

CHAPTER VI

Excess Molar Volumes, Viscosity Deviations and Ultrasonic Speeds of Sound of Binary Mixtures of 2-Butanone with Some Alkoxyethanols and Amines at 298.15 K*

6.1. Introduction

Properties such as density and viscosity of pure chemicals and of their binary mixtures over the whole composition range at a particular temperature or 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 non-thermodynamic ones of binary liquid mixtures are fundamental for understanding of interactions between molecules in these types of binary mixtures. So in recent years, there has been considerable interest in theoretical and experimental investigations of the excess thermodynamic properties of binary mixtures. The present chapter is a continuation of our systematic study¹⁻³ of the physicochemical properties of non-aqueous binary liquid mixtures and it reports density (ρ), viscosity (η) and ultrasonic speeds (u) for the binary mixtures of 2-butanone (BU) + alkoxyethanols (2-methoxy-, 2-ethoxy-, 2-butoxyethanol), and 2-butanone + amines (isopropyl-, cyclohexyl-, diethylamine) over the entire range of composition at 298.15 K and at atmospheric pressure.

It is well known that 2-butanone has numerous applications both in pure chemistry and industry. Also, alkoxyethanols have wide use as monomers in the production of polymers and emulsion formulations. They are also of considerable interest for studying the hetero-proximity effects of the etheric oxygen on the -O-H bond and, hence, its influence on the associated nature of the species in these molecules. Isopropylamine, cyclohexylamine and diethylamine are also important in characterizing the associated nature of the liquids in mixtures, because of the presence of both a proton donor and a proton acceptor and they form water insoluble compounds of medical importance.⁴

To explore the nature of the interactions various thermodynamic parameters, e.g., intermolecular free length, specific acoustic impedance etc. and their deviations have been derived from the density and ultrasonic speed data. The excess or deviation properties were fitted to Redlich-Kister polynomial equation to obtain their coefficients and standard deviations and have been interpreted in terms of molecular interactions and structural effects. This work provides a test of various empirical equations to correlate viscosity and acoustic data of binary mixtures in terms of pure component properties.

6.2. Experimental Section

6.2.1. Chemicals

2-methoxy-, 2-ethoxy- and 2-butoxyethanol (S. D. Fine Chemicals, AR, India) were purified as described in the literature.⁵ 2-butanone, isopropylamine, cyclohexylamine and diethylamine were procured from Merck, India and were used as purchased. The pure chemicals were stored over activated 4Å molecular sieves to reduce water content before use. The chemicals after purification were 99.8% pure and their purity was ascertained by GLC and also by comparing experimental values of densities and viscosities with those reported in the literature,^{2,5,7-8} when available, as presented in Table 1.

6.2.2. Measurements

Densities (ρ) were measured with an Ostwald -Sprengel type pycnometer having a bulb volume of about 25 cm³ and an internal diameter of the capillary of about 0.1 cm. The measurements were done in a thermostated bath controlled to ± 0.01 K. The viscosity was measured by means of a suspended Ubbelohde type viscometer, calibrated at 298.15 K with triply distilled water and purified methanol using density and viscosity values from the literature. The flow times were accurate to ± 0.1 s, and the uncertainty in the viscosity measurements, based on our work on several pure liquids, was $\pm 2 \times 10^{-4}$ mPa.s. The mixtures were prepared by mixing known volume of pure liquids in airtight-stopper bottles and each solution thus prepared was distributed into three recipients to perform all the measurements in triplicate, with the aim of the determining possible dispersion of the results obtained. Adequate precautions were taken to

minimize evaporation losses during the actual measurements. The reproducibility in mole fraction was within ± 0.0002 units. The mass measurements were done on a Mettler AG-285 electronic balance with a precision of $\pm 0.01\text{mg}$. The precision of density measurements was $\pm 3 \times 10^{-4} \text{ g cm}^{-3}$.

Ultrasonic speeds of sound (u) were determined by a multi-frequency ultrasonic interferometer (Mittal enterprise, New Delhi, M-81) working at 2 MHz, calibrated with triply distilled and purified water, methanol and benzene at 298.15 K. The precision of ultrasonic speed measurements was $\pm 0.2 \text{ m s}^{-1}$. The details of the methods and techniques had been described elsewhere.^{1,2}

6.3. Results and Discussion

The experimental values of density and viscosity for all the binary mixtures at 298.15 K were fitted to a polynomial equation⁹ of the following type:

$$y = \sum_{i=0}^3 a_i x_1^i \quad (1)$$

where y is a selected property of the mixture, a_i is an adjustable coefficient and x_1 is the mole fraction of 2-butanone in the mixtures. The values of the coefficients a_i are listed in Table 2. In Table 2, a_0 values are found to be equal/almost equal to the experimental value of a particular property for the second component in the mixtures at $x_1=0$. The experimental densities ρ , viscosities η , excess molar volumes V^E , and viscosity deviations $\Delta\eta$, for the binary mixtures studied at 298.15 K are reported in Table 3.

The excess molar volumes V^E , for the mixtures were calculated using the following equation,

$$V^E = \sum_{i=1}^2 x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right) \quad (2)$$

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where ρ is the density of the mixture and M_i , x_i , η_i are the molecular weight, mole fraction and viscosity of i^{th} component in the mixture, respectively. The estimated uncertainties for excess molar volumes V^E , is $\pm 0.005 \text{ cm}^3 \cdot \text{mol}^{-1}$.

Figure 1 illustrates that the excess molar volumes, V^E for all six systems under investigation are negative over the entire range of composition at the experimental temperature. The values of excess volumes, V^E for the six systems irrespective of their sign are in the order:



The negative values of excess molar volume, V^E suggest specific interactions^{1-3, 9} between the mixing components in the mixtures while its positive values suggest dominance of dispersion forces^{1-3, 9} between them. The negative V^E values indicate the specific interactions such as intermolecular hydrogen bonding between the mixing components and also the interstitial accommodation of the mixing components because of the difference in molar volume. Several effects may contribute to the value of V^E and three different effects may be considered as being important- a) disruption of liquid order on mixing and unfavorable interactions between unlike molecules producing a positive contribution to V^E , b) differences in molecular volumes and free volumes¹⁰ between liquid components and c) the possible association due to hydrogen bond interactions between the unlike molecules. The actual volume change would, therefore, depend on the relative strength of these opposing effects. The negative V^E values may also be due to the difference in the dielectric constants of the liquid components of the binary mixture.²

The very large negative values of V^E for the amines may be attributed to the presence of strong intermolecular hydrogen bond interactions between the amine molecules and the 2-butanone molecules and this effect decreases as one shifts from primary to secondary amines,^{11, 12} so diethylamine has less negative values of V^E , compared to those of isopropylamine and cyclohexylamine.

This is probably due to decreased proton donating ability of diethylamine, thereby decreasing hydrogen bond interaction (-C=O---H-N-) between the 2-butanone and diethylamine. Less negative values of V^E for cyclohexylamine, as compared to isopropylamine, may also be due to steric and other effects because of its cyclic nature.⁴ The alkoxyethanols undergo less self-association in contrast to the high tendency of amines to undergo self-association through intermolecular hydrogen bonding. But, the presence of etheric oxygen in alkoxyethanols facilitates the formation of intramolecular hydrogen bonding between the etheric oxygen and hydrogen of -OH group of the same molecule. Infrared, microwave and calorimetric studies¹³⁻¹⁵ have also shown the presence of mostly ten membered dimmers and linear associates in R-O-C₂H₅OH (where R = -CH₃, -C₂H₅, -C₄H₉, etc) and the small negative values of V^E for the alkoxyethanols under investigation may be attributed to the presence of weak hydrogen bond or dispersive forces^{1,2} between the mixing liquids. These effects further decreases or increases as the chain length of the alkoxyethanols increase. This may be due the increased +I effect of the alkyl group, thereby decreasing the polarity of the -O-H bond in the alkoxyethanol molecules.

Absolute viscosity η , and deviation viscosity $\Delta\eta$, for the mixtures were calculated using the following equations,

$$\eta = \nu\rho \quad (3)$$

$$\Delta\eta = \eta - \sum_{i=1}^2 x_i\eta_i \quad (4)$$

where ρ , ν are the density, kinematic viscosity of the mixtures and x_i , η_i are the mole fraction and viscosity of i^{th} component in the mixture, respectively. The estimated uncertainties for deviations viscosity $\Delta\eta$, is ± 0.004 mPa.s.

Table 3 shows that $\Delta\eta$ values are negative for all the mixtures, except for the mixture containing isopropylamine and cyclohexylamine, over the entire composition range at the experimental temperature. The negative $\Delta\eta$ values indicate the presence of weak interactions or dispersion forces between the unlike molecules in the mixture. The decrease of mixture viscosities indicates the weakening of self-association of alkoxyethanols in

presence of 2-butanone. According to Fort and Moore¹⁶ excess viscosities are negative in mixtures of components having unequal size and in which dispersion forces are present. As expected, the values of $\Delta\eta$ become more negative as the chain length of the alkoxyethanol⁵ molecules increases and one shifts from primary amine to secondary amine. This suggests that the strength of interaction in the mixtures is in the order: BU + BE < BU + EE < BU + ME < BU + DEA < BU + CHA < BU + IPA. Thus Figure 1 illustrating the variation of V^E complements with Figure 2 showing the variation of $\Delta\eta$, both plotted against x_1 of 2-butanone at 298.15 K, in describing the behaviors of the binary mixtures under investigation.

Isentropic compressibility, K_s and deviation in isentropic compressibility ΔK_s , were calculated using the following relations:

$$K_s = (u^2 \rho)^{-1} \quad (5)$$

$$\Delta K_s = K_s - \sum_{i=1}^j x_i K_{s,i} \quad (6)$$

where u and K_s are the speed of sound, isentropic compressibility of the mixture and $K_{s,i}$, the isentropic compressibility of i^{th} component in the mixture, respectively.

The experimental speed of sound, isentropic compressibility and deviation in isentropic compressibility are listed in Table 4 and are graphically represented in Figure 3 as a function of mole fraction of 2-butanone.

Figure 3 shows that ΔK_s values are negative for all the mixtures, except those containing 2-butoxyethanol and 2-ethoxyethanol and increase in the order: BU + IPA < BU + CHA < BU + DEA < BU + ME < BU + EE < BU + BE; this trend is justified by the presence of weak interaction or structure disruptive effects between the mixing liquids for the binary mixtures of alkoxyethanols and by the presence of strong hydrogen bond interactions between the mixing liquids for the binary mixtures of amines. As stated earlier alkoxyethanols in pure state associate predominantly to form ten-membered dimeric rings. These structures can thus resist the structure disruptions in presence of 2-butanone and this effect probably increases

with the number of carbon atoms in the alkoxyethanols. Similar type of results was reported earlier by Sastry et al.⁵

In an attempt to explore the nature of the interactions, various thermodynamic parameters intermolecular free length, L_f ²¹; specific acoustic impedance, Z ²²; Van der Waal's constant, b ²³; molecular radius, r ²¹; geometrical volume, B ; molar surface area, Y ²²; available volume, V_a ²¹; molar speed of sound, R_s ¹²; relative association, R_A ^{5, 12, 24} and molecular association, M_A ^{5, 12, 24} of the binary mixtures have been calculated using the following equations:

$$L_f = K \cdot K_S^{1/2} \quad (7)$$

$$Z = u\rho \quad (8)$$

$$b = (M/\rho) - (RT/\rho^2 u^2) \{ [1 + (Mu^2/3RT)]^{1/2} - 1 \} \quad (9)$$

$$r = (3b/16\pi N_A)^{1/3} \quad (10)$$

$$B = \frac{4}{3}\pi r^3 N_A \quad (11)$$

$$Y = [36\pi N_A B^2]^{1/3} \quad (12)$$

$$V_a = V(1 - u/u_\infty) \quad (13)$$

$$V_0 = V - V_a \quad (14)$$

$$R_s = \overline{M} u^{1/3} / \rho \quad (15)$$

$$R_A = (\rho_{mix} / \rho)(u / u_{mix})^{1/3} \quad (16)$$

$$M_A = \left[\left(\frac{u_{mix}}{\sum_{i=1}^2 x_i u_i} \right)^2 - 1 \right] \quad (17)$$

where K is a temperature dependent constant,²¹ T is the absolute temperature, R is the universal gas constant, N_A is the Avogadro's number, \overline{M} is the average molecular weight, V_0 is the volume at absolute zero, ρ_{mix} and u_{mix} are the density and speed of sound of the mixture, respectively and u_∞ is taken as 1600 ms⁻¹. These parameters are presented in Table 5 for the pure components and in Table 6 for the mixtures as a function of x_1 of 2-butanone, respectively. Figures 4-5 represent that the values of the specific

acoustic impedance Z , and intermolecular free length L_f , for the mixtures behave in an opposite manner and while molecular association M_A , and relative association R_A , values decrease for the mixtures containing alkoxyethanols, they increase for the amine mixtures with the mole fraction of 2-butanone. The relative association R_A , measures the ability of molecules to build associates or supramolecular structure by intermolecular interactions. The increase in chain length is expected to lower self-association, hence R_A values decreases nonlinearly from ME to BE as 2-butanone is added to these mixture. This implies the presence of dissociative interactions between the unlike molecules in the mixtures of alkoxyethanols, while the situation is different for the mixture of amines.^{5, 12, 24} Similarly the molecular association M_A , measures the degree of molecular association by intermolecular interactions and in Figure 5, the nonlinear variation of M_A values for the mixtures of different amines implies the formation of strong cross-association between the unlike molecules through hydrogen bonding in these mixtures. Thus it can be concluded that the non-ideality of the mixtures varies in the order: ^{12, 24}



Deviations in intermolecular free length ΔL_f , and specific acoustic impedance ΔZ , for the binary mixtures have been calculated using the following relations:

$$\Delta L_f = L_f - \sum_{i=1}^2 x_i L_{f,i} \quad (18)$$

$$\Delta Z = Z - \sum_{i=1}^2 x_i Z_i \quad (19)$$

where L_f and Z are the intermolecular free length and specific acoustic impedance of the mixture and x_i , $L_{f,i}$ and Z_i are the mole fraction, intermolecular free length and specific acoustic impedance of i^{th} component in the mixture, respectively.

Figure 6 reveals that ΔL_f are positive for the mixtures of EE and BE, while for the remaining mixtures ΔL_f are negative and ΔZ behaves in opposite manner to ΔL_f . Positive and negative deviations in these functions from rectilinear dependence on composition of the mixtures indicate the extent of association or dissociation between the mixing components.¹² Thus the graded trend obtained from these results support our earlier results.

Many semi-theoretical and semi-empirical models have been proposed to estimate the isothermal viscosity of the binary liquid mixtures in terms of pure-component data. We have attempted McAllister's multibody interaction model^{25,26} along with Heric-Brewer model²⁷ to correlate kinematic viscosities ν , of the binary mixtures with the mole fraction of 2-butanone.

The models are defined as follows:

McAllister's three-body model:

$$\begin{aligned} \ln \nu = & x_1^3 \ln \nu_1 + x_2^3 \ln \nu_2 + 3x_1^2 x_2 \ln \nu_{12} + 3x_2^3 x_1 \ln \nu_{21} - \ln \left[x_1 + \frac{x_2 M_2}{M_1} \right] \\ & + 3x_1^2 x_2 \ln \left[\frac{2}{3} + \frac{M_2}{3M_1} \right] + 3x_2^3 x_1 \ln \left[\frac{1}{3} + \frac{2M_2}{3M_1} \right] + x_2^3 \ln \left[\frac{M_2}{M_1} \right] \end{aligned} \quad (20)$$

McAllister's four-body model:

$$\begin{aligned} \ln \nu = & x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} + 4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 \\ & - \ln \left[x_1 + x_2 \left(\frac{M_2}{M_1} \right) \right] + 4x_1^3 x_2 \ln \left[\frac{3}{4} + \frac{M_2}{4M_1} \right] + 6x_1^2 x_2^2 \ln \left[\frac{1}{2} + \frac{M_2}{2M_1} \right] \\ & + 4x_1 x_2^3 \ln \left[\frac{1}{3} + \frac{3M_2}{4M_1} \right] + x_2^4 \ln \left[\frac{M_2}{M_1} \right] \end{aligned} \quad (21)$$

Heric-Brewer model:

$$\nu = x_1 \nu_1 + x_2 \nu_2 + x_1 x_2 \left\{ a + b(x_1 - x_2) + c(x_1 - x_2)^2 \right\} \quad (22)$$

where ν , ν_1 and ν_2 are kinematic viscosities of the mixture, the pure component 1 and 2, respectively. ν_{12} , ν_{21} , ν_{1112} , ν_{1122} , ν_{2221} , a , b and c are model parameters and x_1 and M_1 are the mole fraction and molecular

weight of the i^{th} pure component in the mixture, respectively. The various calculated parameters are presented in Table 7 and comparatively smaller σ values for the alkoxyethanols in case of Heric-Brewer model²⁷ and those for amines in case of McAllister's three-body model^{25,26} justifies that respective models are more or less suitable for these mixtures.

The excess properties were correlated with the Redlich-Kister equation:²⁸

$$Y_{ij}^E = x_i x_j \sum_{k=1}^m a_k (x_i - x_j)^k \quad (23)$$

where Y_{ij}^E refers to an excess property for each i-j binary pair, and x_i is the mole fraction of i^{th} component, and a_k represents the coefficients. The values of coefficients a_k , were determined by a multiple-regression analysis based on the least-squares method and were summarized along with their standard deviations between the experimental and fitted values of the respective functions in Table 8.

The standard deviation for all the semi-empirical models or equations used was calculated using the relation:

$$\sigma = \left[\sum_{i=1}^n \frac{(Y_{i,\text{exp}}^E - Y_{i,\text{cal}}^E)^2}{n - p} \right]^{\frac{1}{2}} \quad (24)$$

where n is the number of experimental points and p is the number of adjustable parameters. The small σ values for excess or deviation properties indicated that the fits are good and in the present study, V^E , $\Delta\eta$, ΔK_S , ΔL_f and ΔZ are quite systematic and function of the composition of the binary mixtures.

6.4. Conclusion

In summary, amine systems are characterized by the presence of strong hydrogen bond interaction between the mixing liquids and the strength of interaction follows the order: primary amine > secondary amine; also steric and other effects play a pivotal role in this regard. On the contrary, alkoxyethanols systems are characterized by the presence of weak hydrogen bond interaction or dispersive forces in the studied binary

systems. The reason is probably the formation of intramolecular associates in these molecules by the interaction of the etheric oxygen and hydrogen of -OH group in the same alkoxyethanol molecule.

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Table 1.
Physical properties of pure components at 298.15 K.

Pure Components	$\rho \times 10^{-3} / \text{kg.m}^{-3}$		$\eta / \text{mPa.s}$		$u / (\text{m. s}^{-1})$
	Expt.	Lit.	Expt.	Lit.	
2-butanone	0.7981	0.7981 [2]	0.373	0.373 [2]	1195.4
2-methoxyethanol	0.9603	0.9602 [7]	1.540	1.5414 [7]	1340.2
2-ethoxyethanol	0.9250	0.9256 [8]	1.850	1.850 [8]	1302.8
2-butoxyethanol	0.8965	0.8966 [5]	2.792	2.795 [5]	1303.2
Isopropylamine	0.6815		0.278		1075.6
Cyclohexylamine	0.8668		1.753		1416.4
Diethylamine	0.6984		0.265		1130.6

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Table 2.

Correlation coefficients of density, viscosity and ultrasonic speed of the binary mixtures from equation 1 at 298.15 K.

System	a_0	a_1	a_2	a_3	s
$\rho \times 10^{-3} / \text{kg.m}^{-3}$					
BU (1) + ME (2)	0.9603	-0.1758	0.0219	-0.0074	0.0008
BU (1) + EE (2)	0.9250	-0.1118	-0.0079	-0.0072	0.0007
BU (1) + BE (2)	0.8965	-0.0663	-0.0121	-0.0200	0.0006
BU (1) + IPA (2)	0.6815	0.1569	0.0328	-0.0728	0.0006
BU(1) + CHA (2)	0.8664	-0.0237	0.0287	-0.0739	0.0007
BU (1) + DEA (2)	0.6982	0.1012	0.0070	-0.0086	0.0006
$\eta / \text{mPa.s}$					
BU (1) + ME (2)	1.547	-2.311	1.024	0.129	0.002
BU (1) + EE (2)	1.853	-3.353	2.288	-0.409	0.001
BU (1) + BE (2)	2.785	-4.886	2.708	-0.227	0.002
BU (1) + IPA (2)	0.277	0.132	0.098	-0.136	0.003
BU (1) + CHA (2)	1.752	1.345	0.070	-0.106	0.003
BU (1) + DEA (2)	0.266	0.077	-0.117	0.150	0.002
$u / \text{m.s}^{-1}$					
BU (1) + ME (2)	1340.1	-99.8	-31.9	-13.4	0.1
BU (1) + EE (2)	1304.0	-214.9	103.2	4.3	0.2
BU (1) + BE (2)	1304.5	-264.3	177.0	-19.9	0.3
BU (1) + IPA (2)	1072.9	374.5	-358.9	104.9	0.7
BU (1) + CHA (2)	1410.1	-108.7	-88.7	-25.9	1.5
BU (1) + DEA (2)	1128.4	129.1	-22.9	-41.5	0.5

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Table 3

Experimental values of density, $\rho \times 10^{-3}/\text{kg.m}^{-3}$; viscosity, $\eta / \text{mPa.s}$; excess molar volume, $V^E \times 10^6 / \text{m}^3.\text{mol}^{-1}$; and deviations in viscosity, $\Delta\eta / \text{mPa.s}$ for the binary mixtures under investigation at 298.15 K.

Mole fraction of BU (x_1)	$\rho \times 10^{-3}/\text{kg.m}^{-3}$	$\eta / \text{mPa.s}$	$V^E \times 10^6 / \text{m}^3.\text{mol}^{-1}$	$\Delta\eta / \text{mPa.s}$
BU (1) + ME (2)				
0	0.9603	1.540	0	0
0.1049	0.9420	1.321	-0.071	-0.097
0.2087	0.9245	1.113	-0.150	-0.184
0.3115	0.9074	0.937	-0.210	-0.240
0.4130	0.8908	0.780	-0.253	-0.278
0.5135	0.8746	0.640	-0.277	-0.301
0.6128	0.8588	0.525	-0.282	-0.300
0.7112	0.8433	0.464	-0.264	-0.246
0.8085	0.8281	0.430	-0.223	-0.166
0.9047	0.8131	0.410	-0.137	-0.074
1	0.7981	0.373	0	0
BU (1) + EE (2)				
0	0.9250	1.850	0	0
0.1219	0.9111	1.479	-0.065	-0.191
0.2381	0.8978	1.178	-0.139	-0.320
0.3488	0.8847	0.948	-0.197	-0.387
0.4545	0.8719	0.767	-0.234	-0.411
0.5555	0.8593	0.609	-0.257	-0.420
0.6521	0.8467	0.524	-0.248	-0.363
0.7446	0.8344	0.457	-0.222	-0.293
0.8333	0.8221	0.419	-0.167	-0.200
0.9183	0.8101	0.387	-0.094	-0.106
1	0.7981	0.373	0	0
BU (1) + BE (2)				
0	0.8965	2.792	0	0
0.1540	0.8860	2.106	-0.052	-0.313
0.2906	0.8758	1.609	-0.110	-0.480
0.4126	0.8657	1.234	-0.156	-0.560
0.5221	0.8558	0.939	-0.181	-0.590
0.6211	0.8458	0.727	-0.171	-0.562
0.7108	0.8361	0.607	-0.161	-0.466
0.7927	0.8265	0.519	-0.140	-0.355
0.8676	0.8169	0.466	-0.101	-0.227
0.9365	0.8075	0.416	-0.054	-0.110
1	0.7981	0.373	0	0
BU (1) + IPA (2)				
0	0.6815	0.278	0	0
0.0835	0.6946	0.288	-0.371	0.002
0.1701	0.7089	0.300	-0.845	0.005
0.2600	0.7236	0.314	-1.318	0.011
0.3534	0.7381	0.330	-1.700	0.018
0.4505	0.7522	0.345	-1.993	0.024
0.5515	0.7652	0.358	-2.106	0.028

Contd...

Excess Molar Volumes.....Some Alkoxyethanols and Amines at 298.15 K

0.6567	0.7781	0.370	-2.165	0.030
0.7663	0.7884	0.371	-1.890	0.020
0.8806	0.7962	0.372	-1.299	0.010
1	0.7981	0.373	0	0
BU (1) + CHA (2)				
0	0.8668	1.753	0	0
0.1326	0.8630	1.574	-0.463	0.004
0.2559	0.8609	1.409	-1.100	0.009
0.3708	0.8580	1.257	-1.606	0.016
0.4783	0.8540	1.115	-1.937	0.022
0.5790	0.8482	0.978	-2.034	0.025
0.6735	0.8409	0.846	-1.949	0.022
0.7624	0.8322	0.719	-1.692	0.018
0.8462	0.8222	0.599	-1.279	0.014
0.9252	0.8102	0.481	-0.651	0.005
1	0.7981	0.373	0	0
BU (1) + DEA (2)				
0	0.6984	0.265	0	0
0.1013	0.7083	0.273	-0.153	-0.003
0.2023	0.7185	0.279	-0.317	-0.008
0.3030	0.7293	0.283	-0.505	-0.014
0.4034	0.7393	0.287	-0.556	-0.021
0.5035	0.7500	0.293	-0.660	-0.026
0.6034	0.7605	0.300	-0.695	-0.030
0.7030	0.7697	0.313	-0.540	-0.028
0.8023	0.7789	0.331	-0.350	-0.020
0.9013	0.7882	0.353	-0.153	-0.009
1	0.7981	0.373	0	0

Table 4.

Experimental values of ultrasonic speed, u /m.s⁻¹; isentropic compressibility, $K_S \times 10^{12}$ /Pa⁻¹ and deviation in isentropic compressibility, $\Delta K_S \times 10^{12}$ /Pa⁻¹ of binary mixtures at 298.15 K.

x_1	u /m.s ⁻¹	$K_S \times 10^{12}$ /Pa ⁻¹	$\Delta K_S \times 10^{12}$ /Pa ⁻¹	x_1	u /m.s ⁻¹	$K_S \times 10^{12}$ /Pa ⁻¹	$\Delta K_S \times 10^{12}$ /Pa ⁻¹
BU (1) + ME (2)				BU (1) + EE (2)			
0	1340.2	579.7	0	0	1302.8	636.9	0
0.1049	1329.4	600.6	-10.3	0.1219	1280.8	669.1	2.9
0.2087	1317.6	623.0	-18.7	0.2381	1260.0	701.6	7.5
0.3115	1305.4	646.7	-25.6	0.3488	1241.7	733.1	12.5
0.4130	1292.2	672.3	-30.1	0.4545	1226.9	762.0	16.0
0.5135	1278.9	699.1	-33.2	0.5555	1215.8	787.3	17.2
0.6128	1264.6	728.1	-33.7	0.6521	1208.4	808.8	15.4
0.7112	1248.3	761.0	-30.0	0.7446	1204.0	826.7	11.2
0.8085	1230.9	797.0	-22.9	0.8333	1200.9	843.4	6.6
0.9047	1213.2	835.6	-12.9	0.9183	1198.1	859.9	2.7
1	1195.4	876.8	0	1	1195.4	876.8	0
BU (1) + BE (2)				BU (1) + IPA (2)			
0	1303.2	656.8	0	0	1075.6	1268.3	0
0.1540	1270.0	699.8	9.1	0.0835	1100.1	1189.6	-46.0
0.2906	1243.5	738.4	17.7	0.1701	1124.4	1115.7	-86.0
0.4126	1223.4	771.7	24.2	0.2600	1144.8	1054.5	-112.0
0.5221	1209.4	798.8	27.2	0.3534	1165.2	997.9	-132.0
0.6211	1202.3	817.9	24.4	0.4505	1184.2	947.9	-144.0
0.7108	1199.6	831.2	18.0	0.5515	1190.3	922.4	-130.0
0.7927	1198.0	843.1	11.9	0.6567	1192.2	904.2	-107.0
0.8676	1197.1	854.2	6.5	0.7663	1193.6	890.3	-78.0
0.9365	1196.6	865.0	2.2	0.8806	1194.3	880.5	-43.0
1	1195.4	876.8	0	1	1195.4	876.8	0

Contd...

BU (1) + CHA (2)				BU (1) + DEA (2)			
0	1416.4	575.1	0	0	1130.6	1120.1	0
0.1326	1385.6	603.6	-11.5	0.1013	1139.7	1086.9	-8.6
0.2559	1369.1	619.7	-32.6	0.2023	1150.0	1052.3	-18.6
0.3708	1357.9	632.1	-54.9	0.3030	1163.4	1013.1	-33.3
0.4783	1343.3	648.9	-70.5	0.4034	1176.0	978.1	-43.9
0.5790	1321.2	675.4	-74.4	0.5035	1185.5	948.6	-49.0
0.6735	1290.3	714.3	-64.0	0.6034	1190.5	927.8	-45.5
0.7624	1258.1	759.2	-45.9	0.7030	1192.4	913.8	-35.3
0.8462	1230.2	803.7	-26.7	0.8023	1193.0	902.1	-22.8
0.9252	1209.1	844.2	-10.0	0.9013	1193.7	890.3	-10.5
1	1195.4	876.8	0	1	1195.4	876.8	0

Table 5

Derived values of Vander Waal's constant, b/m^3 ; molecular radius, r/nm ; geometrical volume, $B/(\text{m}^3.\text{mol}^{-1})$; molar speed of sound, $R_s/(\text{m}^3.\text{mol}^{-1}.\text{ms}^{-1})^{1/3}$; available volume, $V_a/(\text{m}^3.\text{mol}^{-1})$; intermolecular free length, $L_f/\text{\AA}$; molar volume at absolute zero, $V_0/(\text{m}^3.\text{mol}^{-1})$; molar surface area, $Y/\text{\AA}^2$; and specific acoustic impedance, $Z \times 10^{-6}/(\text{kg}.\text{m}^{-2}.\text{s}^{-1})$ of the pure components at 298.15 K.

Component	$b \times 10^6$	r	$B \times 10^6$	$R_s \times 10^3$	$V_a \times 10^6$	L_f	$V_0 \times 10^6$	Y	$Z \times 10^{-6}$
BU	84.1	0.203	21.0	958.9	22.8	0.609	67.5	31.1	0.954
ME	74.3	0.195	18.6	873.7	12.9	0.495	66.4	28.6	1.287
EE	91.7	0.209	22.9	1064.1	18.1	0.519	79.3	32.9	1.205
BE	124.8	0.231	31.2	1439.8	24.4	0.527	107.4	40.5	1.168
IPA	79.8	0.199	20.0	888.5	28.4	0.732	58.3	30.0	0.733
CHA	108.3	0.221	27.1	1285.0	13.1	0.493	101.0	36.8	1.228
DEA	97.3	0.213	24.3	1091.0	30.7	0.688	74.0	34.3	0.790

Table 6

Ultrasonic speed, u /(m.s⁻¹); available volume, V_a /(m³.mol⁻¹); relative association, R_A ; specific acoustic impedance, $Z \times 10^{-6}$ /(kg.m⁻².s⁻¹); molar speed of sound, $R_S/\{m^3.mol^{-1}.(m.s^{-1})^{1/3}\}$; molecular association, M_A ; intermolecular free length, L_f /Å; deviation in intermolecular free length, ΔL_f /Å; and deviation in specific acoustic impedance, $\Delta Z \times 10^{-6}$ /(kg.m⁻².s⁻¹) for the binary mixtures at 298.15 K.

x_1	u	$V_a \times 10^6$	R_A	$Z \times 10^{-6}$	$R_S \times 10^3$	M_A	L_f	ΔL_f	$\Delta Z \times 10^{-6}$
BU (1) + ME (2)									
0.1049	1329.4	13.6	1.139	1.252	1042.3	-0.494	0.504	-0.003	0.000
0.2087	1317.6	14.4	1.121	1.218	1033.4	-0.502	0.513	-0.006	0.001
0.3115	1305.4	15.2	1.104	1.185	1024.4	-0.509	0.523	-0.008	0.001
0.4130	1292.2	16.1	1.087	1.151	1015.3	-0.516	0.533	-0.009	0.002
0.5135	1278.9	17.0	1.071	1.118	1006.4	-0.523	0.544	-0.010	0.002
0.6128	1264.6	18.0	1.056	1.086	997.3	-0.529	0.555	-0.010	0.003
0.7112	1248.3	19.1	1.041	1.053	987.8	-0.535	0.567	-0.009	0.002
0.8085	1230.9	20.3	1.027	1.019	978.1	-0.542	0.581	-0.007	0.001
0.9047	1213.2	21.5	1.014	0.986	968.4	-0.548	0.595	-0.004	0.001
BU (1) + EE (2)									
0.1219	1280.8	19.1	0.945	1.167	1190.0	-0.501	0.532	0.002	-0.007
0.2381	1260.0	20.1	0.936	1.131	1151.1	-0.506	0.545	0.004	-0.014
0.3488	1241.7	21.0	0.927	1.099	1116.4	-0.511	0.557	0.006	-0.019
0.4545	1226.9	21.6	0.917	1.070	1085.6	-0.517	0.568	0.008	-0.021
0.5555	1215.8	22.1	0.907	1.045	1058.3	-0.522	0.577	0.008	-0.021
0.6521	1208.4	22.4	0.895	1.023	1034.2	-0.528	0.585	0.007	-0.018
0.7446	1204.0	22.5	0.883	1.005	1012.8	-0.534	0.591	0.005	-0.014
0.8333	1200.9	22.6	0.871	0.987	993.4	-0.541	0.597	0.003	-0.009
0.9183	1198.1	22.7	0.859	0.970	975.5	-0.547	0.603	0.001	-0.004

Contd...

BU (1) + BE (2)									
0.1540	1270.0	25.0	0.921	1.125	1459.8	-0.526	0.544	0.004	-0.010
0.2906	1243.5	25.4	0.917	1.089	1342.9	-0.526	0.559	0.008	-0.017
0.4126	1223.4	25.4	0.912	1.059	1253.3	-0.527	0.571	0.010	-0.021
0.5221	1209.4	25.3	0.905	1.035	1183.0	-0.529	0.581	0.011	-0.021
0.6211	1202.3	24.9	0.896	1.017	1127.2	-0.532	0.588	0.010	-0.018
0.7108	1199.6	24.3	0.886	1.003	1081.9	-0.535	0.593	0.008	-0.013
0.7927	1198.0	23.8	0.876	0.990	1043.9	-0.539	0.597	0.005	-0.008
0.8676	1197.1	23.4	0.867	0.978	1011.5	-0.544	0.601	0.003	-0.004
0.9365	1196.6	23.1	0.857	0.966	983.5	-0.549	0.605	0.0011	-0.001
BU (1) + IPA (2)									
0.0835	1100.1	27.0	0.758	0.764	776.1	-0.591	0.709	-0.013	0.013
0.1701	1124.4	25.6	0.768	0.797	794.4	-0.582	0.687	-0.024	0.026
0.2600	1144.8	24.4	0.779	0.828	812.7	-0.572	0.668	-0.032	0.038
0.3534	1165.2	23.2	0.790	0.860	832.3	-0.564	0.650	-0.039	0.049
0.4505	1184.2	22.2	0.801	0.891	852.7	-0.557	0.633	-0.044	0.058
0.5515	1190.3	22.0	0.813	0.911	871.5	-0.551	0.625	-0.040	0.056
0.6567	1192.2	22.0	0.826	0.928	890.7	-0.546	0.618	-0.033	0.049
0.7663	1193.6	22.1	0.837	0.941	911.5	-0.544	0.614	-0.024	0.039
0.8806	1194.3	22.4	0.845	0.951	934.0	-0.546	0.610	-0.013	0.023
BU (1) + CHA (2)									
0.1326	1385.6	14.7	0.872	1.196	1319.7	-0.613	0.505	-0.003	0.004
0.2559	1369.1	15.2	0.873	1.179	1258.9	-0.599	0.512	-0.011	0.021
0.3708	1357.9	15.3	0.873	1.165	1207.9	-0.587	0.517	-0.019	0.039
0.4783	1343.3	15.8	0.872	1.147	1162.4	-0.575	0.524	-0.025	0.050
0.5790	1321.2	16.7	0.871	1.121	1120.1	-0.567	0.534	-0.026	0.051
0.6735	1290.3	18.2	0.870	1.085	1079.8	-0.560	0.550	-0.021	0.042
0.7624	1258.1	19.8	0.868	1.047	1043.0	-0.555	0.567	-0.015	0.028
0.8462	1230.2	21.2	0.864	1.011	1010.5	-0.553	0.583	-0.008	0.015

Contd...

0.9252	1209.1	22.2	0.857	0.980	982.6	-0.553	0.598	-0.003	0.005
BU (1) + DEA (2)									
0.1013	1139.7	29.7	0.764	0.807	955.8	-0.612	0.678	-0.002	0.001
0.2023	1150.0	28.5	0.772	0.826	957.3	-0.605	0.667	-0.005	0.003
0.3030	1163.4	27.2	0.781	0.848	959.7	-0.598	0.655	-0.010	0.009
0.4034	1176.0	26.1	0.789	0.869	961.7	-0.591	0.643	-0.013	0.013
0.5035	1185.5	25.1	0.798	0.889	962.9	-0.584	0.633	-0.015	0.017
0.6034	1190.5	24.4	0.808	0.905	962.9	-0.577	0.626	-0.014	0.016
0.7030	1192.4	24.0	0.818	0.918	962.1	-0.572	0.622	-0.011	0.013
0.8023	1193.0	23.6	0.827	0.929	960.9	-0.566	0.618	-0.007	0.008
0.9013	1193.7	23.3	0.837	0.941	959.7	-0.560	0.614	-0.003	0.003

Table 7
 Values of McAllister and Heric-Brewer parameters for the binary mixtures at 298.15 K

System	McAllister's Three body model			McAllister's Four body model				Heric-Brewer model			
	v_{12}	v_{21}	s	v_{1112}	v_{1122}	v_{2221}	s	$a \times 10^2$ /(cm ² .s ⁻¹)	$b \times 10^2$ /(cm ² .s ⁻¹)	$c \times 10^2$ /(cm ² .s ⁻¹)	s
BU (1) + ME (2)	0.3686	0.4417	0.072	0.3160	0.2480	1.0701	0.107	-1.4063	-0.9457	-0.4536	0.082
BU (1) + EE (2)	0.3688	0.8540	0.031	0.2449	0.3035	0.2161	0.063	-2.3859	-0.4918	-1.4432	0.092
BU (1) + BE (2)	0.5582	0.8298	0.075	0.4112	0.3728	0.5275	0.093	-3.6866	-1.6348	-1.2126	0.162
BU (1) + IPA (2)	0.4596	0.4247	0.024	0.4257	0.4280	0.3006	0.022	0.0553	-0.1239	-0.4831	0.018
BU (1) + CHA (2)	1.1685	1.0506	0.099	1.0773	1.0316	1.5625	0.025	-0.1249	-0.1309	-0.5902	0.030
BU (1) + DEA (2)	0.3527	0.3083	0.019	0.3725	0.3029	0.3733	0.019	-0.1839	-0.2796	-0.0719	0.013

Table 8
 Redlich-Kister coefficients and standard deviations (σ) for excess/deviation
 properties of the binary mixtures at 298.15 K.

Property	a_0	a_1	a_2	a_3	σ
BU (1) + ME (2)					
$V^E \times 10^6$ /($\text{m}^3 \cdot \text{mol}^{-1}$)	-1.096	-0.329	-0.225	-0.307	0.001
$\Delta\eta$ /($\text{mPa}\cdot\text{s}$)	-1.202	-0.332	0.151	1.447	0.002
$\Delta K_S \times 10^{12}$ / Pa^{-1}	-132.3	-38.9	4.2	23.9	0.2
ΔL_f / \AA	-0.039	-0.008			0.001
$\Delta Z \times 10^{-6}$ /($\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$)	0.009	0.006			0.001
BU (1) + EE (2)					
$V^E \times 10^6$ /($\text{m}^3 \cdot \text{mol}^{-1}$)	-0.991	-0.395	0.108		0.002
$\Delta\eta$ /($\text{mPa}\cdot\text{s}$)	-1.674	0.147	0.127	0.192	0.005
$\Delta K_S \times 10^{12}$ / Pa^{-1}	67.6	25.7	-72.8	-50.8	0.1
ΔL_f / \AA	0.032	0.003	-0.023		0.000
$\Delta Z \times 10^{-6}$ /($\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$)	-0.086	0.007	0.040		0.000
BU (1) + BE (2)					
$V^E \times 10^6$ /($\text{m}^3 \cdot \text{mol}^{-1}$)	-0.693	-0.254	0.134	-0.189	0.003
$\Delta\eta$ /($\text{mPa}\cdot\text{s}$)	-2.360	-0.293	0.480	2.426	0.003
$\Delta K_S \times 10^{12}$ / Pa^{-1}	107.6	14.1	-130.6	-45.8	0.3
ΔL_f / \AA	0.045	-0.003	-0.033		0.001
$\Delta Z \times 10^{-6}$ /($\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$)	-0.085	0.023	0.055		0.000
BU (1) + IPA (2)					
$V^E \times 10^6$ /($\text{m}^3 \cdot \text{mol}^{-1}$)	-8.321	-3.035	-0.993	-2.648	0.019
$\Delta\eta$ /($\text{mPa}\cdot\text{s}$)	0.108	0.088	-0.077	-0.079	0.001
$\Delta K_S \times 10^{12}$ / Pa^{-1}	-552.4	180.0	228.5	-97.2	2.1
ΔL_f / \AA	-0.166	0.034	0.038		0.001

Contd...

Excess Molar Volumes....2-Butanone with Some Alkoxyethanols and Amines at 298.15 K

$\Delta Z \times 10^{-6}$ /(kg.m ⁻² .s ⁻¹)	0.226	0.019	-0.064		0.001
BU (1) + CHA (2)					
$V^E \times 10^6$ /(m ³ .mol ⁻¹)	-7.906	-3.258	2.827	-1.224	0.004
$\Delta \eta$ /(mPa.s)	0.089	0.066	-0.050	-0.044	0.001
$\Delta K_s \times 10^{12}$ /Pa ⁻¹	-290.1	-161.8	338.9	598.1	0.0
ΔL_f /Å	-0.102	-0.042	0.142	0.043	0.000
$\Delta Z \times 10^{-6}$ /(kg.m ⁻² .s ⁻¹)	0.202	0.054	-0.255		0.001
BU (1) + DEA (2)					
$V^E \times 10^6$ /(m ³ .mol ⁻¹)	-2.671	-0.753	1.498	1.333	0.026
$\Delta \eta$ /(mPa.s)	-0.105	-0.100	0.025	0.085	0.000
$\Delta K_s \times 10^{12}$ /Pa ⁻¹	-191.6	-20.0	156.6		1.0
ΔL_f /Å	-0.058	-0.009	0.052		0.000
$\Delta Z \times 10^{-6}$ /(kg.m ⁻² .s ⁻¹)	0.065	0.022	-0.076		0.000

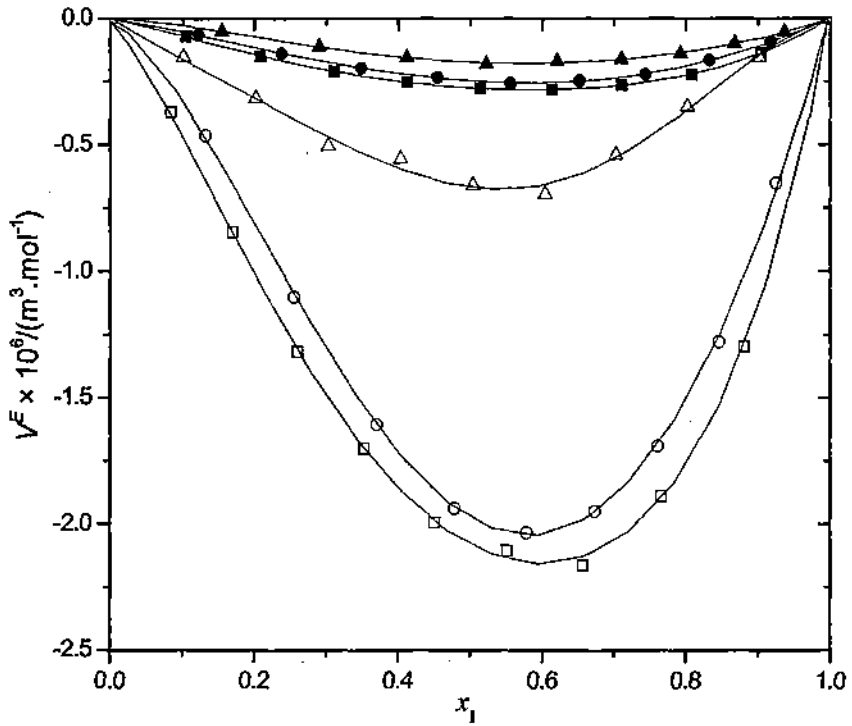


Figure 1. Variation of excess molar volumes, $V^E \times 10^6 / (\text{m}^3 \cdot \text{mol}^{-1})$ against mole fraction (x_1) of 2-butanone at 298.15 K with 2-methoxyethanol (■), 2-ethoxyethanol (●), 2-butoxyethanol (▲), isopropylamine (□), cyclohexylamine (○) and diethylamine (Δ).

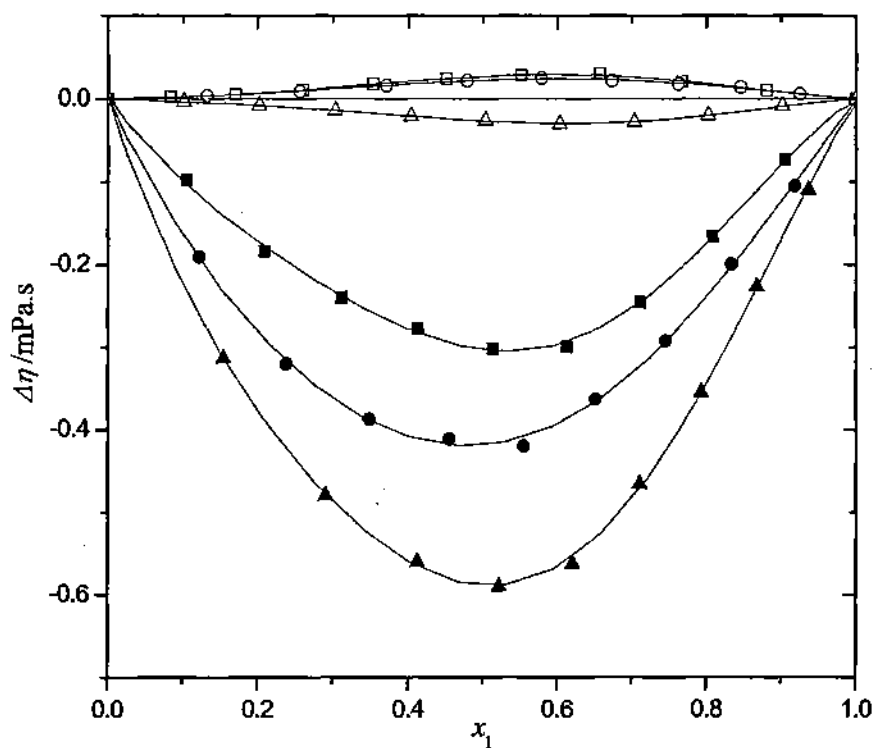


Figure 2. Variation of viscosity deviations, $\Delta\eta$ /(mPa.s) against mole fraction (x_1) of 2-butanone at 298.15 K with 2-methoxyethanol (■), 2-ethoxyethanol (●), 2-butoxyethanol (▲), isopropylamine (□), cyclohexylamine (○) and diethylamine (Δ).

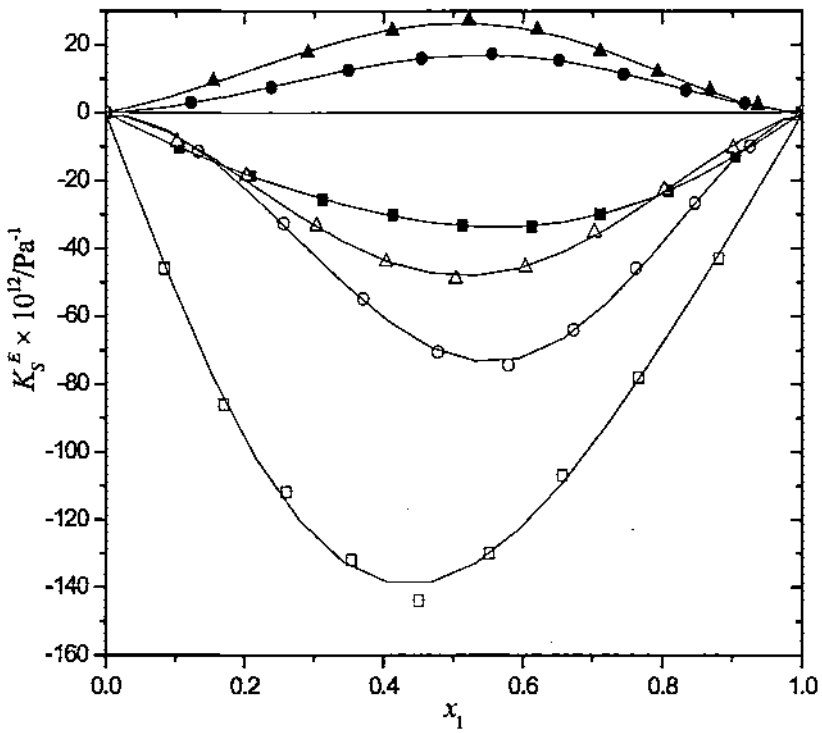


Figure 3. Variation of deviations in isentropic compressibility, $\Delta K_s \times 10^{12}/\text{Pa}^{-1}$ against mole fraction (x_1) of 2-butanone at 298.15 K with 2-methoxyethanol (■), 2-ethoxyethanol (●), 2-butoxyethanol (▲), isopropylamine (□), cyclohexylamine (○) and diethylamine (△).

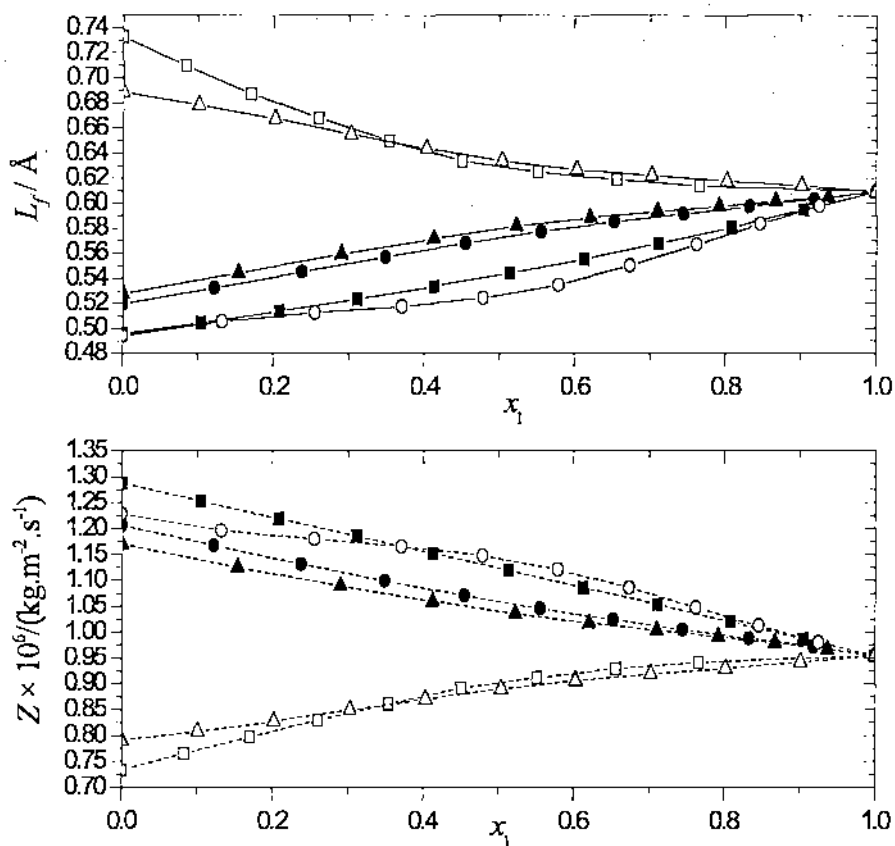


Figure 4. Variation of intermolecular free length, $L_f / \text{Å}$ and specific acoustic impedance, $Z \times 10^6 / (\text{kg.m}^{-2}.\text{s}^{-1})$ against mole fraction (x_1) of 2-butanone at 298.15 K with 2-methoxyethanol (■), 2-ethoxyethanol (●), 2-butoxyethanol (▲), isopropylamine (□), cyclohexylamine (○) and diethylamine (Δ).

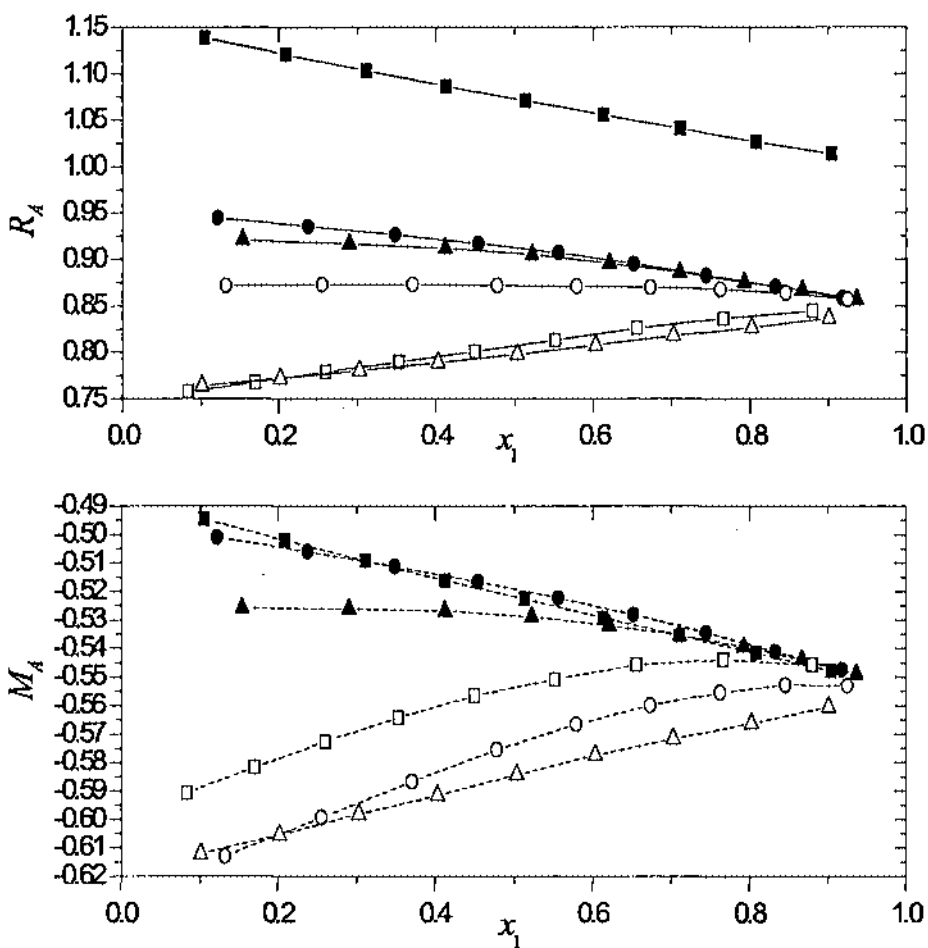


Figure 5. Variation of relative association, R_A and molecular association, M_A against mole fraction (x_1) of 2-butanone at 298.15 K with 2-methoxyethanol (■), 2-ethoxyethanol (●), 2-butoxyethanol (▲), isopropylamine (□), cyclohexylamine (○) and diethylamine (Δ).

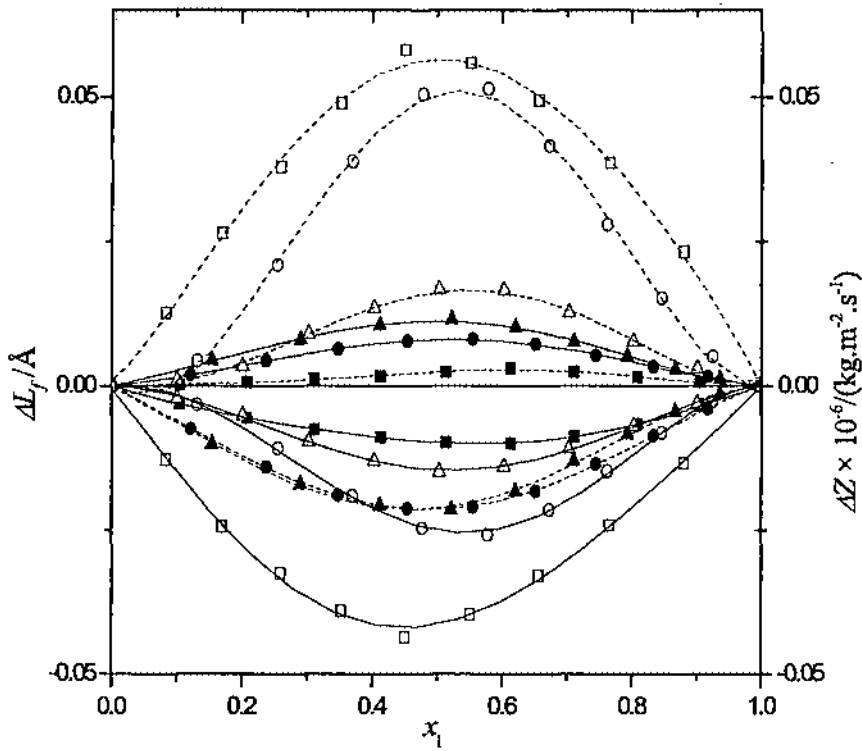


Figure 6. Variation of deviations in intermolecular free length, $\Delta L_f / \text{\AA}$; [solid lines] and deviation in specific acoustic impedance, $\Delta Z \times 10^{-6} / (\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1})$; [dotted lines] against mole fraction (x_1) of 2-butanone at 298.15 K with 2-methoxyethanol (■), 2-ethoxyethanol (●), 2-butoxyethanol (▲), isopropylamine (□), cyclohexylamine (○) and diethylamine (Δ).