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Volumetric, Viscometric and Ultrasonic Study of Binary Liquid Mixture of N, N Dimethyl acetamide and Chloroform at Different Temperatures

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Abstract: Experimental values of ultrasonic velocities(U),densities (ρ) and viscosities (η) in the binary liquid mixture of N,N-dimethyl acetamide (DMAc) with chloroform are presented over the entire composition range at single frequency 2MHz along with two discrete temperatures 300and 303 K . From these fundamental data, the derived thermo-acoustical parameters along with their excess values such as excess velocity (U^E),excess molar volume ($V^E{}_m$), deviations in viscosity (Δη), and excess Gibbs free energy of activation(ΔG^E),excess isothermal compressibility($\kappa^T{}_s$), excess internal pressure(π^E) ,excess enthalpy(ΔH^E) ,excess energy (ΔE^E),excess entropy(ΔS^E),excess internal pressure (π^E),deviations in viscosity (Δη), available volume(V_a), and Lennard -Jones potential repulsive exponent term (n), relative association (R_A) and interaction parameter(χ) have been calculated. These results were fitted to Redlich- Kister polynomial equations to estimate the binary coefficients and standard errors. Jouyban-Acree model is used to correlate the experimental values of densities and ultrasonic velocities at temperatures 300and 303 K. The values of excess molar volume ($V^E{}_m$) have been analyzed using Prigogine-Flory-Patterson (PFP) theory. An analysis of each of the three contributions viz., interactional volume $V_m^E(\text{int})$,free volume $V_m^E(fv)$ and characteristic pressure $V_m^E(P^*)$ to excess molar volume ($V^E{}_m$). The results of the viscosity composition are discussed in the light of various theoretical models. The present investigation also encompass of assessment of the acoustic non-linearity parameter (B/A), excess non linear parameter $(B/A)^E$ and with along with computation of cohesive energy(ΔA), Vander Wall's constant(a) and distance of closest approach(d). The results are discussed in terms of intermolecular interactions between the component molecules of the binary liquid mixture.

Keywords: Ultrasonic velocity, Binary liquid mixture, Lennard -Jones potential repulsive exponent term, PFP theory, Non-linearity parameter (B/A),Cohesive energy, Molecular interactions.

I. INTRODUCTION

The intermolecular interactions in binary and ternary liquid mixtures are estimated by various spectroscopic and non-spectroscopic methods such as Infrared (IR), Raman Effect, Nuclear magnetic resonance (NMR), Dielectric, Ultraviolet (UV) and Ultrasonic method. Among these, ultrasonic method was found to be accurate and sensitive and extensively used by many researchers during last few decades, to elucidate molecular interactions in binary and ternary liquid mixtures. Ultrasonics is an area of enormous scientific and technological research. In view of its extensive scientific and engineering applications, it has drawn attention of large cross section of researchers, non-destructive testing (NDT) professionals, industrialists, technologists, medical researchers, instrumentation engineers, software engineers, material scientists and others. Ultrasound, a mechanical wave, which interacts with matters with a variety of wave modes, longitudinal to several surface waves, which is possible for diverse applications. Ultrasonics started from the basic subject of sound in physics and now represents a vast field of its own with several branches and sub-branches of scientific pursuit and technological importance. Basic measurements of ultrasonics, viz. attenuation and velocity, have been applied in verifying physical theories, microstructural characterization, mechanical property evaluation and others. Prior to ultrasonic application to mixtures/solutions, the spectroscopic and dielectric techniques were the only tool to study the nature and strength of molecular interactions. However, application of ultrasonics has made possible not only the evaluation of physico-chemical properties of the mixtures/solutions but also more reliability on the interpretation of molecular interactions. Due to low cost, easy operational procedure and spontaneous results, the molecular interaction studies through ultrasonics have gained importance all over the world (Raj et al 2007). In liquid mixtures, the possible interactions are between like molecules as well as unlike molecules. These interactions are of two types; namely, long-range and short-range. The long-range interaction includes electrostatic induction and dispersion forces, which arises without the overlap of the electron clouds due to the closer approach of interacting molecules. On the other hand, short-range interactions such as dipole-dipole, dipole-induced dipole, charge transfer, complex formation and hydrogen bonding interactions arise when the molecules come closer together, resulting in a significant overlapping of electron clouds**[1-4]**. The long-range interactions are highly directional. The formation of complexes in the liquid mixtures and solutions can be studied through the intermolecular forces. In the case of an ideal liquid mixture, there is a change in volume or enthalpy while mixing. When two or more liquids are mixed, then the mixture is not ideal. Thus, the deviation from the ideality is explained based on the molecular interactions between the components of the liquid mixtures. The present regarding the study of molecular interaction in binary liquid mixture with N,N dimethyl acetamide (DMAc) with chloroform(CF) as constituents is of particular interest ,because DMAc is a dipolar aprotic solvent with high boiling point along with high thermal and chemical stability. It has large dipole moment and dielectric constant(μ=3.7D and ε=37.8).DMAc is an excellent proton donor as well as proton acceptor and hence it is strongly associated through intermolecular hydrogen bond. It is highly soluble in a variety of polar and non-polar solvents and readily suitable to explore solvent-solvent interactions. It is used as a solvent in the production of acrylic and elasthane fibres, pharmaceuticals, antibiotics and polyimide resins. Chloroform is considered as non polar solvent. It has small dipole moment and dielectric constant(μ=1.15D and ε=4.81).Chloroform is a common solvent in the laboratory because it is relatively non-reactive, miscible with most organic liquids, and conveniently volatile. Chloroform is used as a solvent in the pharmaceutical industry and for producing dyes and pesticides. It is useful for extracting alkaloids (such as morphine) of pharmaceutical importance from plant materials (such as poppies). Chloroform has myriad uses as a reagent and a solvent. Owing to these considerations, an attempt has been made to explain the molecular interaction in binary liquid mixture of dimethyl acetamide (DMAc) and chloroform at different temperatures 300and 303K at constant frequency 2 MHz. Departure from linearity in the velocity versus concentration in liquid mixture of chloroform is taken as an indication of the existence of interaction between different liquid molecules **[5-20]**. The physiochemical properties of liquid mixture can be studied by the non-linear variation of ultrasonic velocity and other ultrasonic parameters with structural changes occurring in a liquid and the liquid mixture.

A. Theory

From the experimental values of fundamental acoustic parameters such as densities($\dot{\rho}$),ultrasonic velocities(U) and viscosities(η),derived thermo-acoustical parameters along with their excess values such as excess velocity (U^E),excess molar volume (V^E_m), deviations in viscosity ($\Delta\eta$), and excess Gibbs free energy of activation(ΔG^E),excess isothermal compressibility(κ^T_s),excess internal pressure(π^E) ,excess enthalpy(ΔH^E) ,excess energy (ΔE^E),excess entropy(ΔS^E),excess internal pressure (π^E),deviations in viscosity ($\Delta\eta$), available volume(V_a), and Lennard -Jones potential repulsive exponent term (n), relative association (R_A) and interaction parameter(χ) have been calculated using following relations.

1) *Available Volume(V_a)*

$$V_a = V_m\left(1-\frac{U}{U_\infty}\right) \tag{1.1}$$

Where V_m is the molar volume and U_∞=1600 ms^{-1}

2) *Relative Association(R_A)*

$$R_A = \left(\frac{\rho}{\rho_0}\right)\left(\frac{U_0}{U}\right) \tag{1.2}$$

Where are ρ_0 and U_0density and velocity of solvent respectively.

3) *Interaction Parameter(χ)*

$$\chi = \left[\left(\frac{U}{U_{ideal}}\right)^2 - 1\right] \tag{1.3}$$

Where $U_{ideal} = x_1U_1 + x_2U_2$ is the ideal mixing velocity, x_1 and x_2 are mole fractions,U_1 and U_2 are ultrasonic velocities of component (1)(DMAc) and (2)(Chloroform) respectively.

4) Excess Molar Volume (V^E_m)

$$V_m^E = \left[\left(\frac{M_1 x_1 + M_2 x_2}{\rho_{1,2}}\right) - \left(\frac{M_1 x_1}{\rho_1} + \frac{M_2 x_2}{\rho_2}\right)\right] \tag{1.4}$$

Where, V_m^E is the excess molar volume of liquid mixture, x_1 and x_2 are mole fractions, M_1 and M_2 are molecular weights, and ρ_1, ρ_2 and $\rho_{1,2}$ are the density of components(1)(DMAc)and (2)(Chloroform) of binary liquid mixture respectively.

5) Viscosity Deviation($\Delta\eta$)

$$\Delta\eta = \eta_{1,2} - (x_1\eta_1 + x_2\eta_2) \tag{1.5}$$

Where x_1, x_2 and η_1 ,η_2 and $\eta_{1,2}$ refers, the mole fractions and viscosities of the components (1)(DMAc) , (2)(Chloroform) and binary mixture respectively.

6) Excess Gibb's free energy(ΔG^E)

The excess Gibbs free energy, ΔG^E of mixtures is given by the following equation:

$$\Delta G^E = \Delta H^E - T\Delta S^E \tag{1.6}$$

Where the excess enthalpy(ΔH^E), and excess entropy (ΔS^E) are computed from π_i and V_f by using the following well established relations of binary mixtures:

$$-\Delta H^E = \pi_m V_m - (x_1\pi_1 V_1 + x_2\pi_2 V_2) \tag{1.7}$$

$$T\Delta S^E = RT\left[x_1 \ln(V_{f,1}) + x_2 \ln(V_{f,2}) - \ln V_{f,m}\right] \tag{1.8}$$

where $\pi_{i,1}$,$\pi_{i,2}$ and π_i are internal pressure of of component(1)(DMAc), component(2) (Chloroform), and mixture, respectively. And $V_{f,1}$,$V_{f,2}$ and $V_{f,m}$ are free volumes of component(1)(DMAc), component (2)(Chloroform),and mixture respectively.

7) Excess isothermal compressibility(κ^E_T)

Isothermal compressibility,(κ_T) can be deduced from the well-known acoustic method and is the convenient and well-established method for(κ_T) is given by

$$\kappa_T = \kappa_S + \left(\frac{\alpha^2 T V_m}{C_P}\right) \tag{1.9}$$

Where α is the coefficient of thermal expansion, which can be deduced by the following theoretical equation suggested by Pandey et.al**[21]**

$$\kappa_T = \left(\frac{75.6 * 10^{-3}}{T^{\frac{1}{9}} U^{\frac{1}{2}} \rho^{\frac{1}{3}}}\right) \tag{1.10}$$

8) Excess internal pressure(π^E): The internal pressure(π), is given by

$$\pi = \left(\frac{\alpha T}{\kappa_T}\right) \tag{1.11}$$

Whereα and κ_T are isobaric thermal expansion and isothermal compressibility of individual component of binary mixture**[22].**The excess internal pressure (π^E) can be deduced by following relation:

$$\pi^E = \left(\frac{\alpha T}{\kappa_T} - \frac{\sum_i \Phi_i \alpha_i T}{\sum_i \Phi_i \kappa_{T,i}}\right) \tag{1.12}$$

9) Excess Velocity(U^E)

Excess velocity can be calculated by the experimentally determined values of ultrasonic velocity for pure component as follows:

$$U^E = U_{mix} - (x_1 U_1 + x_2 U_2) \tag{1.13}$$

10) *Lennard-Jones potential term(n)*

$$n = \left[6\left(\frac{V_m}{V_a} \right) - 13 \right] \qquad (1.14)$$

Where V_m and V_a are molar volume and available volume respectively

11) *Excess enthalpy(H^E)*

Enthalpy(H_i) of pure components can be determined as follows:

$$H_i = \pi_i V_m \qquad (1.15)$$

The excess enthalpy(H^E)can be evaluated by following usual relation:

$$H^E = H_m - (x_1 H_1 + x_2 H_2) \qquad (1.16)$$

Where H_m represents the experimentally calculated value of enthalpy for the mixture and H_1, H_2 represent enthalpy of pure components(1-DMAc) and (2-Chloroform), respectively.

The excess properties have been fitted to a Redlich–Kister type polynomial equation given by-

$$Y^E = x_1 x_2 \sum_j A_j (x_1 - x_2)^j \qquad (1.17)$$

Where $Y^E = \kappa_T^E$, π^E ,H^E ,G^E ,E^E ,S^E and V^E. x_1 and x_2 are the mole fraction of DMAc and Chloroform respectively. Further, A_j are the regulating parameters of the function; and are determined using the least square method. In the present investigation we have limited to j=4 basing on the correlation coefficient. The corresponding standard deviations $\sigma(Y^E)$ have been calculated using the following statistical relation:

$$\sigma(Y^E) = \left[\frac{\sum_j (Y_{\exp} - Y_{cal})^2}{m - n} \right]^{\frac{1}{2}} \qquad (1.18)$$

where 'm' is the total number of investigational points and 'n' is the number of coefficients in Eq. (1.18). The evaluated values of the coefficients A_j and the standard deviations $\sigma(Y^E)$ are tabulated in **Table(4)**and **Table(5).**

II. MATERIAL AND METHODS

Experimental details Liquid mixtures of various concentrations in mole fraction were prepared by taking chemicals (N,N dimethyl acetamide and Chloroform) of analytic grade (AR)with minimum assay of 99.9%(Sisco Research Laboratory ,Mumbai,India)which were used without further purification.Liquid mixture of different mole fractions were prepared on concentration scale with a precision 0.001g using an electronic digital balance.

A. *Density Measurement*

The density of pure liquids and mixtures are measured using a 10ml specific gravity bottle. The weight of the pure liquids and liquid mixtures are determined by using single pan electronic balance supplied by Aczet Pvt.Ltd.India (Model CY 132) with least accuracy up to 1 mg. The specific gravity bottle with the experimental liquid is immersed in a double walled or jacketed cylindrical borosilicate glass beaker supplied by Saber Scientific, Ahmedabad, Gujrat, India, connected via inlet and outlet pipes with temperature controlled water bath Supplied by Mittal enterprises, New Delhi with ± 0.1^0C accuracy to maintain desired temperature for density measurement . An average of triple measurements was taken into account to reduce the possible experimental error. Sufficient care was taken to avoid any air bubble entrapment. The densities of pure liquids thus obtained are found to be in good agreement with standard values. The measured density was measured using the following relation:

$$\rho_2 = \rho_1 \left(\frac{w_2}{w_1} \right) \qquad (1.19)$$

Where w_1= Weight of distilled water,w_2=Weight of experimental liquid,$\dot{\rho}_1$=Density of distilled water,$\dot{\rho}_2$=Density of experimental liquid.

B. Viscosity Measurement

The viscosity of the pure liquids and liquid mixtures are measured using an Ostwald's Viscometer with an accuracy 0.001N-m^{-2}s.The viscometer was pre- calibrated by double distilled water. The Ostwald's Viscometer with the experimental liquid is immersed in a double walled or jacketed cylindrical borosilicate glass beaker supplied by Saber Scientific ,Ahmedabad , Gujrat, India. Connected via inlet and outlet pipes with temperature controlled water bath Supplied by Mittal Enterprises, New Delhi, to maintain desired temperature for viscosity measurement. The efflux time of flow was measured using a digital racer stop watch with an accuracy of 0.1 sec. An average of three or four sets of efflux times for each was taken into consideration to reduce the possible experimental error. The viscosities of pure liquids thus obtained are found to be in good agreement with standard values. Viscosity was determined by using the following relation:

$$\eta_2 = \eta_1 \left[\left(\frac{t_2}{t_1} \right) \left(\frac{\rho_2}{\rho_1} \right) \right] \qquad (1.20)$$

Where η_1=Viscosity of distilled water,η_2=Viscosity of experimental liquid,ρ_1=Density of experimental liquid,ρ_2=Density of experimental,t_1=Time of flow of distilled water,t_2=Time of flow of experimental liquid.

III. RESULT AND DISCUSSION

The experimentally measured values of density (ρ́),coefficient of viscosity (η) and ultrasonic velocity (U) for the binary mixture of N,N dimethyl acetamide (DMAc) +chloroform(CF) at two discrete temperatures 300 and 303 K are presented in **Table-1.**

Table-1

Mole fraction		U(ms^{-1})		ρ́(kg-m^{-3})		η(MPa-s)	
x_1(DMAc)	x_2(CF)	300K	303K	300K	303K	300K	303K
1	0	1432	1424	911.1	908.6	0.9266	0.8802
0.9	0.1	1378.2	1364.2	952.8	950.9	0.8774	0.8339
0.8	0.2	1320	1316.6	1001.1	994.2	0.8309	0.7890
0.7	0.3	1272.4	1257.9	1047.3	1039.2	0.7868	0.7484
0.6	0.4	1228.6	1206.4	1100.5	1086.2	0.7451	0.7090
0.5	0.5	1159.7	1151.3	1140.8	1135.4	0.7055	0.6717
0.4	0.6	1107.5	1100.7	1195.8	1186.7	0.6681	0.6364
0.3	0.7	1073.4	1068.2	1255.4	1240.4	0.6327	0.6029
0.2	0.8	1035.4	1020.8	1315.9	1296.5	0.5991	0.5711
0.1	0.9	998.7	984.6	1367.5	1355.1	0.5673	0.5411
0	1.0	963	955.2	1425.7	1416.4	0.5372	0.5126

The values from Table -1 shows the increase in density with the increase in mole fraction of chloroform (CF) indicates that dipole-induced dipole interactions increases with the increase in concentration of chloroform in the binary liquid mixture under investigation. Further the density values shows decremental pattern with the increase in temperature, affirms the decreases in intermolecular forces due to the increase in the thermal energy of the system. As per Edward Peters[23] a high density or viscosity of a component molecule is a reflection of higher intermolecular interactions. It is evident from Table-1, for a given temperature, ultrasonic velocity decreases with increasing mole fraction of chloroform. Since DMAc is a polar molecule and when it is associated with non polar solute chloroform, the chloroform molecule tends to dissociate the DMAc…..DMAc dipolar association and releases several DMAc dipoles within the solution. These free DMAc dipoles would induce moment in the adjacent chloroform molecules resulting dipole-induced dipole interactions in the binary liquid mixture. With the increase of the mole fraction of chloroform ,concentration of adjacent chloroform molecules increases resulting the dominance of dipole –induced dipole interactions over dipole- dipole interactions within the binary mixture. Hence there exist a weak dipole-dipole attraction. The values from Table-1 show that ultrasonic velocity values decrease with increase in temperature due to the breakage of hetero and homo molecular clusters at high temperatures (Nagargun et al., 2013). From the Table-1, it is evident that The increase in coefficient of viscosity with the increase in mole fraction of DMAc indicates the presence of solute solvent interactions. Which may be the specific interactions between unlike molecules in the binary liquid mixture. As the temperature increases, the magnitude of the viscosity deviation sharply decreases due to the rapid breaking up of the molecular aggregates in the systems and ultimately tends to approach ideality.

Thermophysical properties of mixed solvent are vital to understand the type of the occurring molecular interactions [24] which are helpful in the design and optimization of chemical progressions for a variety of industrial applications. Excess properties are supportive in characterizing the structure, properties of solutions and give precious information concerning solvent–solvent interactions [25].From these fundamental data, the derived thermo-acoustical parameters along with their excess values such as excess velocity (U^E),excess molar volume (V^E_m), deviations in viscosity ($\Delta\eta$), and excess Gibbs free energy of activation(ΔG^E),excess isothermal compressibility($\kappa^T{}_s$), excess enthalpy(ΔH^E) , excess internal pressure (π^E),deviations in viscosity ($\Delta\eta$), available volume(V_a), and Lennard-Jones potential repulsive exponent term (n), relative association (R_A) and interaction parameter(χ) have been calculated at desired frequency 2MHz and two discrete temperatures 300 and 308 K. These derived parameters are presented in Table-2 and Table-3 referred to discrete temperature 300 and 303 K under consideration.

Table-2

System(DMAc+CF)At T=300K											
x_2 (CF)	U^E ($ms^{-1)}$	$\Delta\eta$ (MPa-s)	V^E*10^{-6} (m^3-$mole^{-1}$)	$\kappa^E{}_T$ (T-Pa^{-1})	π^E (MPa)	ΔG^E (J-$mole^{-1}$)	ΔH^E (J-$mole^{-1}$)	V_a*10^{-5} (m^3mole^{-1})	R_A	LJP (n) (J-$mole^{-1}$)	χ
0	0.000	0.0000	0.0000	0.0000	0.0000	0.000	0.000	1.0040	1.0000	44.1428	0.0000
0.1	-7.934	-0.0102	-0.5719	-3.5019	-0.0460	87.169	75.241	1.3011	1.0866	30.2822	-0.0163
0.2	-14.844	-0.0178	-0.9651	-5.3200	-0.0944	135.785	119.18	1.6149	1.1920	21.2857	-0.0187
0.3	-21.088	-0.0230	-1.2056	-5.8897	-0.1398	164.44	152.342	1.8601	1.2937	16.3040	-0.0259
0.4	-28.220	-0.0258	-1.3143	-5.5757	-0.1776	176.781	172.277	2.0787	1.4078	12.8481	-0.0403
0.5	-33.930	-0.0264	-1.3088	-4.6882	-0.2029	172.225	179.092	2.4318	1.5461	8.8033	-0.0560
0.6	-36.830	-0.0248	-1.2034	-3.4949	-0.2117	150.37	170.102	2.6867	1.6970	6.4924	-0.0647
0.7	-34.556	-0.0214	-1.0104	-2.2307	-0.1998	118.12	145.827	2.8400	1.8382	5.2301	-0.0606
0.8	-25.130	-0.0160	-0.7399	-1.1040	-0.1633	75.513	110.918	3.0125	1.9975	4.0032	-0.0429
0.9	-10.849	-0.0088	-0.4006	-0.3030	-0.0980	39.263	65.654	3.1765	2.1521	2.9654	-0.0182
1.0	0.000	0.0000	0.0000	0.0000	0.0000	0.000	0.000	3.3337	2.3269	2.0706	0.0000

Table-3

System(DMAc+CF)At T=303K											
x_2 (CF)	U^E ($ms^{-1)}$	$\Delta\eta$ (MPa-s)	V^E*10^{-6} (m^3-$mole^{-1}$)	$\kappa^E{}_T$ (T-Pa^{-1})	π^E (MPa)	ΔG^E (J-$mole^{-1}$)	ΔH^E (J-$mole^{-1}$)	V_a*10^{-5} (m^3mole^{-1})	R_A	LJP (n) (J-$mole^{-1}$)	χ
0	0.000	0.0000	0.0000	0.0000	0.0000	0.000	0.000	1.0547	1.0000	41.5454	0.0000
0.1	-6.360	-0.0096	-0.5526	-3.2387	-0.0431	85.436	64.348	1.3878	1.0924	27.7125	-0.0182
0.2	-13.353	-0.0167	-0.9321	-4.9209	-0.0886	130.815	111.509	1.6407	1.1835	20.8744	-0.0226
0.3	-20.607	-0.0215	-1.1659	-5.4484	-0.1312	160.482	146.058	1.9508	1.2948	15.0619	-0.0349
0.4	-27.417	-0.0241	-1.2716	-5.1581	-0.1665	171.735	167.157	2.2133	1.4111	11.3902	-0.0521
0.5	-32.809	-0.0247	-1.2667	-4.3371	-0.1902	166.156	172.960	2.4908	1.5456	8.3951	-0.0642
0.6	-35.280	-0.0233	-1.1652	-3.2331	-0.1983	145.870	163.149	2.7386	1.6897	6.2270	-0.0655
0.7	-33.247	-0.0200	-0.9786	-2.0634	-0.1872	110.739	139.604	2.8845	1.8198	5.0518	-0.0619
0.8	-24.194	-0.0150	-0.7168	-1.0211	-0.1529	71.484	105.199	3.1091	1.9904	3.5746	-0.0480
0.9	-9.805	-0.0085	-0.3882	-0.2802	-0.0917	35.732	60.735	3.2715	2.1569	2.5996	-0.0362
1.0	0.000	0.0000	0.0000	0.0000	0.0000	0.000	0.000	3.3337	2.3239	1.8883	0.0000

Table-4

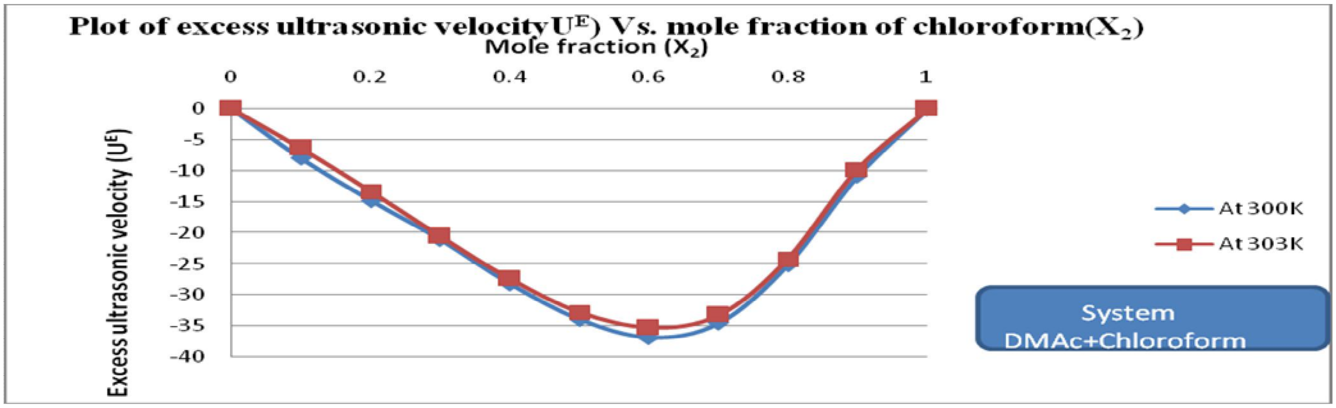
At 300T/K						
Property	A_0	A_1	A_2	A_3	A_4	σ (Y^E)
$K_T{}^E$*10^{-6}(Pa^{-1})	1.5879	10.4742	-66.2685	-15.126	0.0001398	1.4102
π^E*10^6(Pa)	-2.7514	-1.3890	14.3625	7.7035	-21.0185	0.2481
H^E(KJ$mole^{-1}$)	0.6204	0.0780	-1.2409	-0.6924	1.8607	0.0218
G^E(J-$mole^{-1}$)	0.6649	-0.4356	-0.9489	0.1066	1.5879	0.01007
E^E(J-$mole^{-1}$)	-5188.7521	867.9008	90.3156	-52.6449	4.3411	0.0079
S^E($JK^{-1}mole^{-1}$)	5.6518	-0.9422	0.0620	0.0044	-0.00098	1.0306E-05
V^E*10^{-6}(m^3mole^{-1})	-3.6577	11.644	-66.4183	-15.0876	0.0001396	1.4101

Table-5

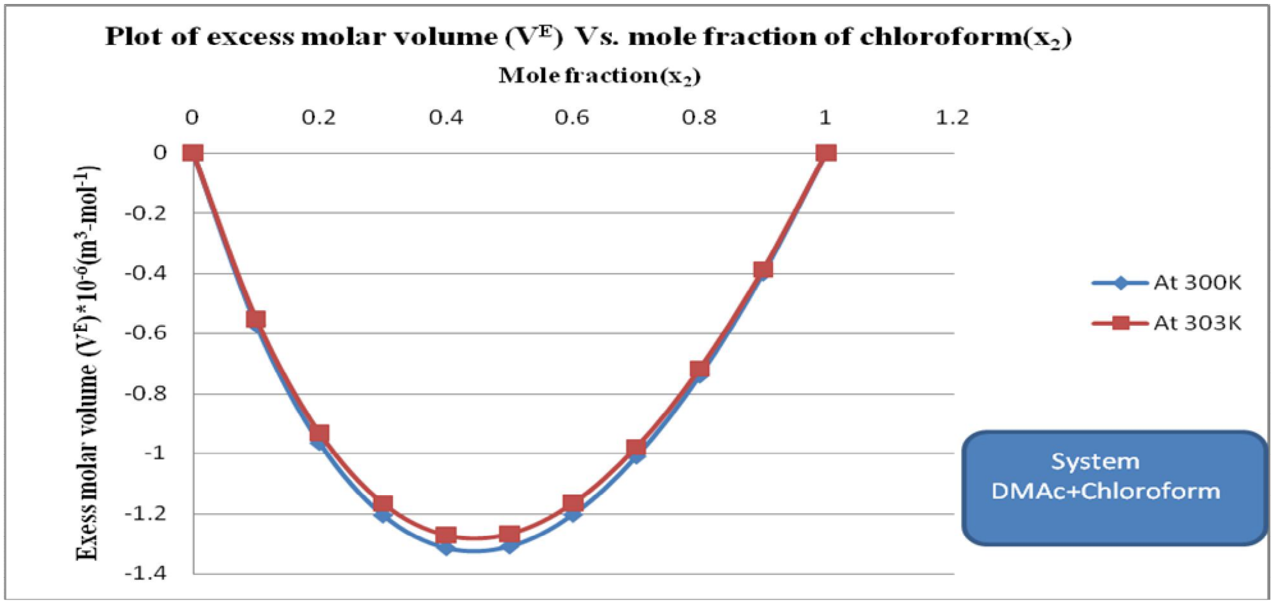
At 303T/K						
Property	A_0	A_1	A_2	A_3	A_4	σ (Y^E)
K_T^E*10^{-6}(Pa^{-1})	1.5880	10.4739	-66.2695	-15.1253	0.0001398	1.41018
π^E*10^6(Pa)	-3.6070	-0.6546	0.7016	-0.7025	-0.7767	0.0241
H^E(J-$mole^{-1}$)	0.6918	-0.0428	-0.0990	0.0278	0.1621	0.001867
G^E(J-$mole^{-1}$)	0.6767	-0.3042	0.5532	0.8791	-1.0355	0.01014
E^E(J-$mole^{-1}$)	-5036.2771	841.3355	79.4084	-48.4455	4.2283	0.01094
S^E($JK^{-1}mole^{-1}$)	5.2432	-0.9058	1.6094	0.003105	-2.1196	0.0339
V^E*10^{-6}(m^3mole^{-1})	-3.4894	11.5976	-66.4081	-15.0905	0.0001397	1.4101

It is evident from Table-2 and Table-3 and figure-1,that the excess velocity (U^E) is negative for the entire range of concentration of chloroform for two discrete temperatures 300 and 303K. It is expected that dipole–dipole, dipole-induced dipole, charge-transfer interaction and hydrogen bonding between unlike components should make negative contributions [26].Table-2 and Table-3 ,figure-1 shows the variation of excess velocity of the solution for different mole fractions of chloroform at two different temperatures. For the whole composition range, the excess velocity becomes increasingly negative with increasing strength of interaction between the component molecules. Since N,N dimethyl acetamide it is a dipolar aprotic liquid with large dipole moment, mixing of DMAc with chloroform will induce the mutual destruction of dipolar structure of the component liquids releasing free dipoles. As a result, strong dipolar interactions between DMAc and chloroform are expected. The observed negative values of velocity (U^E) over the complete range of composition of dimethyl acetamide (DMAc)–chloroform binary mixture indicates the presence dispersive forces between unlike molecules in the binary liquid mixture along with On the other hand, if the predominant effect in the combination is structure breaking.

(figure-1)

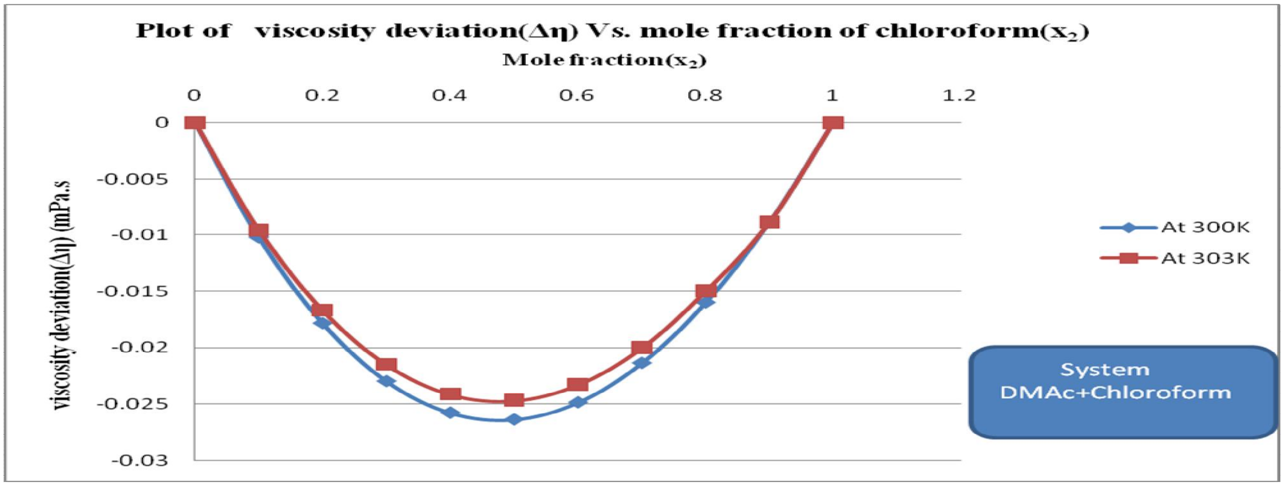


The sign of V_m^E of a system depends upon the relative magnitude of expansion and contraction of the two liquids due to mixing [27]. The negative V_m^E arises due to dominance of the following factors. (a) Chemical interaction between constituent molecules such as heteromolecular association through the formation of H-bond, often termed as strong specific interaction. (b) Association through weaker physical forces such as dipolar force or any other forces of this kind. (c) Accommodation of molecules of one component into the interstitial positions of the structural network of molecules of the other component. (d) Geometry of the molecular structure that favors fitting of the component molecules with each other. It is evident from Table-2,Table-3 and figure-2,the V_m^E values are negative over the entire mole fraction range and at all temperatures investigated for all binary systems under study. The negative V_m^E values are attributed to dipole-dipole interactions between unlike molecules in the mixtures. The electron density at oxygen atom of the carbonyl group of DMAc is generally attributed due to the presence of methyl group at carbon atom of carbonyl group which results the stronger interaction in the system [28, 29]. The strength of interactions between DMAc +chloroform (CF) can be explained due to the fact that negative charge on nitrogen in DMAc is more due to conjugation with C=O group. The observed negative values of V_m^E at the two discrete temperatures (300 &303 K), indicate much stronger interactions between the unlike molecules of components in this binary mixture due to the formation of not only dipole-dipole interactions but also of induced polar interaction like dipole-induced dipole and dipole interactions between chloroform and DMAc. The V_m^E values increase (become less negative) with increase in temperature for the present systems. This is attributed due to the decrease in interaction between chloroform and amide molecules resulting in the less favorable fitting in interestial accommodation leading to relatively less contraction in volume and hence resulting in less negative values of V_m^E with rise in temperature.

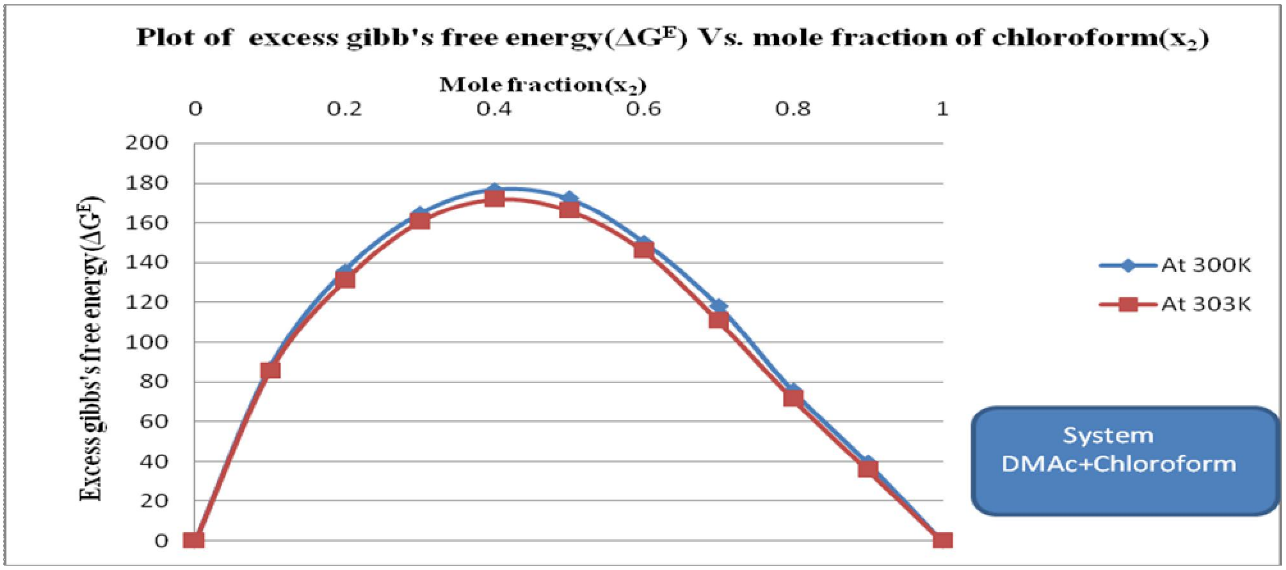


(figure-2)

The variation of the deviation in viscosity (Δη), with mole fraction(x_2), of chloroform (CF) for the binary mixtures at discrete temperatures is shown in Table-2,Table-3 and figure-3. The values of Δη for the system under study are negative over the whole range of composition and temperature investigated. Negative Δη values are observed in systems in which dipole-dipole forces are primarily responsible for interactions between the component molecules[30]. The addition of chloroform to DMAc causes disruption of associates present in the latter releasing more free DMAc dipoles. These free dipoles may interact through dipole-dipole and dipole induced dipole interactions, in addition to hydrogen bonding, with DMAc molecules in the mixture. It is observed from figure(3) that the magnitude of the negative Δη values decreases and tends towards zero as the temperature of the mixtures increases, i.e., the system tends towards ideal behaviour with rise in temperature.

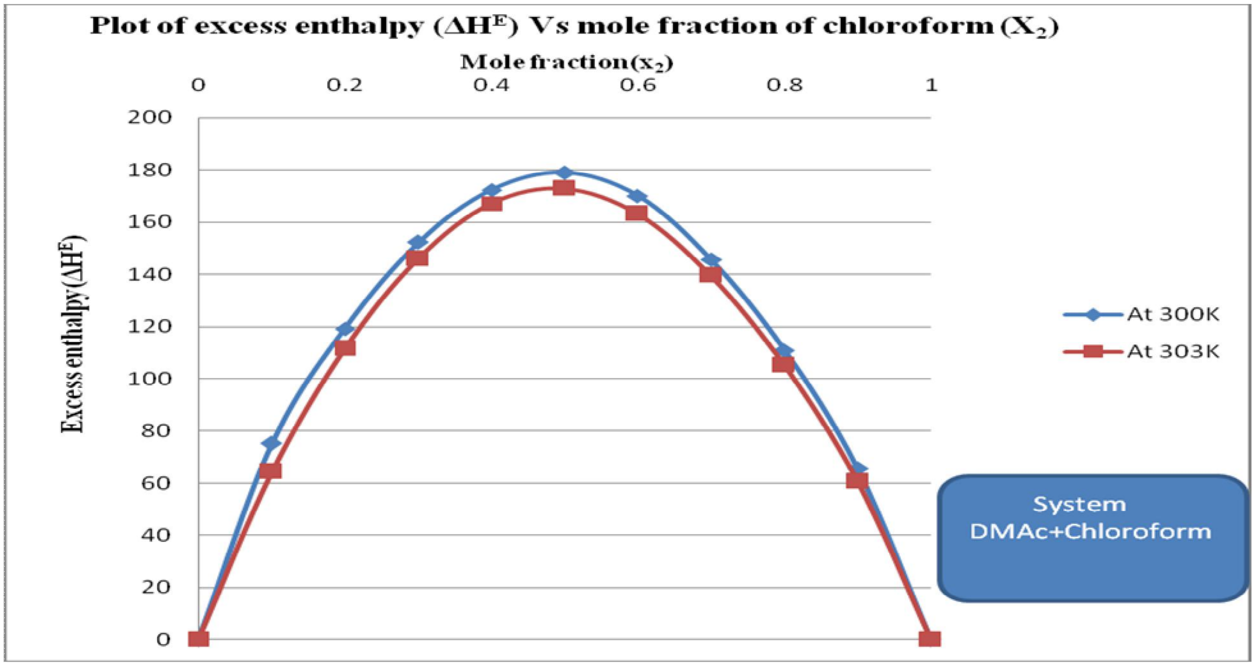


(figure-3)



(figure-4)

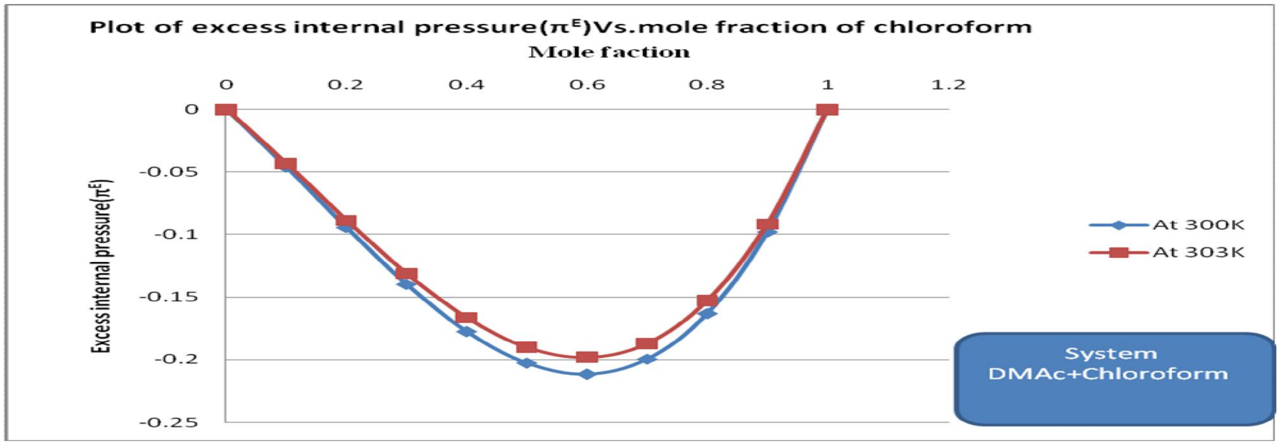
According to Mayer[31] ΔG^E may be considered as a reliable measure to detect the presence of interaction between the molecule. Positive values of ΔG^E can be seen in binary mixture where specific interactions (hydrogen bonding) between the molecules are dominant where as negative ΔG^E indicates characteristic behavior of mixture in, which dispersion forces are dominant[32]. The variation of the excess Gibbs energy of activation of viscous flow(ΔG^E), with mole fraction(x_2),of chloroform for the binary mixtures at different temperatures is presented in Tabe-2,Table-3 and figure(4). The values of ΔG^E are found to be positive over the whole range of composition and temperature investigated. In general, the positive contributions to the ΔG^E values may be attributed to the presence of specific interactions (hydrogen bonding and dipole-dipole interactions), whereas negative contributions may be ascribed to the dispersion forces**.** The observed positive ΔG^E values suggest the formation of hydrogen bonds and dipolar interactions between the unlike molecules in the system under study. As expected, like V^E and $\Delta\eta$, the ΔG^E values tend towards zero with increase in the temperature of the mixture, indicating that the system approaches ideal behaviour with rise in temperature. Thus for the system DMA + chloroform(CF), the functions $V^E{}_m$, $\Delta\eta$ and ΔG^E truly support each other over the entire composition range and at two discrete temperatures investigated.



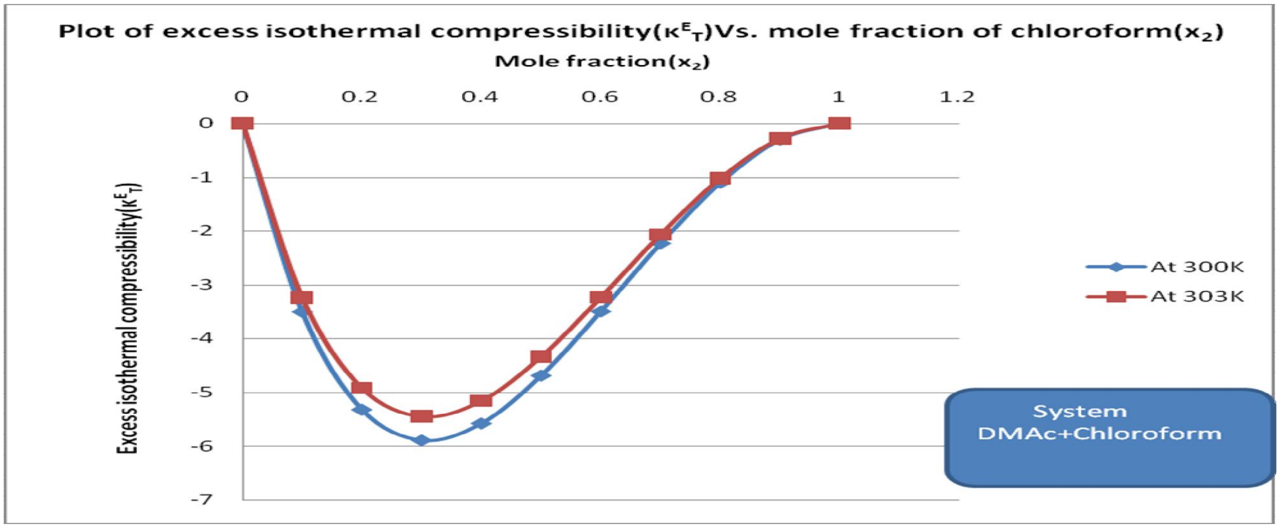
(figure-5)

The enthalpy of mixing (or excess enthalpy) is the enthalpy liberated or absorbed from a substance upon mixing. When a substance or compound is combined with any other substance or compound the enthalpy of mixing is the consequence of the new interactions between the two substances or compounds. This enthalpy if released exothermically can in an extreme case cause an explosion. Enthalpy of mixing can often be ignored in calculations for mixtures where other heat terms exist, or in cases where the mixture is ideal. The sign convention is the same as for enthalpy of reaction: when the enthalpy of mixing is positive, mixing is endothermic while negative enthalpy of mixing signifies exothermic mixing. In ideal mixtures the enthalpy of mixing is null. In non-ideal mixtures the thermodynamic activity of each component is different from its concentration by multiplying with the activity coefficient. Values of excess enthalpy (ΔH^E) were found to be positive in the case of dissociation. In our present study the values of excess enthalpy (ΔH^E) is found to be positive for both temperatures according to Table-2, Table-3 and figure-5. Positive values at both temperatures suggest specific weak interaction between molecules of components mixture. Large positive values in case of DMAc+chloroform suggest that long range dispersive force is prominent in this case. The trend of excess enthalpy (ΔH^E)supports our earlier result of excess internal pressure

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. The observed negative values for excess internal pressure (π^E) for DMAc and chloroform mixtures shown in**Table-2 Table-3** and **figure-6** indicates presence of specific interaction between chloroform and DMAc molecules in these mixtures and is due to existence of dipole-dipole or dipole-induced interactions, proper interstitial accommodation and
orientational ordering.



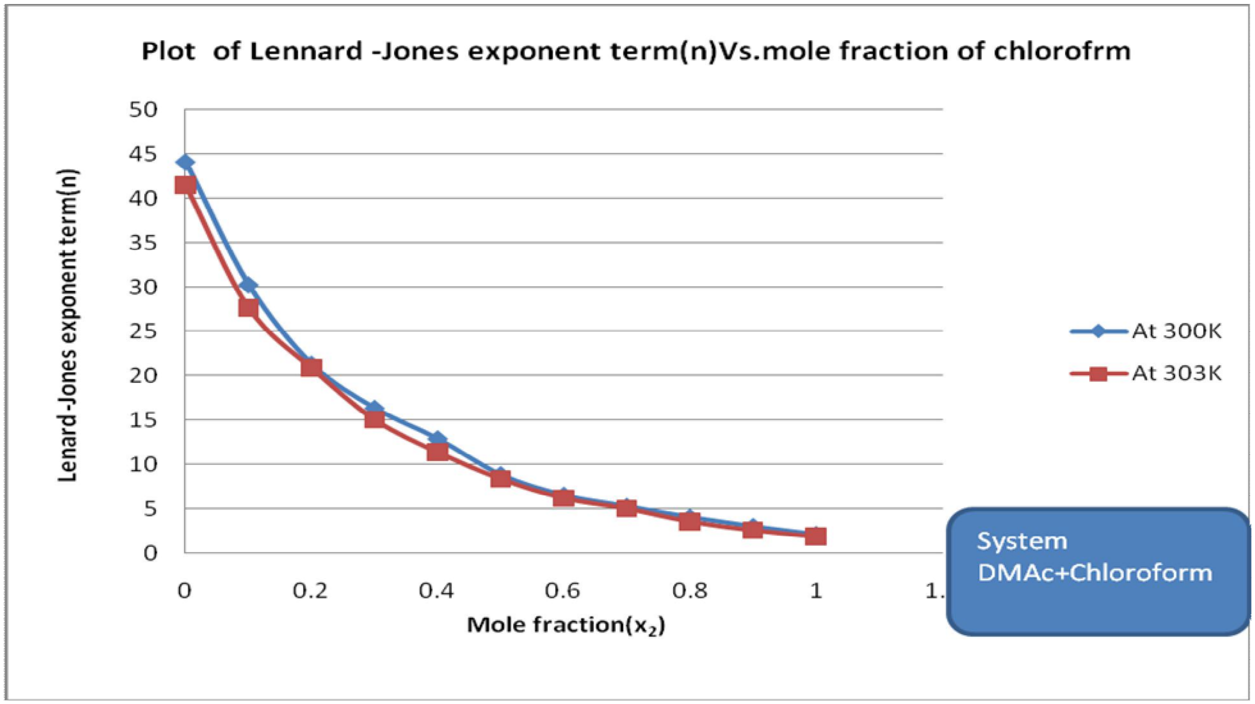
(figure-6)



(figure-7)

The values of $\kappa^E{}_T$ are negative (Table-2,Table-3 and Figure-7) for the whole composition range range of mole fraction of chloroform for the two discrete temperaturs .The negative value of $\kappa^E{}_T$ is associated with a structure forming tendency but the positive value is associated with a structure breaking tendency due to hetero-molecular interaction between the component molecules of liquid mixture . In the present investigation the negative values of $\kappa^E{}_T$ predict the existence of strong molecular interactions in the binary liquid mixture. Hence in the present binary mixture DMAc+chloroform, the specific interaction responsible for association is likely to be through hydrogen bonding, dipole-dipole interactions ,dipole-induce dipole or formation of complexes due to charge transfer.

The Lennard-Jones model consists of two 'parts'; a steep repulsive term, and smoother attractive term, representing the London dispersion forces. Apart from being an important model in itself, the Lennard-Jones potential frequently forms one of 'building blocks' of many force fields. Lennard-Jones model is not the most faithful representation of the potential energy surface, but rather its use is widespread due to its computational expediency. The Lennard-Jones Potential is given by the following equation:



(figure-8)

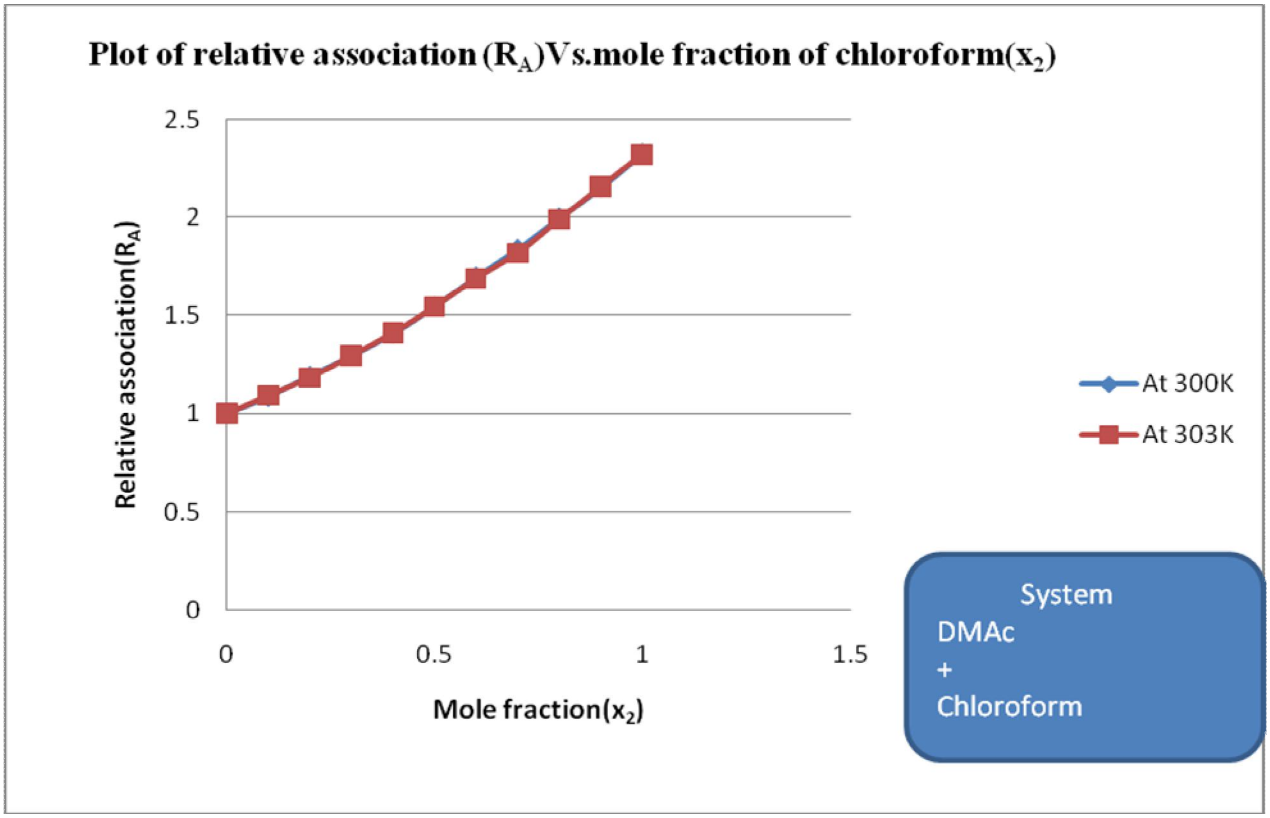
$$V(r) = 4\varepsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] \qquad (1.21)$$

or is sometimes expressed as:

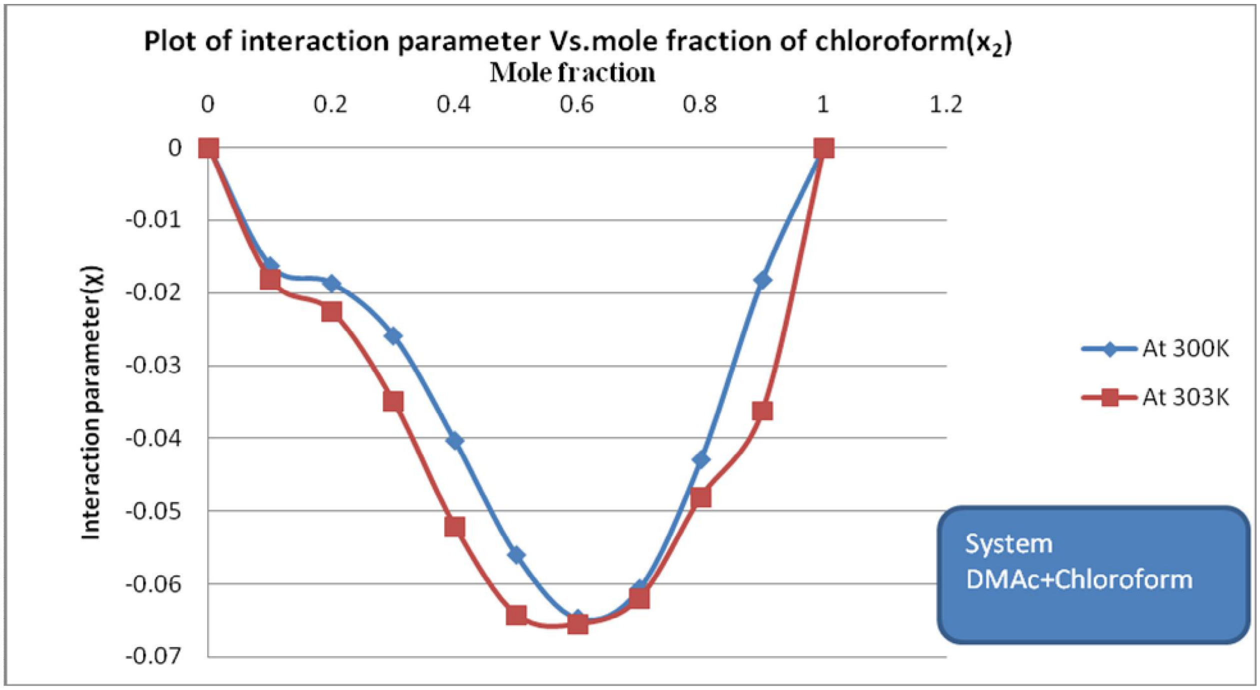
$$V(r) = \frac{A}{r^{12}} - \frac{B}{r^{6}} \qquad (1.22)$$

whereV(r) is the intermolecular potential between the two atoms or molecule, ε is the well depth and a measure of strength with which two paticles attract each other,σ is referred as Vander-Waal's radius and r is referred the distance of separation between both particles. $A=4\varepsilon\sigma^{12}$ and $B=4\varepsilon\sigma^{6}$ Thus large value of n indicates the dominance of attractive forces over repulsive forces.Table-2,Table-3 and figure-8 show that the values of (n) decreases the concentration of chloroform increases for a fixed temperature. The decrease in (n) indicates the decreasing dominance of attractive force over repulsive forces in the binary liquid mixture. Further, the values of n decrease with the increase in temperature for a particular concentration which indicates the increase in repulsive forces due to reduction in molecular interaction in the binary mixture. This also explains the cause for the decrease in ultrasonic velocity with the increase in temperature.

It is evident from Table-2 ,Table-3 and figure-9 that the relative association (R_A) for the mixture DMAc and chloroform increases with the increase in mole fraction of chloroform for two discrete temperatures indicates the presence of molecular interaction between unlike molecules. Such increase in relative association supports the idea that the liquid system is in a more compressed state and the component molecules are much closer to each other at higher concentration of chloroform and there may exist dipole-induced dipole interactions between component molecules in the binary liquid mixture**[33]**. Relative association remains almost constant for all the temperature concerned.

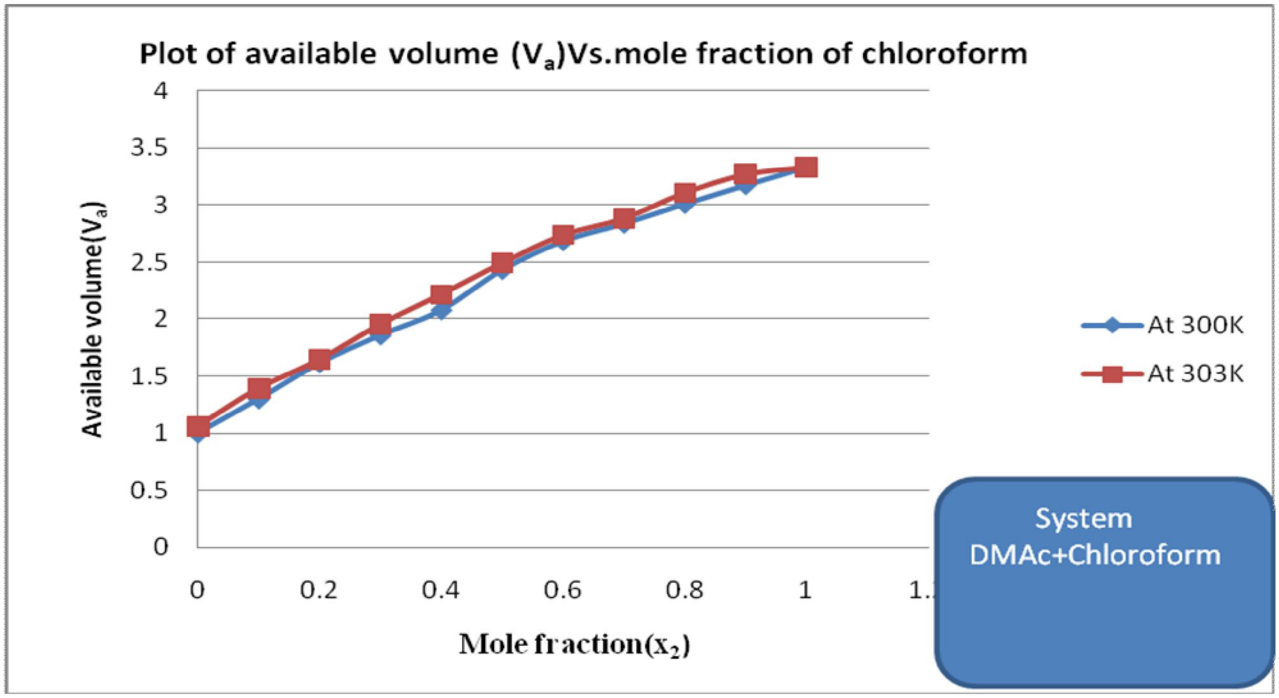


(figure-9)



(figure-10)

Table-2,Table-3 and figure-10 shows that the values of interaction parameter χ are negative for the entire range of mole fraction chloroform at all temperatures under investigation. which indicate the existence of dispersive forces with weak dipole-induced dipole interactions in the binary liquid mixture with the increase in mole fraction of chloroform. With the increase of chloroform concentration in DMAc+chloroform binary mixture induces breaking of DMAc-DMAc dipolar dissociation rate increases ,which results the dominance of dipole-dipole interaction due to availability of free DMAc dipolar concentration within the binary solution. Further interaction parameter shows irregular trend with the increase in temperature. It has limited thermal affects.



(figure-11)

Table-2, Table-3 and figure-11 represents that available volume increases with the increase in mole fraction of chloroform. Available volume is a direct measure of compactness and strength of bonding between the molecules of the liquid mixture .The increase in Va is due to decrese in interstitial accommodation between voids of solvent molecules which may be formed by week complexation between unlike molecules through the reduction in strength of hydrogen bonding in the binary liquid mixture[34]. Thus, the increase in available volume with raising temperature is quite natural phenomenon and it is attributed due to increase in molecular motion and decrease inintermolecular attraction at higher temperatures.

A. Prigogine-Flory-Patterson (PFP) Theory

The PFP model came up from the development of the Prigogine– Flory theory by Patterson and Delmas. The Prigogine-Flory-Patterson (PFP) theory used for quantitative estimation of excess thermodynamic functions of binary liquid mixtures**[35-39]** .The PFP theory is widely used with the aim of establishing the relative importance of different contributions that give rise to the experimentally observed values of the excess thermodynamic functions. The PFP theory considers excess thermodynamic properties of binary mixtures to be the sum of three contributions. The excess molar volumes ($V^E{}_m$) can thus be expressed as a sum of the interactional term, $V_m^E(\text{int})$ calculated from the interactional parameter (χ), the free volume contribution $V_m^E(fv)$ and $P^*(V_m^E)$ contribution which originates from the differences in the internal pressures and the reduced volumes of the pure components. The expression for these three contributions are given as:

$$V_m^E(\text{int}) = \left[\frac{(\tilde{V}^{\frac{1}{3}} - 1)\tilde{V}^{\frac{2}{3}}\psi_1\theta_2}{(\frac{4}{3}\tilde{V}^{-\frac{1}{3}} - 1)P_1^*\chi_{12}} \right] \qquad (1.23)$$

$$V_m^E(fv) = \left[\frac{(V_1^{\approx} - V_2^{\approx})^2 (14/9\, V^{\approx -\frac{1}{3}} - 1)\psi_1\psi_2}{(4/3\, V^{\approx -\frac{1}{3}} - 1)V^{\approx}}\right] \qquad (1.24)$$

$$V_m^E(P^*) = \left[\frac{(V_1^{\approx} - V_2^{\approx})(P_1^* - P_2^*)\psi_1\psi_2}{P_2^*\psi_1 + P_1^* - P_2^*}\right] \qquad (1.25)$$

Thus ,the excess molar volume is given as

$$\frac{V_m^E}{\left(x_1V_1^* + x_2V_2^*\right)} = V_m^E(\text{int}) - V_m^E(fv) + V_m^E(P^*) \qquad (1.26)$$

where the term $V_m^E(\text{int})$ is the interactional contribution which arises from any difference in the chemical natures of the two components in the binary mixture and is proportional to the Flory parameter (χ).The free volume term, $V_m^E(fv)$, arises from dependence of the reduced volume upon the reduced temperature as a result of the difference in the degree of thermal expansion between the two components in the mixture. The term $V_m^E(fv)$,is a measure of geometrical effects which include size, shape and conformation of unlike molecules in the binary mixture. The $V_m^E(P^*)$ term is the characteristic pressure contribution and is proportional to $\left(V_1^{\approx} - V_2^{\approx}\right)\left(P_1^* - P_2^*\right)$ and can bear either sign depending on the relative magnitude of P_i^* and $V_i^{\sim}$ (or the isobaric thermal expansivity,(α_i) of unlike components in the binary mixture.

Reduced volume of binary mixture is defined as:

$$V^{\approx} = \psi_1V_1^{\approx} + \psi_2V_2^{\approx} \qquad (1.27)$$

The characteristic parameters V* and P* are obtained from thermal expansion coefficient (α)and isothermal compressibility(κ_T):

$$V_i^{\approx} = \left[\frac{(1 + 4/3\,\alpha_i T)}{(1 + \alpha_i T)}\right]^3 \qquad (1.28)$$

Where i=1,2 (for binary mixture only).

$$V_i^* = \frac{V_i}{V_i^{\approx}} \qquad (1.29)$$

$$P_i^* = \frac{P_i}{P_i^{\approx}} = \frac{TV_i^{\approx 2}\alpha_i}{\kappa_{T,i}} \qquad (1.30)$$

Where i=1,2 (for binary mixture only).

The molecular contact energy fraction is calculated by:

$$\psi_1 = (1 - \psi_2) = \left[\frac{\phi_1P_1^*}{\phi_1P_1^* + \phi_2P_2^*}\right] \qquad (1.31)$$

$$\phi_1 = (1 - \phi_2) = \left[\frac{x_1V_1^*}{x_1V_1^* + x_2V_2^*}\right] \qquad (1.32)$$

where ϕ is a hard-core volume fraction.

The molecular surface fraction is calculated by:

$$\theta_1 = (1 - \theta_2) = \left[\frac{\phi_1}{\phi_1 + (S_1/S_2)\phi_2}\right] \qquad (1.33)$$

where S_i is the molecular fraction surface/volume ratio for the components determined by Bondi's method**[40]**

$$\frac{S_1}{S_2}=\left(\frac{V_2^*}{V_1^*}\right)^{\frac{1}{3}} \tag{1.34}$$

1) *Properties And Parameters Of Pure Components Used In The Pfp Theory Calculations* **:** The values of Characteristic pressure (P^*),Molar volume (V_m),Characteristic volume (V^*), Thermal expansion coefficient (α), Isothermal compressibility (κ_T) for both the component used in present binary mixture at two discrete temperatures are listed in following tables:

Table-6

At300K					
Component	P^* *10^{-8}(J-m^{-3})	V_m*10^{-5}(m^3-mol^{-1})	V^**10^{-5}(m^3-mol^{-1})	α*10^{-3}(K^{-1})	K_T*10^{-9}(N^{-1}-m^2)
DMAc	1.37269	9.56207	7.1021	1.516251	6.00689
Chloroform	1.55239	8.37343	6.2106	1.527099	5.36443

Table-7

At303K					
Component	P^**10^{-8} (J-m^{-3})	V_m*10^{-5} (m^3-mol^{-1})	V^**10^{-5} (m^3-mol^{-1})	α *10^{-3}(K^{-1})	K_T*10^{-9} (N^{-1}-m^2)
DMAc	1.38203	9.58838	7.12121	1.501701	5.96885
Chloroform	1.55685	8.42841	6.25092	1.512575	5.35198

The interaction parameter χ_{12} required for the calculation of V^E_m using PFP theory has been derived by fitting the V^E_m expression to the experimental equimolar value of V^E_m for given system at different temperatures under study. The values of χ_{12}, , three PFP contributions interactional $V_m^E(\text{int})$ free volume $V_m^E(fv)$, pressure effect $V_m^E(P^*)$ and experimental and calculated (using PFP theory) values at near equimolar composition $x_1 = x_2 = 0.5$ and at different temperatures are presented in Table 8&9.

Table-8

At 300K				
System	χ_{12} (J-m^{-3})	V^E_{int}*10^{-6}(m^3-mol^{-1})	V^E_{fv}*10^{-11}(m^3-mol^{-1})	V^E_{P*}*10^{-9}(m^3-mol^{-1})
DMAc+Chloroform	-0.43210167	-1.31803	6.57179	9.31439

Table-9

At 303K				
System	χ_{12} (J-m^{-3})	V^E_{int}*10^{-6}(m^3-mol^{-1})	V^E_{fv}*10^{-11}(m^3-mol^{-1})	V^E_{P*}*10^{-9}(m^3-mol^{-1})
DMAc+Chloroform	-0.41516087	-1.27577	6.46301	9.10465

Study of the data presented in Table 8, Table-9 reveals that the that the interactional contribution $V_m^E(\text{int})$ and the values of the Flory's interactional parameter(χ_{12}), are negative for the binary mixture under investigation. The interactional contribution $V_m^E(\text{int})$ or the Flory interaction parameter(χ_{12}), when negative, shows the existence of specific intermolecular interactions whereas positive values of each of these two functions indicates a predominance of dispersion forces when a binary mixture is created[41]. It can also be observed that the PFP theory reproduces the main features of the experimental data by using only one adjustable parameter, (χ_{12}) to describe V_m^E for the { DMAc+Chloroform } binary mixture over the entire composition range. Thus it is evident that PFP theory is quite successful in predicting the trend of the dependence of on composition for the present systems.

B. Semi-empirical Models for Analyzing Viscosity of Liquid Mixtures

Several empirical and semi-empirical relations have been used to represent the dependence of viscosity on concentration of components in binary liquid mixtures, and these are classified according to the number of adjustable parameters used to account for the deviation from some average. We will consider here some of the most commonly used semi-empirical models for analyzing viscosity of liquid mixtures based on one, two, and three parameters. An attempt has been made to check the suitability of equations for experimental data fits by taking into account the number of empirical adjustment coefficients.

The equation of Tamara and Kurata, Hind et al., and Katti and Chaudhri has one adjustable parameter.

The one-parameter equation due to Tamura and Kurata [42] gave the equation of the form:

$$\eta_m = x_1\eta_1\phi_1 + x_2\eta_2\phi_2 + 2(x_1x_2\phi_1\phi_2)^{\frac{1}{2}}T_{12} \qquad (1.35)$$

where Φ_1 and Φ_2 are the volume fractions of components 1 and 2, respectively; $T_{12}$is Tamura and Kurata constant.

Hind et al. [43] proposed the following equation:

$$\eta_m = x_1^2\eta_1 + x_2^2\eta_2 + 2x_1x_2H_{12} \qquad (1.36)$$

Where H_{12} is attributed to unlike pair interactions.

Frankel[44] proposed the following equation:

$$\ln\eta_m = x_1^2\ln\eta_1 + x_2^2\ln\eta_2 + 2x_1x_2\ln\eta_{12} \qquad (1.37)$$

Where η_m is the viscosity coefficient, x_1 and x_2 are the mole fractions and η_{12} is the interaction parameter.

Katti and Chaudhri [45] derived the following equation :

$$\ln(\eta V) = x_1\ln(\eta_1V_1^0) + x_2\ln(\eta_2V_2^0) + x_1x_2\frac{W_{vis}}{RT} \qquad (1.38)$$

Where W_{vis} is an interaction term and V_1^0 & V_2^0 are the molar volumes of components 1&2 respectively .

Heric and Brewer [46] equation is as follows:

$$\ln\eta_m = x_1\ln\eta_1 + x_2\ln\eta_2 + x_1\ln M_1 + x_2\ln M_2 - \ln(x_1M_1 + x_2M_2) + x_1x_2[\alpha_{12} + \alpha_{21}(x_1 - x_2)] \qquad (1.39)$$

Where(M_1) and (M_2) are molecular weights of components(1)(DMAc)and(2)(Chloroform) and and α_{12} ,α_{21} are interaction parameters, and other terms involved have their usual meaning. α_{12} and α_{21} are parameters, which can be calculated from the least-squares method.

Krishnan and Laddha (1963) have proposed an equation to predict the viscosities of binary liquid mixtures based on Eyring's theory of absolute reaction rates.

$$\ln\eta_m = x_1\ln(\eta_1M_1) + x_2\ln(\eta_2M_2) - \ln(x_1M_1 + x_2M_2) - 2.303x_1x_2\left[B_{12} + C_{12}(x_1 - x_2) + D_{12}(x_1 - x_2)^2\right] \qquad (1.40)$$

The constants B_{12}, C_{12}& D_{12}can be evaluated if the viscosity data is available for binary system at a particular temperature using least squares method.

McAllister's multibody interaction model [47] was widely used to correlate kinematic viscosity data. The two-parameter McAllister equation based on Eyring's [48] theory of absolute reaction rates has taken into account interaction of both like and unlike molecules by two-dimensional three-body model. The three-body interaction model is as follows:

$$\ln\eta_m = x_1^3\ln\eta_1 + 3x_1^2x_2\ln Z_{12} + 3x_1x_2^2\ln Z_{21} + x_2^3\ln\eta_2 - \ln\left(x_1 + \frac{x_2M_2}{M_1}\right) + 3x_1^2x_2\ln\left(\frac{2}{3} + \frac{M_2}{3M_1}\right) + 3x_1x_2^2\ln\left(\frac{1}{3} + \frac{2M_2}{3M_1}\right) + x_2^3\ln\left(\frac{M_2}{M_1}\right) \qquad (1.41)$$

And four-body model was given by

$$\ln\eta_m = x_1^4\ln\eta_1 + 4x_1^3x_2\ln Z_{1112} + 6{x_1}^2x_2^2\ln Z_{1122} + 4x_1x_2^3\ln Z_{2221} + x_2^4\ln\eta_2 - \ln\left(x_1 + \frac{x_2M_2}{M_1}\right) + 4x_1^{32}x_2\ln\left(3 + \frac{M_2}{4M_1}\right) + 6{x_1}^2x_2^2\ln\left(1 + \frac{M_2}{2M_1}\right) + 4x_1x_2^3\ln\left(1 + \frac{3M_2}{4M_1}\right) + x_2^4\ln\left(\frac{M_2}{M_1}\right) \qquad (1.42)$$

where Z_{12}, Z_{21} , Z_{1112} , Z_{1122}, and Z_{2221} are interaction parameters and and M_1, M_2 & η_1, η_2 are the molecular masses and kinematic viscosity of pure component1(DMAc)&2(Chloroform) respectively. The correlating ability of each of (1.36)–(1.46) was tested as well as their adjustable parameters and standard deviations:

$$\sigma = \left[\frac{1}{n-k}\sum_j\left(\frac{\eta_{\exp} - \eta_{cal}}{\eta_{\exp}}\right)^2\right]^{\frac{1}{2}} \qquad (1.43)$$

where(n) represents the number of data points and(k) is the number of numerical coefficients given from **Table(10-12).**

Table-10

T/K	K-T Model		Hind Model		Frankel Model		K-C Model	
	T_{12}	σ	H_{12}	σ	η_{12}	σ	w_{vis}	σ
300K	0.6823	0.1747	-0.3487	1.8774	0.7056	0.1374	-127.2684	0.0279
303K	0.7344	0.1925	-0.3979	1.9676	0.6717	0.1509	-119.2978	0.0257

Table-11

T/K	H-BModel			K-L Model			
	α_{12}	α_{21}	σ	B_{12}	C_{12}	D_{12}	σ
300K	0.04949	0.00523	0.5857	-0.02145	-0.002252	-0.000267	1.2395
303K	0.04950	0.00524	0.5858	-0.02144	-0.002251	-0.000266	1.2396

Table-12

T/K	McAllister's 3D Model			McAllister's 4D Model			
	Z_{12}	Z_{21}	σ	Z_{1112}	Z_{1122}	Z_{2221}	σ
300K	0.7401	0.6164	0.6305	0.8118	0.7083	0.7886	0.6113
303K	0.778	0.6470	0.7321	0.7720	0.6743	0.7515	0.7386

From this study, it can be concluded that the correlating ability significantly improves for these nonideal systems as number of adjustable parameters is increased.

C. *Jouyban and Acree Model*

The Jouyban-Acree model **[49]** was introduced to correlate the physicochemical properties of the solution in mixed solvents including the dielectric constants, viscosity solvatochromic parameter, and density, speed of sound and more recently molar volumes. The model uses the physicochemical properties of the mono-solvents as input data and a number of curve-fitting parameters representing the effects of solvent-solvent interactions in the solution. It is basically derived for representing the solvent effects on the solubility of non-polar solutes in nearly ideal binary solvent mixtures at isothermal conditions. The proposed equation is as follows:

$$\ln y_{m,T} = x_1 \ln y_{1,T} + x_2 \ln y_{2,T} + x_1 x_2 \sum_j \left[\frac{A_j (x_1 - x_2)^j}{T} \right] \qquad (1.44)$$

Where $y_{m,T}$, $y_{1,T}$, $y_{2,T}$ and is density or velocity of the mixture and component(1)(DMAc) and(2) (Chloroform) at given temperature(T), respectively, and x_1, x_2 are the volume fractions of solvents in case of density and mole fraction in case of viscosity, and A_j are the model constant. The predictive ability of the equation (1.44) was tested by calculating the average percentage deviation (APD) between the experimental and calculated density and speed of sound as follows:

$$APD = \frac{100}{N} \sum \left[\frac{y_{exp} - y_{cal}}{y_{exp}} \right] \qquad (1.45)$$

Where N is the number of data points in each set. The optimum numbers of constants Aj, in each case, were determined from the examination of the average percentage deviation value. The constants Aj calculated from the least square analysis are presented in Table (13) along with the average percentage deviation (APD). Finally, it can be concluded that the expressions used for interpolating the experimental data measured in this work gave good results, as can be seen by inspecting the APD values obtained.

Table-13

Property	T/K	A_0	A_1	A_2	A_3	A_4	APD
ῤ(kgm^{-3})	300	29.8262	4.4000	0.7281	0.1322	0.0263	1.4211
ῤ(kgm^{-3})	303	29.6198	4.3335	0.7116	0.1277	0.0246	1.3970
$U(m\text{-}s^{-1})$	300	-10.4218	39.9591	8.6184	-65.0154	5.0960	0.4955
$U(m\text{-}s^{-1})$	303	-16.1779	19.3437	51.9440	-4.7393	-95.7504	0.7421

D. Statistical theory of Arakawa

The statistical theory of Arakawa et al. **[50]** is based on the assumption that the molecules are rigid spheres and the interaction potential between two molecules is a function of the intermolecular separation. Making use of the partition function and the equation of state, Arakawa et al. **[51]** have derived the following excess functions for binary liquid mixtures.

1) Excess energy(E^E)

$$E^E = 3RT\left[\left(\frac{x_1V_1^0}{V_1^0-\beta r_{1A}^*}+\frac{x_2V_2^0}{V_2^0-\beta r_{2B}^*}\right)-\frac{V}{V-\beta(x_1r_A^{*3}-x_2r_B^{*3})}\right] \tag{1.46}$$

2) Excess entropy(S^E)

$$S^E = 3R\left[x_1\ln\left(\frac{V_1^0-\beta r_{1A}^*}{V-\beta(x_1r_A^{*3}-x_2r_B^{*3}}\left(\frac{r_A^*}{r_{1A}^*}\right)^2\right)-x_2\ln\left(\frac{V_2^0-\beta r_{2B}^*}{V-\beta(x_1r_A^{*3}-x_2r_B^{*3})}\right)\left(\frac{r_B^*}{r_{2B}^*}\right)^2\right] \tag{1.47}$$

3) Molecular Radius(r)

$$r = \sqrt[3]{\frac{M}{\rho N}}\sqrt[3]{\frac{3}{16\pi}\left[1-\frac{\gamma RT}{MU^2}\left(\sqrt{1+\frac{MU^2}{\gamma RT}}-1\right)\right]} \tag{1.48}$$

where R and T denote the gas constant and the absolute temperature; x_1, x_2 the mole fractions of the mixtures; V_1^0, V_2^0 the volume per molecules of the pure components, M is the molecular mass ,ῤ is the density,γ is the specific heat ratio, β the common packing parameter; r_A^*, r_B^* the collision diameter of the molecule within each cell; and r_{1A}^*, r_{2B}^* the diameter of each species in the pure state**[52]**. The molecular diameters are related to the collision diameter of the molecules within each cell by the following formulae :

$$r_A^* = x_1r_{1A}^* + x_2r_{AB}^* \tag{1.49}$$

$$r_B^* = x_1r_{1AB}^* + x_2r_{2B}^* \tag{1.50}$$

Where r^*_{AB} can be evaluated as:

$$r_{AB}^* = \left(\frac{r_{1A}^*+r_{2B}^*}{2}\right) \tag{1.51}$$

The packing parameter is given by the following relations:

$$\beta r_{1A}^{*3} = V_1^0\left[\frac{(1+\alpha_1T)}{\left(1+{}^4\!/_3\,\alpha_1T\right)}\right] \tag{1.52}$$

$$\beta r_{2A}^{*3} = V_2^0\left[\frac{(1+\alpha_2T)}{\left(1+{}^4\!/_3\,\alpha_2T\right)}\right] \tag{1.53}$$

Excess energy (E^E), excess entropy(S^E) and molecular radius predicted by statistical theory of Arakawa for the binary mixture DMAc+Chloroform at temperatures 303&303 K are represented in Table-14.

Table-14

System DMAc+Chloroform						
	Excess energy E^E(J-mole^{-1})		Excess entropyS^E(J-mole^{-1}K^{-1})		Molecular radius(Å)	
x_2	300K	303K	300K	303K	300K	303K
0	0.0000	0.0000	0.0000	0.0000	2.0769	2.0785
0.1	-521.6937	-506.8838	0.5798	0.5585	2.0705	2.0714
0.2	-906.4041	-880.2309	0.9981	0.9612	2.0598	2.0643
0.3	-1158.7797	-1124.9442	1.2680	1.2210	2.0515	2.0563
0.4	-1285.9936	-1248.2274	1.4022	1.3502	2.0396	2.0478
0.5	-1297.1816	-1259.0554	1.4129	1.2559	2.0354	2.0383
0.6	-1202.8616	-1167.6363	1.3118	1.2633	2.0235	2.0283
0.7	-1014.3802	-984.9020	1.1099	1.0689	2.0104	2.0182
0.8	-743.4160	-722.0555	0.8175	0.7875	1.9976	2.0070
0.9	-401.5630	-390.1921	0.4445	0.4283	1.9902	1.9957
1.0	0.0000	0.0000	0.0000	0.0000	1.9800	1.9840

It may be concluded that the statistical mechanical theory can be used as a powerful tool to evaluate the excess functions directly from the ultrasonic velocity and density data.

E. Theoretical evaluation of Beyer's non linearity parameter(B/A)

The nonlinearity parameter (B/A) is a basic parameter for determining the degree of waveform distortion. From the knowledge of this parameter, one can gain information about some physical properties of the liquids such as internal pressure, clustering intermolecular spacing, acoustic scattering and structural behaviour etc. Study of nonlinearity parameter (B/A) of liquid mixtures is important in view of the information it may yield on the interaction in liquid mixtures. B/A values of the liquids have been interpreted as the quantity representing the magnitude of the hardness of liquid which may be considered to be true for the liquid mixtures as well.

In the last few years[52-53],a number of theoretical methods have been proposed for estimating the nonlinearity parameter (B/A) for pure liquids and liquid mixtures. This parameter has been further correlated with other thermo acoustical parameters [53-54], which is used to deduce the available volume and intermolecular free length of liquid mixtures. General formulation for non-linearity in terms of acoustical parameters of liquids has been made using the experiment for sound velocity (U) and introducing the contribution due to isobaric acoustic parameters (k) and the isothermal acoustic parameter ($k^{\approx}$). The expression for B/A is given by[35].

$$\left(\frac{B}{A}\right) = 2k + 2\gamma k^{\approx} \qquad (1.54)$$

Computations of k and $k^{\approx}$ require only the knowledge of isobaric thermal expansion co-efficient(α).

F. Ballou's Method

According to the empirical rule of Ballou**[59]** ,there is a linear relation between the nonlinearity parameter of liquids and reciprocal sound speed as follows:

$$\left(\frac{B}{A}\right) = -0.5 + \left(\frac{1.2*10^4}{U}\right) \qquad (1.55)$$

Where U is the sound velocity expressed in ms^{-1}.

G. Hartmann–Balizar(B&H) Method

Hartmann & Balizar **[60]** has shown theoretically that the intermolecular potential energy is the dominant factor in determining sound speed, and its derivatives in liquids. The expression for nonlinearity parameter due to Hartmann is as follows :

$$\left(\frac{B}{A}\right) = 2 + \left(\frac{0.98 * 10^4}{U}\right) \qquad (1.56)$$

Where U is the sound velocity expressed in ms^{-1}.

The present work deals with the computation of acoustic nonlinearity parameter (B/A) of binary liquid mixture at temperature of 300 K using two different approaches. The different methods used are Ballou's empirical relation and Hartmann's relation.

The excess nonlinearity parameter,$(B/A)^E$ for binary liquid mixtures is given by:

$$\left(\frac{B}{A}\right)^E = \left(\frac{B}{A}\right)_{mix} - \left(\frac{B}{A}\right)_{ideal} \qquad (1.57)$$

where $(B/A)_{mix}$ is the non-linearity parameter of the mixture and is that obtained from the ideal mixture relation:

$$\left(\frac{B}{A}\right)_{ideal} = x_1\left(\frac{B}{A}\right)_1 + x_2\left(\frac{B}{A}\right)_2 \qquad (1.58)$$

where x_1 and x_2, $(B/A)_1$ &$(B/A)_2$are the mole fractions& nonlinearity parameters(B/A) of the component (1)-(DMAc)&(2)-(Chloroform) respectively.

The various molecular parameters of binary liquid mixture are calculated using following Sehgal's relations**[61]** :

1) *Cohesive Energy* (1.59)

$$\Delta A = \frac{MU^2}{\left(\frac{B}{A}+1\right)} \ln\left(\frac{82.051T\rho}{M}\right)$$

2) *Vander Waal's constant* (1.60)

$$a = \left[\frac{\rho U^2 V_m^2}{\left(\frac{B}{A}+1\right)}\right]$$

3) *Distance Of Closest Approach* (1.61)

$$d = \left[\frac{3}{2N\pi}\left(V_m - \frac{RT\left(\frac{B}{A}+1\right)}{\rho U^2}\right)\right]^{\frac{1}{3}}$$

where M is the molecular weight of the binary mixture calculated by ideal mixture relation.

$$M = x_1 M_1 + x_2 M_2 \qquad (1.62)$$

where $x_1$and x_2 are the mole fractions and M_1,M_2 are the molecular weight of the component (1)-(DMAc)&(2)-(Chloroform) respectively. V_m is the molar volume given by:

$$V_m = \frac{M}{\rho_{mix}} \qquad (1.63)$$

where $\dot{\rho}_{mix}$ is the density of the mixture, k is the Boltzmann's constant, N the Avogadro's number, R the universal gas constant.

Table-15

Ballou method					Hartmann–Balizar (B&H) method				
(B/A)	(B/A)^E	ΔA(KJ-mole^{-1})	a(N-m^4-mole^{-2})	d(Á)	(B/A)	(B/A)^E	ΔA(KJ-mole^{-1})	a(N-m^4-mole^{-2})	d(Á)
7.8799	0.0000	389.620	1.9237	4.0491	8.8436	0.0000	351.477	1.7354	4.0283
8.2070	-0.0810	361.117	1.7317	4.0069	9.1107	-0.0662	328.840	1.5769	3.9863
8.5909	-0.1052	329.601	1.5487	3.9635	9.4242	-0.0859	303.253	1.4250	3.9434
8.9310	-0.1732	306.145	1.4092	3.9240	9.7019	-0.1415	284.090	1.3076	3.9045
9.2672	-0.2451	285.531	1.2975	3.8876	9.9766	-0.2002	267.080	1.2136	3.8689
9.8475	-0.0730	248.615	1.1045	3.8272	10.4505	-0.0596	235.524	1.0463	3.8094
10.3350	-0.0066	223.949	0.9858	3.7774	10.8488	- 0.0054	214.243	0.9430	3.7612
10.6790	-0.0573	210.560	0.9221	3.7427	11.1299	-0.0468	202.741	0.8879	3.7279
11.0897	-0.0551	195.013	0.8504	3.7016	11.4649	-0.0450	189.144	0.8248	3.6887
11.5156	-0.0373	180.362	0.7785	3.6555	11.8128	-0.0305	176.180	0.7605	3.6447
11.9610	0.0000	166.554	0.7152	3.6084	12.1765	0.0000	163.830	0.7035	3.6001

In the case of binary mixture of hard spheres in which the molecular diameters differ by a small amount (molecular radius of DMAc is 2.077 Á and that of Chloroform is 1.980Á), Harumi Endo pointed that (B/A) is closely connected with the degree of packing of hard spheres or hardness of the bulk of the liquid system. In the present system, as the mole fraction of chloroform increases, the degree of packing increases and hence(B/A) increases. Negative value of (B/A)^E indicates strong interaction between unlike molecules. Cohesive energy(ΔA) is the measure of the total molecular cohesion and it represents the total strength or stiffness of the solvent structure. In the binary liquid mixture, the magnitude of the cohesive energy decrease with increase in the mole fractions of Chloroform. The effective Vander Waal's constant 'a' represents the effective volume of a molecule. In the present study, the constants 'a' decreases with increase in the mole fraction of Chloroform. The decrease in the value of constant 'a' indicates that the attractive forces in the binary liquid mixtures decrease with the increase in mole fraction of Chloroform. The variation in the values of the distance of closest approach of molecules also have the same nature of variation as those of ‘a’.

IV. CONCLUSION

On the basis of the experimental values of density, viscosity and ultrasonic velocity, related derived acoustic parameters and their excess values for the binary mixture under investigation, it is concluded that there exists dipole-dipole, dipole-induced dipole and dispersion interaction in the binary liquid mixture of DMAc with Chloroform. Further, it is concluded that the molecular interaction becomes more dominant with the increase in mole fraction of chloroform, for a fixed frequency and decrease with the rise in temperature for a fixed concentration of DMAc in the binary mixture. The temperature dependence of the different parameters suggests that the degree of deviation from ideality is strongly temperature sensitive. It can also be observed that the PFP theory reproduces the main features of the experimental data by using only one adjustable parameter, χ_{12} to describe V^E_m for the {DMAc+Chloroform} binary mixtures over the entire composition range. Several empirical and semiempirical relations have been used to represent the dependence of viscosity on concentration of components in binary liquid mixtures. An attempt has been made to check the suitability of empirical and semiempirical relations for experimental viscosities data of DMAc+Chloroform fits by taking into account the number of empirical adjustment coefficients. Also Jouyban - Acree model shows that the good agreement between the estimated and experimental values occurs for given binary systems. Non-linear parameter (B/A) is evaluated using empirical equations and certain molecular properties are calculated with the use of Sehgal's equations. Negative values of excess nonlinear parameter also support the existence of dipole-induced dipole along with weak interaction between unlike molecules.

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