Supporting Information

Table S1. Summary of the MD simulations of alkali halide water mixtures: All simulations were performed at 300 K and 1atm in the *NpT* ensemble. Symbols are N_w, number of water molecules; N_s (= $N_+ = N_- = 1/2Nc$), number of alkali-halide pairs; V, average simulation volume; m_s, salt molality; C_s, salt molarity; ρ, mass density; E_{pot}, average total potential energy per molecule ($N_s + N_w$); and T_{sim}, total simulation time.

	N_s	N_w	m_s (mol/Kg)	V (nm^3)	C_s (mol/l)	ρ (g/cm^3)	E_{pot} (kJ/mol)	T_{sim} (ns)
H2O	0	2170	0.00	65.265	0.00	0.995	-46.45	2
NaF	20	2150	0.52	64.531	0.52	1.018	-54.94	6
	38	2079	1.01	64.519	1.03	1.040	-63.05	6
NaCl	38	2079	1.01	63.595	0.99	1.036	-60.07	6
	77	2048	2.09	63.829	2.00	1.077	-73.99	4
	115	1987	3.21	63.354	3.01	1.114	-88.00	4
	154	1950	4.38	63.783	4.01	1.149	-102.02	4
NaBr	38	2079	1.01	64.089	0.98	1.072	-59.62	5
	77	2048	2.09	64.810	1.97	1.148	-73.05	5
	115	1987	3.21	64.730	2.95	1.222	-86.59	9
	154	1950	4.38	65.584	3.90	1.291	-100.08	5
	231	1730	7.41	63.426	6.05	1.438	-132.53	9
	308	1600	10.69	64.350	7.95	1.562	-163.90	9
NaI	38	2079	1.01	65.051	0.97	1.101	-58.86	5
	77	2048	2.09	66.683	1.92	1.206	-71.52	5
	115	1987	3.21	67.458	2.83	1.305	-84.27	5
	154	1950	4.38	69.151	3.70	1.398	-96.97	5
	231	1730	7.41	68.599	5.59	1.593	-127.35	5
	308	1600	10.69	71.253	7.18	1.748	-156.79	5
LiCl	127	7065	1.00	216.903	0.97	1.016	-62.37	6
	367	6796	3.00	217.842	2.80	1.052	-92.29	6
	589	6541	5.00	219.083	4.47	1.082	-120.13	6
KCl	126	7002	1.00	216.178	0.97	1.041	-58.36	6
	357	6603	3.00	215.924	2.75	1.120	-80.74	6
	561	6228	5.00	216.204	4.31	1.183	-101.56	6
RbCl	125	6963	1.00	216.055	0.96	1.080	-57.79	6
	352	6512	3.00	215.995	2.71	1.229	-79.14	6
	549	6093	5.00	216.324	4.22	1.352	-99.03	6
CsCl	125	6915	1.00	216.032	0.96	1.119	-57.38	6
	345	6385	3.00	215.737	2.66	1.333	-77.74	6
	533	5920	5.00	216.025	4.10	1.510	-96.76	6
KI	124	6880	1.00	217.183	0.95	1.105	-57.18	6
	340	6300	3.00	218.381	2.59	1.292	-77.20	6
	522	5796	5.00	219.331	3.95	1.447	-95.97	6
CsBr	124	6870	1.00	216.248	0.95	1.153	-56.93	6
	339	6275	3.00	216.377	2.60	1.422	-76.47	6
	519	5761	5.00	216.598	3.98	1.640	-94.77	6

Table S2. First shell coordination numbers (n_{ij}^{1st}) and second shell coordination numbers (n_{ij}^{2nd}) as a function of concentration (m) alkali halide aqueous solutions. $R_{max}^{1st}/R_{min}^{1st}$ and R_{max}^{2nd} / R_{min}^{2nd} are the distances (nm) to the first and the second maximum/minimum of the radial distribution functions. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

		m	+/-	+/o	-/o	o/o
NaF	R_{max}^{1st}		0.25	0.23	0.30	0.27
	R_{min}^{1st}		0.326	0.310	0.364	0.342
	n_{ij}^{1st}	0.5	0.06	5.54	7.01	4.80
		1.0	0.03	5.48	7.04	4.75
	R_{max}^{2nd}		0.48	0.44	0.48	0.45
	R_{min}^{2nd}		0.562	0.540	0.592	0.564
	n_{ij}^{2nd}	0.5	0.39	17.51	21.90	19.29
		1.0	0.80	17.38	21.85	19.25
NaCl	R_{max}^{1st}		0.27	0.23	0.32	0.28
	R_{min}^{1st}		0.355	0.315	0.405	0.345
	n_{ij}^{1st}	0.99	0.09	5.52	8.11	5.12
		2.00	0.20	5.39	8.22	5.11
		3.01	0.42	5.11	8.38	5.06
		4.01	0.57	4.93	8.45	5.00
NaBr	R_{max}^{1st}		0.28	0.23	0.33	0.28
	R_{min}^{1st}		0.365	0.315	0.415	0.405
	n_{ij}^{1st}	0.98	0.10	5.49	7.54	5.09
		1.97	0.22	5.34	7.62	6.57
		2.95	0.35	5.17	8.11	7.06
		3.90	0.50	4.97	8.61	8.11
		6.05	0.96	4.33	9.66	8.72
		7.95	1.45	3.67	10.31	8.03
NaI	R_{max}^{1st}		0.29	0.23	0.35	0.28
	R_{min}^{1st}		0.375	0.315	0.425	0.395
	n_{ij}^{1st}	0.97	0.07	5.48	7.91	5.04
		1.92	0.16	5.40	7.98	6.42
		2.83	0.26	5.24	8.04	6.83
		3.70	0.39	5.05	8.59	7.18
		5.59	0.84	4.37	9.60	7.56
		7.18	1.31	3.66	10.80	6.79
LiCl	R_{max}^{1st}		0.23	0.19	0.32	0.28
	R_{min}^{1st}		0.315	0.265	0.398	0.360
	n_{ij}^{1st}	1	0.04	3.96	7.67	5.51
		3	0.12	3.88	7.80	5.40
		5	0.22	3.78	7.91	5.25
	R_{max}^{2nd}		0.46	0.41	0.50	0.45
	R_{min}^{2nd}		0.546	0.502	0.626	0.566
	n_{ij}^{2nd}	1	0.61	14.16	25.07	18.59
		3	1.48	13.29	24.32	17.29
		5	2.26	12.46	23.36	16.27
KCl	R_{max}^{1st}		0.31	0.26	0.32	0.28
	R_{min}^{1st}		0.389	0.342	0.384	0.334
	n_{ij}^{1st}	1	0.20	6.11	7.03	4.32
		3	0.54	5.77	6.79	4.03
		5	0.89	5.39	6.48	3.74

RbCl	R_{max}^{1st}		0.32	0.28	0.32	0.27
	R_{min}^{1st}		0.404	0.361	0.385	0.332
	n_{ij}^{1st}	1	0.24	6.75	7.03	4.23
		3	0.60	6.35	6.72	3.89
		5	0.95	5.95	6.32	3.57
CsCl	R_{max}^{1st}		0.34	0.29	0.32	0.27
	R_{min}^{1st}		0.428	0.376	0.385	0.334
	n_{ij}^{1st}	1	0.34	7.15	6.91	4.28
		3	0.78	6.63	6.48	3.92
		5	1.18	6.15	6.06	3.59
KI	R_{max}^{1st}		0.33	0.26	0.34	0.27
	R_{min}^{1st}		0.418	0.342	0.412	0.332
	n_{ij}^{1st}	1	0.24	6.02	7.34	4.20
		3	0.68	5.47	7.00	3.83
		5	1.10	4.96	6.61	3.51
CsBr	R_{max}^{1st}		0.34	0.29	0.33	0.27
	R_{min}^{1st}		0.438	0.380	0.394	0.332
	n_{ij}^{1st}	1	0.39	7.18	6.90	4.19
		3	0.93	6.47	6.34	3.81
		5	1.38	5.88	5.85	3.49

Table S3. Fitting constants for Equation 4 determined by fitting experimental alkali halide activity coefficients and the corresponding simulated activity derivatives.

		a_1	a_2	a_3	a_4	m_s^{max}
NaF	Exp	1.2759	-0.0410	0	0	1.0
	KBFF	1.2759	0.1757	0	0	1.0
NaCl	Exp	1.3360	0.0810	0	0	6.0
	KBFF	1.3360	0.0827	0	0	5.0
NaBr	Exp	1.4360	0.1041	0	0	4.0
	KBFF	1.4360	0	0.3342	0.0263	5.0
NaI	Exp	1.6350	0.1300	0	0	3.5
	KBFF	1.6350	0	0.4308	0.0133	5.0
LiCl	Exp	1.4644	0	0.2110	0.0102	6.0
	KBFF	1.4644	0	-0.2430	0.0086	5.0
KCl	Exp	1.1720	0.0360	0	0	5.0
	KBFF	1.1720	0.0260	0	0	5.0
RbCl	Exp	1.0643	0.0311	0	0	5.0
	KBFF	1.0643	0.0482	0	0	5.0
CsCl	Exp	0.8402	0.0311	0	0	6.0
	KBFF	0.8402	0.0398	0	0	5.0
KI	Exp	1.4259	0.0480	0	0	4.5
	KBFF					N/A
CsBr	Exp	0.8453	0.0216	0	0	5.0
	KBFF					N/A

Expermental data was fitted to Equation 4

$$\ln \gamma_{\pm} = \frac{-1.178\sqrt{m_s}}{1+a_1\sqrt{m_s}} - \ln(1-a_2 m_s) + a_3 m_s + a_4 m_s^2$$

Simulated data was fitted using the following activity coefficient derivative,

$$\left(\frac{\partial \ln \gamma_{\pm}}{\partial \ln m_s}\right)_{T,P} = \frac{-N_{22}^{+}}{1+N_{22}^{+}} \qquad N_{22}^{+} = N_{22} + m_2(1+N_{11}-2N_{21})$$

which can be then be expressed in terms of the derivative of Equation 4. The fits should be considered approximate due to the relatively low number of points used (typically three). The simulated data for KI and CsBr could not be fitted accurately using the above relationships.

Table S4. Lattice energies (kJ/mol) for alkali halide crystals.

	Exp	Hagler FF	KBFF
NaF	895.4	937.2	1217.7
NaCl	765.7	786.6	808.2
NaBr	728.0	744.8	776.1
NaI	682.0	698.7	751.0
LiCl	832.6	861.9	1178.0
KCl	694.5	711.3	725.3
RbCl	665.3	686.2	692.7
CsCl	648.5	644.3	650.1
KI	627.6	636.0.	663.2
CsBr	627.6	615.0	628.8

Exp: Experimental data taken from (Tosi, M. P.; Fumi, F. G. *J Phys Chem Solids* 1964, *25*, 45)

Hagler FF: Hagler force field data taken from (Peng, Z. W.; Ewig, C. S.; Hwang, M. J.; Waldman, M.; Hagler, A. T. *J Phys Chem A* 1997, *101*, 7243)

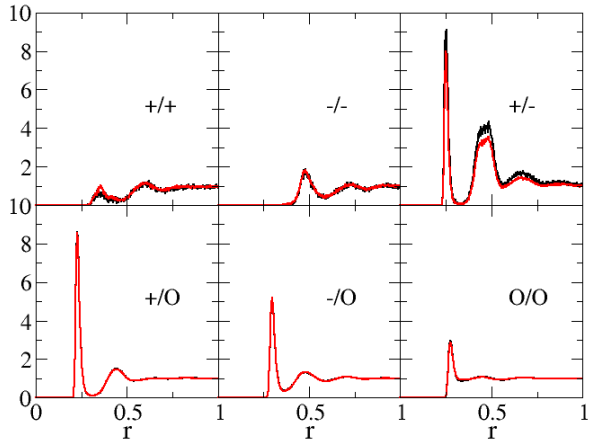

Figure S1. Radial distribution functions of NaF obtained from the 0.5 m (black lines) and 1 m (red lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

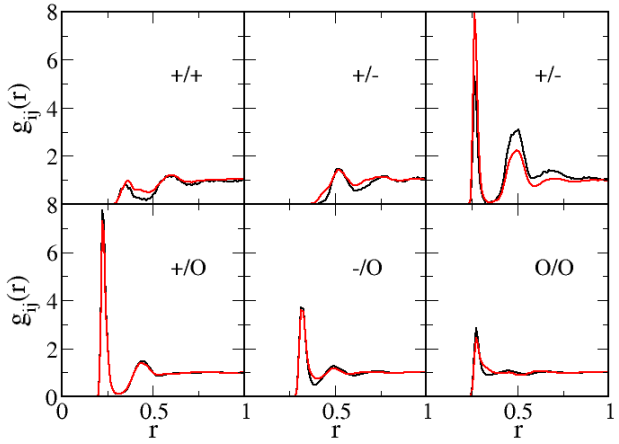

Figure S2. Radial distribution functions of NaCl obtained from the 0.99 m (black lines) and 4.01 m (red lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

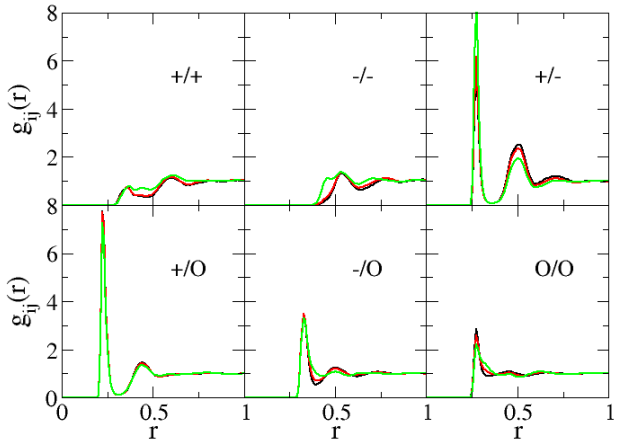

Figure S3. Radial distribution functions of NaBr obtained from the 0.98 m (black lines), 2.95 m (red lines), and 6.05 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

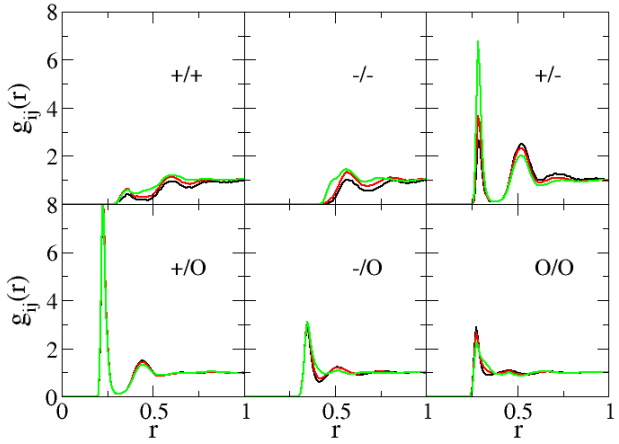

Figure S4. Radial distribution functions of NaI obtained from the 0.97 m (black lines), 2.83 m (red lines), and 5.59 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

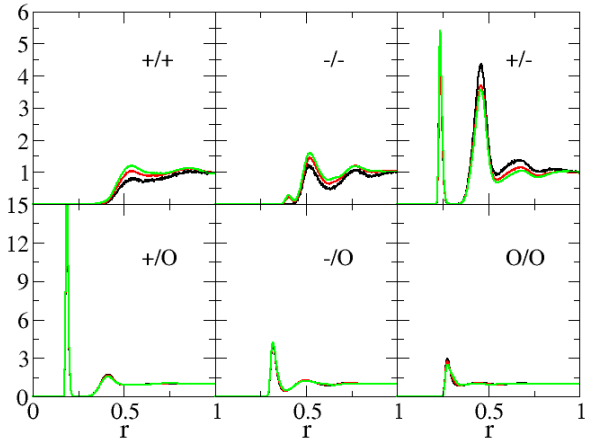

Figure S5. Radial distribution functions of LiCl obtained from the 1 m (black lines), 3 m (red lines), and 5 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

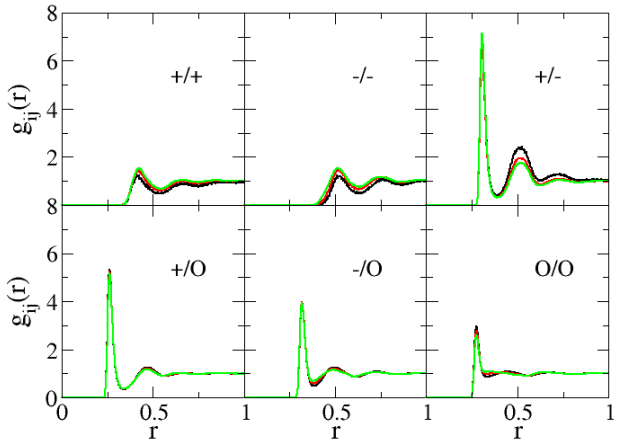

Figure S6. Radial distribution functions of KCl obtained from the 1 m (black lines), 3 m (red lines), and 5 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

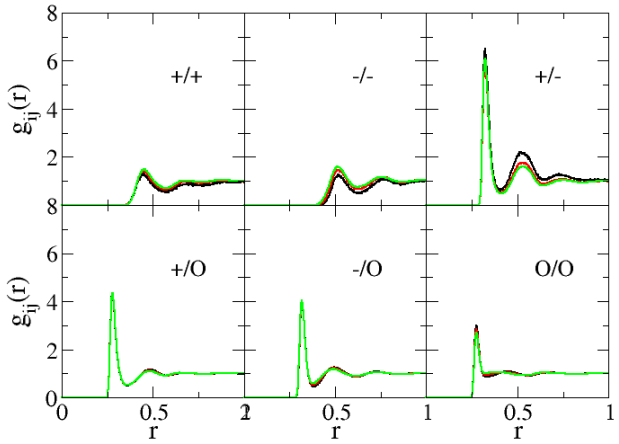

Figure S7. Radial distribution functions of RbCl obtained from the 1 m (black lines), 3 m (red lines), and 5 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

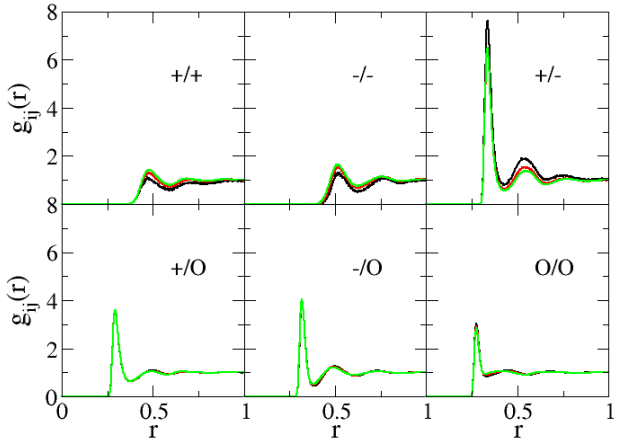

Figure S8. Radial distribution functions of CsCl obtained from the 1 m (black lines), 3 m (red lines), and 5 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

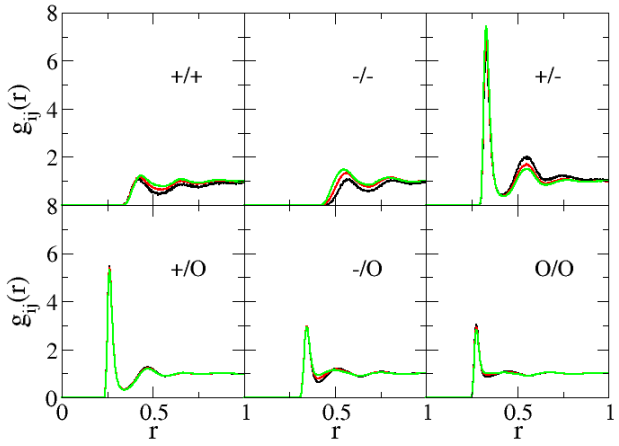

Figure S9. Radial distribution functions of KI obtained from the 1 m (black lines), 3 m (red lines), and 5 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.

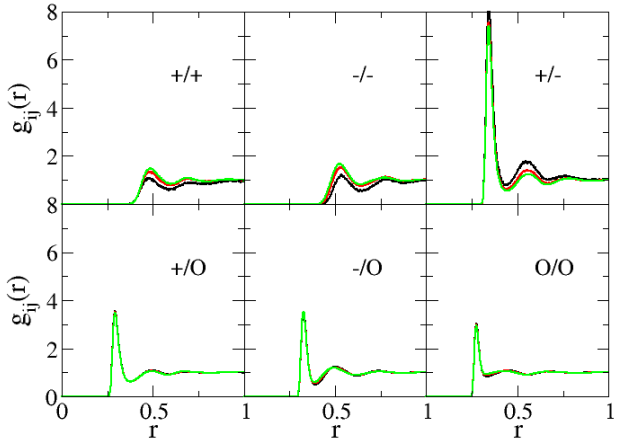

Figure S10. Radial distribution functions of CsBr obtained from the 1 m (black lines), 3 m (red lines), and 5 m (green lines) simulations. Cations, anions, and the water oxygen are denoted by the symbols +, -, and o, respectively.