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Densities, Viscosities, and Ultrasonic Velocity Studies of Binary Mixtures of Chloroform with Octan-1-ol and Decan-1-ol at (303.15 and 313.15) K

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Densities, viscosities, and ultrasonic velocities of binary mixtures of chloroform with octan-1-ol and decan-1-ol have been measured over the entire range of composition at (303.15 and 313.15) K and at atmospheric pressure. From the experimental values of density, viscosity, and ultrasonic velocity, the excess molar volumes V^E, deviations in viscosity $\Delta\eta$, and excess isentropic compressibility κ_s^E have been calculated. The excess molar volumes and excess isentropic compressibility are positive for both the binaries studied over the whole composition, while deviations in viscosities are negative for both the binary mixtures. The excess molar volumes, deviations in viscosity, and deviations in isentropic compressibility have been fitted to the Redlich−Kister polynomial equation. The very recently proposed Jouyban−Acree model is used to correlate the experimental values of density, viscosity, and ultrasonic velocity at different temperatures.

Introduction

Studies on thermodynamic and transport properties of binary liquid mixtures provide information on the nature of interactions in the constituent binaries. Literature provides extensive data on the density and viscosity of liquid mixtures, but a combined study of density, viscosity, and ultrasonic velocity is quite scarce. The effects of molecular size, shape, chain length, and degree of molecular association of normal alkanols and branched alkanols, on the volumetric, viscometric, and acoustic properties of binary mixtures containing acetonitrile, dimethyl sulfoxide, ethyl acetate, and benzonitrile have been reported earlier.[1−5] Alkanols are self-associated, and there is a decrease in the self-association when they are mixed with chloroform as a result of hydrogen bonded interaction between hydroxyl oxygen of alkanols and chloroform and due to the presence of a specific interaction between hydroxyl oxygen of alkanols with the chlorine of chloroform. In continuation with our earlier studies of binary mixtures of chloroform with alkanols,[6,7] we now report density, viscosity, and ultrasonic velocity data for the binary mixtures of chloroform with octan-1-ol and decan-1-ol at (303.15 and 313.15) K.

Experimental Section

Octan-1-ol (E. Merck, purity 99.5 %) and decan-1-ol (s.d. fine chem., purity 99 %) were used after single distillation. Chloroform (s.d. fine chem., purity 99 %) was shaken several times with distilled water to remove ethanol, dried over anhydrous calcium chloride, and then fractionally distilled. The purity of the solvents, after purification, was ascertained by comparing their densities, viscosities, and ultrasonic velocities with the corresponding literature values at 298.15 K (Table 1). Binary mixtures were prepared by mass in airtight stoppered glass bottles. The masses were recorded on an Adairdutt balance to an accuracy of $\pm 1 \times 10^{-4}$ g. Care was taken to avoid evaporation and contamination during mixing. The estimated uncertainty in mole fraction was $< 1 \times 10^{-4}$.

Table 1. Comparison of Experimental Density, Viscosity, and Ultrasonic Velocity of Pure Liquids with Literature Values at 303.15 K

	ρ		η		u	
	$g \cdot cm^{-3}$		mPa·s		$m \cdot s^{-1}$	
pure liquid	exptl	lit	exptl	lit	exptl	lit
chloroform	1.46921	1.47060[a] 1.4692[b]	0.534	0.514[a] 0.534[b]	969	968[c]
octan-1-ol	0.81820	0.81834[d]	6.446	6.298[e]	1335	1339[d]
decan-1-ol	0.82269	0.82292[d]	9.715	-	1370	1366[d]

[a] Reference 13. [b] Reference 22. [c] Reference 23. [d] Reference 24. [e] Reference 25.

Densities were determined by using a 15 cm^3 bicapillary pycnometer as described earlier.[8,9] The pycnometer was calibrated using conductivity water with 0.99705 $g \cdot cm^{-3}$ as its density[10] at 298.15 K. The pycnometer filled with air bubble free experimental liquids was kept in a transparent walled water bath (maintained constant to ±0.01 K) for (10 to 15) min to attain thermal equilibrium. The positions of the liquid levels in the two arms were recorded with the help of a travelling microscope, which could read to 0.01 mm. The estimated uncertainty of density measurements of solvent and binary mixtures was 0.00005 $g \cdot cm^{-3}$. At least three to four measurements were made which had an average deviation of ± 0.00005 $g \cdot cm^{-3}$.

The dynamic viscosities were measured using an Ubbelohde suspended level viscometer,[9] calibrated with conductivity water. An electronic digital stop watch with a readability of ±0.01 s was used for the flow time measurements. At least three repetitions of each data reproducible to ±0.05 s were obtained, and the results were averaged. Since all flow times were greater than 200 s and capillary radius (0.5 mm) was far less than its length (50 to 60) mm, the kinetic energy and end corrections, respectively, were found to be negligible. The uncertainties in dynamic viscosities are of the order of ±0.003 mPa·s.

The ultrasonic velocities (u) were measured at a frequency of 2 MHz in these solutions through interferometric method (using Mittal's F-81 model) at (303.15 and 313.15) K (±0.05

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Table 2. Density ρ, Viscosity η, Ultrasonic Velocity u, Isentropic Compressibility κ_s, Excess Molar Volume V^E, Deviation in Viscosity $\Delta\eta$, and Excess Isentropic Compressibility κ_s^E for Chloroform (1) + Alkanols (2) at (303.15 and 313.15) K

x_1	ρ / g·cm^{-3}	V^E / cm^3·mol^{-1}	η / mPa·s	$\Delta\eta$ / mPa·s	u / m·s^{-1}	κ_s / TPa^{-1}	κ_s^E / TPa^{-1}
Chloroform (1)+ Octan-1-ol (2) 303.15 K							
0.0000	0.81820	0.000	6.446	0.000	1335	686	0
0.0471	0.83414	0.015	5.924	−0.244	1320	688	−1
0.0996	0.85281	0.033	5.360	−0.497	1303	691	−1
0.1490	0.87131	0.052	4.852	−0.713	1286	694	−2
0.1997	0.89124	0.084	4.346	−0.919	1267	699	0
0.2493	0.91182	0.115	3.898	−1.074	1247	705	3
0.3001	0.93412	0.144	3.475	−1.197	1227	711	5
0.3529	0.95870	0.175	3.039	−1.321	1207	716	7
0.4037	0.98384	0.204	2.666	−1.393	1186	723	10
0.4548	1.01069	0.238	2.332	−1.425	1164	730	14
0.5001	1.03604	0.260	2.062	−1.427	1147	734	15
0.5550	1.06885	0.282	1.772	−1.393	1125	739	16
0.5995	1.09736	0.290	1.552	−1.350	1107	744	19
0.6467	1.12961	0.295	1.370	−1.253	1088	748	20
0.7001	1.16888	0.295	1.171	−1.136	1069	749	18
0.7484	1.20734	0.282	1.036	−0.985	1051	750	17
0.7997	1.25163	0.256	0.929	−0.789	1033	749	15
0.8451	1.29414	0.222	0.804	−0.646	1017	747	13
0.9007	1.35120	0.157	0.693	−0.428	999	742	9
0.9494	1.40628	0.084	0.616	−0.217	984	734	4
1.0000	1.46921	0.000	0.534	0.000	969	725	0
Chloroform (1)+ Octan-1-ol (2) 313.15 K							
0.0000	0.81109	0.000	4.604	0.000	1303	726	0
0.0471	0.82677	0.021	4.259	−0.151	1287	730	0
0.0996	0.84516	0.039	3.883	−0.311	1271	732	−2
0.1490	0.86332	0.068	3.545	−0.446	1252	739	2
0.1997	0.88291	0.105	3.212	−0.571	1233	745	3
0.2493	0.90314	0.139	2.902	−0.677	1214	751	5
0.3001	0.92495	0.187	2.593	−0.777	1193	760	10
0.3529	0.94903	0.230	2.315	−0.838	1172	767	13
0.4037	0.97363	0.272	2.070	−0.874	1150	777	18
0.4548	0.99995	0.313	1.823	−0.910	1130	783	20
0.5001	1.02478	0.340	1.648	−0.899	1111	791	24
0.5550	1.05690	0.369	1.426	−0.895	1089	798	26
0.5995	1.08474	0.389	1.273	−0.865	1071	804	28
0.6467	1.11627	0.400	1.134	−0.810	1053	808	28
0.7001	1.15472	0.400	0.987	−0.737	1033	812	28
0.7484	1.19238	0.384	0.883	−0.643	1015	814	27
0.7997	1.23577	0.350	0.769	−0.546	996	816	25
0.8451	1.27750	0.302	0.704	−0.424	980	815	22
0.9007	1.33355	0.216	0.627	−0.272	962	810	16
0.9494	1.38758	0.127	0.555	−0.144	948	802	8
1.0000	1.44974	0.000	0.491	0.000	934	791	0

x_1	ρ / g·cm^{-3}	V^E / cm^3·mol^{-1}	η / mPa·s	$\Delta\eta$ / mPa·s	u / m·s^{-1}	κ_s / TPa^{-1}	κ_s^E / TPa^{-1}
Chloroform (1)+ Decan-1-ol (2) 303.15 K							
0.0000	0.82269	0.000	9.715	0.000	1370	648	0
0.0523	0.83735	0.015	8.774	−0.461	1354	651	−1
0.1025	0.85218	0.055	7.938	−0.836	1338	655	0
0.1519	0.86773	0.086	7.108	−1.212	1322	659	0
0.1992	0.88356	0.115	6.311	−1.575	1305	665	2
0.2501	0.90166	0.155	5.650	−1.769	1284	673	6
0.2992	0.92032	0.197	5.021	−1.947	1264	680	9
0.3497	0.94092	0.233	4.374	−2.130	1244	687	12
0.4005	0.96319	0.272	3.838	−2.200	1222	695	15
0.4510	0.98710	0.306	3.293	−2.281	1201	702	17
0.5000	1.01224	0.327	2.873	−2.252	1180	710	20
0.5504	1.04028	0.347	2.439	−2.223	1158	717	22
0.5991	1.06975	0.364	2.098	−2.117	1137	723	24
0.6496	1.10328	0.362	1.778	−1.973	1116	728	23
0.6990	1.13934	0.350	1.501	−1.796	1096	731	21
0.7508	1.18121	0.322	1.277	−1.545	1074	734	19
0.7990	1.22443	0.291	1.052	−1.327	1054	735	16
0.8513	1.27702	0.232	0.916	−0.983	1033	734	11
0.8999	1.33189	0.176	0.784	−0.669	1013	732	6
0.9501	1.39624	0.091	0.651	−0.341	992	728	1
1.0000	1.46921	0.000	0.534	0.000	969	725	0
Chloroform (1)+ Decan-1-ol (2) 313.15 K							
0.0000	0.81610	0.000	6.819	0.000	1334	689	0
0.0523	0.83045	0.036	6.174	−0.314	1318	693	0
0.1025	0.84507	0.072	5.590	−0.580	1301	699	2
0.1519	0.86035	0.109	5.068	−0.790	1283	706	5
0.1992	0.87581	0.161	4.603	−0.955	1265	714	8
0.2501	0.89354	0.215	4.121	−1.115	1245	722	12
0.2992	0.91184	0.263	3.726	−1.200	1226	730	15
0.3497	0.93202	0.309	3.291	−1.315	1205	739	19
0.4005	0.95390	0.346	2.935	−1.350	1184	748	22
0.4510	0.97726	0.396	2.571	−1.394	1163	757	26
0.5000	1.00185	0.427	2.259	−1.396	1142	765	28
0.5504	1.02931	0.450	1.974	−1.362	1121	773	30
0.5991	1.05822	0.462	1.697	−1.331	1100	781	32
0.6496	1.09100	0.465	1.480	−1.228	1079	787	31
0.6990	1.12633	0.448	1.251	−1.145	1059	792	30
0.7508	1.16724	0.424	1.076	−0.992	1037	797	28
0.7990	1.20966	0.375	0.911	−0.852	1017	799	24
0.8513	1.26095	0.321	0.770	−0.662	995	801	20
0.8999	1.31472	0.247	0.661	−0.463	975	800	14
0.9501	1.37793	0.131	0.561	−0.246	954	797	7
1.0000	1.44974	0.000	0.491	0.000	934	791	0

K). The uncertainty in velocity measurements is ±0.1 %. The other experimental details are the same as those reported earlier.[2−4]

Results and Discussion

Experimental values of densities ρ, viscosities η, and ultrasonic velocities u of mixtures at (303.15 and 313.15) K are listed as a function of mole fraction in Table 2. The density values have been used to calculate excess molar volumes V^E using the following equation

$$V^E/\mathrm{cm^3{\cdot}mol^{-1}} = (x_1M_1 + x_2M_2)/\rho_{12} - (x_1M_1/\rho_1) - (x_2M_2/\rho_2) \tag{1}$$

where ρ_{12} is the density of the mixture and x_1, M_1, ρ_1, and x_2, M_2, ρ_2 are the mole fraction, the molecular weight, and the density of pure components 1 and 2, respectively.

The viscosity deviations $\Delta\eta$ were calculated using

$$\Delta\eta/\mathrm{mPa{\cdot}s} = \eta_{12} - x_1\eta_1 - x_2\eta_2 \tag{2}$$

where η_{12} is the viscosity of the mixture and x_1, x_2 and η_1, η_2 are the mole fraction and the viscosity of pure components 1 and 2, respectively.

The excess isentropic compressibility, κ_s^E, was obtained using the relation

$$\kappa_s^E = \kappa_s - \kappa_s^{id} \tag{3}$$

where κ_s is the isentropic compressibility and was calculated using the Laplace relation, that is, $\kappa_s = (1/u^2\rho)$ and κ_s^{id} was calculated from the relation[11,12]

$$\kappa_s^{id} = \sum \phi_i[\kappa_{s,i} + TV^{\circ}_i(\alpha^{\circ\,2}_i)/C_{p,i}] - [T(\sum x_iV^{\circ}_i)(\sum \phi_i\alpha^{\circ}_i)^2/\sum x_iC_{p,i}] \tag{4}$$

where ϕ_i is the volume fraction of the component i in the mixture stated, T is the temperature, and κ_{si}, V°_i, α°_i, and $C_{p,i}$, are the isentropic compressibility, molar volume, coefficient of thermal expansion, and molar heat capacity, respectively, for pure component i. The values required were taken from the literature.[13,14]

Table 3. Parameters and Standard Deviations σ of Equations 5 and 6 for Chloroform + Octan-1-ol and Chloroform + Decan-1-ol

	T/K	a_0	a_1	a_2	a_3	σ
		Chloroform + Octan-1-ol				
V^E/(cm^3·mol^{-1})	303.15	1.039	0.956	0.026	−0.176	0.003
	313.15	1.358	1.309	0.184	−0.115	0.003
$\Delta\eta$/(mPa·s)	303.15	−5.709	0.522	0.898	−0.013	0.009
	313.15	−3.658	0.180	0.583	0.059	0.007
$\Delta\kappa_s$/(TPa^{-1})	303.15	60.815	91.046	−33.990	−36.103	0.744
	313.15	92.863	109.32	−13.873		0.821
		Chloroform + Decan-1-ol				
V^E/(cm^3·mol^{-1})	303.15	1.330	0.897	−0.196	−0.029	0.004
	313.15	1.689	1.133	0.058	0.054	0.005
$\Delta\eta$/(mPa·s)	303.15	−9.110	1.145	1.063	-	0.025
	313.15	−5.564	0.504	−0.244	0.237	0.010
$\Delta\kappa_s$/(TPa^{-1})	303.15	85.255	96.575	−91.897	−86.280	0.994
	313.15	114.90	88.813	−44.385	−8.144	0.526

The excess molar volumes and deviations in viscosity and isentropic compressibility were fitted to the Redlich−Kister[15] equation of the type

$$Y = x_1 x_2 \sum_i^n a_i (x_1 - x_2)^i \tag{5}$$

where Y is V^E, or $\Delta\eta$, or κ_s^E and n is the degree of the polynomial. Coefficients a_i were obtained by fitting eq 5 to experimental results using a least-squares regression method. In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation σ.

σ was calculated using the relation

$$\sigma(Y) = \left[\frac{\sum (Y_{\text{expt}} - Y_{\text{calc}})^2}{N - n}\right]^{1/2} \tag{6}$$

where N is the number of data points and n is the number of coefficients. The calculated values of the coefficients a_i along with the standard deviations σ are given in Table 3.

The variation of V^E with the mole fraction x_1 of chloroform for octan-1-ol and decan-1-ol at 303.15 K is represented in Figure 1. The observed positive excess molar volumes in the present investigation may be discussed in terms of several effects.[16] These may be divided into three types: chemical, physical, and structural. The physical interactions mainly involve the dispersion forces giving a positive contribution. The specific interactions such as the formation of hydrogen bonds, the formation of charge-transfer complexes, and the dipole−dipole interactions between the components result in volume decrease. Changes in the associative equilibria of alkan-1-ol molecules may lead to contributions of all these types. The disruption of alkanol aggregates through the breaking of hydrogen bonds makes V^E positive, since aggregates have smaller volumes than the sum of their components. As the chain length in alkanols increases, the proton-donating ability and the strength and extent of H-bonds decreases. Thus, the mixtures of chloroform with alkanols show the trend octan-1-ol < decan-1ol.

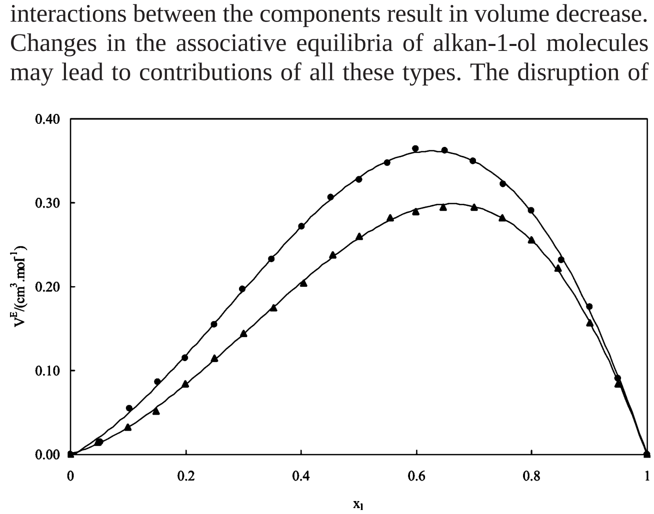

Figure 1. Excess molar volumes V^E at 303.15 K for x_1 chloroform + (1 − x_1) alkanols: ▲, octan-1-ol; ●, decan-1-ol.

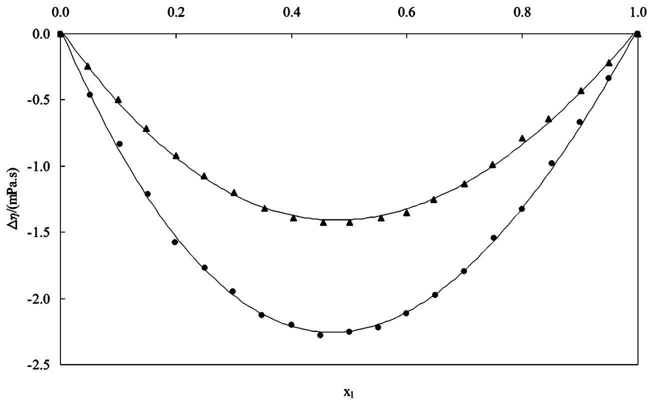

Figure 2. Deviations in viscosity ($\Delta\eta$) at 303.15 K for x_1 chloroform + (1 − x_1) alkanols: ▲, octan-1-ol; ●, decan-1-ol.

Table 4. Interaction Parameters and Standard Deviations σ for Chloroform + Octan-1-ol and Chloroform + Decan-1-ol

system	T/K	G_{12}	100 σ
chloroform + octan-1-ol	303.15	0.4241	3.13
	313.15	0.3281	2.96
chloroform + decan-1-ol	303.15	0.8967	1.70
	313.15	0.7615	2.04

Figure 2 depicts the variation of $\Delta\eta$ with the mole fraction x_1 of chloroform. $\Delta\eta$ values are negative in all systems and become more negative with an increase in chain length of alkanols, suggesting a decrease in hetero association of molecules with an increase in molar mass of alkanols.

The viscosities of the binary mixtures of chloroform with alkanols have been correlated with the help of the Grunberg and Nissan[17] viscosity model.

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12} \tag{7}$$

where G_{12} is an interaction parameter which is a function of the components 1 and 2 as well as temperature.

The correlating ability of eq 7 was tested by calculating the percentage standard deviation σ between the experimental and calculated viscosity as

$$\sigma = \sum [(\eta_{\text{exptl}} - \eta_{\text{calcd}})/\nu_{\text{exptl}})^2/n - m)]^{1/2} \tag{8}$$

where n represents the number of experimental points and m represents the number of coefficients. Table 4 includes the different parameters for the Grunberg−Nissan and percentage standard deviations. From Table 4, it is seen that the values of G_{12} are positive for both the binary mixtures of CHCl$_3$ with octan-1-ol and decan-1-ol.

The variation of κ_s^E with the mole fraction of chloroform, x_1, is represented in Figure 3. Kiyohara and Benson[18] have suggested that κ_s^E is the result of several opposing effects. A strong molecular interaction through charge-transfer, dipole induced dipole, and dipole−dipole[19] interactions, interstitial accommodation, and orientational ordering leads to a more compact structure, making κ_s^E negative, and breaks up the alkanol structures, making κ_s^E positive. The magnitudes of the various contributions depend mainly on the relative molecular size of the components. The positive values of κ_s^E for mixtures of

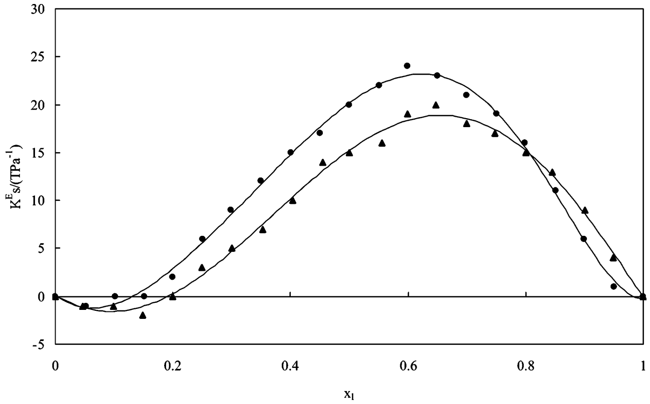

Figure 3. Excess isentropic compressibility (κ_s^E) at 303.15 K for x_1 chloroform + (1 − x_1) alkanols: ▲, octan-1-ol; ●, decan-1-ol.

Table 5. Parameters of Jouyban−Acree Model and Average Percentage Deviation for Densities, Viscosities, and Ultrasonic Velocities at (303.15 and 313.15) K

system	A_o	A_1	A_2	APD
	Density			
chloroform + octan-1-ol	47.590	−11.652	3.667	0.01
chloroform + decan-1-ol	46.169	−10.300	4.019	0.01
	Viscosity			
chloroform + octan-1-ol	109.608	−97.464	32.235	0.89
chloroform + decan-1-ol	264.994	−53.948	−49.933	1.01
	Ultrasonic Velocity			
chloroform + octan-1-ol	9.807	−20.437	−1.530	0.05
chloroform + decan-1-ol	28.653	−6.880	8.644	0.05

chloroform with octan-1-ol and decan-1-ol signify decreasing dipole−dipole interactions due to decreasing proton-donating abilities with increasing chain length of alkan-1-ols. Declustering of alkan-1-ols in the presence of chloroform may also lead to positive κ_s^E values.

Recently, Jouyban and Acree[20,21] proposed a model for correlating the density and viscosity of liquid mixtures at various temperatures. The proposed equation is

$$\ln y_{m,T} = f_1 \ln y_{1,T} + f_2 \ln y_{2,T} + f_1 f_2 \sum [A_j(f_1 - f_2)^j/T] \quad (9)$$

where $y_{m,T}$, $y_{1,T}$, and $y_{2,T}$ are density or viscosity of the mixture and solvents 1 and 2 at temperature T, respectively, $f_1$and f_2 are the volume fractions of solvents in the case of density and mole fraction in the case of viscosity, and A_j values are the model constants.

The correlating ability of the Jouyban−Acree model was tested by calculating the average percentage deviation (APD) between the experimental and calculated density and viscosity as

$$\text{APD} = (1/N) \sum [(|y_{\text{exptl}} - y_{\text{calcd}}|)/y_{\text{exptl}})] \quad (10)$$

where N is the number of data points in each set. The optimum numbers of constants A_j, in each case, were determined from the examination of the APD value.

Since the Jouyban−Acree model was not previously applied to ultrasonic velocity measurements, we extend the Jouyban−Acree model (eq 9) to ultrasonic velocity of the liquid mixtures with f as the mole fraction and again apply eq 10 for the correlating ability of the model.

The constants A_j calculated from the least-squares analysis are presented in Table 5 along with the APD. The proposed model provides reasonably accurate calculations for the density, viscosity, and ultrasonic velocity of binary liquid mixtures at various temperatures and the model could be used in data modeling.

Acknowledgment

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