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Densities of the $NaNO_3 + NaCl + H_2O$ and $NaNO_3 + KCl + H_2O$ Systems and Their Correlation with the Pitzer Model

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Densities for two ternary systems, $NaNO_3$ (1) + NaCl (2) + H_2O (3) and $NaNO_3$ (1) + KCl (2) + H_2O (3), have been measured at (20, 25, and 30) °C. The experimental study was carried out for four total ionic strength values, $I = I_1 + I_2 \approx (0.1, 1, 2, \text{and } 3)\ \text{mol}\cdot\text{kg}^{-1}$, and for each total ionic strength, the values of the ionic strength fraction of the NaCl or KCl were $y_2 \approx 0.2, 0.4, 0.6, \text{and } 0.8$. The parameters of the Pitzer model were fit to the experimental density values. Also, using this model, we determined the volume of mixing (ΔV_M) at constant ionic strength and at 25 °C, for the two ternary systems studied in the present work. In general, the values for ΔV_M are positive, which indicates that the mixed solutes possess an opposite structural form, resulting in an attraction with an increase in volume in every ternary system.

Introduction

Experimental density values of aqueous ternary solutions containing $NaNO_3$ + NaCl and $NaNO_3$ + KCl are scarce. Padova and Lewkowicz[1] carried out a study of volumetric properties at 25 °C for the first system in which they derived diverse equations to determine the mean apparent molal volume of the ternary system and the partial molal volume of the $NaNO_3$ and NaCl. Nevertheless, the experimental values of density are not given in this work. For the second ternary system, we could not find studies that collect data on density. Laliberté and Cooper[2] mention that despite the importance of density data of aqueous electrolyte solutions in engineering calculations, finding information on this property in the scientific literature can be a frustrating task.

Considering the importance of the density values in the design and control of chemical processes, we think that it is useful to contribute experimental information on these systems. Moreover, both ternary systems are of great industrial interest to us because they are present in the production of natural fertilizers from mineral deposits located in northern Chile.

The measurement of density of the systems $NaNO_3$ (1) + NaCl (2) + H_2O (3) and $NaNO_3$ (1) + KCl (2) + H_2O (3) was carried out at (20, 25, and 30) °C and at four total ionic strengths: $I \approx (0.1, 1, 2, \text{and } 3)\ \text{mol}\cdot\text{kg}^{-1}$. For each of these total ionic strengths, four ionic strength fractions were studied for NaCl or KCl: $y_2 \approx 0.2, 0.4, 0.6, \text{and } 0.8$. We correlated the density values by using the thermodynamic model of Pitzer.[3] Subsequently, the volume of mixing (ΔV_M) was determined at constant total ionic strength at 25 °C for each of the ternary systems.

Table 1. Densities for the $NaNO_3$ (1) + NaCl (2) + H_2O (3) Ternary System

			20 °C	25 °C	30 °C
I / $\text{mol}\cdot\text{kg}^{-1}$	m_2 / $\text{mol}\cdot\text{kg}^{-1}$	y_2	ρ / $\text{g}\cdot\text{cm}^{-3}$	ρ / $\text{g}\cdot\text{cm}^{-3}$	ρ / $\text{g}\cdot\text{cm}^{-3}$
0.1001	0.0201	0.2008	1.00359	1.00244	1.00097
0.1000	0.0400	0.4000	1.00330	1.00201	1.00065
0.1000	0.0600	0.6000	1.00301	1.00176	1.00031
0.1000	0.0800	0.8000	1.00270	1.00150	1.00002
0.9998	0.2003	0.2003	1.05023	1.04749	1.04538
0.9998	0.3998	0.3999	1.04735	1.04473	1.04271
1.0001	0.6000	0.5999	1.04453	1.04200	1.04005
1.0001	0.8001	0.8000	1.04160	1.03920	1.03728
1.9999	0.3996	0.1998	1.09551	1.09329	1.09092
1.9993	0.8001	0.4002	1.09019	1.08805	1.08578
2.0003	1.2002	0.6000	1.08485	1.08278	1.08059
1.9993	1.5986	0.7996	1.07951	1.07750	1.07541
2.9989	0.6009	0.2004	1.13720	1.13459	1.13238
2.9989	1.1998	0.4001	1.13000	1.12754	1.12499
2.9995	1.7992	0.5998	1.12269	1.12037	1.11792
2.9999	2.3994	0.7998	1.11520	1.11302	1.11067

Experimental Section

All reagents used in this research were of analytical grade (Merck) and were directly used without further purification (sodium nitrate ≥ 99.5 %, sodium chloride ≥ 99.5 %, and potassium chloride ≥ 99.5 %). Before their use, all reagents were dried in an oven for 24 h at 110 °C. Distilled deionized water was used in all procedures. The solutions were prepared by mass by the use of an analytical balance with a precision of $\pm 1\cdot10^{-4}$ g (Denver Instrument, Model AA-200). The density of the solutions was measured in triplicate with a Mettler Toledo DE-50 vibrating tube densimeter; an uncertainty of less than $\pm 5\cdot10^{-5}\ \text{g}\cdot\text{cm}^{-3}$ was always obtained. We calibrated the densimeter using air and distilled deionized water as a reference substance prior to the initiation of each run of measurements at a given temperature. The densimeter had self-contained Peltier systems for temperature control (to ± 0.1 °C). The time to reach the temperature stability was 600 s.

Thermodynamic Framework

For a mixture of aqueous electrolytes at a fixed temperature, the equation for density is

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$$\rho = \frac{1000 + \sum_{\mathrm{j}} m_{\mathrm{j}} M_{\mathrm{j}}}{1000/\rho_0 + \sum_{\mathrm{j}} m_{\mathrm{j}} \phi^0_{V\mathrm{j}} + V^{\mathrm{ex}}} \tag{1}$$

where m_{j}, M_{j}, and $\phi^0_{V\mathrm{j}}$ are the molality, the molecular weight, and the apparent molar volume at infinite dilution of the electrolyte j in the mixture, respectively. ρ_0 is the water density and V^{ex} represents the excess volume of the mixture. The property V^{ex} is determined with the Pitzer model, whose general equation is mentioned elsewhere.[4,5] When the general equation of the excess volume to the $NaNO_3$ + KCl + H_2O system is applied, the following expression is obtained

$$\begin{aligned} V^{\mathrm{ex}} = {} & A_V\left(\frac{I}{b}\right)\ln(1 + b\sqrt{I}) + 2RT\{m_{\mathrm{Na}} m_{\mathrm{Cl}}[B^V_{\mathrm{NaCl}} + \\ & (m_{\mathrm{Na}} + m_{\mathrm{K}})C^V_{\mathrm{NaCl}}] + m_{\mathrm{Na}} m_{\mathrm{NO_3}}[B^V_{\mathrm{NaNO_3}} + (m_{\mathrm{Na}} + \\ & m_{\mathrm{K}})C^V_{\mathrm{NaNO_3}}] + m_{\mathrm{K}} m_{\mathrm{Cl}}[B^V_{\mathrm{KCl}} + (m_{\mathrm{Na}} + m_{\mathrm{K}})C^V_{\mathrm{KCl}}] + \\ & m_{\mathrm{K}} m_{\mathrm{NO_3}}[B^V_{\mathrm{KNO_3}} + (m_{\mathrm{Na}} + m_{\mathrm{K}})C^V_{\mathrm{KNO_3}}]\} + \\ & RT[m_{\mathrm{Na}} m_{\mathrm{K}}(2\theta^V_{\mathrm{NaK}} + m_{\mathrm{Cl}}\psi^V_{\mathrm{NaKCl}} + m_{\mathrm{NO_3}}\psi^V_{\mathrm{NaKNO_3}}) + \\ & m_{\mathrm{Cl}} m_{\mathrm{NO_3}}(2\theta^V_{\mathrm{ClNO_3}} + m_{\mathrm{Na}}\psi^V_{\mathrm{NaClNO_3}} + m_{\mathrm{K}}\psi^V_{\mathrm{KClNO_3}})] \end{aligned} \tag{2}$$

with

$$B^V_{\mathrm{j}} = \beta^{(0)V}_{\mathrm{j}} + \beta^{(1)V}_{\mathrm{j}} g(2\sqrt{I}) \tag{3}$$

$$g(2\sqrt{I}) = \frac{1}{2I}[1 - (1 + 2\sqrt{I})\mathrm{e}^{-2\sqrt{I}}] \tag{4}$$

$$I = \frac{1}{2}(m_{\mathrm{Na}} + m_{\mathrm{K}} + m_{\mathrm{Cl}} + m_{\mathrm{NO_3}}) \tag{5}$$

where the symbols have the usual meanings.[4,5] Equations 2, 3, 4, and 5 are also applicable to the $NaNO_3$ + NaCl + H_2O ternary system, making $m_{\mathrm{K}} = 0$.

The volume of mixing ΔV_{M} at constant total ionic strength is given by[6]

$$\Delta V_{\mathrm{M}} = V^{\mathrm{ex}}(\mathrm{mixture}) - [y_2 V^{\mathrm{ex}}(2) + (1 - y_2)V^{\mathrm{ex}}(1)] \tag{6}$$

where y_2 is the ionic strength fraction of electrolyte 2 in the ternary system and is defined by $y_2 = I_2/(I_1 + I_2)$.

For the ternary system without a common ion, $NaNO_3$ (1) + KCl (2) + H_2O (3), the substitution of the various terms in eq 6 yields

$$\begin{aligned} \frac{\Delta V_{\mathrm{M}}}{I^2 RT} = {} & y_2(1 - y_2)[2(B^V_{\mathrm{NaCl}} + IC^V_{\mathrm{NaCl}} + B^V_{\mathrm{KNO_3}} + \\ & IC^V_{\mathrm{KNO_3}} - B^V_{\mathrm{NaNO_3}} - IC^V_{\mathrm{NaNO_3}} - B^V_{\mathrm{KCl}} - IC^V_{\mathrm{KCl}}) + \\ & 2(\theta^V_{\mathrm{NaK}} + \theta^V_{\mathrm{ClNO_3}}) + y_2 I(\psi^V_{\mathrm{NaKCl}} + \psi^V_{\mathrm{KClNO_3}}) + \\ & (1 - y_2)I(\psi^V_{\mathrm{NaKNO_3}} + \psi^V_{\mathrm{NaClNO_3}})] \end{aligned} \tag{7}$$

Because the volume of mixing is commonly expressed as

$$\Delta V_{\mathrm{M}} = y_2(1 - y_2)I^2 RT[v_0 + (1 - 2y_2)v_1] \tag{8}$$

it is convenient to rewrite the above equation in Pitzer formalism for v_0 and v_1, giving

$$\begin{aligned} v_0 = {} & 2[B^V_{\mathrm{NaCl}} + B^V_{\mathrm{KNO_3}} - B^V_{\mathrm{NaNO_3}} - B^V_{\mathrm{KCl}} + \theta^V_{\mathrm{NaK}} + \theta^V_{\mathrm{ClNO_3}}] + \\ & 2I\left[C^V_{\mathrm{NaCl}} + C^V_{\mathrm{KNO_3}} - C^V_{\mathrm{NaNO_3}} - C^V_{\mathrm{KCl}} + \frac{1}{2}(\psi^V_{\mathrm{NaKCl}} + \right. \\ & \left. \psi^V_{\mathrm{KClNO_3}} + \psi^V_{\mathrm{NaKNO_3}} + \psi^V_{\mathrm{NaClNO_3}})\right] \end{aligned} \tag{9}$$

$$v_1 = \frac{I}{2}[\psi^V_{\mathrm{NaKNO_3}} + \psi^V_{\mathrm{NaClNO_3}} - \psi^V_{\mathrm{NaKCl}} - \psi^V_{\mathrm{KClNO_3}}] \tag{10}$$

Proceeding in a similar fashion with the ternary system with a common ion, $NaNO_3$ (1) + NaCl (2) + H_2O (3), less complex equations are obtained

$$v_0 = 2\theta^V_{\mathrm{ClNO_3}} + I\psi^V_{\mathrm{NaClNO_3}} \tag{11}$$

$$v_1 = 0 \tag{12}$$

Results and Discussion

Tables 1 and 2 show the experimental values of density for the two ternary systems studied in the present work. In these tables, I represents the total ionic strength and m_2 represents the molality of the second electrolyte, NaCl or KCl. Because both ternary systems are constituted by 1:1 electrolytes, $I = m_1 + m_2$ and $y_2 = m_2/(m_1 + m_2)$.

From these Tables, it is clear that (for a constant I value) the densities of both ternary systems decrease as temperature increases. This is due to the expansion of the volume of the solution, which in turn is affected by the temperature increase. It is also clear that for constant I and temperature values the density of both ternary systems decreases with the increase

Table 2. Densities for the $NaNO_3$ (1) + KCl (2) + H_2O (3) Ternary System

I	m_2		20 °C	25 °C	30 °C
			ρ	ρ	ρ
$\mathrm{mol \cdot kg^{-1}}$	$\mathrm{mol \cdot kg^{-1}}$	y_2	$\mathrm{g \cdot cm^{-3}}$	$\mathrm{g \cdot cm^{-3}}$	$\mathrm{g \cdot cm^{-3}}$
0.0999	0.0200	0.2002	1.00370	1.00249	1.00106
0.1000	0.0400	0.4000	1.00352	1.00231	1.00087
0.1000	0.0600	0.6000	1.00332	1.00213	1.00066
0.1000	0.0800	0.8000	1.00314	1.00194	1.00050
0.9999	0.2000	0.2000	1.05027	1.04846	1.04652
1.0001	0.3999	0.3999	1.04836	1.04662	1.04475
0.9999	0.6001	0.6002	1.04646	1.04476	1.04294
0.9998	0.7997	0.7999	1.04464	1.04315	1.04135
1.9999	0.3998	0.1999	1.09743	1.09513	1.09310
2.0001	0.8000	0.4000	1.09371	1.09156	1.08959
2.0002	1.2000	0.5999	1.09031	1.08827	1.08615
1.9999	1.5999	0.8000	1.08680	1.08489	1.08289
3.0002	0.6006	0.2002	1.14051	1.13809	1.13679
2.9995	1.2000	0.4001	1.13544	1.13339	1.13222
2.9987	1.7992	0.6000	1.13059	1.12831	1.12592
2.9992	2.3984	0.7997	1.12548	1.12334	1.12110

Table 3. Volumetric Pitzer Parameters at Different Temperatures

parameter	20 °C	25 °C	30 °C
	NaCl		
ϕ^0_V ($\mathrm{cm^3 \cdot mol^{-1}}$)	16.131	16.620	17.009
$10^5(\beta^{(0)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	1.3769	1.2335	1.1024
$10^6(\beta^{(1)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	9.7694	4.3540	0.42863
$10^7(C^V)$ ($\mathrm{kg^2 \cdot (mol^2\ bar)^{-1}}$)	−7.3342	−6.5780	−5.8245
	KCl		
ϕ^0_V ($\mathrm{cm^3 \cdot mol^{-1}}$)	26.432	26.848	27.160
$10^5(\beta^{(0)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	1.3705	1.2793	1.1819
$10^6(\beta^{(1)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	15.828	8.9480	4.8048
$10^8(C^V)$ ($\mathrm{kg^2 \cdot (mol^2\ bar)^{-1}}$)	−8.3413	−7.1310	−2.5373
	$NaNO_3$		
ϕ^0_V ($\mathrm{cm^3 \cdot mol^{-1}}$)	26.614	27.668	28.667
$10^5(\beta^{(0)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	1.4271	1.4650	1.5343
$10^5(\beta^{(1)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	4.3739	1.0333	−2.3518
$10^7(C^V)$ ($\mathrm{kg^2 \cdot (mol^2\ bar)^{-1}}$)	−3.3654	−4.3421	−5.4967
	KNO_3		
ϕ^0_V ($\mathrm{cm^3 \cdot mol^{-1}}$)	37.404	38.173	38.870
$10^5(\beta^{(0)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	2.0403	2.1620	1.8002
$10^5(\beta^{(1)V})$ ($\mathrm{kg \cdot (mol\ bar)^{-1}}$)	3.1611	1.1867	0.64208
$10^6(C^V)$ ($\mathrm{kg^2 \cdot (mol^2\ bar)^{-1}}$)	−2.1232	−2.7043	−2.1155

Table 4. Mixing Parameters at Different Temperatures

10^6 (θ_{ClNO_3})	10^6 (Ψ_{NaClNO_3})	SD_1	10^5 (Ψ_{NaK})	10^5 (Ψ_{NaKNO_3})	10^6 (Ψ_{NaKCl})	10^6 (Ψ_{KClNO_3})	SD_2
			20 °C				
0	0	0.00059	0	0	0	0	0.00029
−16.242	16.773	0.00042	4.3599	−3.0542	−9.6415	−9.6415	0.00007
7.3986	0	0.00046	1.9958	−1.3769	−19.283	0	0.00007
			25 °C				
0	0	0.00045	0	0	0	0	0.00023
9.5446	−1.348	0.00020	1.7946	−1.6089	−9.5649	−9.5649	0.00008
7.6574	0	0.00020	1.9845	−1.7446	−19.138	0	0.00008
			30 °C				
0	0	0.00038	0	0	0	0	0.00054
14.166	−5.8007	0.00014	1.2070	−2.8486	−5.9566	−5.9566	0.00016
6.6212	0	0.00014	1.9615	−3.4287	−11.913	0	0.00016

in y_2, which tends to reach the value of density of the second electrolyte ($y_2 = 1$). This behavior occurs because the density value of the NaCl + H_2O or KCl + H_2O system is lower than that of the $NaNO_3$ + H_2O system within the concentration interval evaluated in this work. A comparison of values of density for these three binary systems confirms this.[7−9]

The experimental data of Tables 1 and 2 were correlated by the use of the Pitzer model. However, to carry out this correlation, previous knowledge of the ϕ_V^0, $\beta^{(0)V}$, $\beta^{(1)V}$, and C^V parameters for each electrolyte is necessary. For the ternary system with a common ion, it is necessary to know the parameters of $NaNO_3$ and NaCl, whereas for the system without a common ion in addition to the two previous ions, it is necessary to include KCl and KNO_3.

Krumgalz et al.[10] reports the values of Pitzer parameters according to temperature for the electrolytes NaCl and KCl. Nevertheless, for the electrolytes $NaNO_3$ and KNO_3, values[11] of only 25 °C are reported. To safeguard this problem, we have used information on density reported by Isono[9] to obtain the parameters of both nitrates at (20, 25, and 30) °C. In the data fitting process, the expression of the apparent molal volume was used for binary systems,[10] and the following standard deviations were obtained for $NaNO_3$: (0.13, 0.17, and 0.19) $\text{cm}^3\cdot\text{mol}^{-1}$ at (20, 25, and 30) °C, respectively. For the same temperatures, the standard deviations for the KNO_3 were (0.07, 0.06, and 0.08) $\text{cm}^3\cdot\text{mol}^{-1}$, respectively. The values of the ϕ_V^0, $\beta^{(0)V}$, $\beta^{(1)V}$, and C^V parameters are shown in Table 3. These parameters were used in the correlation of the two ternary systems.

According to eq 2, the mixing parameters to be adjusted for the system without a common ion are θ_{NaK}^V, θClNO3^V, ψ_{NaKCl}^V, $\psi_{\text{NaKNO}_3}^V$, $\psi_{\text{NaClNO}_3}^V$, and $\psi_{\text{KClNO}_3}^V$. However, for the system with a common ion, only two of these parameters are necessary ($\theta_{\text{ClNO}_3}^V$ and $\psi_{\text{NaClNO}_3}^V$). First, we correlated the system with a common ion ($NaNO_3$ + NaCl + H_2O). Once the two mixing parameters for this system were obtained, the system $NaNO_3$ + KCl + H_2O was correlated, and the four remaining mixing parameters were obtained.

The correlation of the density was carried out by the use of eq 1, for which V^{ex} is derived from eq 2. The values of density of the water and of A_V (Debye−Hückel constant) used in the fit were obtained by the use of the equations developed by Kell[12] and Krumgalz et al.,[10] respectively. The objective function (OF) to be

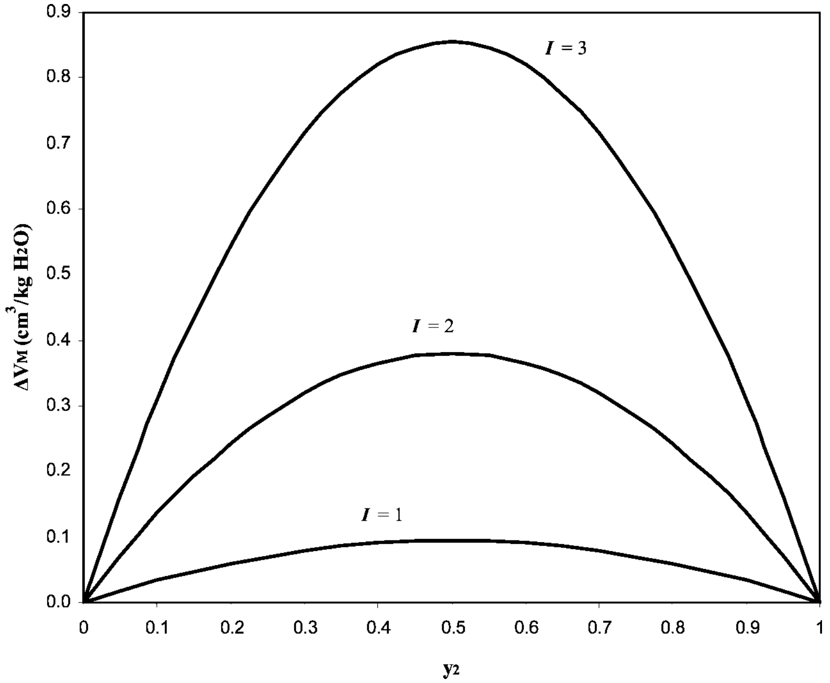

Figure 1. Volume of mixing of the $NaNO_3$ + NaCl + H_2O system at 25 °C.

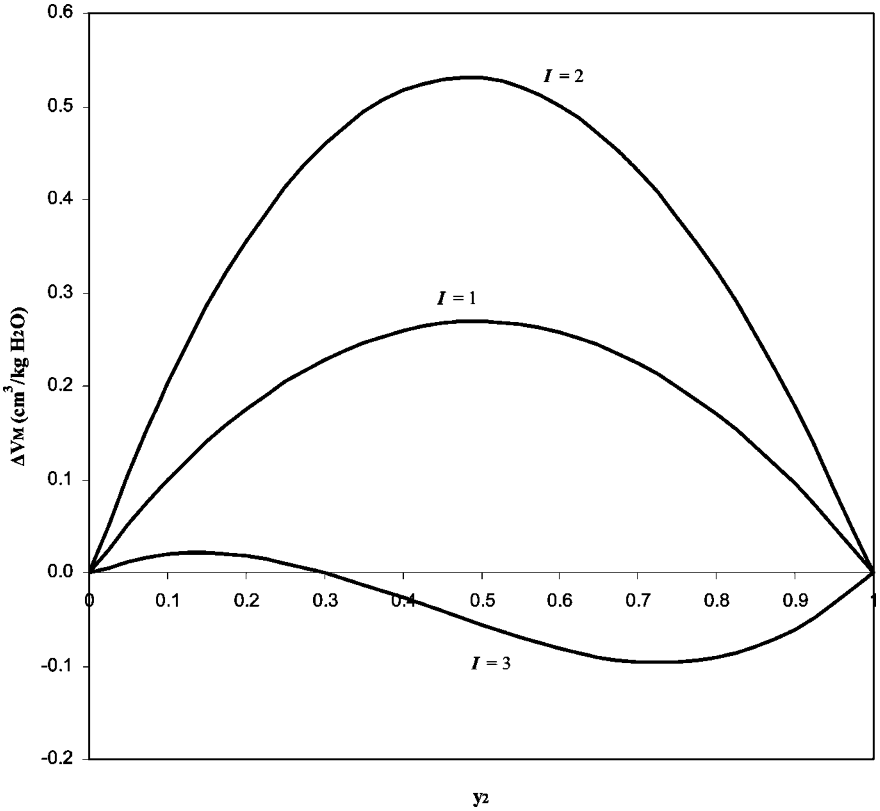

Figure 2. Volume of mixing of the $NaNO_3 + KCl + H_2O$ system at 25 °C.

minimized in the estimate of the parameters of the mixture was the following

$$\mathrm{OF} = \sum_{i=1}^{n} (\rho_{\mathrm{exptl}} - \rho_{\mathrm{calcd}})^2 \qquad (13)$$

Table 4 gives the results that are obtained when the density data are adjusted to the Pitzer model. In this table, SD_1 and SD_2 are the standard deviations of the systems $NaNO_3 + NaCl + H_2O$ and $NaNO_3 + KCl + H_2O$, respectively. In general, the quality of the fit is better when the mixing parameters are considered (option 2) than when they are not considered at all (option 1). Nevertheless, option 3 (with $\psi^V_{\mathrm{NaClNO_3}} = \psi^V_{\mathrm{NaKCl}} = 0$) is better because by using a minor number of parameters, the values of SD_1 and SD_2 are maintained as option 2. If a new fit was undertaken that discarded another ternary mixing parameter (ψ^V), then an increase in SD_2 was observed.

By using eqs 8, 9, 10, 11, and 12, we obtained the values of volume of mixing at 25 °C for the two systems studied in this work. Figures 1 and 2 show the values of ΔV_M versus y_2 for the systems $NaNO_3 + NaCl + H_2O$ and $NaNO_3 + KCl + H_2O$, respectively. In each graph, different curves correspond to the different I values. In Figure 1, the system with the common ion that presents positive values of the volume of mixing, ΔV_M, is observed in the entire interval of y_2. Also, the values of ΔV_M are more positive as I increases.

The fact that the ΔV_M value may be positive, according to eq 6, indicates that the excess volume of the two binary mixtures before mixing is less than that obtained after the mixing process. This indicates that the final ternary mixture reaches a less-compact structure, resulting in a volume expansion. The presence of two electrolytes probably does not greatly disturb the water structure because an overall decrease in the structure of water implies a breakage of hydrogen bonds, which contributes to a decrease in volume.[13] According to the structural hydration interaction model, the positive values of ΔV_M indicate that the two solutes that have been mixed in this system have different structural abilities for orientating molecules of water (one is a structure maker and the other is a structure breaker), resulting in an attraction with an increased volume.[14] Some studies show that sodium nitrate acts as a structure breaker;[15] therefore, it is probable that potassium chloride acts as structure maker in this ternary system.

The values of ΔV_M in the system without a common ion (Figure 2) show behavior similar to that of the previous example for $I = (1 \text{ and } 2)\ \mathrm{mol\cdot kg^{-1}}$; however, for $I = 3\ \mathrm{mol\cdot kg^{-1}}$, a different behavior is observed. For this total ionic strength, the values of ΔV_M tend to be negative for y_2 values greater than 0.3, which would indicate that starting from these conditions, both solutes tend to possess the same ability to orientate water molecules, resulting in a repulsion with a decrease in volume.[14] On the basis of what was previously stated, it is probable that the behavior of potassium chloride changed to a structure breaker.

Conclusions

This work provides reliable information on density for the systems $NaNO_3 + NaCl + H_2O$ and $NaNO_3 + KCl + H_2O$. This information is important in the design and control of chemical processes. The experimental data were correlated by the Pitzer model, and the values for volume of mixing (ΔV_M) were determined at 25 °C. In general, the values for ΔV_M are positive, which indicates that the mixed solutes possess an opposite structural form, resulting in an attraction with an increase in volume in every ternary system.

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