

CHAPTER IV

Studies on Thermodynamic and Transport Properties of Binary Mixtures of Acetonitrile with Some Cyclic Ethers at Different Temperatures by Volumetric, Viscometric, and Interferometric Techniques*

4.1. *Introduction*

The mixing of different solvents gives rise to solutions that generally do not behave ideally. This deviation from ideality is expressed by many thermodynamic variables, particularly by excess properties. Excess thermodynamic properties of solvent mixtures correspond to the difference between the actual property and the property if the system behaves ideally and, thus, are useful in the study of molecular interactions and arrangements. In particular, they reflect the interactions that take place between solute-solute, solute-solvent, and solvent-solvent species.¹

This work is a part of our program to provide data for the characterization of the molecular interactions between solvents in binary systems.^{2,3} Acetonitrile is a dipolar aprotic solvent lacking strong specific intermolecular forces, where dipole-dipole forces predominate,^{4,5} and tetrahydrofuran, 1,3-dioxolane, and 1,4-dioxane are cyclic ethers differing in the number and position of oxygen atom and methylene group.^{6,7} Tetrahydrofuran, 1,3-dioxolane, and 1,4-dioxane are versatile solvents used in the separation of saturated and unsaturated hydrocarbons, in pharmaceutical synthesis, and serve as solvents for many polymers. Acetonitrile has important technological applications, namely, in battery industry and plating techniques.^{8,9}

In the present chapter, we report densities, viscosities, and ultrasonic speeds for the binary systems of acetonitrile + tetrahydrofuran, acetonitrile + 1,3-dioxolane, and acetonitrile + 1,4-dioxane at the temperatures of (298.15, 308.15, and 318.15) K and atmospheric pressure over the entire composition range. The experimental data are used to calculate excess molar volumes (V^E), deviations in viscosity ($\Delta\eta$), and deviations in isentropic compressibility (ΔK_s) of the mixtures. Various thermodynamic parameters

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(e.g., intermolecular free length, specific acoustic impedance, etc.) and their deviations have also been derived from the density and ultrasonic speed data. These results are useful for the interpretation of the nature of interactions that occur between acetonitrile and the cyclic ethers. The work also provides a test of various empirical equations to correlate viscosity and acoustic data of binary mixtures in terms of pure component properties.

4.2. Experimental Section

4.2.1. Materials

Acetonitrile (Merck, India) was distilled from P_2O_5 and then from CaH_2 in an all-glass distillation apparatus,¹⁰ the middle fraction was collected. 1,4-Dioxane (Merck, India) was kept several days over potassium hydroxide (KOH), refluxed for 24 h, and distilled over lithium aluminum hydride ($LiAlH_4$) as described earlier.² 1,3-Dioxolane (LR) was purified by standard methods. It was refluxed with PbO_2 and then fractionally distilled after addition of xylene.¹² Tetrahydrofuran (Merck, India) was kept several days over potassium hydroxide (KOH), refluxed for 24 h, and distilled over $LiAlH_4$ as described earlier.¹¹ The methods of purification of the chemicals are also discussed in details under chapter III. The purity of the solvents was ascertained by comparing experimental values of densities and viscosities with available literature as listed in Table 1.

4.2.2. Apparatus and Procedure

The densities were measured with an Ostwald-Sprengel type pycnometer having a bulb volume of 25 cm^3 and an internal diameter of the capillary of about 0.1cm, calibrated at (298.15, 308.15, and 318.15) K with doubly distilled water and benzene. The pycnometer with the test solution was equilibrated in a thermostatic water bath maintained at $\pm 0.01\text{ K}$ of the desired temperature, removed from the bath, properly dried, and weighed in an electronic balance. The evaporation losses remained insignificant during the time of actual measurements. Averages of triplicate measurements were taken into account. The mixtures were prepared by mixing known volume of pure liquids in airtight stoppered bottles. The reproducibility in mole fraction was within ± 0.0002 . The mass measurements, accurate to $\pm 0.01\text{ mg}$, were made on a digital electronic analytical balance (Mettler, AG 285, Switzerland). The total uncertainty of density is $\pm 3 \times 10^{-4}\text{ gm.cm}^{-3}$, and that of temperature is $\pm 0.01\text{ K}$. The

viscosity was measured by means of a suspended Ubbelohde type viscometer, which was calibrated at 298.15 K with triple-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 within ± 0.03 % of the reported value. Details of the methods and techniques of density and viscosity measurements have been described earlier.^{11,13,14} Speeds of sound were determined by a multifrequency ultrasonic interferometer (Mittal Enterprise, New Delhi) working at 5 MHz, calibrated with water, methanol, and benzene at 298.15 K. The details of the methods^{11,13} and techniques have been described earlier in chapter III. The uncertainty of ultrasonic speed measurements is ± 0.2 m.s⁻¹.

4.3. Results and Discussion

The physical properties of the pure liquids along with their literature values are recorded in Table 1. However no literature data for viscosity of 1,3-dioxolane at (308.15 K and 318.15 K) and sound speed of acetonitrile at 298.15 K were available to us. Table 2 lists the experimental values of densities (ρ_i) and viscosities (η_i) of the binary mixtures along with the corresponding mole fractions of acetonitrile (x_1), excess molar volumes (V^E), viscosity deviations ($\Delta\eta$), and interaction parameters (d_{12}, T_{12}, H_{12}) at all the experimental temperatures. The plots of V^E and $\Delta\eta$ against x_1 at 298.15 K are represented in Figure 1 and Figure 2 respectively. Because of similarity in nature, the plots at the other two temperatures are not presented here. The excess molar volumes (V^E) were calculated using equation (1):^{15, 16}

$$V^E = \sum_{i=1}^j x_i M_i (1/\rho - 1/\rho_i) \quad (1)$$

where ρ is the density of the mixture, M_i, x_i and ρ_i are the molecular weight, mole fraction, and density of i th component. The estimated uncertainty for V^E is from (0.001 to 0.014) cm³.mol⁻¹. $\Delta\eta$ can be computed using equation (2),^{16,17}

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

where η is the absolute viscosity of the mixture and x_i, η_i are the mole fraction and viscosity of i th component in the mixture, respectively. The estimated uncertainty for

$\Delta\eta$ ranges from (0.001 to 0.003) mPa·s.

It is seen that, the values of V^E and $\Delta\eta$ (see Table 2) for all the experimental binary mixtures are negative over the entire range of composition and temperature. The negative values of V^E for the three systems are in the following order:

Acetonitrile + 1,4-dioxane > Acetonitrile + 1,3-dioxolane > Acetonitrile + tetrahydrofuran

Negative values of V^E ¹⁸ indicate a specific interaction between the mixing components. The chemical or specific interaction between the mixing molecules results in a volume decrease. The negative values of V^E for the binary mixtures of acetonitrile with the ethers may be attributed to the dipole-induced dipole interactions between the mixing components.¹⁹

The molar volumes of acetonitrile are (52.84, 53.63, 54 cm³·mol⁻¹) and those of tetrahydrofuran, 1,3-dioxolane and 1,4-dioxane are (81.87, 82.74, 83.77, cm³·mol⁻¹); (70.03, 70.80, 71.62 cm³·mol⁻¹) and (85.65, 86.66, 87.70 cm³·mol⁻¹) at 298.15 K, 308.15 K and 318.15 K, respectively. It is clear that, the molar volume values of acetonitrile and the other components differ considerably; hence, nonassociated acetonitrile molecules are interstitially accommodated into clusters of ethers yielding a negative contribution to observed V^E values. This implies that the complex-forming interactions are almost absent in the experimental binary systems and therefore observed $\Delta\eta$ values are also negative.²⁰

From close observation of Table 2, it is seen that the negative V^E values are much higher than those of $\Delta\eta$ for all the binary systems under consideration. This clearly supports mere addition of acetonitrile molecules into aggregates of the other components.²⁰

Isentropic compressibility (K_s) values were calculated from experimental densities, ρ and speeds of sound u , using equation (3)

$$K_s = 1/(u^2 \rho) \quad (3)$$

Where K_s gives the isentropic compressibility for the i th component of the mixture.

We also derived the deviations in isentropic compressibility (ΔK_s), deviations in intermolecular free length (ΔL_f) and deviations in specific acoustic impedance (ΔZ) for the binary mixtures using equations (4), (5) and (6).

$$\Delta K_s = K_s - \sum_{i=1}^2 x_i K_{s,i} \quad (4)$$

$$\Delta L_f = L_f - \sum_{i=1}^2 x_i L_{f,i} \quad (5)$$

$$\Delta Z = Z - \sum_{i=1}^2 x_i Z_i \quad (6)$$

Where K_s, L_f and Z are the isentropic compressibility, intermolecular free length and specific acoustic impedance of the mixture and $x_i, K_{s,i}, L_{f,i}$ and Z_i are the mole fraction, isentropic compressibility, intermolecular free length and specific acoustic impedance of i th component in the mixture, respectively. Experimental values of $u, K_s, \Delta K_s, \Delta L_f, \Delta Z$ are listed in Table 3 and the plots of $\Delta K_s, \Delta L_f$ and ΔZ against x_1 are shown in Figure 3 to Figure 5.

For the investigated binary mixtures, the deviations in isentropic compressibility are negative. The composition dependence of ΔK_s for the investigated binary mixtures is shown in Figure 3; it shows that ΔK_s decrease in the order:

Acetonitrile + 1, 4-dioxane > acetonitrile + 1, 3-dioxolane > acetonitrile + tetrahydrofuran

These results can be explained in terms of molecular interactions and structural effects. There is a parallel in the qualitative behaviour of the ΔK_s and V^E curves (Figure 1 and Figure 3).

Figure (4) and Figure (5) shows that ΔL_f are positive for all the binary mixture and ΔZ behaves in opposite manner to ΔL_f . Positive and negative deviations in these functions from linear dependence on composition of the mixtures indicate the extent of association or dissociation between the mixing components.²¹ The observed values of

ΔK_s and ΔL_f can be qualitatively explained by considering the factors - (i) the mutual disruption of associates present in pure liquids, (ii) dipole-induced dipole interaction between the mixing liquids and (iii) interstitial accommodation of one component into another. The second two factors contribute negative ΔK_s and ΔL_f values. Observed negative value of ΔK_s and ΔL_f for the mixtures over the entire range of composition implies that the weak dipole-induced dipole interactions are predominant between the unlike molecules along with interstitial accommodation between the components.^{22, 15} Thus the graded behaviors of these functions support the results obtained earlier.

In an attempt to explore the nature of the interactions occurring between the mixing components, various thermodynamic parameters²¹⁻²⁵ such as intermolecular free length (L_f), specific acoustic impedance (Z), Vander Waal's constant (b), molecular radius (r), geometrical volume (B), molar surface area (Y), available volume (V_a), molar speed of sound (R), relative association (R_A) and molecular association (M_A) of the binary mixtures have been calculated using

$$L_f = K \sqrt{K_s} \quad (7)$$

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

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

$$r = \left(\frac{3b}{16\pi N}\right)^{\frac{1}{3}} \quad (10)$$

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

$$Y = (36\pi NB^2)^{\frac{1}{3}} \quad (12)$$

$$V_a = V - \left(1 - \frac{u}{u_\infty}\right) \quad (13)$$

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

$$R = Mu^{\frac{1}{3}} / \rho \quad (15)$$

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

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

where K is a temperature dependent constant, V_0 is volume at absolute zero, u_{∞} is taken as 1600 ms^{-1} . These parameters are listed in Table 4 for the pure components and in Table 5 for the binary mixtures. Plots of Z , L_f , R , M_A and R_A against x_1 are shown in Figure 6 to Figure 10.

The values of Z and L_f for the mixtures behave in opposite manner and the Figures do not exhibit any sudden variation in their behaviour. This implies the absence of any complex formation²⁴ between the mixing components; that is further supported by the linear variations of R , R_A and M_A of the binary mixtures against mole fraction of acetonitrile.²⁴ M_A and R_A values decrease for the mixtures with increasing mole fraction of acetonitrile. This implies strongly dissociative interactions between the unlike molecules in the mixtures.^{24,25}

The mixing functions V^E , $\Delta\eta$, ΔK_S , ΔL_f and ΔZ were represented mathematically by the following type of Redlich-Kister equation²⁶ [equation (18)] for correlating the experimental data:

$$Y_{ij}^E = x_i x_j \sum_{k=1}^m a_k (x_i - x_j)^k \quad (18)$$

where Y_{ij}^E refers to an excess property (V^e , $\Delta\eta$, ΔK_S , ΔL_f and ΔZ) 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 the standard deviations between the experimental and fitted values of the respective functions in Table 6. The standard deviation was calculated using equation (19).

$$\sigma = \left[\sum_{i=1}^n \frac{(Y_{i,\text{exp}}^E - Y_{i,\text{cal}}^E)^2}{n - p} \right]^{\frac{1}{2}} \quad (19)$$

where n is the number of experimental points and p is the number of adjustable parameters. The small σ values for excess properties indicates that the fits are good for the present study,

Several semi-empirical models have been proposed from time to time to estimate the dynamic viscosity of the binary liquid mixtures in terms of pure-component data^{16, 17} and to interpret the molecular interactions in these mixtures. Some of them we examined are as follows:

Grunberg and Nissan²⁷ have suggested the following logarithmic relation between the viscosity of the binary mixtures and the pure components:

$$\eta = \exp \left[\sum_{i=1}^j (x_i \ln \eta_i) + d_{12} \prod_{i=1}^j x_i \right] \quad (20)$$

where d_{12} is a constant proportional to the interchange energy. It may be regarded as an approximate measure of the strength of molecular interactions between the mixing components. The values of the interchange parameter (d_{12}) have been calculated using equation (20) as a function of the composition of the binary liquid mixtures of acetonitrile with tetrahydrofuran, 1, 3-dioxolane and 1,4-dioxane and were listed in Table 2.

Tamura-Kurata²⁸ put forward the following equation for the viscosity of the binary liquid mixtures:

$$\eta = \sum_{i=1}^j x_i \phi_i \eta_i + 2T_{12} \prod_{i=1}^j [x_i \phi_i]^{\frac{1}{2}} \quad (21)$$

where T_{12} is the interaction parameter and ϕ_i is the volume fraction of i th pure component in the mixture.

The following viscosity model of Hind et al²⁹ may also interpret the molecular interactions:

$$\eta = \sum_{i=1}^j x_i^2 \eta_i + 2H_{12} \prod_{i=1}^j x_i \quad (22)$$

where H_{12} is Hind interaction parameter. In the present study, the values of interaction parameter T_{12} and H_{12} have been calculated from equation (21) and (22), respectively and were listed in Table 2. It is observed that for a given binary mixture T_{12} and H_{12} do not differ appreciably from each other; this is in agreement with the view put forward by Fort and Moore¹⁵ in regard to the nature of parameter T_{12} and H_{12} .

McAllister multibody interaction model³⁰ is widely used to correlate the kinematic viscosities $\left(\nu = \frac{\eta}{\rho}\right)$ of the binary mixtures with mole fraction.

The three-body model is defined as:

$$\begin{aligned} \ln \nu = & x_1^3 \ln \nu_1 + x_2^3 \ln \nu_2 + 3x_1^2 x_2 \ln \nu_{12} + 3x_2^3 x_1 \ln \nu_{21} - \ln \left[x_1 + \frac{x_2 M_2}{M_1} \right] \\ & + 3x_1^2 x_2 \ln \left[\frac{2}{3} + \frac{M_2}{3M_1} \right] + 3x_2^3 x_1 \ln \left[\frac{1}{3} + \frac{2M_2}{3M_1} \right] + x_2^3 \ln \left[\frac{M_2}{M_1} \right] \end{aligned} \quad (23)$$

The four-body model is given by

$$\begin{aligned} \ln \nu = & x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln \nu_{1112} + 6x_1^2 x_2^2 \ln \nu_{1122} + 4x_1 x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 \\ & - \ln \left[x_1 + x_2 \left(\frac{M_2}{M_1} \right) \right] + 4x_1^3 x_2 \ln \left[\frac{3}{4} + \frac{M_2}{4M_1} \right] + 6x_1^2 x_2^2 \ln \left[\frac{1}{2} + \frac{M_2}{2M_1} \right] \\ & + 4x_1 x_2^3 \ln \left[\frac{1}{3} + \frac{3M_2}{4M_1} \right] + x_2^4 \ln \left[\frac{M_2}{M_1} \right] \end{aligned} \quad (24)$$

Where ν, ν_1 and ν_2 are kinematic viscosities of the mixture, the pure component 1 and 2 respectively, $\nu_{12}, \nu_{21}, \nu_{1112}, \nu_{1122}, \nu_{2221}$ are model parameters and M_i are the mole fraction and molecular weight of the i th pure component in the mixture, respectively.

Table 7 records the parameters calculated using equation (23) and (24) along with the standard deviations. It is seen that the values of both the parameters are positive and adequate for all of the binary mixtures.

Heric and Brewer³¹ have proposed an equation for the kinematic viscosity of the binary liquid mixtures

$$v = x_1v_1 + x_2v_2 + x_1x_2 \{a + b(x_1 - x_2) + c(x_1 - x_2)^2\} \quad (25)$$

Where, a , b and c are model parameters and M_i, x_i are the molecular weight and mole fraction of the i th pure component in the mixture, respectively.

The percentage standard deviation²⁴ was calculated by using equation (26).

$$\sigma \% = \left[\frac{\sum \left(100 \left(\frac{v_{\text{exp } il} - v_{\text{calcd}}}{v_{\text{exp } il}} \right)^2 \right)}{(n - m)} \right]^{\frac{1}{2}} \quad (26)$$

where n represents the number of experimental points and m the number of coefficients.

The values of parameters a, b, c have been calculated using equation (25) and listed in Table 7 along with the standard deviations. A perusal of this Table 7 shows that the values of a, b are negative for all of the binary mixtures.

4.4. Conclusion:

In summary, the study provided us with some idea about the type and nature of molecular interactions between the mixing components. Strong specific interaction between acetonitrile and ethers molecules seems to be present. Dipole-induced dipole interactions are present between the unlike molecules along with the interstitial accommodation of non associated acetonitrile molecules in to the clusters of ether molecules. This type of interaction is largest in acetonitrile 1,4-OO mixture and least in case of accetonitrile tetrahydrofuran mixture. This study on excess or deviation properties along with acoustic properties seems to be of much use in exploring the nature of interactions present in the experimental binary mixtures, however, more extensive study will have a better in this field.

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TABLE 1

 Comparison of density (ρ), viscosity (η) and sound speed (u) with Literature Data at the experimental temperatures

Pure Solvent	T / K	$\rho \times 10^3$ $/ (\text{kg} \cdot \text{m}^{-3})$		η $/ (\text{mPa} \cdot \text{s})$		u $/ (\text{m} \cdot \text{s}^{-1})$	
		Expt.	Lit.	Expt.	Lit.	Expt.	Lit.
Acetonitrile	298.15	0.7768	0.77686 ¹⁰	0.3443	0.3446 ¹⁰	1713.2	-
	308.15	0.7654	0.76564 ¹⁰	0.3124	0.3125 ¹⁰		
	318.15	0.7547	0.75498 ¹⁰	0.2891	0.2893 ¹⁰		
Tetrahydro- -furan	298.15	0.8808	0.8807 ³³	0.4631	0.4630 ³³	1292.2	1294 ³⁴
	308.15	0.8715	0.8712 ³³	0.4276	0.4277 ³³		
	318.15	0.8608	0.8614 ³³	0.3903	0.3902 ³³		
1,4-Dioxane	298.15	1.0287	1.0282 ³⁴	1.1779	1.178 ³⁴	1344.4	1358 ³⁴
	308.15	1.0168	1.0168 ³⁴	0.9985	0.999 ³⁴		
	318.15	1.0047	1.00526 ³⁷	0.8909	0.901 ³⁸		
1,3 Dioxolane	298.15	1.0577	1.05862 ³⁵	0.5878	0.5886 ³⁵	1338.2	1338.8 ³⁶
	308.15	1.0463	1.04620 ³⁷	0.5128			
	318.15	1.0344	1.03364 ³⁷	0.4580			

TABLE 2

Values of density (ρ), viscosity (η), excess molar volume (V^E), viscosity deviation ($\Delta\eta$), Grunberg-Nissan, Tamura-Kurata and Hind interaction parameter (d_{12} , T_{12} , H_{12}) for binary mixtures

x_1	$\rho \times 10^3 /$ ($\text{kg} \cdot \text{m}^{-3}$)	$\eta /$ ($\text{mPa} \cdot \text{s}$)	$V^E \times 10^6 /$ ($\text{m}^3 \cdot \text{mol}^{-1}$)	$\Delta\eta /$ ($\text{mPa} \cdot \text{s}$)	d_{12}	T_{12}	H_{12}
Acetonitrile + Tetrahydrofuran							
298.15 K							
0	0.8808	0.4631	0	0	0	0	0
0.1633	0.8703	0.4419	-0.104	-0.002	0.011	0.393	0.397
0.3052	0.8595	0.4236	-0.142	-0.003	0.006	0.392	0.396
0.4295	0.8486	0.4075	-0.145	-0.005	-0.002	0.390	0.394
0.5394	0.8376	0.3932	-0.124	-0.006	-0.015	0.389	0.392
0.6372	0.8268	0.3808	-0.093	-0.007	-0.029	0.387	0.389
0.7249	0.8162	0.3701	-0.062	-0.007	-0.047	0.384	0.387
0.8039	0.8058	0.3611	-0.037	-0.007	-0.067	0.382	0.383
0.8754	0.7959	0.3539	-0.020	-0.005	-0.087	0.380	0.380
0.9405	0.7862	0.3481	-0.007	-0.003	-0.119	0.376	0.375
1	0.7768	0.3443	0	0	0	0	0
Acetonitrile + Tetrahydrofuran							
308.15 K							
0	0.8715	0.4276	0	0	0	0	0
0.1633	0.8604	0.4066	-0.070	-0.002	0.006	0.357	0.361
0.3052	0.8492	0.3875	-0.099	-0.005	-0.013	0.353	0.358
0.4295	0.8380	0.3701	-0.110	-0.008	-0.039	0.349	0.354
0.5394	0.8270	0.3550	-0.106	-0.012	-0.067	0.345	0.349
0.6372	0.8162	0.3425	-0.096	-0.012	-0.095	0.342	0.345
0.7249	0.8057	0.3321	-0.082	-0.012	-0.126	0.338	0.339
0.8039	0.7953	0.3242	-0.066	-0.011	-0.155	0.336	0.336
0.8754	0.7852	0.3184	-0.049	-0.008	-0.184	0.333	0.332

Contd.

Studies on Thermodynamic andTechniques

0.9405	0.7752	0.3145	-0.028	-0.005	-0.214	0.331	0.328
1	0.7654	0.3124	0	0	0	0	0

Acetonitrile + Tetrahydrofuran

318.15 K

0	0.8608	0.3903	0	0	0	0	0
0.1633	0.8510	0.3683	-0.201	-0.005	-0.106	0.313	0.319
0.3052	0.8405	0.3497	-0.290	-0.010	-0.135	0.311	0.317
0.4295	0.8296	0.3340	-0.311	-0.013	-0.169	0.309	0.313
0.5394	0.8184	0.3202	-0.288	-0.016	-0.218	0.305	0.308
0.6372	0.8073	0.3089	-0.245	-0.017	-0.277	0.301	0.303
0.7249	0.7963	0.3001	-0.196	-0.017	-0.349	0.298	0.298
0.8039	0.7855	0.2938	-0.141	-0.015	-0.443	0.294	0.292
0.8754	0.7750	0.2901	-0.094	-0.012	-0.581	0.291	0.287
0.9405	0.7647	0.2879	-0.045	-0.007	-0.960	0.284	0.276
1	0.7547	0.2890	0	0	0	0	0

Acetonitrile + 1,3-Dioxolane

298.15 K

0	1.0577	0.5878	0	0	0	0	0
0.1670	1.0227	0.5233	-0.124	-0.024	-0.193	0.355	0.380
0.3109	0.9899	0.4737	-0.233	-0.038	-0.231	0.355	0.376
0.4361	0.9585	0.4360	-0.277	-0.046	-0.266	0.355	0.373
0.5461	0.9282	0.4046	-0.268	-0.0502	-0.328	0.350	0.364
0.6435	0.8994	0.3783	-0.239	-0.053	-0.421	0.340	0.351
0.7302	0.8721	0.3591	-0.197	-0.051	-0.519	0.330	0.337
0.8081	0.8465	0.3444	-0.160	-0.047	-0.660	0.314	0.315
0.8783	0.8221	0.3380	-0.111	-0.036	-0.782	0.301	0.298
0.9420	0.7989	0.3366	-0.062	-0.022	-0.982	0.277	0.266
1	0.7768	0.3443	0	0	0	0	0

Contd.

Studies on Thermodynamic andTechniques

Acetonitrile + 1,3-Dioxolane

298.15 K

0	0	0	0	0	0	0	0
0.1670	0.1670	0.1670	0.1670	0.1670	0.1670	0.1670	0.1670
0.3109	0.9792	0.4235	-0.295	-0.027	-0.174	0.333	0.349
0.4361	0.9478	0.3913	-0.352	-0.034	-0.221	0.329	0.343
0.5461	0.9177	0.3646	-0.357	-0.034	-0.284	0.323	0.334
0.6435	0.8890	0.3419	-0.334	-0.042	-0.377	0.312	0.321
0.7302	0.8619	0.3251	-0.297	-0.041	-0.477	0.302	0.307
0.8081	0.8361	0.3134	-0.248	-0.038	-0.593	0.290	0.292
0.8783	0.8116	0.3077	-0.190	-0.029	-0.706	0.279	0.277
0.9420	0.7882	0.3070	-0.114	-0.017	-0.845	0.264	0.257
1	0.7654	0.3124	0	0	0	0	0

Acetonitrile + 1,3-Dioxolane

318.15 K

0	1.0344	0.4580	0	0	0	0	0
0.1670	1.0003	0.4132	-0.202	-0.017	-0.188	0.297	0.313
0.3109	0.9679	0.3791	-0.336	-0.026	-0.215	0.298	0.311
0.4361	0.9366	0.3502	-0.390	-0.034	-0.275	0.292	0.304
0.5461	0.9065	0.3263	-0.392	-0.039	-0.354	0.284	0.293
0.6435	0.8780	0.3065	-0.369	-0.043	-0.460	0.274	0.280
0.7302	0.8511	0.2910	-0.342	-0.044	-0.597	0.260	0.262
0.8081	0.8255	0.2817	-0.290	-0.040	-0.737	0.246	0.245
0.8783	0.8009	0.2780	-0.219	-0.032	-0.890	0.231	0.225
0.9420	0.7775	0.2797	-0.128	-0.019	-1.093	0.210	0.198
1	0.7547	0.2891	0	0	0	0	0

Acetonitrile + 1,4-Dioxane

298.15 K

0	1.0287	1.1780	0	0	0	0	0
0.1926	0.9976	0.9468	-0.100	-0.071	0.119	0.53	0.41

Contd.

Studies on Thermodynamic andTechniques

0.3492	0.9686	0.7720	-0.193	-0.115	0.031	0.51	0.40
0.4791	0.9410	0.6349	-0.258	-0.144	-0.115	0.47	0.38
0.5886	0.9146	0.5315	-0.289	-0.156	-0.297	0.44	0.36
0.6822	0.8891	0.4573	-0.275	-0.152	-0.494	0.41	0.35
0.7630	0.8645	0.4099	-0.237	-0.132	-0.648	0.40	0.35
0.8336	0.8412	0.3679	-0.191	-0.115	-0.998	0.35	0.32
0.8957	0.8185	0.3482	-0.115	-0.0831	-1.253	0.32	0.31
0.9508	0.7972	0.3389	-0.057	-0.0464	-1.632	0.27	0.28
1	0.7768	0.3443	0	0	0	0	0

Acetonitrile + 1,4-Dioxane

308.15 K

0	1.0168	0.9985	0	0	0	0	0
0.1926	0.9812	0.8244	-0.194	-0.042	0.207	0.432	0.520
0.3492	0.9484	0.6862	-0.288	-0.073	0.135	0.415	0.495
0.4791	0.9187	0.5802	-0.342	-0.090	0.055	0.405	0.475
0.5886	0.8914	0.5030	-0.357	-0.092	-0.007	0.405	0.466
0.6822	0.8661	0.4477	-0.331	-0.083	-0.044	0.412	0.464
0.7630	0.8430	0.4079	-0.301	-0.067	-0.048	0.424	0.469
0.8336	0.8212	0.3775	-0.229	-0.049	-0.029	0.438	0.478
0.8957	0.8014	0.3531	-0.168	-0.031	0.014	0.453	0.490
0.9508	0.7828	0.3340	-0.090	-0.012	0.207	0.486	0.525
1	0.7654	0.3124	0	0	0	0	0

Acetonitrile + 1,4-Dioxane

318.15 K

0	1.0047	0.8909	0	0		0	0
0.1926	0.9755	0.7189	-0.254	-0.056	0.015	0.322	0.409
0.3492	0.9468	0.5994	-0.364	-0.081	-0.014	0.337	0.411
0.4791	0.9194	0.5014	-0.430	-0.101	-0.143	0.324	0.387
0.5886	0.8930	0.4305	-0.450	-0.106	-0.268	0.318	0.370

Contd.

Studies on Thermodynamic andTechniques

0.6822	0.8675	0.3854	-0.421	-0.095	-0.324	0.328	0.371
0.7630	0.8430	0.3542	-0.364	-0.078	-0.352	0.341	0.375
0.8336	0.8193	0.3354	-0.276	-0.054	-0.279	0.366	0.395
0.8957	0.7968	0.3194	-0.188	-0.033	-0.190	0.389	0.416
0.9508	0.7752	0.3076	-0.093	-0.011	0.142	0.438	0.471
1	0.7547	0.2891	0	0		0	0

TABLE 3.

 Values of Ultrasonic Speeds u , isentropic compressibility K_s , Deviations in isentropic compressibility ΔK_s for 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	ΔL_f / Å	ΔZ / (Kg·m ² ·s ⁻¹)
Acetonitrile + Tetrahydrofuran					
0	1292.2	679.94	0	0	0
0.1633	1344.3	635.81	-4.72	-0.0004	0.3119
0.3052	1395.2	597.70	-8.16	-0.0013	2.2063
0.4295	1443.9	565.25	-10.64	-0.0020	4.3811
0.5394	1490.4	537.47	-12.11	-0.0025	6.2800
0.6372	1533.8	514.14	-11.73	-0.0027	7.2298
0.7249	1573.6	494.81	-9.96	-0.0023	6.5553
0.8039	1610.3	478.56	-7.36	-0.0015	4.5467
0.8754	1645.5	464.04	-4.62	-0.0008	2.8429
0.9405	1679.0	451.20	-1.75	-0.0001	0.6821
1	1713.2	438.59	0	0	0
Acetonitrile + 1,3-Dioxolane					
0	1338.2	527.95	0	0	0
0.1670	1388.9	506.89	-6.14	-0.0006	2.7010
0.3109	1436.2	489.75	-10.42	-0.0013	8.2001
0.4361	1481.5	475.34	-13.65	-0.0024	15.5741
0.5461	1524.4	463.62	-15.54	-0.0039	25.0101
0.6435	1563.5	454.83	-15.63	-0.0048	29.1401
0.7302	1597.8	449.15	-13.56	-0.0045	27.6649
0.8081	1628.8	445.28	-10.47	-0.0037	23.1053
0.8783	1658.2	442.39	-7.09	-0.0026	16.1342
0.9420	1686.4	440.14	-3.65	-0.0015	8.6832
1	1713.2	438.61	0	0	0
Acetonitrile + 1,4-Dioxane					
0	1344.4	537.84	0	0	0

Contd.

Studies on Thermodynamic andTechniques

0.1926	1399.3	511.94	-6.79	-0.0007	3.7000
0.3492	1451.0	490.37	-12.82	-0.0015	9.1001
0.4791	1498.4	473.32	-16.98	-0.0025	15.5741
0.5886	1540.2	460.91	-18.52	-0.0036	25.0101
0.6822	1575.7	453.00	-17.15	-0.0045	29.1401
0.7630	1606.5	448.20	-13.93	-0.0042	27.6649
0.8336	1633.5	445.52	-9.60	-0.0034	23.1053
0.8957	1660.1	443.32	-5.64	-0.0023	16.1342
0.9508	1686.3	441.13	-2.36	-0.0012	8.6832
1	1713.2	438.61	0	0	0

TABLE 4.

Vander Wall's Constant b , Molecular Radius r , Geometrical Volume B , Collision Factor S , Molar Speed of Sound R , Available Volume V_a , Intermolecular Free Length L_f , Molar Volume at Absolute Zero V_0 , Molar Surface Area Y and Specific Acoustic Impedance Z of the Pure Components at 298.15 K.

Pure Solvent	$b \times 10^5$ /m ³	r /nm	$B \times 10^5$ /(m ³ ·mol ⁻¹)	S	$R \times 10^6$ /{m ³ ·mol ⁻¹ (m·s ⁻¹) ^{1/3} }	$V_a \times 10^5$ /(cu·m)	L_f /Å	$V_0 \times 10^5$ /(cu·m)	$Y \times 10^4$ /Å	$Z \times 10^{-3}$ /(Kg·m ² ·s ⁻¹)
Acetonitrile	4.94	0.170	1.24	4.58	632.32	-3.74	0.431	5.658	21.82	1330.81
Tetrahydrofuran	7.66	0.197	1.91	3.45	891.72	1.57	0.536	6.612	29.22	1138.17
1,3-Dioxolane	6.57	0.187	1.64	3.57	771.81	1.15	0.473	5.858	26.38	1415.41
1,4-Dioxane	8.07	0.200	2.02	3.57	945.32	1.37	0.477	7.197	30.26	1382.98

TABLE 5.

Intermolecular Free Length L_f , Molar Speed of Sound R , Relative association R_A ,
Molecular association M_A , Available Volume V_a , Specific Acoustic Impedance Z of
Binary Mixtures at 298.15 K

x_1	L_f / Å	$R \times 10^6$ /{ $m^3 \cdot mol^{-1}$ ($m \cdot s^{-1}$) ^{1/3} }	R_A	M_A	$V_a \times 10^5$ / (cu·m)	$Z \times 10^{-3}$ /(Kg·m ² ·s ⁻¹)
Acetonitrile + Tetrahydrofuran						
0.1633	0.5186	911.85	1.2147	-0.5911	1.18	1169.94
0.3052	0.5029	842.69	1.1848	-0.634	8.72	1199.17
0.4295	0.4890	791.88	1.1565	-0.6681	6.26	1225.29
0.5394	0.4768	753.03	1.1295	-0.6961	4.19	1248.36
0.6372	0.4664	722.31	1.1043	-0.7193	2.43	1268.15
0.7249	0.4575	697.28	1.0809	-0.7389	9.41	1284.37
0.8039	0.4500	676.55	1.059	-0.7558	-3.58	1297.58
0.8754	0.4431	659.27	1.0385	-0.7703	-1.55	1309.65
0.9405	0.4369	644.61	1.0189	-0.7831	-2.65	1320.03
Acetonitrile + 1,3-Dioxolane						
0.1670	0.4654	937.98	1.3773	-0.4928	8.64	1403.98
0.3109	0.4587	860.66	1.3224	-0.5567	6.26	1397.31
0.4361	0.4523	804.77	1.2715	-0.6074	4.32	1394.09
0.5461	0.4462	762.5	1.2241	-0.6487	2.66	1394.22
0.6435	0.4412	729.26	1.1801	-0.6831	1.25	1390.11
0.7302	0.4378	702.28	1.1391	-0.7124	7.42	1381.30
0.8081	0.4354	679.94	1.1013	-0.7373	-9.61	1370.15
0.8783	0.4335	661.37	1.0652	-0.7591	-1.93	1357.24
0.9420	0.4319	645.71	1.0317	-0.7779	-2.85	1344.40
Acetonitrile + 1,4-Dioxane						
0.1926	0.4654	1039.22	1.3739	-0.5033	9.08	1395.94
0.3492	0.4555	917.01	1.3179	-0.5677	6.05	1405.44

Contd.

Studies on Thermodynamic andTechniques

0.4791	0.4475	837.8	1.2667	-0.6173	3.84	1409.99
0.6822	0.4378	740.65	1.1769	-0.6897	8.45	1400.95
0.7630	0.4354	708.61	1.1370	-0.7173	-2.21	1388.82
0.8336	0.4341	683.07	1.1002	-0.7407	-1.12	1374.1
0.8957	0.4331	662.64	1.0648	-0.7611	-2.00	1358.79
0.9508	0.4320	645.97	1.0317	-0.7788	-2.85	1344.32

TABLE 6

Redlich-Kister coefficients a_n and standard deviations σ for the binary mixtures

Binary mixture	Excess property	T/K	a_0	a_1	a_2	a_3	σ	
Acetonitrile + Tetrahydrofuran	$V^E \times 10^6$ / ($m^3 \cdot mol^{-1}$)	298.15	-0.535	0.439	0.238	-0.157	0.004	
		308.15	-0.435	0.081	0.000	-0.046	0.000	
		318.15	-1.198	0.489	0.054	-0.116	0.001	
	$\Delta\eta$ / ($mPa \cdot s$)	298.15	-0.023	-0.023	-0.014	-	0.000	
		308.15	-0.041	-0.044	-	-	0.001	
		318.15	-0.060	0.047	-0.023	-	0.000	
	$\Delta K_S \times 10^{12}$ / (Pa^{-1})	298.15	-46.921	-20.28	16.041	28.30	0.101	
		298.15	-0.001	-0.0096	0.008	0.0125	0.000	
		298.15	22.952	33.3804	-8.691	-31.4791	0.195	
	Acetonitrile + 1,3- Dioxolane.	$V^E \times 10^6$ / ($m^3 \cdot mol^{-1}$)	298.15	-1.104	0.187	0.333	-0.650	0.002
			308.15	-1.431	-0.288	-	-	0.014
			318.15	-1.580	-0.014	-0.254	-0.777	0.002
		$\Delta\eta$ / ($mPa \cdot s$)	298.15	-0.195	-0.080	-0.108	-0.059	0.001
			308.15	-0.141	-0.090	-0.090	-	0.002
			318.15	-0.148	-0.105	-0.111	-0.022	0.000
$\Delta K_S \times 10^{12}$ / (Pa^{-1})		298.15	-59.821	-34.96	6.201	66.24	0.063	
		298.15	-0.013	0.015	-	-	0.001	
		298.15	86.912	106.635	-	-	1.712	

Contd.

Studies on Thermodynamic andTechniques

Acetonitrile + 1,4-Dioxane	$V^E \times 10^6$ / (m ³ ·mol ⁻¹)	298.15	-1.069	-0.714	0.219	0.424	0.005
		308.15	-1.380	-0.368	-0.180	-	0.006
		318.15	-1.771	-0.377	-	-	0.009
	$\Delta\eta$ / (mPa·s)	298.15	-0.583	-0.279	-0.115	-	0.003
		308.15	-0.365	-0.112	0.131	0.102	0.001
		318.15	-0.408	-0.168	0.066	0.332	0.001
	$\Delta K_S \times 10^{12}$ / (Pa ⁻¹)	298.15	-70.221	-41.531	31.130	46.671	0.101
	ΔL_f / Å	298.15	-0.031	-0.021	0.014	0.023	0.001
	ΔZ / (Kg· m ² ·s ⁻¹)	298.15	213.983	113.683	-36.983	-77.647	0.163

Table 7

Parameters of McAllister Model, Heric and Brewer Parameters and Standard Deviations for Kinematic Viscosities at Various Temperatures.

Binary System	T/K	McAllister (three body model)			McAllister (four body model)				Heric and Brewer parameter			
		ν_{12}	ν_{21}	σ	ν_{1112}	ν_{1122}	ν_{2221}	σ	$a \times 10^2$ / $\text{cm}^2 \text{s}^{-1}$	$b \times 10^2$ / $\text{cm}^2 \text{s}^{-1}$	$c \times 10^2$ / $\text{cm}^2 \text{s}^{-1}$	σ
Acetonitrile + Tetrahydrofuran	298.15	0.4472	0.4953	0.004	0.4422	0.4809	0.3976	0.001	-0.1092	-0.0851	-0.1719	0.009
	308.15	0.3992	0.4621	0.004	0.3961	0.4358	0.4554	0.001	-0.6990	-0.0797	-0.0048	0.002
	318.15	0.3481	0.4175	0.015	0.3499	0.3967	0.1921	0.003	-0.2939	-0.2417	-0.4706	0.025
Acetonitrile + 1,3-Dioxolane	298.15	0.3181	0.4772	0.025	0.3339	0.4365	0.0950	0.011	-0.7160	-0.5678	-1.2469	0.063
	308.15	0.3028	0.4280	0.021	0.3204	0.4244	0.2383	0.005	-0.4759	-0.2327	-0.7290	0.026
	318.15	0.2541	0.3887	0.023	0.2739	0.3861	0.1297	0.006	-0.4963	-0.4096	-0.9001	0.042

Contd.

Acetonitrile + 1,4-Dioxane	298.15	0.3131	0.8765	0.054	0.2817	0.5868	0.0175	0.025	-1.8590	-2.0151	-2.7610	0.187
	308.15	0.4955	0.0428	0.031	0.4467	0.2183	0.8251	0.025	-0.5120	-3.4702	-2.8854	0.154
	318.15	0.3815	0.2167	0.046	0.3034	0.0921	0.6748	0.046	-0.7564	-5.1889	-4.3310	0.230

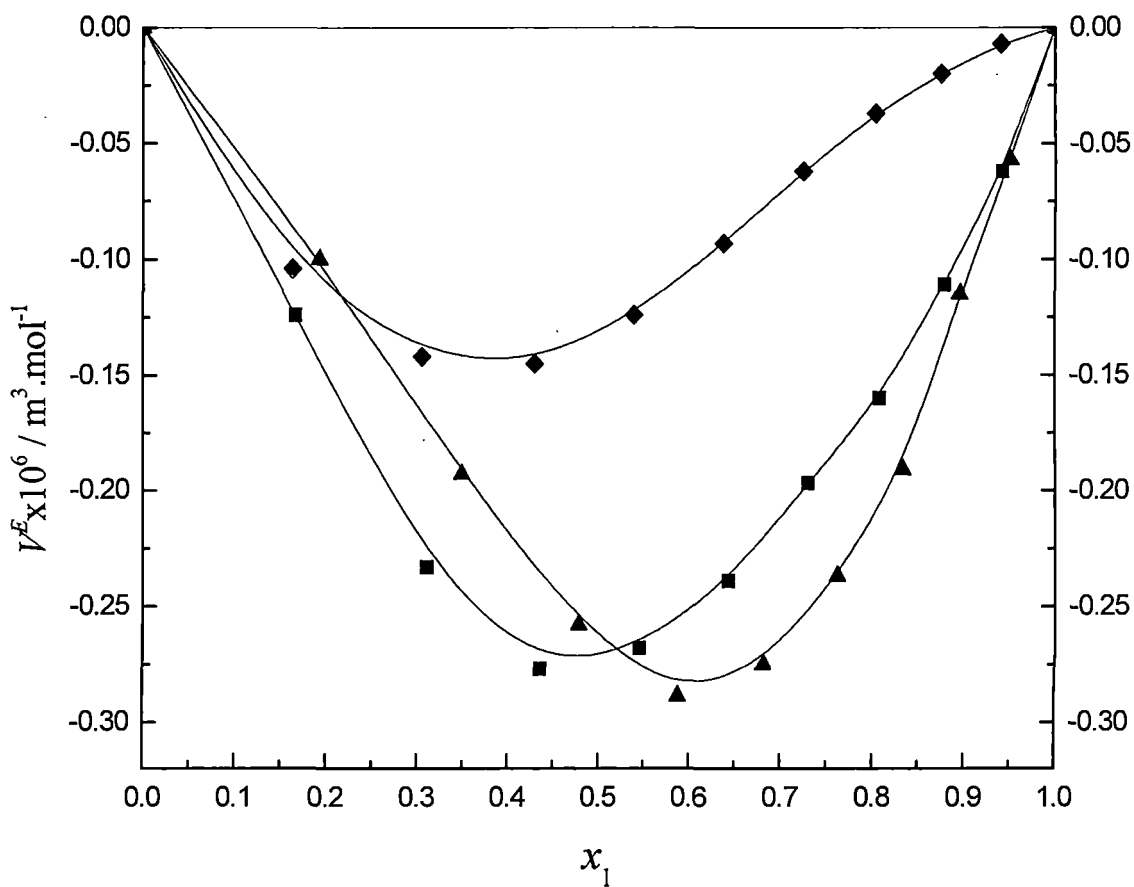


Figure1. Excess molar volumes (V^E) for binary mixtures of Acetonitrile (1) with \blacklozenge , Tetrahydrofuran; \blacksquare , 1,3-Dioxolane; \blacktriangle , 1,4-Dioxane at 298.15 K.

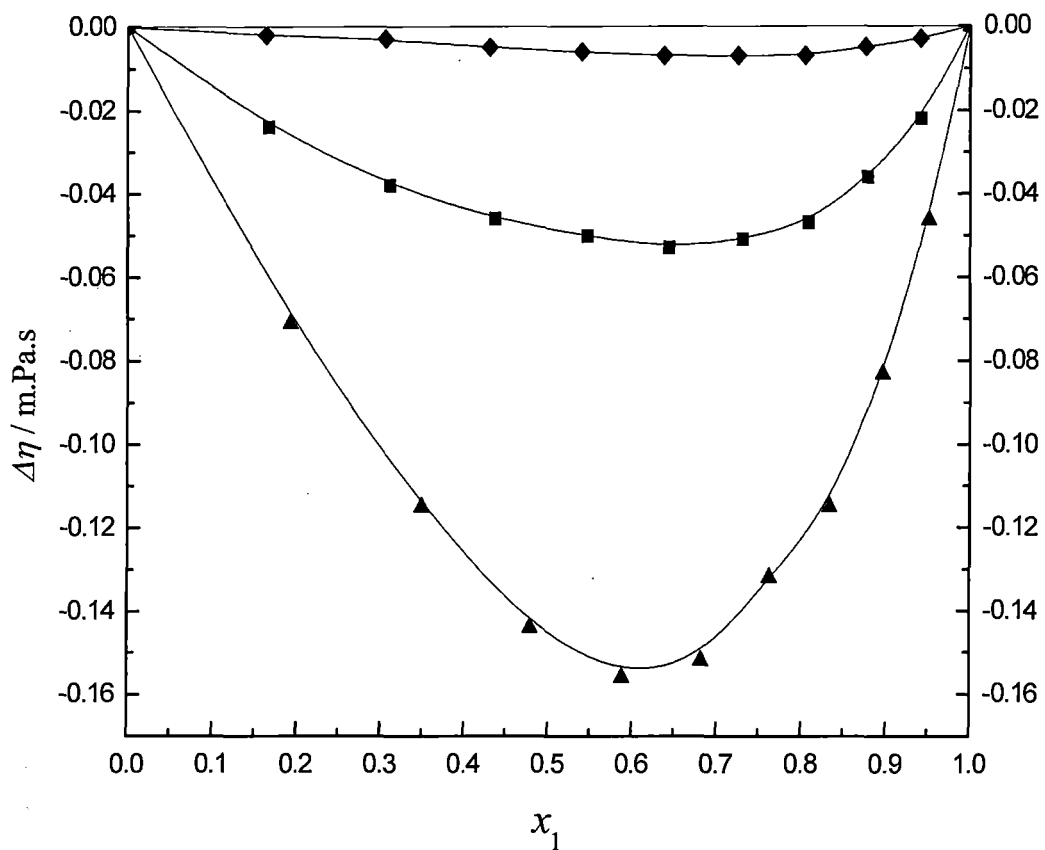


Figure 2. Viscosity deviations ($\Delta\eta$) for binary mixtures of Acetonitrile (1) with \blacklozenge , Tetrahydrofuran; \blacksquare , 1,3-Dioxolane; \blacktriangle , 1,4-Dioxane at 298.15 K.

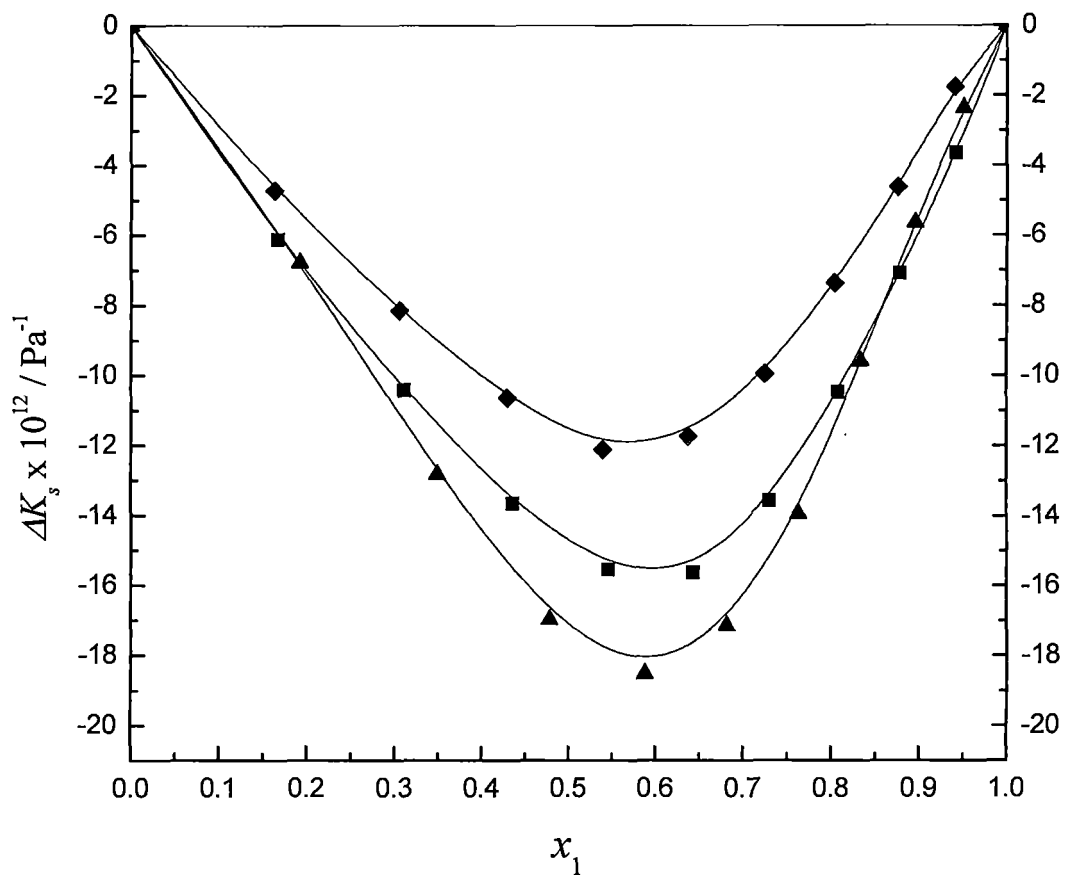


Figure3.

Deviations in isentropic compressibility (ΔK_s) for binary mixtures of Acetonitrile (1) with ♦, Tetrahydrofuran; ■, 1,3-Dioxolane; ▲, 1,4-Dioxane at 298.15 K.

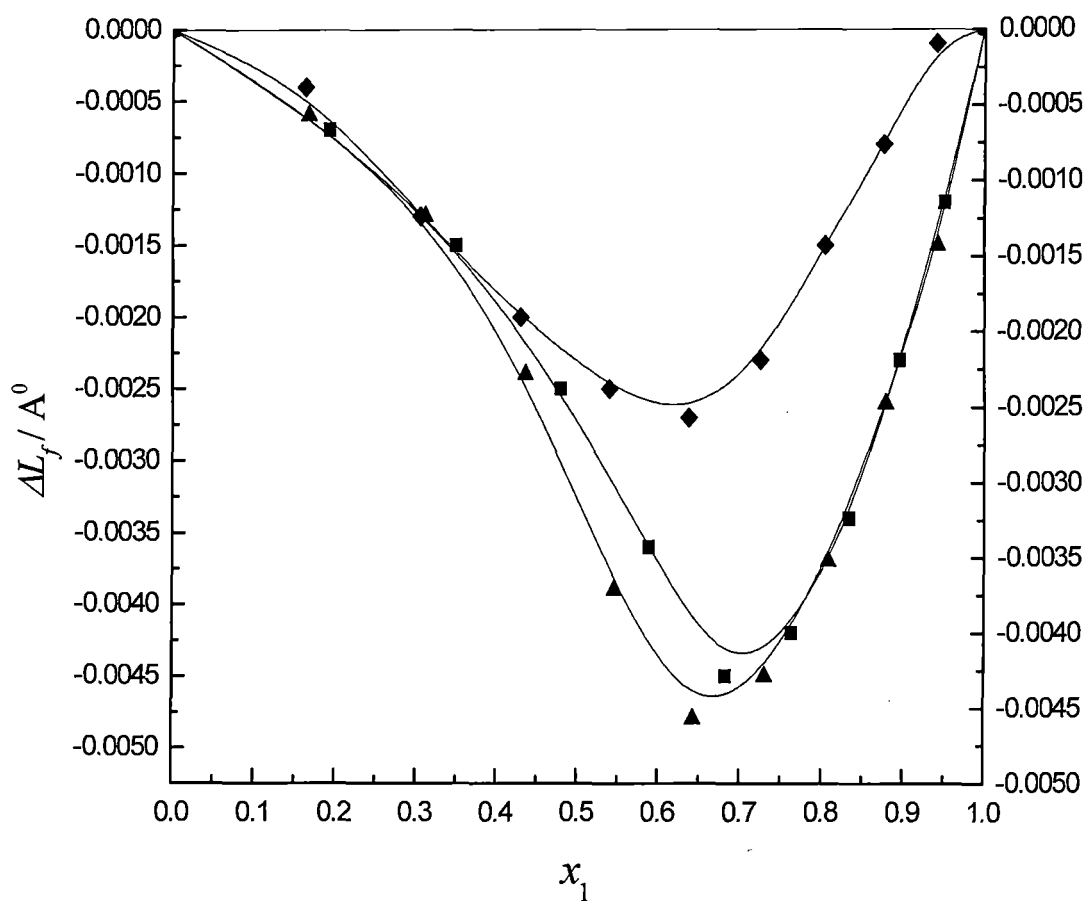


Figure4. Deviations in intermolecular free length (ΔL_f) for binary mixtures of Acetonitrile (1) with◆, Tetrahydrofuran; ■, 1,3-Dioxolane; ▲, 1,4-Dioxane at 298.15 K.

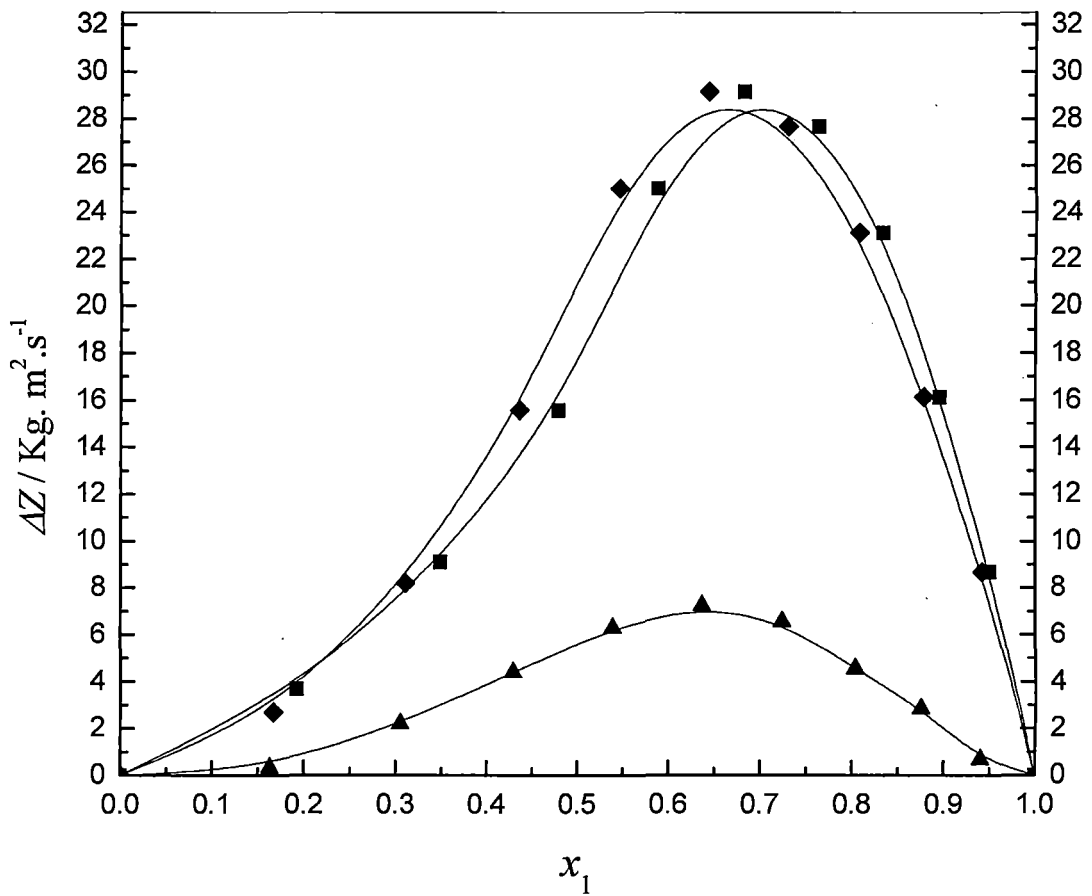


Figure5. Deviations in specific acoustic impedance (ΔZ) for binary mixtures of Acetonitrile (1) with♦, Tetrahydrofuran; ■, 1,3-Dioxolane; ▲, 1,4-Dioxane at 298.15 K.

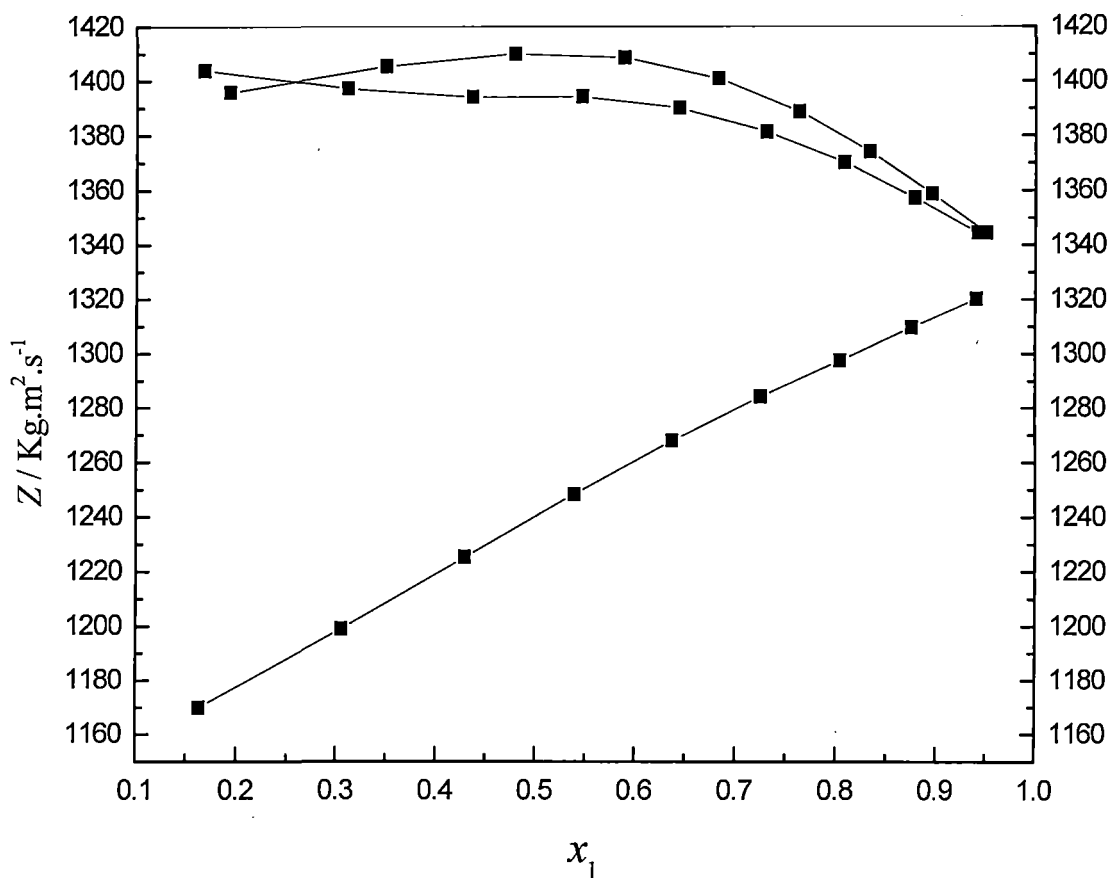


Figure 6. Z values for binary mixtures of Acetonitrile (1) with ♦, Tetrahydrofuran; ■, 1,3-Dioxolane; ▲, 1,4-Dioxane at 298.15 K.

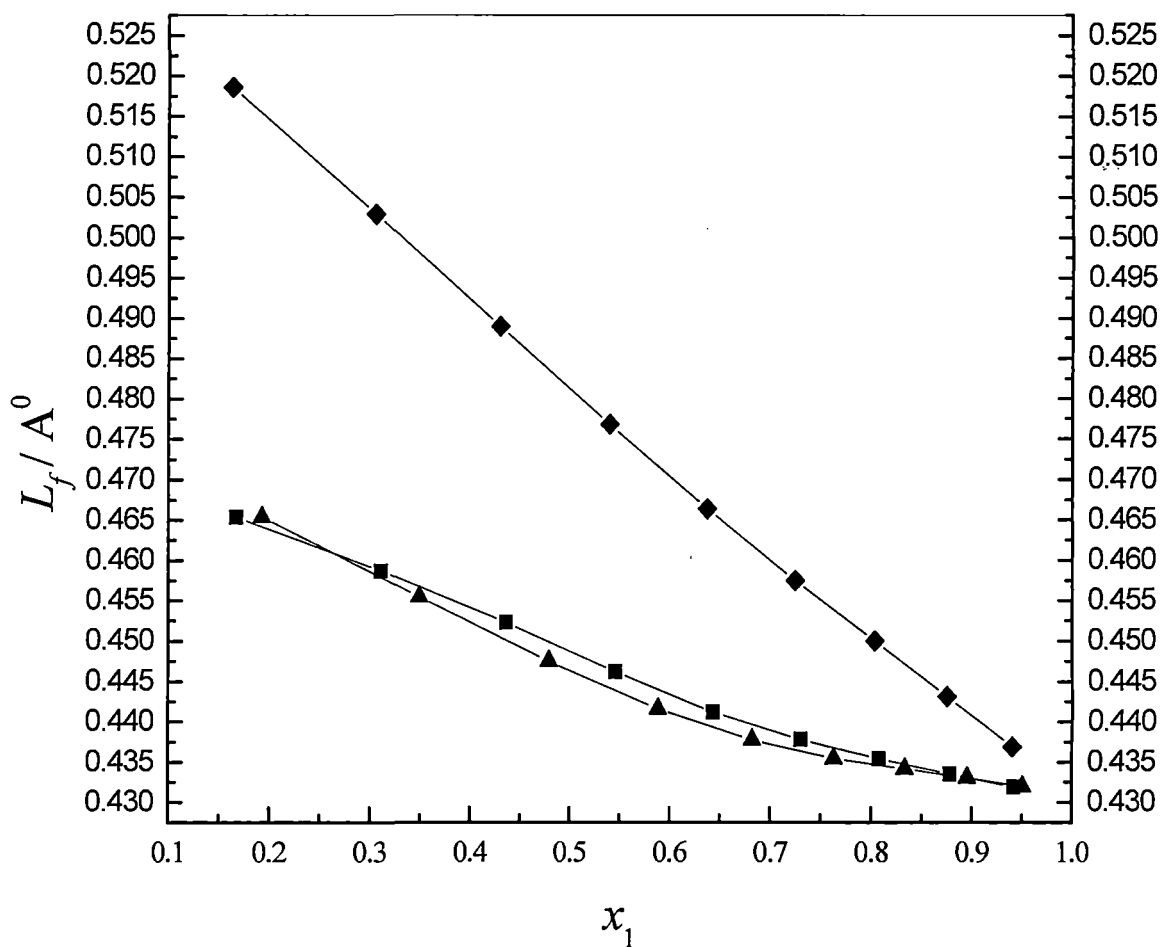


Figure7. L_f values for binary mixtures of Acetonitrile (1) with \diamond , Tetrahydrofuran; \square , 1,3-Dioxolane; \blacktriangle , 1,4-Dioxane at 298.15 K.

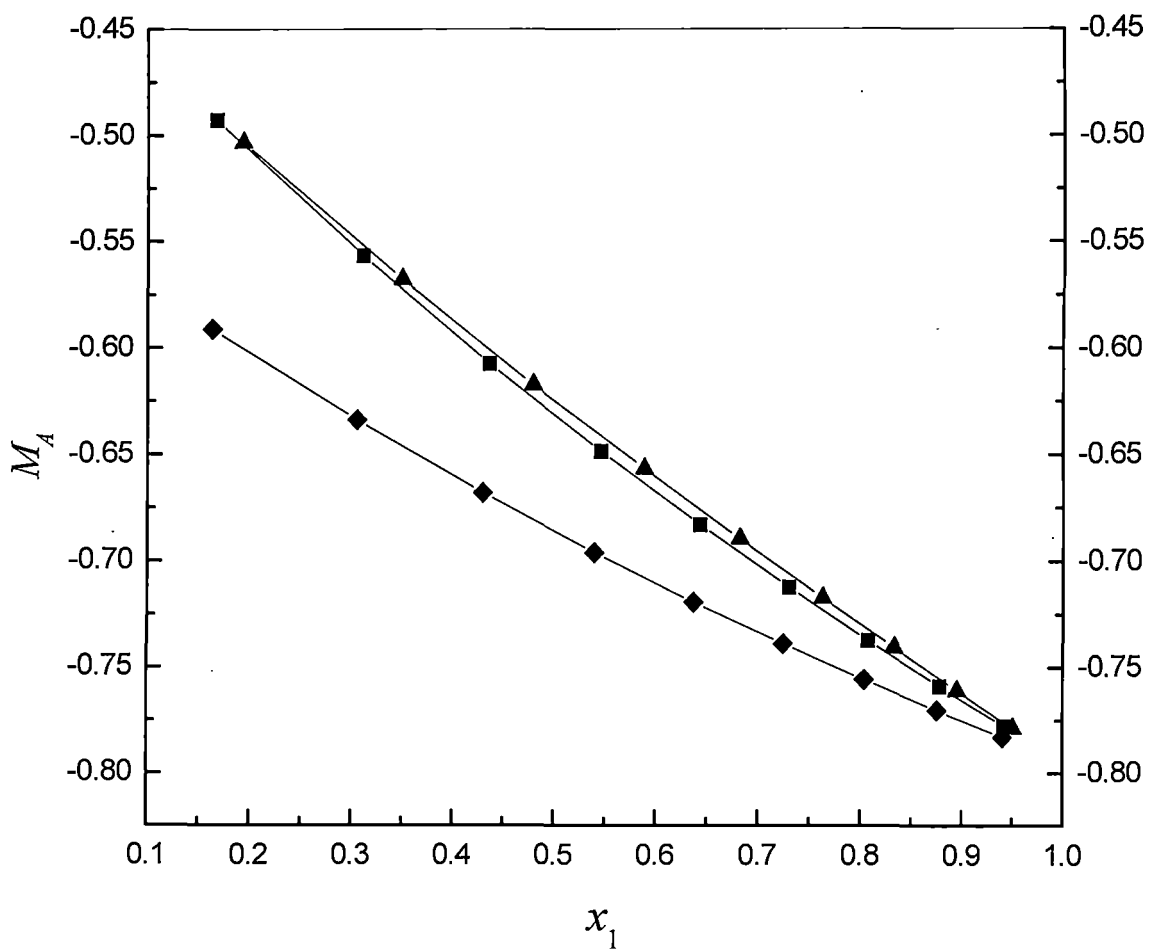


Figure9. M_A values for binary mixtures of Acetonitrile (1) with \diamond , Tetrahydrofuran; \blacksquare , 1,3-Dioxolane; \blacktriangle , 1,4-Dioxane at 298.15 K.