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Densities and Viscosities of Ternary Systems of NaCl−Sucrose−Water from 283.15 to 303.15 K

P. Bohuon,*,† M. Le Maguer,‡ and A. L. Raoult-Wack†

CIRAD-SAR, 73, rue J. F. Breton, BP 5035, 34030 Montpellier Cedex, France, and Department of Food Science, University of Guelph, Guelph, Ontario, Canada N1G 2W1

Densities and viscosities of ternary systems of NaCl−sucrose−water were measured in the range 283.1 to 303.1 K. NaCl and sucrose molalities ranged from 0 to 6 mol·kg^{-1} and 0 to 5.55 mol·kg^{-1}, respectively. For density, experimental data were fitted by a five-parameter correlation with a goodness of fit approximating experimental error. The presence of NaCl was found to be responsible for excess volume. For viscosity, a five-parameter polynomial correlation gave the dependence of dynamic viscosity on aqueous molalities and temperature, with a deviation less than 3.8%. There was a major contribution of interaction terms in the calculation of the ternary viscosity, particularly for the high viscosity level.

Introduction

Direct contacting of foodstuff with aqueous concentrated solutions has been widely used in the field of food engineering. Sodium chloride or sucrose solutions have generally been used. Such soaking operations include candying and semicandying, osmotic dehydration, brining and curing, immersion chilling, and freezing. A renewal of interest was recently raised in the field of soaking operations by the used of mixed blends (water−sucrose−NaCl) at low temperatures (below 303 K) either for vegetable (Islam and Flink, 1982; Bolin *et al.*, 1983; Jayaraman *et al.*, 1990) or animal tissues (Collignan and Raoult-Wack, 1992; Collignan and Raoult-Wack, 1994). In fact the use of mixed blends makes it possible to take benefit from respective advantages of each solute, and low temperatures are particularly interesting for processing fragile biological material (Raoult-Wack, 1994). However, further understanding of soaking operations with mixed blends needs to take into account the physical properties of water−NaCl−sucrose systems. Sucrose aqueous solutions have a Newtonian behavior, even for highly dry matter and low temperatures (Drago and Delavier, 1967). However, there is no available information on the behavior of such solutions when NaCl is added. In binary systems, empirical correlations were proposed to predict the dynamic viscosity of sucrose aqueous solution (Misra and Varshni, 1961; Maurandi, 1989; Eszterle, 1990; Nabetani *et al.*, 1992) or NaCl aqueous solution (Robinson and Stokes, 1959; Korosi and Fabuss, 1968; Sawamura *et al.*, 1992). Viscosity and densities of concentrated water−NaCl−sucrose have been little studied to date. Few data on viscosities (Mulcahy and Steel, 1985), densities (Sangster *et al.*, 1976; Mulcahy and Steel, 1985; Vitagliano *et al.*, 1986), and solute solubilities (West, 1933; Robinson *et al.*, 1970) in ternary solutions at 25 °C have been published. Some models, based on Eyring theory (Glasstone *et al.*, 1941) were proposed in order to evaluate the bulk viscosity of solvent blends, from the respective properties of each solvent in the pure state (Chandramouli and Laddha, 1962; Kalidas and Laddha, 1964; Prolongo *et al.*, 1984; Reid *et al.*, 1986). These models are usually cubic polynomial equations of composition for the ternary system, which take into account the respective fractions of total occurrences for different types of interactions. Excess volume can be predicted by thermodynamic models for blends of strong electrolyte solutions (Woolley and Burchfield, 1984) or by semiempirical theories for blends of solvent (Prolongo *et al.*, 1984). However, when electrolytes and nonelectrolytes are associated in an aqueous system, the use of such predictive models becomes impossible. The present work deals with the characterization of thermophysical properties (dynamic viscosity, density) of the concentrated ternary system water−NaCl−sucrose in the temperature range 283.15−303.15 K. The objectives were twofold: to acquire experimental data and to propose a practical set of empirical correlations between thermophysical properties and solution composition.

Experimental Section

Ternary solutions were prepared by completely dissolving NaCl (Prolabo product) and sucrose (Merck product) in demineralized water at room temperature. The three components of the mixed blends thus obtained were denoted as $i = 1$ (NaCl), $i = 2$ (sucrose), and $i = 3$ (water). Solutions were made up with preweighed amounts ($\pm 0.1 \times 10^{-3}$ g) of solutes and water. Concentrations, noted m_i, were expressed as aqueous molality (mol·kg^{-1}). The NaCl molality ranged from 0 to 6 mol·kg^{-1}, and the sucrose molality ranged from 0 to 5.55 mol·kg^{-1}. Density and viscosity measurements were made at temperatures ranging from 283.1 to 303.1 K at 5 K intervals, on solutions which had previously been stored at the measurement temperature for 24 h. The density, noted ρ (kg·m^{-3}), was measured by densimeters (M50 ISO 649 and ISO 387). The solution temperature was thermostated to within ±0.2 K. The maximum error of calibration was ±1 kg·m^{-3}, and the maximum deviation from the mean, ±0.6 kg·m^{-3}. The viscosities, noted η (Pa·s), were measured by a Rheomat RM 180 viscometer (Metler). The solution temperature was controlled within ±0.2 deg. The measurement system (standard DIN 53019) was constituted of a coaxial cylinder, and two spindle sizes (∅ 30×10^{-3} and 31.5×10^{-3} m) were required to cover the whole viscosity range. Measurements were made at six shearing speeds: 100, 140, 180, 220, 260, and 300 rad·s^{-1}. The results were expressed as means of these six viscosity values. The viscometer was tested at 25 °C with four Brookfield standard oils to within ±1%.

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* Tel: +(33) 04 67 61 57 33. Fax: +(33) 04 67 61 12 23. E-mail: bohuon@cirad.fr.

† CIRAD-SAR.

‡ University of Guelph.

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The maximum error of calibration was 1.5%, and the maximum deviation from the mean, 1%.

Correlations Used

Density. If V is the volume of solution (m^3) containing 1 kg of water, m_1 moles of NaCl, and m_2 moles of sucrose, then density ρ(kg·m^{-3}) may be easily calculated by

$$\rho = (1 + 0.05844 m_1 + 0.3423 m_2)/V \qquad (1)$$

The experimental volume of the mixed blend was fitted to a polynomial, as given by

$$V = (1/\rho_{H_2O}) + a_1 m_1 + a_2 m_2 + a_{11} m_1^2 + a_{22} m_2^2 + a_{12} m_1 m_2 \qquad (2)$$

where ρ_{H_2O} is the water density (kg·m^{-3}) and a_i and a_{ii} are parameters. Let us note that $(a_i + a_{ii} m_i)$ is the apparent molal volume (m^3·mol^{-1}) of solute i in aqueous solution and a_i is the apparent molal volume at infinite dilution.

Dynamic Viscosity. Experimental dynamic viscosities η (Pa·s) of the mixed blend were fitted to a polynomial, as given by

$$\log_{10}(\eta/\eta_{H_2O}) = b_1 m_1 + b_2 m_2 + b_{11} m_1^2 + b_{22} m_2^2 + b_{12} m_1 m_2 + b_{111} m_1^3 + b_{222} m_2^3 + b_{122} m_1 m_2^2 + b_{211} m_2 m_1^2 \qquad (3)$$

where η_{H_2O} is the water dynamic viscosities (Pa·s) and $\log_{10}$ is the decimal logarithm. The equation consisted of nine parameters, including six binary parameters (b_i, b_{ii}, b_{iii}) and three ternary parameters (b_{ij}, b_{ijj}). The use of a logarithmic form was suggested by that of empirical correlations available in the literature (Reid *et al.*, 1986). Let us note that when $m_1 = 0$ and $m_2 = 0$, eq 3 leads to $\eta = \eta_{H_2O}$, which is correct.

The identification procedure used to calculated the parameters of eqs 2 and 3 consisted of estimating parameters for the NaCl−water system from literature data at first (ASHRAE-Handbook, 1981; Weast, 1986), then parameters for the sucrose−water system from literature data (Baloh, 1967; Weast, 1986), and parameters for the ternary system at last. The temperature dependence for each viscosity parameter (b_{index}) was obtained by

$$b_{\text{index}} = \theta^0_{\text{index}} + \frac{\theta^1_{\text{index}}}{T} \qquad (4)$$

where T is the temperature (K). The coefficients (θ^1_{index}, θ^1_{index}) of eq 4 were estimated from parameter values of b_{index}, previously identified. At each stage of the procedures, the three or two coefficients were identified by least-squares regression. Fitting adequacy was estimated by the correlation coefficient, and by the root mean square relative deviation, noted RMSD, of the density or dynamic viscosity.

Table 1. Density of Water−NaCl−Sucrose Systems

m/mol·kg^{-1}		ρ/kg·m^{-3}		
sucrose	NaCl	T = 283.15 K	T = 293.15 K	T = 303.15 K
0.37	1.50	1102	1097	1094
0.37	4.50	1190	1185	1181
0.80	6.00	1255	1248	1243
1.39	0.40	1154	1149	1146
1.39	3.00	1218	1214	1210
1.39	5.60	1273	1269	1264
2.21	1.50	1231	1226	1223
2.21	4.50	1289	1284	1279
2.78	3.00	1284	1280	1275
3.34	1.50	1281	1277	1271
3.34	4.50	1330	1321	1316
4.16	0.40	1292	1287	1283
4.16	3.00	1331	1324	1317
4.16	5.60	1364	1358	1353
4.75	6.00	1385	1377	1372
5.18	1.50	1338	1332	1327
5.18	4.50	1376	1366	1363
5.55	0.75	1337	1332	1327
5.55	3.00	1365	1358	1353
5.55	5.25	1391	1384	1380
5.55	6.00	1396	1388	1385

Table 2. Coefficients for the Fit of V/m^3·kg$^{-1}_{\text{water}}$ = $(1/\rho_{H_2O}) + a_1 m_1 + a_2 m_2 + a_{11} m_1^2 + a_{22} m_2^2 + a_{12} m_1 m_2$

	T = 283.15 K	T = 293.15 K	T = 303.15 K
ρ_{H_2O}/kg·m^{-3}	999.73	998.23	995.68
	Regressions Coefficients		
$a_1 \times 10^5$	1.74	1.79	1.84
$a_{11} \times 10^7$	5.75	5.60	5.22
$a_2 \times 10^4$	2.10	2.11	2.13
$a_{22} \times 10^7$	7.81	6.70	6.24
$a_{12} \times 10^7$	4.70	7.50	5.00
	RMSD^a (%)		
NaCl + water	0.018	0.027	0.031
sucrose + water	0.032	0.032	0.033
NaCl + sucrose + water	0.152	0.114	0.137

[a] RMSD: root mean square relative deviation of density.

Table 3. Comparisons between Ideal and Experimental Density Values (E_{id}) and Experimental and Correlated Density Values (E_{cor})^a

m/mol·kg^{-1}		T = 283.15 K		T = 293.15 K		T = 303.15 K	
NaCl	sucrose	E_{id}	E_{cor}	E_{id}	E_{cor}	E_{id}	E_{cor}
0.40	4.16	−0.6	0.04	−0.3	−0.11	−0.2	0.13
0.40	1.39	−0.9	0.16	−0.5	−0.04	−0.5	0.10
1.50	5.18	−0.8	0.10	−0.5	0.01	−0.2	0.06
1.50	2.21	−1.2	−0.06	−1.0	−0.10	−0.9	0.05
3.00	5.55	−1.0	0.10	−0.6	0.02	−0.3	0.05
3.00	2.78	−1.5	−0.19	−1.2	−0.16	−1.0	−0.15
4.50	5.18	−1.4	−0.16	−0.7	−0.01	−0.6	0.10
4.50	2.21	−2.0	0.20	−1.8	0.08	−1.5	−0.14
6.00	5.55	−1.3	0.10	−0.8	0.19	−0.7	0.21
6.00	2.78	−2.4	0.09	−1.7	−0.05	−1.5	−0.05
mean		−1.3		−0.9		−0.7	

[a] $E_{id} = 100\,(\rho_{id} - \rho_{expt})/\rho_{expt}$. $E_{cor} = 100\,(\rho_{expt} - \rho_{correlated})/\rho_{expt}$.

Results and Discussion

Density. The density data of the NaCl−sucrose−water mixture are included in Table 1, and polynomial coefficients for a least-squares regression are presented Table 2. For all regressions, the R^2 value was very close to 1. The RMSD was lower than 0.03% and 0.03% for the NaCl and sucrose binary systems, respectively. For the ternary system, the RMSD ranged from 0.11 to 0.15% for the three temperatures. This deviation was mainly due to the experimental apparatus which allowed a ±0.1% accuracy only. Table 3 gives the relative difference (%) between the ideal and experimental density (E_{id}) and the relative difference between the experimental and correlated density (E_{cor}) by eqn 1. Whatever the composition and the temperature of the ternary system, its density was higher than that of the ideal system, which accounts for a negative excess volume. Moreover, deviation with respect to ideality was all the more marked as the NaCl concentration was higher and sucrose concentration and temperature were lower. This nonideal behavior of the ternary system led to experimental densities higher than ideal densities. The relative difference was less than 2.4%. Moreover, the

Table 4. Dynamic Viscosity of Water−NaCl−Sucrose Systems

m/mol·kg^{-1}		$10^3\eta$/Pa·s		
sucrose	NaCl	$T =$ 283.15 K	$T =$ 293.15 K	$T =$ 303.15 K
0.37	1.50	2.0 ± 0.5	1.7 ± 0.2	1.2 ± 0.2
0.37	4.50	2.9 ± 0.3	2.4 ± 0.1	1.9 ± 0.3
0.80	6.00	6.1 ± 0.4	4.3 ± 0.2	3.1 ± 0.4
1.39	0.40	5.3 ± 0.4	3.7 ± 0.3	2.8 ± 0.3
1.39	3.00	6.9 ± 0.5	6.1 ± 0.4	3.6 ± 0.2
1.39	5.60	10.0 ± 0.4	7.5 ± 0.4	4.9 ± 0.3
2.21	1.50	14.7 ± 0.5	9.1 ± 0.3	6.3 ± 0.4
2.21	4.50	20.9 ± 0.3	13.4 ± 0.3	9.7 ± 0.1
2.78	3.00	30.1 ± 0.3	19.0 ± 0.1	13.8 ± 0.4
3.34	1.50	46.1 ± 0.3	27.4 ± 0.4	18.0 ± 0.1
3.34	4.50	67.7 ± 0.7	38.3 ± 0.4	23.2 ± 0.4
4.16	0.40	95.3 ± 0.6	52.0 ± 0.1	30.8 ± 0.4
4.16	3.00	126.6 ± 0.7	67.8 ± 0.4	40.9 ± 0.5
4.16	5.60	185.2 ± 0.9	93.9 ± 0.3	52.8 ± 0.4
4.75	6.00	350.0 ± 1.4	170.1 ± 0.4	88.2 ± 0.6
5.18	1.50	302.2 ± 1.5	146.8 ± 0.4	81.0 ± 0.5
5.18	4.50	451.5 ± 2.3	210.1 ± 0.3	114.5 ± 0.5
5.55	0.75	394.0 ± 1.7	197.3 ± 0.5	104.3 ± 0.5
5.55	3.00	542.4 ± 2.2	258.4 ± 0.8	122.3 ± 0.4
5.55	5.25	729.3 ± 2.5	340.0 ± 1.1	166.8 ± 0.7
5.55	6.00	796.2 ± 2.6	347.9 ± 1.7	162.9 ± 0.3

Table 5. Coefficients for the Fit of $\log_{10}(\eta/\eta_{H_2O}) = b_1m_1 + b_2m_2 + b_{11}m_1^2 + b_{12}m_1m_2 + b_{211}m_2m_1^2$

	$T =$ 283.15 K	$T =$ 293.15 K	$T =$ 298.15 K	$T =$ 303.15 K
$10^3\eta_{H_2O}$/Pa·s	1.30	1.00	0.89	0.79
	Regressions Coefficients			
$b_1 \times 10^2$	3.080	3.584	3.785	4.025
$b_{11} \times 10^2$	2.968	2.219	1.701	1.076
b_2	0.4381	0.4024	0.3857	0.3713
$b_{12} \times 10^3$	6.031	4.823		4.459
$b_{211} \times 10^4$	−6.684	−7.201		−7.631
	RMSD (%)			
NaCl + water	0.08	0.22	0.22	0.02
sucrose + water	1.23	0.53	0.67	0.85
NaCl + sucrose + water	2.90	3.11		3.80

[a] RMSD: root mean square relative deviation of dynamic viscosity.

relative deviation between experimental and correlated densities was not greater than ± 0.2%. Hence, taking into account excess volume led to accurate prediction, with respect to experimental error.

Viscosity. The viscosity data of the NaCl−sucrose−water mixture are included in Table 4, and polynomial coefficients for a least-squares regression are presented Table 5. For all regressions, the R^2 value was very close to 1. The RMSD was lower than 0.22% and 1.23% for the NaCl and sucrose binary systems, respectively. In the case of salt, cubic coefficients were nonsignificant at 99%, hence $b_{111} = 0$. Let us point out that the other correlation given by the literature generally uses a cubic polynomial form (Djelveh *et al.*, 1988; Sawamura *et al.*, 1992). In the case of sucrose, quadratic and cubic polynomial coefficients were nonsignificant at 99% ($b_{22} = 0$; $b_{22} = 0$), then eq 3 could be reduced to a linear form which is well-known for sucrose (Eszterle, 1990). Figure 1 gives simultaneously the literature data and predicted dynamic viscosity of the NaCl and sucrose binary systems, as a function of molality, for different temperatures. For NaCl binary systems, a 10 K temperature decrease led to an increase in dynamic viscosity by a factor ranging from 1.23 to 1.33. For sucrose binary systems, a 10 K temperature decrease led to an increase in dynamic viscosity by a factor ranging from 1.23 to 2.1.

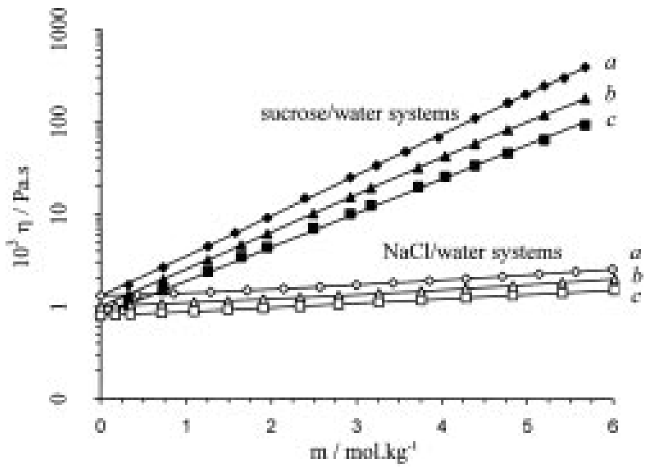

Figure 1. Dynamic viscosity as a function of molality for the binary NaCl and sucrose system at 283.15 K (*a*), 293.15 K (*b*), and 303.15 K (*c*), as correlated by eq (3), and dynamic viscosity literature data: (◆, ■) Baloh (1967); (◇, □) ASHRAE-Handbook (1981); (▲, △) Weast (1986).

Table 6. Regression Coefficient of Temperature Correlation Eq 4

coefficient	θ^0	θ^1	R^2 [a]
b_1	0.1707	−39.60	0.994
b_{11}	−0.0240	7.66	0.970
b_2	−0.5932	291.87	0.997
b_{12}	−0.0182	6.81	0.952
b_{211}	−0.0021	0.39	0.994

[a] R^2: least-squares correlation coefficient.

Viscosity data showed that in the whole experimental concentration range, the viscosities of the ternary systems ranged from 41 to 796 × 10^{-3} Pa·s. The variation of the shearing speed during experimental measurements led to a viscosity relative variation lower than 1%. Therefore, in the whole experimental concentration range, the ternary systems proved to have a Newtonian rheological behavior. The RMSD value showed that the fitting adequacy was better at 283.15 K than at 303.15 K. However, in all cases, the RMSD for ternary systems was lower than 3.8%. Coefficients b_{122} were nonsignificant at 99%; so whatever the temperature, $b_{122} = 0$.

At 283.15 K, for a ternary system composed of 6 and 5.55 *m* NaCl and sucrose, respectively, eq 3 predicted $\eta = 806 \times 10^{-3}$ Pa·s, whereas eq 3 deprived of interaction terms predicted $\eta = 691 \times 10^{-3}$ Pa·s (relative difference = 14.3%). Therefore, there was a major contribution from interaction terms in the calculation of the dynamic ternary viscosity, particularly for the high viscosity level. At 283.15 K, for a binary sucrose system with 5.55 *m*, $\eta = 353 \times 10^{-3}$ Pa·s, whereas for a binary NaCl system with 6 *m*, $\eta = 2.56 \times 10^{-3}$ Pa·s. When 6 *m* NaCl was added to the sucrose binary system, the overall viscosity increased by 128%. This suggests some kind of attractive interaction existing between NaCl and sucrose, being responsible for the enhanced viscosity observed in this system.

Table 6 gives the temperature dependence of the regression coefficients of eq 3 according to eq 4. R^2 values were greater than 0.95, which showed that a linear relationship between coefficients and 1/*T* was adequate in all cases. In order to validate eq 4 for the interaction coefficient, coefficients of eq 3 were estimated at 288.15 and 298.15 K by eq 4. For eleven distinct solute molalities, Table 7 compares predicted and experimental dynamic viscosity values. Results showed that the mean relative deviation was lower than 3%, which validated the 1/*T* dependence

Table 7. Comparison of Correlated Eq 3 and Experimental Dynamic Viscosity Values at 288.15 and 298.15 K

m/mol·kg^{-1}		$10^3\eta$/Pa·s			
		T = 288.15 K		T = 298.15 K	
sucrose	NaCl	expt[a]	correl[b]	expt	correl
2.21	4.50	16.8	16.1	10.9	10.4
2.78	3.00	23.6	23.5	14.9	14.7
2.78	6.00	35.2	34.1	19.2	20.4
3.34	1.50	35.0	34.4	20.1	20.7
3.34	4.50	48.3	49.5	28.5	29.0
4.16	3.00	90.6	92.8	50.7	51.7
4.16	6.00	129.7	127.3	67.5	67.7
5.20	1.50	209.3	208.6	106.1	108.3
5.20	4.50	299.1	304.8	148.3	151.7
5.55	3.00	358.8	366.2	182.0	181.4
5.55	6.00	513.4	522.6	237.3	242.0

[a] Expt: experimental viscosity measurement. [b] Correl: correlated viscosity.

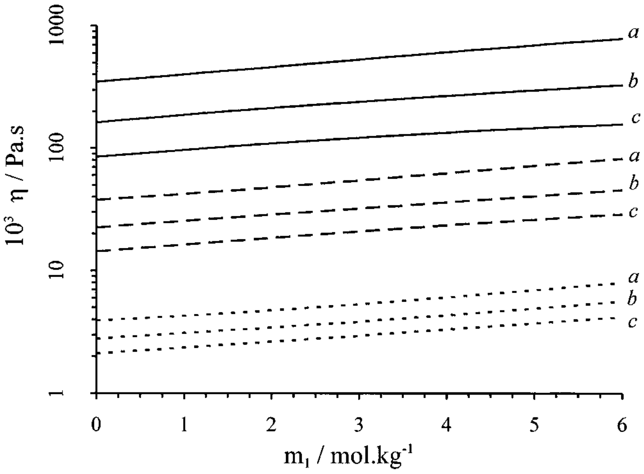

Figure 2. Ternary system dynamic viscosity as a function of NaCl molality for three sucrose molalities [(—) 5.55 mol·kg^{-1}; (– – –) 3.33 mol·kg^{-1}; (···) 1.11 mol·kg^{-1}] and three temperatures [283.15 K (*a*), 293.15 K (*b*), and 303.15 K (*c*)].

relationship. Figure 2 gives the ternary system dynamic viscosity as a function of NaCl molality, for three sucrose molalities (1.11, 3.33, and 5.55 mol·kg^{-1}) and three temperatures. Results showed that the presence of sucrose had a prevailing effect on viscosity, and whatever the sucrose molality, the effect of NaCl molality was similar which is in accordance with previous observations in Table 6 (slight effect of temperature on the interaction coefficient; prevailing effect of sucrose). For a ternary system with 6 and 5.55 m NaCl and sucrose, respectively, the dynamic viscosity was reduced by 2.35 when the temperature increased from 283.15 to 293.15 K and by 2.11 when the temperature increased from 293.15 to 303.15 K.

Conclusion

The present work led to experimental measurements of physical properties (density, viscosity) of mixed blends (ternary systems: NaCl−sucrose−water), for a wide range of concentration (zero to saturation) and temperature (283.15 to 303.15 K). The present work also led to correlations which allow an accurate prediction of these physical properties, even for high concentration levels. The main interests of the results are twofold: to provide mathematical forms of density and viscosity evolution against composition and temperature, for heat and mass transport in soaking processes, and to provide a practical set of tools and scientific criteria for soaking process implementation at the industrial scale (equipment design, process control, ...).

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