*J. Chem. Eng. Data* **2002,** *47,* 927−931 **927**

# Measurement of the Viscosity of Binary and Ternary Systems Containing Benzaldehyde, Toluene, and Benzyl Alcohol at 293.15 K and 0.1 MPa

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In this work we measure the density and viscosity of the ternary system benzyl alcohol (1) + benzaldehyde (2) + toluene (3) at 293.15 K and 0.1 MPa for 21 different ternary composition sets. Values of the viscosity and density of the respective binary systems, over the whole composition range at the same temperature and pressure conditions, are also presented. Experimental data of binary systems were used to determine the excess molar volumes and the viscosity deviations which were fitted by means of Redlich−Kister polynomials. The experimental data of the dynamic viscosity of the binary systems have been used to obtain the binary interaction parameters of a model recently proposed in the literature by our group. The overall mean relative standard deviation between experimental and calculated viscosities for the ternary system was 1.1%.

## Introduction

In this study we have measured the viscosity of ternary systems containing monosubstituted aromatics in order to evaluate the effect of the substituent in the solvent−solvent interactions of aromatic mixtures. We present the experimental results of the viscosity of the ternary system benzyl alcohol + benzaldehyde + toluene and the respective binaries. To our knowledge there are no viscosity data available in the literature for the binary and ternary systems measured in this work. The solvents selected are miscible over the whole composition range and have large differences in viscosity.

The experimental data have been used to evaluate whether a previously developed viscosity model by our group[1,2] would be adequate for systems constituted by monosubstituted aromatic hydrocarbons. This model is based on Eyring's theory for viscous flow[3] and the UNIQUAC equation.[4,5]

Using Eyring's approach for liquid viscosity and classical thermodynamics, it is possible to obtain an expression that considers the viscosity of real systems as a combination of a reference term contribution and a deviation one, as follows:[1,2]

$$\ln(\eta V) = \ln(\eta V)_{\mathrm{ref}} + \frac{G_{\mathrm{E}}^{\ddagger}}{RT} \tag{1}$$

where $\eta$ is the solvent mixture dynamic viscosity, $V$ is the solvent mixture molar volume, $G_{\mathrm{E}}^{\ddagger}$ is the molar excess Gibbs free energy of activation for the flow, $R$ is the gas constant, and $T$ is the absolute temperature.

Kendall and Monroe's[6] equation has been adopted to describe the reference term, and the UNIQUAC model[4,5] has been chosen to describe the deviation one. Thus, the final expression for nonelectrolyte liquid mixture viscosity can be written as follows:[1,2]

$$\ln(\eta V) = \sum_{i=1}^{N_{\mathrm{SOL}}} x_i \ln(\eta_i) + \ln\Big(\sum_{i=1}^{N_{\mathrm{SOL}}} x_{\mathrm{i}} V_{\mathrm{i}}^{\circ}\Big) + \sum_{i=1}^{N_{\mathrm{SOL}}} x_{\mathrm{i}} \ln\left(\frac{\phi_i}{\theta_i}\right) + \frac{z}{2}\sum_{i=1}^{N_{\mathrm{SOL}}} q_i x_i \ln\left(\frac{\theta_i}{\phi_i}\right) - \sum_{i=1}^{N_{\mathrm{SOL}}} x_i q_i \ln\left[\sum_{k=1}^{N_{\mathrm{SOL}}} \theta_i \exp\left(-\frac{\alpha_{ki}}{T}\right)\right] \tag{2}$$

where $x_i$ is the mole fraction of component $i$, $V_i^{\circ}$ is the molar volume of the pure liquid $i$ at the same temperature and pressure of the mixture, $\phi_i$ is the volume fraction, $\theta_i$ is the surface area fraction, $q_i$ is the surface area parameter of the UNIQUAC model, $N_{\mathrm{SOL}}$ is the number of solvents in the mixture, $z$ is the coordination number, and $\alpha_{ki}$ is the UNIQUAC binary interaction parameter obtained by means of fitting experimental viscosity data of the different binary liquid mixtures.

## Experimental Section

Benzyl alcohol PA (Merck, purity > 99.5%), toluene PA (Merck, purity > 99.5%), and benzaldehyde for synthesis (Merck, purity > 99%) were used without further purification. The purity of the solvents was confirmed by gas chromatography and by comparing their densities and viscosities with the corresponding literature values at 293.15 K or 298.15 K and 0.1 MPa (Table 1). The solutions were prepared by mass using a digital balance (Chyo, model JK-180), with a precision of ±0.1 mg, in airtight stopped bottles. To avoid evaporation and solvent contamination, the solutions were used immediately after preparation. The estimated error in mole fraction is less than $1 \times 10^{-4}$.

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10.1021/je010327v CCC: $22.00

**Table 1. Comparison of Experimental Results of Densities and Viscosities of Pure Solvents with Literature Values at 0.1 MPa**

| | | $\rho$/g·cm$^{-3}$ | | | $\eta$/mPa·s | |
|---|---|---|---|---|---|---|
| | $T$/K | this work | lit. value | $T$/K | this work | lit. value |
| benzyl alcohol | 298.15 | 1.041 26 | 1.0414[a] | 298.15 | 5.553 | 5.737[a]<br>5.054[b] |
| benzaldehyde | 293.15 | 1.044 67 | 1.04463[c]<br>1.04437[d] | 298.15 | 1.390 | 1.40[e] |
| toluene | 298.15 | 0.862 22 | 0.8625[f] | 298.15 | 0.556 | 0.5549[f] |

[a] Experimental data from ref 12. [b] Experimental data from ref 13. [c] Experimental data from ref 14. [d] Experimental data from ref 15. [e] Experimental data from ref 16. [f] Experimental data from ref 17.

**Table 2. Experimental Densities and Experimental and Calculated Viscosities of Binary Mixtures at 293.15 K and 0.1 MPa**

| $x_1$ | $\rho$/g·cm$^{-3}$ | $\eta^{\text{exp}}$/mPa·s | $\eta^{\text{calc}}$/mPa·s | $\delta\eta$[a] |
|---|---|---|---|---|
| | Benzyl Alcohol (1) + Benzaldehyde (2) | | | |
| 0 | 1.044 67 | 1.495 | | |
| 0.1032 | 1.046 36 | 1.695 | 1.712 | 1.019 |
| 0.2000 | 1.047 00 | 1.882 | 1.872 | 0.570 |
| 0.3007 | 1.047 32 | 2.114 | 2.073 | 1.980 |
| 0.4023 | 1.047 26 | 2.322 | 2.339 | 0.738 |
| 0.4989 | 1.047 04 | 2.633 | 2.668 | 1.343 |
| 0.6013 | 1.046 72 | 3.114 | 3.117 | 0.098 |
| 0.6880 | 1.046 60 | 3.591 | 3.597 | 0.171 |
| 0.8000 | 1.046 06 | 4.366 | 4.382 | 0.369 |
| 0.8984 | 1.045 69 | 5.395 | 5.267 | 2.377 |
| 1 | 1.045 16 | 6.421 | | |
| | Benzyl alcohol (1) + Toluene (3) | | | |
| 0 | 0.866 82 | 0.591 | | |
| 0.1006 | 0.884 82 | 0.699 | 0.701 | 0.322 |
| 0.1997 | 0.902 62 | 0.851 | 0.850 | 0.172 |
| 0.3007 | 0.920 80 | 1.051 | 1.054 | 0.296 |
| 0.4020 | 0.939 10 | 1.329 | 1.330 | 0.038 |
| 0.5020 | 0.957 05 | 1.717 | 1.694 | 1.348 |
| 0.6000 | 0.974 67 | 2.158 | 2.170 | 0.578 |
| 0.6925 | 0.991 13 | 2.764 | 2.763 | 0.019 |
| 0.7994 | 1.010 11 | 3.659 | 3.681 | 0.616 |
| 0.8995 | 1.027 68 | 4.821 | 4.847 | 0.530 |
| 1 | 1.045 16 | 6.421 | | |
| | Benzaldehyde (2) + Toluene (3) | | | |
| 0 | 0.866 82 | 0.591 | | |
| 0.1053 | 0.885 84 | 0.652 | 0.647 | 0.862 |
| 0.2063 | 0.904 17 | 0.721 | 0.707 | 1.998 |
| 0.2998 | 0.921 00 | 0.768 | 0.768 | 0.023 |
| 0.4022 | 0.939 46 | 0.844 | 0.842 | 0.145 |
| 0.4995 | 0.957 04 | 0.920 | 0.921 | 0.073 |
| 0.6003 | 0.975 20 | 1.017 | 1.012 | 0.480 |
| 0.6939 | 0.991 72 | 1.108 | 1.106 | 0.178 |
| 0.8082 | 1.011 18 | 1.256 | 1.235 | 1.636 |
| 0.9002 | 1.027 24 | 1.367 | 1.352 | 1.092 |
| 1 | 1.044 67 | 1.495 | | |

[a] $\delta\eta = 100|(\eta_{\text{exp}} - \eta_{\text{calc}})/\eta_{\text{exp}}|$.

**Table 3. Experimental Densities and Experimental and Calculated Viscosities of Ternary (Benzyl Alcohol (1) + Benzaldehyde (2) + Toluene (3)) Mixtures at 293.15 K and 0.1 MPa**

| $x_1$ | $x_2$ | $\rho$/g·cm$^{-3}$ | $\eta^{\text{exp}}$/mPa·s | $\eta^{\text{calc}}$/mPa·s | $\delta\eta$[a] |
|---|---|---|---|---|---|
| 0.7446 | 0.1274 | 1.023 84 | 3.547 | 3.545 | 0.045 |
| 0.6225 | 0.2528 | 1.024 60 | 2.890 | 2.877 | 0.440 |
| 0.6246 | 0.1268 | 1.002 36 | 2.607 | 2.584 | 0.854 |
| 0.4982 | 0.2532 | 1.002 85 | 2.128 | 2.107 | 0.970 |
| 0.4996 | 0.1276 | 0.980 24 | 1.893 | 1.886 | 0.282 |
| 0.3760 | 0.2591 | 0.982 08 | 1.610 | 1.584 | 1.613 |
| 0.3692 | 0.1283 | 0.957 03 | 1.392 | 1.382 | 0.699 |
| 0.2525 | 0.6192 | 1.025 54 | 1.745 | 1.720 | 1.430 |
| 0.2508 | 0.4992 | 1.003 01 | 1.541 | 1.515 | 1.732 |
| 0.2436 | 0.3791 | 0.979 38 | 1.322 | 1.324 | 0.111 |
| 0.2510 | 0.2510 | 0.957 72 | 1.191 | 1.190 | 0.102 |
| 0.2473 | 0.1291 | 0.936 13 | 1.082 | 1.056 | 2.435 |
| 0.1288 | 0.4992 | 0.980 38 | 1.172 | 1.180 | 0.666 |
| 0.1293 | 0.3741 | 0.958 52 | 1.042 | 1.045 | 0.321 |
| 0.1276 | 0.2502 | 0.935 51 | 0.923 | 0.926 | 0.360 |
| 0.1253 | 0.1264 | 0.912 74 | 0.817 | 0.823 | 0.688 |
| 0.1252 | 0.7349 | 1.022 50 | 1.464 | 1.498 | 2.360 |
| 0.1270 | 0.6224 | 1.002 84 | 1.316 | 1.335 | 1.425 |
| 0.3769 | 0.4962 | 1.025 79 | 2.043 | 1.995 | 2.328 |
| 0.3647 | 0.3837 | 1.002 03 | 1.772 | 1.736 | 2.061 |
| 0.4996 | 0.3742 | 1.025 20 | 2.414 | 2.368 | 1.931 |

[a] $\delta\eta = 100|(\eta_{\text{exp}} - \eta_{\text{calc}})/\eta_{\text{exp}}|$.

The densities of pure solvents and solutions were measured with a DMA 58 Anton Paar digital density meter, with an uncertainty of 1 × $10^{-5}$ g/cm$^3$. The cell temperature of the density meter was controlled to within ±0.01 K. For all pure solvents and solutions, the density was measured at least twice and the results were averaged. The estimated uncertainty of the measurements was 1 × $10^{-5}$ g/cm$^3$. Tridistilled water and dry air were used as calibrating substances.

Viscosities were measured using Cannon-Fenske viscometers (Schott-Geräte, internal diameters of 0.44 mm, 0.54 mm, 0.63 mm, and 0.77 mm) immersed in a thermostated bath (Schott-Geräte, model CT 1450/2), controlled to within ±0.01 K, and coupled to an automatic module AVS-350 (Schott-Geräte). The uncertainty in terms of efflux time was 0.01 s. At least 10 repetitions for each solution composition were performed, and the results were averaged.

The efflux time was converted to dynamic viscosity by means of the following equation

$$\eta = k(t - \omega)\rho \tag{3}$$

where $k$ is the viscometer constant, $t$ is the efflux time, $\omega$ is the Hagenbach correction, and $\rho$ is the density. The uncertainty in dynamic viscosity was less than 2 × $10^{-3}$ mPa·s.

## Results and Discussion

Table 2 presents the densities and dynamic viscosities calculated (using eq 3) from the experimental data obtained at 293.15 K and 0.1 MPa for the three binary mixtures studied in this work as well as the calculated values obtained by means of eq 2 and the percentage relative deviation. Table 3 presents dynamic viscosities and densities of the ternary system benzyl alcohol (1) + benzaldehyde (2) + toluene (3) under the same temperature and pressure conditions for 21 different composition sets, as well as the values calculated by means of eq 2.

The experimental viscosity data of the binary systems have been correlated through eq 2 in order to determine the necessary binary interaction parameter for the used model. Table 4 shows the model interaction parameters for each binary system along with the mean relative standard deviation and maximum deviation obtained. The mean relative standard deviation is given by

$$\text{MRSD} = \left[\frac{1}{\text{ND}}\sum_{i=1}^{\text{ND}}\left(\frac{\eta_{\text{calc}} - \eta_{\text{exp}}}{\eta_{\text{exp}}}\right)^2\right]^{1/2} \tag{4}$$

where ND is the number of data correlated. The good correlation agreement between experimental and calculated values for the viscosity of the binary systems can be observed in Figure 1 and Table 4.

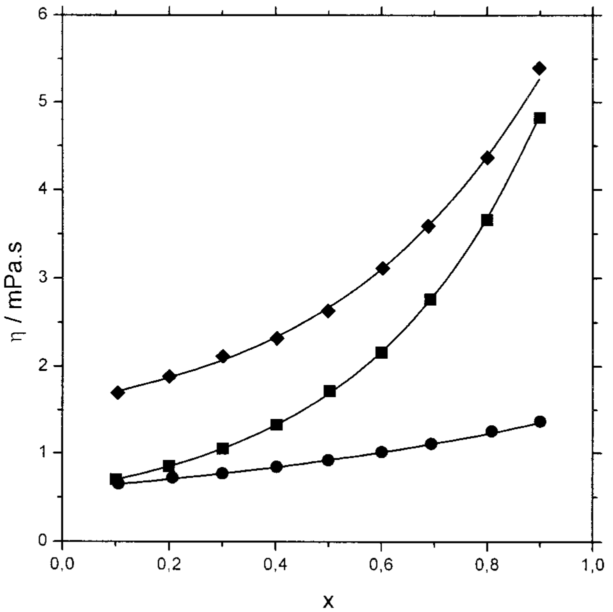

**Figure 1.** Dynamic viscosity, $\eta$, at 293.15 K and 0.1 MPa, as a function of the mole fraction of the first component of each binary system. Experimental results: ◆, benzyl alcohol (1) + benzaldehyde (2); ■, benzyl alcohol (1) + toluene (3); ●, benzaldehyde (2) + toluene (3). —, Calculated values from eq 2.

**Table 4. Interaction Energy Parameters**

| system | $\alpha_{21}$/K | $\alpha_{12}$/K | MRSD/% | max. dev/% |
|---|---|---|---|---|
| benzyl alcohol + benzaldehyde | −308.20 | 588.14 | 1.2 | 2.4 |
| benzyl alcohol + toluene | 275.03 | −226.21 | 0.6 | 1.3 |
| benzaldehyde + toluene | −88.01 | 92.81 | 1.0 | 2.0 |

The previously fitted parameters were used to predict the dynamic viscosity of the ternary system benzyl alcohol (1) + benzaldehyde (2) + toluene (3) at 293.15 K and 0.1 MPa, by means of the proposed model. The agreement between experimental and calculated viscosity values is rather good. The overall MRSD obtained for the ternary system was 1.1% with a maximum deviation of 2.4%. It seems reasonable to conclude that the proposed model is able to predict the viscosity behavior of solutions containing benzyl alcohol, benzaldehyde, and toluene at 293.15 K, at 0.1 MPa, and at different compositions. It has been found that no significant improvement in the performance of the model could be achieved by including experimental ternary viscosity data for fitting the model parameters.

The excess molar volumes ($V^E$) of the three binary solvent mixtures have been calculated by means of the following equation:

$$V^E = \sum_i \frac{x_i M_i}{\rho} - \frac{x_i M_i}{\rho_i^\circ} \qquad (5)$$

where $V^E$ is the excess molar volume, $M_i$ is the molar mass of component $i$, and $\rho_i^\circ$ is the density of the pure component $i$ at the temperature and pressure of the solvent mixture.

The viscosity deviations have been determined according to the equation

$$\Delta\eta = \eta - \sum_i \eta_i x_i \qquad (6)$$

where $\Delta\eta$ is the dynamic viscosity deviation and $\eta_i$ is the dynamic viscosity of pure solvent $i$ at the same temperature and pressure of the solvent mixture.

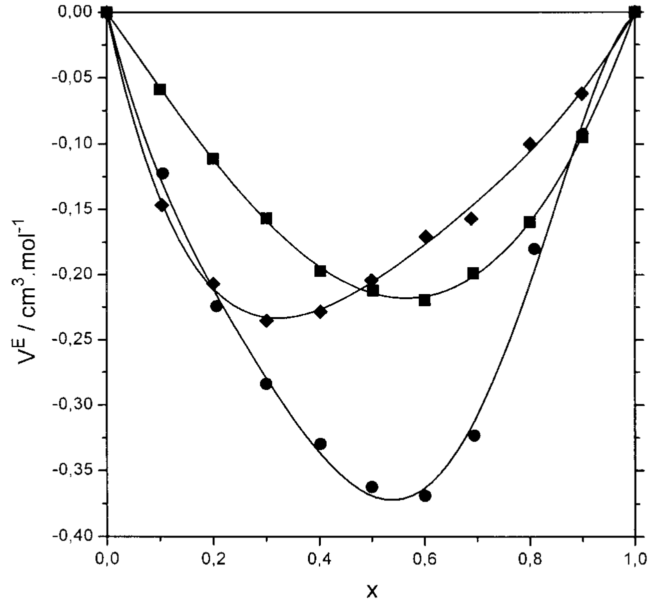

**Figure 2.** Excess molar volumes, $V^E$, at 293.15 K and 0.1 MPa, as a function of the mole fraction of the first component of each binary system: ◆, benzyl alcohol (1) + benzaldehyde (2); ■, benzyl alcohol (1) + toluene (3); ●, benzaldehyde (2) + toluene (3); —, calculated values from eq 7.

**Table 5. Redlich-Kister Coefficients and Standard Deviations ($\sigma$) for Binary Mixtures at 293.15 K and 0.1 MPa**

| | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $\sigma$ |
|---|---|---|---|---|---|
| Benzyl Alcohol (1)−Benzaldehyde (2) | | | | | |
| $V^E$ (cm$^3$·mol$^{-1}$) | −0.821 | −0.519 | −0.458 | −0.081 | 0.005 |
| $\Delta\eta$ (mPa·s) | −5.233 | 2.430 | 0.706 | −0.943 | 0.050 |
| Benzyl Alcohol (1)−Toluene (3) | | | | | |
| $V^E$ (cm$^3$·mol$^{-1}$) | −0.855 | −0.239 | 0.030 | −0.020 | 0.002 |
| $\Delta\eta$ (mPa·s) | −7.248 | −3.311 | −1.552 | −0.642 | 0.011 |
| Benzaldehyde (2)−Toluene (3) | | | | | |
| $V^E$ (cm$^3$·mol$^{-1}$) | −1.477 | −0.311 | −0.480 | 0.962 | 0.012 |
| $\Delta\eta$ (mPa·s) | −0.489 | −0.096 | 0.203 | 0.074 | 0.005 |

The deviation properties for each binary system were fitted by means of the Redlich−Kister equation:[7]

$$Y = x_1 x_2 \sum_j A_j (x_1 - x_2)^j \quad \text{with} \quad j = 0, ..., 3 \qquad (7)$$

where $Y$ is the property to be fitted and $j$ is the polynomial degree.

The coefficients were obtained by least-squares fitting of the property according to eq 7. Table 5 presents the values of the Redlich−Kister coefficients obtained for excess volume and viscosity deviation along with their standard deviations, defined by

$$\sigma = \left[\frac{\sum (Y_{\text{exp}} - Y_{\text{RK}})^2}{(n - m)}\right]^{1/2} \qquad (8)$$

where $\sigma$ is the standard deviation, $Y_{\text{exp}}$ is the property derived from experimental data, $Y_{\text{RK}}$ is the property calculated from the fitted Redlich−Kister equation, $n$ is the number of data points, and $m$ is the number of coefficients in the Redlich−Kister polynomial.

Figure 2 shows the behavior of the excess molar volumes for the three binary systems. All the curves show a negative

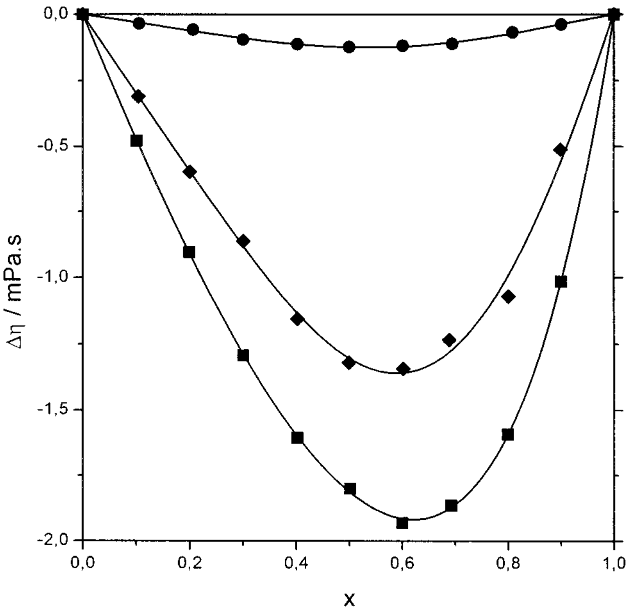

**Figure 3.** Viscosity deviations, $\Delta\eta$, at 293.15 K and 0.1 MPa, as a function of the mole fraction of the first component of each binary system: ◆, benzyl alcohol (1) + benzaldehyde (2); ■, benzyl alcohol (1) + toluene (3); ●, benzaldehyde (2) + toluene (3); —, calculated values from eq 7.

deviation over the whole composition range. In the benzyl alcohol + benzaldehyde system, this minimum is close to the benzaldehyde rich region. In the benzaldehyde + toluene system, the minimum value is near the equimolar composition, while in the benzyl alcohol + toluene system this minimum is slightly shifted toward the more toluene rich composition.

It is often stated in the literature[8−11] that the excess molar volume values of binary liquid mixtures are dependent on the chemical, physical, and structural factors of the liquids. Owing to the similarity in shape and size of the compounds studied in this work, we might suppose that the most important contribution to the $V^E$ would be due to the chemical effects.

The chemical effects in the excess molar volume take into account a compromise between the positive contribution, due to net destruction of order during the mixing (e.g. the disruption of H-bonds), and the negative contribution, due to net creation of order during the mixing (e.g. the decrease in free volume by new H-bonds formed between the component molecules in solution).[10,11] Thus, the experimental negative $V^E$ values suggest that there is a reduction in the free volume as a result of the formation of new H-bonds between molecules in solution.

Figure 3 shows the behavior of $\Delta\eta$ for the three binary systems studied. As in the $V^E$ diagram, the viscosity deviation curves present a minimum. In the case of the benzaldehyde + toluene system this minimum is found near equimolar composition, while for benzyl alcohol + toluene and benzyl alcohol + benzaldehyde systems it is slightly shifted in the direction of pure benzyl alcohol.

## Conclusions

The viscosities and densities of the binary systems benzyl alcohol + benzaldehyde, benzyl alcohol + toluene, and benzaldehyde + toluene and of the ternary system were experimentally measured at 293.15 K and 0.1 MPa. Using the binary density and viscosity data, it was possible to obtain the binary interaction parameters of the model used, eq 2. The model showed excellent agreement with the experimental data. The mean relative standard deviation was 1.2% for the system benzaldehyde + benzyl alcohol, 1.0% for the system benzaldehyde + toluene, and 0.6% for the system benzyl alcohol + toluene. In the case of the ternary system the MRSD obtained was 1.1%.

The excess properties of the binary systems were determined and subsequently fitted by means of a Redlich−Kister polynomial. The derived excess molar volumes and viscosity deviation curves showed a minimum for all binary systems studied. The experimental negative $V^E$ values might suggest the formation of new H-bonds between solvent molecules in solution.

## Acknowledgment

We would like to acknowledge one of the anonymous reviewers for the references about the chemical effects on the excess volume.

## Nomenclature

$A_j$ = Redlich−Kister coefficients
$G_E^{\ddagger}$ = molar Gibbs free energy of activation for the flow process
$j$ = degree of the Redlich−Kister polynomial
$k$ = viscometer constant
$m$ = number of coefficients in the Redlich−Kister polynomial
$M_i$ = molar weight of pure component $i$
MRSD = mean relative standard deviation
$n$ = number of data points
ND = number of data correlated
$N_{SOL}$ = number of solvents in the mixture
$q_i$ = surface area parameter of component $i$
$R$ = gas constant (8.314 510 J·mol$^{-1}$·K$^{-1}$)
$T$ = absolute temperature
$t$ = efflux time
$V$ = molar volume of the liquid mixture
$V_i^{\circ}$ = molar volume of the pure liquid $i$
$V^E$ = excess molar volume
$Y$ = property to be ajusted by a Redlich−Kister polynomial
$x_i$ = mole fraction of component $i$
$z$ = coordination number

*Greek Letters*

$\alpha_{ki}$ = UNIQUAC binary interaction parameter between species $k$ and $i$
$\Delta\eta$ = viscosity deviation
$\eta$ = dynamic viscosity
$\eta_i$ = dynamic viscosity of pure solvent $i$
$\theta_i$ = surface area fraction
$\rho$ = density
$\sigma$ = standard deviation in Redlich−Kister coefficients
$\phi_i$ = volume fraction
$\omega$ = Hagenbach correction (eq 3)

*Subscripts*

calc = calculated value
exp = experimental value
$i$ = pure component $i$
ref = reference term
RK = property calculated by Redlich−Kister polynomial

*Superscript*

‡ = activated state

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Received for review December 19, 2001. Accepted April 29, 2002. The authors are grateful to the Brazilian agencies CAPES, CNPq, FAPERJ, FINEP, FUJB, and FUNDCCMN for financial support.

JE010327V