Densities, Viscosities, and Refractive Indices of Binary Mixtures of Methyl Ethyl Ketone + Pentanol Isomers at Different Temperatures

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Densities, viscosities, and refractive indices of mixing of methyl ethyl ketone with 1-pentanol, 2-pentanol, and 3-pentanol have been measured as a function of composition range at temperatures (298.15, 308.15, and 318.15) K and ambient pressure. Excess molar volumes V_m^E, viscosity deviations $\Delta\eta$, and refractive index deviations Δn_D were calculated and correlated by the Redlich−Kister-type function to derive the coefficients and estimate the standard error. From the experimental data, partial molar volumes, $\bar{V}_{m,i}$, and partial molar volumes at infinite dilution, $\bar{V}^0_{m,i}$, were also calculated. The latter values are interesting from a theoretical point of view since at infinite dilution the only interactions present are solute−solvent interactions. The effect of temperature and −OH group position of pentanol isomers on the excess molar volumes, viscosity, and refractive index deviations of its mixtures with methyl ethyl ketone is discussed in terms of the nature and type of intermolecular interactions in binary mixtures.

Introduction

It is well-known that the thermodynamic properties of liquid mixtures depend on the way in which molecules of pure solvents are associated during the mixing process. From this point of view, a study of the thermomechanical behavior of solvent mixtures should provide an important source of structural information at the microscopic level.

Properties such as density, viscosity, and refractive index deviation of pure chemicals and of their binary liquid mixtures over the whole composition range measured at several temperatures are useful for a full understanding of their thermodynamic and transport properties as well as for practical chemical engineering purposes. On the other hand, excess thermodynamic functions and deviations of nonthermodynamic ones of binary liquid mixtures are fundamental for understanding of the interactions between molecules in these types of mixtures, particularly when polar components are involved. These functions have also been used as a qualitative and quantitative guide to predict the extent of complex formation in this kind of mixtures.[1–3]

Excess and deviation properties of binary mixtures are complex properties because they depend not only on solute−solute, solvent−solvent, and solute−solvent interactions but also on the structural effects arising from interstitial accommodation. Knowledge of several properties at different temperatures is required for engineering design and for subsequent operations.[4]

The main purpose of the preceding papers of this series[5,6] was to provide a set of thermodynamic data for the analysis and characterization of molecular interactions in binary liquid mixtures containing alkanols with nitrils and ketones. In continuation with these investigations, the present paper reports the excess molar volumes, viscosity, and refractive index deviations for methyl ethyl ketone + 1-pentanol, methyl ethyl ketone + 2-pentanol, and methyl ethyl ketone + 3-pentanol at temperatures (298.15, 308.15, and 318.15) K, with the aim of analyzing the influence of temperature and −OH group of pentanol isomers upon the aforementioned excess properties.

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Table 1. Experimental and Literature Values of Densities ρ, Viscosities η, and Refractive Indices n_D of Methyl Ethyl Ketone, 1-Pentanol, 2-Pentanol, and 3-Pentanol, at $T = 298.15$ K

	ρ / g·cm^{-3}		η / mPa·s		n_D	
compd	exptl	lit.	exptl	lit.	exptl	lit.
methyl ethyl ketone	0.79976	0.7999[a]	0.391	0.3855[b]	1.3750	1.3769[c]
1-pentanol	0.81085	0.81083[d]	3.540	3.5344[c]	1.4068	1.4078[d]
2-pentanol	0.80530	0.80540[d]	3.478	3.478[e]	1.4032	1.4044[f]
3-pentanol	0.81579	0.81552[g]	4.625	4.598[h]	1.4071	1.4079[d]

[a] From ref 7. [b] From ref 8. [c] From ref 9. [d] From ref 11. [e] From ref 5. [f] From ref 10. [g] From ref 12. [h] From ref 13.

Experimental Section

Chemicals. Methyl ethyl ketone, 1-pentanol, 2-pentanol, and 3-pentanol were purchased from Merck with mass fraction higher than 99 % and used without further purifications. The purity of reagents was checked by comparing the measured densities, viscosities, and refractive indices at 298.15 K with those reported in the literature.[5,7–13] The results are in good agreement with values found in the literature and reported in Table 1.

Apparatus and Procedure. The density of the pure compounds and mixtures was measured by an Anton Parr DMA 4500, provided with automatic viscosity correction. The uncertainty of the density measurements was estimated to be $\pm 1 \cdot 10^{-5}$ g·cm^{-3}. The apparatus was calibrated once a day with dry air and bidistillated water. The temperature in the cell was regulated to ± 0.01 K with a solid state thermostat. Viscosities were measured with an Ubbleohde viscometer with an uncertainty of $\pm 2 \cdot 10^{-3}$ mPa·s. The equation for viscosity, according to Poiseuille's law, is

$$\eta = \rho(kt - c/t) \quad (1)$$

were k and c are the viscometer constants and t and are the efflux time and dynamic viscosity, respectively. The k and c

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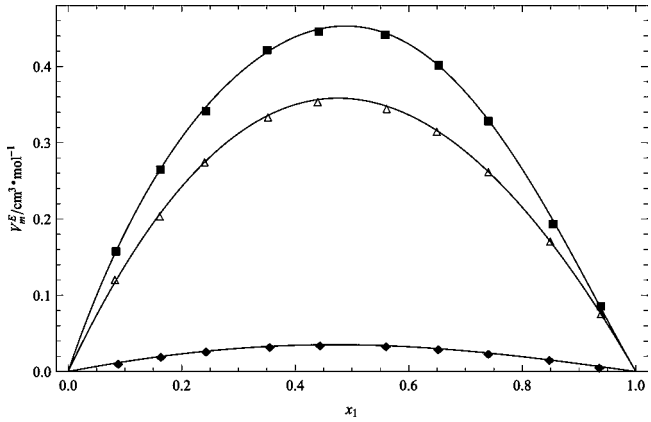

Figure 1. Excess molar volumes V_m^E vs mole fraction of methyl ethyl ketone for binary mixtures of methyl ethyl ketone with ♦, 1-pentanol; Δ, 2-pentanol; and ■, 3-pentanol at $T = 298.15$ K. The solid curves were calculated from coefficients of eq 3 given in Table 3.

parameters were obtained by measurements on double distilled water and benzene at 298.15 K. The temperature in the cell was regulated to ± 0.01 K.

Refractive indices were measured using a digital Abbe refractometer (model: DR-A1). The uncertainty of refractive index measurement was estimated to be ± 0.0004 units. The measurement method relies on an optical detection of the critical angle at the wavelength of the sodium D line (589.6 nm). The mixtures were prepared by weighing known masses of pure liquids in airtight, narrow-mouth ground stoppered bottles taking due precautions to minimize evaporation losses. All of the mass measurements were performed on an electronic balance (model: Mettler AE 163, Switzerland) accurate to 0.01 mg. The uncertainty in the mole fraction was estimated to be $\pm 1 \cdot 10^{-4}$.

Results and Discussion

Densities and Excess Molar Volumes. The excess molar volumes of the solutions of molar composition x measured at temperatures (298.15, 308.15, and 318.15) K are calculated from the densities of the pure liquids and their mixtures according to following equation

$$V_m^E = \sum_{i=1}^{N} x_i M_i (\rho^{-1} - \rho_i^{-1}) \quad (2)$$

where ρ is the density of the mixture; ρ_i is the density of pure component i; x_i is the mole fraction; M_i is the molar mass of component i; and N stands for the number of components in the mixture.

The corresponding V_m^E values of binary mixtures of methyl ethyl ketone (1) + pentanols (2) plotted against mole fraction of methyl ethyl ketone at $T = 298.15$ K are presented in Figure 1. The uncertainty for excess molar volume is $\pm 1 \cdot 10^{-3}$ cm^3·mol^{-1}. The values of densities for binary mixtures, at different temperatures, are reported in Table 2. Each set of results was fitted using a Redlich−Kister polynomial,[14] which for binary mixtures is

Table 2. Densities ρ, Viscosities η, and Refractive Indices n_D of Mixing for the Binary Mixtures as a Function of the Mole Fraction x_1 of Methyl Ethyl Ketone

x_1	ρ / g·cm^{-3}	η / mPa·s	n_D	x_1	ρ / g·cm^{-3}	η / mPa·s	n_D	x_1	ρ / g·cm^{-3}	η / mPa·s	n_D
	Methyl Ethyl Ketone (1) + 1-Pentanol (2)				Methyl Ethyl Ketone (1) + 2-Pentanol (2)				Methyl Ethyl Ketone (1) + 3-Pentanol (2)		
T/K = 298.15				T/K = 298.15				T/K = 298.15			
0.0000	0.81085	3.540	1.4068	0.0000	0.80530	3.478	1.4032	0.0000	0.81579	4.625	1.4071
0.0880	0.80994	2.557	1.4049	0.0824	0.80400	2.664	1.4014	0.0843	0.81343	2.842	1.4053
0.1632	0.80915	1.997	1.4031	0.1612	0.80299	2.066	1.3998	0.1622	0.81150	2.007	1.4035
0.2427	0.80831	1.592	1.4010	0.2401	0.80204	1.690	1.3980	0.2428	0.80972	1.494	1.4014
0.3549	0.80711	1.202	1.3979	0.3519	0.80097	1.347	1.3952	0.3505	0.80745	1.084	1.3983
0.4434	0.80615	0.983	1.3953	0.4396	0.80031	1.131	1.3929	0.4415	0.80582	0.860	1.3956
0.5597	0.80487	0.773	1.3915	0.5608	0.79965	0.886	1.3895	0.5584	0.80392	0.677	1.3918
0.6518	0.80385	0.648	1.3883	0.6494	0.79934	0.743	1.3868	0.6526	0.80264	0.591	1.3885
0.7401	0.80285	0.554	1.3851	0.7403	0.79920	0.623	1.3840	0.7399	0.80172	0.508	1.3853
0.8477	0.80159	0.485	1.3810	0.8491	0.79926	0.505	1.3803	0.8540	0.80080	0.445	1.3809
0.9355	0.80055	0.420	1.3776	0.9384	0.79949	0.436	1.3772	0.9383	0.80016	0.407	1.3774
1.0000	0.79976	0.391	1.3750	1.0000	0.79976	0.391	1.3750	1.0000	0.79976	0.391	1.3750
T/K = 308.15				T/K = 308.5				T/K = 308.15			
0.0000	0.80345	2.658	1.4031	0.0000	0.79700	2.361	1.3990	0.0000	0.80691	2.864	1.4029
0.0880	0.80226	2.011	1.4014	0.0824	0.79554	1.778	1.3974	0.0843	0.80458	1.965	1.4012
0.1632	0.80125	1.616	1.3997	0.1612	0.79434	1.381	1.3959	0.1622	0.80260	1.476	1.3995
0.2427	0.80016	1.304	1.3976	0.2401	0.79329	1.105	1.3941	0.2428	0.80057	1.151	1.3975
0.3549	0.79862	1.009	1.3943	0.3519	0.79197	0.865	1.3914	0.3505	0.79807	0.869	1.3944
0.4434	0.79739	0.835	1.3914	0.4396	0.79107	0.731	1.3890	0.4415	0.79623	0.717	1.3916
0.5597	0.79576	0.667	1.3873	0.5608	0.79015	0.587	1.3854	0.5584	0.79410	0.578	1.3876
0.6518	0.79444	0.570	1.3838	0.6494	0.78963	0.518	1.3826	0.6526	0.79264	0.506	1.3841
0.7401	0.79314	0.492	1.3804	0.7403	0.78923	0.462	1.3796	0.7399	0.79155	0.448	1.3806
0.8477	0.79154	0.427	1.3760	0.8491	0.78902	0.406	1.3757	0.8540	0.79031	0.396	1.3760
0.9355	0.79021	0.380	1.3725	0.9384	0.78909	0.371	1.3724	0.9383	0.78970	0.365	1.3725
1.0000	0.78921	0.361	1.3700	1.0000	0.78921	0.361	1.3700	1.0000	0.78921	0.361	1.3700
T/K = 318.5				T/K = 318.15				T/K = 318.15			
0.0000	0.79590	2.288	1.3996	0.0000	0.78837	1.772	1.3950	0.0000	0.79766	1.902	1.3981
0.0880	0.79444	1.608	1.3976	0.0824	0.78683	1.344	1.3937	0.0843	0.79503	1.382	1.3965
0.1632	0.79321	1.321	1.3957	0.1612	0.78550	1.094	1.3923	0.1622	0.79288	0.991	1.3949
0.2427	0.79189	1.097	1.3937	0.2401	0.78432	0.897	1.3906	0.2428	0.79072	0.724	1.3929
0.3549	0.78999	0.852	1.3905	0.3519	0.78276	0.721	1.3877	0.3505	0.78817	0.520	1.3901
0.4434	0.78850	0.721	1.3878	0.4396	0.78166	0.642	1.3852	0.4415	0.78628	0.417	1.3873
0.5597	0.78650	0.585	1.3838	0.5608	0.78047	0.514	1.3813	0.5584	0.78416	0.331	1.3835
0.6518	0.78488	0.500	1.3804	0.6494	0.77991	0.457	1.3782	0.6526	0.78270	0.294	1.3800
0.7401	0.78331	0.443	1.3770	0.7403	0.77915	0.407	1.3750	0.7399	0.78162	0.292	1.3767
0.8477	0.78136	0.381	1.3724	0.8491	0.77867	0.366	1.3711	0.8540	0.78016	0.297	1.3720
0.9355	0.77973	0.345	1.3686	0.9384	0.77855	0.338	1.3679	0.9383	0.77921	0.312	1.3684
1.0000	0.77852	0.334	1.3657	1.0000	0.77852	0.334	1.3657	1.0000	0.77852	0.334	1.3657

$$Y^{E} = x_1(1 - x_1)\sum_{k=0}^{N} A_k(1 - 2x_1)^k \qquad (3)$$

where $Y^{E} \equiv V_m^{E}$ or $\Delta\eta$ or Δn_D and x_1 is the mole fraction of ethyl methyl ketone. A_k is the adjustable parameter obtained by the least-squares method, and k is the degree of the polynomials. In each case, the optimum number of coefficients was ascertained from an examination of the variation of standard deviation σ. Table 3 presents the values of the parameters A_k together with the standard deviation σ.

Excess molar volumes of all binary mixtures are positive and increase slightly with increasing temperature. When alcohols, which exist in a highly associated form in the pure state, are mixed with polar solvents (ketones), the monomerization occurs and new specific interactions appear in the solution. The disruption of the hydrogen-bonded alcohol structure gives rise to a positive contribution to V_m^{E}, which depends: (i) on the dielectric constant of the ketone[15] and (ii) on the chain length[16] and degree of branching in the alcohol which decrease the self-association in the pure state. On the other hand, the interactions between unlike molecules in both systems are surely weaker than the sum of the interactions between like molecules. These effects produce positive excess molar volumes.

As seen, at each temperature V_m^{E} values are in the sequence: 3-pentanol > 2-pentanol > 1-pentanol. Excess molar volumes for each binary mixture increase with increasing temperature. As an example, Figure 2 shows the effect of temperature on V_m^{E} of methyl ethyl ketone + 1-pentanol. Reddy and Naidu[17] also reported the density of methyl ethyl ketone + 1-pentanol at temperatures (303.15 and 308.15) K which were in good consistency comparing our results. The data show that the density of the mixture and pure state decreases with an increase in the temperature. Partial molar quantities are defined as the rate of change with concentration of extensive functions and account for binary and higher-order interactions between components.

The partial molar volumes $\bar{V}_{m,i}{}^{0}$ in these mixtures were calculated over the whole composition range using eqs 4 and 5.[7,18]

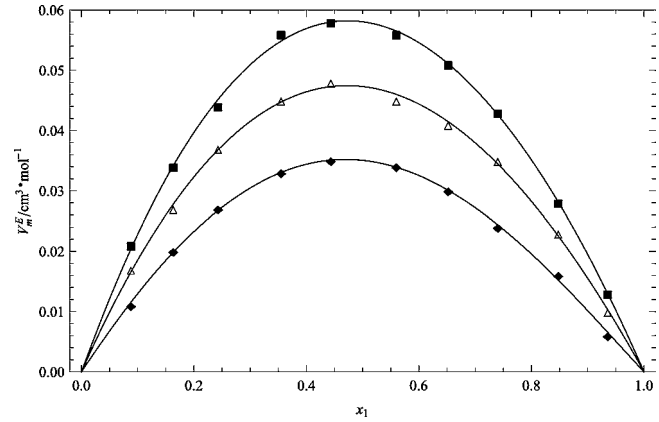

Figure 2. Excess molar volumes V_m^{E} vs mole fraction of methyl ethyl ketone for binary mixtures of methyl ethyl ketone with 1-pentanol at: ♦, $T = 298.15$ K; Δ, $T = 308.15$ K; ■, $T = 318.15$ K. The solid curves were calculated from coefficients of eq 3 given in Table 3.

$$\bar{V}_{m,1} = V_m^{E} + V_{m,1}^{*} + (1 - x_1)(\partial V_m^{E}/\partial x_1)_{T,P} \qquad (4)$$

$$\bar{V}_{m,2} = V_m^{E} + V_{m,2}^{*} - x_1(\partial V_m^{E}/\partial x_1)_{T,P} \qquad (5)$$

where $V_{m,1}^{*}$ and $V_{m,1}^{*}$ are pure molar volumes of components 1 and 2, respectively.

Values of partial molar volumes at infinite dilution, $\bar{V}_{m,i}^{0}$, are in Table 4.

The partial properties at infinite dilution are of interest since at the limit of infinite dilution the solute−solute interactions disappear and the only interactions present are solute−solvent interactions. Since the partial molar volumes at infinite dilution of each component are not very different from the corresponding molar volumes $V_{m,1}^{*}$, interaction between methyl ethyl ketone and alcohols is not very favorable. $\bar{V}_{m,i}^{0}$ increases slightly with increasing temperature.

Dynamic Viscosities. The viscosity deviation can be calculated as

Table 3. Parameters A_k and Standard Deviations σ for Methyl Ethyl Ketone + Pentanol Isomers at Different Temperatures

			T/K	A_0	A_1	A_2	A_3	σ
methyl ethyl ketone + 1-pentanol	V_m^{E}	cm^3·mol^{-1}	298.15	0.1402	0.0194	−0.0184		0.0005
			308.15	0.1892	0.0211			0.0006
			318.15	0.2327	0.038			0.0007
	$\Delta\eta$	mPa·s	298.15	−4.354	−2.660	−2.086	−1.394	0.006
			308.15	−3.044	−1.9149	−1.152		0.009
			318.15	−2.595	−1.429	−1.786	−1.6075	0.01
	Δn_D		298.15	0.0102	0.001			0.00002
			308.15	0.0116	0.0049			0.00005
			318.15	0.013	0.0001			0.00005
methyl ethyl ketone + 2-pentanol	V_m^{E}	cm^3·mol^{-1}	298.15	1.423	0.1457			0.002
			308.15	1.489	0.0962			0.004
			318.15	1.4875	0.0563			0.007
	$\Delta\eta$	mPa·s	298.15	−3.730	−2.356	−2.093	−1.039	0.01
			308.15	−2.823	−1.833	−1.33	−0.5236	0.006
			318.15	−1.915	−1.094	−1.106	−0.668	0.009
	Δn_D		298.15	0.008	−0.0004			0.00003
			308.15	0.0112	0.0005			0.00004
			318.15	0.0118	0.0057			0.00003
methyl ethyl ketone + 3-pentanol	V_m^{E}	cm^3·mol^{-1}	298.15	1.8119	0.2104	−0.065		0.007
			308.15	1.884	0.022	−0.1838		0.004
			318.15	1.9435	0.4188	−0.1983		0.005
	$\Delta\eta$	mPa·s	298.15	−6.864	−5.146	−5.752	−4.543	0.03
			308.15	−3.843	−2.658	−2.493	−1.564	0.01
			318.15	−3.028	−1.9032	−0.967		0.01
	Δn_D		298.15	0.0107	0.0011			0.00004
			308.15	0.0127	0.003			0.00005
			318.15	0.0142	0.0018			0.00004

Table 4. Partial Molar Volumes at Infinite Dilution, $\bar{V}^0_{m,i}$, for Methyl Ethyl Ketone + Pentanols at Different Temperatures

system		T/K = 298.15	T/K = 308.15	T/K = 318.15
		Methyl Ethyl Ketone + 1-Pentanol		
$\bar{V}^0_{m,1}$	$cm^3 \cdot mol^{-1}$	90.31	91.57	92.87
$\bar{V}^0_{m,2}$	$cm^3 \cdot mol^{-1}$	108.83	109.88	110.97
		Methyl Ethyl Ketone + 2-Pentanol		
$\bar{V}^0_{m,1}$	$cm^3 \cdot mol^{-1}$	91.69	92.92	94.17
$\bar{V}^0_{m,2}$	$cm^3 \cdot mol^{-1}$	110.79	112.03	113.26
		Methyl Ethyl Ketone + 3-Pentanol		
$\bar{V}^0_{m,1}$	$cm^3 \cdot mol^{-1}$	92.05	93.22	94.83
$\bar{V}^0_{m,2}$	$cm^3 \cdot mol^{-1}$	109.69	111.04	112.04

$$\Delta\eta = \eta - x_1\eta_1 - x_2\eta_2 \quad (6)$$

where η is the viscosity of the mixture and η_1 and η_2 are pure component viscosity. The values η for binary systems at temperatures (298.15, 308.15, and 318.15) K are listed in Table 2. The values of $\Delta\eta$ at T = 298.15 K are shown in Figure 3. The uncertainty for viscosity deviation is $\pm 4 \cdot 10^{-3}$ mPa·s. $\Delta\eta$ values were fitted to Redlich−Kister eq 3, and the adjustable parameters, A_k, and standard deviations, σ, are given in Table 3.

The values of $\Delta\eta$ for the system containing methyl ethyl ketone + pentanol isomers are asymmetrical and all negative throughout the whole concentration range at all temperatures.

According to Kauzman and Eyring,[19] the viscosity of a mixture strongly depends on the entropy of the mixture, which is related with the liquid's structure and enthalpy (and consequently with molecular interactions between the components of the mixture). Therefore, the viscosity deviation depends on molecular interactions as well as on the size and shape of the molecules. Vogel and Weiss[20] affirm that mixtures with positive deviation of Raoult's law and without strong specific interactions present the negative viscosity deviation. The negative values of $\Delta\eta$ for all the binary systems fall in the order: 3-pentanol > 1-pentanol > 2-pentanol.

Refractive Index. Refractive index deviations were defined by

$$\Delta n_D = n_D - x_1 n_{D1} - x_2 n_{D2} \quad (7)$$

where n_D and n_{Di} are the refractive index of the mixture and the refractive index of the pure component i, respectively. The experimental refractive indices of the binary mixtures, n_D, at temperatures (298.15, 308.15, and 318.15) K are listed in Table 2. The values of Δn_D at T = 298.15 K are shown in Figure 4. The results were fitted by eq 3, and the adjustable parameters and standard deviations are given in Table 3.

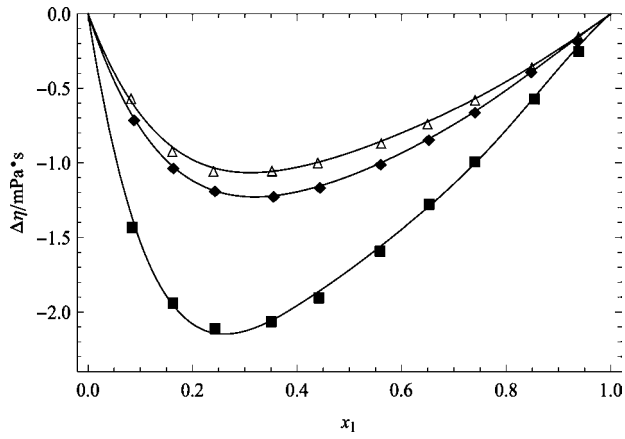

Figure 3. Viscosity deviations $\Delta\eta$ vs mole fraction of methyl ethyl ketone, for binary mixtures of methyl ethyl ketone with with ♦, 1-pentanol; Δ, 2-pentanol; and ■, 3-pentanol at T = 298.15 K. The solid curves were calculated from coefficients of eq 3 given in Table 3.

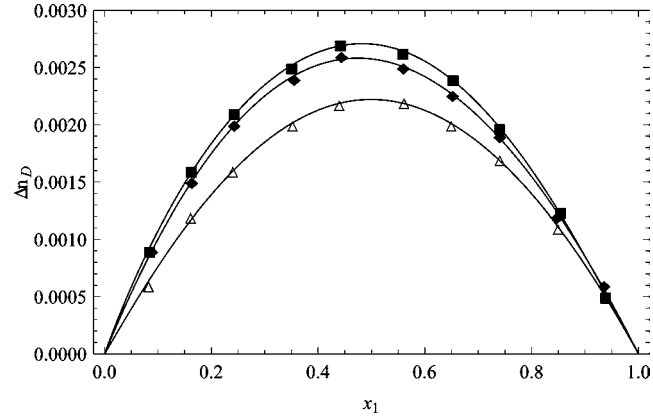

Figure 4. Refractive index deviations Δn_D vs mole fraction of methyl ethyl ketone, for binary mixtures of methyl ethyl ketone with ♦, 1-pentanol; Δ, 2-pentanol; and ■, 3-pentanol at T = 298.15 K. The solid curves were calculated from coefficients of eq 3 given in Table 3.

It can be seen that the changes in refractive indices of pentanol isomers with methyl ethyl ketone are positive throughout the entire composition range; while the temperature increases, the change in the refractive index becomes more positive. The algebraic values of Δn_D for all the binary systems fall in the order: 3-pentanol > 1-pentanol > 2-pentanol.

Conclusions

Excess molar volumes, viscosity, and refractive index deviations for mixtures of methyl ethyl ketone and pentanol isomers at temperatures (298.15, 308.15, and 318.15) K were calculated from experimental results and fitted by the Redlich−Kister type equation. Excess molar volumes and refractive index deviations are positive for all binary mixtures. Positive quantities show that dominant factors are physical interactions. Viscosity deviations are negative for all of the mixtures. The negative values for all systems are due to the dissociation of the associated structures of pentanols in methyl ethyl ketone. Excess molar volume and refractive index deviations increase with increasing temperature, but the values of viscosity deviations decrease.

Acknowledgment

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Supporting Information Available:

Excess molar volumes, partial molar volumes, viscosity, and refractive index deviations for the mixtures of methyl ethyl ketone and pentanol isomers. This material is available free of charge via the Internet at http://pubs.acs.org.

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