Supplementary material

Scaled charges for ions: an improvement but not the final word for modeling electrolytes in water

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I. DENSITIES

The numerical results for the densities of NaCl and KCl solutions obtained in this work with all the developed models are collected in this supplementary material.

TABLE I. Model q = 0.75 Madrid-Transport simulation results for density obtained for NaCl solutions in TIP4P/2005 water at temperature T = 298.15 K and pressure P = 1 bar for different concentrations below experimental solubility. Numbers in parentheses are the uncertainty in the results. Expt data were taken from ref[1].

Molality (mol/kg)	Density kg/m^3	
m	Sim	Expt
0	997.3(5)	997.043
1	1035.2(5)	1036.21
2	1070.31(5)	1072.27
3	1103.77(5)	1105.76
4	1135.44(5)	1136.9
5	1165.64(5)	1165.91
6	1194.5(5)	1192.88

TABLE II. Model q = 0.8 simulation results for density obtained for NaCl solutions in TIP4P/2005 water at temperature T = 298.15 K and pressure P = 1 bar for different concentrations below experimental solubility. Numbers in parentheses are the uncertainty in the results. Expt data were taken from ref[1].

Molality (mol/kg)	Density kg/m^3	
m	Sim	Expt
0	997.043(5)	997.043
1	1035.76(5)	1036.21
2	1071.66(5)	1072.27
3	1105.15(5)	1105.76
4	1137.08(5)	1136.91
5	1167.16(5)	1165.91
6	1195.67(5)	1192.88

TABLE III. Model q = 0.92 Madrid-Interfacial simulation results for density obtained for NaCl solutions in TIP4P/2005 water at temperature T = 298.15 K and pressure P = 1 bar for different concentrations below experimental solubility. Numbers in parentheses are the uncertainty in the results. Expt data were taken from ref[1].

Molality (mol/kg)	Density kg/m^3	
m	Sim	Expt
0	997.3(5)	997.043
1	1037.24(5)	1036.21
2	1072.78(5)	1072.27
3	1105.72(5)	1105.76
4	1135.83(5)	1136.9
5	1163.81(5)	1165.91
6	1189.79(5)	1192.88

TABLE IV. Model q = 0.75 Madrid-Transport simulation results for density obtained for KCl solutions in TIP4P/2005 water at temperature T = 298.15 K and pressure P = 1 bar for different concentrations below experimental solubility. Numbers in parentheses are the uncertainty in the results. Expt data were taken from ref[1].

Molality (mol/kg)	Density kg/m^3	
m	Sim	Expt
0	997.3(5)	997.043
1	1041.10(5)	1041.4
2	1081.18(5)	1081.5
3	1118.18(5)	1118.3
4	1152.28(5)	1152.2
4.5	1168.44(5)	1168.2

TABLE V. Model q = 0.92 Madrid-Interfacial simulation results for density obtained for KCl solutions in TIP4P/2005 water at temperature T = 298.15 K and pressure P = 1 bar for different concentrations below experimental solubility. Numbers in parentheses are the uncertainty in the results. Expt data were taken from ref[1].

Molality (mol/kg)	Density kg/m^3	
m	Sim	Expt
0	997.3(5)	997.043
1	1043.36(5)	1041.4
2	1083.85(5)	1081.5
3	1120.55(5)	1118.3
4	1153.88(5)	1152.2
4.5	1169.10(5)	1168.2

II. VISCOSITIES OF KCL AQUEOUS SOLUTIONS

In this section we show the results for the viscosities of the KCl solutions at 298.15 K and 1 bar using Madrid-Transport and Madrid-2019 force fields.

TABLE VI. Results for viscosity obtained with the Madrid-2019 and Madrid-Transport models for KCl solutions in TIP4P/2005 water at temperature T = 298.15 K and pressure p = 1 bar for different concentrations below experimental solubility. Expt data were taken from refs[2,3]

Molality (mol/kg)	Viscosity mPa·s		
m	Expt	q = 0.85 Madrid-2019	q = 0.75 Madrid-Transport
1	0.89	0.94	
2	0.90	1.03	
4	0.94	1.22	1.03

III. SURFACE TENSION

The numerical results of the surface tensions evaluated in this work for NaCl and KCl with different force fields are collected in this section.

TABLE VII. Surface tension of NaCl aqueous solutions relative to that of pure water values evaluated at 298.15 K and 1 bar (γ_{water}=65.486 mN·m^{-1}). Results obtained with different models. The asterisk (*) indicates that the system has been evaluated with a cutoff of 2.5 nm (pure water has been also evaluated with that cutoff, obtaining γ^{*}_{water}=67.5 mN·m^{-1}). The continuous lines are the fit of experimental data taken from refs[4].

		$\Delta\gamma$			
		q = ±1	q = ±0.92	q = ±0.85	q = ±0.75
Molality	Expt	JC-TIP4P/2005	Madrid-Interfacial	Madrid-2019	Madrid-Transport
(mol/kg)			mN·m^{-1}		
0	0	0	0	0	0
1	1.55				
2	3.12		2.679		
5	9.20	14.11	8.345	6.050	3.594
5*	9.20		8.03		

TABLE VIII. Surface tension of KCl aqueous solutions relative to that of pure water values evaluated at 298.15 K and 1 bar (γ_{water}=65.486 mN·m^{-1}). Results obtained with different models. The asterisk (*) indicates that the system has been evaluated with a cutoff of 2.5 nm (pure water has been also evaluated with that cutoff, obtaining γ^{*}_{water}=67.5 mN·m^{-1}) The continuous lines are the fit of experimental data taken from ref[5]

			$\Delta\gamma$		
		q = ±1	q = 0.92	q = ±0.85	q = ±0.75
Molality	Expt	JC-TIP4P/2005	Madrid-Interfacial	Madrid-2019	Madrid-Transport
(mol/kg)			mN·m^{-1}		
0	0	0	0	0	0
1	1.45				
2	3.00				
4	6.05	6.98	6.60	4.83	3.37
4*	6.05		6.63		

IV. TMD

The numerical results of the TMDs evaluated in this work for NaCl and KCl with different force fields are collected in this section.

TABLE IX. Temperatures of maximum densities and absolute values of maximum densities for NaCl aqueous solutions evaluated at 1 bar and different temperatures with different force fields.

Molality			q = 0.75		q = 0.8		q = 0.85		q = 0.92		q = 1		q = 1	
	Expt		Madrid-Transport				Madrid-2019		Madrid-Interfacial		JC-TIP4P/2005		Yagasaki Model	
m	TMD	ρ	TMD	ρ	TMD	ρ	TMD	ρ	TMD	ρ	TMD	ρ	TMD	ρ
(mol/kg)	K	kg/m^3	K	kg/m^3	K	kg/m^3	K	kg/m^3	K	kg/m^3	K	kg/m^3	K	kg/m^3
1	262.7	1044.0	270.5	1039.47	264.4	1042.36	261.2	1044.18	262.0	1044.28	258.93	1051.13	258.1	1050.18

TABLE X. Temperatures of maximum densities and absolute values of maximum densities for KCl aqueous solutions evaluated at 1 bar and different temperatures with different force fields.

Molality			q = ±0.75		q = ±0.85		q = ±1		q = ±1	
	Expt		Madrid-Transport		Madrid-2019		JC-TIP4P/2005		Yagasaki Model	
m	TMD	ρ	TMD	ρ	TMD	ρ	TMD	ρ	TMD	ρ
(mol/kg)	K	kg/m^3	K	kg/m^3	K	kg/m^3	K	kg/m^3	K	kg/m^3
1	265.00	1047.5	267.6	1046.45	266.7	1047.90	261.8	1055.04	261.6	1053.98

V. EVALUATION OF CIP

In Fig. 1 we have plotted the Na-Cl RDFs multiplied by r^2 at 6 m for the different force fields developed in this work. The integral of these plots (multiplied by $4\pi r^2$ and ρ_-) are the number of CIP (which are specified in the legend).

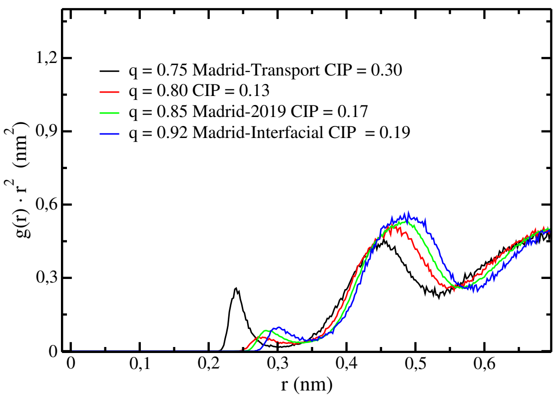

FIG. 1. Na-Cl RDFs multiplied by r^2 at T = 298.15 K and 1 bar for 6 m NaCl aqueous solutions. We show the results of this work for the different developed force fields and the Madrid-2019.

VI. RELATIVE CHANGE OF THE DIELECTRIC CONSTANT

In Figure 2 we show the behavior of the relative change of the dielectric constant. We define $\Delta\epsilon_r = (\epsilon_{solution} - \epsilon_{H_2O})/\epsilon_{H_2O}$, being $\epsilon_{solution}$ the dielectric constant of the NaCl solution (at each concentration) and ϵ_{H_2O} the dielectric constant of pure water.

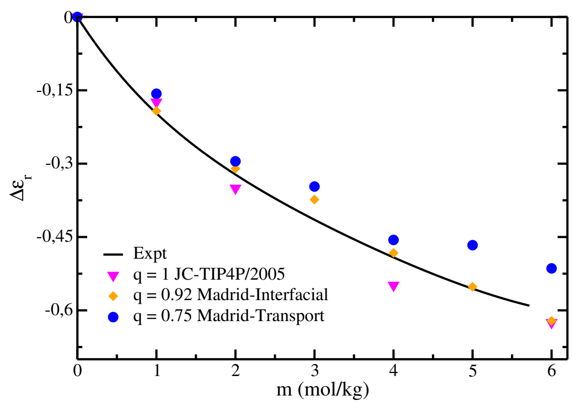

FIG. 2. Relative change of the dielectric constant as a function of molality for NaCl aqueous solutions at 1 bar and 298.15 K evaluated with different force fields. Magenta triangles: q = ±1 JC-TIP4P/2005. Orange diamonds: q = ±0.92 Madrid-Interfacial. Blue circles: q = ±0.75 Madrid-Transport. Experimental values are also shown with a fitted curve of the data[6,7].

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