

Volumetric and Thermodynamic Studies of Molecular Interactions in Ternary Liquid Mixtures at 303, 308 and 313K

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Abstract: *Ultrasonic velocity, density and viscosity were measured for mixtures of 1-alkanols, namely, 1-propanol, 1-butanol, 1-pentanol and 1-hexanol, with N, N-dimethylformamide (DMF) in cyclohexanone at 303, 308 and 313K. The experimental data were used to calculate the excess free volume (V_f^E), excess internal pressure (π_i^E), and Gibb's free energy (ΔG^*), which were discussed in the light of molecular interaction existing in the mixtures. It was observed that the addition of DMF to mixtures caused the dissociation of the hydrogen-bonded structure of 1-alkanols. Furthermore, the DMF-alkanol interactions were weaker than the alkanol-ketone interactions in the mixtures.*

Keywords: alkanols, ultrasonic velocity, Gibb's free energy, excess free volume

Abstrak: *Halaju ultrasonik, ketumpatan dan kelikatan telah diukur bagi 1-alkanol, iaitu 1-propanol 1, 1-butanol, 1-pentanol dan 1-hexanol, dengan dimetilformamida (DMF) dalam Cyclohexanone pada 303, 308 dan 313 K. Data-data dari eksperimen telah digunakan untuk menghitung isipadu bebas berlebihan (V_f^E) tekanan dalaman berlebihan (π_i^E), dan tenaga bebas Gibb (ΔG^*), yang telah didiskusikan dalam interaksi molekular yang wujud di dalam 'mixtures'. Telah diperhatikan bahawa penambahan DMF ke dalam 'mixtures' telah mengakibatkan pelebaran ikatan hidrogen struktur 1-alkanols. Interaksi DMF-alkanol lebih lemah daripada interaksi alkanol-ketone.*

Kata kunci: alkohol, halatuju ultrasonik, tenaga bebas Gibb, isipadu bebas lebihan

1. INTRODUCTION

In recent years, the measurement of ultrasonic velocity has been successfully employed in understanding the nature of molecular interactions in pure liquids and liquid mixtures. Ultrasonic velocity measurements are highly sensitive to molecular interactions and can be used to provide qualitative information about the physical nature and strength of molecular interaction in liquid mixtures.¹⁻³ The ultrasonic velocity of a liquid is fundamentally related to the binding forces between atoms or molecules, and has been successfully employed in understanding the nature of molecular interactions in pure liquids and binary and ternary mixtures.⁴⁻⁶ Variations in ultrasonic velocity and related parameters have shed much light upon the structural changes associated with

liquid mixtures of weakly⁷ or strongly interacting components.⁸ The study of molecular associations in organic ternary mixtures having an alcohol as one component is of particular interest since alcohols are strongly self-associated liquids with a three-dimensional network of hydrogen bonds⁹ and can be associated with any other group having some degree of polar attraction.¹⁰ A survey of the literature has shown that a few attempts have been made to obtain ultrasonic velocity data for ternary liquid mixtures.¹¹⁻¹³

However, no thermodynamic studies have been conducted for ternary mixtures of N,N-dimethylformamide (DMF), cyclohexanone and 1-alkanols. Hence, experimental studies were carried out by the authors to characterize N,N-dimethylformamide + cyclohexanone + 1-propanol, 1-butanol, 1-pentanol or 1-hexanol through ultrasonic velocity measurements at 303, 308 and 313K. The main purpose of this study is to characterize the molecular interactions in these systems and subsequently to determine the effect of the chain length of 1-alkanols.

2. EXPERIMENTAL

The chemicals used in the present work were analytical reagent (AR) and spectroscopic reagent (SR) grades with a minimum assay of 99.9%, obtained from Sd Fine Chemicals, (India) and E-merck, (Germany), without further purification. In all systems, the various concentrations of the ternary liquid mixtures were prepared in terms of mole fraction, out of which the mole fraction of the second component, cyclohexanone ($X_2 = 0.4$), was kept fixed while the mole fractions of the remaining two (X_1 and X_3) were varied from 0.0 to 0.6. The densities of pure liquids and liquid mixtures were determined using a specific gravity bottle via the relative measurement method with an accuracy of ± 0.1 mg (Model: SHIMADZU AX-200). An Ostwald's viscometer with 10 ml capacity was used for the viscosity measurement of pure liquids and liquid mixtures. The viscometer was calibrated with fresh conductivity water immersed in a water bath that was maintained at the experimental temperature. The flow time of water (t_w) and the flow time of solution (t_s) were measured with a digital stop clock with an accuracy of 0.01 s (Model: RACER HS-10W). An ultrasonic interferometer (Model: F81) supplied by M/s. Mittal Enterprises, New Delhi, with frequency of 3 MHz and overall accuracy of ± 2 ms⁻¹ was used for velocity measurement.

3. THEORY AND CALCULATION

3.1 Free Volume (V_f)

Suryanarayana et al.¹⁴ obtained a formula for free volume in terms of the ultrasonic velocity (U) and the viscosity of the liquid (η) as

$$V_f = \left(\frac{M_{eff} U}{K \eta} \right)^{3/2} \quad (1)$$

Where, M_{eff} is the effective molecular weight ($M_{eff} = \sum m_i x_i$, in which m_i and x_i are the molecular weight and the mole fraction of the individual constituents respectively) and K is a temperature-independent constant equal to 4.28×10^9 for all liquids.

3.2 Internal Pressure (π_i)

On the basis of statistical thermodynamics, Suryanarayana¹⁵ derived an expression for the determination of internal pressure through use of the concept of free volume:

$$\pi_i = bRT \left(\frac{K \eta}{U} \right)^{1/2} \left(\frac{\rho^{2/3}}{M_{eff}^{7/6}} \right) \quad (2)$$

Where, T is the absolute temperature, ρ is the density, and R is the gas constant. It is stated that in the case of liquid systems, including electrolytic solutions, there is no serious harm in assuming cubic packing and equating b to 2.

3.3 Gibb's Free Energy (ΔG*)

On the basis of Eyring rate process theory, the Gibb's Free Energy can be computed as

$$-\Delta G^* = -2.30KT \log \frac{h}{KT\tau} \quad (3)$$

Where, $\tau = \frac{4}{3} \eta \beta$, K is Boltzmann's constant, r is the relaxation time and h is Planck's constant.

3.4 Excess Parameters (A^E)

In order to study the non-ideality of the liquid mixtures, the difference between the values of the real mixture (A_{exp}) and those corresponding to an ideal mixture (A_{id}), namely the excess parameters (A^E) of some of the acoustic parameters, were computed using the equation

$$A^E = A_{\text{exp}} - A_{id} \quad (4)$$

where, $A_{id} = \sum_{i=1}^n A_i X_i$, A_i , are any acoustical parameters and X_i are the mole fractions of the liquid components.

4. RESULTS AND DISCUSSION

The experimentally determined values of the density (ρ), viscosity (η) and ultrasonic velocity (U) for all of the pure liquids at 303, 308, and 313K are presented in Table 1, and the same values for the ternary systems (I to IV) are listed in Table 2. The excess values of free volume (V_f^E), excess internal pressure (π_i^E) and Gibbs Free Energy (ΔG^*) have been evaluated and are presented in Table 3.

Table 1: Density (ρ), viscosity (η) and ultrasonic velocity (U) of pure liquids at 303, 308 and 313K.

Liquids	$\rho/(\text{kgm}^{-3})$			$\eta/(\times 10^{-3} \text{ Nsm}^{-2})$			$U/(\text{ms}^{-1})$		
	Temperature (K)								
	303	308	313	303	308	313	303	308	313
N-N dimethyl formamide (DMF)	947.6	942.1	935.1	0.7679	0.7262	0.6797	1459.6	1434.7	1420.8
cyclohexanone	944.3	939.7	934.6	1.7571	1.6012	1.4559	1408.8	1362.8	1348.8
1-propanol	800.1	795.3	790.1	1.6111	1.4172	1.2581	1192.6	1181.0	1164.0
1-butanol	804.4	802.1	798.5	2.1502	1.8643	1.6308	1229.1	1211.0	1198.4
1-pentanol	807.2	801.5	798.1	2.7656	2.4088	2.0934	1253.2	1242.9	1218.9
1-hexanol	810.2	807.6	803.2	3.5130	3.1824	2.7804	1289.0	1272.5	1255.4

Table 2: The values of density (ρ), viscosity (η) and ultrasonic velocity (U) at 303, 308 and 313K.

X1	X3	$\rho/(\text{kgm}^{-3})$			$\eta/(\times 10^{-3} \text{ Nsm}^{-2})$			U/(ms ⁻¹)		
		303K	308K	313K	303K	308K	313K	303K	308K	313K
System I: 1-propanol+ Cyclohexanone + DMF										
0.0000	0.6004	943.8	942.2	937.5	1.0003	0.9353	0.8654	1454.6	1395.6	1389.9
0.0998	0.4995	924.8	923.3	921.9	1.0236	0.9489	0.8829	1405.2	1380.9	1362.6
0.1997	0.4002	915.1	913.4	909.8	1.0561	0.9814	0.9028	1373.4	1350.8	1339.9
0.2999	0.2999	906.6	902.6	896.8	1.0996	1.0120	0.9313	1355.4	1332.0	1320.0
0.3997	0.2005	895.4	890.7	886.5	1.1704	1.0715	0.9717	1328.7	1311.8	1293.8
0.4998	0.1003	882.5	877.2	872.9	1.2054	1.0859	0.9972	1314.8	1297.2	1287.4
0.6060	0.000	870.3	865.9	860.5	1.2708	1.1529	1.0425	1286.16	1280.58	1260.6
System II: 1-butanol + Cyclohexanone + DMF										
0.0000	0.6002	943.1	940.2	936.5	0.9995	0.9333	0.8645	1484.4	1464.0	1372.6
0.0997	0.4998	928.1	926.2	920.1	1.0601	0.9844	0.8979	1386.4	1365.9	1354.8
0.2003	0.3997	919.0	914.7	908.2	1.1147	1.0255	0.9431	1363.6	1344.6	1329.8
0.2996	0.3002	903.6	898.5	893.7	1.1705	1.0703	0.9899	1347.6	1323.2	1301.6
0.4066	0.2035	890.3	884.9	881.1	1.2580	1.1471	1.0370	1328.9	1310.6	1288.2
0.4996	0.1002	880.6	876.2	870.7	1.3584	1.2280	1.1153	1315.4	1302.6	1279.4
0.6000	0.000	863.9	862.3	858.3	1.4649	1.3193	1.1884	1296.3	1280.2	1264.8
System III: 1-pentanol+ Cyclohexanone + DMF										
0.0000	0.6003	944.3	940.8	933.4	0.9897	0.9229	0.8508	1404.6	1392.6	1380.0
0.0997	0.5002	927.4	923.5	915.9	1.0811	0.9922	0.9194	1385.7	1365.4	1353.5
0.1994	0.4000	909.9	907.5	903.5	1.1464	1.0598	0.9800	1363.4	1346.4	1333.8
0.2994	0.3002	898.3	894.0	889.7	1.2376	1.1277	1.0369	1347.9	1330.0	1317.2
0.3993	0.2008	886.3	879.3	877.7	1.3672	1.2426	1.1444	1331.8	1320.2	1298.6
0.5003	0.1001	875.8	869.2	865.9	1.5161	1.3705	1.2389	1321.5	1299.9	1286.2
0.6006	0.0000	863.9	858.7	853.1	1.6785	1.5143	1.3584	1309.6	1290.6	1272.9
System IV: 1-hexanol+ Cyclohexanone + DMF										
0.0000	0.6003	945.5	940.4	934.4	1.0020	0.9225	0.8625	1408.4	1392.8	1366.4
0.1061	0.4680	927.9	925.8	919.7	1.1146	1.0272	0.9445	1384.6	1361.4	1348.5
0.1995	0.4009	913.2	909.4	902.3	1.2474	1.1364	1.0141	1364.7	1345.6	1331.6
0.2998	0.3004	897.7	893.4	888.7	1.3849	1.2625	1.1485	1362.8	1335.9	1314.4
0.3997	0.1997	886.2	881.3	874.6	1.5652	1.4101	1.2716	1336.9	1318.9	1299.9
0.5001	0.0998	873.5	870.5	865.5	1.7898	1.6064	1.4380	1325.2	1309.5	1296.4
0.5994	0.000	861.9	859.6	854.2	2.0098	1.7971	1.6077	1320.9	1301.1	1284.9

Table3: The values of excess free volume (V_f^E), excess internal pressure (π_i^E) and Gibb's free energy (ΔG^*) at 303, 308 and 313K.

X1	X3	$V_f^E / (\times 10^{-7} \text{m}^3 \text{mol}^{-1})$			$\pi_i^E / (\times 10^6 \text{Nm}^{-2})$			$\Delta G^* / (\times 10^{-20} \text{KJ mol}^{-1})$		
		303K	308K	313K	303K	308K	313K	303K	308K	313K
System I: 1-propanol+ Cyclohexanone + DMF										
0.0000	0.6004	0.0741	0.0345	0.0493	-022	-021	-015	0.2609	0.2598	0.2581
0.0998	0.4995	0.0841	0.1022	0.0972	-045	-042	-035	0.2793	0.2748	0.2718
0.1997	0.4002	0.0873	0.1081	0.1362	-061	-056	-053	0.2973	0.2962	0.2941
0.2999	0.2999	0.1192	0.1497	0.2118	-080	-073	-073	0.3115	0.3112	0.2927
0.3997	0.2005	0.1197	0.1545	0.1871	-089	-083	-078	0.3319	0.3298	0.3259
0.4998	0.1003	0.1945	0.2474	0.2713	-110	-105	-097	0.3438	0.3410	0.3380
0.6060	0.0000	0.2386	0.2829	0.3131	-128	-121	-112	0.3639	0.3575	0.3545
System II: 1-butanol + Cyclohexanone + DMF										
0.0000	0.6002	0.1230	0.1601	0.0402	-027	-028	-022	0.2537	0.2518	0.2505
0.0997	0.4998	0.0114	0.0394	0.0859	-031	-030	-031	0.2922	0.2920	0.2868
0.2003	0.3997	0.0444	0.2575	0.1021	-047	-046	-043	0.3089	0.3077	0.3055
0.2996	0.3002	0.0990	0.1371	0.1294	-067	-058	-054	0.3252	0.3248	0.3239
0.4066	0.2035	0.1215	0.1598	0.1880	-079	-074	-068	0.3461	0.3440	0.3409
0.4996	0.1002	0.1666	0.2137	0.2289	-090	-087	-076	0.3657	0.3608	0.3593
0.6000	0.0000	0.2181	0.2559	0.2860	-101	-093	-085	0.3882	0.3833	0.3783
System III: 1-pentanol+ Cyclohexanone + DMF										
0.0000	0.6003	0.0213	0.0649	0.0958	-015	-018	-019	0.2717	0.2701	0.2671
0.0997	0.5002	0.0068	0.0595	0.0702	-029	-031	-030	0.2959	0.2942	0.2925
0.1994	0.4000	0.0584	0.0945	0.1052	-050	-047	-042	0.3159	0.3148	0.3126
0.2994	0.3002	0.0967	0.1497	0.1698	-065	-062	-056	0.3363	0.3335	0.3307
0.3993	0.2008	0.1131	0.1643	0.1630	-074	-072	-062	0.3612	0.3572	0.3571
0.5003	0.1001	0.1503	0.1867	0.2183	-083	-077	-070	0.3849	0.3832	0.3781
0.6006	0.0000	0.2023	0.2341	0.2656	-091	-083	-077	0.4092	0.4065	0.4021
System IV: 1-hexanol+ Cyclohexanone + DMF										
0.0000	0.6003	0.0003	0.0661	0.0342	-013	-018	-014	0.2727	0.2700	0.2632
0.1061	0.4680	0.0367	0.0791	0.1066	-034	-036	-035	0.3016	0.3012	0.2982
0.1995	0.4009	0.0248	0.0309	0.1077	-038	-043	-046	0.3303	0.3274	0.3198
0.2998	0.3004	0.0216	0.0599	0.0785	-055	-056	-052	0.3529	0.3528	0.3509
0.3997	0.1997	0.0335	0.0877	0.1170	-060	-066	-061	0.3846	0.3805	0.3772
0.5001	0.0998	0.0647	0.1140	0.1513	-066	-070	-067	0.4146	0.4095	0.4033
0.5994	0.0000	0.1356	0.1795	0.2903	-075	-077	-070	0.4392	0.4349	0.4300

In all of the mixtures, the density and the ultrasonic velocity decreased with increasing mole fractions of 1-alkanol, as well as with temperature. However, the value of viscosity increased with increasing concentrations of 1-alkanols and decreased with increasing temperature. As the number of hydrocarbon groups or the chain-length of the alcohol increased, a gradual decrease in sound velocity was observed. This behaviour at these concentrations is different from the behaviour of ideal mixtures, and can be attributed to intermolecular interactions in the systems studied.¹⁶

N-N-dimethyl formamide (DMF), as a polar solvent, is certainly to some extent associated by dipole-dipole interactions, and is of particular interest because of the absence of any significant structural effects due to the lack of hydrogen bonds; therefore, it may work as an aprotic, protophilic solvent with a large dipole moment and high dielectric constant ($\mu = 3.24\text{D}$ and $\epsilon = 36.71$). On the other hand, alkanols are polar liquids strongly associated with hydrogen bonding, with an extent of polymerisation that may differ depending on temperature, chain length and position of the OH group. Due to the polar natures of DMF, cyclohexanone and alcohols, dipole-dipole interactions were present in these mixtures. When the compounds were mixed, the changes that occur in association equilibria were evidently due to the rupture of the hydrogen bonds in pure cyclohexanone and 1-alkanols and DMF-DMF, dipole-dipole interactions, and the formation of $\text{O-H}\dots\text{C=O}$ and perhaps even $\text{O-H}\dots\text{N}(\text{CH}_3)_2$ hydrogen bonds between the components.

In order to understand the nature of the molecular interactions between the components of the liquid mixtures, it is of interest to discuss the same in terms of excess parameters rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their concentrations, and this has been interpreted to arise from the presence of strong or weak interactions. The extent of deviation depends upon the nature of the constituents and composition of the mixtures.

Figure 1 shows the variation in excess free volume as a function of the concentration of 1-alkanols in all systems. The values of excess free volume were almost positive in all of the systems and decreased with increasing concentrations of DMF. This was due to the weakening of the hydrogen bonding interaction between the ketone (cyclohexanone) and alcohols, and also due to the dissociation of alkanol molecules. The observed positive value for excess free volume also suggests that the DMF-alkanol association is weaker than the alkanol-cyclohexanone interactions.

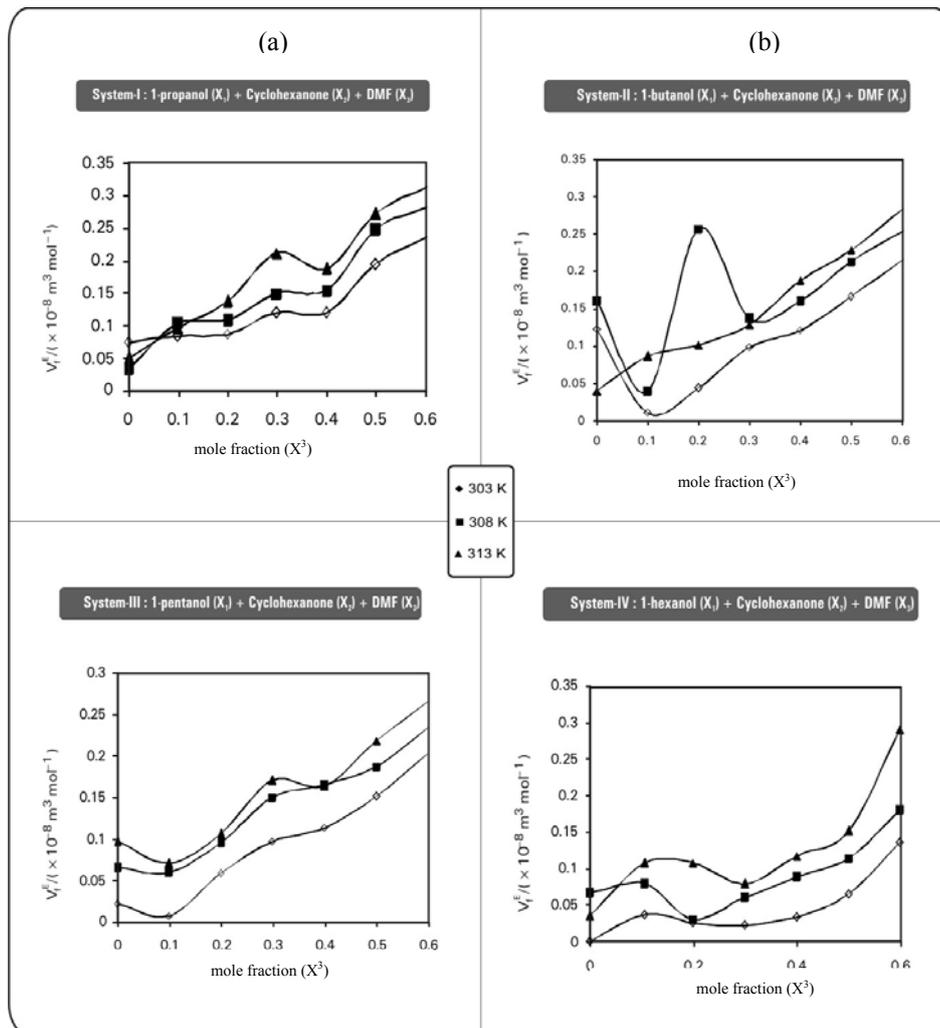
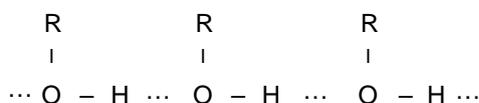


Figure 1: Variation of excess free volume (V_f^E) versus mole fraction of system 1-alcohol at 303, 308 and 313 K.

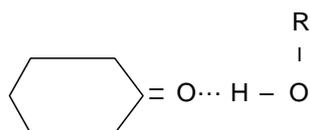
A plausible qualitative explanation of the behaviour of these mixtures has been suggested. The mixing of DMF with 1-alkanols causes the dissociation of the hydrogen-bonded structure of 1-alkanols and the subsequent formation of (new) H-bonds [C=O ...H-O] between the proton acceptor oxygen atom (with lone pair of electrons) of the C=O group of DMF and the proton of the OH group of 1-alkanols. The first (dissociation) effect leads to an increase in free volume, resulting in positive values, whereas the second effect leads to a reduction in free volume, resulting in negative values of (V_f^E). The observed positive (V_f^E) values for the four liquid ternary systems over the entire composition range suggest that the effect due to the disruption of H-bonded associations of 1-alkanols dominates that of H-bonding between unlike molecules, i.e., the DMF-alkanol interaction is weaker than the DMF-DMF or alkanol-alkanol interactions.

From Table 3, it can be observed that the excess values of (V_f^E) were more positive for System-I [1-propanol-cyclohexanone-DMF] than the other systems, suggesting that the strengths of the hydrogen bonds formed should follow the order 1-pentanol > 1-butanol > 1-hexanol > 1-propanol. Furthermore, an increase in temperature also induces the rupture of hydrogen bonds between unlike molecules.

Generally, 1-alkanols are associated through hydrogen bonding.

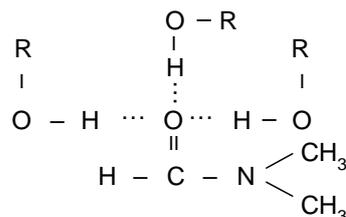


Cyclohexanone–1-alkanol interactions are due to hydrogen bonding between the oxygen atom of the ketone (cyclohexanone) and the proton of the hydroxyl group of the alkanol.



Furthermore, the addition of N, N-dimethylformamide (DMF) to mixtures causes the dissociation of the hydrogen-bonded structures of 1-alkanols, as well as a decrease in the interactions between ketones and alkanols. The subsequent formation of new hydrogen bonds between the proton acceptor

oxygen atoms of the $\text{C}=\text{O}$ group of DMF and the proton of the $-\text{OH}$ group of 1-alkanols $17 \left[\begin{array}{c} \diagup \\ \text{C}=\text{O} \cdots \text{H}-\text{O}- \\ \diagdown \end{array} \right]$.



The internal pressure is a cohesive force, which is the result of attractive and repulsive forces between the molecules. The attractive forces mainly consist of hydrogen bonding, dipole-dipole, and dispersion interactions. Repulsive forces, acting over very small intermolecular distances, play a minor role in the cohesion process under normal circumstances. Such a negative excess internal pressure in all the systems (Fig. 2) clearly confirms the above prediction.

The value of Gibb's free energy (ΔG^*) (Table 3) exhibited positive deviations, increased with increasing concentrations of 1-alkanols in all of the systems and decreased with increasing temperature. The increasing positive values of Gibb's function suggest the existence of molecular associations between unlike molecules.¹⁸⁻¹⁹ The decrease in $-\Delta G^*$ with increase in temperature in all of the mixtures indicates the need for a shorter time for the co-operative process or the rearrangement of the molecules in the mixture.²⁰

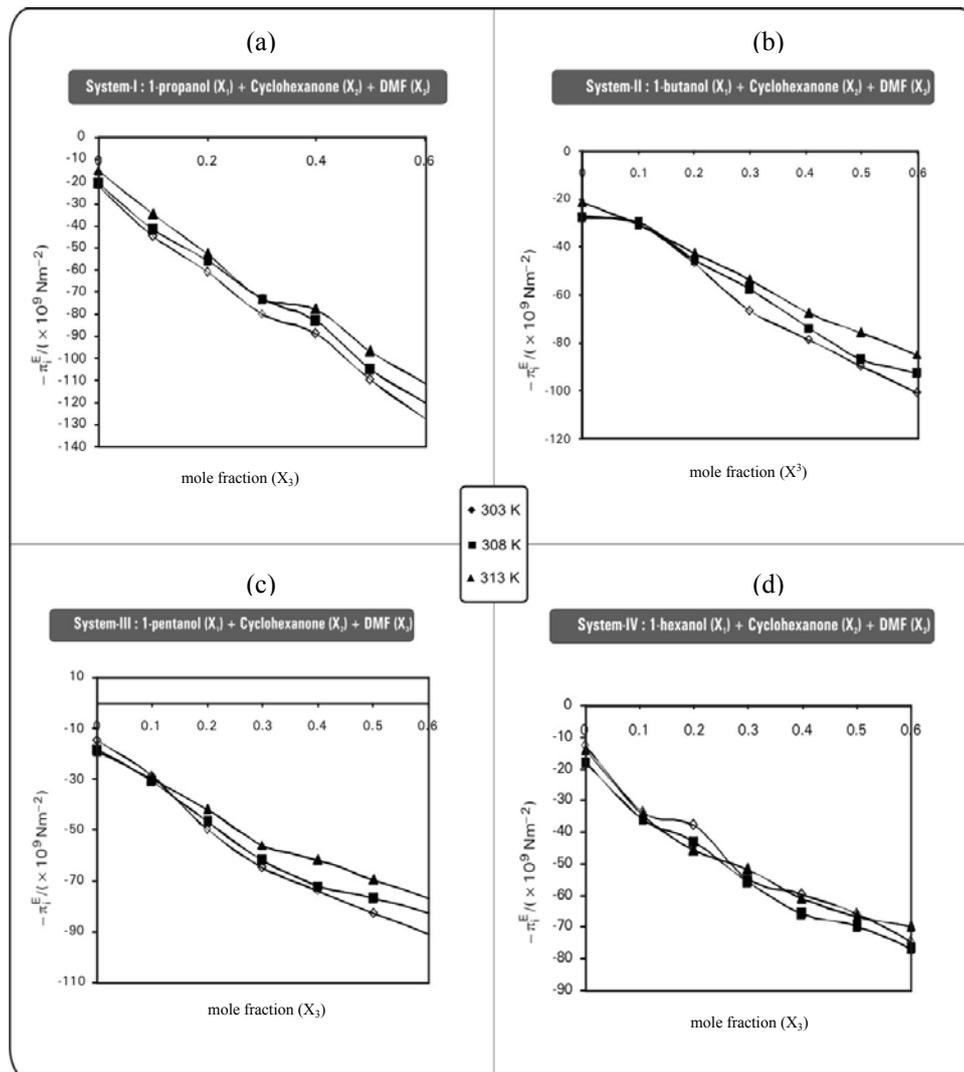


Figure 2: Variation of excess internal pressure (π_i^E) versus mole fraction of system 1-alcohol at 303, 308 and 313 K.

5. CONCLUSION

From ultrasonic velocity, related acoustical parameters and their excess values for ternary liquid mixtures of 1-alkanols with DMF in cyclohexanone at different concentrations and at varying temperatures, It is concluded that there exist a molecular interaction between DMF (proton acceptor) and 1-alkanols due to hydrogen bonding and the observed positive excess values of free volume indicate that the effect due to rupture of hydrogen bonded association of 1-alkanols and decrease in interaction between ketone and 1-alkanols influences over that hydrogen bonding between DMF-1-alkanols.

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