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Thermophysical Properties of Binary Mixtures of 2-Methyl-1-propanol with Hexane, Octane, and Decane at 298.15 K

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Densities, viscosities, and speeds of sound have been measured for the binary mixtures of 2-methyl-1-propanol $\{(CH_3)_2CHCH_2OH\}$ with *n*-hexane (C_6H_{14}), *n*-octane (C_8H_{18}), and *n*-decane ($C_{10}H_{22}$) over the entire composition range at 298.15 K and at atmospheric pressure. The experimental density (ρ), viscosity (η) and speed of sound (u) values were used to calculate the excess molar volumes (V^E), viscosity deviation ($\Delta\eta$), deviation in speed of sound (Δu), isentropic compressibility (κ_s), and deviation in isentropic compressibility ($\Delta\kappa_s$). The values of V^E, $\Delta\eta$, Δu and $\Delta\kappa_s$ were fitted to the Redlich−Kister polynomial equation and have been used to discuss the presence of significant interactions between alcohols and alkane molecules.

Introduction

In mixtures of the associating molecules, alcohols, and non-associating molecules, the alkanes are of particular interest from a theoretical viewpoint of the models of the hydrogen-bonded systems. Alcohol molecules associate mainly by building up linear chains. The extent of this association depends in a specific way on the concentration of the alcohols in the liquid state and on temperature.[1−10] Systematic studies of the thermodynamic excess properties of alkane and alkanol mixtures are important from the viewpoint of understanding the molecular liquid structure and the intermolecular interactions dominated by the self-association of alkanol molecules due to hydrogen bonding. Alcohols are the well-known solvents with protic and self-associated properties, which are used to study the hydrophobic effects. Thus accurate knowledge of their excess thermodynamic mixing properties such as volume, enthalpy, and Gibbs free energy has great relevance in theoretical and applied areas of research. Alkanols and ethers are usually added to gasoline as oxygenates and replace lead antiknock agents to inhibit the combustion emission and replace pollution. 2-Methyl-1-propanol is added in the oxygenated gasoline with ethanol to avoid the formation of the liquid phases in the presence of a small quantity of water.[11] Alkanes are an important series of homologous, nonpolar, organic solvents. They have often been used in the study of solute dynamics because their physicochemical properties as a function of chain length are well-known. They are also employed in a large range of chemical processes.[12−14]

The present work is a part of our systematic studies on thermodynamic properties for mixtures of great interest in several chemical industries. This study concerns experimentally measured values of densities, ρ, viscosities, η, and speed of sound u for binary mixtures of 2-methyl-1-propanol with *n*-hexane, *n*-octane, and *n*-decane at the temperature of 298.15 K and at atmospheric pressure. Furthermore, using the experimental results, excess molar volumes V^E, deviation in viscosity $\Delta\eta$, deviation in speed of sound Δu, isentropic compressibility, κ_s, and deviation in isentropic compressibility, $\Delta\kappa_s$, have been calculated in order to gain a better understanding of the intermolecular interactions between the component molecules. These excess properties have been correlated with composition using the Redlich−Kister polynomial equation.[15]

Experimental Section

Materials. 2-Methyl-1-propanol, hexane, and octane were obtained from S.D. Fine Chemicals Ltd. whereas decane was supplied from Spectrochem. The stated label purities of these reagents ranged from (99 to 99.9) % in mass. All the chemicals were of AR grade and were partially degassed before use. The experimentally measured values of densities, viscosities, and speeds of sound of all the pure components at 298.15 K are given in Table 1. It can be easily seen that the experimental values of all the measured properties are in close agreement with those reported in the literature.[16−25]

Apparatus and Procedure. Densities were measured by a bicapillary pycnometer with a bulb of about 10 cm^3 and a capillary with an internal diameter of about 1 mm. The precision in density measurements is $\pm$ 0.0002 $\times$ 10^3 $kg{\cdot}m^{-3}$. The pycnometer was calibrated with deionized double-distilled water with 0.9970 $\times$ 10^3 $kg{\cdot}m^{-3}$ as its density and compared with literature[26] at 298.15 K. The pycnometer, filled with the desired liquid, was kept in a transparent-walled water bath with a thermal stability of $\pm$ 0.01 K, as checked by means of a calibrated thermometer, to attain thermal equilibrium.

The kinematic viscosities ν ($= \eta/\rho$) of pure liquids and liquid mixtures were measured at 298.15 K and at atmospheric pressure using an Ubbelohde suspended level viscometer.[27] The viscometer was calibrated so as to determine the two constants A and B in the equation $\eta/\rho = At - B/t$, obtained by measuring the flow time t with double distilled water and cyclohexane.[28] The viscometer is filled with liquid or liquid mixtures, and its limbs were closed with Teflon caps, taking due precautions to minimize the evaporation losses. The flow time measurements were made by using an electronic stopwatch with a precision of $\pm$ 0.01 s. An average of four or five sets of flow times for each liquid or liquid mixtures was taken for the purpose of calculations of viscosity. The measured values of kinematic viscosities were converted to dynamic viscosities after multiplication by the density. The reproducibility of the viscosity estimates was found to be within $\pm$ 0.006 mPa·s.

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Table 1. Comparison of Experimental Densities ρ, Viscosities η, Speeds of Sound u, and Isentropic Compressibility κ_s with Literature Data for Pure Liquids at 298.15 K

	$\rho \times 10^{-3}$/kg·m^{-3}		η/mPa·s		u/m·s^{-1}		κ_s/TPa^{-1}	
pure liquid	expt.	lit.	expt.	lit.	expt.	lit.	expt.	lit.
hexane	0.6549	0.65489[16] 0.65484[17]	0.313	0.3000[20] 0.2968[18]	1078.50	1078 [22] 1078.1[23]	1312.8	1314[22] 1318[17]
octane	0.6986	0.69862[17]	0.518	0.5151[17]	1170.88	1172.02[24]	1044.1	1042.5[17]
decane	0.7259	0.72635[17] 0.72614[18]	0.845	0.8406[18]	1235.44	1234.7[25]	902.6	904.82[17]
2-methyl-1-propanol	0.7982	0.7978[17,18] 0.7982[17,19]	3.404	3.4098[21] 3.3330[17]	1191.35	1191[19]	882.7	

For the measurement of speed of sound, a crystal controlled variable path ultrasonic interferometer MX-3 (supplied by Mittal Enterprises, New Delhi) was used with accuracy about ± 0.4 %. The principle used in the measurement of velocity (u) is based on the accurate determination of the wavelength (λ) in the medium. Ultrasonic waves of known frequency (f) are produced by a quartz plate fixed at the bottom of the cell. The waves are reflected by a movable metallic plate kept parallel to the quartz plate. If the separation between these two plates is exactly a whole multiple of the sound wavelength, standing waves are formed in the medium. The acoustic resonance gives rise to an electrical reaction on the generator driving the quartz plate, and the anode current of the generator becomes maximum. If the distance is now increased or decreased and the variation is exactly one-half wavelength ($\lambda/2$) or a multiple of it, the anode current again becomes maximum. From the knowledge of wavelength, the velocity can be obtained by the relation $u = \lambda \times f$.

Results and Discussion

The experimental densities, viscosities, speed of sound, excess molar volumes, deviation in viscosity, deviation in speed of sound, isentropic compressibility, and deviation in isentropic compressibilities for the different binary mixtures at 298.15 K are summarized in Table 2. The experimental values of density are used to calculate the excess molar volumes, V^E, of the mixtures as

$$V^E/\text{m}^3\cdot\text{mol}^{-1} = (M_1x_1 + M_2x_2)/\rho_m - (M_1x_1/\rho_1 + M_2x_2/\rho_2) \quad (1)$$

where M_i, x_i, and ρ_i ($i = 1, 2$) are the molecular weights, the mole fractions, and the densities of pure component, respectively. ρ_m is the density of the mixture. The reproducibility in V^E values due to estimated errors in density (0.0002×10^3 kg·m^{-3}) is about $\pm 3 \times 10^6$ m^3·mol^{-1}. The viscosity deviations, $\Delta\eta$, were calculated using

$$\Delta\eta/\text{mPa}\cdot\text{s} = \eta_m - (x_1\eta_1 + x_2\eta_2) \quad (2)$$

where η_m is the viscosity of the mixture and η_i represents that for the ith component. Deviation in speed of sound, Δu, was calculated by

$$\Delta u/\text{m}\cdot\text{s}^{-1} = u_m - (x_1u_1 + x_2u_2) \quad (3)$$

where u_m is the speed of sound of the mixture. The isentropic compressibility, κ_s, values can be estimated from

$$\kappa_s = (\rho u^2)^{-1} \quad (4)$$

Defining the ideal isentropic solution property, κ_s^{id}, in terms of the volume fraction average

$$\kappa_s^{id} = \phi_1\kappa_{s,1} + \phi_2\kappa_{s,2} \quad (5)$$

where $\kappa_{s,i}$ is the isentropic compressibility of pure component i and $\phi_i = x_iV_i/\sum x_iV_i$ is the volume fraction of component i. The deviation in isentropic compressibility, $\Delta\kappa_s$, from the ideal mixing values can be estimated from the following equation:

$$\Delta\kappa_s = \kappa_s^{mix} - (\phi_1\kappa_{s,1} + \phi_2\kappa_{s,2}) \quad (6)$$

The composition dependence of V^E, $\Delta\eta$, and Δu was represented by the Redlich–Kister equation:[15]

$$Y(V^E, \Delta\eta, \text{ or } \Delta u) = x_1x_2\sum_{i=1}^{k} A_i(2x_1 - 1)^{i-1} \quad (7)$$

where k is the number of estimated parameters and A_i is the polynomial coefficients, which were obtained by fitting the equation to the experimental results with least-squares regression method. The standard deviation, σ, for V^E, $\Delta\eta$, and Δu is defined as

$$\sigma = [\sum(Y_{exp} - Y_{cal})^2/(n - k)]^{1/2} \quad (8)$$

where n is the number of measurements. The $\Delta\kappa_s$ values were correlated with the composition data by means of the equation

$$\Delta\kappa_s = \phi_1\phi_2\sum_{i=1}^{k} A_i(2\phi_1 - 1)^{i-1} \quad (9)$$

and the standard deviations $\sigma(\Delta\kappa_s)$ may be given as

$$\sigma(\Delta\kappa_s) = [\sum(\Delta\kappa_{s(exp)} - \Delta\kappa_{s(cal)})^2/(n - k)]^{1/2} \quad (10)$$

For all the binary mixtures investigated, $\sigma(V^E) < 0.006 \times 10^6$ m^3·mol^{-1}, $\sigma(\Delta\eta) < 0.004$ mPa·s, $\sigma(\Delta u) < 0.5$ m·s^{-1}, and $\sigma(\Delta\kappa_s) < 0.6$ TPa^{-1}. The parameters of eqs 7 and 9 and standard deviations σ of eqs 8 and 10 are given in Table 3.

The V^E values fitted with eq 7 are plotted as a function of x_1 in Figure 1. A close examination of Figure 1 shows that binary mixtures of hexane, octane, and decane with 2-methyl-1-propanol exhibit positive V^E values for the whole range of composition increasing magnitude with the length of hydrocarbon chain. The maximum positive value is obtained for the $(CH_3)_2CHCH_2OH + C_{10}H_{22}$ mixture. The positive values of V^E can be visualized as being due to a closer approach of the unlike molecules having significantly different molecular sizes. Due to the presence of nonpolar molecules like alkanes, the existing H-bonding in alcohol molecules breaks and the system shows weak intermolecular interactions. The longer the chain length of the alkane molecules, the weaker will be the interaction between liquid components. The positive V^E values show that volume expansion is taking place when 2-methyl-1-propanol mixes with alkanes because the addition of alkanes, which are nonpolar, causes rupture of H-bonds in self-associated alcohols. The V^E values for the binary liquid mixtures of $(CH_3)_2CHCH_2OH + C_6H_{14}$ and $(CH_3)_2CHCH_2OH + C_8H_{18}$ have been

Table 2. Experimental Densities ρ, Viscosities η, Speeds of Sound u, Isentropic Compressibility κ_s, Excess Molar Volume V^E, Viscosity Deviation $\Delta\eta$, Deviation in Speeds of Sound Δu, and Deviation in Isentropic Compressibility $\Delta\kappa_s$ for the Binary Mixtures at 298.15 K

x_1	$\rho \times 10^{-3}$/kg·m^{-3}	η/mPa·s	u/m·s^{-1}	κ_s/TPa^{-1}	$V^E \times 10^6$/m^3·mol^{-1}	$\Delta\eta$/mPa·s	Δu/m·s^{-1}	$\Delta\kappa_s$/TPa^{-1}
$(CH_3)_2CHCH_2OH$ (1) + C_6H_{14} (2)								
0.0000	0.6549	0.313	1078.50	1312.8	0.000	0.000	0.0	0.0
0.0554	0.6601	0.323	1077.54	1304.7	0.097	−0.161	−7.2	9.0
0.1040	0.6649	0.337	1077.48	1295.5	0.163	−0.297	−12.8	15.3
0.2000	0.6751	0.373	1080.12	1269.7	0.237	−0.558	−21.0	21.4
0.3274	0.6901	0.462	1086.69	1227.1	0.248	−0.862	−28.8	24.3
0.4173	0.7016	0.547	1094.14	1190.3	0.232	−1.056	−31.4	21.9
0.4666	0.7083	0.606	1099.20	1168.5	0.208	−1.149	−32.0	19.9
0.5057	0.7138	0.659	1103.48	1150.5	0.185	−1.217	−32.1	18.0
0.5589	0.7215	0.748	1110.04	1124.8	0.160	−1.293	−31.5	15.1
0.6090	0.7290	0.859	1116.92	1099.6	0.138	−1.336	−30.3	12.0
0.7073	0.7446	1.162	1132.47	1047.9	0.088	−1.337	-−25.8	6.2
0.8051	0.7612	1.627	1150.02	993.3	0.053	−1.175	−19.3	0.8
0.8610	0.7712	2.004	1161.26	961.6	0.041	−0.970	−14.4	−1.2
0.9149	0.7813	2.454	1172.51	930.9	0.026	−0.687	−9.2	−1.9
0.9376	0.7857	2.670	1177.52	917.9	0.018	−0.541	−6.8	−1.9
1.0000	0.7982	3.404	1191.35	882.7	0.000	0.000	0.0	0.0
$(CH_3)_2CHCH_2OH$ (1) + C_8H_{18} (2)								
0.0000	0.6986	0.518	1170.88	1044.1	0.000	0.000	0.0	0.0
0.0589	0.7015	0.527	1168.32	1044.4	0.118	−0.161	−3.8	5.8
0.1165	0.7045	0.535	1166.53	1043.1	0.229	−0.319	−6.7	10.2
0.2050	0.7098	0.576	1164.78	1038.4	0.320	−0.534	−10.3	14.9
0.2792	0.7147	0.617	1163.96	1032.8	0.374	−0.707	−12.6	17.8
0.3371	0.7189	0.663	1163.07	1028.3	0.392	−0.828	−14.7	20.4
0.4081	0.7246	0.719	1162.72	1020.8	0.377	−0.977	−16.5	22.1
0.4662	0.7296	0.788	1163.28	1012.9	0.362	−1.075	−17.1	22.3
0.5126	0.7339	0.850	1163.72	1006.2	0.337	−1.147	−17.7	22.4
0.6126	0.7441	1.028	1166.33	987.9	0.263	−1.258	−17.1	20.2
0.6615	0.7495	1.154	1168.40	977.3	0.233	−1.273	−16.0	18.1
0.7069	0.7549	1.300	1170.05	967.6	0.191	−1.258	−15.3	16.5
0.7855	0.7650	1.634	1173.55	949.2	0.121	−1.151	−13.4	14.1
0.8615	0.7758	2.102	1178.62	927.9	0.056	−0.902	−9.9	9.6
0.9523	0.7901	2.875	1187.04	898.2	0.003	−0.391	−3.3	2.4
1.0000	0.7982	3.404	1191.35	882.7	0.000	0.000	0.0	0.0
$(CH_3)_2CHCH_2OH$ (1) + $C_{10}H_{22}$ (2)								
0.0000	0.7259	0.845	1235.44	902.6	0.000	0.000	0.0	0.0
0.0728	0.7281	0.848	1228.67	909.8	0.102	−0.183	−3.6	7.9
0.1021	0.7290	0.859	1226.63	911.7	0.152	−0.247	−4.3	10.1
0.2032	0.7326	0.890	1219.92	917.2	0.261	−0.475	−6.6	16.7
0.2976	0.7364	0.934	1215.18	919.6	0.356	−0.673	−7.1	20.3
0.3547	0.7390	0.976	1212.24	920.8	0.396	−0.777	−7.6	22.3
0.4171	0.7421	1.023	1209.44	921.2	0.434	−0.889	−7.6	23.6
0.5016	0.7470	1.119	1206.13	920.2	0.433	−1.010	−7.2	24.0
0.6084	0.7542	1.297	1201.81	918.0	0.416	−1.105	−6.8	23.8
0.6575	0.7580	1.415	1200.16	915.9	0.395	−1.113	−6.3	22.8
0.7027	0.7619	1.543	1198.37	913.9	0.356	−1.100	−6.1	21.8
0.8004	0.7716	1.942	1195.10	907.4	0.245	−0.951	−5.1	17.8
0.8952	0.7829	2.503	1192.92	897.6	0.129	−0.633	−3.1	10.9
0.9497	0.7905	2.928	1191.98	890.3	0.053	−0.347	−1.6	5.6
0.9848	0.7958	3.249	1191.78	884.7	0.014	−0.116	−0.2	1.4
1.0000	0.7982	3.404	1191.35	882.7	0.000	0.000	0.0	0.0

Table 3. Estimated Parameters of Equations 7 and 10 and Standard Deviations σ for Excess Properties of the Binary Mixtures

binary mixture	excess property	A_0	A_1	A_2	A_3	A_4	σ
$(CH_3)_2CHCH_2OH$ (1) + C_6H_{14} (2)	$V^E \times 10^6$/m^3·mol^{-1}	0.762	−0.990	0.423	0.153		0.003
	$\Delta\eta$/mPa·s	−4.830	−3.176	−1.699	−0.352		0.004
	Δu/m·s^{-1}	−128.4	5.1	1.9	8.8		0.1
	$\Delta\kappa_s$/TPa^{-1}	54.0	−116.3	42.2	−33.5	18.7	0.3
$(CH_3)_2CHCH_2OH$ (1) + C_8H_{18} (2)	$V^E \times 10^6$/m^3·mol^{-1}	1.373	−1.077	0.082	−0.122	−0.441	0.006
	$\Delta\eta$/mPa·s	−4.522	−3.028	−1.776	−0.115	0.352	0.003
	Δu/m·s^{-1}	−69.2	−11.8	−5.2			0.4
	$\Delta\kappa_s$/TPa^{-1}	78.1	−53.5	23.5	−15.1		0.7
$(CH_3)_2CHCH_2OH$ (1) + $C_{10}H_{22}$ (2)	$V^E \times 10^6$/m^3·mol^{-1}	1.774	0.048	−0.490	−0.331		0.006
	$\Delta\eta$/mPa·s	−4.040	−2.512	−1.126	−0.056		0.003
	Δu/m·s^{-1}	−29.1	6.4	−18.4	5.2		0.2
	$\Delta\kappa_s$/TPa^{-1}	90.0	−38.6	23.4	−54.5	35.5	0.3

compared with the literature.[29,30] For the binary mixture of $(CH_3)_2CHCH_2OH + C_{10}H_{22}$, no V^E data could be found for comparison.

The variation of the viscosity deviations $\Delta\eta$ with the mole fraction of the first component for the binary mixture is presented in Figure 2. The $\Delta\eta$ values are negative for all three binary mixtures over the whole composition range. It shows a decreasing trend with an increase in chain length of alkane molecules from C_6 to C_{10}. The $\Delta\eta - x_1$ curves show maximum negative values at $x_1 \approx 0.65$. The viscosity of pure 2-methyl-

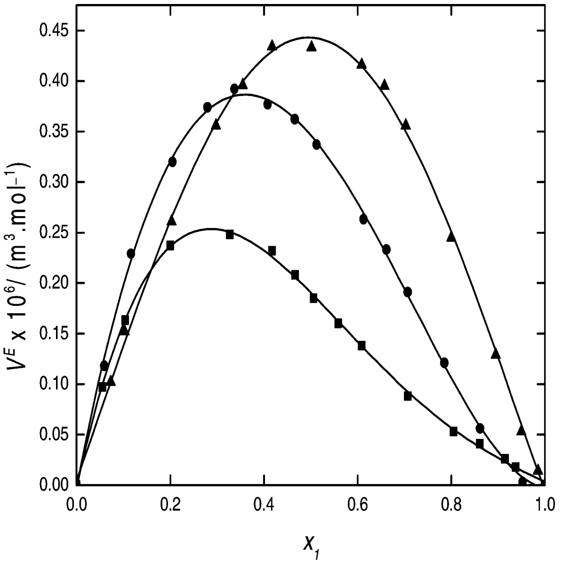

Figure 1. Excess molar volumes V^E at 298.15 K for binary mixtures of 2-methyl-1-propanol (1) with ■, *n*-hexane; ●, *n*-octane; ▲, *n*-decane; —, for Redlich−Kister eq 7.

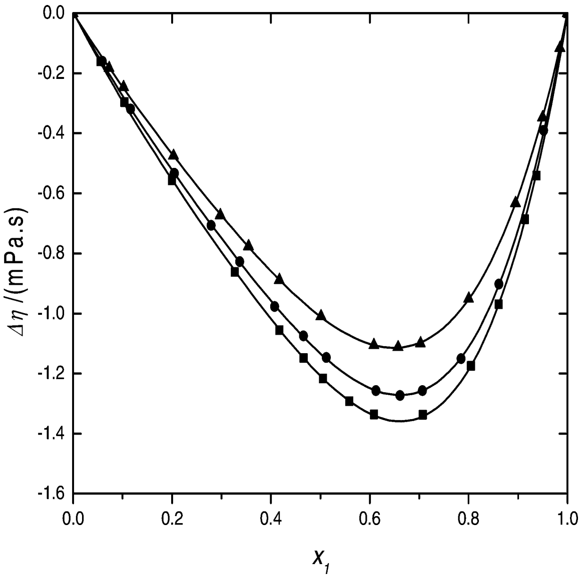

Figure 2. Viscosity deviations $\Delta\eta$ at 298.15 K for binary mixtures of 2-methyl-1-propanol (1) with ■, *n*-hexane; ●, *n*-octane; ▲, *n*-decane; —, for Redlich−Kister eq 7.

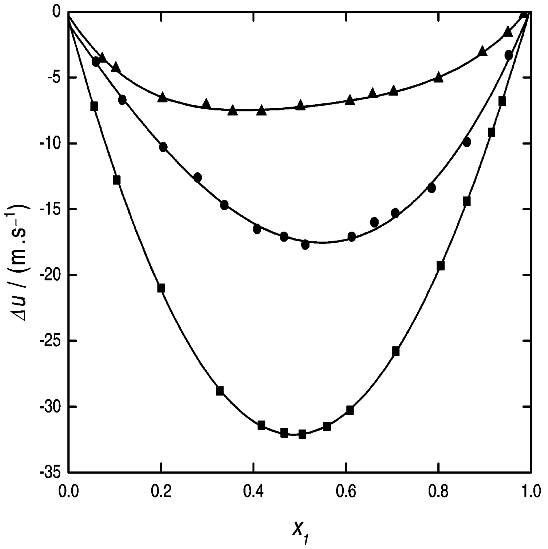

Figure 3. Deviation in speed of sound Δu at 298.15 K for binary mixtures of 2-methyl-1-propanol (1) with ■, *n*-hexane; ●, *n*-octane; ▲, *n*-decane; —, for Redlich−Kister eq 7.

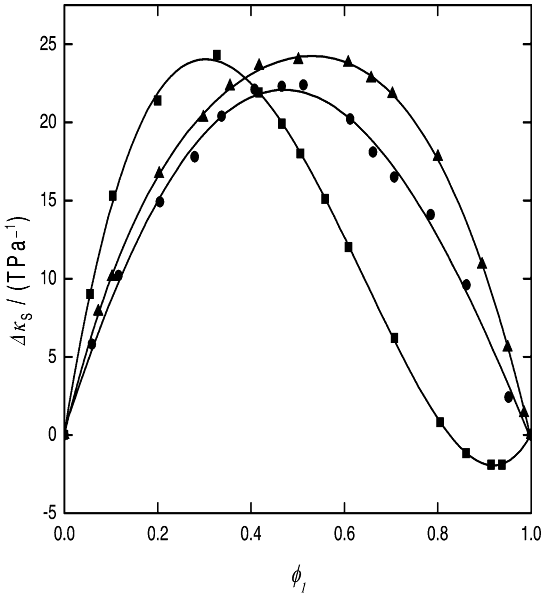

Figure 4. Deviation in isentropic compressibility $\Delta\kappa_S$ at 298.15 K for binary mixtures of 2-methyl-1-propanol (1) with ■, *n*-hexane; ●, *n*-octane; ▲, *n*-decane; —, for Redlich−Kister eq 9.

1-propanol is found to be about an order of magnitude larger (≈ 3.404 mPa•s) than that of decane (≈ 0.845 mPa•s). The negative values of viscosity deviations further support the presence of dispersion forces between the mixing components in these binary mixtures.

The deviation in speed of sound on mixing Δu for all three binary liquid mixtures at 298.15 K is shown in Figure 3. The Δu values for all the three systems studied are negative for the entire composition range. The Δu values show a decreasing trend with increasing chain length of hydrocarbons like $\Delta\eta$ values. The negative Δu values show the importance of interstitial accommodation of alcohol molecules between the alkane molecules. Thus as the size of alkane molecules increases, the molecules of the mixing component cannot be accommodated easily.

The deviation in isentropic compressibility $\Delta\kappa_S$ shows a positive trend for $(CH_3)_2CHCH_2OH + C_8H_{18}$ and $(CH_3)_2CHCH_2OH + C_{10}H_{22}$ systems over the entire composition range. For the system $(CH_3)_2CHCH_2OH + C_6H_{14}$, $\Delta\kappa_S$ is positive, but it becomes negative at higher x_1 values of 2-methyl-1-propanol as shown in Figure 4. The $\Delta\kappa_S$ values can be interpreted in terms of (i) an increase in free volume in the mixture as compared to those in pure components due to rupture of alcohol aggregates with the addition of alkanes and (ii) interstitial accommodation of alkane molecules in the aggregates of alcohols.

The positive $\Delta\kappa_S$ values for $(CH_3)_2CHCH_2OH + C_8H_{18}$ and $(CH_3)_2CHCH_2OH + C_{10}H_{22}$ show that the first factor predominates. The positive values of $\Delta\kappa_S$ also suggest that a mixture is more compressible than the corresponding ideal mixture. In these binary mixtures, an expansion in free volume makes the mixture more compressible than ideal mixtures. Negative $\Delta\kappa_S$ means that the mixture is less compressible than the ideal mixture.

Conclusions

In this paper, an attempt is made to measure densities, viscosities, and speed of sound at 298.15 K over the entire range of mixture composition of 2-methyl-1-propanol with hexane, octane, and decane. Out of these measured data, the excess molar volume, deviations in viscosity, speed of sound, and isentropic compressibility have been calculated and correlated by a Redlich−Kister type polynomial equation to derive the coefficients and standard errors.

Positive deviations are observed in the case of V^E whereas $\Delta\eta$ and Δu values show negative deviations for all binary mixtures. Both positive and negative deviations are observed in the case of $\Delta\kappa_S$. On the whole, it can be concluded that the strength of bonding is expected to decrease with an increase in the chain length of alkanes. The present results of V^E, $\Delta\eta$, or Δu and $\Delta\kappa_S$ corroborate this fact.

Acknowledgment

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