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MEASUREMENTS AND CORRELATION OF OSMOTIC COEFFICIENTS AND EVALUATION OF VAPOR PRESSURE FOR SOLUTIONS OF KCH_3COO AND $NaCH_3COO$ IN METHANOL AT 25 °C†

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†This paper is dedicated to Prof. Josef Barthel in honour of his 75th birthday.

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Abstract

The osmotic coefficients of potassium acetate and sodium acetate in methanol have been measured by the isopiestic method at 25 °C. Sodium iodide was used as isopiestic standard for the calculation of osmotic coefficients. The molality ranges covered in this study correspond to about 0.17-2.51 $mol \cdot kg^{-1}$ for potassium acetate and 0.25-1.76 $mol \cdot kg^{-1}$ for sodium acetate. The system of equations of Pitzer–Mayorga and MSA-NRTL were used to fit osmotic coefficients. The parameters from the fit were used to calculate the vapor pressures. The osmotic coefficient data are successfully correlated with these models, which provide reliable predictions of vapor pressures.

Key words: osmotic coefficient, isopiestic, methanol, models

Introduction

Thermodynamic properties of electrolyte solutions are important for a variety of applications in the chemical processes in industries. Electrolytes are involved in numerous processes including environmental applications such as chemical waste disposal, separation process and electrochemical process. Osmotic coefficient data of binary electrolyte solutions are required to describe the thermodynamic behavior of electrolyte solutions with organic solvents. These data are also useful to predict thermodynamic properties of electrolytes in mixed solvents.[1-3] However; accurate thermodynamic data are very scare for non-aqueous electrolyte solutions. Barthel and his co-workers[4-9] have made accurate vapor pressure-lowering measurements on a few non-aqueous electrolyte solutions from which osmotic coefficient values may be calculated. There are also other reports on the vapor pressure of some electrolytes in methanol.[10,11] The reported data for NaI in methanol[4,11] solutions have been used as the isopiestic reference standards and the osmotic coefficients of some solutes in methanol have been measured.[12-14]

In this work the osmotic coefficients and vapor pressures of solutions of potassium acetate and sodium acetate in methanol are reported. For solutions of KCH_3COO in methanol, a few vapor pressure data has been reported; however, for concentrations lower than 1 $mol\cdot kg^{-1}$, only one data point has been given.[11] Information for the activity and osmotic coefficients of the solution of sodium acetate in methanol has not been reported.

The osmotic coefficients have been measured using an improved isopiestic apparatus. For isopiestic reference sodium iodide in methanol solutions were used as described previously.[12] Vapor pressures for the solutions of investigated electrolytes in methanol have been calculated from the osmotic coefficient data by the relevant thermodynamic relations.

The Pitzer-Mayorga model[15] and MSA-NRTL model[16] were successfully used to reproduce the experimental osmotic coefficients and to derive the vapor pressures.

Experimental

Apparatus and procedure. The isopiestic apparatus employed in this research is essentially similar to the one used previously.[17] Recently, this technique has been used for the measurement of osmotic coefficients of some inorganic salts in methanol.[12-14] This apparatus consisted of a five-leg manifold attached to round-bottom flasks. The five flasks were typically used as follows. Two flasks contained the standard NaI solutions, two flasks contained either potassium acetate or sodium acetate solutions, and the central flask was used as a methanol reservoir. The apparatus was held in a constant temperature bath for at least 120 hours for equilibration at (298.15 ± 0.005) K.

Chemicals. The methanol and salts were obtained from Merck. They were all supra pure reagents (methanol GR., min. 99.8%; NaI, GR., min. 99.5%; KCH_3COO GR., min. 99.5%, $NaCH_3COO$, GR, min 99.5%). All chemicals were used without further purification. The salts were dried in an electrical oven at about 393 K for 24 h prior to use.

Results and Discussions

Experimental results. Isopiestic equilibrium molalities with reference standard solutions of NaI in methanol as reported in Tables 1 and 2 enabled the calculation of the osmotic coefficient, ϕ, of the solutions of potassium acetate and sodium acetate in methanol from

$$\phi = \left(\nu^* \phi^* m^* / \nu m\right) \tag{1}$$

where ν^* and ν are the sum of stoichiometric numbers of anion and cation, $\nu_+ + \nu_-$, in the reference solution and in the solutions of potassium acetate or sodium acetate, respectively, m^* is the molality of the reference standard in isopiestic equilibrium with these solutions, and ϕ^* is the osmotic coefficient of the isopiestic reference standard, calculated at m^*. The necessary ϕ^* values at any m^* were obtained from the fitted Pitzer and Mayorga equation, including the $\beta^{(2)}$ term,[15] as described by Zafarani-Moattar and Nasirzadeh.[12] It was shown that,[15] using $\alpha_{(1)}$=2, $\alpha_{(2)}$=1.4, $\beta^{(0)}$=0.40830, $\beta^{(1)}$=1.04430, $\beta^{(2)}$=-0.875 and C^{ϕ}=-0.02224, the osmotic coefficients of the isopiestic reference standard solutions, ϕ^*, are reproducible with standard deviation of 0.005 for NaI in methanol solutions in the range (0.02 to 4.33) mol·kg^{-1} at 25 °C.

Table 1. Experimental isopiestic molalities, osmotic coefficients, vapor pressures and activity of methanol for KCH_3COO in methanol at 25 °C.

m_{NaI} / (mol·kg^{-1})	$m_{KCH3COO}$/ (mol·kg^{-1})	ϕ_{exp}	ϕ_{calc}	P_{exp} / (kPa)	a_s
0.0000	0.0000	1.000	1.000	16.958	1.0000
0.1720	0.1783	0.799	0.799	16.801	0.9909
0.2762	0.2915	0.793	0.793	16.705	0.9853
0.3760	0.4044	0.794	0.794	16.607	0.9796
0.4719	0.5171	0.798	0.798	16.509	0.9739
0.5640	0.6295	0.802	0.802	16.410	0.9682
0.6527	0.7416	0.807	0.807	16.311	0.9624
0.7383	0.8534	0.812	0.812	16.211	0.9566
0.8210	0.9649	0.817	0.817	16.111	0.9507
0.9010	1.0761	0.821	0.822	16.011	0.9449
1.0540	1.2976	0.830	0.830	15.811	0.9333
1.1634	1.4630	0.836	0.836	15.662	0.9246
1.2688	1.6281	0.842	0.842	15.512	0.9159
1.3371	1.7380	0.845	0.846	15.413	0.9101
1.3708	1.7929	0.847	0.847	15.364	0.9072
1.3641	1.7819	0.847	0.847	15.374	0.9078
1.4699	1.9577	0.852	0.853	15.215	0.8986
1.5345	2.0676	0.856	0.856	15.116	0.8928
1.5981	2.1778	0.859	0.859	15.017	0.8870
1.6296	2.2329	0.861	0.860	14.968	0.8841
1.7227	2.3990	0.865	0.865	14.819	0.8755
1.6918	2.3435	0.863	0.863	14.869	0.8784
1.7533	2.4545	0.866	0.866	14.770	0.8726
1.7838	2.5102	0.868	0.868	14.720	0.8697

From the calculated osmotic coefficient data, the activity of methanol in potassium acetate and sodium acetate solutions and the vapor pressure of methanol over these solutions were determined at isopiestic equilibrium molalities, with the help of the following thermodynamic relations:

$$\phi = -\ln a_s / \nu m M_s \tag{2}$$

$$\ln a_s = \ln\left(p/p^*\right) + \left(B_s - V_s^*\right)\left(p - p^*\right)/RT \tag{3}$$

In these equations, a_s is the activity of solvent, B_s, V^*_s and p^* are second virial coefficient, molar volume and vapor pressure of pure methanol, respectively. The values of M_s=0.032042, B_s=-2.075×10^{-3} m^3·mol^{-1}, V_s^*= 4.073×10^{-5} m^3·mol^{-1} and p^*= 16957.7 Pa (taken from Barthel et al.[7]) were used at 298.15 K.

Table 2. Experimental isopiestic molalities, osmotic coefficients, vapor pressures and activity of methanol for $NaCH_3COO$ in methanol at 25 °C.

m_{NaI} / (mol·kg^{-1})	$m_{NaCH3COO}$ / (mol·kg^{-1})	ϕ_{exp}	ϕ_{calc}	P_{exp} / (kPa)	a_s
0.0000	0.0000	1.000	1.000	16.958	1.0000
0.2369	0.2558	0.776	0.774	16.743	0.9874
0.2582	0.2822	0.769	0.773	16.721	0.9862
0.2691	0.2943	0.770	0.772	16.711	0.9854
0.3042	0.3319	0.776	0.772	16.679	0.9836
0.3512	0.3878	0.773	0.773	16.632	0.9810
0.3732	0.4154	0.771	0.773	16.608	0.9797
0.3989	0.4436	0.776	0.774	16.585	0.9782
0.4406	0.4948	0.775	0.776	16.541	0.9757
0.4914	0.5554	0.779	0.778	16.489	0.9726
0.5103	0.5789	0.780	0.779	16.469	0.9715
0.5903	0.6799	0.783	0.783	16.381	0.9664
0.6347	0.7385	0.784	0.786	16.330	0.9636
0.6550	0.7601	0.790	0.787	16.312	0.9622
0.7324	0.8676	0.790	0.791	16.217	0.9570
0.7491	0.8863	0.794	0.792	16.201	0.9559
0.7623	0.9084	0.791	0.793	16.182	0.9550
0.7869	0.9424	0.792	0.794	16.152	0.9533
0.8441	1.0246	0.793	0.797	16.079	0.9492
0.8593	1.0441	0.796	0.797	16.062	0.9481
0.8958	1.0964	0.798	0.799	16.016	0.9455
0.9812	1.2215	0.802	0.803	15.906	0.9391
1.0254	1.2877	0.804	0.805	15.848	0.9358
1.1160	1.4175	0.814	0.808	15.735	0.9287
1.1575	1.4997	0.807	0.810	15.664	0.9254
1.1869	1.5384	0.813	0.810	15.631	0.9230
1.3163	1.7631	0.813	0.815	15.443	0.9123

A comparison of our vapor pressure data to that of Tomasula et al.[11] for KCH_3COO in methanol is given in Figure 1. Figure 1 shows that the data of Tomasula et al. are somewhat higher than those obtained in this work. However, close examination of the Tomasula et al. data indicates that these authors have used p^*=17.08 kPa for vapor pressure of the pure methanol which is slightly higher (0.12 kPa) than the value of 16.96 kPa used in this work. If the Tomasula et al. data were corrected according to this value (0.12 kPa), the obtained values are in a good agreement with those of this work.

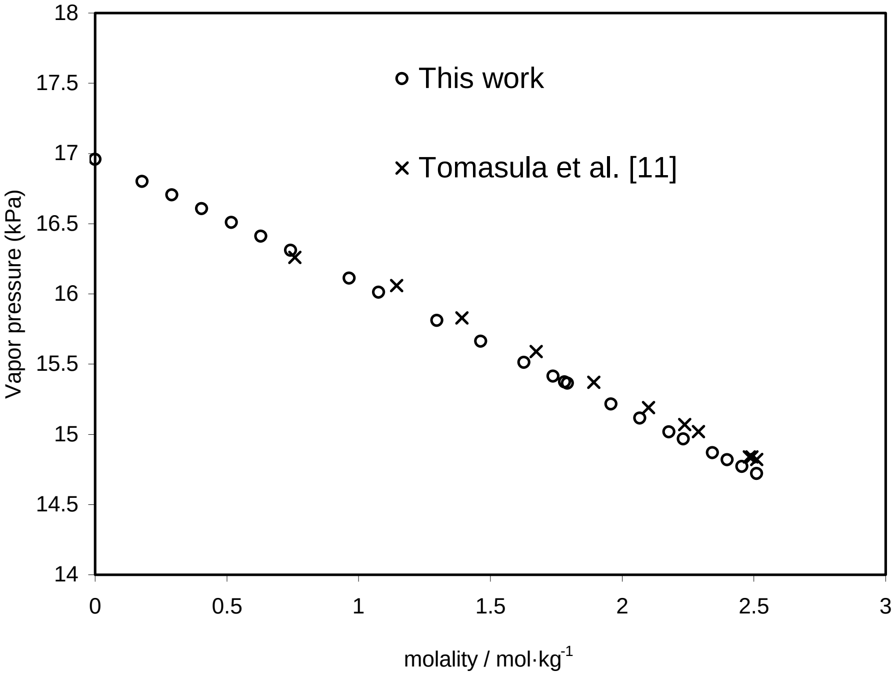

Figure 1. Comparison of vapor pressures for KCH_3COO in methanol solutions at 25 °C.

Correlation of data

Pitzer model. Several models are available in the literature for the correlation of osmotic coefficients as a function of molalities. The model of Pitzer and Mayorga[15] has been successfully used for aqueous and in a few cases, for non-aqueous electrolyte solutions.[12-14]

The experimental osmotic coefficient data were correlated with the model of Pitzer and Mayorga[15] for solutions of KCH_3COO and $NaCH_3COO$ in methanol. This model has the following form[15]

$$\phi - 1 = f^{\phi} + mB^{\phi} + m^2C^{\phi} \tag{4}$$

where

$$f^{\phi} = -A_{\phi} I^{1/2} \Big/ \left(1 + b I^{1/2}\right) \tag{5}$$

$$A_{\phi} = (1/3)(2\pi N_A d_s)^{1/2} \left(e^2 / 4\pi\varepsilon_0 \varepsilon k T\right)^{3/2} \tag{6}$$

$$\mathrm{B}^{\phi} = \beta^{(0)} + \beta^{(1)} \exp\left[-\alpha_{(1)} I^{1/2}\right] + \beta^{(2)} \exp\left[-\alpha_{(2)} I^{1/2}\right] \tag{7}$$

In these equations $\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(2)}$ and C^{ϕ} are Pitzer's ionic-interaction parameters; $\alpha_{(1)}$, $\alpha_{(2)}$ and b are adjustable parameters, and A_{ϕ} is the Debye-Hückel constant for the osmotic coefficient on the molal basis. The remaining symbols have their usual meaning. For methanol solutions A_{ϕ}=1.294 kg$^{1/2}$·mol$^{-1/2}$ was calculated using equation (6). From the analysis of the experimental osmotic coefficient data, we found that the values of b=3.2 kg$^{1/2}$.mol$^{-1/2}$, $\alpha_{(1)}$=2.0 kg$^{1/2}$.mol$^{-1/2}$ and $\alpha_{(2)}$=1.4 kg$^{1/2}$.mol$^{-1/2}$ were satisfactory at 298.15 K. The ion-interaction parameters obtained from the experimental osmotic coefficient data for the investigated systems are shown in Table 3.

Table 3. Pitzer parameters for methanol solutions of KCH_3COO and $NaCH_3COO$ calculated from osmotic coefficients[a] at 25 °C.

no. of data	molality range	$\beta^{(0)}$	$\beta^{(1)}$	$\beta^{(2)}$	C^{ϕ}	sd$_{(\phi)}$[b]
			KCH_3COO			
23	0.17– 2.51	0.008128	-0.687219	0.838449	0.004572	0.0001
			$NaCH_3COO$			
26	0.25-1.76	0.026218	-0.128391	-2.118794	1.9988	0.002

[a] A_{ϕ}=1.294 kg$^{1/2}$·mol$^{-1/2}$; b=3.2 kg$^{1/2}$·mol$^{-1/2}$; $\alpha_{(1)}$=2 kg$^{1/2}$·mol$^{-1/2}$; $\alpha_{(2)}$=1.4 kg$^{1/2}$·mol$^{-1/2}$; [b] standard deviation of osmotic coefficients.

MSA-NRTL model. The MSA-NRTL model has been developed[16] as a semi-empirical model for electrolyte solutions. The model calculates the excess Gibbs energy of the electrolyte solution. The Gibbs energy is divided in two parts. The first long-range part corresponds to the electrostatic contribution of ion charges to the excess Gibbs energy. The second contribution is a short-range contribution corresponding to all short-range forces existing between ions and solvent molecules. The molar excess Gibbs energy, g^{ex}, is written as follows:

$$g^{ex} = g^{ex}_{Lr} + g^{ex}_{Sr} \tag{8}$$

Therefore, the activity coefficient for a solvent (s) is written as:

$$\ln \gamma_s = \ln \gamma_s^{Lr} + \ln \gamma_s^{Sr} \tag{9}$$

Kunz et al.[16] have focused on the so-called RPM-MSA (restricted primitive model-mean spherical approximation) and have used it in place of the PDH (Pitzer-Debye-Hückel) equation to calculate the long-range distribution to the activity coefficient of species (i) in solution. The MSA which was originally applied to RPM electrolyte solutions is known to account for electrostatic interactions between ions in a better way than the DH model[18] and it has been found quite accurate enough to be of great practical ability.[19] Kunz et al.[16] have ignored the electro-neutrality assumption and have made two modifications in using NRTL which lead them to have four different adjustable parameters, that is, τ_{am}, τ_{cm}, $\tau_{\mathrm{mc,ac}}$ and $\tau_{\mathrm{ma,ca}}$. A combination of all adopted expressions that have been given by Kunz et al.[16] gave us the following equation for MSA-NRTL model.

$$\phi = -\frac{1}{\nu m M_s}\left\{\begin{array}{l} \dfrac{x_s(\tau_{am}\exp(-\alpha\tau_{am})+\tau_{cm}\exp(-\alpha\tau_{cm}))}{x_s(\exp(-\alpha\tau_{am})+\exp(-\alpha\tau_{cm}))+x_m}\cdot\left[1-\dfrac{x_m}{x_s(\exp(-\alpha\tau_{am})+\exp(-\alpha\tau_{cm}))+x_m}\right] \\ +\dfrac{x_s^2\tau_{mc,ac}\exp(-\alpha\tau_{mc,ac})}{(x_s+x_m\exp(-\alpha\tau_{mc,ac}))^2}+\dfrac{x_s^2\tau_{ma,ca}\exp(-\alpha\tau_{ma,ca})}{(x_s+x_m\exp(-\alpha\tau_{ma,ca}))}+ \\ \left[\begin{array}{l} x_s(1-\dfrac{x_s}{x_s+x_m\exp(-\alpha\tau_{mc,ac})})\cdot(1-\dfrac{a\tau_{mc,ac}x_s}{x_s+x_m\exp(-\alpha\tau_{mc,ac})}) \\ +x_s(1-\dfrac{x_s}{x_s+x_m\exp(-\alpha\tau_{ac,ca})})\cdot(1-\dfrac{a\tau_{ma,ca}x_s}{x_s+x_m\exp(-\alpha\tau_{ma,ca})}) \end{array}\right] \\ \cdot(1-x_m)\tau_{mc,ac}^{(2)}+\dfrac{\Gamma^3}{3\pi}\cdot\dfrac{M_s}{N_A\sigma}+\ln x_m \end{array}\right\} \qquad (10)$$

where M_{s}, N_{A}, x_{m}, x_{s} and α are the molar mass of solvent (kg·mol^{-1}), Avogadro constant, solvent mole fraction, solute mole fraction and non randomness factor, respectively. The parameter α varies between 0 and 1. The value of 1 corresponds to a full random distribution of species in the solution. A commonly accepted value for α is 0.2 in aqueous solutions,[20] even though it can be adjusted.[16] Therefore, this parameter is not adjusted in this work. The NRTL model is built by considering the different possible configurations around a particle. The system is then divided in as many cells as there are different species in the solution. Each cell is centered on a species. Γ is the MSA screening parameter,

$$\Gamma = (1/2\sigma)\left(\sqrt{(1+2\kappa\sigma)}-1\right) \qquad (11)$$

κ is the screening parameter in the Debye-Hückel theory

$$\kappa = \sqrt{4\pi\lambda \Sigma \rho_i z_i^2} \tag{12}$$

and

$$\lambda = \left(\beta e^2 / 4\pi\varepsilon_0 \varepsilon\right) \tag{13}$$

$\beta = 1/kT$ with k the Boltzmann constant, e is the charge of a proton; ε_0 and ε are the permittivity of a vacuum and of solvent, respectively. ρ is the number density of ion i, σ is the mean ionic diameter of the ions and z_i is the number of ionic charge. The mean ionic diameter σ is the only adjusted parameter in MSA part.

$$\tau_{ma,ca} = \tau_{mc,ac} + \tau_{am} - \tau_{cm} \tag{14}$$

$$\tau_{mc,ac} = \tau^{(1)}_{mc,ac} + \tau^{(2)}_{mc,ac} x_m \tag{15}$$

There are five adjustable parameters (one MSA parameter, σ and four NRTL parameters, τ_{am}, τ_{cm}, $\tau^{(1)}_{mc,ac}$ and $\tau^{(2)}_{mc,ac}$ in equation (10) which have to be calculated by fitting the model to the experimental data. MSA-NRTL parameters were obtained from the fitting experimental osmotic coefficient data for the investigated systems and are shown in Table 4.

Table 4. MSA-NRTL model parameters for methanol solutions of KCH_3COO and $NaCH_3COO$ calculated from osmotic coefficients at 25 °C.

no. of data	molality range	τ_{cm}	τ_{am}	$\tau^{1}_{mc,ac}$	$\tau^{2}_{mc,ac}$ [a]	σ [b]	$sd_{(\phi)}$ [c]
			KCH_3COO				
23	0.17– 2.51	2.723	-5.328	16.275	-6.916	4.653	0.0003
			$NaCH_3COO$				
26	0.25-1.76	5.640	-3.245	10.351	-4.700	4.184	0.0025

[a] in units of kg.mol^{-1}. [b] in units of 10^{-10} m, [c] standard deviation of osmotic coefficients.

Figure 2 shows the molality dependence of osmotic coefficient obtained from the isopiestic experiments and those generated using various models for KCH_3COO, $NaCH_3COO$ and $LiCH_3COO$[12] in methanol. Figure 2 show that both the MSA-NRTL and the Pitzer-Mayorga models have nearly the same prediction accuracy. The curves reveal the typical pattern of the dependence of the osmotic coefficients. At every concentration, the osmotic coefficients decrease in the order $KCH_3COO > NaCH_3COO > LiCH_3COO$, which indicates increasing ion–ion interactions from KCH_3COO to $LiCH_3COO$. There is, indeed, evidence for a higher ion pairing of

$LiCH_3COO$ compared to $NaCH_3COO$ in methanol from conductometric studies.[21] For instance, Barthel and Neueder[21] have reported association constants of 90.6 and 23.2 for $LiCH_3COO$ and $NaCH_3COO$, respectively. This result indicates strong ion–ion interaction in $LiCH_3COO$ + methanol solutions in compared to $NaCH_3COO$ in methanol. There aren't any conductance data for KCH_3COO in literature.

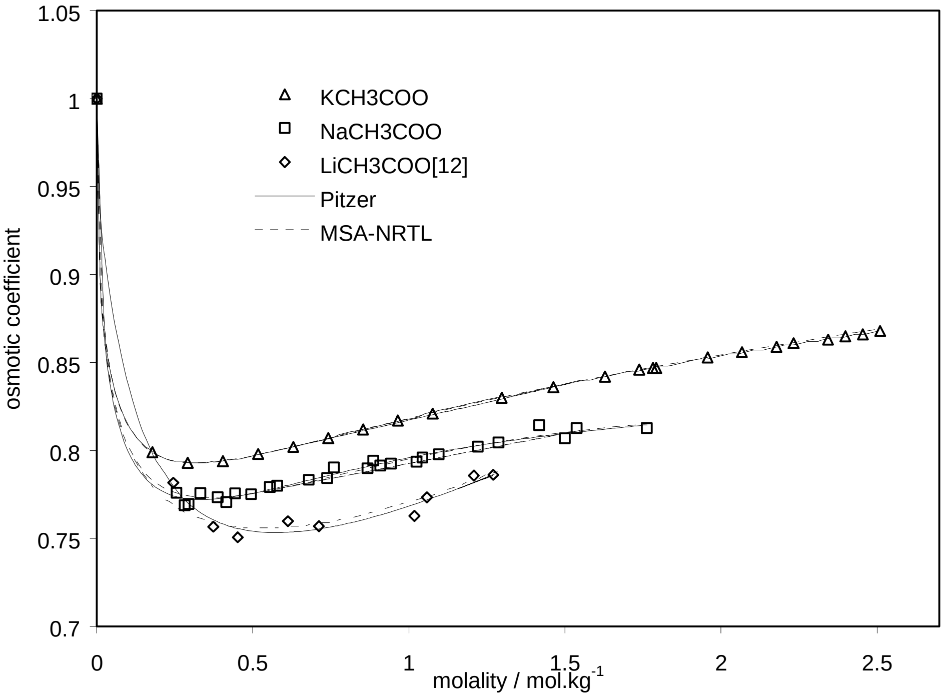

Figure 2. Experimental osmotic coefficients of KCH_3COO, $NaCH_3COO$ and $LiCH_3COO$ in methanol solutions at 25 °C. Lines were generated using the Pitzer and MSA-NRTL models.

Conclusions

Experimental osmotic coefficient measurements have been reported for KCH_3COO and $NaCH_3COO$ in methanol solutions by an improved isopiestic method at 25 °C. Experimental data of the investigated systems are satisfactorily correlated using the Pitzer-Mayorga and MSA-NRTL models. Model parameters are obtained and used to calculate vapor pressure for methanol salt systems.

The models of MSA-NRTL and Pitzer-Mayorga have been shown to correlate the experimental osmotic coefficient data with very good accuracy. For the Pitzer and Mayorga model, data analysis shows that the values $\alpha_{(1)}$=2.0, $\alpha_{(2)}$=1.4, b=3.2 based on the best representation of some lithium salts in methanol, also give a good overall results for KCH_3COO and $NaCH_3COO$ in methanol solutions. The fit accuracy obtained with the MSA-NRTL model is the same as that obtained with the Pitzer model. The main advantage of the MSA-NRTL model is the relative physical significance of the parameters, since the σ_{MSA} corresponds to the mean ionic solvated diameter, and the NRTL τ parameters correspond to the interaction energies between the different species in solution.

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Povzetek

Z izopiestično metodo smo pri 25 ºC izmerili osmozne koeficiente kalijevega in natrijevega acetata v metanolu v koncentracijskem obsegu med 0,17-2,51 $mol\cdot kg^{-1}$ za raztopine kalijevega in 0,25-1,76 $mol\cdot kg^{-1}$ za raztopine natrijevega acetata. Kot izopiestični standard smo za izračun osmoznih koeficientov uporabili natrijev jodid. Izmerjene vrednosti smo primerjali z vrednostmi dobljenimi s Pitzer-Mayorga in MSA-NRTL setom enačb in s dobljenimi parametri izračunali tudi parni tlak. Ugotovili smo, da se eksperimentalno določeni osmozni koeficienti zadovoljivo ujemajo z izračunanimi, kar nam omogoča tudi zanesljivo oceno vrednosti parnega tlaka.