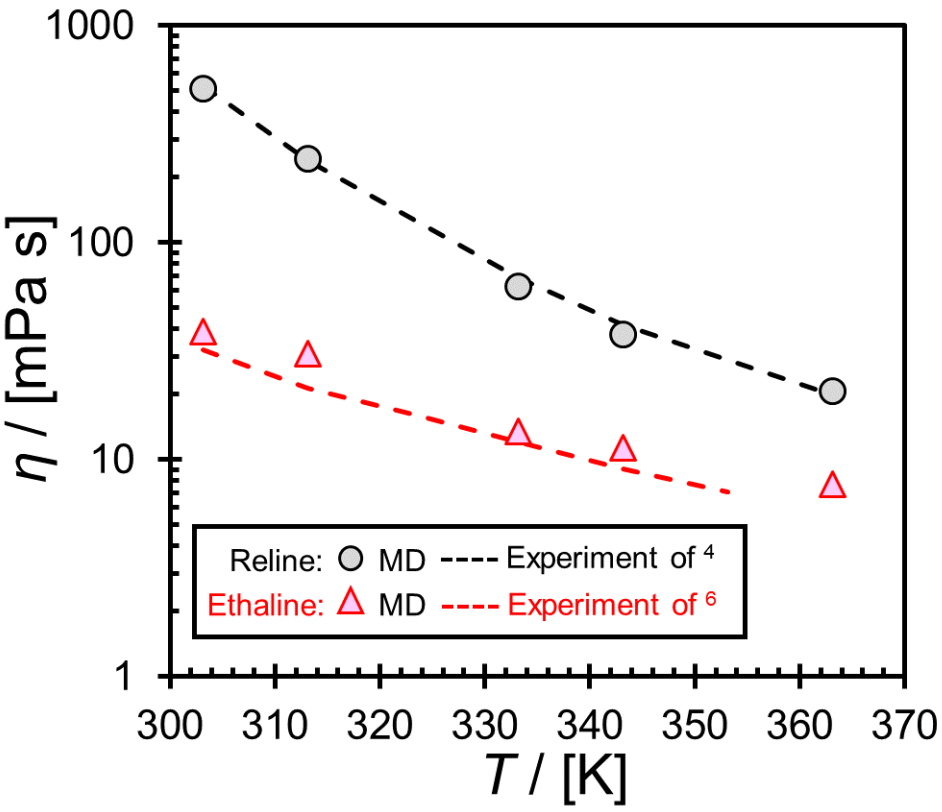

Figure S3: Viscosities of neat reline and neat ethaline as a function of temperature. Black-gray circles represent simulation results for reline. The black dashed line represents experiment results for reline.[4] Red-pink triangles represent simulation results for ethaline. Red dashed line is experimental data for ethaline.[6]

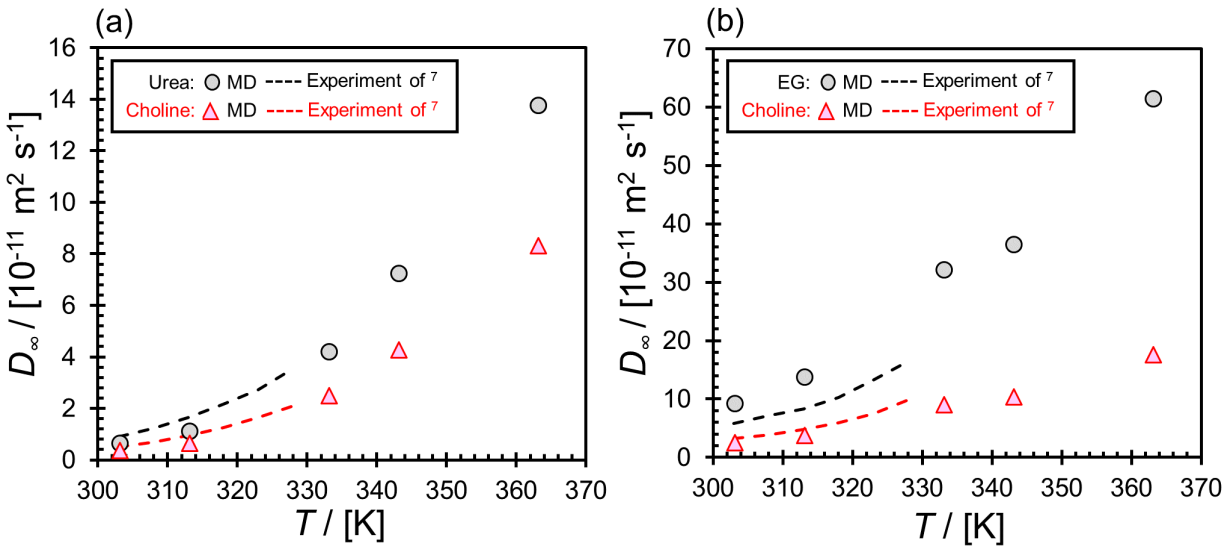

Figure S4: Finite-size corrected self-diffusion coefficients of (a) neat reline, and (b) neat ethaline as a function of temperature. Black-gray circles represent simulation results for urea in reline solution and ethylene glycol in ethaline solution. The black dashed line indicates experiment results.[7] Red-pink triangles represent simulation results for choline cation in reline and ethaline solutions.The red dashed line indicates experiment results.[7]

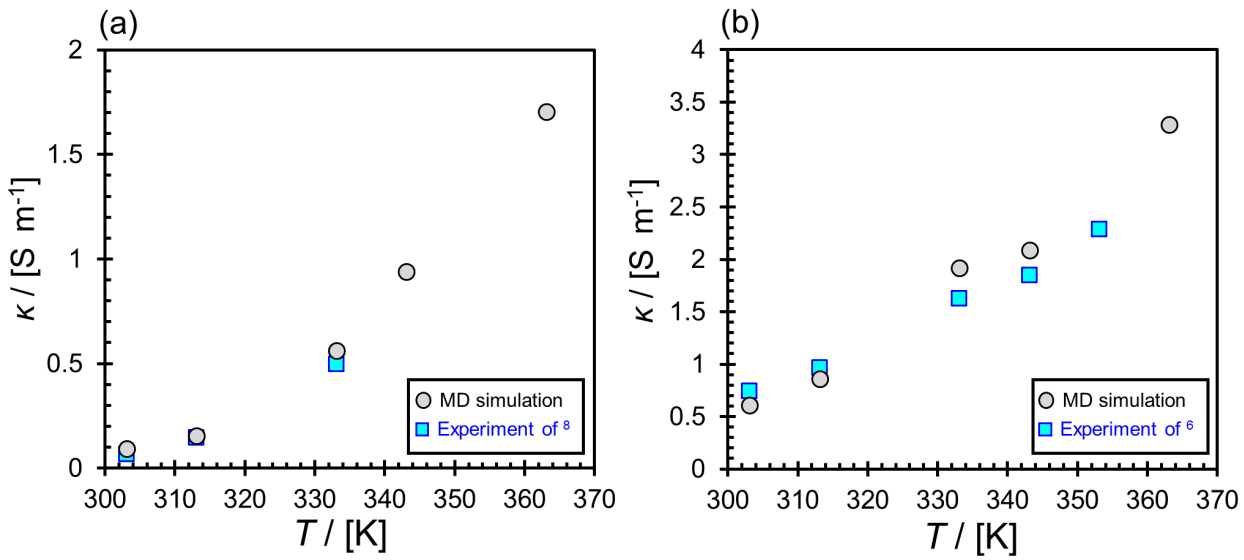

Figure S5: Ionic conductivity of (a) neat reline and (b) neat ethaline as a function of temperature. Black-gray circles represent computed ionic conductivities. Blue-cyan squares are experiment results for reline[8] and ethaline.[6]

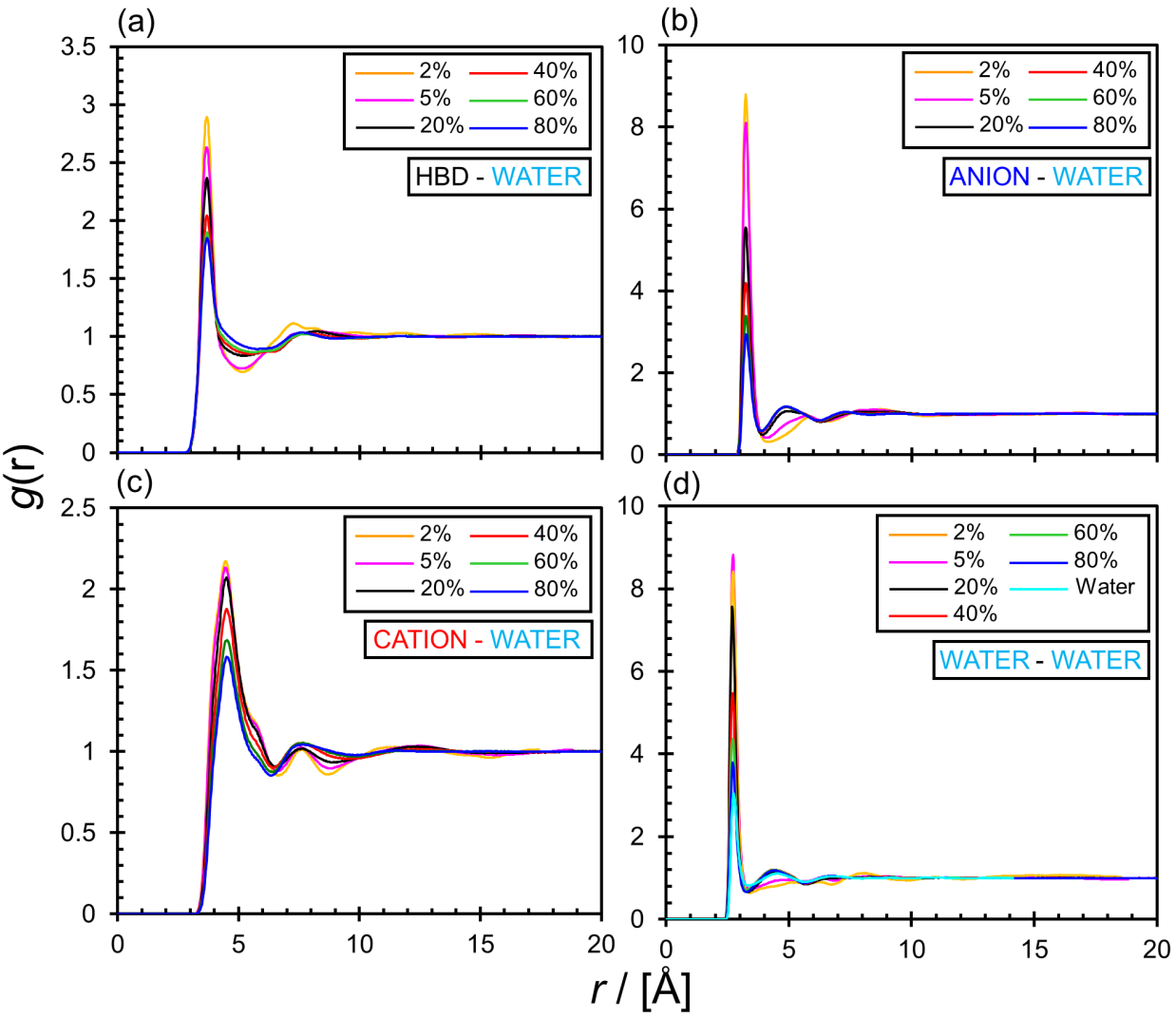

Figure S6: Radial distribution functions of (a) urea-water, (b) Cl^--water, (c) Ch^+-water, and (d) water-water in aqueous reline solution as a function of the mass fraction of water at 303.15 K and 1 atm.

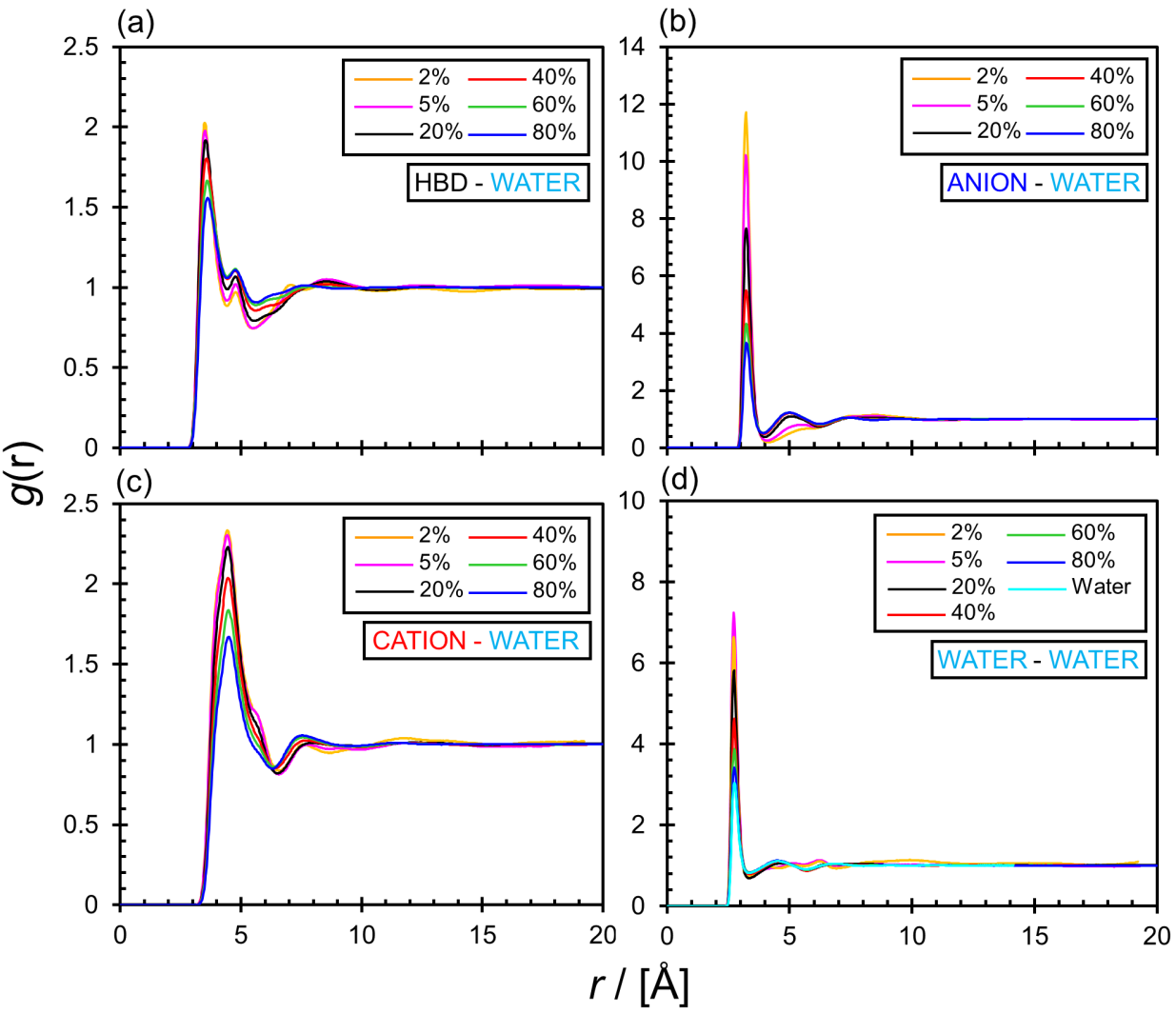

Figure S7: Radial distribution functions of (a) EG-water, (b) Cl^--water, (c) Ch^+-water, and (d) water-water in aqueous ethaline solution as a function of the mass fraction of water at 303.15 K and 1 atm.

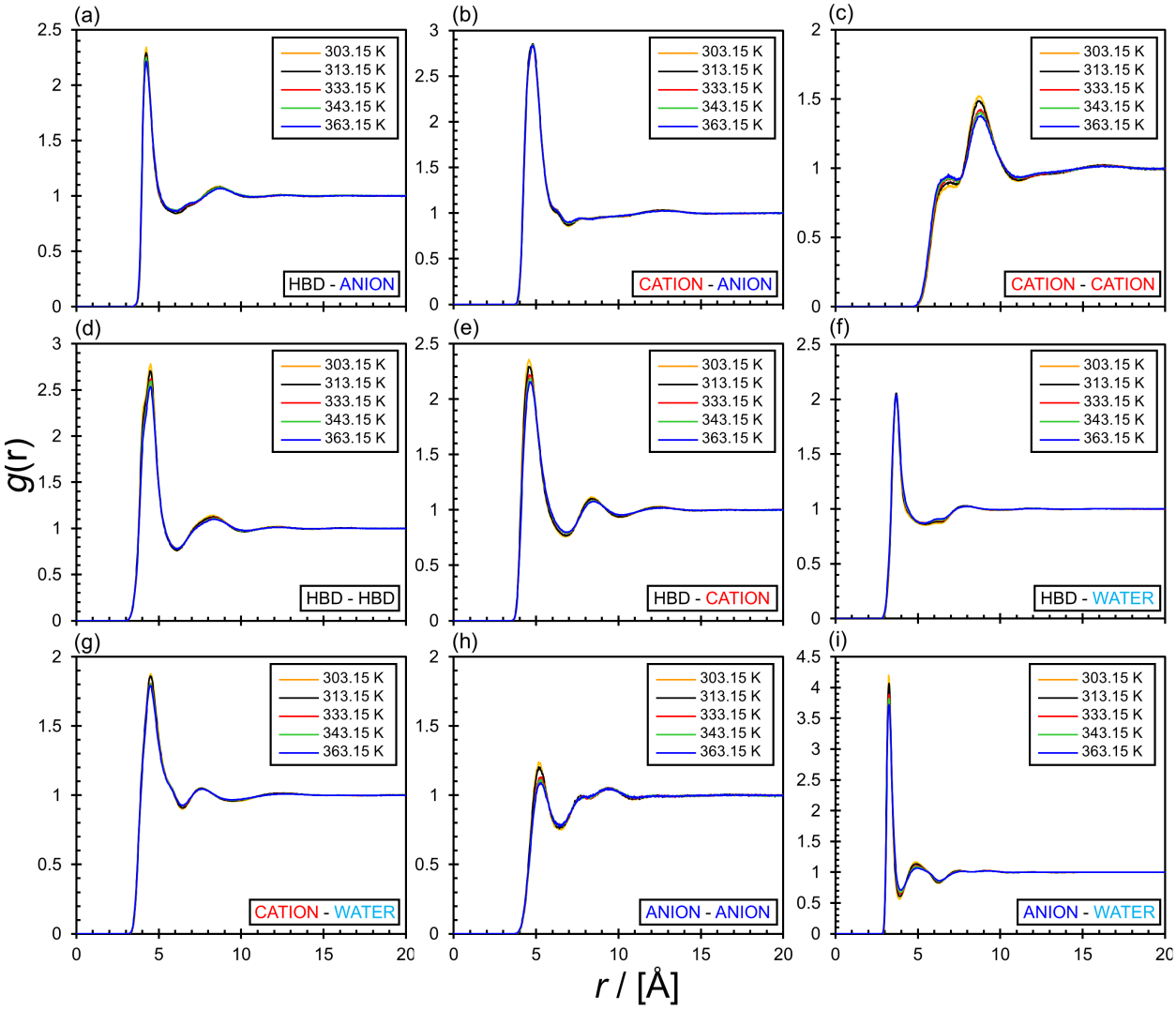

Figure S8: Influence of temperature on radial distribution functions of (a) urea-Cl^-, (b) Ch^+-Cl^-, (c) Ch^+-Ch^+, (d) urea-urea, (e) urea-Ch^+, (f) urea-water, (g) Ch^+-water, (h) Cl^--Cl^-, and (i) Cl^--water in an aqueous reline mixture with mass fraction of water of 40%.

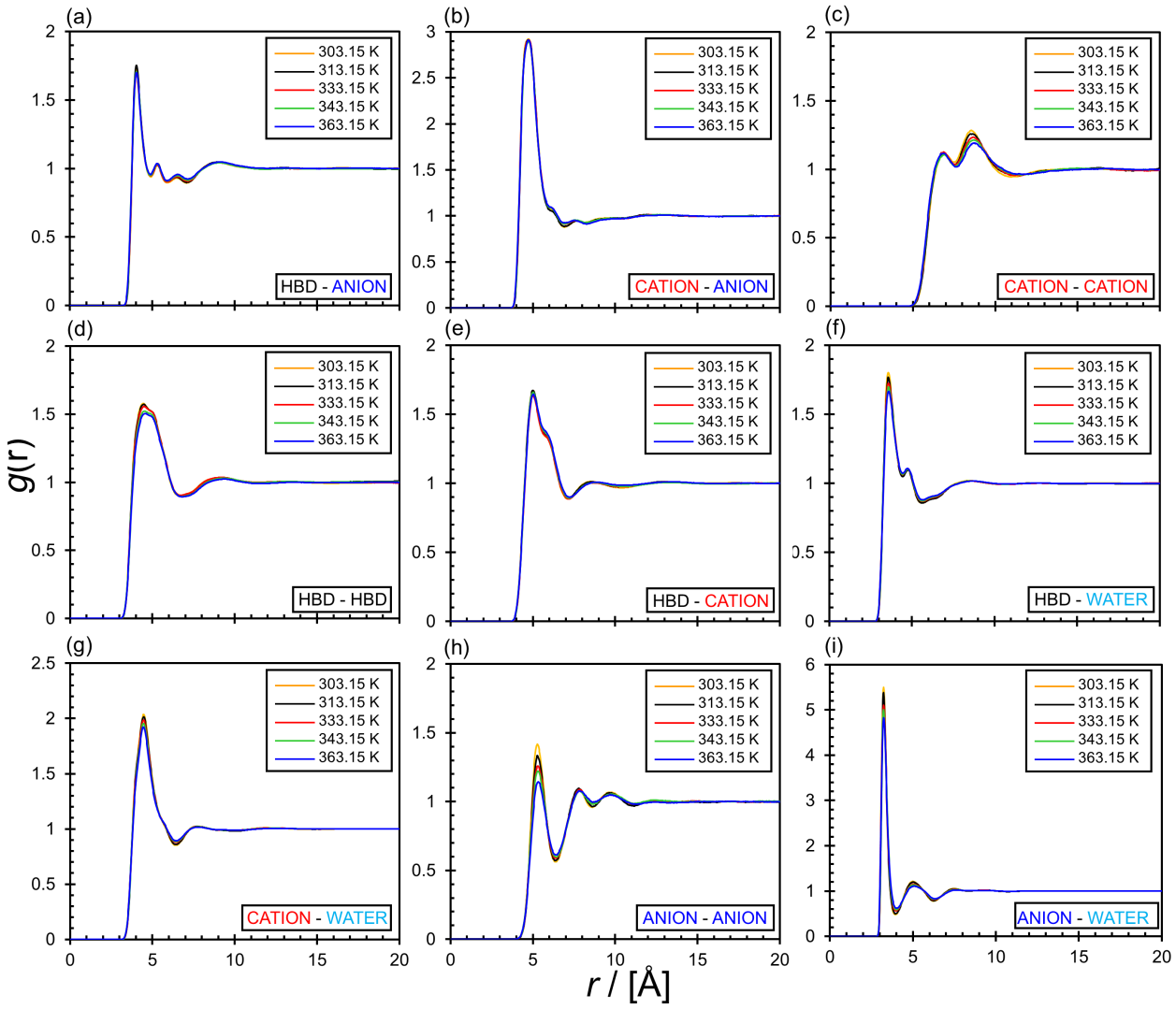

Figure S9: Influence of temperature on radial distribution functions of (a) EG-Cl^-, (b) Ch^+-Cl^-, (c) Ch^+-Ch^+, (d) EG-EG, (e) EG-Ch^+, (f) urea-water, (g) Ch^+-water, (h) Cl^--Cl^-, and (i) Cl^--water in an aqueous ethaline mixture with mass fraction of water of 40%.

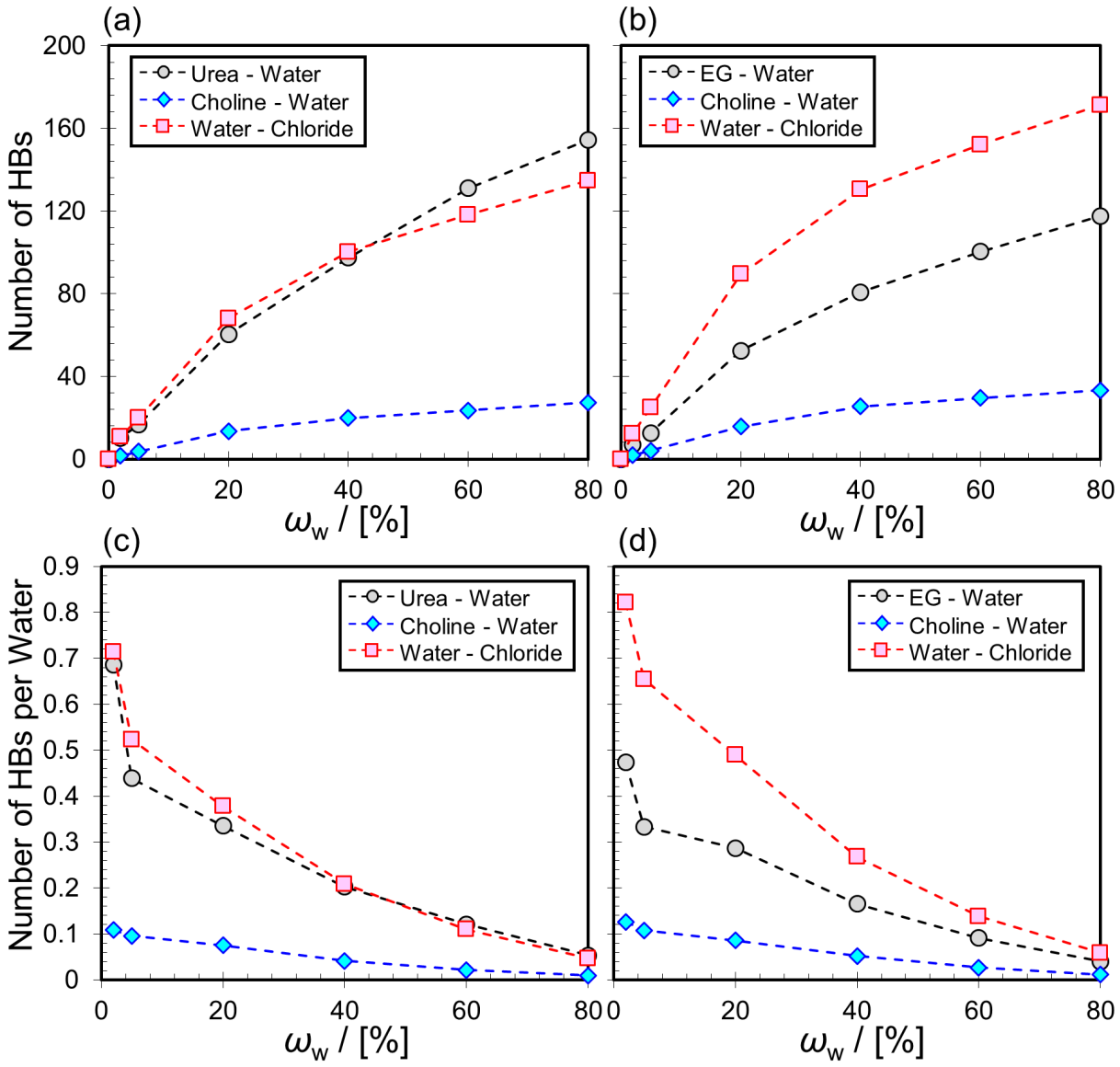

Figure S10: Number of hydrogen bonds as a function of the mass fraction of water between (a) water molecules and components of reline and (b) water molecules and components of ethaline. Number of hydrogen bonds per water molecule as a function of the mass fraction of water between (c) water molecules and components of reline and (d) water molecules and components of ethaline. Each simulation of reline contains 50 choline chloride molecules and 100 urea molecules. Each simulation of ethaline simulation contains 50 choline chloride molecules and 100 ethylene glycol molecules. The number of water molecules are presented in Table S12.

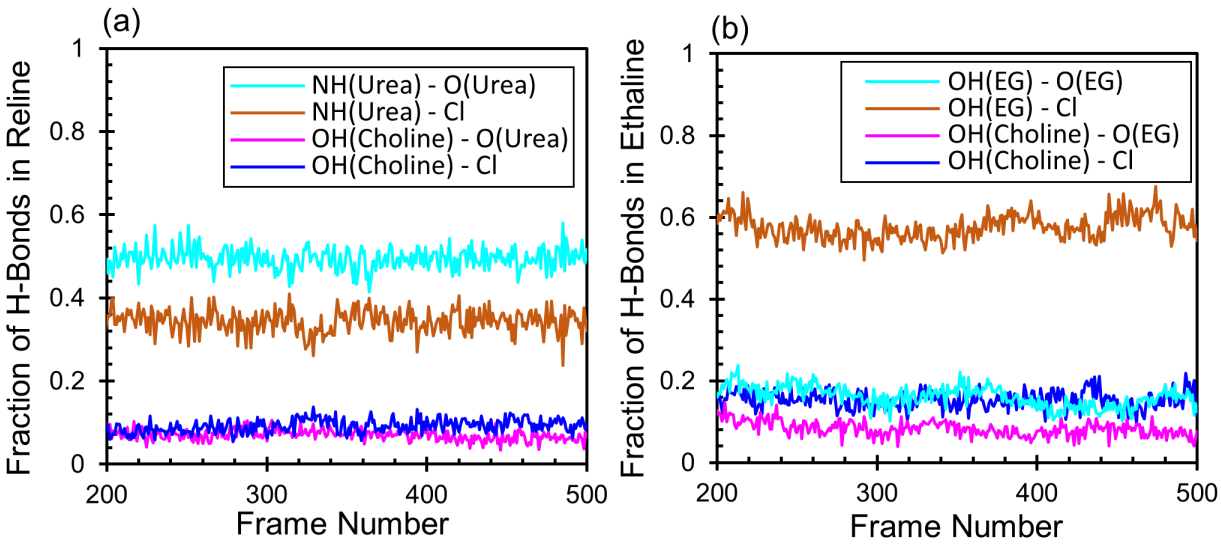

Figure S11: Fraction of hydrogen bonds for (a) neat reline and (b) neat ethaline at 303.15K temperature.

References

(1) Perkins, S. L.; Painter, P.; Colina, C. M. Experimental and Computational Studies of Choline Chloride-based Deep Eutectic Solvents. *J. Chem. Eng. Data* **2014**, *59*, 3652–3662.

(2) Mark, P.; Nilsson, L. Structure and Dynamics of the TIP3P, SPC, and SPC/E Water Models at 298 K. *Journal of Physical Chemistry A* **2001**, *105*, 9954–9960.

(3) Wang, J.; Wolf, R. M.; Caldwell, J. W.; Kollman, P. A.; Case, D. A. Development and Testing of a General Amber Force Field. *J. Comput. Chem.* **2004**, *25*, 1157–1174.

(4) Yadav, A.; Pandey, S. Densities and Ciscosities of (Choline Chloride+ Urea) Deep Eutectic Solvent and its Aqueous Mixtures in the Temperature Range 293.15 K to 363.15 K. *J. Chem. Eng. Data* **2014**, *59*, 2221–2229.

(5) Yadav, A.; Kar, J. R.; Verma, M.; Naqvi, S.; Pandey, S. Densities of Aqueous Mixtures of (Choline Chloride+ Ethylene Glycol) and (Choline Chloride+ Malonic Acid) Deep Eutectic Solvents in Temperature Range 283.15–363.15 K. *Thermochim. Acta* **2015**, *600*, 95–101.

(6) Mjalli, F.; Ahmed, O. Ethaline and Glyceline Binary Eutectic Mixtures: Characteristics and Intermolecular Interactions. *Asia-Pac. J. Chem. Eng.* **2017**, *12*, 313–320.

(7) D'Agostino, C.; Harris, R. C.; Abbott, A. P.; Gladden, L. F.; Mantle, M. D. Molecular Motion and Ion Diffusion in Choline Chloride Based Deep Eutectic Solvents Studied by 1 H Pulsed Field Gradient NMR Spectroscopy. *Phys. Chem. Chem. Phys.* **2011**, *13*, 21383–21391.

(8) Agieienko, V.; Buchner, R. Densities, Viscosities, and Electrical Conductivities of Pure Anhydrous Reline and Its Mixtures with Water in the Temperature Range (293.15 to 338.15) K. *J. Chem. Eng. Data* **2019**, *64*, 4763–4774.