# Dynamic Viscosity Study of Binary Mixtures Triethylamine + Water at Temperatures Ranging from (283.15 to 291.35) K

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Densities, $\rho$, and dynamic viscosities, $\eta$, of triethylamine + water (TEA−W) binary mixtures have been determined experimentally in the single phase region and over the whole composition range at $T = (283.15$ to $291.35)$ K. The experimental measurements of these properties were carried out at atmospheric pressure. Results have been fitted to obtain the adjustable parameters and standard deviations between the measured and fitted values, respectively. The viscosity data have been also correlated with the equations of Grunberg−Nissan, Hind et al., Katti−Chaudhri, Heric−Brewer, McAllister, and Auslander.

## Introduction

The properties of liquid mixtures have attracted much attention from both theoretical and practical points of view. Many engineering problems require quantitative data on the density and viscosity of liquid mixtures.

The present work is a continuation of our earlier studies[1,2] on thermodynamic and transport properties of binary liquid mixtures that split into two phases from single phase upon changing temperature. Here we continue our study of the binary mixture triethylamine + water (TEA−W). This studied mixture is very interesting from an experimental as well as theoretical point of view because the TEA−W mixture is known to be a typical partially miscible system that has a lower critical solution temperature (LCST) that splits into two phases with a temperature increase.[3,4] Moreover, trialkylamines are industrially important liquids that are used as intermediates in the production of polymers such as phenolic resins, corrosion inhibitors, agrochemicals, lubricating additives, pharmaceuticals, textile dyes, and paints.[5]

A detailed search in the literature shows that some measurements have been made on the density and the viscosity for the TEA−W mixture.[6−8] However, they are still not available with the necessary accuracy. Furthermore, these experiments were carried out on one hand in the single phase and far away from the critical region[6] and on the other hand in the two-phase region.[8] This prompted us to carry out precise measurements of these physical properties under atmospheric pressure in the neighborhood of the critical temperature $T_c$ and relatively far from this temperature in the single-phase region ($T < T_c$).

In the present article, we report densities, $\rho$, and viscosities, $\eta$, of (TEA−W) system over the entire range of composition from $T = (283.15$ to $291.35)$ K (very close to the (TEA−W) critical temperature that was identified visually in our laboratory to be $T_c = 291.426$ K when the critical opalescence was well developed) at the critical composition $C_c = 32.1$ % of TEA mass fraction.[12] This work will also provide a test of various semiempirical equations to correlate viscosity of binary mixtures. The types of used relations are Grunberg−Nissan, Hind et al., Katti−Chaudhri, Heric−Brewer, McAllister, and Auslander.

## Experimental Section

***Chemicals.*** The triethylamine (TEA) was purchased from Prolabo. The purity was stated to be 99.95 mol %. The TEA was used as received without further purification. The water used in this study was obtained from deionized and three-times-distilled operations.

***Apparatus and Procedure.*** All mixtures were prepared from weighed amounts of the pure components. The weight was performed by using a digital electronic balance (Sartorius BP 221S) with an uncertainty of $10^{-4}$ g covering the whole composition range of the mixture. Some care was taken to avoid moisture and dust in the final sample, namely, baking the syringes and the cells overnight under vacuum and preparing the mixtures in a dust-free area. The mole fraction was accurate up to the fourth place of decimal. The cell, in which TEA and water were mixed together, was immersed in a thermally stabilized water bath with thermal regulation on the order of 0.002 K over hours. We measured the temperature by using a quartz thermometer (HP 2804 A) giving an uncertainty of 0.001 K that was calibrated on an absolute scale within 0.01 K.

The densities were measured with an Anton Paar digital precision densimeter (model DMA46) that was modified in our laboratory by controlling temperature with an accuracy of ± 0.01 K across a computer processing using a graphical interface. The density, $\rho$ ($\text{g}\cdot\text{cm}^{-3}$), of a solution was calculated from the electronically measured frequency of a mechanical oscillator filled with the fluid. The filling was operated by means of medical syringes. The oscillator was a U-shaped glass tubing (volume of 0.7 $\text{cm}^3$) placed in a metal block. The published density values data for both water[10,11] and methanol[12] were used to calibrate the densimeter. The response time of the apparatus to temperature changes is much higher than the time needed to reach a stable signal. To avoid systematic errors when measuring densities of pure liquids at different temperatures, each temperature change was followed by a waiting time of 2 h. The accuracy of density measurements was $\pm\ 10^{-4}\ \text{g}\cdot\text{cm}^{-3}$.

The kinematic viscosities of the pure liquids and their mixtures were measured with an (AVS/N-Schott-Geräte) Ubbelhode vis-

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**Table 1. Comparison of Measured Densities and Viscosities of Pure Components: Water and TEA with Literature Values**

| | | $\rho/\mathrm{g\cdot cm^{-3}}$ | | $\eta/\mathrm{mPa\cdot s}$ | |
|---|---|---|---|---|---|
| liquid | $T$/K | experimental | literature[10] | experimental | literature[10] |
| water | 283.15 | 0.9995 | 0.9997 | 1.3054 | 1.3055 |
| | 286.15 | 0.9993 | 0.9993 | 1.2002 | 1.2001 |
| | 288.15 | 0.9990 | 0.9991 | 1.1372 | 1.1372 |
| | 290.15 | 0.9987 | 0.9987 | 1.0797 | 1.0795 |
| | 291.15 | 0.9985 | 0.9985 | 1.0524 | 1.0524 |
| | | experimental | literature | experimental | literature |
| TEA | 288.15 | 0.7318 | 0.7318[6]<br>0.7319[14] | 0.3989 | 0.3830[6] |
| | 289.15 | 0.7308 | 0.7304[15] | 0.3940 | 0.3937[15] |
| | 293.15 | 0.7270 | 0.7272[16] | 0.3770 | 0.3608[8] |
| | 298.15 | 0.7237 | 0.72367[17] | 0.3560 | 0.356[18]<br>0.3563[19] |

cometer suspended in a thermostatically controlled water bath at required temperature controlled to ± 0.002 K. Care was taken to prevent evaporation during the measurements. The viscometer was calibrated using viscosity values taken from published values for methanol[12] and water.[13] An average of four or five sets of flow times was taken for each liquid and liquid mixture. The measured values of kinematic viscosities were converted to dynamic viscosities ($\eta$) after multiplication by the density. The flow-time measurements were made with an electronic stop watch having an uncertainty of ± 0.01 s. Viscosity values are accurate within the range ± 0.003 mPa·s.

Experimental physical properties of pure components, when available, were in accordance with those published in the literature, as shown in Table 1, with the exception of some viscosity values for triethylamine.[6,8] The factors that lead to the observed differences are unknown; however, they may simply reflect slight differences in the purity of triethylamine.

## Results and Discussion

The experimental densities values of TEA−W binary mixtures are shown in Figure 1. The experimental densities, $\rho$, and viscosities, $\eta$, at 283.15 K, 286.15 K, 288.15 K, 290.15 K, 291.15 K, and 291.35 K are listed in Table 2. Densities, $\rho$, and viscosities, $\eta$, have been fitted to the following polynomial equation of the form

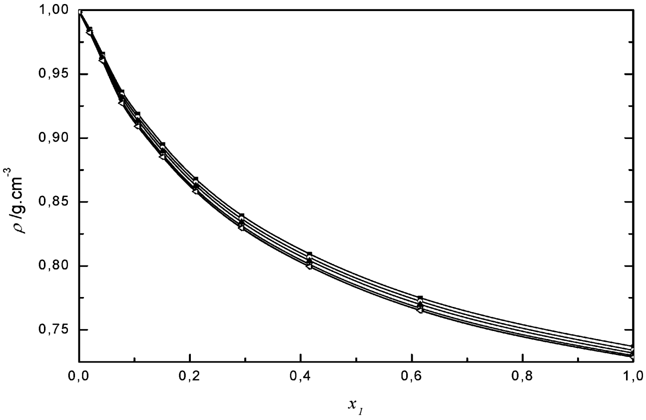

**Figure 1.** Density, $\rho$, variation against TEA mole fraction for triethylamine (1) + water (2) mixtures at: ■, 283.15 K; ○, 286.15 K; ▲, 288.15 K; ∇, 290.15 K; tilted solid triangle, 291.15 K; open solid triangle, 291.35 K. Polynomial correlations are plotted as solid lines.

**Table 2. Densities, $\rho$, and Viscosities, $\eta$, for the Mixtures Triethylamine (1) + Water (2) from $T$ = (283.15 to 291.35) K**

| $w_1$ | $x_1$ | $\rho/\mathrm{g\cdot cm^{-3}}$ | $\eta/\mathrm{mPa\cdot s}$ |
|---|---|---|---|
| | $T$ = 283.15 K | | |
| 0.0000 | 0.0000 | 0.9995 | 1.3054 |
| 0.0993 | 0.0192 | 0.9853 | 2.5131 |
| 0.2013 | 0.0429 | 0.9658 | 4.0642 |
| 0.3021 | 0.0775 | 0.9362 | 5.5315 |
| 0.4011 | 0.1060 | 0.9190 | 6.6209 |
| 0.5007 | 0.1510 | 0.8951 | 6.6426 |
| 0.6025 | 0.2117 | 0.8680 | 5.6286 |
| 0.6996 | 0.2930 | 0.8395 | 4.2307 |
| 0.8000 | 0.4167 | 0.8093 | 2.8128 |
| 0.8998 | 0.6150 | 0.7750 | 1.8354 |
| 1.0000 | 1.0000 | 0.7370 | 0.4229 |
| | $T$ = 286.15 K | | |
| 0.0000 | 0.0000 | 0.9993 | 1.2002 |
| 0.0993 | 0.0192 | 0.9843 | 2.2072 |
| 0.2013 | 0.0429 | 0.9642 | 3.5021 |
| 0.3021 | 0.0775 | 0.9340 | 4.7479 |
| 0.4011 | 0.1060 | 0.9163 | 5.4685 |
| 0.5007 | 0.1510 | 0.8924 | 5.4898 |
| 0.6025 | 0.2117 | 0.8653 | 4.7923 |
| 0.6996 | 0.2930 | 0.8368 | 3.6689 |
| 0.8000 | 0.4167 | 0.8066 | 2.5041 |
| 0.8998 | 0.6150 | 0.7723 | 1.5270 |
| 1.0000 | 1.0000 | 0.7340 | 0.4085 |
| | $T$ = 288.15 K | | |
| 0.0000 | 0.0000 | 0.9990 | 1.1372 |
| 0.0993 | 0.0192 | 0.9836 | 2.0274 |
| 0.2013 | 0.0429 | 0.9628 | 3.1903 |
| 0.3021 | 0.0775 | 0.9317 | 4.2304 |
| 0.4011 | 0.1060 | 0.9138 | 4.7414 |
| 0.5007 | 0.1510 | 0.8899 | 4.7628 |
| 0.6025 | 0.2117 | 0.8628 | 4.2959 |
| 0.6996 | 0.2930 | 0.8343 | 3.3571 |
| 0.8000 | 0.4167 | 0.8041 | 2.3269 |
| 0.8998 | 0.6150 | 0.7698 | 1.3497 |
| 1.0000 | 1.0000 | 0.7318 | 0.3989 |
| | $T$ = 290.15 K | | |
| 0.0000 | 0.0000 | 0.9987 | 1.0797 |
| 0.0993 | 0.0192 | 0.9828 | 1.8697 |
| 0.2013 | 0.0429 | 0.9614 | 2.9395 |
| 0.3021 | 0.0775 | 0.9290 | 3.7533 |
| 0.4011 | 0.1060 | 0.9109 | 4.0647 |
| 0.5007 | 0.1510 | 0.8870 | 4.0871 |
| 0.6025 | 0.2117 | 0.8599 | 3.8470 |
| 0.6996 | 0.2930 | 0.8314 | 3.1064 |
| 0.8000 | 0.4167 | 0.8012 | 2.1694 |
| 0.8998 | 0.6150 | 0.7669 | 1.1920 |
| 1.0000 | 1.0000 | 0.7298 | 0.3900 |
| | $T$ = 291.15 K | | |
| 0.0000 | 0.0000 | 0.9985 | 1.0524 |
| 0.0993 | 0.0192 | 0.9824 | 1.7985 |
| 0.2013 | 0.0429 | 0.9605 | 2.8380 |
| 0.3021 | 0.0775 | 0.9275 | 3.5209 |
| 0.4011 | 0.1060 | 0.9092 | 3.7470 |
| 0.5007 | 0.1510 | 0.8853 | 3.7683 |
| 0.6025 | 0.2117 | 0.8582 | 3.6430 |
| 0.6996 | 0.2930 | 0.8297 | 3.0050 |
| 0.8000 | 0.4167 | 0.7995 | 2.0985 |
| 0.8998 | 0.6150 | 0.7652 | 1.1210 |
| 1.0000 | 1.0000 | 0.7287 | 0.3855 |
| | $T$ = 291.35 K | | |
| 0.0000 | 0.0000 | 0.9985 | 1.0466 |
| 0.0993 | 0.0192 | 0.9823 | 1.7830 |
| 0.2013 | 0.0429 | 0.9604 | 2.8190 |
| 0.3021 | 0.0775 | 0.9271 | 3.4703 |
| 0.4011 | 0.1060 | 0.9090 | 3.6788 |
| 0.5007 | 0.1510 | 0.8851 | 3.6999 |
| 0.6025 | 0.2117 | 0.8580 | 3.6025 |
| 0.6996 | 0.2930 | 0.8295 | 2.9851 |
| 0.8000 | 0.4167 | 0.7993 | 2.0846 |
| 0.8998 | 0.6150 | 0.7650 | 1.1071 |
| 1.0000 | 1.0000 | 0.7285 | 0.3847 |

$$Y = \sum_{i=0}^{n} A_i x_1^i \tag{1}$$

where $Y$ stands for each of the above properties and $x_i$ is mole fraction of pure component $i$. The standard deviations $\sigma$

$$\sigma = [\sum_{i=1}^{N} (Y_{i,\text{calcd}} - Y_{i,\text{exptl}})^2/(N - j)]^{1/2} \tag{2}$$

where $N$ is the number of experimental data points and $j$ is the number of parameters.

The coefficients, $A_i$, and the standard deviations, $\sigma$, are shown in Table 3.

It is found that the density decreases with increasing TEA concentration at constant temperature and decreases with a thermal increase in temperature at fixed TEA mole fractions. Figure 2 shows the plots of viscosities of TEA−W mixtures against TEA mole fraction at different temperatures. As can be appreciated, the addition of TEA to water rapidly increases the viscosity, which passes through a maximum and then declines. The decline, however, continues at a slower rate up to the pure state of TEA. The effect of temperature on viscosity is seen to be quite significant, particularly in the region at or around the maximum viscosity. The position of the maximum apparently remains unchanged with the variation of temperature. We observe that the maximum occurred at around 0.1271 TEA mole fraction (45 % TEA mass fraction). This value is in good agreement with literature values. In fact, Kartzmark[15] found that the maxima occurred at 0.1128 mol fraction (42 % TEA mass fraction), whereas Dudley et al.[6] found that the maxima occurred at 0.151 mol fraction (50 % TEA mass fraction).

However, concerning the vicinity of critical temperature, it can be clearly seen from close observation of Figures 1and 2 that $\rho$ and $\eta$ have not been affected by the correlation of the concentration fluctuations. Indeed, we did not detect any anomaly near the critical composition $x_c \approx 0.078$ mol fraction of triethylamine.

Several semiempirical relations have been proposed to estimate the dynamic viscosity, $\eta$, of liquid mixtures in terms of pure-component data.[20−22] We have examined equations proposed by Grunberg and Nissan;[23] Hind, McLaughlin, and Ubbelohde;[24] Katti and Chaudhri;[25] Heric and Brewer;[26] McAllister;[27] and Auslander.[28] In general, Irving[20] found that using mole fraction instead of volume or mass does not significantly improve the accuracy of the calculated mixture viscosity. Nevertheless, the opposite is true, and mass or volume fractions generally give better results.[29] This was true for most mixing rules and studied systems. For this reason, all of the following mixing rules used for the determination of the interaction parameters are expressed in mass fraction, $w_i$, of component $i$

**Table 3. Parameters, $A_i$, from Equation *1* and Standard Deviations, $\sigma(Y)$, for Density, $\rho$/g·cm$^{-3}$, and Viscosity, $\eta$/mPa·s, for the Binary Mixture Triethylamine (1) + Water (2) from $T$ = (283.15 to 291.35) K**

| | $T$/K | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $\sigma(Y)$ |
|---|---|---|---|---|---|---|---|
| $\rho$ | 283.15 | 1.001 | −0.936 | 1.804 | −1.907 | 0.774 | 0.0013 |
| | 286.15 | 1.000 | −0.966 | 1.925 | −2.086 | 0.860 | 0.0018 |
| | 288.15 | 1.000 | −0.995 | 2.043 | −2.264 | 0.948 | 0.0025 |
| | 290.15 | 0.999 | −1.030 | 2.183 | −2.476 | 1.053 | 0.0018 |
| | 291.15 | 0.999 | −1.051 | 2.264 | −2.598 | 1.113 | 0.0016 |
| | 291.35 | 0.999 | −1.054 | 2.279 | −2.622 | 1.125 | 0.0023 |
| $\eta$ | 283.15 | 9.319 | −8.804 | 16.717 | −44.868 | 50.661 | 0.0041 |
| | 286.15 | 8.423 | −9.562 | 7.849 | −31.973 | 54.134 | 0.0034 |
| | 288.15 | 7.949 | −10.253 | 2.269 | −23.075 | 56.791 | 0.0056 |
| | 290.15 | 7.585 | −11.077 | −3.434 | −14.821 | 59.814 | 0.0047 |
| | 291.15 | 7.434 | −11.569 | −6.069 | −10.389 | 61.648 | 0.0033 |
| | 291.35 | 7.419 | −11.640 | −6.844 | −10.206 | 61.459 | 0.0038 |

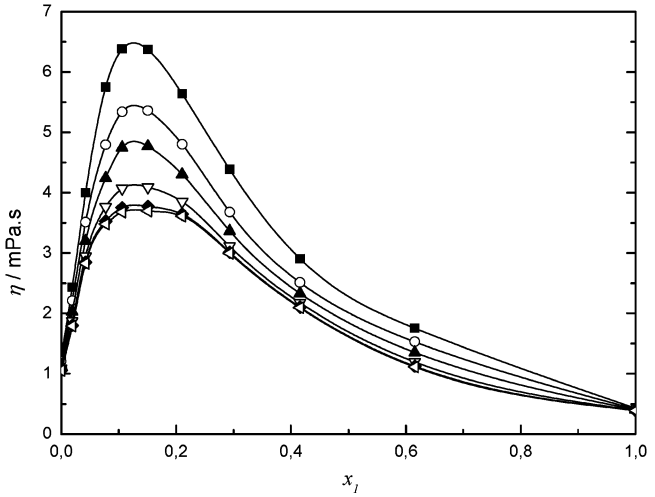

**Figure 2.** Viscosity, $\eta$, variation versus TEA mole fraction for triethylamine (1) + water (2) mixtures at: ■, 283.15 K; ○, 286.15 K; ▲, 288.15 K; ∇, 290.15 K; ◆, 291.15 K; open tilted triangle, 291.35 K. Polynomial correlations are plotted as solid lines.

Grunberg and Nissan[23]

$$\ln \eta = w_1 \ln \eta_1 + w_2 \ln \eta_2 + w_1 w_2 G_{12} \tag{3}$$

where $G_{12}$ is a parameter proportional to the interchange energy.

Hind et al.[24]

$$\eta = w_1^2 \eta_1 + w_2^2 \eta_2 + 2 w_1 w_2 \eta_{12} \tag{4}$$

where $\eta_{12}$ is attributed to unlike pair interactions.

Katti and Chaudhri[25]

$$\ln(\eta V) = w_1 \ln(\eta_1 V_1) + w_2 \ln(\eta_2 V_2) + w_1 w_2 W_{\text{vis}}/RT \tag{5}$$

where $W_{\text{vis}}$ is an interaction term and $V_i$ is the molar volume of pure component $i$.

Heric and Brewer[26]

$$\ln(\eta V) = w_1 \ln(\eta_1 V_1) + w_2 \ln(\eta_2 V_2) + w_1 w_2(\alpha_1 + \alpha_2(w_1 - w_2)) \tag{6}$$

$\alpha_1$ and $\alpha_2$ are adjustable parameters.

McAllister[27]

$$\begin{aligned} \ln \nu = {} & w_1^3 \ln \nu_1 + 3 w_1^2 w_2 \ln Z_{12} + 3 w_1 w_2^2 \ln Z_{21} + \\ & w_2^3 \ln \nu_2 - \ln(w_1 + w_2 M_2/M_1) + 3 w_1^2 w_2 \ln(2/3 + \\ & M_2/3M_1) + 3 w_1 w_2^2 \ln(1/3 + 2M_2/3M_1) + w_2^2 \ln(M_2/M_1) \end{aligned} \tag{7}$$

where $Z_{12}$ and $Z_{21}$ are interaction parameters and $M_i$ and $\nu_i$ are the molecular mass and kinematic viscosity of pure component $i$, respectively.

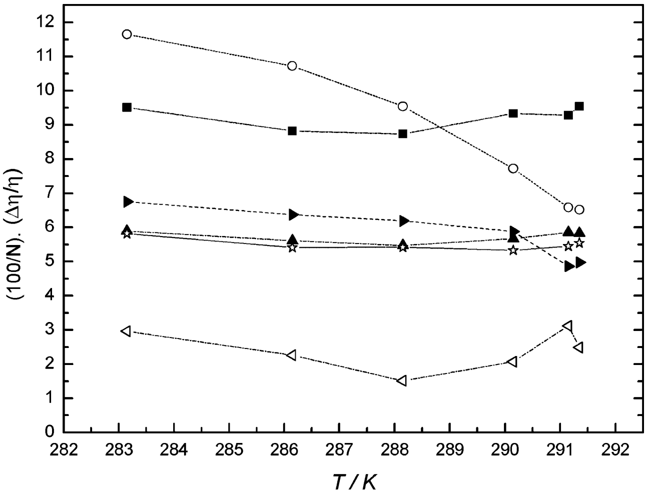

**Figure 3.** Standard percentage deviations, $(100/N)\cdot\Delta\eta/\eta$, where $\Delta\eta/\eta = |\eta_{\mathrm{exptl}} - \eta_{\mathrm{calcd}}|/\eta_{\mathrm{exptl}}$ represents the fractional deviation from the experimental viscosities ($\eta_{\mathrm{exptl}}$) of the calculated values ($\eta_{\mathrm{calcd}}$) obtained as a function of temperature, $T$, with the correlation by: ■, Grunberg−Nissan; ○, Hind et al.; ▲, Katti−Chaudhri; ☆, Heric−Brewer; solid tilted triangle, McAllister; open tilted triangle, Auslander. $N$ is the number of experimental viscosity data points in each temperature set.

Auslander[28]

$$\eta = [w_1\eta_1(w_1+B_{12}w_2) + w_2\eta_2A_{21}(w_2 + B_{21}w_1)]/[w_1(w_1 + B_{12}w_2) + w_2A_{21}(w_2 + B_{21}w_1)] \quad (8)$$

where $B_{12}$, $A_{21}$, and $B_{21}$ are the parameters representing binary 1−2 interactions.

We tested the correlating ability of eqs 3, 4, 5, 6, 7, and 8 by calculating the standard percentage deviation (PD) between the experimental and the calculated viscosity to be

$$\mathrm{PD} = \frac{100}{N}\sum_{1}^{N}\frac{|\eta_{\mathrm{calcd}} - \eta_{\mathrm{exptl}}|}{\eta_{\mathrm{exptl}}} \quad (9)$$

where $N$ is the number of experimental viscosity data points in each set.

In general, the models tested showed a good ability to fit the experimental data. As depicted in Figure 3, the Grunberg−Nissan and Hind et al. models provided the worst results; however, the deviations were only less than 12 %, and the parameters deduced can be used to predict viscosity data of multicomponent systems. The other models exhibit very good correlation ability with deviations better than 6 %; in particular, the excellent ability of the Auslander model with an average error of only 2.4 % must be focused on.

## Conclusions

Densities and viscosities of triethylamine + water mixture are reported at 283.15 K, 286.15 K, 288.15 K, 290.15 K, 291.15 K, and 291.35 K as a function of triethylamine composition under atmospheric pressure. Results have been fitted to obtain the adjustable parameters and standard deviations between the measured and fitted values. Moreover, the viscosity data have been correlated with some semiempirical equations of one, two, or three parameters.

**Table 4. Parameters Values Determined from Equations *3*, *4*, *5*, *6*, *7*, and *8* and Percentage Deviations (PD) in Correlating Viscosities for the Binary Mixture Triethylamine (1) + Water (2) from $T$ = (283.15 to 291.35) K**

Single-Parameter Equations

| | Grunberg and Nissan | | Hind et al. | | Katti and Chaudhri | |
|---|---|---|---|---|---|---|
| $T$/K | $G_{12}$ | PD | $\eta_{12}$ | PD | $W_{\mathrm{vis}}/RT$ | PD |
| 283.15 | 9.01 | 9.51 | 11.01 | 11.65 | 11.82 | 5.89 |
| 286.15 | 8.56 | 8.82 | 9.25 | 10.72 | 11.33 | 5.61 |
| 288.15 | 8.20 | 8.73 | 8.11 | 9.54 | 11.03 | 5.47 |
| 290.15 | 7.83 | 9.33 | 7.13 | 7.72 | 10.58 | 5.67 |
| 291.15 | 7.64 | 9.28 | 6.75 | 6.58 | 10.39 | 5.85 |
| 291.35 | 7.60 | 9.54 | 6.60 | 6.51 | 10.37 | 5.83 |
| average PD | | 9.20 | | 8.78 | | 5.72 |

Two-Parameter Equations

| | Heric and Brewer | | | McAllister | | |
|---|---|---|---|---|---|---|
| $T$/K | $\alpha_1$ | $\alpha_2$ | PD | $Z_{12}$ | $Z_{21}$ | PD |
| 283.15 | 11.85 | 1.83 | 5.82 | −11.08 | 16.35 | 6.75 |
| 286.15 | 11.30 | 1.72 | 5.41 | −10.41 | 15.32 | 6.37 |
| 288.15 | 10.91 | 1.65 | 5.42 | −10.11 | 14.90 | 6.19 |
| 290.15 | 10.62 | 1.67 | 5.33 | −9.89 | 14.67 | 5.88 |
| 291.15 | 10.37 | 1.64 | 5.45 | −9.83 | 14.64 | 4.86 |
| 291.35 | 10.33 | 1.69 | 5.54 | −9.84 | 14.72 | 4.97 |
| average PD | | | 5.49 | | | 5.84 |

Three-Parameter Equation

| | Auslander | | | |
|---|---|---|---|---|
| $T$/K | $B_{12}$ | $B_{21}$ | $A_{21}$ | PD |
| 283.15 | −9.18 | 10.00 | 0.89 | 2.96 |
| 286.15 | −9.33 | 10.15 | 0.91 | 2.26 |
| 288.15 | −9.76 | 10.60 | 0.93 | 1.51 |
| 290.15 | −10.45 | 11.42 | 0.95 | 2.07 |
| 291.15 | −10.95 | 12.02 | 0.96 | 3.12 |
| 291.35 | −11.10 | 12.19 | 0.96 | 2.49 |
| average PD | | | | 2.40 |

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