Vapor-Liquid Equilibrium Properties of Sodium n-Heptyl Sulfonate in Water and in Aqueous Solutions of Poly(ethylene glycol) at Different Temperatures

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The vapor-liquid equilibria properties of sodium n-heptyl sulfonate (C_7SO_3Na) in pure water and in aqueous poly(ethylene glycol) (PEG) solutions were determined at different temperatures below and above the critical micelle concentration (CMC). Vapor-liquid equilibrium data such as water activity, vapor pressure, osmotic coefficient, activity coefficient and Gibbs free energies were obtained through isopiestic method. The concentration dependence of all investigated thermodynamic properties exhibit a change in slope at the concentration in which micelles are formed. It was found that the vapor pressure depression for a ternary aqueous C_7SO_3Na + PEG system is more than the sum of those for the corresponding binary solutions and, at higher temperatures, the higher concentration of PEG is in equilibrium with a certain concentration of surfactant.

INTRODUCTION

It has been generally recognized that the study of thermodynamic and transport properties of surfactants is important to understand their behavior in solutions [1]. Therefore, the limited interest of scientists working in this field for direct measurement of some fundamental solution properties, such as the activity and osmotic coefficients, enthalpy and volume changes on dilution, electrical conductivity, transport numbers, *etc.*, is somewhat unjustifiable. These properties are indispensable for testing various theories and models of surfactant solutions [2]. The interaction between polymers and surfactant in aqueous solutions has also become a very interesting topic for widespread application in such areas as oil recovery, colloid stability, surface modification, wetting, and the physiological transport and metabolism of lipids, and their interaction with

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proteins as well as theoretical studies, and has been investigated for several decades and extensively documented [3]. Surfactants may bind cooperatively to nonionic water-soluble polymers to form micelle-polymer aggregates [4-6], and these interactions are largely confined to anionic surfactants.

Of all the mixed polymer-surfactant systems, sodium alkyl sulfates $[CH_3-(CH_2)_{n-1}-SO_4Na]$, especially sodium dodecyl sulfate ($C_{12}SO_4Na$), are the most frequently used anionic surfactants. Sodium alkyl sulfonates $[CH_3-(CH_2)_{n-1}-SO_3Na]$ are among the most important classes of anionic surfactants which can form the micelle in aqueous solutions, and may also interact with water soluble polymers such as poly(ethylene glycol) (PEG) in aqueous solutions. Some limited thermodynamic properties for binary aqueous solutions of some sodium alkyl sulfonates have been reported in the literature [7-18]. Regarding the mixed polymer-surfactant systems, only few investigations involved sodium alkyl sulfonate [15,19-23]. Although Ortona *et al.* [16] have

performed the measurements of osmotic coefficients on the binary system sodium n-octyl sulfonate + water at 298.15 K, there is, however, no information about vapor-liquid equilibria (VLE) properties of aqueous solutions of sodium alkyl sulfonates at different temperatures as there is none about VLE of polymer-surfactant systems in the literature. In this study, the isopiestic method was used for determination of the osmotic coefficient, water activity, vapor pressure and activity coefficient of binary C_7SO_3Na + water and ternary C_7SO_3Na + PEG + water systems at different temperatures. The activity or osmotic coefficients of different components is of great interest for understanding the interactions in the system. They are the most relevant thermodynamic reference data and are often the starting point of any modeling. However, in colloidal chemistry, these values have been rarely determined, experimentally. Since C_7SO_3Na (or any other anionic surfactants) has some properties similar to the 1:1 electrolytes or polymers, the obtained thermodynamic properties of investigated systems and those of 1:1 electrolyte solutions and of polymer solutions have also been compared.

EXPERIMENTAL

Materials

Sodium n-heptyl sulfonate (C_7SO_3Na) (purity > 99%), sodium chloride (purity > 99.5%), and poly(ethylene glycol) (PEG) were obtained from Merck and were used without further purification. PEG had a nominal molecular weight of 6000. The manufacturer has characterized this polymer with charge/lot number S35317 203. Double distilled, deionized water was used.

Methods

In this study, the isopiestic method was used to obtain the osmotic coefficient of aqueous surfactant mixtures. The apparatus used for determination of water activity in the binary water + C_7SO_3Na solutions consisted of five-leg manifold attached to round-bottom flasks. Two flasks contained the standard pure NaCl solutions, two flasks contained the pure surfactant solutions and the central flask was used as a water reservoir. However, the apparatus used for determination of water activity in the ternary water + PEG + C_7SO_3Na solutions consisted of eight-leg manifold attached to

round-bottom flasks. Two flasks contained the standard pure NaCl solutions, one flask contained the pure PEG solution, one flask contained the pure surfactant solution, three flasks contained the PEG + surfactant solutions and the central flask was used as a water reservoir. The apparatus was kept in a constant-temperature bath for at least 15 days (depending on surfactant concentration) for equilibrium. The temperature was controlled to within ± 0.05 K. After equilibrium had been reached, the manifold assembly was removed from the bath, and each flask was weighed with an analytical balance with a precision of $\pm 1 \times 10^{-4}$ g. From the weight of each flask after equilibrium and the initial weight of salt and polymer, the mass fraction of each solution was calculated. The water activities for the standard aqueous NaCl solutions at different concentrations and temperatures were calculated from the correlation of Colin *et al*. [25]. It was assumed that the equilibrium condition was reached when the differences between the mass fractions of each duplicate were less than 1%. In all cases, averages of the duplicate are reported as the total isopiestic mass fraction. The uncertainty in the measurement of solvent activity was estimated to be $\pm 2 \times 10^{-4}$. The activity of the solvent, a_w, in the reference and in the surfactant solutions must be the same as isopiestic equilibrium and is related to osmotic coefficient, ϕ, as [26]

$$\Phi = -\frac{1000 \ln a_w}{\nu m M_w} \tag{1}$$

where ν is the stoichiometric numbers of the solute, m is the molality of solution, and M_w is the molecular weight of the solvent. The isopiestic equilibrium molalities of the solutions of C_7SO_3Na, which are plotted versus the reference molalities of the standard solutions of NaCl in Fig. 1, enabled the calculation of the osmotic coefficient, ϕ, of the solutions of C_7SO_3Na by using

$$\Phi = \frac{\nu^* \Phi^* m^*}{\nu m} \tag{2}$$

where ν^* and ν are the sums of the stoichiometric numbers of anions and cations in the reference solution and the solution of C_7SO_3Na, respectively, m is the molality of the C_7SO_3Na solution, m^* is the molality of the reference standard in isopiestic equilibrium with this solution, and ϕ^* is the osmotic
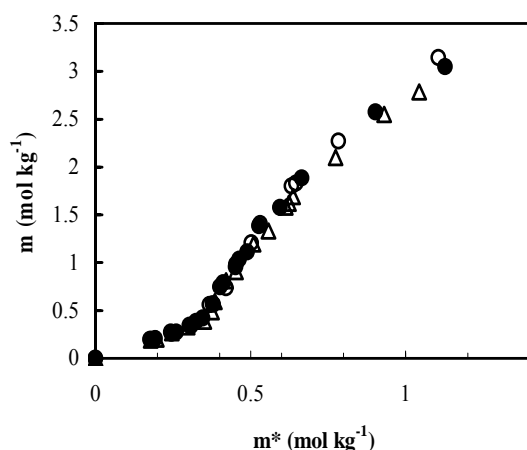
Fig. 1. Plot of the isopiestic equilibrium molality of surfactant, m, against those of NaCl, m^*, for $C_7SO_3Na + H_2O$ system at different temperatures: ○, $T = 298.15$ K; ●, $T = 308.15$ K; △, $T = 318.15$ K.

coefficient of the isopiestic reference standard, calculated at m^*. The necessary ϕ^* values at any m^* and temperatures were obtained from the correlation of Colin *et al*. [25]. From the calculated osmotic coefficient data, the activity of water in surfactant solution and the vapor pressure of this solution were determined at isopiestic equilibrium molalities, with the help of Eq. (1) and the following relation:

$$\ln a_w = \ln\left(\frac{p}{p_w^\circ}\right) + \frac{\left(B_w^\circ - V_w^\circ\right)\left(p - p_w^\circ\right)}{RT} \qquad (3)$$

where B_w° is the second virial coefficient of water vapor, V_w° is the molar volume of liquid water, and p_w° is the vapor pressure of pure water. The second virial coefficients of water vapor were calculated using the equation provided by Rard and Platford [27]. Molar volumes of liquid water were calculated using density of water at different temperatures [28]. The vapor pressures of pure water were calculated using the equation of state of Saul and Wagner [29].

The experimental osmotic coefficients are related to the mean molal activity coefficients, $\gamma_\pm$, at molality m' by the relation [26]:

$$\ln \gamma_\pm = \Phi' - 1 + \int_0^{m'} \frac{\Phi - 1}{m} dm \qquad (4)$$

where ϕ' is the osmotic coefficient of the solution at molality m'.

RESULTS AND DISCUSSION

As can be understood from Fig. 1, at each temperature, the plots of the isopiestic equilibrium molality of surfactant, m, against those of NaCl, m^*, exhibit a change in slope at the concentration in which micelles are formed. Similarly, for the concentrations higher than CMC, the concentration of surfactant which is in equilibrium with a certain concentration of NaCl is larger than those we expect in the absence of formation of micelle. In fact, the confinement of a fraction of the counterions to the micellar surface results in an effective loss of ionic charges, and therefore, the hydration number of monomeric state of surfactant is larger than the hydration number of micellar form of surfactant so that micellization lowers the NaCl concentrations required to achieve a certain water activity.

The experimental water activities, a_w, and the vapor pressure depressions, $p - p_w^\circ$, for binary aqueous surfactant solutions are plotted against the molality of surfactant, in Figs. 2 and 3, respectively. As can be seen from these figures, once micelles are formed, a_w and $p - p_w^\circ$ undergo an abrupt change in concentration dependence. Although water activities of investigated binary solutions do not vary significantly with variation of temperature, as Fig. 3 shows, the vapor pressure depression of these solutions is very sensitive to the variation of temperature. In fact, thermodynamic study of aqueous electrolyte solutions shows that the temperature has only minute effect on the water activity of aqueous electrolyte solutions. However, water activity of aqueous polymer solutions increases with increasing temperature [30-33]. Similar to the aqueous surfactant solutions, vapor pressure depression of both of binary aqueous electrolyte and aqueous polymer solutions increase with increasing temperature [30,33]. In Figs. 4 and 5, respectively, water activities and vapor pressure depressions of aqueous C_7SO_3Na solutions obtained in this work have been compared with those of aqueous 1:1 electrolyte (NaCl and potassium di-hydrogen citrate (KH_2Cit)) and aqueous poly(vinyl pyrrolidone) (PVP) solutions at 308.15 K. As can be seen from Figs. 4 and 5, for NaCl, KH_2Cit and PVP, the data points are smoothly curved
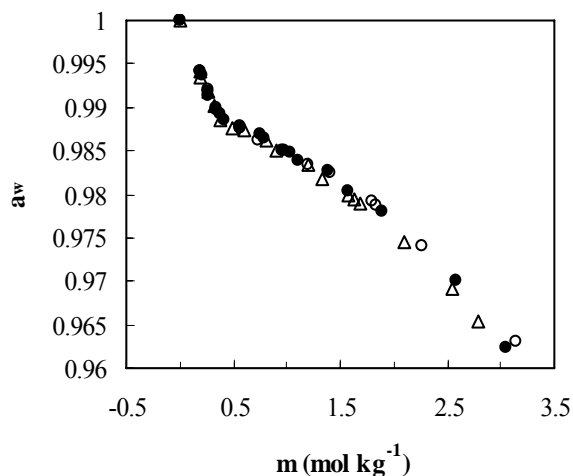
Fig. 2. Plot of water activity, a_w, data against surfactant molality, m, for $C_7SO_3Na + H_2O$ system at different temperatures: ○, $T = 298.15$ K; ●, $T = 308.15$ K; △, $T = 318.15$ K.
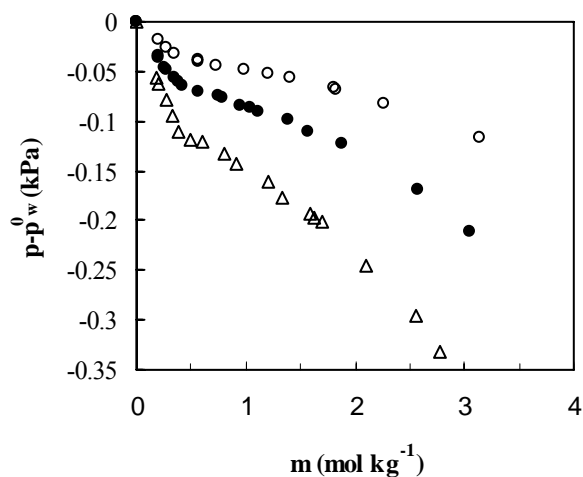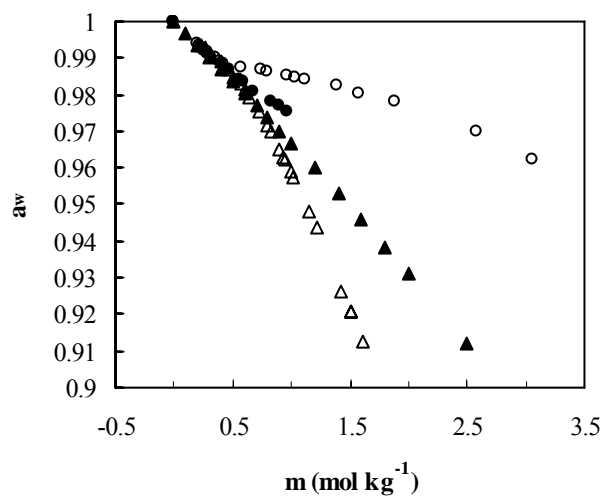

Fig. 4. Plot of water activity data, a_w, against molality of solute, m, at $T = 308.15$ K: ○, $C_7SO_3Na + H_2O$; ●, $KH_2Cit + H_2O$[33]; ▲, $NaCl + H_2O$[34]; △, PVP + H_2O[35].

Fig. 3. Plot of vapor pressure depression data, $p - p_w^\circ$ / kPa, against surfactant molality, m, for $C_7SO_3Na + H_2O$ system at different temperatures: ○, $T = 298.15$ K; ●, $T = 308.15$ K; △, $T = 318.15$ K.

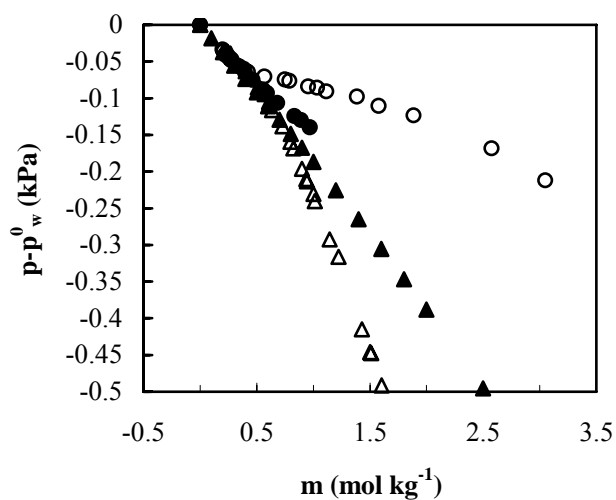**Fig. 5.** Plot of vapor pressure depression data, $p - p_w^\circ$ / kPa, against molality of solute, m, at $T = 308.15$ K: ○, $C_7SO_3Na + H_2O$; ●, $KH_2Cit + H_2O$[33]; ▲, $NaCl + H_2O$[34]; △, PVP + H_2O[35].

and there is no sudden change of slope. Figures 4 and 5 also show that, for concentrations below the CMC of C_7SO_3Na, water activities and vapor pressure depressions of aqueous C_7SO_3Na, NaCl, KH_2Cit and PVP solutions have similar values; however, for concentrations higher than CMC of C_7SO_3Na, water activities and vapor pressure depressions of aqueous C_7SO_3Na solutions are larger than those of other solutions, which indicate that by formation of micelles the interactions between water and surfactant become weaker. The variations of the experimental osmotic coefficient, ϕ, as a function of surfactant molality, for all temperatures, are given in Fig. 6. As can be seen, similar to the other investigated thermodynamic properties, the values of ϕ exhibit a change in slope at the CMC of surfactant.

The water activity was used to obtain the solvent activity coefficient, γ_w. Calculated water activity coefficients, γ_w, of aqueous solutions of C_7SO_3Na are presented as a function of square root of surfactant molality at different temperatures in Fig. 7. As can be seen, the water activity coefficients slightly depend on concentration up to the CMC and at concentrations higher than the CMC, the values of the water activity coefficient increase as a result of the micelle formation. Figure 7 also shows that for concentrations higher than the CMC, γ_w > 1 (positive deviations from ideal-solution behavior) indicate that the solvent-solvent interactions are more favorable than the micelle-solvent interactions.

The variation of $\gamma_\pm$ with the surfactant concentration is shown in Fig. 8. The concentration dependence of the mean molal activity coefficients exhibits a change in slope at the concentration in which micelles are formed. The values of $\gamma_\pm$ for aqueous C_7SO_3Na solutions decreased along with an increase in the surfactant concentration. According to the pseudo-phase model for micelle formation, below the CMC, the concentrations of the free monomers and counterions are equal to total concentration. However, the concentration of surfactant in monomer form at total concentration higher than CMC is constant and equals to the CMC value [36-42]. On the basis of this model, using an extended Pitzer-Debye-Hückel type [43] parametrically linear equation and following the method of Zielinski, *et al.* [42], we can obtain the following equations for the dependence of the mean molal activity coefficients of surfactant solutions on the molality of surfactant below and above CMC:
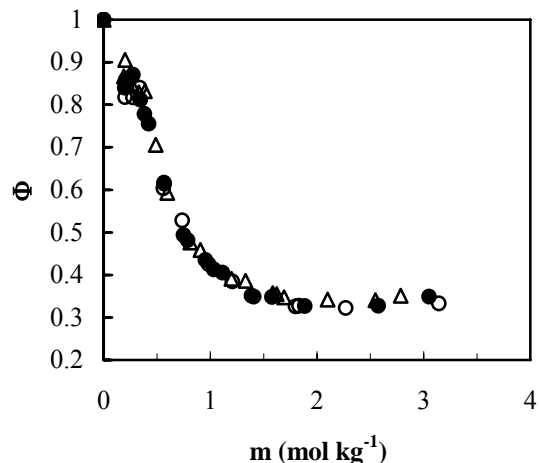
Fig. 6. Plot of osmotic coefficient data, ϕ, against surfactant molality, m, for C_7SO_3Na + H_2O system at different temperatures: $\circ$, $T = 298.15$ K; $\bullet$, $T = 308.15$ K; $\triangle$, $T = 318.15$ K.
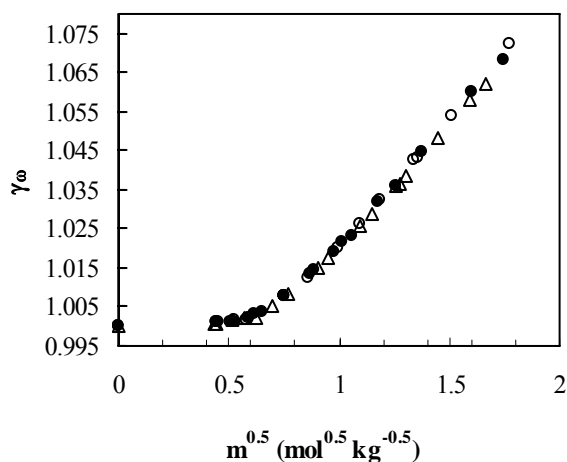
Fig. 7. Variation of water activity coefficient, γ_w, as a function of square root of surfactant molality, $m^{0.5}$, for C_7SO_3Na + H_2O system at different temperatures: $\circ$, $T = 298.15$ K; $\bullet$, $T = 308.15$ K; $\triangle$, $T = 318.15$ K.
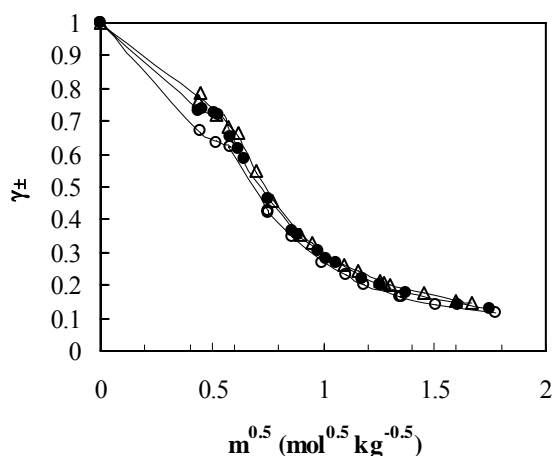
Fig. 8. Variation of mean molar activity coefficient, $\gamma_\pm$, as a function of square root of surfactant molality, $m^{0.5}$, for $C_7SO_3Na + H_2O$ system at different temperatures: $\circ$, $T = 298.15$ K; $\bullet$, $T = 308.15$ K; $\triangle$, $T = 318.15$ K; The solid lines show the results of the fitting of the experimental data to Eq. (5).

$$\ln \gamma_\pm = \frac{-2.303 A_\gamma m^{\frac{1}{2}}}{1 + m^{\frac{1}{2}}} + \sum_{i=2}^{3} A_i m^{\frac{i}{2}}$$
$$m \leq CMC \tag{5a}$$

$$\ln \gamma_\pm = \frac{-2.303 A_\gamma (CMC)^{\frac{1}{2}}}{1 + (CMC)^{\frac{1}{2}}} + \sum_{i=2}^{3} A_i (CMC)^{\frac{i}{2}} + \sum_{i=4}^{7} A_i (m - CMC)^{\frac{i-3}{2}}$$
$$m \geq CMC \tag{5b}$$

where A_γ is the Debye-Hückel constant equal to 0.510, 0.519 and 0.529 kg$^{0.5}$ mol$^{-0.5}$ at 298.15, 308.15 and 318.15 K,

respectively. The coefficients of Eq. (5) containing both A_i and CMC were obtained by the method of least-squares and are given in Table 1. In fact, at each temperature, the CMC as well as A_i values were taken as adjustable parameters. As can be seen from Table 1, the CMC values for C_7SO_3Na in water were obtained as 0.3321, 0.3059 and 0.3959 mol kg^{-1}, respectively, at 298.15, 308.15 and 318.15 K. It was found that the U-shaped temperature dependence of CMC appears to be a rule which has been observed on several occasions [44-47], and the curves of CMC against temperature have a minimum around 303.15-318.15 K.

The following relation was used to obtain the mole fraction mean ionic activity coefficient, $\gamma_\pm^{(x)}$ from the molal mean ionic activity coefficient $\gamma_\pm$:

$$\ln \gamma_\pm^{(x)} = \ln \gamma_\pm + \ln \left(1 + \frac{M_w \upsilon m}{1000}\right) \tag{6}$$

The activity coefficient data, which had been converted to mole fraction scale, were used to calculate the molar excess Gibbs free energy (g^{ex}), and the molar Gibbs free energy change due to mixing (Δg^{mix}) of aqueous surfactant solutions, whose values are shown in Fig. 9. The molar excess Gibbs free energy and the molar Gibbs free energy change due to mixing are negative and decrease with an increase in concentration of the surfactant. As expected, the excess free energies for the solutions are less negative than Δg^{mix}, indicating the importance of enthalpy and entropy effects in these solutions.

In this work, the isopiestic equilibrium weight fractions of different $C_7SO_3Na(s) + PEG (p) + H_2O (w)$ solutions were also obtained at 298.15, 308.15, and 318.15 K. It was found that, water activity of the $PEG + H_2O$ system increases with

Table 1. Coefficients of Eq. (5) for Aqueous Solutions of C_7SO_3Na at Different Temperatures

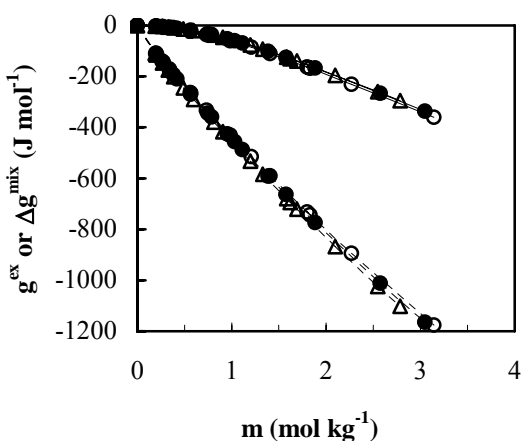| T (K)	A_2	A_3	A_4	A_5	A_6	A_7	CMC (mol kg^{-1})
298.15	-0.4772	0.5971	-3.3552×10^{-5}	-2.3327	1.5829	-0.3269	0.3321
308.15	0.2875	0.0469	-3.3013×10^{-5}	-2.4971	1.7410	-0.3676	0.3059
318.15	1.6874	-2.5399	-3.3591×10^{-5}	-2.4090	1.8193	-0.4300	0.3959

Fig. 9. Variation of molar excess Gibbs free energy, g^{ex}, (solid lines) and molar Gibbs free energy change due to mixing, Δg^{mix}, (dotted lines) as a function of surfactant molality, m, for $C_7SO_3Na + H_2O$ system at different temperatures: $\circ$, $T = 298.15$ K; $\bullet$, $T = 308.15$ K; $\triangle$, $T = 318.15$ K.

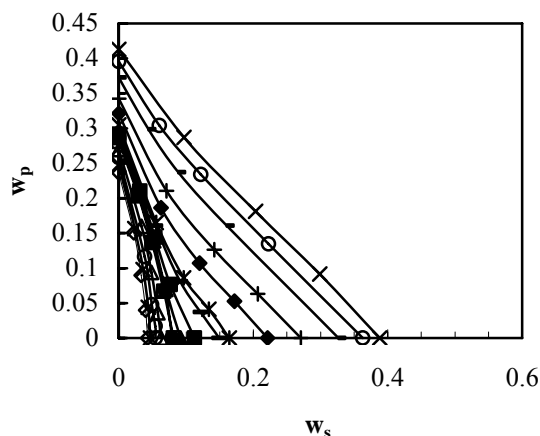**Fig. 10.** Plot of weight fraction of PEG, w_p, against weight fraction of C_7SO_3Na, w_s, for constant water activity curves of $C_7SO_3Na + PEG + H_2O$ system at $T = 298.15$ K: $\diamond$, 0.9925; $\times$, 0.9913; $\circ$, 0.9892; $\triangle$, 0.9883; $\square$, 0.9881; $\bullet$, 0.9876; $\blacktriangle$, 0.9868; $\blacksquare$, 0.9868; $\boldsymbol{-}$, 0.9856; $*$, 0.9851; $\blacklozenge$, 0.9826; +, 0.9783; $\blacksquare$, 0.9728; $\circ$, 0.9675; $\times$, 0.9621.

increasing temperature. From Eq. (3), we conclude that by increasing temperature, the rise of vapor pressure of the PEG + H_2O solution is larger than that of pure water. This occurs because the PEG becomes more hydrophobic with increasing temperature.

The lines of constant water activity or vapor pressure of $C_7SO_3Na + PEG + H_2O$ system at 298.15 K are plotted in Fig. 10. In fact, 5 points on each line in this figure have a constant water activity or chemical potential and thus these points are in equilibrium. Figure 10 shows that, the constant water activity lines exhibit a change in slope at the concentration in which micelles are formed. In other words, for surfactant concentrations higher than CMC, the constant water activity lines have a concave slope, but for concentrations below the CMC, the constant water activity lines have a convex slope. Table 2 shows that the vapor pressure depression for a ternary aqueous $C_7SO_3Na + PEG$ system is more than the sum of those for the corresponding binary solutions. In the aqueous solutions, hydration of segments of PEG will result in a reduction in the free water content, and consequently, in an increase in the effective concentration of the surfactant. Similarly, ionic species of surfactant in the aqueous solutions are hydrated which will result in an increase in the effective concentration of the polymer. Thus, it can be expected that the vapor pressure depression for an aqueous $C_7SO_3Na + PEG$ system to be more than the sum of those for corresponding binary solutions.

Figure 11 shows that, the slope of the constant water activity lines increases with increasing temperature. In other words, at higher temperatures, the higher concentration of PEG is in equilibrium with a certain concentration of surfactant. This is because the composition of the systems in equilibrium changes with varying temperature. At higher temperatures, hydrogen bond interactions of PEG are weakened, and therefore, by increasing the temperature, water is driven from the PEG-rich solutions to vapor phase, and thus, by increasing the temperature of these 5 systems in equilibrium, increase of concentration for the system with higher PEG concentration is larger than those for the systems with less PEG concentration. Figure 11 also shows that the effect of temperature on the constant water activity lines at surfactant concentrations below the CMC is smaller than those at concentrations higher than CMC.

Table 2. Vapor Pressure Depression for Several $C_7SO_3Na(s)$ + PEG (p) + H_2O (w) Solutions Along with Those for Corresponding Binary Solutions

w_p	w_s	$(p - p^0)/kPa$		
		PEG + C_7SO_3Na + H_2O	PEG + H_2O	C_7SO_3Na + H_2O
			$T = 298.15$ K	
0.2872	0.0969	-0.121	-0.039	-0.037
0.2339	0.1222	-0.103	-0.023	-0.042
0.3041	0.0603	-0.103	-0.046	-0.029
			$T = 308.15$ K	
0.3545	0.0434	-0.192	-0.112	-0.039
0.3190	0.0474	-0.159	-0.085	-0.043
0.2981	0.0443	-0.136	-0.071	-0.040
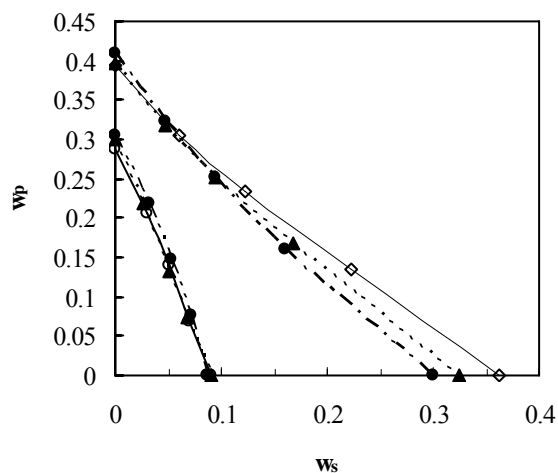
Fig. 11. Constant water activity curves for the C_7SO_3Na (s) + PEG (p) + H_2O (w) system: ○, 298.15 K; ▲, 308.15 K; ●, 318.15 K.

REFERENCES

[1] G.C. Kresheck, in: F. Franks (Ed.), In Water-A Comprehensive Treatise, Plenum Press, New York, Vol. 4, 1975, p. 95.

[2] J. Skerjanc, K. Kogej, J. Cerar, Langmuir 15 (1999) 5023.

[3] E.D. Goddard, In Interactions of Surfactants with Polymers and Proteins, CRC Press: Boca Raton, FL, 1993.

[4] E.D. Goddard, Colloids Surf. 19 (1986) 301.

[5] B. Cabane, J. Phys. Chem. 18 (1977) 1639.

[6] M.L. Fishman, F.R. Elrich, J. Phys. Chem. 79 (1975) 2740.

[7] O. Annunziata, L. Costantino, G. D'Errico, L. Paduano, V. Vitagliano, J. Colloid Interface Sci. 216 (1999) 16.

[8] K. Holmberg, B. Jonsson, B. Kronberg, B. Lindman, Surfactants and Polymers in Aqueous Solutions, John Wiley & Sons Ltd., West Sussex, 2003.

[9] K.A. Wright, A.D. Abbott, V. Sivertz, H.V. Tartar, J. Am. Chem. Soc. 61 (1939) 549.

[10] K.A. Wright, H.V. Tartar, J. Am. Chem. Soc. 61 (1939) 544.

[11] E.C. Lingafelter, O.L. Wheeler, H.V. Tartar, J. Am. Chem. Soc. 68 (1946) 1490.

[12] M. Saito, Y. Moroi, R. Matuura, J. Colloid Interface Sci. 76 (1980) 256.

[13] K. Matsuoka, Y. Moroi, M. Saito, J. Phys. Chem. 97 (1993) 13006.

[14] O. Ortona, L. Paduano, L. Costantino, V. Vitagliano, J. Mol. Liq. 63 (1995) 291.

[15] O. Annunziata, L. Costantino, G. D'Errico, L. Paduano, V. Vitagliano, J. Colloid Interface Sci. 216 (1999) 8.

[16] O. Ortona, G. D'Errico, L. Paduano, V. Vitagliano, J. Solut. Chem. 32 (2003) 1103.

[17] P. Roscigno, L. Paduano, G. D'Errico, V. Vitagliano, Langmuir 17 (2001) 4510.

[18] D.M. Bloor, J. Gormally, E. Wyn-Jones, J. Chem. Soc., Faraday Trans. 1, 80 (1984) 1915.

[19] F. Li, G.Z. Li, G.Y. Xu, H.Q. Wang, M. Wang, Colloid Polym. Sci. 276 (1998) 1.

[20] Z. Hou, Z. Li, H. Wang, Colloid Polym. Sci. 277 (1999) 1011.

[21] O. Ortona, G. D'Errico, L. Paduano, R. Sartorio, Phys. Chem. Chem. Phys. 4 (2002) 2604.

[22] P. Roscigno, G. D'Errico, O. Ortona, R. Sartorio, L. Paduano, Prog. Colloid Polym. Sci. 122 (2003) 113.

[23] P. Yan, J.-X. Xiao, Colloids and Surfaces A: Physicochem. Eng. Aspects 244 (2004) 39.

[24] L.R. Ochs, M. Kabiri Badr, H. Cabezas, AIChE J. 36 (1990) 2908.

[25] E. Colin, W. Clarke, D.N. Glew, J. Phys. Chem. Ref. Data 14 (1985) 489.

[26] G.N. Lewis, M. Randall, in: L. Brewer, K.S. Pitzer (Eds.), Thermodynamics, 2nd ed., McGraw-Hill, New York, 1965.

[27] J.A. Rard, R.F. Platford, in: K.S. Pitzer (Ed.), Activity Coefficients in Electrolyte Solutions, CRC Press, Boca Raton, 1991, pp. 209.

[28] G.S. Kell, J. Chem. Eng. Data 20 (1975) 97.

[29] A. Saul, W.J. Wagner, J. Phys. Chem. Ref. Data 16 (1987) 893.

[30] R. Sadeghi, F. Ziamajidi, J. Chem. Thermodyn. 38 (2006) 1335.

[31] R. Sadeghi, F. Ziamajidi, Fluid Phase Equilib. 249 (2006) 165.

[32] R. Sadeghi, F. Ziamajidi, Fluid Phase Equilib. 255 (2007) 46.

[33] R. Sadeghi, B. Goodarzi, Biophys. Chem. 135 (2008) 116.

[34] R.A. Robinson, R.H. Stokes, Electrolyte Solutions, 2nd ed., Butterworths, London, 1965.

[35] R. Sadeghi, M.T. Zafarani-Moattar, J. Chem. Thermodyn. 36 (2004) 665.

[36] P. Mukerjee, Adv. Coll. Interface Sci. 1 (1967) 242.

[37] G. Stainsby, A.E. Alexander, Trans. Faraday Soc. 46 (1952) 587.

[38] K. Shinoda, E. Hutchinson, J. Phys. Chem. 66 (1962) 577.

[39] K. Shinoda, T. Nakagawa, B. Tamamushi, T. Isemura, Colloid Surfactants, Some Physicochemical Properties, Academic Press, New York, 1963.

[40] M.E. Friedmann, H.A. Scheraga, J. Phys. Chem. 69 (1965) 3795.

[41] D.M. Bloor, J. Gormally, E. Wyn-Jones, J. Chem. Soc., Faraday Trans. 1, 80 (1984) 1915.

[42] R. Zielinski, S. Ikeda, H. Nomura, S. Kato, J. Chem. Soc., Faraday Trans. 1, 84 (1988) 151.

[43] K.S. Pitzer, J. Phys. Chem. 77 (1973) 268.

[44] J.L. del Castillo, J. Czapkiewicz, A. Gonzalez Perez, J.R. Rodriguez, Colloids and Surfaces A: Physicochem. Eng. Aspects 166 (2000) 161.

[45] A. Gonzalez-Perez, J.L. del Castillo, J. Czapkiewicz, J.R. Rodriguez, J. Phys. Chem. B 105 (2001) 1720.

[46] T.-M. Perger, M. Bester-Rogac, J. Colloid Interface Sci. 313 (2007) 288.

[47] R. Zielinski, J. Colloid Interface Sci. 235 (2001) 201.