

Densities, Viscosities, Refractive Indices, and Surface Tensions for the Ternary Mixtures of 2-Propanol + Benzyl Alcohol + 2-Phenylethanol at $T = 308.15$ K

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Densities, viscosities, refractive indices, and surface tensions for the ternary system of 2-propanol + benzyl alcohol + 2-phenylethanol were measured at $T = 308.15$ K and atmospheric pressure. Densities were determined using a vibrating-tube density meter. Viscosities were measured with an automatic Ubbelohde capillary viscometer. Refractive indices were measured using a digital Abbe-type refractometer. Surface tensions were determined by the Wilhelmy-plate method. The experimental data are used to calculate excess molar volumes V^E , deviations in the viscosity $\Delta\eta$, deviations in the refractive index Δn_D , and deviations in the surface tension $\Delta\sigma$. The calculated quantities V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ were fitted to the variable-degree polynomials.

Introduction

The flavor alcohols are the essential compounds contained in flavoring, perfumery, artificial essences, cosmetics, and industrial solvents. In a previous paper, we have determined densities, viscosities, refractive indices, and surface tensions for three binary systems formed by two flavor alcohols, such as benzyl alcohol and 2-phenylethanol, with 2-propanol at $T = (298.15, 308.15, \text{ and } 318.15)$ K.¹ In this work, we continue to measure the same properties for the ternary mixtures of 2-propanol + benzyl alcohol + 2-phenylethanol at $T = 308.15$ K. The experimental data are used to calculate excess molar volumes V^E , deviations in the viscosity $\Delta\eta$, deviations in the refractive index Δn_D , and deviations in the surface tension $\Delta\sigma$. Smooth representations of the results are described and used to construct the curves of constant V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ for the ternary mixtures. As far as we know, there is no literature data regarding the properties for the system proposed in this study.

Experimental Section

The mass purities and source of the chemicals employed are as follows: 2-propanol (Tedia, > 99.5 %); benzyl alcohol (Merck, > 99.5 %); 2-phenylethanol (Merck, > 99 %). The chemicals were used directly as received without further purification. The measured densities, viscosities, refractive indices, and surface tensions of pure components at $T = 308.15$ K along with the literature data available were shown in Table 1. The table indicates a substantial amount of differences in the refractive indices between this work and the literature values. These differences may not be solely due to the present experimental method but may more or less result from the experimental apparatus or compounds used.

Densities ρ were measured with an Anton Paar DMA-5000 vibrating-tube densimeter (Anton-Paar, Graz, Austria). The kinematic viscosities, ν , were determined with commercial Ubbelohde capillary viscometers of (0.46, 0.58, and 0.78) mm diameter (Cannon Instrument Co., State College, PA). The absolute viscosity, η , was calculated from the density ρ by the relation $\eta = \nu\rho$. Refractive indices, n_D , were measured with a

digital Abbe refractometer RX-5000 (ATAGO, Tokyo, Japan). Surface tensions σ were measured with an automatic surface tension meter model CBVP-A3 (Kyowa, Japan) using the Wilhelmy-plate method. The detailed measuring procedure has been described in a previous study (ref 1).

All samples were prepared by mass using a Precisa 262SMA balance with a precision of 10^{-5} g. The uncertainty in the composition is estimated to within $\pm 1 \cdot 10^{-4}$ mole fraction. All liquids were thermostatically controlled to within ± 0.01 K, ± 0.01 K, ± 0.05 K, and ± 0.05 K for the ρ , η , n_D , and σ measurements, respectively. All measurements were performed at least four times at atmospheric pressure (100.8 ± 0.4) kPa, and the results were averaged to give the final values. The uncertainty in the ρ , η , n_D , and σ measurements was estimated to be $\pm 4 \cdot 10^{-5}$ g·cm⁻³, ± 0.5 %, $\pm 8 \cdot 10^{-5}$, and ± 0.05 mN·m⁻¹, respectively.

Results and Discussion

Table 2 lists the experimental densities ρ , viscosities η , refractive indices n_D , surface tensions σ , excess molar volumes V^E , deviations in the viscosity $\Delta\eta$, deviations in the refractive index Δn_D , and deviations in the surface tension $\Delta\sigma$ for the 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) ternary system at $T = 308.15$ K. The molar excess volumes, V^E , were calculated from density data according to the following equation

$$V^E = \sum_{i=1}^3 x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right) \quad (1)$$

where x_i , M_i , and ρ_i are the mole fraction, molar mass, and density of the pure component i , respectively. ρ is the density of the mixture. The deviations in the viscosity, $\Delta\eta$, are given by

$$\Delta\eta = \eta - \sum_{i=1}^3 x_i \eta_i \quad (2)$$

where η is the absolute viscosity of the mixtures and η_i is the absolute viscosity of pure component i . The deviations in the refractive index, Δn_D , as stated by Fialkov,¹¹ were calculated

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Table 1. Comparison of Measured Densities, Viscosities, Refractive Indices, and Surface Tensions of Pure Components with Literature Values at $T = 308.15$ K

components	ρ g·cm ⁻³		η mPa·s		n_D		σ mN·m ⁻¹	
	exptl	lit.	exptl	lit.	exptl	lit.	exptl	lit.
2-propanol	0.77253	0.772559 ^a 0.77246 ^b 0.7726 ^c	1.550	1.550 ^a 1.542 ^b 1.5529 ^c	1.37063	1.3716 ^d	20.05	
benzyl alcohol	0.77243 ^d 1.03360 1.0337 ^f 1.03366 ^g	1.0336 ^e	1.5317 ^d 4.093 4.125 ^g	4.004 ^f 3.877 ^h	1.53421 1.5318 ^b	1.5350 ^e	37.73 37.9 ⁱ	37.63 ^e
2-phenylethanol	1.00863	1.0330 ^h 1.00879 ^g	7.518	7.438 ^g	1.52852	1.5267 ^g	38.93	

^a Contreras, 2001.² ^b Nikam et al., 1998.³ ^c Bhuiyan and Uddin, 2008.⁴ ^d Aralaguppi and Baragi, 2006.⁵ ^e Azizian and Bashavard, 2005.⁶ ^f Nikam and Kharat, 2003.⁷ ^g Francesconi et al., 2005.⁸ ^h Nayak et al., 2003.⁹ ⁱ Glinski et al., 1995.¹⁰

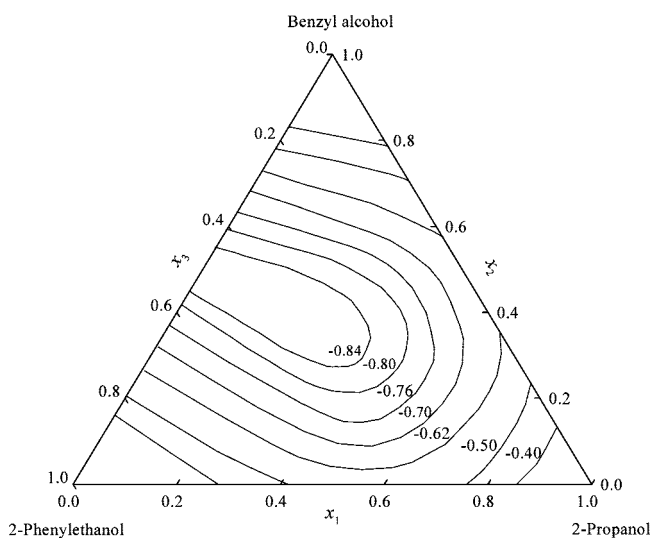


Figure 1. Iso-lines of constant V^E (cm³·mol⁻¹) for the 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) ternary system at $T = 308.15$ K.

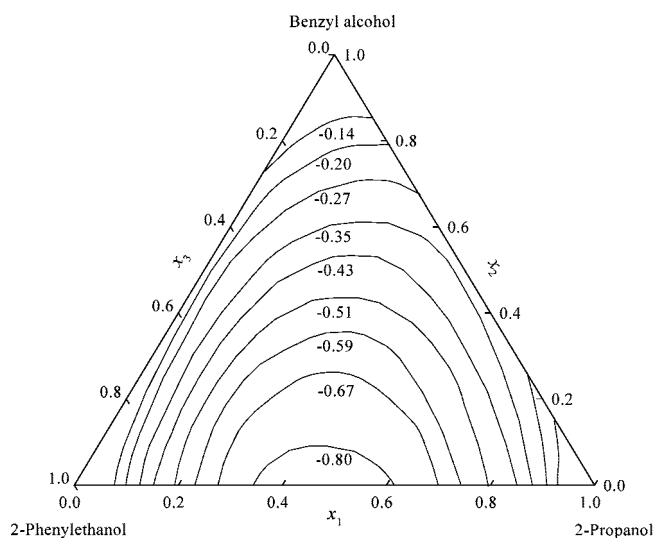


Figure 2. Iso-lines of constant $\Delta\eta$ (mPa·s) for the 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) ternary system at $T = 308.15$ K.

from the refractive indices of the pure liquids, n_{Di} , and their mixtures, n_D , as

$$\Delta n_D = n_D - \sum_{i=1}^3 \phi_i n_{Di} \quad (3)$$

where ϕ_i is the volume fraction of the i th component. The deviations in the surface tension $\Delta\sigma$ were calculated from the following equation

$$\Delta\sigma = \sigma - \sum_{i=1}^3 x_i \sigma_i \quad (4)$$

where σ is the surface tension of the mixture and σ_i is the surface tension of pure component i . These ternary V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ data at $T = 308.15$ K were included in Table 2.

As can be seen from Table 2, the V^E values at $T = 308.15$ K are negative over the whole mixtures investigated. These negative V^E values vary from -0.867 cm³·mol⁻¹ ($x_1 = 0.3001$, $x_2 = 0.3500$, $x_3 = 0.3499$) to -0.194 cm³·mol⁻¹ ($x_1 = 0.0500$, $x_2 = 0.9000$, $x_3 = 0.0500$). From the previous binary results (ref 1), the V^E values of 2-propanol + benzyl alcohol mixtures are more negative than those of 2-propanol + 2-phenylethanol mixtures. It may be said that the former system can form stronger hydrogen bonds between unlike molecules because of the shorter chain length of the benzyl alcohol and thus is a less spatial hindrance to the hydrogen

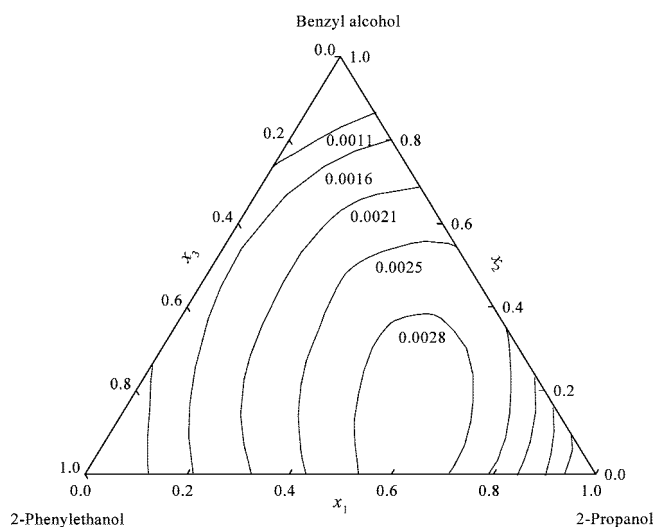


Figure 3. Iso-lines of constant Δn_D for the 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) ternary system at $T = 308.15$ K.

bonding formation. Alternately, the V^E values of benzyl alcohol + 2-phenylethanol mixtures are more negative than those of the 2-propanol + benzyl alcohol system. Probably the components, benzyl alcohol + 2-phenylethanol, strongly interact to form complex species upon mixing because of the π -electrons of benzene rings in both molecules. Thus, it

Table 2. Experimental Densities ρ , Viscosities η , Refractive Indices n_D , Surface Tensions σ , Excess Molar Volumes V^E , Deviations in the Viscosity $\Delta\eta$, Deviations in the Refractive Index Δn_D , and Deviations in the Surface Tension $\Delta\sigma$ for 2-Propanol (1) + Benzyl Alcohol (2) + 2-Phenylethanol (3) Ternary Mixtures at $T = 308.15$ K

x_1	x_2	ρ g·cm ⁻³	η mPa·s	n_D	σ mN·m ⁻¹	V^E cm ³ ·mol ⁻¹	$\Delta\eta$ mPa·s	$\Delta n_D \cdot 10^3$	$\Delta\sigma$ mN·m ⁻¹
0.0500	0.9000	1.02430	4.052	1.52848	36.66	-0.194	-0.085	0.71	-0.25
0.0500	0.8000	1.02375	4.339	1.52823	36.70	-0.419	-0.140	1.03	-0.32
0.0500	0.7000	1.02317	4.616	1.52795	36.75	-0.637	-0.206	1.27	-0.40
0.0501	0.5999	1.02183	4.940	1.52762	36.81	-0.774	-0.225	1.46	-0.46
0.0500	0.5000	1.01993	5.245	1.52719	36.88	-0.846	-0.262	1.53	-0.51
0.0500	0.4001	1.01699	5.597	1.52674	36.98	-0.799	-0.253	1.57	-0.53
0.0500	0.3000	1.01403	5.935	1.52613	37.14	-0.740	-0.257	1.43	-0.49
0.0499	0.2001	1.00995	6.297	1.52550	37.34	-0.541	-0.237	1.25	-0.41
0.0500	0.1000	1.00574	6.644	1.52470	37.55	-0.316	-0.233	0.91	-0.32
0.0500	0.0501	1.00382	6.852	1.52429	37.64	-0.218	-0.196	0.72	-0.29
0.1000	0.8500	1.01521	3.865	1.52260	35.59	-0.288	-0.145	1.11	-0.43
0.1000	0.7500	1.01482	4.132	1.52244	35.66	-0.516	-0.220	1.41	-0.48
0.1001	0.6500	1.01389	4.406	1.52217	35.71	-0.689	-0.288	1.59	-0.55
0.1000	0.5500	1.01259	4.715	1.52194	35.73	-0.816	-0.323	1.78	-0.65
0.1000	0.4500	1.01048	5.005	1.52163	35.81	-0.855	-0.375	1.89	-0.69
0.1000	0.3500	1.00751	5.350	1.52108	35.95	-0.792	-0.372	1.75	-0.67
0.1000	0.2500	1.00423	5.660	1.52057	36.11	-0.685	-0.405	1.63	-0.63
0.1000	0.1500	1.00026	6.034	1.51993	36.33	-0.488	-0.374	1.37	-0.53
0.1000	0.0500	0.99639	6.377	1.51926	36.52	-0.288	-0.373	1.08	-0.46
0.2000	0.7500	0.99578	3.543	1.51015	33.57	-0.430	-0.213	1.71	-0.68
0.2000	0.6500	0.99545	3.813	1.51010	33.59	-0.637	-0.285	1.92	-0.78
0.2000	0.5500	0.99453	4.042	1.51006	33.62	-0.783	-0.398	2.14	-0.87
0.2000	0.4500	0.99275	4.301	1.50994	33.65	-0.836	-0.483	2.26	-0.96
0.2001	0.3500	0.99052	4.612	1.50970	33.75	-0.837	-0.514	2.27	-0.98
0.2000	0.2500	0.98736	4.901	1.50931	33.91	-0.725	-0.568	2.09	-0.94
0.2001	0.1500	0.98426	5.214	1.50890	34.08	-0.611	-0.596	1.90	-0.89
0.1999	0.0501	0.98023	5.564	1.50849	34.32	-0.380	-0.590	1.71	-0.77
0.3000	0.6500	0.97514	3.200	1.49716	31.53	-0.564	-0.302	2.50	-0.96
0.3000	0.5500	0.97485	3.429	1.49710	31.55	-0.746	-0.415	2.48	-1.06
0.3000	0.4500	0.97372	3.679	1.49709	31.60	-0.841	-0.508	2.50	-1.13
0.3001	0.3500	0.97194	3.932	1.49707	31.69	-0.867	-0.597	2.53	-1.15
0.3000	0.2500	0.96966	4.205	1.49699	31.85	-0.829	-0.666	2.47	-1.12
0.3000	0.1500	0.96615	4.508	1.49682	32.01	-0.652	-0.706	2.33	-1.08
0.3000	0.0500	0.96285	4.808	1.49638	32.23	-0.485	-0.748	1.92	-0.98
0.4000	0.5500	0.95260	2.898	1.48250	29.62	-0.633	-0.349	2.40	-1.10
0.4000	0.4500	0.95210	3.131	1.48302	29.65	-0.760	-0.458	2.70	-1.19
0.4000	0.3500	0.95105	3.348	1.48333	29.68	-0.829	-0.584	2.81	-1.28
0.4000	0.2599	0.94962	3.655	1.48358	29.81	-0.836	-0.619	2.87	-1.26
0.4000	0.1499	0.94688	3.856	1.48352	29.95	-0.738	-0.761	2.61	-1.25
0.4001	0.0499	0.94373	4.158	1.48331	30.12	-0.566	-0.801	2.22	-1.20
0.5000	0.4501	0.92855	2.645	1.46711	27.83	-0.687	-0.348	2.41	-1.12
0.5000	0.3501	0.92803	2.829	1.46791	27.87	-0.774	-0.506	2.72	-1.20
0.4999	0.2501	0.92703	3.048	1.46869	27.90	-0.811	-0.629	3.03	-1.29
0.5000	0.1500	0.92520	3.297	1.46893	28.01	-0.757	-0.724	2.81	-1.30
0.5000	0.0499	0.92258	3.540	1.46897	28.15	-0.613	-0.823	2.41	-1.28
0.6000	0.3500	0.90228	2.372	1.45108	26.15	-0.669	-0.367	2.71	-1.03
0.6000	0.2500	0.90190	2.567	1.45208	26.18	-0.726	-0.514	2.91	-1.12
0.6000	0.1501	0.90095	2.778	1.45291	26.22	-0.723	-0.646	2.97	-1.20
0.5999	0.0500	0.89908	3.002	1.45352	26.25	-0.619	-0.764	2.82	-1.29
0.7000	0.2500	0.87395	2.143	1.43349	24.62	-0.608	-0.341	2.46	-0.79
0.7000	0.1500	0.87367	2.328	1.43486	24.68	-0.625	-0.499	2.68	-0.85
0.7001	0.0500	0.87263	2.529	1.43599	24.70	-0.574	-0.640	2.78	-0.95
0.8000	0.1500	0.84280	1.945	1.41471	23.08	-0.445	-0.285	2.13	-0.57
0.8000	0.0500	0.84273	2.118	1.41635	23.11	-0.423	-0.455	2.23	-0.66
0.9000	0.0500	0.80960	1.780	1.39400	21.55	-0.266	-0.196	1.40	-0.33

is expected to have more negative V^E values for the ternary mixtures when a larger amount of benzyl alcohol or 2-phenylethanol is involved in the mixtures.

It is observed that the $\Delta\eta$ values at $T = 308.15$ K are negative over the whole mole fractions studied. The values of $\Delta\eta$ vary from -0.823 mPa·s ($x_1 = 0.5000$, $x_2 = 0.0499$, $x_3 = 0.4501$) to -0.085 mPa·s ($x_1 = 0.0500$, $x_2 = 0.9000$, $x_3 = 0.0500$). The value of Δn_D was calculated on a volume fraction basis and in which case it has simple theoretical significance as reflecting changes in free volume.¹² As shown in the results, the Δn_D values at $T = 308.15$ K are positive

over the whole compositions investigated. These positive Δn_D values vary from 0.00071 ($x_1 = 0.0500$, $x_2 = 0.9000$, $x_3 = 0.0500$) to 0.00297 ($x_1 = 0.6000$, $x_2 = 0.1501$, $x_3 = 0.2499$). For the deviations in the surface tensions, the $\Delta\sigma$ values at $T = 308.15$ K are negative over the whole concentrations studied. The values of $\Delta\sigma$ vary from -1.30 mN·m⁻¹ ($x_1 = 0.500$, $x_2 = 0.1500$, $x_3 = 0.350$) to -0.25 mN·m⁻¹ ($x_1 = 0.0500$, $x_2 = 0.9000$, $x_3 = 0.050$).

In the previous paper (ref 1), we have reported the data of V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ for the binary systems of 2-propanol + benzyl alcohol, 2-propanol + 2-phenylethanol, and benzyl

Table 3. Binary Interaction Coefficients a_k of the Redlich–Kister Equation and Standard Deviations δ for V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ of Three Binary Systems at $T = 308.15$ K (Ref 1)

ΔQ	a_0	a_1	a_2	a_3	a_4	$\delta \cdot 10^3$
2-propanol (1) + benzyl alcohol (2)						
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	-2.567	-0.547	-0.186	0.115	-0.451	2.2
$\Delta\eta/\text{mPa} \cdot \text{s}$	-1.303	-0.060	0.398	-0.097	-0.660	3.0
Δn_D	0.00987	0.00193	0.00315			0.026
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	-4.433	0.607	4.232	0.259	-3.281	16.7
2-propanol (1) + 2-phenylethanol (3)						
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	-2.170	-0.768	-0.266	0.224	-0.627	3.1
$\Delta\eta/\text{mPa} \cdot \text{s}$	-3.438	0.336	-0.516	-0.175	0.676	5.1
Δn_D	0.01083	0.00457	0.00409	-0.00050		0.039
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	-5.390	-1.858	-0.283	3.283	1.322	16.2
benzyl alcohol (2) + 2-phenylethanol (3)						
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	-3.413	0.090	1.268	0.643		4.8
$\Delta\eta/\text{mPa} \cdot \text{s}$	-0.664	-0.154	0.323	0.034	-0.333	2.2
Δn_D	0.00531	-0.00013	-0.00023	-0.00113		0.026
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	-1.460	0.371	1.344	-0.396	-1.330	9.2

Table 4. Ternary Coefficients C_i 's of Equation 7 and Standard deviations δ of V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ for the 2-Propanol (1) + Benzyl Alcohol (2) + 2-Phenylethanol (3) Ternary System at $T = 308.15$ K

ΔQ_{123}	C_0	C_1	C_2	C_3	C_4	$\delta \cdot 10^2$
$V^E/\text{cm}^3 \cdot \text{mol}^{-1}$	6.467	-24.280	-5.969	17.712	26.982	1.8
$\Delta\eta/\text{mPa} \cdot \text{s}$	-2.839	4.207	4.185			14.1
Δn_D	-0.0123	-0.00181	0.0155			0.010
$\Delta\sigma/\text{mN} \cdot \text{m}^{-1}$	3.409	-6.531	-3.286	-7.504	15.808	3.5

alcohol + 2-phenylethanol at $T = 308.15$ K. These binary V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ data have been fitted to the Redlich–Kister type equation¹³

$$\Delta Q(x_i, x_j) = x_i x_j \sum_{k=0}^m a_k (x_i - x_j)^k \quad (5)$$

where $\Delta Q(x_i, x_j)$ refers to binary V^E , $\Delta\eta$, Δn_D , or $\Delta\sigma$ expressed as a function of mole fractions x_i and x_j of pure components i and j , respectively. The values of the binary interaction coefficients a_k 's for these three binary systems were shown in Table 3.

The ternary V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ data of 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) at $T = 308.15$ K were correlated, respectively, using the equation

$$\Delta Q_{123} = \Delta Q(x_1, x_2) + \Delta Q(x_1, x_3) + \Delta Q(x_2, x_3) + x_1 x_2 x_3 \Delta_{123} \quad (6)$$

where ΔQ_{123} refers to ternary V^E , $\Delta\eta$, Δn_D , or $\Delta\sigma$ and $x_3 = 1 - x_1 - x_2$. ΔQ 's are the binary contribution functions for V^E , $\Delta\eta$, Δn_D , or $\Delta\sigma$ as defined in eq 5. The ternary contribution term Δ_{123} was correlated using the expression of the following form

$$\Delta_{123} = C_0 + C_1 x_1 + C_2 x_2 + C_3 x_1 x_2 + C_4 x_1^2 \quad (7)$$

The ternary parameters C_i 's were determined from a least-squares procedure, and the result is shown in Table 4. The standard deviation, δ , is defined as

$$\delta = \left[\sum_{i=1}^n \frac{(\Delta Q_{123i}^{\text{exptl}} - \Delta Q_{123i}^{\text{calcd}})^2}{n - p} \right]^{1/2} \quad (8)$$

where n is the number of data points and p is the number of adjustable parameters.

The curves of constant V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ for the 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) mixtures at $T = 308.15$ K were calculated from eqs 6 and 7 and plotted in Figures 1 to 4, respectively. As can be seen from Figure 1, the ternary system shows the negative values of V^E at all compositions. The minimum V^E value is found near 2-propanol + benzyl alcohol at $x \approx 0.55$ of 2-propanol.

Figure 2 shows the negative values for ternary $\Delta\eta$ at all compositions, with a minimum value near the 2-propanol + 2-phenylethanol at $x \approx 0.50$ of 2-propanol. Curves of constant Δn_D in Figure 3 show the positive values of Δn_D at all compositions, with a maximum value close to 2-propanol + benzyl alcohol at $x \approx 0.60$ of 2-propanol. Curves of constant $\Delta\sigma$ in Figure 4 show the negative values of $\Delta\sigma$ at all compositions, with a minimum value close to 2-propanol + benzyl alcohol at $x \approx 0.55$ of 2-propanol.

Conclusion

This paper reports experimental data of densities, viscosities, refractive indices, and surface tensions for the 2-propanol + benzyl alcohol + 2-phenylethanol ternary system at $T = 308.15$ K and atmospheric pressure. Over the concentration range studied, the excess molar volume V^E , the deviations in the viscosity $\Delta\eta$, and the deviations in the surface tension

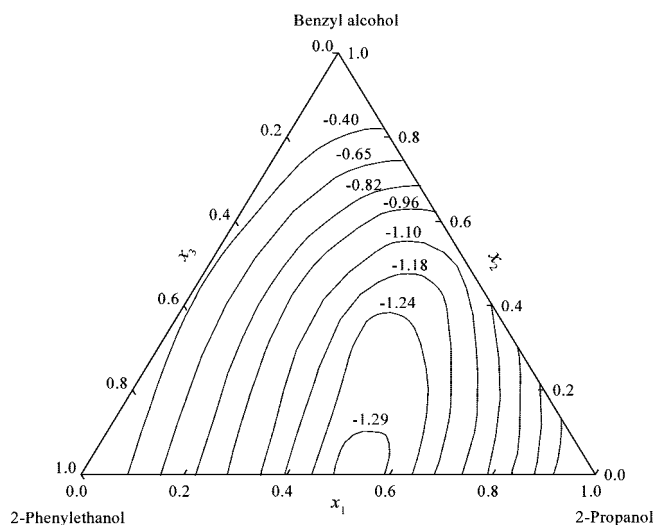


Figure 4. Iso-lines of constant $\Delta\sigma$ ($\text{mN} \cdot \text{m}^{-1}$) for the 2-propanol (1) + benzyl alcohol (2) + 2-phenylethanol (3) ternary system at $T = 308.15$ K.

$\Delta\sigma$ are negative, whereas the deviations in the refractive index Δn_D are positive. The calculated ternary V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ data were correlated using variable-degree polynomials along with a predetermined binary result. The correlation results are used to construct the curves of constant V^E , $\Delta\eta$, Δn_D , and $\Delta\sigma$ for the ternary mixtures.

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