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Prediction of the diffusion coefficients in multicomponent liquid refrigerant solutions

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ABSTRACT

Immersion chilling and freezing (ICF) consist of soaking foods in a low freezing point liquid medium maintained at a low temperature that are considered secondary refrigerants. Data related to transport properties for the refrigerating media used in ICF processes are scarce, particularly for ternary and higher order systems. The objective of this work was to develop a model for the prediction of diffusion coefficients for systems of interest for ICF processes. The generalized Maxwell–Stefan equations were used in the model development. Data obtained from literature corresponding to Fick diffusion coefficients of the binary systems NaCl–H₂O and KCl–H₂O, and the ternary system NaCl–KCl–H₂O, were compared with predicted values. Results were satisfactory, errors of the main diffusion coefficients less than 13.5% being obtained. These results may be extended to the typical range of conditions of ICF, where no data are available in literature to date.

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1. Introduction

The mass transfer from a liquid medium to a food is a common phenomenon in the food industry. Therefore, it is crucial to understand and model such phenomenon. Cheese brining, immersion cooling and freezing (ICF) in secondary refrigerants and osmotic dehydration are examples of such process. Particularly, ICF have important advantages when it is compared with other alternatives of cooling or freezing, such as high energy transfer rates, great versatility, lower annual costs and lower environmental impact (Lucas and Raoult-Wack, 1998; Chourot et al., 2003). The liquid media can be a binary or multicomponent solution of NaCl, CaCl₂, KCl, ethanol, acetic acid, sugars, etc.

A mass transfer phenomenon due to a diffusive flux is generally described by the constitutive equation called Fick's law, where the flux is proportional to a local concentration gradient and the proportionality coefficient is called fickian diffusion coefficient, which it is generally assumed constant. That equation is valid for special cases and it is less than useful in the case of electrolyte solutions (Wesselingh et al., 1995) due to the diffusion coefficients depend on the system, composition, composition gradients and temperature. Therefore, to predict the changes in concentration due to diffusive fluxes, models based on the thermodynamics of the irreversible processes connected with the Onsager reciprocal relations (Dunlop and Gosting, 1959) and extended models of the

equations developed by Maxwell and Stefan around 1870 (Wesselingh et al., 1995) are generally used. All those models need diffusion coefficients (fickian diffusion coefficients, thermodynamical or phenomenological coefficients and Maxwell–Stefan coefficients) that are related each other (Wendt, 1965; Curran et al., 1967).

Generally, it is very important that diffusion coefficients were available in a wide range of concentrations. Such information is scarce, particularly in the case of multicomponent systems. Wendt (1965) derived equations for predicting the phenomenological coefficients and diffusion coefficients for ternary systems. Good agreements between estimated and observed values were found for low concentration mixtures. Miller (1966) derived rigorous expressions for predicting the diffusion coefficients of binary electrolyte systems using irreversible thermodynamics and the Onsager reciprocal relations. Zorrilla and Rubiolo (1994) obtained effective diffusion coefficients associated with the diffusion of multicomponents in a food using the generalized Fick's law. The values obtained were considered constant in the range of concentration studied.

A model that is consistent with the thermodynamics of irreversible processes is the extended Maxwell–Stefan (MS) model. In this model, the MS coefficients are provided for each pair of species, are symmetric, are less concentration-dependent than Fick coefficients and do not depend on the concentration gradient (Graham and Dranoff, 1982; Krishna and Wesselingh, 1997). The MS diffusivity has the physical significance of an inverse drag coefficient. The MS model takes into account the effect of strong composition through a thermodynamic correction factor, which changes with

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$$(J^n) = -c_t [B]^{-1} (d) \quad (12)$$

Eq. (12) allows visualizing that:

$$J_i^n = -c_t \sum_{j=1}^{n-1} b_{ij} d_j \quad (13)$$

where b_{ij} are the elements of the matrix $[B]^{-1}$.

The total current carried by the mixture of electrolytes is:

$$i = F \sum_{i=1}^{n-1} z_i N_i \quad (14)$$

For pure diffusion, there is no current flowing through the mixture:

$$i = 0 \quad (15)$$

Therefore, using Eqs. (15), (14), (4), (13), and (3), the diffusion potential $\nabla\phi$ can be obtained:

$$F\nabla\phi = - \frac{\sum_{k=1}^{n-1} \sum_{l=1}^{n-1} z_k x_l b_{kl} \nabla_{T,P} \mu_l}{\sum_{k=1}^{n-1} \sum_{l=1}^{n-1} z_k x_l b_{kl} z_l} \quad (16)$$

Eq. (16) shows that even when $i = 0$, there exists a finite electrical potential. The electrical potential gradient tends to slow down the fastest ion and to accelerate the other so that they move together, satisfying the electroneutrality condition (Krishna and Wesselingh, 1997).

Replacing Eqs. (16) and (3) in Eq. (13):

$$J_i^n = - \frac{c_t}{RT} \frac{\sum_{j=1}^{n-1} \sum_{k=1}^{n-1} \sum_{l=1}^{n-1} x_j x_l b_{ij} b_{kl} z_k (z_l \nabla_{T,P} \mu_j - z_j \nabla_{T,P} \mu_l)}{\sum_{k=1}^{n-1} \sum_{l=1}^{n-1} z_k x_l b_{kl} z_l} \quad (17)$$

2.1. Case of a binary system

Considering a system consisting of a neutral solvent such as water and an electrolyte s completely ionized:

$$C_{v1} A_{v2} \rightarrow v_1 C^{z1} + v_2 A^{z2} \quad (18)$$

where C is the cation, A is the anion and subscripts 1 and 2 refer to the cation and anion, respectively.

It is common to define the flux of the electrolyte s as:

$$J_s^n = -c_t D_s^n \nabla x_s \quad (19)$$

Taking into account that the cation and anion motions are coupled through Eq. (2), the electrolyte diffuses as if it were a nonelectrolyte (Miller, 1966), resulting that:

$$J_s^n = \frac{J_i^n}{v_i}, \quad i = 1, 2 \quad (20)$$

Moreover, taking into account that:

$$\mu_s = v_1 \mu_1 + v_2 \mu_2 \quad (21)$$

$$\mu_i = \mu_i^0 + RT \ln(\gamma_i x_i) \quad (22)$$

$$\sum_{i=1}^2 v_i z_i = 0 \quad (23)$$

$$\gamma_s = (\gamma_1^{v_1} \gamma_2^{v_2})^{\frac{1}{v}} \quad (24)$$

$$v = v_1 + v_2 \quad (25)$$

Eq. (26) can be obtained:

$$D_s^n = \frac{L}{c_t} \frac{\partial \mu_s}{\partial x_s} \Big|_{T,P} \quad (26)$$

where

$$L = - \frac{c_t}{RT} \frac{z_1 z_2 x_1 x_2 (b_{11} b_{22} - b_{12} b_{21})}{v_1 v_2 \sum_{k=1}^2 \sum_{l=1}^2 z_k b_{kl} x_l z_l} \quad (27)$$

$$\frac{\partial \mu_s}{\partial x_s} \Big|_{T,P} = vRT \left(\frac{\partial \ln \gamma_s}{\partial x_s} \Big|_{T,P} + \frac{1}{x_s [1 + (v-1)x_s]} \right) \quad (28)$$

$$\frac{\partial \ln \gamma_s}{\partial x_s} \Big|_{T,P} = \frac{1}{v [1 + (v-1)x_s]^2} \sum_{i=1}^2 v_i^2 \frac{\partial \ln \gamma_i}{\partial x_i} \Big|_{T,P} \quad (29)$$

L is the thermodynamic diffusion coefficient when the solvent velocity is used as reference velocity (Miller, 1966).

2.2. Case of a ternary system

Considering a system consisting of a neutral solvent such as water and two electrolytes completely ionized with a common ion:

$$C_{v1} A_{v3} \rightarrow v_1 C^{z1} + v_3 A^{z3} \quad (30)$$

$$B_{v2} A_{v3} \rightarrow v_2 B^{z2} + v_3 A^{z3} \quad (31)$$

where C and B are the cations, A is the anion and subscripts 1 and 2 refer to the cations and 3 to the anion, respectively.

It is common to define the fluxes of the electrolytes as:

$$J_{si}^n = -c_t \sum_{j=1}^2 D_{sij}^n \nabla x_{sj} \quad (32)$$

The subscripts in D_s^n refer to the salt for which the cation is i or j . Taking into account that Eqs. (20)–(25) are valid for each electrolyte, Eq. (33) is obtained:

$$D_{sij}^n = \frac{1}{c_t} \sum_{k=1}^2 L_{ik} \frac{\partial \mu_{sk}}{\partial x_{sj}} \Big|_{T,P} \quad (33)$$

where

$$L_{11} = \frac{c_t}{v_1^2 RT} \left[\frac{z_2^2 x_1 x_2 (b_{11} b_{22} - b_{12} b_{21}) + 2z_2 z_3 x_1 x_3 (b_{11} b_{23} - b_{13} b_{21})}{\sum_{k=1}^3 \sum_{l=1}^3 z_k b_{kl} x_l z_l} + \frac{z_3^2 x_1 x_3 (b_{11} b_{33} - b_{13} b_{31})}{\sum_{k=1}^3 \sum_{l=1}^3 z_k b_{kl} x_l z_l} \right] \quad (34)$$

$$L_{22} = \frac{c_t}{v_2^2 RT} \left[\frac{z_1^2 x_1 x_2 (b_{11} b_{22} - b_{12} b_{21}) + 2z_1 z_3 x_2 x_3 (b_{22} b_{13} - b_{23} b_{12})}{\sum_{k=1}^3 \sum_{l=1}^3 z_k b_{kl} x_l z_l} + \frac{z_3^2 x_2 x_3 (b_{22} b_{33} - b_{23} b_{32})}{\sum_{k=1}^3 \sum_{l=1}^3 z_k b_{kl} x_l z_l} \right] \quad (35)$$

$$L_{12} = L_{21} = \frac{c_t}{v_1 v_2 RT} \left[\frac{z_1 z_2 x_1 x_2 (b_{12} b_{21} - b_{11} b_{22}) + z_2 z_3 x_2 x_3 (b_{12} b_{23} - b_{13} b_{22})}{\sum_{k=1}^3 \sum_{l=1}^3 z_k b_{kl} x_l z_l} + \frac{z_1 z_3 x_1 x_2 (b_{12} b_{31} - b_{11} b_{32}) + z_2^2 x_2 x_3 (b_{12} b_{33} - b_{13} b_{32})}{\sum_{k=1}^3 \sum_{l=1}^3 z_k b_{kl} x_l z_l} \right] \quad (36)$$

$$\frac{\partial \mu_{si}}{\partial x_{sj}} \Big|_{T,P,x_{sj}} = v_{sj} RT \left\{ \frac{\partial \ln \gamma_{si}}{\partial x_{sj}} \Big|_{T,P,x_{sj}} + \frac{v_i^2}{v_{si} x_i} \frac{[1 + x_{sj}(v_{sj} - 1)]}{[(v_{s1} - 1)x_{s1} + (v_{s2} - 1)x_{s2} + 1]^2} + \frac{v_3}{v_{si} x_3} \frac{[1 + x_{sj}(v_{sj} - v_{s3})]}{[(v_{s1} - 1)x_{s1} + (v_{s2} - 1)x_{s2} + 1]^2} \right\} \quad (37)$$

$$\left. \frac{\partial \mu_{si}}{\partial x_{sj}} \right|_{T,P,x_{si}} = v_{si}RT \left\{ \left. \frac{\partial \ln \gamma_{si}}{\partial x_{sj}} \right|_{T,P,x_{si}} + \frac{v_i^2}{v_{si}x_i} \frac{[x_{si}(v_{si}-1)]}{[(v_{s1}-1)x_{s1} + (v_{s2}-1)x_{s2} + 1]^2} + \frac{v_3}{v_{si}x_3} \frac{[1 + x_{sj}(v_{sj}-v_{si})]}{[(v_{s1}-1)x_{s1} + (v_{s2}-1)x_{s2} + 1]^2} \right\} \quad (38)$$

$$\left. \frac{\partial \ln \gamma_{si}}{\partial x_{si}} \right|_{T,P,x_{sj}} = \frac{1}{v_{si}[(v_{s1}-1)x_{s1} + (v_{sj}-1)x_{sj} + 1]^2} \left\{ v_i^2[(v_{sj}-1)x_{sj} + 1] \frac{\partial \ln \gamma_i}{\partial x_i} + v_3^2[(v_{sj}-v_{si})x_{sj} + 1] \frac{\partial \ln \gamma_3}{\partial x_3} \right\} \quad (39)$$

$$\left. \frac{\partial \ln \gamma_{si}}{\partial x_{sj}} \right|_{T,P,x_{si}} = \frac{1}{v_{si}[(v_{s1}-1)x_{s1} + (v_{sj}-1)x_{sj} + 1]^2} \left\{ v_i(1-v_{sj}) \frac{\partial \ln \gamma_i}{\partial x_i} + v_3^2[(v_{sj}-v_{si})x_{sj} + 1] \frac{\partial \ln \gamma_3}{\partial x_3} \right\} \quad (40)$$

2.3. Model for the prediction of $\frac{\partial \ln \gamma_i}{\partial x_i}$

The derivatives of the activity coefficients of the ionic species in Eqs. (29), (39), and (40) can be evaluated by the extended UNIQUAC model (Peralta et al., 2007) taking into account the electro-neutrality condition given by Eq. (2). Thus:

$$\frac{\partial \ln \gamma_i}{\partial x_i} = \frac{\partial \ln \gamma_i^{D-H}}{\partial x_i} + \frac{\partial \ln \gamma_i^C}{\partial x_i} + \frac{\partial \ln \gamma_i^R}{\partial x_i} \quad (41)$$

where

$$\ln \gamma_i^{D-H} = -A_{DH}z_i^2 \frac{I^{1/2}}{1 + bI^{1/2}} \quad (42)$$

$$I = \frac{1}{2} \frac{\sum_{i=1}^n x_i z_i^2}{x_w M_w} \quad (43)$$

$$\ln \gamma_i^C = \ln \left(\frac{\phi_i}{x_i} \right) + \frac{z}{2} q_i \ln \left(\frac{\theta_i}{\phi_i} \right) + l_i - \frac{\phi_i}{x_i} \left(\sum_{j=1}^n x_j l_j \right) \quad (44)$$

$$l_i = \frac{z}{2} (r_i - q_i) - (r_i - 1) \quad (45)$$

$$\theta_i = \frac{x_i q_i}{\sum_{j=1}^n x_j q_j} \quad (46)$$

$$\phi_i = \frac{x_i r_i}{\sum_{j=1}^n x_j r_j} \quad (47)$$

$$\ln \gamma_i^R = q_i \left[1 - \ln \left(\sum_{j=1}^n \theta_j \tau_{ji} \right) - \sum_{j=1}^n \frac{\theta_j \tau_{ij}}{\sum_{k=1}^n \theta_k \tau_{kj}} \right] \quad (48)$$

$$\tau_{ji} = \exp \left[\frac{u_{ii} - u_{ji}}{T} \right] \quad (49)$$

$$u_{ji} = u_{ji}^o + u_{ji}^t (T - 298.15) \quad (50)$$

3. Materials and methods

3.1. Experimental data

To verify the validity of the model, experimental data obtained from the literature were compared with the predicted values. The experimental values correspond to systems with potentiality to be

Table 1

Experimental values of $D_{sij}^n \times 10^9$ (m² s⁻¹) in NaCl–KCl–H₂O (Dunlop, 1959; Dunlop and Gosting, 1959; Cussler, 1976). The subscripts in D_s^n mean 1: NaCl and 2: KCl according to Eq. (32). $X_{NaCl} = c_{NaCl}/(c_{NaCl} + c_{KCl})$.

C_{NaCl} (kmol m ⁻³)	C_{KCl} (kmol m ⁻³)	X_{NaCl}	D_{s11}^n	D_{s12}^n	D_{s21}^n	D_{s22}^n
0.25	0.25	0.50	1.38	-0.011	0.150	1.836
0.25	0.50	0.33	1.351	0.018	0.221	1.866
0.50	0.25	0.66	1.429	0.015	0.104	1.838
0.50	0.50	0.50	1.403	0.026	0.173	1.859
1.50	1.50	0.50	1.458	0.201	0.394	1.899

Table 2

MS diffusion coefficients (Graham and Dranoff, 1982).

i	j	$D_{ij} \times 10^9$ (m ² s ⁻¹)
Na ⁺	H ₂ O	1.28
K ⁺	H ₂ O	1.93
Cl ⁻	H ₂ O	2.08
Na ⁺	K ⁺	-0.15
Na ⁺	Cl ⁻	0.09
K ⁺	Cl ⁻	0.11

Table 3

UNIQUAC q and r parameters (Thomsen et al., 1996; García et al., 2005).

Species	q	r
H ₂ O	1.400	0.9200
Na ⁺	1.1990	1.4034
K ⁺	2.4306	2.2304
Cl ⁻	10.197	10.386

Table 4

UNIQUAC interaction parameters u_{ij}^o (Christensen and Thomsen, 2003; García et al., 2005).

ij	H ₂ O	Na ⁺	K ⁺	Cl ⁻
H ₂ O	0	733.286	535.023	1523.39
Na ⁺		0	-46.194	1443.23
K ⁺			0	1465.18
Cl ⁻				2214.81

Table 5

UNIQUAC interaction parameters u_{ji}^t (Christensen and Thomsen, 2003; García et al., 2005).

ij	H ₂ O	Na ⁺	K ⁺	Cl ⁻
H ₂ O	0	0.48719	0.99356	14.631
Na ⁺		0	0.11899	15.635
K ⁺			0	15.329
Cl ⁻				14.436

used in the food industry, particularly in ICF processes. Experimental Fick diffusion coefficients of the binary systems NaCl–H₂O (Rard and Miller, 1979) and KCl–H₂O (Rard and Miller, 1980), and of the ternary system NaCl–KCl–H₂O (Table 1) (Dunlop, 1959; Dunlop and Gosting, 1959; Cussler, 1976) were used. All experimental data were obtained at 298.15 K.

3.2. Model parameters

Table 2 shows the MS diffusion coefficients used (Graham and Dranoff, 1982).

The extended UNIQUAC parameters for the study cases were obtained from Thomsen et al. (1996), Christensen and Thomsen (2003) and García et al. (2005) (Tables 3–5).

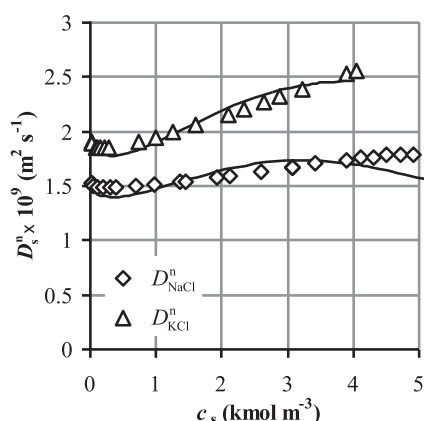


Fig. 1. Diffusion coefficients for the binary systems NaCl–H₂O (Rard and Miller, 1979) and KCl–H₂O (Rard and Miller, 1980). Symbols correspond to experimental data and lines to calculated values.

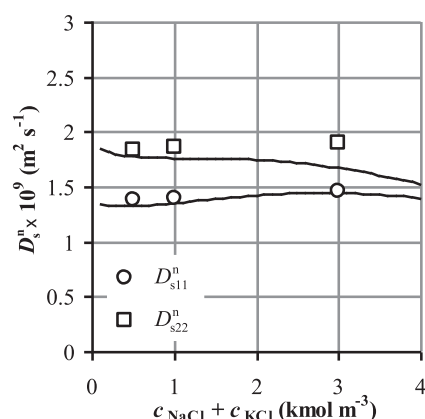


Fig. 2. Main diffusion coefficients for the ternary system NaCl–KCl–H₂O for $X_{\text{NaCl}} = 0.5$. Symbols correspond to experimental data (Dunlop, 1959; Cussler, 1976) and lines to calculated values.

The Debye–Hückel parameter as proposed by Nicolaisen et al. (1993) was used.

$$A_{DH} = 35.765 + 4.222 \times 10^{-2}(T - 273.15) + 3.681 \times 10^{-4}(T - 273.15)^2 \quad (51)$$

3.3. Physical properties

Leaist and Al-Dhafer (2000) indicated that the concentration dependence of diffusion coefficients is dominated by changes in the viscosity and thermodynamic driving forces. In this study, the predicted diffusion coefficients were corrected by the viscosity effect according to Horvath (1985) using the factor η^0/η , where η^0 is the viscosity of pure water and η is the viscosity of the solution. The values of viscosity of the study cases were obtained from the data reported by Hai-Lang and Shi-Jun (1996).

The density of the solutions is necessary to convert electrolyte concentration to ion molar fraction and to convert volume-fixed frame diffusion coefficients to solvent-fixed frame coefficients. The values of density of the study cases were obtained from the data reported by Hai-Lang and Shi-Jun (1996).

4. Results and discussion

In Fig. 1, predicted diffusion coefficient values are compared with the experimental ones for the systems NaCl–H₂O and KCl–H₂O. Due to Rard and Miller (1979, 1980) obtained volume-fixed frame diffusion coefficients, those values were converted to solvent-fixed frame coefficients following the procedure suggested by Dunlop and Gosting (1959). In Fig. 2, predicted main diffusion coefficient values are compared with the experimental ones corresponding to $X_{\text{NaCl}} = 0.5$ for the system NaCl–KCl–H₂O. A satisfactory accuracy can be observed.

In Fig. 3, the percentage errors of the model related to the experimental data for the binary systems are shown. The highest absolute percentage errors were 13.4% and 4% for the NaCl–H₂O and KCl–H₂O systems, respectively. The highest absolute percentage error in the prediction of the main coefficients for the system NaCl–KCl–H₂O was 11.8%, while the error for the cross-term coefficients was 118% (Table 6). It is worth mentioning that the magnitude of the cross-term coefficients is remarkably smaller than the main coefficients, showing at consequence a higher uncertainty. Moreover, the errors observed may be related to some of the assumptions made in the model development. The MS coefficients were considered constant although those coefficients show a weak

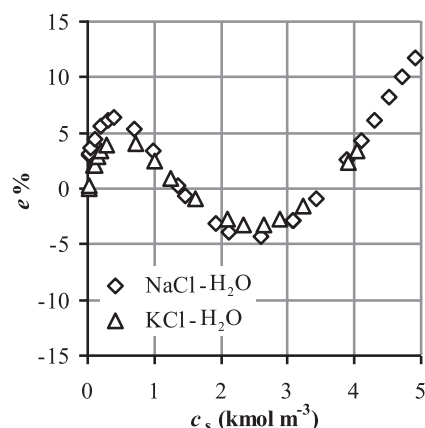


Fig. 3. Percentage error $e\% = 100 \times (D_{\text{exp}} - D_{\text{calc}})/D_{\text{exp}}$ for the binary systems NaCl–H₂O and KCl–H₂O.

Table 6

Percentage error for the system NaCl–KCl–H₂O. The subscripts in e% mean 1: NaCl and 2: KCl.

C_{NaCl} (kmol m ⁻³)	C_{KCl} (kmol m ⁻³)	$e_{11}\%$	$e_{12}\%$	$e_{21}\%$	$e_{22}\%$
0.25	0.25	3.61	–117.87	34.38	3.05
0.25	0.50	3.75	41.70	15.35	4.23
0.50	0.25	3.73	73.77	22.78	4.72
0.50	0.50	3.29	–90.95	–0.47	5.08
1.50	1.50	0.74	–82.54	–31.03	11.80

dependence with the media concentration (Graham and Dranoff, 1982) and with the ion solvation. The errors in the predictions associated with the extended UNIQUAC model may also be considered as a source of error in this case (Peralta et al., 2007).

5. Conclusions

The Maxwell–Stefan equations were adequate for predicting diffusion coefficients of systems with potentiality to be used in ICF processes. A satisfactory accuracy was observed for the systems studied. Although the MS equations are useful for electrolyte and non-electrolyte systems of any level of concentrations, it is not commonly used because of the complex mathematical developments and the lack of activity coefficient data. In this case, the extended UNIQUAC model strongly helped in the prediction of

activity coefficients, resulting in a very promising model for predicting diffusion coefficients for multicomponent systems.

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