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# Isobaric Vapor−Liquid Equilibria and Excess Volumes for the Binary Mixtures Water + Sulfolane, Water + Tetraethylene Glycol, and Benzene + Tetraethylene Glycol

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Isobaric vapor−liquid equilibrium data were determined at 101.33 kPa for the binary mixtures of water + sulfolane and water + tetraethylene glycol. Calculations of the nonideality of the vapor phase were made with the second virial coefficients obtained from the Hayden−O'Connell method. The boiling points and vapor-phase compositions reported were well correlated by the Wilson, NRTL, and UNIQUAC models. In addition, the densities of binary mixtures of water + sulfolane, water + tetraethylene glycol, and benzene + tetraethylene glycol were determined over the entire concentration range at 298.15 K and atmospheric pressure by using an oscillating tube densimeter. Excess volumes were calculated for each data point. All mixtures exhibit negative excess volumes with a minimum which occurs at $x \cong 0.5$ for the benzene + tetraethylene glycol system; it is shifted toward the water-rich region for the water + sulfolane and water + tetraethylene glycol systems. The experimental excess volumes were correlated using the Redlich−Kister equation.

## Introduction

Solvent extraction is one of the most important methods to produce high-purity aromatic extracts from catalytic reformates. In recent years, sulfolane or tetraethylene glycol has been employed more and more in new or improved extraction processes. Therefore, it is necessary to have complete thermodynamic data for these systems. Several researchers[1−4] have studied the vapor−liquid and liquid−liquid equilibria for mixtures containing an aromatic and sulfolane or tetraethylene glycol. Herskowitz and Gottlieb[5] reported the activity coefficients of the solvent in the water + tetraethylene glycol system by an isopiestic method at 298.15 K.

For the volumetric properties, excess volumes for a nitrile + sulfolane[6,7] and an aromatic hydrocarbon + sulfolane[2,8] have been well documented for a reasonable range of temperature. To our knowledge, for the systems involving tetraethylene glycol, only aqueous solutions and solutions with HFC-134a have been studied.[9] Lepori and Mollica[10] have reported the volumetric properties of dilute aqueous solutions of tetraethylene glycol, and some density data for this system have been given by Müller and Rasmussen.[11]

In this work, the isobaric vapor−liquid equilibrium data for the mixtures of water + sulfolane and water + tetraethylene glycol were measured at 101.33 kPa, and the excess volumes of water + sulfolane, water + tetraethylene glycol, and benzene + tetraethylene glycol systems were reported at 298.15 K, covering the whole concentration range.

## Experimental Section

***Chemicals.*** Pure benzene and sulfolane were obtained from Beijing Chemical Engineering Plant. Tetraethylene glycol was procured from Beijing Yanshan Petrochemical Company. All the chemicals were purified by distillation (sulfolane and tetraethylene glycol distilled under vacuum), and their middle fractions were collected. Then they were dried using type 0.5 nm molecular sieves and the water content was found to be <0.01 mass%, as determined with a Mitsubishi moisture meter (Model CA-05). To minimize the contact of these deliquescent reagents with moist air, all the purified chemicals were kept in sealed bottles in desiccators. Doubly distilled water was used in all solutions.

**Table 1. Densities ($\rho$) and Refractive Indices ($n_D$) of Pure Components and Comparison with Literature Values at 298.15 K**

| | $\rho$/g·cm$^{-3}$ | | $n_D$ | |
|---|---|---|---|---|
| component | exptl | lit. | exptl | lit. |
| benzene | 0.873 51 | 0.873 60[a] | 1.4979 | 1.49792[a] |
| sulfolane | 1.265 64[b] | 1.264 0[a,b]<br>1.266 00[c] | 1.4819[d] | 1.4816[a,d] |
| tetraethylene glycol | 1.120 06 | 1.120 30[e]<br>1.119 3[f] | 1.4570 | 1.4570[g] |

[a] Riddick et al.[12] [b] Extrapolated value. [c] Interpolated data from Jannelli et al.[6] [d] Data at 303.15 K. [e] Data from Müller and Rasmussen.[11] [f] Interpolated data from Kumagai et al.[9] [g] Data from Daubert and Danner.[13]

The purity was tested by GLC, which indicated a minimum purity of 99.8 mol %, and no appreciable peaks of impurities were detected. The densities and refractive indices of the purified chemicals are reported in Table 1 in comparison with the literature data at 298.15 K and atmospheric pressure.

***Vapor−Liquid Equilibrium Measurements.*** An inclined ebulliometer with a pumplike stirrer developed by Zhou et al.[14] and described by Yu et al.[3] was used. It is a recirculation type, in which both liquid and vapor phase recirculate continuously, and the equilibrium compositions of both phases can be determined. The ebulliometer was operated at atmospheric pressure (101.33 kPa). The steady state was usually reached after 1.5 h of operation. The temperature in the equilibrium chamber was measured

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using a mercury thermometer. The uncertainty in the temperature measurement is ±0.05 K.

The compositions of the sampled liquid phase and vapor phase were analyzed by measuring their densities at 298.15 K with a vibrating tube densimeter, after calibration with gravimetrically prepared standard solutions. The accuracy of the equilibrium composition measurements was ±0.0003 mole fraction. Since there is a large difference between the boiling points of water and of tetraethylene glycol, the amount of tetraethylene glycol present in the vapor phase was so small that is was not detected by the densimeter. Therefore, the vapor-phase composition was not measured for the water + tetraethylene glycol system.

***Density Measurements.*** An Anton Paar (Graz, Austria) DMA60 oscillating tube densimeter equipped with a density measuring cell (DMA602) was used to measure densities of pure components and binary mixtures at 298.15 K. The temperature of the U-shaped tube was checked continuously using a calibrated digital thermometer (Anton Paar DT100−20) with an accuracy of ±0.01 K. The system was maintained at constant temperature to within ±0.005 K by means of a Hetotherm ultrathermostated water bath (Heto, Type CB7). All the measurements were carried out at atmospheric pressure.

The density determination is based on measuring the period of oscillation of a vibrating U-shaped hollow tube that is filled with the sample. The relation between the period $\tau$ and density $\rho$ is

$$\rho = A(\tau^2 - B) \tag{1}$$

where $A$ and $B$ are temperature-dependent constants determined by calibration with doubly distilled water and dry air. The precision for the measured period $\tau$ is $1 \times 10^{-6}$ s, which leads to a precision in densities and excess molar volumes of $\pm 5 \times 10^{-5}$ g·cm$^{-3}$ and $\pm 3 \times 10^{-3}$ cm$^3$·mol$^{-1}$, respectively.

The apparatus, the mixture standard sample preparation, and the procedure of density measurements have been described elsewhere.[15,16]

The density $\rho$ of the binary solutions was used to calculate the excess molar volume $V^E$, according to

$$V^E/\text{cm}^3\cdot\text{mol}^{-1} = \frac{x_1M_1 + x_2M_2}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \tag{2}$$

where $x_i$ is the mole fraction, $M_i$ is the molar mass, and $\rho_i$ is the density of component $i$.

## Results and Discussion

Isobaric vapor−liquid equilibria were measured at 101.33 kPa for water + sulfolane and water + tetraethylene glycol systems. The results are shown in Tables 2 and 3, respectively. The liquid-phase activity coefficients $\gamma_i$ were calculated with

$$\phi_i y_i P = \gamma_i x_i P_i^{\text{sat}}\, \phi_i^{\text{sat}} \exp\left[\frac{V_i^L(P - P_i^{\text{sat}})}{RT}\right] \tag{3}$$

where $\phi_i$ and $\phi_i^{\text{sat}}$ are the fugacity coefficients of component $i$ in the mixture and pure vapor, respectively. $T$ is the equilibrium temperature, $P$ is the pressure, $P_i^{\text{sat}}$ is the saturated vapor pressure, $V_i^L$ is the saturated liquid molar volume, $x_i$ is the liquid-phase molar fraction, and $y_i$ is the vapor-phase molar fraction of component $i$.

The vapor pressures of the pure components were obtained using fitted parameters from Reid et al.,[17] Riddick et al.,[12] and Stephenson and Malanowski.[18] These are reproduced in Table 4. The fugacity coefficient was calculated by using the virial equation of state truncated after the second virial term. The second virial coefficient was determined by the Hayden−O'Connell method.[19] The critical properties and other parameters required for estimating the second virial coefficients, obtained from Domanska et al.,[20] Nikitin et al.,[21] Prausnitz et al.,[22] Steele et al.,[23] and Yu et al.,[3] are listed in Table 5. The value of the solvation parameter is taken as zero for the water + sulfolane system and 1.55 for the water + tetraethylene glycol system. The liquid molar volumes were calculated from the Hankinson−Brobst−Thomson equation.[17]

**Table 2. Vapor−Liquid Equilibrium Data for Water (1) + Sulfolane (2) Mixtures at 101.33 kPa**

| $T$/K | $x_1$ | $y_1$ [a] | $\gamma_1$ [b] | $\gamma_2$ [b] |
|---|---|---|---|---|
| 373.15 | 1.0000 | 1.0000 | 1.000 | |
| 373.45 | 0.9902 | 0.9999 | 1.000 | 5.318 |
| 373.77 | 0.9786 | 0.9998 | 1.001 | 4.786 |
| 374.18 | 0.9620 | 0.9997 | 1.003 | 3.955 |
| 375.18 | 0.9153 | 0.9995 | 1.018 | 2.803 |
| 376.51 | 0.8372 | 0.9993 | 1.063 | 1.902 |
| 379.00 | 0.6751 | 0.9988 | 1.210 | 1.433 |
| 381.86 | 0.5502 | 0.9982 | 1.348 | 1.339 |
| 383.20 | 0.5041 | 0.9979 | 1.407 | 1.323 |
| 385.78 | 0.4331 | 0.9975 | 1.504 | 1.210 |
| 390.48 | 0.3423 | 0.9968 | 1.636 | 1.058 |
| 396.82 | 0.2653 | 0.9952 | 1.731 | 1.050 |
| 402.58 | 0.2192 | 0.9934 | 1.760 | 1.041 |
| 413.38 | 0.1522 | 0.9885 | 1.853 | 1.040 |
| 423.11 | 0.1112 | 0.9825 | 1.940 | 1.009 |
| 454.23 | 0.0484 | 0.9409 | 2.019 | 1.005 |
| 560.51 | 0.0000 | 0.0000 | | 1.000 |

[a] Experimental values. [b] Calculated from the experimental data.

**Table 3. Vapor−Liquid Equilibrium Data for Water (1) + Tetraethylene Glycol (2) Mixtures at 101.33 kPa**

| $T$/K | $x_1$ | $y_1$ [a] | $\gamma_1$ [b] | $\gamma_2$ [a] |
|---|---|---|---|---|
| 373.15 | 1.0000 | 1.000 00 | 1.000 | |
| 373.58 | 0.9937 | 1.000 00 | 0.992 | 0.686 |
| 373.75 | 0.9891 | 1.000 00 | 0.991 | 0.690 |
| 374.79 | 0.9596 | 1.000 00 | 0.985 | 0.713 |
| 375.21 | 0.9465 | 1.000 00 | 0.984 | 0.723 |
| 376.10 | 0.9196 | 1.000 00 | 0.982 | 0.743 |
| 377.11 | 0.8907 | 1.000 00 | 0.980 | 0.763 |
| 378.65 | 0.8506 | 1.000 00 | 0.973 | 0.790 |
| 384.11 | 0.7242 | 0.999 99 | 0.952 | 0.862 |
| 387.36 | 0.6668 | 0.999 98 | 0.931 | 0.888 |
| 395.60 | 0.5175 | 0.999 93 | 0.926 | 0.941 |
| 404.77 | 0.3938 | 0.999 82 | 0.924 | 0.969 |

[a] Calculated values from NRTL equation. [b] Calculated from the experimental data when tetraethylene glycol in the vapor phase was neglected.

**Table 4. Coefficients Used in the Determination of the Pure Vapor Pressure of the Component**

| component | $A$ | $B$ | $C$ | $D$ |
|---|---|---|---|---|
| water[a] | −7.764 51 | 1.45838 | −2.77580 | −1.233 03 |
| sulfolane[b] | 27.807 3 | 4350.7 | 6.5633 | |
| tetraethylene glycol[c] | 9.447 60 | 4051.160 | −36.521 | |

[a] Coefficients[17] of the equation $\ln(P^{\text{sat}}/P_c) = (1 - x)^{-1}(Ax + Bx^{1.5} + Cx^3 + Dx^6)$, where $x = 1 - T/T_c$, $P^{\text{sat}}$ is the saturated pressure, $P_c$ is the critical pressure, and $T_c$ is the critical temperature. [b] Coefficients[12] of the equation $\log(P^{\text{sat}}/\text{kPa}) = A - B/(T/\text{K}) - C \log(T/\text{K})$. [c] Coefficients[18] of the equation $\log(P^{\text{sat}}/\text{kPa}) = A - B/(T/\text{K} + C)$.

The calculated activity coefficients for the water + sulfolane mixture are also listed in Table 2. The experimental vapor−liquid equilibrium data for the water + sulfolane system were shown to be thermodynamically

**Table 5. Critical Properties and Parameters Used in the Determination of the Second Virial Coefficients in the Vapor Phase, Where $T_c$ Is the Critical Temperature, $P_c$ Is the Critical Pressure, RD Is the Mean Radius of Gyration, DM Is the Dipole Moment, and $\eta$ Is the Association Parameter**

| component | $T_c$/K | $P_c$/MPa | RD/nm | DM/D | $\eta$ |
|---|---|---|---|---|---|
| water[a] | 647.37 | 22.120 | 0.0615 | 1.83 | 1.70 |
| sulfolane | 855.0[b] | 7.290[b] | 0.2910[c] | 4.10[c] | 0.00 |
| tetraethylene glycol | 800.0[d] | 3.200[d] | 0.5054[e] | 5.84[e] | 1.55[e] |

[a] Data from Prausnitz et al.[22] [b] Data from Steele et al.[23] [c] Data from Domanska et al.[20] [d] Data from Nikitin et al.[21] [e] Data from Yu et al.[3]

**Table 6. UNIQUAC Pure Component Parameters for Volume ($r$) and Surface Area ($q$) of Each Compound[29]**

| component | $r$ | $q$ |
|---|---|---|
| water | 0.920 | 1.400 |
| sulfolane | 4.036 | 3.206 |
| tetraethylene glycol | 7.187 | 6.208 |

**Table 7. Correlation Parameters and Absolute Average Deviations in Boiling Temperature ($\Delta T$) and Vapor-Phase Mole Fraction ($\Delta y$) for the Binary Mixtures**

| model[a] | $A_{12}$/K[b] | $A_{21}$/K[b] | $\alpha_{12}$ | $\Delta y$[c] | $\Delta T$ (%)[d] |
|---|---|---|---|---|---|
| Water (1) + Sulfolane (2) | | | | | |
| Wilson | 824.61 | −265.24 | | 0.0006 | 0.165 |
| NRTL | 585.55 | 12.43 | 0.47 | 0.0002 | 0.041 |
| UNIQUAC | −269.99 | 453.30 | | 0.0002 | 0.139 |
| Water (1) + Tetraethylene Glycol (2) | | | | | |
| Wilson | 865.91 | −959.28 | | | 0.146 |
| NRTL | 283.04 | −294.44 | 0.47 | | 0.118 |
| UNIQUAC | −401.62 | 890.08 | | | 0.160 |

[a] All models in ln $\gamma$ form. [b] Energy parameters $A_{ji} = (\lambda_{ji} - \lambda_{ii})/R$ for the Wilson model, $A_{ji} = (g_{ji} - g_{ii})/R$ for the NRTL model, and $A_{ji} = (U_{ji} - U_{ii})/R$ for the UNIQUAC model. [c] $\Delta y = \sum_1^N |y_i^{\text{exp}} - y_i^{\text{calc}}|/N$. [d] $\Delta T = (100/N)\sum_1^N |T_i^{\text{exp}} - T_i^{\text{calc}}|/T_i^{\text{exp}}$, where $N$ is the number of experimental points.

consistent by using a Herington analysis[24] with $(D - J) < 10$ and the test described by Fredenslund et al.[25] The activity coefficients of water in tetraethylene glycol were calculated by neglecting the amount of tetraethylene glycol present in the vapor phase. The values of $\gamma_i$ for the water + tetraethylene glycol system are <1. This is in agreement with the result of Herskowitz and Gottlieb[5] with an isopiestic method at 298.15 K.

The experimental vapor−liquid equilibrium data were then used to obtain the binary parameters in various activity coefficient models. The Wilson,[26] NRTL,[27] and UNIQUAC[28] models were used in the regressions. The UNIQUAC pure component parameters for the surface area ($q$) and volume ($r$) of the molecules were calculated from Walas[29] and are listed in Table 6. The following objective function was minimized in the regressions:

$$\text{OF} = \sum_{j=1}^{N} 100\left[\frac{|T_j^{\text{exp}} - T_j^{\text{calc}}|}{T_j^{\text{exp}}}(1 - w) + w\left|y_j^{\text{exp}} - y_j^{\text{calc}}\right|\right] \quad (4)$$

where $N$ is the number of experimental points, $w = 0.5$ for the water + sulfolane system, and $w = 0$ for the water + tetraethylene glycol system because the vapor-phase composition for the latter system was not measured in this work. The regression results are shown in Table 7. The deviations in vapor-phase compositions and equilibrium temperatures are reasonably small, and this indicates that all three activity coefficient models are suitable to represent the binary experimental data. The comparison between the experimental activity coefficients and the prediction by the NRTL model for the systems of water + sulfolane and water + tetraethylene glycol is shown in Figures 1 and 2.

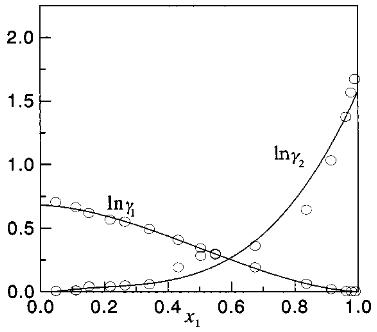

**Figure 1.** Activity coefficient curves for the system water (1) + sulfolane (2): comparison of experimental data (○) with the prediction of the NRTL model (—).

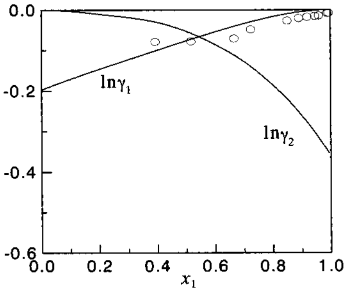

**Figure 2.** Activity coefficient curves for the system water (1) + tetraethylene glycol (2): comparison of experimental data (○) with the prediction of the NRTL model (—).

**Table 8. Densities ($\rho$) and Excess Molar Volumes ($V^E$) for Binary Mixtures at 298.15 K**

| $x_1$ | $\rho$/ g·cm$^{-3}$ | $V^E$/ cm$^3$·mol$^{-1}$ | $x_1$ | $\rho$/ g·cm$^{-3}$ | $V^E$/ cm$^3$·mol$^{-1}$ |
|---|---|---|---|---|---|
| Water (1) + Sulfolane (2) | | | | | |
| 0.0725 | 1.261 75 | −0.003 | 0.6025 | 1.209 39 | −0.156 |
| 0.1148 | 1.259 24 | −0.005 | 0.6790 | 1.193 61 | −0.181 |
| 0.1764 | 1.255 26 | −0.009 | 0.7374 | 1.177 76 | −0.184 |
| 0.2149 | 1.252 52 | −0.011 | 0.8331 | 1.141 22 | −0.174 |
| 0.2730 | 1.248 01 | −0.017 | 0.8645 | 1.127 45 | −0.162 |
| 0.3438 | 1.241 92 | −0.035 | 0.9008 | 1.101 55 | −0.141 |
| 0.4169 | 1.234 61 | −0.058 | 0.9351 | 1.074 05 | −0.112 |
| 0.4651 | 1.229 21 | −0.082 | 0.9754 | 1.031 17 | −0.052 |
| 0.5011 | 1.224 73 | −0.101 | 0.9872 | 1.015 84 | −0.031 |
| 0.5532 | 1.217 33 | −0.125 | 0.9955 | 1.003 99 | −0.013 |
| Water (1) + Tetraethylene Glycol (2) | | | | | |
| 0.0098 | 1.120 01 | −0.012 | 0.7667 | 1.109 08 | −0.999 |
| 0.0882 | 1.119 68 | −0.121 | 0.8402 | 1.099 54 | −0.898 |
| 0.1675 | 1.119 46 | −0.254 | 0.8761 | 1.090 85 | −0.786 |
| 0.2370 | 1.119 24 | −0.371 | 0.9054 | 1.080 17 | −0.653 |
| 0.2925 | 1.119 04 | −0.464 | 0.9373 | 1.063 50 | −0.480 |
| 0.4229 | 1.118 23 | −0.664 | 0.9659 | 1.040 56 | −0.278 |
| 0.4359 | 1.118 10 | −0.681 | 0.9797 | 1.025 67 | −0.170 |
| 0.5363 | 1.117 10 | −0.828 | 0.9890 | 1.013 68 | −0.093 |
| 0.6047 | 1.116 08 | −0.921 | 0.9943 | 1.006 01 | −0.048 |
| 0.7168 | 1.112 42 | −1.006 | | | |
| Benzene (1) + Tetraethylene Glycol (2) | | | | | |
| 0.0667 | 1.111 89 | −0.090 | 0.5371 | 1.030 79 | −0.377 |
| 0.1235 | 1.104 35 | −0.146 | 0.6183 | 1.010 68 | −0.341 |
| 0.1685 | 1.098 08 | −0.195 | 0.6670 | 0.997 42 | −0.310 |
| 0.2134 | 1.091 50 | −0.242 | 0.7296 | 0.978 81 | −0.252 |
| 0.2539 | 1.085 24 | −0.279 | 0.7970 | 0.956 68 | −0.184 |
| 0.3326 | 1.072 13 | −0.336 | 0.8664 | 0.931 36 | −0.119 |
| 0.3860 | 1.062 45 | −0.365 | 0.8920 | 0.921 26 | −0.093 |
| 0.4368 | 1.052 54 | −0.379 | 0.9936 | 0.903 89 | −0.052 |
| 0.5015 | 1.038 87 | −0.383 | | | |

**Table 9. Values of the Coefficients of the Correlation Equation and Standard Deviations for Excess Molar Volumes at 298.15 K**

| system | $a_1$ | $a_2$ | $a_3$ | $a_4$ | $a_5$ | $\sigma$/cm$^3$·mol$^{-1}$ |
|---|---|---|---|---|---|---|
| water + sulfolane | −0.4055 | 0.9482 | −0.4255 | −1.9606 | −0.3636 | 0.0047 |
| water + tetraethylene glycol | −3.1010 | 2.9614 | −2.8107 | 1.2578 | 0.8707 | 0.0051 |
| benzene + tetraethylene glycol | −1.5248 | −0.1804 | 0.6230 | −0.1367 |  | 0.0036 |

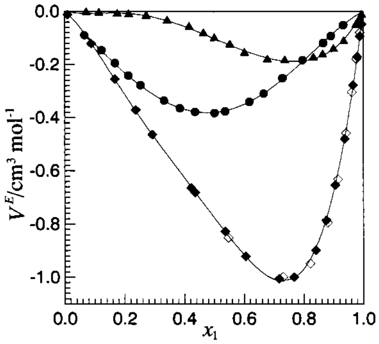

**Figure 3.** Excess molar volumes as a function of composition at 298.15 K for binary systems. Experimental points: (▲) water (1) + sulfolane (2) (this work); (●) benzene (1) + tetraethylene glycol (2) (this work); (◆) water (1) + tetraethylene glycol (2) (this work); (◇) water (1) + tetraethylene glycol (2) (Müller and Rasmussen[11]). The curves were calculated with eq 3.

The experimental densities and excess molar volumes for the binary mixtures of water + sulfolane, water + tetraethylene glycol, and benzene + tetraethylene glycol at 298.15 K are given in Table 8. The excess molar volume as a function of composition is graphically represented in Figure 3. The measured excess molar volumes $V^E$ were fitted to the following expression by a least-squares method:

$$V^E = x_1(1 - x_1)\sum_{j=1}^{m} a_j(1 - 2x_1)^{j-1} \tag{5}$$

Coefficients $a_j$ are given in Table 9 along with the standard deviation $\sigma$ for each system, defined as

$$\sigma^2 = \frac{1}{N - m}\sum_{j=1}^{N}(V_j^{E,\text{calc}} - V_j^{E,\text{exp}})^2 \tag{6}$$

where $m$ is the number of adjustable parameters $a_j$ in eq 5. The superscripts calc and exp refer to the values calculated by using eqs 5 and 2, respectively.

The literature excess volumes for the water + tetraethylene glycol system reported by Müller and Rasmussen[11] are also included in Figure 3. Good agreement was seen between the literature values and the present data.

It can be seen from Table 8 that, for all cases, the excess volumes are negative. For the benzene + tetraethylene glycol system, the minimum excess molar volume occurs at $x \cong 0.5$, and it shifted toward the water-rich region for the water + sulfolane and water + tetraethylene glycol systems, as shown in Figure 3. Excess molar volumes for aqueous tetraethylene glycol solutions are smaller than those for the other two systems studied; this can be interpreted as an increase in the order of the system due to the formation of hydrogen bonds between water and tetraethylene glycol molecules.

## Conclusion

Vapor−liquid equilibria of two binary mixtures of water + sulfolane and water + tetraethylene glycol were measured at 101.33 kPa and tested by using the Herington analysis[24] and the method of Frendenslund et al.[25] Analysis of the experimental vapor−liquid equilibrium data for the two binary systems (by using the Wilson, NRTL, and UNIQUAC models) shows that all three models generally give satisfactory results. Densities and excess volumes for three mixtures of water + sulfolane, water + tetraethylene glycol, and benzene + tetraethylene glycol were determined at 298.15 K. The excess volumes for the three binary systems were successfully correlated using the Redlich−Kister equation.

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