

Acoustical and Thermodynamical Properties of Ternary Liquid Mixtures at 303.15 K

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Abstract: The observed experimental parameters such as density (ρ), viscosity (η) and ultrasonic velocity (U) of ternary liquid mixtures of primary alkanols such as 1-propanol, 1-butanol, 1-pentanol and 1-hexanol with *N-N* dimethylformamide (DMF) in acetophenone at 303.15 K were determined. The observed experimental data have been utilized to evaluate some of the thermo acoustical parameters and also their excess values such as adiabatic compressibility (β^E), intermolecular free length (L_f^E), free volume (V_f^E), internal pressure (π^E), Gibb's energy (ΔG^E) and viscosity (η^E). Our present investigation observes that the interaction between DMF (Proton acceptor) and 1-alkanols is due to hydrogen bonding. There seems to be a dissociation of associated structure of 1-alkanols and decrease in interaction (hydrogen bonding) between acetophenone and 1-alkanols on further addition of DMF. It is also noticed that on further addition of DMF leads to subsequent decrease in interaction between ketone (acetophenone) and 1-alkanols as well as between DMF and 1-alkanol, which eventually resulting in existence of weak molecular interactions in the present system of liquid mixtures.

Keywords: Adiabatic compressibility, Dipole-dipole interaction, Hydrogen bonding, Internal pressure, Intermolecular free length

Introduction

In recent years, the measurement of ultrasonic velocity has been adequately employed in understanding the nature of molecular interactions in pure liquids and liquid mixtures. The 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 the liquid mixtures¹⁻³. Ultrasonic velocity of a liquid is fundamentally related to the binding forces between the atoms or the molecules and has been adequately employed in understanding the nature of molecular interaction in pure liquids^{4,5} binary and ternary mixture. The variation of ultrasonic velocity and related parameters throw much light upon the structural changes associated with the liquid mixtures having weakly interacting components⁶⁻⁷ as well as strongly interacting components.

The study of molecular association in organic ternary mixtures having alcohol as one of the components is of particular interest, since alcohols are strongly self-associated liquid having a three dimensional network of hydrogen bond⁸ and can be associated with any other group having some degree of polar attractions⁹. Although several investigations¹⁰⁻¹¹, were carried out in liquid mixtures having alcohol as one of the components, a systematic study in a series of primary alcohols in ternary systems has been scarcely reported.

The ultrasonic studies for the binary mixtures of *N,N*-Dimethylformamide (DMF) with substituted benzenes¹² with alcohols¹³⁻¹⁴ with branched alcohols¹⁵ with aliphatic esters¹⁶ with tetrahydrofuran¹⁷ with chloroethanes and chloromethanes¹⁸ and with vinylpyrrolidine¹⁹ are already reported in the literature. An exhaustive survey of literature has shown that a few attempts have been made for the ultrasonic velocity data of ternary mixtures²⁰. However, no effort appears to have been made to collect the ultrasonic velocity data for the ternary mixtures of *N,N*-Dimethylformamide, (DMF) Acetophenone with 1-alkanols. Hence, the above literature survey has helped the authors which motivated them to carry out the present work. The present ternary liquid systems taken up by the authors for investigation at 303.15 K are

System I	1-propanol	+	Acetophenone	+	DMF
System II	1-butanol	+	Acetophenone	+	DMF
System III	1-pentanol	+	Acetophenone	+	DMF
System IV	1-hexanol	+	Acetophenone	+	DMF

Experimental

In the present work, we have used chemicals, which are analytical reagent (AR) and spectroscopic reagent (SR) grades of minimum assay of 99.9% obtained from E-Merk, Germany and Sd fine chemicals, India. The purities of the above chemicals were checked and were compared with available literature values. The ternary liquid mixtures of different known composition were prepared by mole fraction basis. In all systems, the mole fraction of the second component, acetophenone ($X_2 = 0.2$) was kept fixed while the mole fractions of the remaining two (X_1 and X_3) were varied from 0.0 to 0.9 so as to have the mixtures of different compositions. The density was determined using a specific gravity bottle by relative measurement method. The weight of the sample was measured using a electronic digital balance with an accuracy of ± 0.1 mg (Model: Shimadzu, Japan Make, AX-200). An Ostwald's viscometer (10 mL) was used for the viscosity measurement. Efflux time was determined by using a digital chronometer to within ± 0.001 s. An Ultrasonic Interferometer having a frequency of 2MHz (Mittal Enterprises, New Delhi. Model: F-81) has been used for velocity measurement. The overall accuracy in the velocity measurement is ± 2 ms⁻¹. An electronically digital operated constant temperature water bath (RAAGA Industries, Chennai) has been used to circulate water through the double walled measuring cell made up of steel containing the experimental solution at the desired temperature. The accuracy in the temperature measurement is ± 0.1 K.

Theory

Various acoustical and thermodynamical parameters are calculated from the measured data such as

$$\text{Adiabatic Compressibility } \beta = \frac{1}{U^2 \rho} \quad (1)$$

$$\text{Intermolecular free length } L_f = K\sqrt{\beta} \quad (2)$$

Where, K is a temperature dependent constant.

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

Where M_{eff} is the effective molecular weight K^* is a temperature independent constant for all liquids.

$$\text{Internal Pressure } \pi_i = bRT \left(\frac{K\eta}{U} \right)^{1/2} \left(\frac{\rho^{2/3}}{M_{\text{eff}}^{7/6}} \right) \quad (4)$$

Where, the terms have usual meaning. The Gibb's free energy can be estimated from the following relation.

$$\Delta G = KT \ln \left(\frac{KT\tau}{h} \right) \quad (5)$$

Where K is the Boltzmaan's constant and τ is the relaxation time. Excess values of the above parameters can be determined using

$$A^E = A_{\text{exp}} - A_{\text{id}} \quad (6)$$

Where $A_{\text{id}} = \sum A_i X_i$, A_i is any acoustical parameters and X_i the molefraction of the liquid component.

Results and Discussion

The experimentally determined values of density (ρ), viscosity (η) and ultrasonic velocity (U) of all the pure liquids at 303.15 K are presented in Table 1²¹⁻²³ and the same for the ternary systems (I to IV) are listed in Table 2.

The values of adiabatic compressibility (β), free length (L_f), free volume (V_f) and internal pressure (π_i), for the four liquid systems are reported at 303.15 K are given in Table 3, whereas, the Tables 4 & 5 summarise those excess values of some of these above parameters.

Table 1. Values of Density (ρ), Viscosity (η) and Ultrasonic Velocity (U) of pure organic liquids at 303.15 K

Organic Liquids	Density $\rho/(\text{kg m}^{-3})$		Viscosity $\eta/(\times 10^{-3} \text{ Nsm}^{-2})$		Ultrasonic Velocity U/ms^{-1}	
	Literature Value	Present Value	Literature Value	Present Value	Literature Value	Present Value
1-Propanol	800.1 ²¹	797.962	1.6111 ²¹	1.40018	1192 ²¹	1192
1-Butanol	804.4 ²¹	800.782	2.1502 ²¹	1.8146	1229 ²¹	1213
1-Pentanol	807.2 ²¹	806.825	2.7656 ²¹	2.3232	1253 ²¹	1255
1-Hexanal	810.2 ²¹	810.854	3.5130 ²¹	3.2259	1289 ²¹	1288
Acetophenone	1019.7 ²²	1017.52	1.532 ²²	1.3433	1454 ²²	1454
N,N-Dimethyl formamide (DMF)	947.6 ²³	939.371	0.7679 ²³	0.7522	1459 ²³	1441

The perusal of Table 2 exhibits that in all the four liquid systems, the ultrasonic velocity decreases with increase in mole fraction of alcohols. It is observed that as the number of hydrocarbon group or chain-length of alcohol increases, a gradual decrease in sound velocity is noticed. This behaviour at such concentrations is different from the ideal mixtures behaviour can be attributed to intermolecular interactions in the systems studied²⁴⁻²⁵. However, the adiabatic compressibility (β) increases with increase of concentration as shown in Table 3. It is primarily the compressibility that increases due to structural changes of molecules in the mixture leading to a decrease in ultrasonic velocity²⁶⁻²⁷. Such a continuous increase in adiabatic compressibility with respect to the concentration of alkanols has been qualitatively ascribed to the effect of hydrogen bonding or dipole-dipole interactions²⁸.

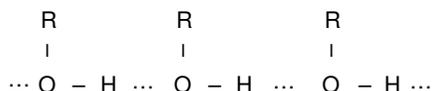
Table 2. Values of density (ρ), viscosity (η) and ultrasonic velocity (U) at 303.15 K

Mole fraction		Density	Viscosity	Ultrasonic
X_1	X_3	$\rho/(\text{kgm}^{-3})$	$\eta/(\times 10^{-3} \text{ Nsm}^{-2})$	velocity U/(ms ⁻¹)
System - I : 1 - Propanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)				
0.0000	0.7999	960.992	0.8759	1444
0.1998	0.6002	937.357	0.9023	1412
0.3997	0.3999	910.230	0.9460	1359
0.5996	0.1998	885.117	1.0405	1318
0.7999	0.0000	857.722	1.1910	1264
System - II : 1 - Butanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)				
0.0000	0.8001	960.992	0.8759	1444
0.1999	0.6002	932.119	0.93701	1401
0.2849	0.5725	902.844	1.0229	1358
0.6002	0.1996	877.463	1.1063	1311
0.7955	0.0000	855.43	1.1951	1281
System -III : 1 - Pentanol +Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)				
0.0000	0.8000	960.992	0.8759	1444
0.1999	0.6004	926.16	0.9656	1397
0.4005	0.3997	899.62	1.2108	1365
0.5998	0.1998	873.702	1.3620	1333
0.8006	0.0000	849.933	1.7667	1307
System -IV : 1 - Hexanol +Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)				
0.0000	0.8000	960.992	0.8759	1444
0.1999	0.6001	924.59	1.0239	1391
0.3999	0.4004	895.18	1.2506	1361
0.6000	0.1999	870.211	1.6087	1339
0.7999	0.0000	849.933	2.1649	1317

Alkanols are liquids which are associated through the hydrogen bonding and in the pure state, they exhibit equilibrium between the monomer and multimer species. Also, they can be associated with any other groups having some degree of polar attractions⁹. Similarly, *N,N* dimethylformamide (DMF) is belonging to the amide group which is highly polar, aprotic solvent and its molecules are associated through dipolar interactions²⁹. Mixing of DMF with alkanols would release the dipoles of alkanols due to the breaking of hydrogen bonds. As a result, the free dipoles released from the alkanols in association with DMF molecules forming strong hydrogen bonds, hence stronger molecular association existing between the

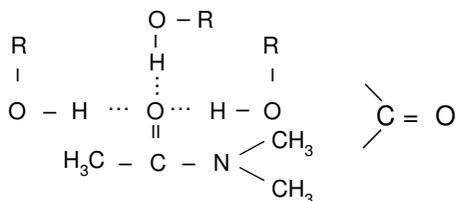
DMF and alkanols molecules through hydrogen bonding²⁸. Further, mixing of such DMF with 1-alkanols causes dissociation of hydrogen bonded structure of 1-alkanols and subsequent formation of (new) H-bond (C=O ...H...O) between proton acceptor oxygen atom (with lone pair of electrons) of C=O group of DMF and proton of OH- group of 1-alkanols³⁰.

As, 1-alkanols are associated through hydrogen bonding,



Acetophenone – 1-alkanols interactions are due to hydrogen bonding between the oxygen atom of the ketone and the proton of hydroxyl group of alkanols. The ultrasonic velocity in ternary liquid mixtures of *N,N*-dimethyl formamide in cyclohexanone with primary alcohols was investigated by Kannappan *et al.*²³ and discussed in terms of hydrogen bond formation between the ketone and alcohols. Thirumaran *et al.*³¹ also pointed out from their study of ternary mixtures of *N,N*-dimethyl acetamide (DMA) in cyclohexanone with 1-alkanols that there is formation of hydrogen bonding between the ketone and 1-alkanols.

Further, the addition of *N,N*-Dimethylformamide (DMF) with mixtures causes dissociation of hydrogen bonded structure of 1-alkanols as well as decrease in interaction between ketone and alkanols leading to subsequent decrease in ultrasonic velocity. The subsequent formation of new hydrogen bonding between proton acceptor oxygen atoms of group of DMF and proton of –OH group of 1-alkanols³⁰ $\left[\text{C} = \text{O} \cdots \text{H} - \text{O} - \right]$.



It is further observed that the same trend in free length too in all the three systems, as shown in Table 3. The increase in free length is due to the loose packing of the molecules inside the shield, which may be brought by weakening of molecular interactions³². In all the four liquid systems, free volume (V_f) decreases with the increasing concentration of alcohols which are represented in Table 3. As discussed earlier, the dissociation of hydrogen bonded structure of 1-alkanols due to the mixing of DMF with it and subsequent formation of new bonds. Such a dissociation effect leads to a contraction in volume, which resulting an decrease in free volume. Further, the increasing trend of internal pressure (π_i) in all the four liquid systems with increasing concentration of alcohols, which are tabulated in Table 3 clearly supporting this prediction.

In order to understand the nature of molecular interactions between the components of the liquid mixtures, it is of interest to discuss the same in terms of excess parameter rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their physical behaviour with respect to concentration and these have been interpreted as arising from the presence of strong or weak interactions. The effect of deviation depends upon the nature of the constituents and composition of the mixtures.

Table 3. Values of adiabatic compressibility (β), free length (L_f), free volume (V_f) and internal pressure (π_i) at 303.15 K

Mole Fraction		Adiabatic Compressibility $\beta/(\times 10^{-10} \text{ m}^2 \text{ N}^{-1})$	Free length $L_f (\times 10^{-10} \text{ m})$	Free Volume V_f ($\times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$)	Internal Pressure π_i ($\times 10^6 \text{ N m}^{-2}$)
X_1	X_3				
System - I : 1 - Propanol + Acetophenone + <i>N,N</i> - Dimethyl formamide (DMF)					
0.0000	0.7999	4.9905	0.4573	1.7914	459.294
0.1998	0.6002	5.3508	0.4615	1.5802	481.164
0.3997	0.3999	5.9485	0.4866	1.3211	520.364
0.5996	0.1998	6.5038	0.5088	1.0389	557.180
0.7999	0.0000	7.2972	0.5389	0.7560	621.156
System - II : 1 - Butanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.8001	4.9905	0.4457	0.1404	459.20
0.1999	0.6002	5.4657	0.4664	0.1217	471.163
0.2849	0.5725	6.0060	0.4880	0.1090	487.778
0.6002	0.1996	6.6307	0.5137	0.0792	520.516
0.7955	0.0000	7.1238	0.5320	0.0594	562.236
System -III : 1 - Pentanol +Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.8000	4.9905	0.4457	1.7916	459.21
0.1999	0.6004	5.5146	0.4685	1.5548	459.55
0.4005	0.3997	5.9658	0.4873	1.2640	489.35
0.5998	0.1998	6.4413	0.5064	0.9571	495.75
0.8006	0.0000	6.8875	0.5236	0.5181	538.45
System -IV : 1 - Hexanol +Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.8000	4.9905	0.4457	0.1403	455.359
0.1999	0.6001	5.5890	0.4717	0.1163	459.200
0.3999	0.4004	6.0307	0.4899	0.0918	462.094
0.6000	0.1999	6.4093	0.5051	0.0671	483.640
0.7999	0.0000	6.7833	0.5196	0.0456	521.370

The excess values of adiabatic compressibility (β^E), intermolecular free length (L_f^E), free volume (V_f^E), internal pressure (π_i^E) Gibb's Free Energy (ΔG^E) and viscosity (η^E) for all the four liquid systems are furnished in Tables 4 & 5. In the present investigation, one can observe that all the excess value parameters exhibit both positive as well as negative deviations over the entire range of composition in all four liquid systems. It is learnt that negative values of β^E is an indication of strong intermolecular interactions in the liquid mixtures, which is attributed to charge transfer, dipole-induced dipole and dipole-dipole interactions and hydrogen bonding. While positive excess values to weak interactions which result from dispersion forces. Rastogi²⁷ shown from his studies of excess thermodynamic function in ternary mixtures that the addition of third component weakens the strength of the interaction and the ternary mixture tends to approach ideal behaviour. Further, it also suggested that the negative excess compressibility has been due to a closed packed molecules and positive excess values to weak interaction between unlike molecules.

Table 4. Excess values of adiabatic compressibility (β^E), free length (L_f^E), free volume (V_f^E) and internal pressure (π_i^E) at 303.15 K

Mole Fraction		Excess Adiabatic Compressibility $\beta^E / (\times 10^{-10} \text{ m}^2 \text{ N}^{-1})$	Excess Free length L_f^E ($\times 10^{-10} \text{ m}$)	Excess Free Volume V_f^E ($\times 10^{-7} \text{ m}^3 \text{ mol}^{-1}$)	Excess Internal Pressure π_i^E ($\times 10^6 \text{ Nm}^{-2}$)
X_1	X_3				
System - I : 1 - Propanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.7999	-0.0399	0.0100	-0.0405	313.069
0.1998	0.6002	-0.4181	-0.0140	0.0397	140.461
0.3997	0.3999	-0.5566	-0.0168	0.0725	108.568
0.5996	0.1998	-0.6875	-0.0227	0.08217	12.5728
0.7999	0.0000	-0.7386	-0.0210	0.0904	-56.635
System - II : 1 - Butanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.8001	-0.0410	-0.0017	-1.6919	312.934
0.1999	0.6002	-0.2375	-0.0067	-1.4152	210.805
0.2849	0.5725	-0.2766	-0.0222	-1.4096	170.1518
0.6002	0.1996	-0.4162	-0.0113	-0.8656	31.7346
0.7955	0.0000	-0.5574	-0.0163	-0.5888	-37.355
System -III : 1 - Pentanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.8000	-0.0405	-0.0016	-0.0405	313.3
0.1999	0.6004	-0.0661	-0.0081	0.0219	211.75
0.4005	0.3997	-0.1646	-0.0184	0.0323	139.696
0.5998	0.1998	-0.2326	-0.0280	0.0256	44.984
0.8006	0.0000	-0.4379	-0.0405	-0.1139	-14.435
System -IV : 1 - Hexanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)					
0.0000	0.8000	-0.0405	-0.0016	-1.6918	312.74
0.1999	0.6001	0.0955	0.0058	-1.4006	209.82
0.3999	0.4004	0.0730	0.0053	-1.1102	117.24
0.6000	0.1999	-0.0092	0.0022	-0.8186	39.55
0.7999	0.0000	-0.0976	-0.0017	-0.5248	-21.95

Our present study depicts that the values of excess adiabatic compressibility (β^E) are negative for the first three liquid systems and it becomes negative on going to higher mole fraction range in system IV. However, the value of β^E decreases on further addition of alcohols in all the four systems and such a decreasing values suggest that the weakening of molecular interactions in the liquid mixtures. The Table 4 shows the variation of the excess free volume as a function of concentration in all systems. In the present investigation, the excess free volume values are positive in Systems I & III and negative in II & IV, which exhibit that the free volume in all the systems weaken due to hydrogen bonding interaction between ketone and alcohols and also due to dissociation of alcohol molecules. The observed positive values of excess free volume also suggest that the DMF - alkanols interaction is weaker than the alkanol-alkanol and alkanols-ketone interactions. The decreasing trend of positive excess values of internal pressure in all the four systems clearly confirms this.

One should observe from the Table 5, the variation of excess Gibbs energy ΔG^E for the four ternary liquid systems. The values of ΔG^E are all negative in all the systems concerned and increase with molar concentration of alcohols. According to Reed *et al.*³³ the positive values of excess Gibbs energy values may be attributed to specific interactions like hydrogen bonding and charge transfer, while negative ΔG^E values may be ascribed to the dominance

of dispersion forces. In the present investigation, our close observation suggests that the increasing negative values of ΔG^E shows that the strength of interaction gets weakened with increasing of 1-alkanols. According to Fort *et al.*³⁴ the variation of excess viscosity (η^E) gives the strength of molecular interaction between the molecules.

Table 5. Excess values of excess Gibb's free energy (ΔG^*) and excess viscosity (η^E) at 303.15 K

Mole fraction		Excess Gibb's free energy (ΔG^{*E}) ($\times 10^{-20}$ k Jmol ⁻¹)	Excess viscosity(η^E) ($\times 10^{-3}$ Nsm ⁻¹)
X ₁	X ₃		
System - I : 1 - Propanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)			
0.0000	0.7999	-0.9507	0.0055
0.1998	0.6002	-0.8142	-0.0961
0.3997	0.3999	-0.6543	-0.1831
0.5996	0.1998	-0.4817	-0.2179
0.7999	0.0000	-0.2826	-0.1976
System - II : 1 - Butanol + Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)			
0.0000	0.8001	-0.9510	0.00540
0.1999	0.6002	-0.8081	-0.1458
0.2849	0.5725	-0.7827	-0.1933
0.6002	0.1996	-0.4790	-0.4016
0.7955	0.0000	-0.2964	-0.5170
System -III : 1 - Pentanol +Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)			
0.0000	0.8000	-0.9509	0.0055
0.1999	0.6004	-0.8983	-4.2836
0.4005	0.3997	-0.8003	-0.2889
0.5998	0.1998	-0.7476	-0.4504
0.8006	0.0000	-0.6419	-0.3619
System -IV : 1 - Hexanol +Acetophenone + <i>N,N</i> -Dimethyl formamide (DMF)			
0.0000	0.8000	-0.9509	0.0055
0.1999	0.6001	-0.7985	-0.3412
0.3999	0.4004	-0.6440	-0.6092
0.6000	0.1999	-0.4729	-0.7458
0.7999	0.0000	-0.2853	-0.6841

For systems, where dispersion, induction and dipolar forces which are operated by the values of excess viscosity are found to be negative, whereas the existence of specific interactions leading to the formation of complexes in liquid mixtures tends to make excess viscosity positive. In our present investigation, from Table 5, the excess viscosity values are negative in all the four ternary liquid systems and decrease with the further addition of alkanols (except in System III) Hence, our present close observation of excess viscosity reveals that presence of weak dipolar forces are existing in the liquid mixtures. The possession of such negative values of η^E further suggests that the dominance of dispersive interaction resulting from the breaking up of the hydrogen bonds of alkanols resulting in weakening of molecular interactions in the liquid mixtures.

Conclusion

From ultrasonic velocity, related acoustical parameters and their excess values, we firmly conclude that the interaction between DMF and 1-alkanols is due to hydrogen bonding. We

also further noticed that on further addition of DMF causes the rupturing of hydrogen bonded association of 1-alkanols and subsequent decrease in interaction between ketone (acetophenone) and 1-alkanols as well as between DMF and 1-alkanols. Hence, it very obvious that existence of a weak molecular interaction prevails in the present system of liquid mixtures. No donor-acceptor complexities were identified in the present study.

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