

Statistical Associating Fluid Theory Coupled with Restrictive Primitive Model Extended to Bivalent Ions. SAFT2: 2. Brine/Seawater Properties Predicted

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Statistical associating fluid theory coupled with restricted primitive model (SAFT2) represents the properties of aqueous multiple-salt solutions, such as brine/seawater. The osmotic coefficients, densities, and vapor pressures are predicted without any additional parameters using the salt hydrated diameters obtained for single-salt solutions. For a given ion composition of brine, the predicted vapor pressure, osmotic coefficient, activity of water, and density are found to agree with the experimental data.

1. Introduction

The properties of electrolyte solutions, such as the activity coefficients, osmotic coefficients, densities, and vapor pressures, can be estimated from a predictive equation of state, for example, from statistical associating fluid theory (SAFT) coupled with restricted primitive model (SAFT1-RPM).^{1,2} SAFT1-RPM was used to represent the properties of single-salt and multiple-salt solutions in water, but its application was limited to monovalent ions due to a limited range of the square-well width implied in SAFT1.^{1,2} This limitation is removed in SAFT2 developed in the first part of this work,³ which makes it applicable to multivalent ions and represent the properties of aqueous single-salt solutions composed of pairs of five cations (Li^+ , Na^+ , K^+ , Ca^{2+} , Mg^{2+}) and six anions (Cl^- , Br^- , I^- , NO_3^- , HCO_3^- , SO_4^{2-}). Similar to SAFT1-RPM, SAFT2 has three ion parameters (energy, volume, and square-well width) and one salt parameter (hydrated diameter). These parameters were developed on the basis of the activity coefficients and densities of single-salt solutions alone and shown by Tan et al.³ to predict the osmotic coefficients for these solutions.

The purpose of this work is to understand how this approach works for multiple-salt solutions, without additional fitting, and to develop a predictive molecular model that is applicable within broad ranges of brine and seawater salinities.

2. Mixing Rules

As described in the first part of this work,³ SAFT2 is defined in terms of the dimensionless residual Helmholtz energy

$$\tilde{a}^{\text{res}} = \tilde{a}^{\text{hs}} + \tilde{a}^{\text{disp}} + \tilde{a}^{\text{chain}} + \tilde{a}^{\text{assoc}} + \tilde{a}^{\text{ion}} \quad (1)$$

where the superscripts refer to terms accounting for the residual, hard sphere, dispersion, chain, association, and ionic or electrostatic interactions, respectively.

For aqueous salt solutions, the segment numbers (m) for water and ions are set equal to unity, which make the chain term equal to zero. Water is modeled as a single-segmented molecule with four association sites (type 4C—Huang and Radosz⁴). Salt is

modeled as a molecule composed of two charged, but nonassociating, spherical segments, of which one represents the cation and the other represents the anion. For each segment, there are three parameters: segment volume (v), segment energy (u/k), and reduced well range of the square-well potential (λ). There is another parameter for each salt, that is, the effective salt hydrated diameter (d) that is used in the ionic term.

The mixing rules for v , u , and λ between two segments α and β are the same as those in SAFT1⁵ as follows

$$v_{\alpha\beta} = \left[\frac{(v_\alpha)^{1/3} + (v_\beta)^{1/3}}{2} \right]^3 \quad (2)$$

$$u_{\alpha\beta} = (u_\alpha u_\beta)^{1/2} \quad (3)$$

$$\lambda_{\alpha\beta} = \frac{\lambda_\alpha + \lambda_\beta}{2} \quad (4)$$

It is assumed that there are no dispersive interactions between ions, so eq 3 applies to the interaction between water molecules and ions. The dispersive binary interaction parameter (k_{ij}) between water and ions, if any, is implicitly accounted for by the ion energy parameter (u), which is obtained from binary (salt + water) properties. Therefore, the ion energy parameter is in fact the effective energy of the ions interacting with water molecules.

The mixing rule for the effective salt hydrated diameter d is

$$d = \sum_i x'_i d_i \quad (5)$$

where

$$x'_i = \frac{C_i^m}{\sum_i C_i^m} \quad (6)$$

The summation is over all salts in the mixture and C_i^m is the molality of salt i , so that x' is the salt mole fraction in a solvent-free basis.

3. Results and Discussion

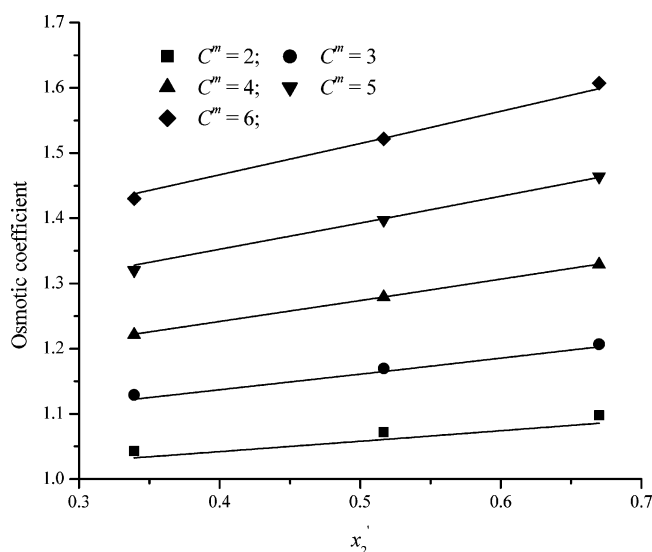
The SAFT2 model described above is used to predict the properties of osmotic coefficients, vapor pressures, and liquid

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TABLE 1: ARD for Osmotic Coefficient of Aqueous Two-salt Solutions and Including Solutions Containing K^+ and Another Cation

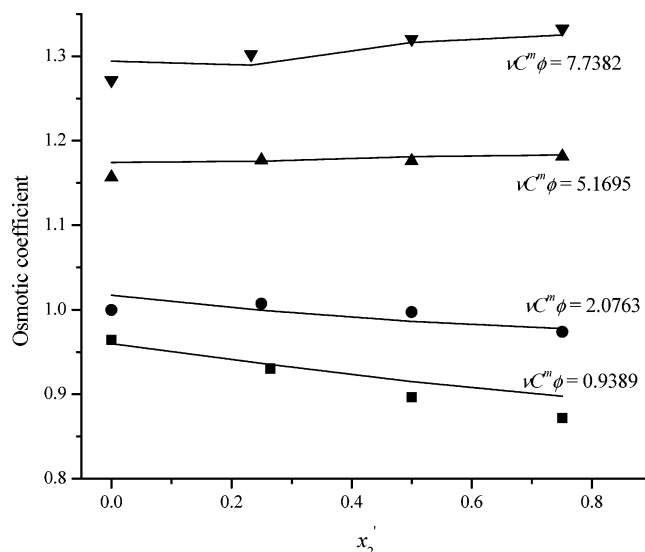
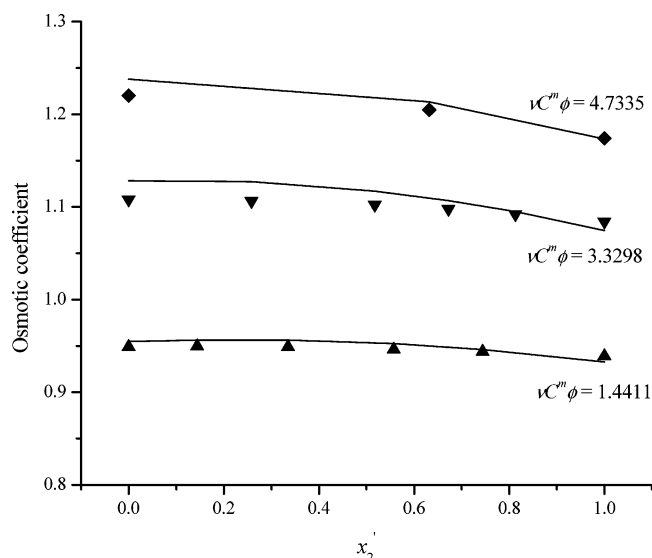
solution	ARD, %	ref	solution	ARD, %	ref
(a) ARD for Osmotic Coefficient of Aqueous Two-salt Solutions					
LiCl + NaCl	0.47	^a	KCl + KBr	4.93	^d
LiCl + CaCl ₂	0.99	^b	KCl + K ₂ SO ₄	1.18	ⁱ
LiCl + MgCl ₂	0.87	^c	KCl + KNO ₃	1.86	^e
NaCl + NaBr	0.29	^d	NaCl + MgCl ₂	1.68	^j
NaCl + NaNO ₃	0.98	^e	NaCl + Na ₂ SO ₄	1.02	^j
NaCl + CaCl ₂	0.49	^f	Na ₂ SO ₄ + MgSO ₄	2.90	^j
CaCl ₂ + MgCl ₂	0.78	^g	MgSO ₄ + MgCl ₂	2.44	^j
MgCl ₂ + Mg(NO ₃) ₂	1.28	^h	NaCl + MgSO ₄	1.82	^k
CaCl ₂ + Mg(NO ₃) ₂	2.84	^h	Na ₂ SO ₄ + MgCl ₂	1.65	^k
(b) ARD for Osmotic Coefficient of Aqueous Two-salt Solutions Containing K^+ and Another Cation					
KCl + LiCl	13.4	^a	KBr + NaCl	5.18	^d
KCl + NaCl	5.17	^{d,l}	KBr + NaBr	3.53	^d
KCl + NaBr	4.32	^d	KNO ₃ + NaCl	5.91	^e
KCl + NaNO ₃	10.3	^e	KNO ₃ + NaNO ₃	4.84	^e
KCl + Na ₂ SO ₄	8.62	ⁱ	K ₂ SO ₄ + NaCl	6.66	ⁱ
KCl + CaCl ₂	8.39	^m	K ₂ SO ₄ + Na ₂ SO ₄	6.92	ⁱ
KCl + MgCl ₂	7.20	ⁿ			

^a Robinson and Lim.¹⁹ ^b Long et al.²⁰ ^c Yao et al.²¹ ^d Covington et al.²² ^e Bezboruah et al.²³ ^f Robinson and Bower.²⁴ ^g Robinson and Bower.²⁵ ^h Platford.²⁶ ⁱ Robinson et al.²⁷ ^j Wu et al.⁸ ^k Wu et al.⁹ ^l Robinson.²⁸ ^m Robinson and Covington.²⁹ ⁿ Padova and Saad.³⁰

**Figure 1.** Comparison of the predicted and experimental osmotic coefficients for aqueous NaCl(1) + LiCl(2) solution: symbols, experimental data;¹⁹ solid lines, prediction.

densities for multiple-salt solutions in water at 25 °C using the parameters obtained from single-salt solutions³ without any adjustable parameters.

3.1. Osmotic Coefficient of Two-Salt Solutions. The average relative deviation (ARD) of the model predictions of the osmotic coefficients in the aqueous two-salt solutions is summarized in Table 1. Examples of typical results are shown in Figures 1–3. Figure 1 shows an example of the model prediction for a solution containing two 1-1 type salts with common ions, that is, for the aqueous LiCl + NaCl solution, where C^m is the total molality of salts ($= \sum C_i^m$). Figure 2 shows an example of the prediction for a solution containing 1-1 and 2-1 type salts, that is, for the aqueous LiCl + CaCl₂ solution, where $\nu C^m = \sum \nu_i C_i^m$, ν_i is the number of moles of ions per mole of salt i , and ϕ is the osmotic coefficient of solution. Figure 3 shows an example for a solution containing two 2-1 type salts, that is, for the aqueous MgCl₂ + Mg(NO₃)₂ solution. As shown in Figures 1–3, the predicted

**Figure 2.** Comparison of the predicted and experimental osmotic coefficients for aqueous LiCl(1) + CaCl₂(2) solution: symbols, experimental data;²⁰ solid lines, prediction.**Figure 3.** Comparison of the predicted and experimental osmotic coefficients for aqueous MgCl₂(1) + Mg(NO₃)₂(2) solution: symbols, experimental data;²⁶ solid lines, prediction.

data agree with the experimental data. Note that most of the experimental data for osmotic coefficients presented here were obtained using isopiestic technique, that is, $\nu C^m \phi = \nu' C'^m \phi' = \text{constant}$, where prime (') denotes the property of a reference salt.

In Table 1a, the ARD for each of the systems is typically less than 3%, except for the KCl + KBr solution, which is shown in Figure 4. Figure 4 illustrates that the experimental osmotic coefficient used for parameter fitting for aqueous KBr solution at the same ionic strength, for example, $\nu C^m \phi = 8.4930$ (shown as square at $x_2' = 1$), is much larger than that reported for this two-salt solution (triangle at $x_2' = 1$). Therefore, the discrepancies between model predictions and experimental data for this system are attributed to the inconsistency of the experimental data from different sources.

Table 1b shows the ARD for the solutions containing two different cations, one of which is K^+ ; Figures 5 and 6 show examples of the model predictions for NaBr + KBr and KCl + MgCl₂. The error increases with the total molality of the salts. We suspect that this is due to the peculiar parameters of K^+ .

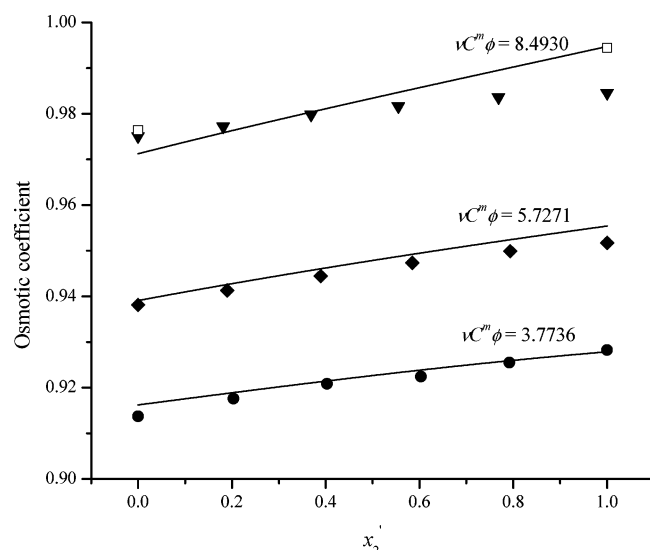


Figure 4. Comparison of the predicted and experimental osmotic coefficients for aqueous KCl(1) + KBr(2) solution: symbols, experimental data;²² solid lines, prediction.

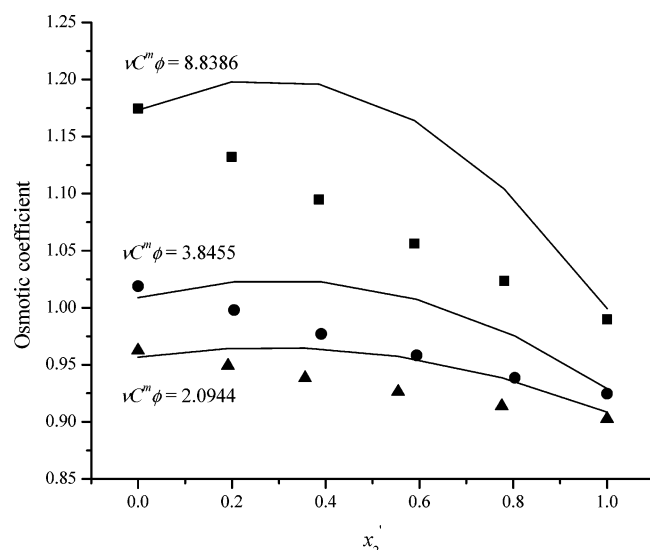


Figure 5. Comparison of the predicted and experimental osmotic coefficients for aqueous NaBr(1) + KBr(2) solution: symbols, experimental data;²² solid lines, prediction.

K^+ has much lower ion energy than the other ions³; (u/k) is 123 K, while those for the other cations are always larger than 1000 K, for example, 1485 K for Na^+ and 2598 K for Li^+ . However peculiar, this is consistent with the (u/k) values obtained by others, for example, for SAFT-VRE⁶ and PC-SAFT.⁷ Therefore, two-salt solutions containing K^+ and another cation pose a special challenge, which is being addressed in our current work.

Since our goal is to study the properties of brine and the amount of Na^+ , Mg^{2+} , Cl^- , and SO_4^{2-} in brine are the largest, first we test our model on the experimental data obtained for two-salt solutions containing these ions.

There are six two-salt solutions composed of Na^+ , Mg^{2+} , Cl^- , and SO_4^{2-} . Their osmotic coefficients have been measured by Wu et al.^{8,9} The predicted osmotic coefficients for these solutions are shown in Figure 7 along with the experimental data, where the horizontal axis y_i is the ionic strength fraction of salt i given by

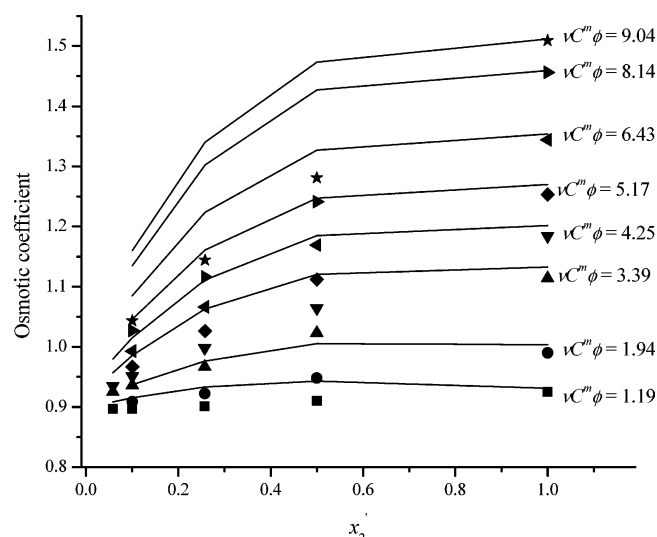


Figure 6. Comparison of the predicted and experimental osmotic coefficients for aqueous KCl(1) + $MgCl_2$ (2) solution: symbols, experimental data;³⁰ solid lines, prediction.

$$y_i = \frac{I_i}{\sum_i I_i} \quad (7)$$

$$I_i = \frac{C_i^m}{2} (\nu_+ z_+^2 + \nu_- z_-^2)_i \quad (8)$$

where z is the valence of an ion and subscripts $+$ and $-$ denote cation and anion, respectively. Their corresponding ARD is listed in Table 1a for each solution. For the solutions with $MgSO_4$, the prediction quality is affected by the fit error for aqueous $MgSO_4$.³

3.2. Density and Vapor Pressure of Two-Salt Solutions.

Most of the densities reported to date are for solutions with K^+ and other cations that are not subject of this work. However, Zhang et al.¹⁰ measured the density of $NaCl$ + $CaCl_2$ solutions at different mole ratios of $NaCl$ to $CaCl_2$. The density predicted in this work is shown to agree with their experimental data in Figure 8.

Vapor pressure for the $NaCl$ + $CaCl_2$ solutions has also been measured.¹¹ The experimental and predicted vapor pressures are listed in Table 2. These predictions agree with the experimental data as well.

3.3. Solution Properties uniquely Depend on the Ion Composition, not the Salt Composition. Since the effective hydrated diameter is a salt property in this work, for a given ion composition, the question is if one can obtain a unique solution properties based on a nonunique set of salts and their composition giving the ion composition. For example, consider an aqueous solution of Na^+ , Mg^{2+} , Cl^- , and SO_4^{2-} with comparable mole ratios, as shown in Table 3. Table 4 shows two examples of salt composition that are consistent with the ion composition from Table 3. In case 1, the solution contains two salts, that is, $NaCl$ and $MgSO_4$, with solvent-free mole fractions of 0.6 and 0.4, respectively. In case 2, the solution contains three salts, that is, Na_2SO_4 , $MgSO_4$, and $MgCl_2$ with solvent-free mole fractions of 0.3, 0.1, and 0.3, respectively.

The osmotic coefficient (ϕ), the activity of water (a_w), and the density of the solution (ρ) for these two cases are predicted up to 5 molality of ionic strength, and the results are listed in Table 5. It turns out that the results predicted for these two cases

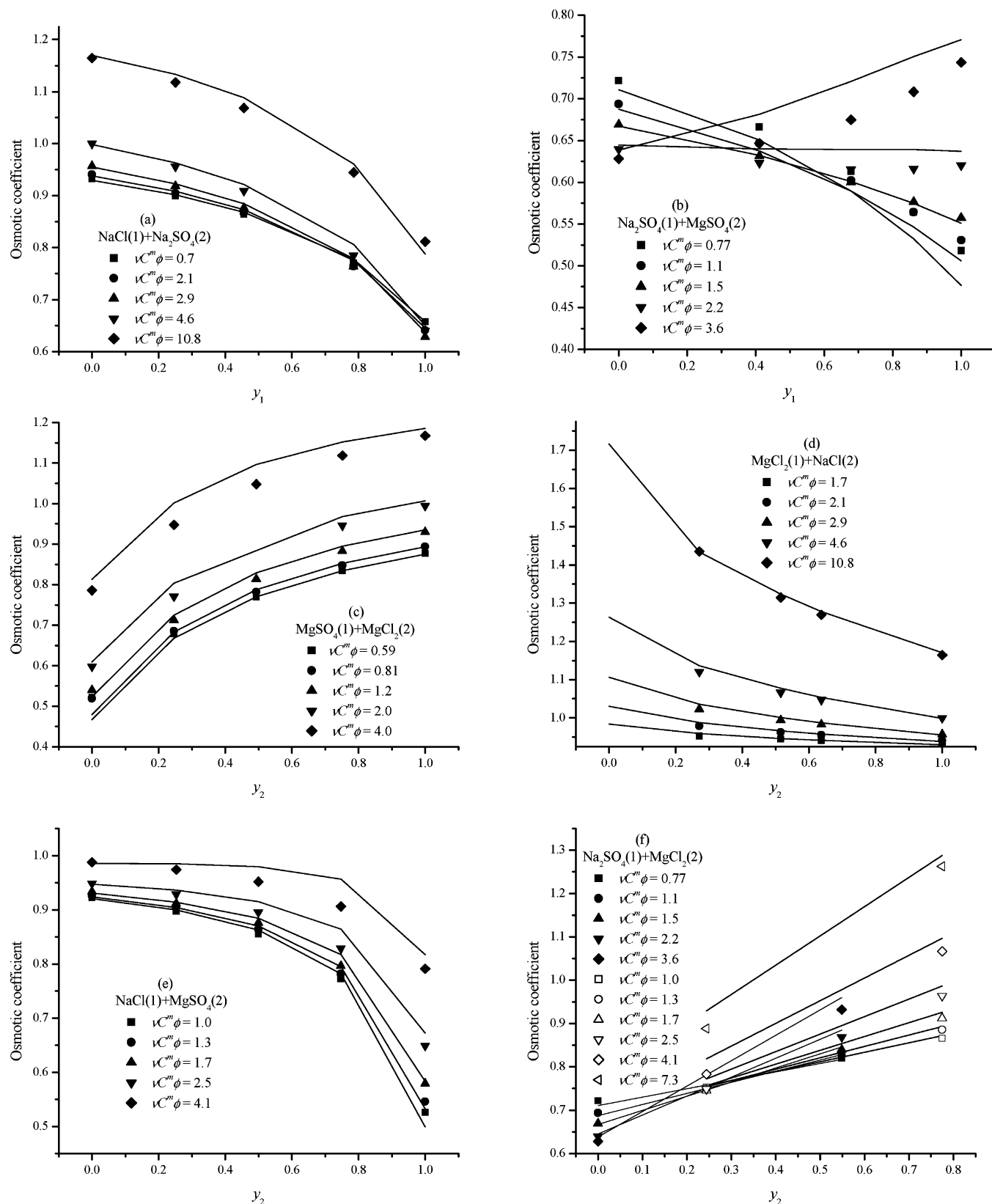


Figure 7. Comparison of the predicted and experimental osmotic coefficients for the following: (a) NaCl + Na₂SO₄ + H₂O, (b) Na₂SO₄ + MgSO₄ + H₂O, (c) MgSO₄ + MgCl₂ + H₂O, (d) MgCl₂ + NaCl + H₂O, (e) NaCl + MgSO₄ + H₂O, (f) Na₂SO₄ + MgCl₂ + H₂O. Symbols, experimental data;^{8,9} solid lines, prediction.

are essentially the same. The results imply that, for a given ion composition, the calculated results do not depend on the choice of salt composition. Hence, the type of salt and their concentration in the electrolyte solution can be selected arbitrarily as long as they are consistent with the ion composition.

3.4. Properties of Brine/Seawater. The composition and the number of brine components may be different from one place to another, but the main ions are Na⁺, K⁺, Ca²⁺, Mg²⁺, Cl⁻, and SO₄²⁻. A typical composition is listed in Table 6.¹²

For this typical brine listed in Table 6, two cases are

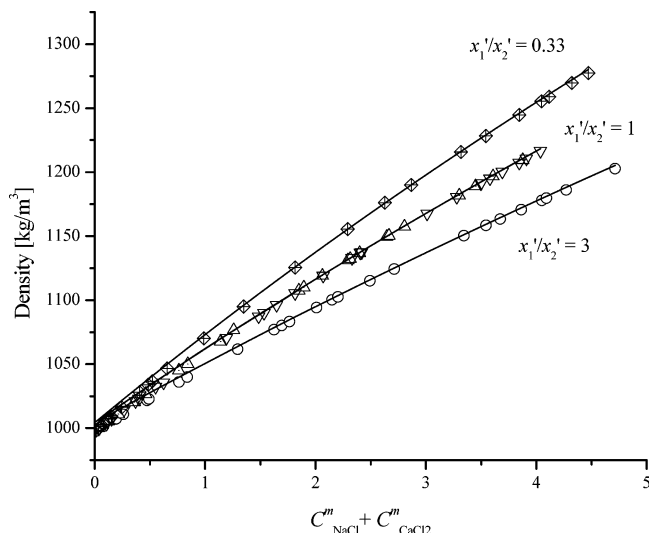


Figure 8. Comparison of the predicted and experimental densities for aqueous NaCl(1) + CaCl₂(2) solution: symbols, experimental data;¹⁰ solid lines, prediction.

TABLE 2: Comparison of the Predicted and Experimental Vapor Pressures (P)¹¹ for Aqueous NaCl + CaCl₂ Solutions

C^m_{NaCl} mol/kgH ₂ O	$C^m_{\text{CaCl}_2}$ mol/kgH ₂ O	P_{exp} kPa	P_{cal} kPa	$ P_{\text{exp}} - P_{\text{cal}} $ kPa
1.023	0.59	2.920	2.958	0.038
1.391	0.455	2.893	2.940	0.047
1.423	0.572	2.840	2.911	0.071
1.536	0.822	2.773	2.835	0.062

TABLE 3: Mole Ratio for Aqueous Na⁺, Mg²⁺, Cl⁻, and SO₄²⁻ Solutions

ion	Na	Mg	Cl	SO ₄
moles	0.6	0.4	0.6	0.4

TABLE 4: Salt Mole Ratio for Aqueous Na⁺, Mg²⁺, Cl⁻, and SO₄²⁻ Solutions

case 1		case 2	
salt	moles	salt	moles
NaCl	0.6	Na ₂ SO ₄	0.3
MgSO ₄	0.4	MgSO ₄	0.1
		MgCl ₂	0.3

TABLE 5: Prediction of These Two Cases with SAFT2

I mol/kgH ₂ O	case 1			case 2		
	ϕ	a_w	ρ (kg/m ³)	ϕ	a_w	ρ (kg/m ³)
0.01	0.9048	0.9997	1004.9	0.9046	0.9997	1004.9
0.05	0.8406	0.9985	1007.6	0.8401	0.9985	1007.6
0.1	0.8127	0.9971	1010.9	0.8120	0.9971	1010.9
0.5	0.7842	0.9860	1037.6	0.7829	0.9860	1037.6
1	0.8177	0.9710	1070.3	0.8161	0.9710	1070.3
2	0.9282	0.9353	1133.9	0.9264	0.9354	1133.9
3	1.0642	0.8913	1195.3	1.0624	0.8915	1195.3
4	1.2140	0.8395	1254.5	1.2122	0.8397	1254.5
5	1.3718	0.7810	1311.8	1.3700	0.7813	1311.8

constructed, as shown in Table 7. The difference between the two cases is the cation paired with SO₄²⁻; case 1 is obtained when SO₄²⁻ is paired with Na⁺ and case 2 is obtained when SO₄²⁻ is paired with Mg²⁺. This will also serve as a second test on the conclusion obtained in the previous section.

Again, the calculated vapor pressures, osmotic coefficients, activities of water, and densities of brine in both cases are essentially the same. A sample of calculated osmotic coefficients

TABLE 6: Main Ions and Typical Composition of Brine

ion	Na ⁺	Mg ²⁺	Ca ²⁺	K ⁺	Cl ⁻	SO ₄ ²⁻
moles	0.4822	0.0553	0.0105	0.0094	0.5650	0.0291

TABLE 7: Salt-based Composition of Brine

case 1		case 2	
salt	moles	salt	moles
NaCl	0.4240	NaCl	0.4822
MgCl ₂	0.0553	MgCl ₂	0.0262
CaCl ₂	0.0105	CaCl ₂	0.0105
KCl	0.0094	KCl	0.0094
Na ₂ SO ₄	0.0291	MgSO ₄	0.0291

TABLE 8: Predicted Osmotic Coefficients and Densities of Brine

salinity	osmotic coefficient (ϕ)		density (ρ) (kg/m ³)	
	case 1	case 2	case 1	case 2
1	0.9498	0.9498	1004.9	1004.9
3	0.9282	0.9283	1006.2	1006.2
5	0.9179	0.9179	1007.5	1007.5
10	0.9063	0.9064	1010.8	1010.8
15	0.9019	0.9019	1014.1	1014.1
20	0.9007	0.9008	1017.4	1017.4
25	0.9015	0.9015	1020.8	1020.8
30	0.9035	0.9036	1024.2	1024.2
35	0.9063	0.9064	1027.6	1027.6
40	0.9098	0.9099	1031.0	1031.0
50	0.9186	0.9187	1038.0	1038.0
75	0.9472	0.9473	1055.8	1055.8
100	0.9832	0.9833	1074.3	1074.3
125	1.0255	1.0256	1093.6	1093.6
150	1.0738	1.0739	1113.6	1113.6
175	1.1281	1.1282	1134.5	1134.5
200	1.1883	1.1885	1156.3	1156.3
225	1.2548	1.2549	1179.0	1179.0
250	1.3274	1.3276	1202.8	1202.8
270	1.3902	1.3904	1222.6	1222.6

and densities at different salinities (= total mass of salts in 1 kg solution) is given in Table 8.

Since the calculated results of brine of the two cases considered are essentially the same, the comparison with the experimental data is performed for one of these two cases only (case 1, noted as model brine in Figure 9). For the vapor pressure and activity of water at different salinities, the prediction agrees well with the experimental data,^{13–16} as it does for the brine osmotic coefficients^{15–18} at ionic strengths I up to 7.4 molality, which exceeds that used for parameter fitting (up to 6 molality).

Since the molality of NaCl in brine and seawater is much higher than that of the other salts, the properties of brine have often been approximated with the properties of a corresponding NaCl solution (that is, an NaCl solution that has the same salt concentration but no other salts). To test this approximation, the predicted properties of a corresponding NaCl solution are also included in Figure 9, which illustrates the discrepancies between the brine and NaCl properties, for example, for the osmotic coefficients at low ionic strengths ($I = 0–1$) and for the brine density. The calculated density of the NaCl solution is much lower than the experimental brine density,^{13,14} which is well predicted by SAFT2 at different chlorinity (mass of chlorine in 1 kg solution).

For the record, the predicted data for the solutions containing K⁺ with other cations generally are less accurate. However, for brine, the concentration of K⁺ is much lower than those of the other ions. Therefore, the impact of the less accurate representation of K⁺ on the brine properties is negligible.

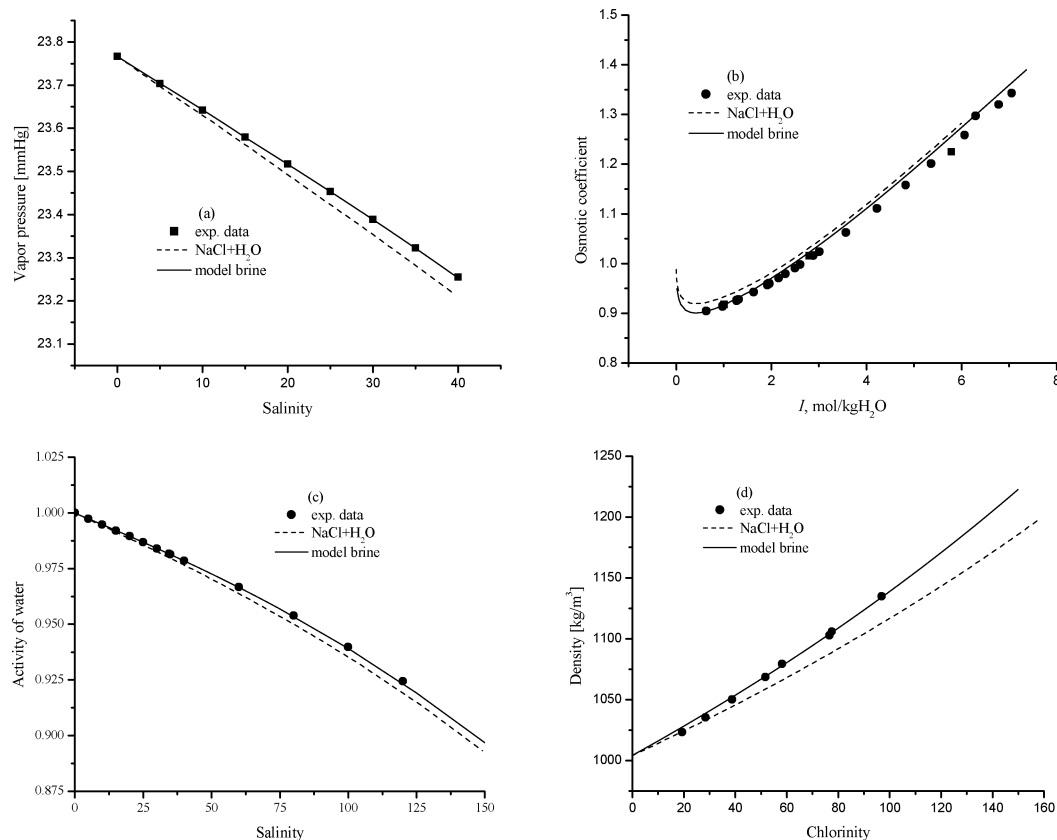


Figure 9. Comparison of the predicted and experimental properties of brine:^{13–18} (a) vapor pressure, (b) osmotic coefficient, (c) activity of water, and (d) density.

4. Conclusions

SAFT2 is found to represent the properties of aqueous multiple-salt solutions, such as brine and seawater. The osmotic coefficients, densities, and vapor pressures are predicted using the salt parameters previously obtained for single-salt solutions, without any additional parameters. For a given ion composition of brine, the predicted osmotic coefficients, activity of water, and density are found to agree with the experimental data. The choice of salt type and composition for a given ion composition does not affect the brine properties as long as it is consistent with its ion composition.

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Nomenclature

\tilde{a} = dimensionless residual molar Helmholtz energy
 a_w = water activity
 C_i^m = molality of salt i , mol/kgH₂O
 C_{ji}^m = molality of ion j in salt i , mol/kgH₂O
 d = effective salt hydrated diameter, Å
 I = ionic strength, mol/kgH₂O
 k = Boltzmann constant
 P = pressure, kPa
 u = segment energy, K
 v = segment volume, cc/mol
 x' = salt mole fraction in a solvent-free basis
 y = ionic strength fraction
 z_{ji} = valence of ion j in salt i
 Greek letters
 α, β = segment types
 ϕ = osmotic coefficient

λ = reduced well width of the square-well potential
 ν_i = the number of moles of ions per mole of salt i
 ρ = solution density, kg/m³

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