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Study of Internal Pressure and Related Parameters in Ternary Liquid Mixtures Using Ultrasonic Velocity Measurement

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Abstract

Ultrasonic velocity(U), density(ρ) and viscosity(η) measurements have been taken in two ternary liquid mixtures Ethylene glycol + cyclohexanone + Isopropyl acetate and Ethylene glycol + cyclohexanone + Isobutyl acetate with fixed mole fraction (0.4) of cyclohexanone, at different temperatures with increasing mole fraction of ethylene glycol. The experimental data has been used to calculate the free volume, molar volume, internal pressure, enthalpy, Gibbs free energy, Molar sound velocity and their excess values. They have been analysed to discuss the nature and strength of intermolecular interactions in these mixtures. The compositional variation of excess parameters have been correlated using the Redlich-Kister equation.

Keywords: *Ternary mixtures, internal pressure, molar sound velocity, excess parameters intermolecular interactions*

1. Introduction

The study of thermodynamic properties of binary and ternary mixtures contributes to an understanding of the behavior of different liquids and functional groups. Number of studies on the thermodynamic properties of ternary mixtures has increased in recent years due to industrial applications[1-5]. A third component added to a mixture of two immiscible components brings about the phenomenon in certain ternary liquid mixtures has been studied earlier [6-9] by observing on the distribution of molecules of one component between the other two components and the formation of hitherto unknown compounds from a study of the change in free volume, molar volume, internal pressure and other thermodynamic parameters.

The measurement of ultrasonic velocity has been adequately employed in understanding the molecular interactions in liquid mixtures. Molecular interaction studies can be carried out by both spectroscopic and non–spectroscopic techniques. However, ultrasonic velocity and viscosity measurements have been widely used in the field of interactions and structural aspect evaluation studies.

Internal pressure has gained significant interest by chemists, physicists and chemical engineers in past, as it provides a measure of explaining molecular interactions, internal structure, clustering phenomenon and dipolar interactions. Internal pressure has been a subject of active interest among several researchers during recent past [7–10]. Several attempts have been made by a number of investigators [11–15] to calculate the internal pressure of liquids and liquid mixtures theoretically.

The measurement of internal pressure is important in the study of the thermodynamic properties of liquids. The internal pressure is the cohesive force, which is a resultant of force of attraction and force of repulsion between the molecules. Cohesion creates pressure within the liquid. So internal pressure gives an idea of the solubility characteristics. Dissolved solutes exist under the internal pressure of the medium and their interactions with the solvent arise through hydrogen bonding, charge transfer, Columbic (or) Vanderwaal's interaction.

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2. Experimental Details

Ultrasonic velocity (U), measurements were made with a single crystal variable path ultrasonic interferometer (Model F-81; Mittal enterprises) at 2 MHz frequency with an accuracy of ±0.01%.The densities (ρ)were measured using specific gravity bottles by relative measurement method with accuracy of ±0.01kgm^{-3} and compared at the working temperature with the corresponding literature values. The viscosity (η) measurements were made with Ostwald viscometer with an accuracy of ±0.001Nsm^{-2}.

The chemicals used in the present work are AR grade of purity >99% from E-Merck and Hi-Media and they were used without further purification.

1. Monoethylene glycol + Cyclohexanone + Isopropyl acetate (MEG+CY+IPA)
2. Monoethylene glycol + Cyclohexanone + Isobutyl acetate (MEG+CY+IBA).

Actually monoetyhlene glycol and isopropyl acetate/ isobutyl acetate liquids are immiscible and by the addition of cyclohexanone only the ternary mixtures are formed. In all the four systems, the mole fraction of the second component, cyclohexanone(CY) is kept fixed at 0.4(X_2), while the mole fractions of the remaining two have been varied from 0.0 to 0.6 (X_1 and X_3) so as to have the mixtures of different compositions. There is no significance in fixing mole fraction of the second liquid component (CY) at X_2 = 0.4.

3. Theoretical Aspects

3.1. Molar Volume of the Liquid Mixture (V_m)

The molar volumes of the mixtures have been computed using its relationship with the molecular weight and mole fraction of the constituent is given below

$$V_m = \frac{M_{eff}}{\rho_{mix}} \tag{1}$$

Where $M_{eff} = \frac{M_1X_1 + M_2X_2 + M_3X_3}{X_1 + X_2 + X_3}$ here M_1, M_2, M_3 are the molecular weights of corresponding liquids and X_1, X_2, X_3 are their mole fractions.

3.2. Free Volume (V_f)

The free volumes of the mixtures have been computed using its relationship with the ultrasonic velocity and viscosity is given below

$$V_f = \left(\frac{M_{eff}U}{k\eta} \right)^{3/2} \tag{2}$$

where k is a constant, independent of temperature and it's value is 4.28 X 10^9 for all liquids.

3.3. Internal Pressure (π_i)

On the basis of dimensional analysis, using free volume concept, the following expression can be used for calculating internal pressure

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

here b is packing factor (b=2), k is a constant, independent of temperature and it's value is 4.28 X 10^9 for all liquids, R is universal gas constant and T is absolute temperature.

3.4. Enthalpy (H)

The enthalpy of the liquid mixture is given by

$$H = \pi_i V_m \tag{4}$$

3.5. Gibb's Free Energy for Activation of Flow (ΔG)

The Gibb's free energy for activation of flow (**ΔG**) can be calculated using relation

$$\Delta G = RT \ln(\eta V_m) \tag{5}$$

where R is the Universal gas constant (8.31432×10^7 JK^{-1}), and T is the absolute temperature

3.6. Molar Sound Velocity or Rao's Constant (R)

Rao's constant can be calculated by the relation

$$R = V_m U^{1/3} \tag{6}$$

3.7. Excess Thermodynamic Parameters

The excess thermodynamic functions (Y^E) provide a way to represent directly the deviation of a solution from ideal behaviour. The difference between the thermodynamic function of mixing for a real system and the value corresponding to a perfect solution at the same temperature, pressure and composition is called the thermodynamic excess function, denoted by Y^E. Excess values for all the parameters are computed using the general formula

$$Y^E = Y_{exp} - \left(X_1 Y_1 + X_2 Y_2 + X_3 Y_3 \right) \tag{7}$$

The excess properties Y^E were fitted by non linear least square method to Redlich - Kister type polynomial

$$Y^E = x_1 (1 - x_1) \sum_{i=0}^{p} A_i (2x_1 - 1)^i \tag{8}$$

In each case, the optimum number of coefficients A_i was determined from an examination of the variation of standard deviation (σ) as calculated by

$$\sigma\left(Y^E\right) = \left[\frac{\sum \left(Y^E_{\exp} - Y^E_{cal} \right)}{(n - p)} \right]^{\frac{1}{2}} \tag{9}$$

where 'n' represents the number of experimental points and 'p' is the number of coefficients.

4. Results and Discussions

The two ternary systems contain an aliphatic cyclic compound *i.e.*, cyclohexanone along with straight or branch chained liquids. An exhaustive survey of literature reveals that a few researchers studied the ternary mixtures using ultrasonics. Hence a systematic study has been taken up for two ternary mixtures at four temperatures 303, 308, 313 and 318K.

The measured values of density, ultrasonic velocity, viscosity and the evaluated parameters are presented in Table 1.1 for the ternary system-I and Table 2.1 for the ternary system-II. From tables it can be noticed that, at all the four temperatures, the values of density(ρ), ultrasonic velocity(U) and viscosity (η) increase with increase in mole fraction of monoethylene glycol. This type of trend is observed by S Thirumaran *et.*

al., [16] for the ternary mixtures they studied. It is evident that the pronounced increase or decrease in these parameters with composition of mixtures indicates the presence of interactions between the component molecules in the ternary mixtures. It can also be observed that the ultrasonic velocity is increasing with increase in mole fraction of monoethylene glycol and is decreasing with increase in temperature, for the ternary systems under study. This trend indicates specific interactions among the constituents of the mixtures. This behaviour at such concentrations which is different from the ideal mixture behaviour can be attributed to intermolecular interactions [17, 18]. The chemical interaction may involve the association due to hydrogen bonding or due to dipole-dipole interaction or may be due to the formation of charge-transfer complexes. All these processes may lead to strong interaction forces [19]. The increase in velocity in these liquid mixtures suggest that molecular interactions among the molecules of the components of liquid mixture.

An analysis of the viscosity values from the tables it can be observed that the viscosity is in increasing trend with increase in mole fraction of monoethylene glycol and is decreasing with increase of temperature. Similar trend is also observed for the density values. This kind of non-linearity indicates the presence of molecular interactions.

It can be observed that the values of molar volume (V_m) decreases with increase in concentration of MEG in both the ternary liquid systems. This is because of the fact, that molecular weight is directly proportional to the molar volume. The molecular weights of components of the mixtures are IPA-102.13, IBA-116.16 (alkyl acetates), CY-98.15, MEG-62.07. Since alkyl acetates have higher molecular weights in the solution comparing with others, whose concentration is decreasing in the mixture supports the calculation. Moreover, molar volume (V_m) also increases with rise in temperature in the present study, which probably from the fact that thermal energy facilitates an increase in the molecular separation in the liquid mixtures and thus leads to an increase in molar volume (V_m) with increase of temperature [20].

It is observed that for the ternary liquid systems, the free volume (V_f) decrease and the internal pressure (π_i) increase with increase in concentration of MEG. Further, the decrease in free volume and increase in internal pressure with rise in concentration of MEG in all the systems clearly show the increasing magnitude of interactions. Similar trend was observed by R Thyagarajan *et al.*, [21] also supports the present discussion.

Here the variation of enthalpy (H) with mole fraction of MEG follows the same trend as that of the internal pressure. The increase in enthalpy with the increase in MEG and also with increase in temperature suggests specific interactions among the molecules of the liquid mixtures.

The Gibb's free energy of activation flow (ΔG) for the ternary mixtures under study are shown in the tables, form which it is inferred that the values of ΔG are negative. The decrease in Gibb's free energy in all the systems indicates that, the need for shorter time for the cooperative process or the rearrangement of the molecules in the mixtures.

Rao's constant (R) constant show a decreasing trend with increase of mole fraction of MEG as can be seen from Figure 1.3, Figure 2.3. More over in both the cases the values increase with increase in temperature. These non-linear trends may be attributed to possibility of molecular interactions that are taking place.

4.1. Excess Acoustical and Thermodynamic Parameters

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 parameters rather than actual values. Non-ideal liquid mixtures show considerable deviation from linearity in their concentrations and this can be interpreted as the presence of strong or weak interactions.

Table 1.1. Ternary Liquid Mixture – I: MEG +CY + IPA Density, Ultrasonic Velocity, Viscosity and Related Parameters at Temperatures 303,308,313 and 318K

MEG X_1	IPA X_3	ρ Kgm^{-3}	U ms^{-1}	η x 10^{-3} Ns/m^2	V_m x 10^{-5} m^3/mol	V_f x 10^{-8} m^3/mol	π_i x 10^{16} N/m^2	H x 10^{12} Joule/ mol	ΔG x 10^{11} Joule/ mol	R x 10^{-7} m^3/mol $(m/s)^{1/3}$
303K										
0.000				0.6652			3.3594	3.8407	-4.13	2.564
0.108	0.600	879.4	1149.1	0.7502	11.432	25.848	3.7383	4.0119	-4.11	2.391
0.194	0.492	896.6	1189.8	0.8553	10.731	21.289	4.1632	4.2157	-4.09	2.234
0.267	0.405	915.8	1226.2	0.9712	10.126	17.312	4.5954	4.4267	-4.07	2.109
0.379	0.333	932.8	1260.6	1.2620	9.6327	14.224	5.5233	4.9117	-4.03	1.928
0.423	0.220	959.6	1328.3	1.4620	8.8926	9.6132	6.0780	5.2147	-4.00	1.850
0.497	0.176	973.9	1361.3	2.0640	8.5795	7.7502	7.4701	6.0287	-3.93	1.726
0.527	0.103	999.3	1430.2	2.5830	8.0705	4.7180	8.4850	6.6545	-3.88	1.669
0.553	0.073	1012.9	1463.2	3.3571	7.8427	3.4092	9.7948	7.4896	-3.82	1.622
0.578	0.046	1024.8	1496.4	4.9421	7.6464	2.3314	12.034	8.9822	-3.73	1.576
0.600	0.021	1036.7	1526.3	10.190	7.4636	1.3193	17.464	12.731	-3.55	1.533
	0.000	1049.5	1559.6		7.2899	0.4525				
308K										
0.000				0.6210	11.741		3.3279	3.8404	-4.21	1.406
0.108	0.600	871.2	1114.8	0.6981	11.061	33.277	3.6874	4.0019	-4.20	1.324
0.194	0.492	886.6	1158.6	0.7890	10.471	28.224	4.0761	4.1851	-4.18	1.246
0.267	0.405	903.2	1197.4	0.8940	10.001	24.034	4.4803	4.3858	-4.16	1.186
0.379	0.333	917.9	1234.6	1.1441	9.2804	20.463	5.3307	4.8279	-4.12	1.097
0.423	0.220	942.2	1303.5	1.3030	8.9672	15.032	5.8091	5.0836	-4.09	1.055
0.497	0.176	954.8	1336.6	1.8071	8.4608	12.766	7.0579	5.8172	-4.02	0.990
0.527	0.103	978.5	1409.2	2.2050	8.2396	8.7504	7.8949	6.3375	-3.98	0.962
0.553	0.073	989.6	1445.2	2.7551	8.0460	7.0800	8.9394	7.0022	-3.93	0.937
0.578	0.046	1000.4	1475.2	3.7220	7.8721	5.4381	10.504	8.0526	-3.86	0.913
0.600	0.021	1009.3	1504.4	8.7931	7.7187	3.9115	16.308	12.247	-3.64	0.892
	0.000	1018.8	1532.2			1.6261				
313K										
0.000				0.5530	11.629	31.808	3.2017	3.7234	-4.31	1.387
0.108	0.600	864.5	1096.4	0.6181	10.954	26.872	3.5246	3.8609	-4.29	1.304
0.194	0.492	878.4	1143.8	0.6920	10.368	22.556	3.8759	4.0187	-4.28	1.226
0.267	0.405	894.4	1183.8	0.7780	9.8969	18.943	4.2376	4.1939	-4.26	1.165
0.379	0.333	907.9	1222.5	0.9791	9.1648	13.564	4.9856	4.5692	-4.22	1.073
0.423	0.220	931.1	1296.2	1.1021	8.8550	11.404	5.4048	4.7860	-4.20	1.031
0.497	0.176	943.6	1327.5	1.4860	8.3547	7.4678	6.4702	5.4057	-4.14	0.967
0.527	0.103	965.3	1398.5	1.7621	8.1283	5.8800	7.1364	5.8007	-4.10	0.938
0.553	0.073	977.3	1435.5	2.1330	7.9344	4.4692	7.9467	6.3052	-4.05	0.912
0.578	0.046	987.6	1467.2	2.8112	7.7639	2.9819	9.2269	7.1637	-3.99	0.890
0.600	0.021	996.6	1495.2	5.7222	7.5878	1.0331	13.339	10.121	-3.81	0.864
	0.000	1008.3	1518.4							
318K										
0.000				0.5281			3.1839	0.3512	-4.38	1.406
0.108	0.600	856.3	1079.8	0.5901	11.741	33.277	3.5000	0.2110	-4.37	1.324
0.194	0.492	869.9	1129.6	0.6560	11.061	28.224	3.8300	0.1138	-4.36	1.246
0.267	0.405	885.6	1170.4	0.7320	10.471	24.034	4.1665	0.0410	-4.34	1.186
0.379	0.333	898.4	1210.8	0.9070	10.001	20.463	4.8539	-0.058	-4.30	1.097
0.423	0.220	919.5	1286.3	1.0150	9.2804	15.032	5.2443	-0.093	-4.28	1.055
0.497	0.176	931.8	1318.2	1.3281	8.9672	12.766	6.1830	-0.147	-4.23	0.990
0.527	0.103	953.2	1389.2	1.5490	8.4608	8.7504	6.7536	-0.100	-4.19	0.962
0.553	0.073	964.1	1428.3	1.8621	8.2396	7.0800	7.4922	-0.185	-4.15	0.937
0.578	0.046	973.9	1459.8	2.3272	8.0460	5.4381	8.4848	-0.201	-4.10	0.913
0.600	0.021	982.9	1483.2	4.1952	7.8721	3.9115	11.518	-0.215	-3.95	0.892
	0.000	991.2	1506.3		7.7187	1.6261				

Table 2.1. Ternary Liquid Mixture – II: MEG +CY + IBA Density, Ultrasonic Velocity, Viscosity and Related Parameters at Temperatures 303,308,313 and 318K

MEG X_1	IBA X_3	ρ Kgm^{-3}	U ms^{-1}	η x 10^{-3} Ns/m^2	V_m x 10^{-5} m^3/mol	V_f x 10^{-8} m^3/mol	π_i x 10^{16} N/m^2	H x 10^{12} Joule/ mol	ΔG x 10^{11} Joule/ mol	R x 10^{-7} m^3/mol $(m/s)^{1/3}$
303K										
0.000	0.600	875.4	1195.6	0.7252	12.447	27.194	3.1210	3.8849	-4.088	1.509
0.096	0.503	891.6	1206.8	0.7823	11.633	22.857	3.4597	4.0247	-4.086	1.389
0.248	0.352	922.8	1251.7	0.9405	10.354	16.196	4.1938	4.3426	-4.069	1.209
0.308	0.291	938.2	1283.5	1.0530	9.8335	13.467	4.6161	4.5392	-4.053	1.139
0.360	0.239	953.3	1323.2	1.2020	9.3828	11.034	5.0898	4.7757	-4.032	1.080
0.407	0.192	967.5	1363.2	1.3920	8.9839	8.8693	5.6351	5.0625	-4.006	1.029
0.479	0.120	992.5	1430.6	1.9221	8.3624	5.4837	6.9385	5.8022	-3.942	0.949
0.517	0.082	1006.9	1468.4	2.4830	8.0388	3.7402	8.0925	6.5054	-3.888	0.907
0.548	0.051	1020.8	1497.6	3.2571	7.7682	2.4865	9.4863	7.3692	-3.828	0.870
0.574	0.025	1034.7	1527.4	4.6420	7.5265	1.4649	11.557	8.6984	-3.747	0.837
0.600	0.000	1049.5	1559.6	10.190	7.2893	0.4524	17.465	12.731	-3.557	0.805
308K										
0.000	0.600	869.2	1156.5	0.6586	12.536	29.892	3.0594	3.8355	-4.17	1.513
0.096	0.503	883.6	1169.8	0.7112	11.738	25.165	3.3854	3.9739	-4.17	1.399
0.248	0.352	911.2	1211.5	0.8513	10.486	17.908	4.0879	4.2868	-4.15	1.226
0.308	0.291	923.9	1242.3	0.9434	9.9857	15.123	4.4684	4.4620	-4.14	1.161
0.360	0.239	936.2	1277.6	1.0641	9.5542	12.570	4.8944	4.6762	-4.12	1.107
0.407	0.192	948.8	1317.4	1.2082	9.1610	10.420	5.3583	4.9087	-4.10	1.058
0.479	0.120	971.5	1382.8	1.6031	8.5432	6.8417	6.4587	5.5178	-4.04	0.979
0.517	0.082	985.6	1419.6	2.0151	8.2125	4.8633	7.4299	6.1019	-4.00	0.936
0.548	0.051	997.4	1456.3	2.6350	7.9505	3.2767	8.6605	6.8855	-3.94	0.903
0.574	0.025	1008.3	1488.2	3.5220	7.7235	2.1318	10.189	7.8699	-3.87	0.874
0.600	0.000	1018.8	1532.2	8.7910	7.5090	0.5499	16.309	12.247	-3.64	0.849
313K										
0.000	0.600	860.5	1132.2	0.6027	12.663	33.076	2.9858	3.7811	-4.26	1.533
0.096	0.503	874.4	1146.8	0.6453	11.861	28.263	3.2867	3.8987	-4.26	1.419
0.248	0.352	900.4	1189.6	0.7566	10.612	20.797	3.9210	4.1611	-4.25	1.248
0.308	0.291	912.9	1216.4	0.8281	10.106	17.817	4.2652	4.3104	-4.24	1.181
0.360	0.239	924.5	1252.4	0.9169	9.6751	15.251	4.6245	4.4742	-4.22	1.128
0.407	0.192	936.1	1289.2	1.0320	9.2853	12.778	5.0418	4.6814	-4.20	1.079
0.479	0.120	957.3	1353.6	1.3041	8.6699	9.0313	5.9249	5.1368	-4.16	1.001
0.517	0.082	969.3	1393.8	1.6020	8.3506	6.6743	6.7193	5.6110	-4.12	0.962
0.548	0.051	980.6	1426.6	2.0330	8.0867	4.6879	7.7226	6.2451	-4.06	0.928
0.574	0.025	991.6	1462.2	2.6911	7.8536	3.1087	9.0302	7.0920	-4.00	0.898
0.600	0.000	1006.3	1518.4	5.7221	7.6023	1.0330	13.323	10.128	-3.81	0.868
318K										
0.000	0.600	851.3	1109.4	0.5590	12.800	35.914	2.9303	3.7509	-4.35	1.556
0.096	0.503	864.9	1125.8	0.5980	11.992	30.813	3.2208	3.8625	-4.35	1.442
0.248	0.352	889.5	1170.3	0.6956	10.742	23.020	3.8193	4.1034	-4.34	1.272
0.308	0.291	901.4	1195.5	0.7582	10.235	19.814	4.1473	4.2447	-4.33	1.205
0.360	0.239	912.5	1227.6	0.8331	9.8023	17.089	4.4843	4.3956	-4.31	1.150
0.407	0.192	923.8	1264.8	0.9263	9.4089	14.602	4.8564	4.5693	-4.29	1.101
0.479	0.120	943.2	1328.2	1.1491	8.7995	10.611	5.6483	4.9702	-4.26	1.025
0.517	0.082	954.9	1366.9	1.3790	8.4765	8.1163	6.3322	5.3675	-4.22	0.985
0.548	0.051	965.9	1402.6	1.6921	8.2098	6.0191	7.1464	5.8670	-4.17	0.951
0.574	0.025	977.9	1438.3	2.2270	7.9636	4.0284	8.3374	6.6396	-4.11	0.919
0.600	0.000	991.2	1506.3	4.1951	7.7181	1.6259	11.519	8.8910	-3.95	0.892

The extent of deviation depends upon the nature of the constituents and composition of the mixtures. The thermodynamic excess properties are found to be more sensitive towards intermolecular interaction among the component molecules of liquid mixtures. The sign and extent of deviation of excess parameters depend on the strength of interaction between unlike molecules [22].

So various excess acoustic and thermodynamic parameters have been evaluated and corresponding graphs are also given. More over the excess parameters were fitted to Redlich – Kister polynomial and evaluated the coefficients by least square method. These coefficients are reported in the Tables 1.3 and 2.3 followed by the graphs for excess values for the ternary mixtures under study.

The sign and magnitude of excess ultrasonic velocity(U^E) play an important role in describing molecular rearrangement as a result of the molecular interactions between the component molecules in the mixtures. The excess ultrasonic velocity (U^E) curves at different temperatures varying with mole fraction of monoethylene glycol are represented in the figures, Figure 1.1, and Figure 2.1 for the two ternary systems. The U^E values starts from negative value reaches negative maximum at around 0.3 mole fraction of MEG and then begins to increase and reaches to positive value at the mole fraction 0.6 of MEG.

Actually the negative trend describes weak interactions among the molecules of the components of the systems. Even with the addition of the third component the same trend is observed, but with the increase in mole fraction of MEG, resultant interactions gradually become stronger, after the mole fraction of 0.3 of MEG. The reason may be stated as, the MEG is self-associated and more polar than the alkyl acetates and the reduction of mole fraction of the IPA/IBA. After the particular mole fraction of MEG, as the concentration of alkyl acetates is decreasing the specific interactions play dominant role than the rupture of hydrogen bond.

Table 1.2. Ternary Liquid Mixture – I : MEG +CY + IPA Excess Values of Acoustic and Thermodynamic Parameters at Temperatures 303,308,313 and 318K

MEG X_1	IPA X_3	U^E m/sec	η^E x 10^{-3} Ns/m^2	$V_m^E x10^{-7}$ m^3/mole	V_f^E x 10^{-8} m^3/mole	$\pi_i^E x10^{14}$ N/m^2	H^E x10^{11} Joule/mol	ΔG^E x 10^{10} Joule/mol
303K								
0.0000	0.6000	-72.240	-0.3596	16.419	2.2999	-0.5641	-0.4988	-0.6143
0.1080	0.4921	-91.146	-0.1476	12.473	1.3767	-2.4587	-1.4011	-1.1006
0.1948	0.4052	-102.45	-0.2331	7.4090	0.3282	-3.8604	-2.0594	-1.4305
0.2672	0.3330	-108.15	-0.3028	4.0872	-0.3265	-4.9526	-2.5686	-1.6529
0.3798	0.2203	-102.38	-0.3991	-0.5311	-1.1402	-6.3944	-3.2021	-1.8596
0.4239	0.1761	-93.566	-0.4281	-4.1765	-1.5137	-6.7676	-3.3369	-1.8423
0.4970	0.1032	-65.156	-0.4493	-9.6740	-2.0894	-6.9147	-3.2500	-1.5482
0.5272	0.0730	-48.793	-0.4311	-13.596	-2.3805	-6.5354	-2.9242	-1.2324
0.5536	0.0464	-29.918	-0.3830	-16.500	-2.5619	-5.7806	-2.3508	-0.7990
0.5782	0.0218	-13.570	-0.2519	-19.421	-2.7450	-4.0586	-1.1027	-0.2979
0.6001	0.0000	7.5552	0.2484	-23.229	-2.8772	0.9095	2.4283	1.6103
308K								
0.0000	0.6000	-67.200	-0.300	18.920	2.5445	-0.5517	-0.4820	-0.6028
0.1080	0.4921	-85.303	-1.300	15.225	1.7331	-2.3280	-1.3630	-1.0947
0.1948	0.4052	-96.065	-2.100	11.355	0.8707	-3.6555	-2.0170	-1.4424
0.2672	0.3330	-100.50	-2.700	8.8623	0.1909	-4.6835	-2.5155	-1.6667
0.3798	0.2203	-95.927	-3.600	3.8869	-0.6051	-6.0594	-3.1596	-1.9067
0.4239	0.1761	-87.956	-3.900	0.5482	-0.9075	-6.4528	-3.3290	-1.9273
0.4970	0.1032	-57.395	-4.100	-5.6789	-1.6572	-6.6500	-3.3015	-1.6684
0.5272	0.0730	-38.676	-4.000	-8.5884	-1.9791	-6.4102	-3.0726	-1.4053
0.5536	0.0464	-23.570	-3.700	-11.566	-2.2113	-5.8872	-2.6620	-1.0628
0.5782	0.0218	-8.4460	-3.000	-13.118	-2.4354	-4.8090	-1.8489	-0.4931
0.6001	0.0000	6.7169	1.860	-15.438	-3.0894	0.5619	2.1337	1.5299
313K								
0.0000	0.6000	-65.060	-0.300	20.158	3.3458	-0.5558	-0.4882	-0.6337
0.1080	0.4921	-80.825	-0.900	17.428	2.7483	-1.8441	-1.1150	-1.0411
0.1948	0.4052	-91.404	-1.300	13.393	1.9260	-2.7872	-1.5709	-1.3289
0.2672	0.3330	-95.186	-1.600	10.377	1.2166	-3.5061	-1.9085	-1.5018
0.3798	0.2203	-87.129	-2.100	6.2776	0.3696	-4.4373	-2.3293	-1.6763
0.4239	0.1761	-81.475	-2.200	2.4488	-0.1309	-4.6756	-2.4241	-1.6841
0.4970	0.1032	-53.366	-2.300	-3.0316	-1.0188	-4.7011	-2.3222	-1.4170

0.5272	0.0730	-34.000	-2.200	-7.1644	-1.3922	-4.4854	-2.1409	-1.1972
0.5536	0.0464	-17.507	-2.000	-10.140	-1.7333	-4.0683	-1.8224	-0.9044
0.5782	0.0218	-3.8710	-1.400	-12.120	-2.2314	-3.1551	-1.1380	-0.3663
0.6001	0.0000	6.4378	1.340	-16.430	-3.3037	0.6307	1.6649	1.3184
318K								
0.0000	0.6000	-62.960	-0.299	22.515	3.7816	-0.5418	-0.4692	-0.6244
0.1080	0.4921	-77.522	-0.702	19.254	3.1392	-1.5439	-0.9431	-0.9538
0.1948	0.4052	-88.266	-0.900	14.786	2.5009	-2.2727	-1.2912	-1.2010
0.2672	0.3330	-91.148	-1.201	11.883	1.8805	-2.8202	-1.5420	-1.3385
0.3798	0.2203	-82.543	-1.401	7.7990	1.0560	-3.5066	-1.8367	-1.4632
0.4239	0.1761	-76.780	-1.506	4.6023	0.5963	-3.6540	-1.8861	-1.4529
0.4970	0.1032	-49.479	-1.500	-1.5370	-0.4405	-3.6078	-1.7687	-1.2045
0.5272	0.0730	-28.348	-1.400	-5.1663	-0.8767	-3.4058	-1.6050	-0.9989
0.5536	0.0464	-12.352	-1.200	-8.1243	-1.4312	-2.9887	-1.2890	-0.6986
0.5782	0.0218	-3.5890	-0.800	-10.461	-1.9524	-2.2963	-0.7761	-0.2737
0.6001	0.0000	6.3786	0.950	-12.517	-3.3469	0.4699	1.3120	1.1422

Table 1.3. Values of the Coefficients of Redlich - Kister Polynomial and Standard Deviation (σ) for Ternary Mixture - I

Temp. , K	A_0	A_1	A_2	A_3	σ
Excess Ultrasonic velocity U^E m/sec					
303	-3599.51	24882.8	-274883.8	832898.4	7.9011
308	-3365.88	23029.2	-263196.9	889103.2	7.6231
313	-3179.94	26803.2	-332720.9	1196557.4	6.1202
318	-3060.55	22366.8	-254834.1	938043.6	6.4454
Excess Viscosity η^E x 10^{-3} Ns/m^2					
303	-7.6046	-103.152	1016.77	-6507.35	0.0144
308	-69.677	-1871.62	27286.6	-133829.4	0.4609
313	-43.536	-582.991	7450.47	-43809.3	0.1481
318	-31.932	-246.593	2611.23	-18668.5	0.0788
V_m^E x 10^{-7} m^3/mole					
303	312.264	-24402.3	367117	-1545944	4.4013
308	452.631	-19086.8	261716	-1078210	3.0017
313	527.816	-19700.3	267295	-1080540	2.9890
318	519.962	-21681.5	293053	-1128860	4.0830
V_f^E x $10^{-7}$$m^3$/mole					
303	22.8699	-3487.99	49523.4	-205072	0.5777
308	39.5962	-3324.12	46252.6	-189604	0.5042
313	77.5122	-3780.41	50806.1	-200452	0.5889
318	96.0603	-3626.21	48680.6	-190893	0.5394
π_i^E x 10^{15}N/ m^2					
303	-126.466	-2001.88	23649.0	-129703.8	0.3749
308	-122.109	-30229.5	44740.7	-217327.8	0.7630
313	-93.1224	-1795.98	25768.6	-129299.2	0.4527
318	-76.0328	-1238.08	17445.1	-88712.8	0.3279
H^E x 10^{11} Joule/mole					
303	-66.4654	69.3146	-7940.10	19420.0	0.1493
308	-66.7816	-821.644	9561.68	-54507.7	0.1873
313	-52.1960	-320.838	2527.21	-21311.0	0.0868
318	-42.9999	-134.041	137.474	-8340.58	0.0559
ΔG^E x 10^{10} Joule/mole					
303	-47.6747	250.113	-6565.54	23497.4	0.0625
308	-48.0224	139.160	-4445.23	12420.2	0.0603
313	-44.5569	249.735	-5562.25	17162.0	0.0771
318	-40.3550	259.691	-5319.47	17337.0	0.0613

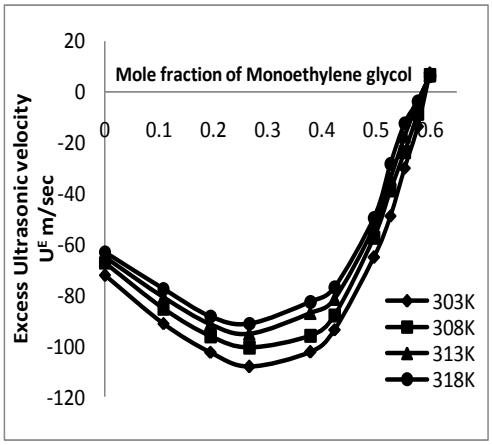

Figure 1.1. Variation of Excess Ultrasonic Velocity with Mole Fraction of Monoethylene Glycol

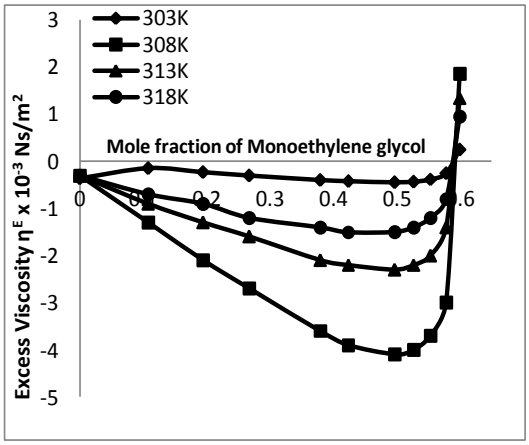

Figure 1.2. Variation of Excess Viscosity with Mole Fraction of Monoethylene Glycol

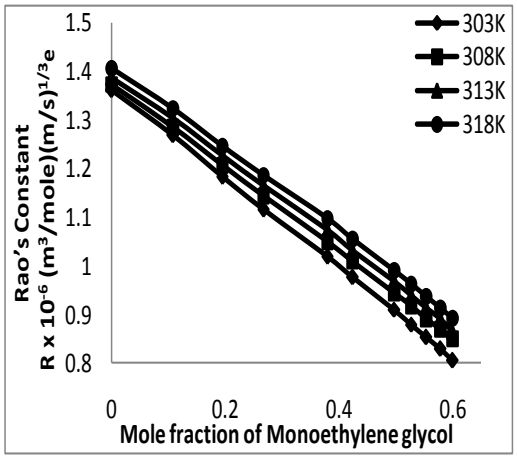

Figure 1.3. Variation of Rao's Constant with Mole Fraction of Monoethylene Glycol

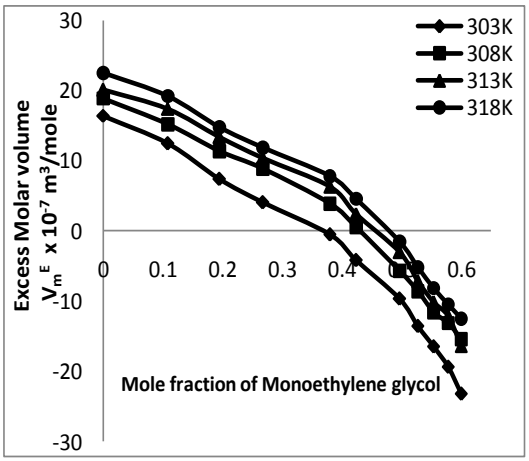

Figure 1.4. Variation of Excess Molar Volume with Mole Fraction of Monoethylene Glycol

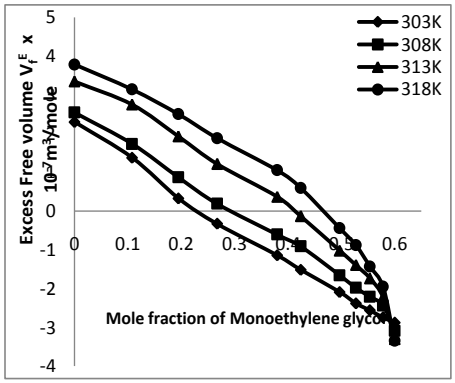

Figure 1.5. Variation of Excess Free Volume with Mole Fraction of Monoethylene Glycol

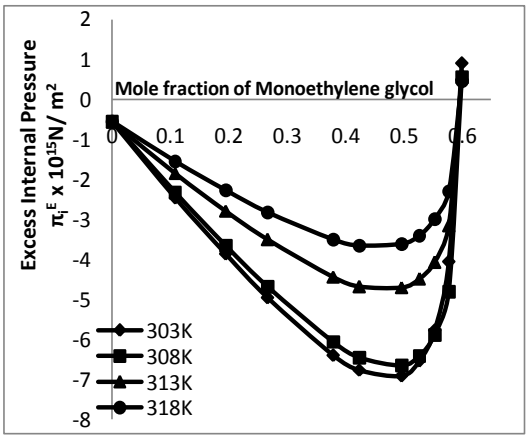

Figure 1.6. Variation of Excess Internal Pressure with Mole Fraction of Monoethylene Glycol

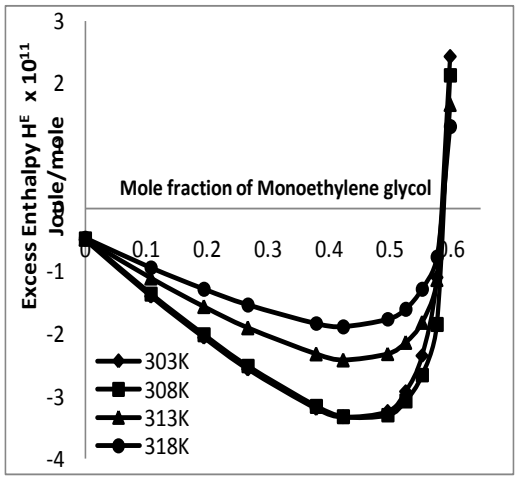

Figure 1.7. Variation of Excess Enthalpy with Mole Fraction of Monoethylene Glycol

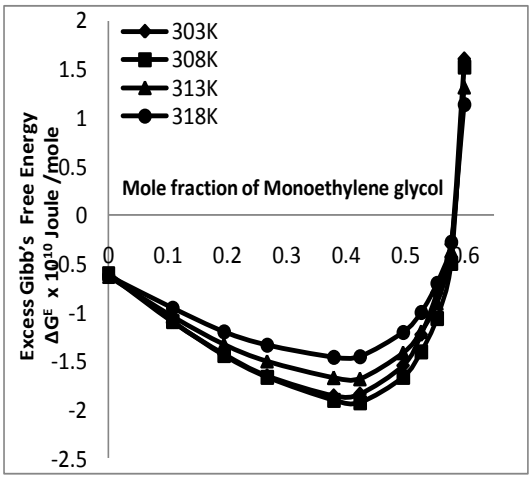

Figure 1.8. Variation of Excess Gibb's Free Energy with Mole Fraction of Monoethylene Glycol

Table 2.2. Ternary Liquid Mixture – II : MEG +CY + IBA Excess Values of Acoustic and Thermodynamic Parameters at Temperatures 303,308,313 and 318K

Mole fraction of MEG X_1	Mole fraction of IBA X_3	U^E m/sec	η^E x 10^{-3} Ns/m^2	V_m^Ex10^{-7} m^3/mole	V_f^Ex 10^{-8} m^3/mole	π_i^Ex10^{14} N/m^2	H^E x10^{11} Joule/mol	ΔG^E x 10^{10} Joule/mol
303K								
0.0000	0.6001	-48.593	-0.3541	18.690	3.4347	-0.6477	-0.5421	-0.6113
0.0968	0.5032	-86.938	-1.3658	13.789	2.3967	-2.3711	-1.3498	-1.0935
0.2480	0.3521	-119.71	-2.8772	5.2153	0.8798	-4.8587	-2.5127	-1.6973
0.3087	0.2913	-118.94	-3.4348	1.1653	0.2211	-5.7295	-2.9100	-1.8594
0.3609	0.2392	-106.13	-3.8623	-2.8232	-0.4382	-6.3683	-3.1850	-1.9089
0.4074	0.1926	-89.876	-4.1857	-5.8582	-1.0171	-6.8134	-3.3531	-1.8920
0.4797	0.1202	-59.452	-4.4539	-10.782	-1.9379	-7.0502	-3.3209	-1.6360
0.5179	0.0821	-41.362	-4.3148	-13.118	-2.3844	-6.7103	-2.9922	-1.282
0.5483	0.0517	-27.757	-3.8765	-16.168	-2.6032	-5.9643	-2.4260	-0.8416
0.5748	0.0253	-11.665	-2.7842	-19.554	-2.7261	-4.4584	-1.3567	-0.1606
0.6000	0.0000	7.7200	2.4856	-23.232	-2.8772	0.9136	2.4299	1.6065
308K								
0.0000	0.6001	-52.910	-0.3	21.157	4.4226	-0.6601	-0.5594	-0.6639
0.0968	0.5032	-90.485	-1.2	16.658	3.2206	-2.2740	-1.3430	-1.1450
0.2480	0.3521	-128.53	-2.6	8.7773	1.4604	-4.6019	-2.4712	-1.7582
0.3087	0.2913	-129.59	-3.1	5.9808	0.8866	-5.4377	-2.8741	-1.9414
0.3609	0.2392	-121.90	-3.5	3.2340	0.2295	-6.0581	-3.1576	-2.0147
0.4074	0.1926	-106.48	-3.8	0.1665	-0.2255	-6.5259	-3.3678	-2.0436
0.4797	0.1202	-79.048	-4.1	-5.3221	-1.1703	-6.8742	-3.4473	-1.8796
0.5179	0.0821	-62.478	-4.1	-8.8565	-1.7627	-6.6688	-3.2276	-1.5899
0.5483	0.0517	-41.790	-3.8	-11.447	-2.2435	-6.0475	-2.7337	-1.1443
0.5748	0.0253	-23.958	-3.2	-13.696	-2.4280	-5.0498	-2.0021	-0.6092
0.6000	0.0000	6.8800	1.86	-15.441	-3.0894	0.5657	2.1352	1.5260
313K								
0.0000	0.6001	-58.829	-0.3	22.895	5.0564	-0.6386	-0.5364	-0.6780
0.0968	0.5032	-95.889	-0.8	18.416	4.0675	-1.8026	-1.0862	-1.0828
0.2480	0.3521	-134.06	-1.6	11.296	2.5648	-3.4576	-1.8671	-1.5915
0.3087	0.2913	-139.61	-1.9	8.1706	1.9841	-4.0321	-2.1361	-1.7438
0.3609	0.2392	-131.64	-2.1	5.6661	1.4746	-4.4633	-2.3327	-1.8079
0.4074	0.1926	-119.60	-2.2	3.1022	0.8406	-4.7497	-2.4459	-1.8074
0.4797	0.1202	-93.755	-2.4	-1.8851	-0.0490	-4.9608	-2.4888	-1.6859
0.5179	0.0821	-74.093	-2.3	-4.1478	-0.9024	-4.7450	-2.2785	-1.4045
0.5483	0.0517	-57.551	-2.1	-6.8203	-1.6891	-4.2018	-1.8541	-0.9959

0.5748	0.0253	-36.232	-1.5	-9.5902	-2.2265	-3.2957	-1.1904	-0.4494
0.6000	0.0000	6.6000	1.34	-14.924	-3.3037	0.6161	1.6728	1.3195
318K								
0.0000	0.6001	-63.707	-0.3	24.729	5.2850	-0.6267	-0.5201	-0.6866
0.0968	0.5032	-99.917	-0.6	20.027	4.3272	-1.5444	-0.9417	-1.0342
0.2480	0.3521	-137.87	-1.1	13.554	2.9935	-2.8332	-1.5346	-1.4665
0.3087	0.2913	-145.62	-1.3	10.589	2.3878	-3.2634	-1.7275	-1.5899
0.3609	0.2392	-142.05	-1.5	8.1465	1.8893	-3.5783	-1.8647	-1.6388
0.4074	0.1926	-130.07	-1.6	5.4305	1.3953	-3.7864	-1.9469	-1.6388
0.4797	0.1202	-105.94	-1.6	1.3647	0.5000	-3.8968	-1.9443	-1.5109
0.5179	0.0821	-88.148	-1.5	-1.0974	-0.3666	-3.6901	-1.7579	-1.2608
0.5483	0.0517	-69.004	-1.3	-3.9201	-1.1642	-3.2554	-1.4259	-0.9139
0.5748	0.0253	-47.843	-0.9	-7.8782	-2.0262	-2.3954	-0.7998	-0.3589
0.6000	0.0000	6.5400	0.95	-12.520	-3.3469	0.4725	1.3130	1.1379

Table 1.3. Values of the Coefficients of Redlich - Kister Polynomial and Standard Deviation (σ) for Ternary Mixture - II

Temp , K	A_0	A_1	A_2	A_3	σ
Excess Ultrasonic velocity U^E m/sec					
303	-4459.68	18773.2	--95614.2	-124037.8	7.7175
308	-4222.06	22720.2	-167286.9	306955.1	5.4511
313	-4459.68	18773.2	-95614.2	-124037.8	7.7175
318	-4627.17	14485.9	-26755.3	-521828.2	10.104
Excess Viscosity η^E x 10^{-3} Ns/m^2					
303	-76.2102	-986.691	9871.98	-64190.1	0.1680
308	-68.0309	-1436.02	20315.3	-109808.0	0.3742
313	-43.6504	-465.430	4931.43	-33992.6	0.0867
318	-31.2847	-272.830	2255.65	-16727.2	0.0771
V_m^E x 10^{-7} m^3/mole					
303	450.625	-25198.1	331014	-1357711	3.6607
308	581.072	-21412.0	259632	-1028359	2.8546
313	662.521	-21044.1	242845	-897992.6	2.8208
318	737.518	-21700.6	252009	-903904.1	2.9101
V_f^E x $10^{-7}$$m^3$/mole					
303	78.4723	-3690.46	42909.3	-172698	0.4534
308	109.179	-4141.61	47716.1	-183860	0.5186
313	147.001	-4559.57	54453.1	-206358	0.5725
318	159.594	-4725.74	57748.0	-215470	0.5879
π_i^E x 10^{15}N/ m^2					
303	-130.409	-1711.36	19427.6	-115352	0.3696
308	-124.427	-2172.48	30468.7	-166056	0.6248
313	-95.9974	-1187.57	15333.8	-91071.7	0.3462
318	-80.5264	-685.852	8010.22	-54713.2	0.2089
H^E x 10^{11} Joule/mole					
303	-70.6459	-156.481	-3470.98	2883.83	0.0606
308	-70.0072	-587.030	5949.06	-41595.5	0.1771
313	-54.8128	-135.360	-707.311	-8605.36	0.0635
318	-46.4136	76.1483	-3460.36	5154.56	0.0489
ΔG^E x 10^{10} Joule/mole					
303	-52.6987	418.875	-9426.03	36577.2	0.1253
308	-54.9616	181.581	-4486.72	11940.2	0.0554
313	-51.0470	311.461	-6369.21	20052.9	0.0645
318	-48.0843	386.898	-7202.90	23406.8	0.0800

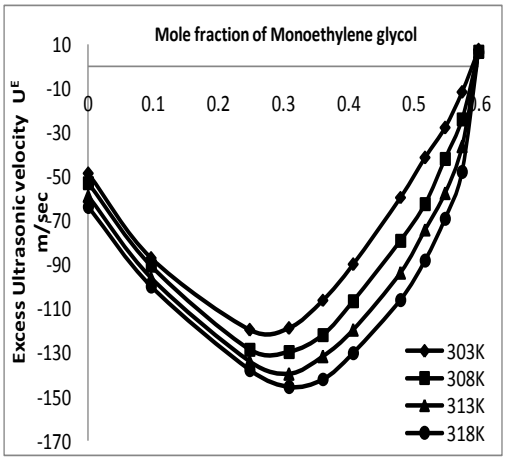

Figure 2.1. Variation of Excess Ultrasonic Velocity with Mole Fraction of Monoethylene Glycol

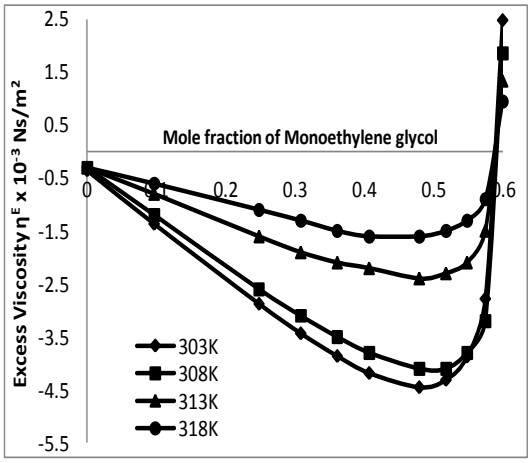

Figure 2.2. Variation of Excess Viscosity with Mole Fraction of Monoethylene Glycol

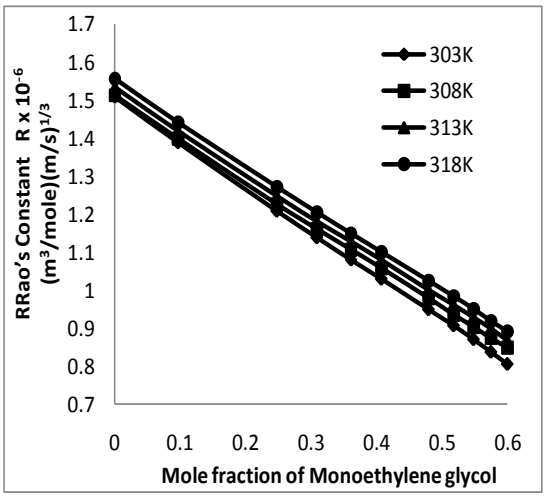

Figure 2.3. Variation of Rao's Constant with Mole Fraction of Monoethylene Glycol

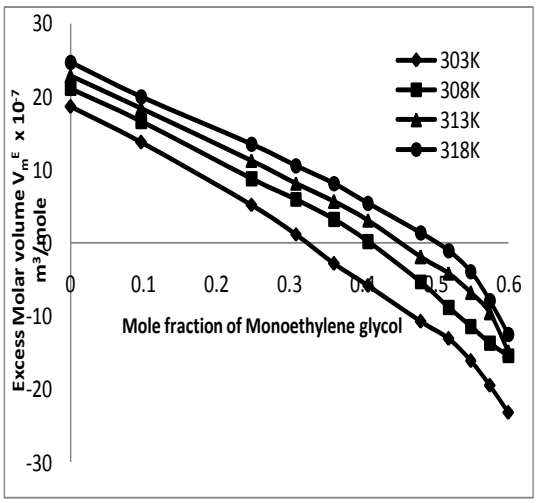

Figure 2.4. Variation of Excess Molar Volume with Mole Fraction of Monoethylene Glycol

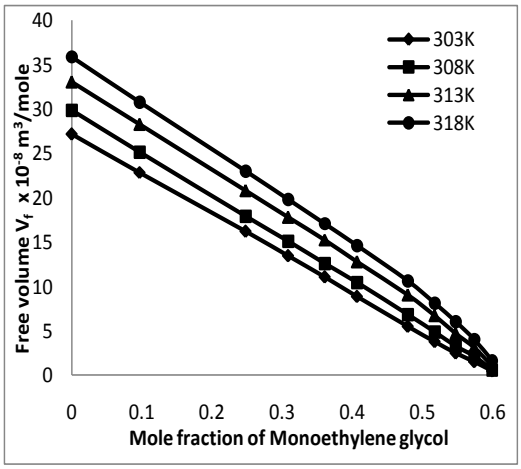

Figure 2.5. Variation of Excess Free Volume with Mole Fraction of Monoethylene Glycol

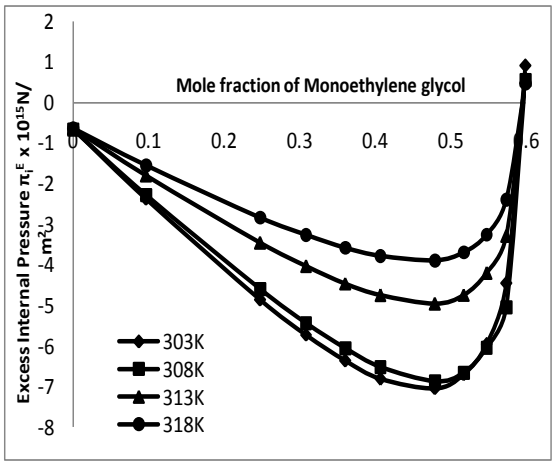

Figure 2.6. Variation of Excess Internal Pressure with Mole Fraction of Monoethylene Glycol

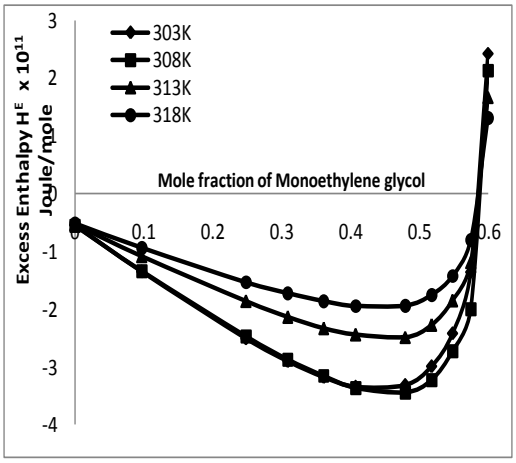

Figure 2.7. Variation of Excess Enthalpy with Mole Fraction of Monoethylene Glycol

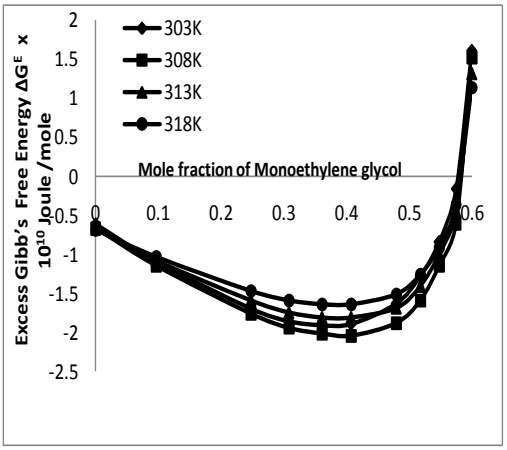

Figure 2.8. Variation of Excess Gibb's Free Energy with Mole Fraction of Monoethylene Glycol

The measurement of viscosity in ternary liquid mixture gives some reliable information in the study of intermolecular interaction. The molecules of one or more components forming the ternaries are either polar, associating and accordingly show non-ideal behaviors in mixtures. Negative values of η^E in most of the cases are the consequence of lower viscosity contributions of similar non-specific interactions and H-bonding effects of molecular species in real mixtures rather than those in the corresponding ideal mixtures.

In the present study, it is observed that, for the two ternary systems the η^E values gradually decrease up to the mole fraction around 0.5 and then begins to increase (can be seen form Figure 1.2 and Figure 2.2. More over it is observed that the η^E values decrease as the temperature increases. The negative values imply the presence of dispersion forces between the mixing components in the mixtures, and the positive values may be attributed to the presence of specific interactions.

The excess molar volume (V_m^E) is another important parameter through which molecular interactions can be explained. In the present investigation, for the ternary systems it is observed that, as the mole fraction of MEG increases, the V_m^E values continuously decreasing (*begins with positive, ends with negative values*) (Figure 1.4, Figure 2.4). So this kind of trend may be interpreted as, the interactions among the molecules of the component mixtures, are moving from weak to strong. As the temperature is increasing, the V_m^E values are decreasing in all the four ternary systems.

These effects may be arbitrarily divided into three types, namely chemical, physical and structural. A physical contribution, that is from non-specific interactions between the real species present in the mixture, contributes a positive trend to V_m^E. The chemical or specific intermolecular interactions result in a volume decrease, and these interactions include charge transfer type forces and other complex-forming interactions. This effect yields negative contributions to V_m^E. The structural contributions are mostly negative and arise from several effects, especially from interstitial accommodation changes the free volume. In other words, structural contributions arising from geometrical fitting of one component into the other because of differences in the free volume and molar volume between components lead to a overall negative contribution to V_m^E. Here in the present mixtures, it may be concluded that the chemical or specific intermolecular interactions dominate over the other types of interactions [23, 24].

In the ternary mixtures, the excess free volume (V_f^E) values decreases non-linearly beginning from positive to the negative as the mole fraction of MEG increases, which can be seen from the plots Figure 1.5, Figure 2.5. Adgaonkar *et. al.*, and others [26] showed positive values of V_f^E indicate the existence of weak molecular interactions in the liquid mixtures and the negative values of excess free volume suggest the specific interactions among unlike molecules. The negative values of excess free volume (V_f^E) in the systems

assert that the combined effect of the factors responsible for volume contraction and vice-versa. More over, the interactions include charge transfer type forces and other complex-forming interactions resulting specific interactions dominate over the other forces acting among the molecules.

A qualitative explanation of the behaviour of the ternary mixtures has been suggested as follows: The mixing of CY with glycols causes the dissociation of the hydrogen-bonded structure of glycols 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 CY and the proton of the OH group of glycol. 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. So here it is observed that the interactions are moving form weak to strong as the concentration of MEG increases.

It is an interesting fact to note that the variation of excess internal pressure, excess enthalpy and excess Gibb's free energy exhibit the same trend as that of the excess viscosity and behave exactly in a reverse trend as that of the free volume at all the four temperatures.

The values of π_i^E are almost negative and gradually decrease and move towards the positive values by the increase of mole fraction of MEG. More over the π_i^E decreases with increase in temperature. This situation is observed for all the four ternary systems under study and can be viewed from plots, Figure 1.6, Figure 2.6. This suggests that dipole and dispersive forces are operative in these systems, when the MEG concentration is low. When the concentration of MEG increased, the corresponding decrease in concentration of alkyl acetates leads to specific interactions; *i.e.*, the interactions move from weak to strong which supports the above arguments in case of other parameters.

The variation of excess enthalpy (H^E) with the mole fraction of MEG can be observed in the plots, Figure 1.7, Figure 2.7 for all the four mixtures. The H^E values decrease with increase in temperature is observed in the present study. The gradual increase from negative to positive values of H^E, indicates that interactions are moving from exothermic to endothermic effect. As the concentration of MEG increases the endothermic effect dominates the exothermic effect leads to strong interactions among the components of the mixture.

Next, the excess Gibb's free energy (ΔG^E) is negative at low concentration of MEG in the mixtures and positive in the MEG rich concentration which can be seen from the figures, Figure 1.8 and Figure 2.8. The increase of ΔG^E values with increase in temperature is also observed. In the negative region the trend is due to the rupture of hydrogen bond and dominance of dispersive forces. Where as the positive trend suggest the complex formation among the components of the mixture indicating strong interactions. So the present argument is complimented by the above discussion. The reduction of negative excess values with increase in concentration of MEG, in all the four ternary systems indicates the need for smaller time for the cooperative process; or the rearrangement of molecules in mixtures decreases energy that leads to dissociation. Recently Ali and Nain[26] have attributed the increasing positive values of Gibb's free energy in few ternary liquid mixtures to hydrogen bond formation between the unlike molecules, which supports the present investigation.

5. Conclusion

The above discussion reveal that the existence of molecular interactions in the ternary liquid mixtures. The present investigation shows that greater molecular interaction exists in the mixtures which may be due to hydrogen bonding formation. Also the weak molecular interaction that exists which may be due to the dominance of dispersion forces and dipolar interaction between the unlike molecules. In this manner all the parameters which have been evaluated form the measured values support one another to give a conclusion that the interactions becoming strong starting from weak interactions among the component molecules of the mixtures.

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