The Study of Physicochemical Properties of Binary Liquid Mixtures of Triethylamine with 2-Methoxyethanol, 2-Ethoxyethanol, 2-Butoxyethanol, 2-(2-Methoxyethoxy)ethanol, 2-(2-Ethoxyethoxy)ethanol, and 2-(2-Butoxyethoxy) ethanol at 308.15 K

S. Bahadur Alisha[1]*, S. Nafeesa Banu[1], K.S.V. Krishna Rao[2], M.C.S. Subha[3], K. Chowdoji Rao[4]

[1]Department of Chemistry, S.L.S. Degree College, Pullareddypeta, Andhra Pradesh, India, [2]Department of Chemistry, Yogi Vemana University, Vemanapuram, Kadapa, Andhra Pradesh, India, [3]Department of Chemistry, Sri Krishnadevaraya University, Ananthapuramu, Andhra Pradesh, India, [4]Departmet of Polymer Science & Technology, Sri Krishnadevaraya University, Ananthapuramu, Andhra Pradesh, India

ABSTRACT

Densities and viscosities of binary liquid mixtures of triethylamine with cellosolves [2-methoxyethanol (EEL), 2-EEL, 2-butoxyethanol] and carbitols [2-(2-methoxyethoxy)ethanol, 2-(2-ethoxyethoxy)ethanol, 2-(2-butoxyethoxy)ethanol] have been measured at 308.15K. From the experimental data, the excess volume (V^E), excess viscosity (η^E), and the excess molar Gibbs free energy of the activation of viscous flow (G^{*E}) have been computed and presented as function of composition. The observed variations of the properties for the above mixtures conclude that the interactions between unlike molecules predominate over the dissociation effects in the individual components. It is also evident that the presence of strong interactions between unlike molecules is predominant and characterized by the negative V^E and positive η^E and G^{*E} values.

Key words: Binary liquid mixtures, Trietylamine, Cellosolves and carbitols, Viscosity, Density, excess thermodynamic parameters, Molecular interactions.

1. INTRODUCTION

The formation of hydrogen bond in solutions and its effect on the physical properties of the mixtures have received much attention. Hydrogen bonding plays an important role in fundamental sciences and in industrial applications. Although many experimental and theoretical studies have been directed toward understanding of hydrogen bonding, it remains an area of active research. Knowledge of physicochemical properties of liquid mixtures formed by two or more components associated through hydrogen bonds is important from theoretical and process design aspects. Properties such as densities, viscosities, and the excess properties and their variation with composition of the binary mixtures are useful to design engineering processes and in chemical and biological industries. The investigation on the possible changes in these properties of mixtures has been found to be an excellent qualitative and quantitative way to elicit the information about molecular structure and intermolecular forces present in the liquid mixtures. In continuation of on our ongoing program of studies

on binary liquid mixtures [1-3], we present here some new experimental data on the density (ρ) and viscosity (η) for the binary liquid mixture systems of triethylamine (TEA) with different cellosolves and carbitols at 308.15K.

TEA is a weakly polar [4] liquid with a low dielectric constant (ε = 2.42 at 298.15 K). Whereas cellosolves and carbitols are polar and associative in nature. TEA shows more molecular interactions with other molecules due to the presence of lone pair of electrons on N-atom. From the theoretical point of view, cellosolves and carbitols are drawn a special interest in the recent years.[1-3,5,6] The study of molecular interactions of cellosolves and carbitols is of interest because of investigating the effect of simultaneous presence of etheric and hydroxyl oxygen atoms in the same molecule. At the same time, it is also important to note that the presence of etheric oxygen enhances the ability of the -OH group to form hydrogen bonds with other organic solvents [7-10]. The binary liquid mixtures of TEA with organic liquids possessing

*Corresponding Author:
E-mail: bahaduralisha@gmail.com

hydroxyl group may be useful to study the molecular interactions. These interactions also form a basis to study the properties of these binary solvent systems, systematically. Thus, the different types of interactions between TEA with cellosolves/carbitols are discussed from the present viscosity studies. In view of the above, the present research aims to measure densities and viscosities of binary mixtures of TEA with cellosolves (2-methoxtethanol [MEL], 2-ethoxyethanol [EEL], 2-butoxyethanol [BEL]) and carbitols [methylcarbitol (MC), ethylcarbitol (EC), and butylcarbitol (BC)] at 308.15 K and using the data the excess volume (V^E), excess viscosity (η^E), and the excess molar gibbs free energy of the activation of viscous flow ($G^{*.E}$) were calculated. The results were discussed in terms of intermolecular interactions between unlike molecules.

2. EXPERIMENTAL

2.1. Materials
TEA, 2-EEL (MC), 2-EEL (EC), 2-BEL (BC), MC, ethyl carbitol (EC), and butylcarbitol (BC) were purchased from E-Merck, Mumbai and used as recieved.

2.2. Methods
Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of closed system using electonic balance with the precision of ±0.01 mg. Mixtures were allowed to stand for some time before every measurement so as to avoid air bubbles. The purities of the liquids were checked by comparing the values of densities with literature data (Table 1) [8,11-15]. The measurements were made with proper care in an AC room to avoid evaporation loss.

The densities (ρ) of liquids and their mixtures were measured using bicapillary pycnometer having a capillary diameter of 0.85 mm, which was calibrated using double distilled water. The necessary buoyancy corrections were applied. The density values were reproducible within ±0.2 Kg m^{-3}. Viscosity

measurements were carried out with a precision of ±0.2% using calibrated Schott-Gerate AVS 400 viscometer, whose flow time for doubly distilled water was found to be 375 s at 298 K. The temperature of the test liquids during the measurements was maintained within an uncertainty of ±0.01 K in an electronically controlled thermostatic water bath. The measurements were made with proper care in an AC room to avoid evaporation loss.

3. RESULTS AND DISCUSSION
The experimental results of measurements of densities and viscosities of binary mixtures are presented in Table 2. The molar volume (V), excess volume (V^E), excess viscosity (η^E), excess gibbs energy of activation of viscous flow (G^{*E}), and Grunberg-Nissan interaction parameter (d^1) were calculated from the measured data using the following equations 1-5, respectively.

$$V = (X_1M_1+X_2M_2)/\rho \tag{1}$$

$$V^E = V-(X_1V_1+X_2V_2) \tag{2}$$

$$\eta^E = \eta-(X_1\eta_1+X_2\eta_2) \tag{3}$$

$$G^{*E} = RT[\ln \eta V-(X_1\ln \eta_1 V_1+X_2 \ln \eta_2 V_2)] \tag{4}$$

$$d^1 = \ln \eta/[X_1\ln \eta_1+X_2\ln \eta_2+X_1X_2] \tag{5}$$

Where ρ, η and V are the density, viscosity and molar volume of the mixture, M_1 and M_2 are the molar masses, η_1 and η_2 are the viscosities, V_1 and V_2 are the molar volumes of the TEA with cellosolves and TEA with carbitols, respectively.

3.1. TEA with Cellosolves
The values of the excess molar volumes of the mixtures at 308.15 K are presented in Table 2. The data are represented graphically in Figures 1-3. An examination of V^E data in Table 2 shows that the values of V^E are negative for the mixtures of TEA with cellosolves over the entire composition range

Table 1: Comparison of experimental density(ρ) of pure liquids with literature values at 308.15K.

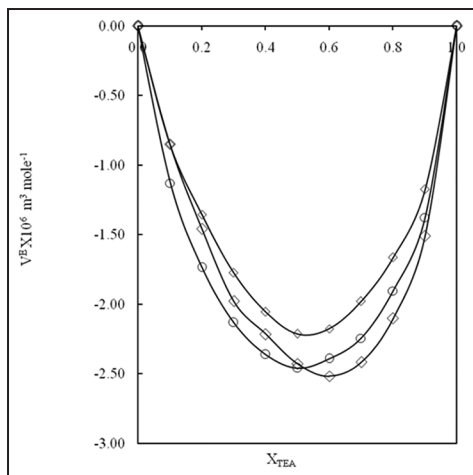| Liquid	Density ($\rho\times10^{-3}$) (Kg m^{-3})		Reference
	Experimetal	Litarature	
Triethylamine	0.7144	0.7144	[11]
2-methoxyethanol	0.9518	0.9529	[8,13]
		0.9511	
2-ethoxyethanol	0.9167	0.9163	[13]
2-butoxyethanol	0.8877	0.8887	[8]
2-(2-methoxyethoxy) etanol (Methylcarbitol)	1.0073	1.0065	[14]
2-(2-ethoxyethoxy) etanol (Ethylcarbitol)	0.9838	0.9839	[15]
2-(2-butoxyethoxy) etanol (Butylcarbitol)	0.9480	0.9479	[15]

Figure 1: Plots of excess volumes (VE) vs molefraction of triethylaminr (TEA) at 308.15K for the binary mixtures of oTEA with MEL (Δ), EEL (O) and BEL (◊).

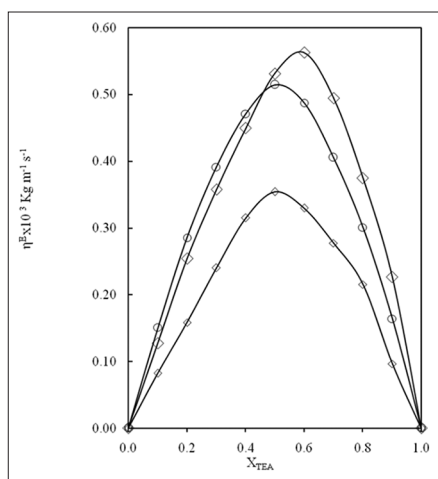**Figure 2:** Plots of excess viscosity (η^E) versus molefraction of triethylamine (TEA) at 308.15K for the binary mixtures of oTEA with MEL (Δ), EEL (O) and BEL (◊).
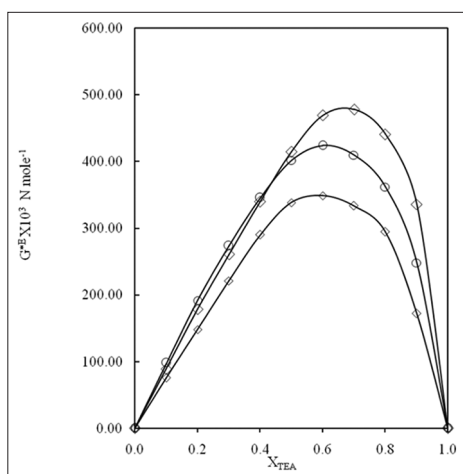

Figure 3: Plots of excess gibbs free energy of activation ($G*^E$) versus molefraction of triethylaminr (TEA) at 308.15K for the binary mixtures of oTEA with MEL (Δ), EEL (O) and BEL (◊).

at 308.15 K. In general, the excess volume data of liquid mixtures may be explained in terms of various factors [16-18].

i. Chemical or specific interaction, which includes charge-transfer complexes, formation of hydrogen bonding, dipole-dipole interaction, and other complex forming between dissimilar molecules.

ii. Loss of dipolar association and differences in size and shape.

iii. Physical interaction consisting dispersion forces or weak dipole-dipole interactions.

The first effect leads to contraction in volume making negative V^E value in liquid mixtures, whereas the remaining effects lead to expansion in volume resulting in positive V^E values. The actual volumes of V^E depend on the resultant of these opposing contributions [19-21]. It is observed from Figure 2 that the negative excess molar volumes were found to be large and approximately symmetrical with a minimum value around 0.6 mole fraction of TEA. It is also observed that the negative values of V^E fall in the sequence.

TEA+MEL<TEA+EEL<TEA+BEL

From the above sequence, it is concluded that the that the former effect is dominant in this investigation for binary mixtures of TEA with cellosolves. Hydrogen bonding ability of the -OH group of cellosolves increase with increase in the chain length of the alkoxy group of cellosolves, which in turn results in more negative V^E values for these mixtures. This further indicates a complex formation between the component molecules [22-24].

Figure 2 shows that the η^E values are positive over the whole composition range for all the systems under study. A corelation between the signs of η^E has been observed for a number of binary solvent systems [25]. η^E being positive where V^E is negative or vice versa. In general for the systems, where dispersion and dipolar interactions are operating, η^E values are found to be negative whereas charge transfer and hydrogen bonding interactions lead to the formation of complex species between unlike molecules resulting in positive values of η^E. The actual values depend on the dominance of these factors. The experimental values show that the latter effect is predominant over the former effect to make η^E values positive [26] for all the systems under study. The algebraic values of η^E for the mixtures of TEA + cellosolves fall in the order:

TEA+MEL<TEA+EEL<TEA+BEL

This order suggests that the hydrogen bonding between unlike molecules increase with increase in the chain length of the alkoxy group of cellosolves

196

Table 2: Values of density (ρ), viscosity (η), excess viscosity(η^E), molar volume (V), excess molar volumes (V^E), exces gibbs free energy of actiation of viscousflow (G*^{E}), and Grunberg-Nissan interaction parameter (d^1) for the binary liquid mixtures of triethylamine + cellosolves and triethylamine + carbitols at 308.15 K.

Mole fraction of TEA (X$_{TEA}$)	ρ×10^{-3}	η×10^3	η^E×10^3	V×10^5	V^E×10^6	G*^{E}	d^1
	(Kg m$_{-3}$)	(Kg m^{-1}s^{-1})	(Kg m^{-1}s^{-1})	(m^3 mole^{-1})	(m^3 mole^{-1})	(N mole^{-1})	
TEA+2-methoxyethanol (MEL)							
0.0000	0.9518	1.2373	0.0000	79.95	0.0000	0.00	-
0.1001	0.9218	1.2331	0.0821	85.28	−0.8485	75.48	1.2878
0.2004	0.8919	1.2230	0.1584	90.96	−1.3555	148.08	1.4193
0.3001	0.8649	1.2186	0.2400	96.69	−1.7741	221.21	1.6319
0.4001	0.8397	1.2076	0.3151	102.58	−2.0533	289.89	1.8873
0.4998	0.8164	1.1607	0.3542	108.57	−2.2120	338.31	2.1292
0.5999	0.7941	1.0497	0.3292	114.79	−2.1754	348.88	2.2963
0.6998	0.7731	0.9113	0.2772	121.15	−1.9781	333.30	2.5177
0.8003	0.7534	0.7627	0.2152	127.66	−1.6633	294.57	2.9457
0.9005	0.7647	0.5576	0.0964	134.33	−1.1735	172.03	3.0918
1.0000	0.7144	0.3754	0.0000	141.64	0.0000	0.00	-
TEA+2-ethoxyethanol (EEL)							
0.0000	0.9167	1.4563	0.0000	98.31	0.0000	0.00	-
0.1006	0.8985	1.4987	0.1511	101.54	−1.1289	98.41	1.8248
2.002	0.8773	1.5253	0.2854	105.25	−1.7342	191.59	1.9845
0.3002	0.8558	1.5228	0.3910	109.19	−2.1299	273.74	2.1501
0.4001	0.8346	1.4946	0.4708	113.29	−2.3604	345.46	2.3684
0.5003	0.8139	1.4297	0.5142	117.53	−2.4584	401.58	2.6397
0.5998	0.7937	1.2952	0.4872	121.91	−2.3912	423.83	2.8996
0.6999	0.7743	1.1050	0.4052	126.40	−2.2434	409.42	3.2037
0.8001	0.7551	0.8924	0.3009	131.08	−1.9027	361.61	3.7203
0.9004	0.7362	0.6471	0.1640	135.95	−1.3758	247.95	4.5669
1.0000	0.7144	0.3754	0.0000	141.64	0.0000	0.00	-
TEA+2-butoxyethanol (BEL)							
0.0000	0.8877	2.1929	0.0000	133.13	0.0000	0.00	-
0.1001	0.8749	2.1380	0.1270	133.13	−0.8483	88.90	1.6802
0.2005	0.8605	2.0825	0.2540	133.38	−1.4574	178.64	1.8857
0.3003	0.8457	2.0042	0.3571	133.71	−1.9777	260.77	2.0946
0.4002	0.8292	1.9147	0.4492	134.32	−2.2144	339.79	2.3779
0.5001	0.8127	1.8151	0.5311	134.96	−2.4262	414.18	2.7748
0.6002	0.7956	1.6644	0.5624	135.72	−2.5152	468.95	3.2661
0.7004	0.7776	1.4142	0.4943	136.68	−2.4157	477.98	3.8014
0.7998	0.7588	1.1139	0.3746	137.84	−2.1012	440.64	4.5866
0.9006	0.7386	0.7822	0.2261	139.29	−1.5082	335.68	6.2420
1.0000	0.7144	0.3754	0.0000	141.64	0.0000	0.00	-
TEA+2-(2-methoxyethoxy) ethanol (MC)							
0.0000	1.0073	2.5471	0.0000	119.98	0.0000	0.00	-
0.1006	0.9792	2.4608	0.1322	120.75	−0.7748	93.80	1.7482
0.1997	0.9504	2.3624	0.2490	122.44	−1.3088	183.07	1.9218

(Contd...)

Table 2: (*Continued*).

Mole fraction of TEA (X_{TEA})	$\rho \times 10^{-3}$	$\eta \times 10^3$	$\eta^E \times 10^3$	$V \times 10^5$	$V^E \times 10^6$	G^{*E}	d^1
	(Kg m$_{-3}$)	(Kg m^{-1}s^{-1})	(Kg m^{-1}s^{-1})	(m^3 mole^{-1})	(m^3 mole^{-1})	(N mole^{-1})	
0.3001	0.9210	2.2347	0.3393	124.28	−1.7126	265.35	2.1131
0.4003	0.8915	2.0793	0.4015	126.26	−1.9721	337.85	2.3479
0.5002	0.8627	1.8933	0.4325	128.28	−2.1869	396.81	2.6449
0.5998	0.8340	1.6639	0.4194	130.43	−2.2642	434.21	3.0111
0.7004	0.8047	1.3858	0.3598	132.81	−2.1353	440.65	3.4909
0.8001	0.7760	1.0753	0.2658	135.28	−1.8891	403.02	4.1874
0.9003	0.7462	0.7338	0.1419	138.14	−1.2733	288.74	5.3413
1.0000	0.7144	0.3754	0.0000	141.64	0.0000	0.00	-
TEA+2-(2-ethoxyethoxy) ethanol (EC)							
0.0000	0.9748	2.7662	0.0000	137.65	0.0000	0.00	-
0.1003	0.9545	2.6474	0.2114	137.11	−0.9398	91.74	1.7654
0.1999	0.9314	2.5458	0.2575	136.98	−1.4650	187.20	1.9775
0.3001	0.9071	2.4001	0.3514	137.01	−1.8398	272.03	2.1781
0.4002	0.8822	2.2293	0.4199	137.13	−2.1159	348.06	2.4313
0.5001	0.8573	2.0197	0.4491	137.27	−2.3763	408.66	2.7377
0.6002	0.8313	1.7648	0.4336	137.59	−2.4553	448.15	3.1232
0.7001	0.8047	1.4721	0.3797	138.04	−2.4017	459.54	3.6560
0.8009	0.7769	1.1411	0.2897	138.70	−2.1451	428.01	4.4791
0.9008	0.7475	0.7686	0.1560	139.75	−1.4978	311.03	5.8029
1.0000	0.7144	0.3754	0.0000	141.64	0.0000	0.00	-
TEA+2-(2-butoxyethoxy) ethanol (BC)							
0.0000	0.9397	3.4815	0.0000	172.64	0.0000	0.00	-
0.1005	0.9262	3.2804	0.1111	168.53	−0.9918	98.09	1.8182
0.2009	0.9095	3.1127	0.2552	164.89	−1.5234	201.69	2.0901
0.3004	0.8917	2.9310	0.3826	161.37	−1.9588	299.42	2.3650
0.3998	0.8725	2.7199	0.4802	157.97	−2.2807	388.23	2.6825
0.5002	0.8521	2.4626	0.5348	154.56	−2.5788	463.12	3.0718
0.6004	0.8299	2.1473	0.5307	151.32	−2.7083	515.08	3.5601
0.6999	0.8057	1.7827	0.4752	148.33	−2.6172	536.66	4.2357
0.7993	0.7796	1.3706	0.3718	145.51	−2.3530	511.55	5.2871
0.9005	0.7501	0.8981	0.2136	143.00	−1.7286	392.29	7.2634
1.0000	0.7144	0.3754	0.0000	141.64	0.0000	0.00	-

MEL: 2-methoxtethanol, EEL: 2-ethoxyethanol, BEL: 2-butoxyethanol, MC: Methylcarbitol, EC: Ethylcarbitol, BC: Butylcarbitol, TEA: Triethylamine
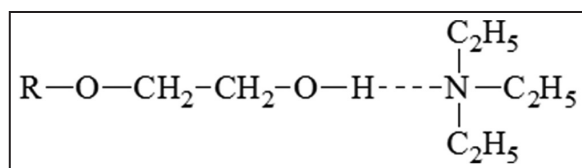
Scheme 1: Schematic representation of interaction between TEA and cellosolves.

(Scheme 1). A similar observation was reported by Krishnaiah *et al*. [8] from their viscosity studies of cellosolves with water.

The variation of excess gibbs free energy of activation of viscous flow G^{*E} with X_{TEA} is shown graphically in Figure 3. The G^{*E} values for these systems under study are positive which is an indication of the presence of strong specific interactions and are in the following order:

TEA+MEL<TEA+EEL<TEA+BEL

A similar observation was made by Pal and Singh [27,28] from their viscosity studies. Grunberg-

Nissan interaction parameter (d^1) gives an idea about the quantitative strength of interactions between unlike molecules. On the basis of the positive d^1 values (Table 2), we can conclude that the interactions are stronger in TEA + cellosolve mixtures. The results suggest that A-B type of interaction forms between the component molecules.

Where R may the $-CH_3$ or $-C_2H_5$ or $-C_4H_9$. In conclusion, it may be said that the observed variation in excess functions of these mixtures studied support the view that the predominance of interactions between unlike molecules over the dissociation effect in the individual components.

The strength of bonding is expected to increase with the increase in chain length of alkoxy group of cellosolves, and the results of this study corroborate this fact.

3.2. TEA with Carbitols

The measured density (ρ) and viscosity (η) data at 308.15 K for the mixtures of TEA+MC, + EC and +BC are used to calculate the excess molar volume (V^E), excess viscosity (η^E), excess gibbs free energy of activation of viscous flow (G^{*E}) and Grunberg-Nissan interaction parameter (d^1) and the results are presented in Table 2. Excess molar volume versus X_{TEA} plots Figure 4 show that V^E is negative over the entire range of composition which suggests the dominating nature of hydrogen bonding between unlike molecules (Scheme 1). Cobos *et al.* [29] reported negative excess molar volumes for the binary system of 2-BEL + 1-butanol at 298.15 K. The negative values of V^E for all the systems under study fall in the order.
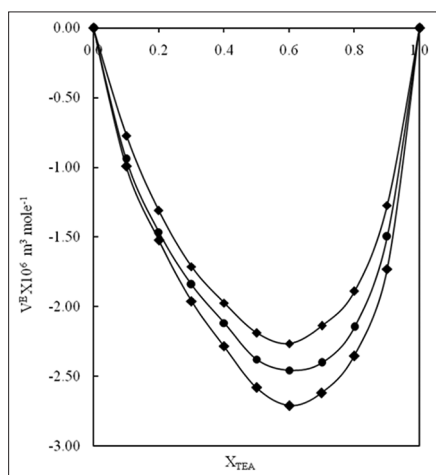
TEA+MC<TEA+EC<TEA+BC

This order suggests that the interactions due to hydrogen bonding increase with the increase in chain length of the alkoxy group of carbitols. This conclusion receives support from the work of Cobos *et al.* [29] and Aminabhavi *et al.* [14].

Figure 5 shows the variation of η^E drawn as a function of X_{TEA} and observed positive values over the whole composition range. The emergence of new structural species in solution formed out of intermolecular hydrogen bonding (Scheme 1) causes the solution to possess less flow capability and hence viscosity of the mixture is increased. This is evident from the curves (Figure 5) that a maxima in η^E is noticed at 0.6 mole fraction of TEA in all the three mixtures under study.

The algebraic values of η^E for the mixtures TEA + carbitols fall in the order

TEA+BC>TEA+EC>TEA+MC

This order suggests that η^E increases and becomes more positive with increase in the chain length of the alkoxy group of carbitols.

The excess gibbs free energy of activation of viscous flow (G^{*E}) is a measure of free flow of the mixture. The variation of G^{*E} with X_{TEA} is shown graphically in Figure 6. From this figure, it is observed that all the values of G^{*E} are found to be positive which further indicates the presence of strong intermolecular interactions between the unlike molecules. The G^{*E} values for the systems under study suggest the following order:

TEA+BC>TEA+EC>TEA+MC

The above order is in accordance with the viscosity results of these systems.

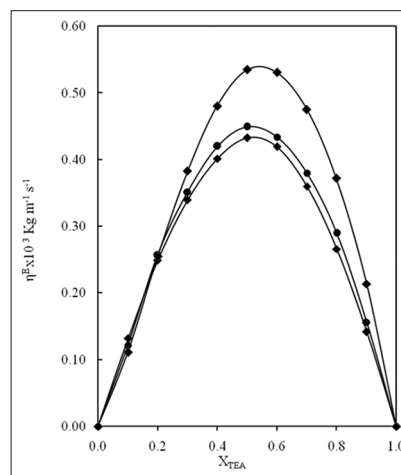**Figure 4:** Plots of excess volumes (V^E) versus molefraction of triethylaminr (TEA) at 308.15K for the binary mixtures of TEA with MC ($-\blacktriangle-$), EC ($-\bullet-$), and BC ($-\blacklozenge-$).
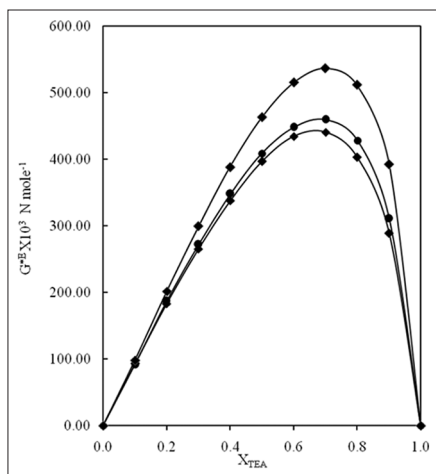
Figure 5: Plots of excess viscosity (η^E) versus molefraction of triethylaminr (TEA) at 308.15K for the binary mixtures of oTEA with MC ($-\blacktriangle-$), EC ($-\bullet-$), and BC ($-\blacklozenge-$).

Figure 6: Plots of excess gibbs free energy of activation (G^{*E}) versus molefraction of triethylaminr (TEA) at 308.15K for the binary mixtures of TEA with MC (−▲−), EC (−●−), and BC (−♦−).

According to Fort and Moore [30], systems exhibit strong interaction if the parameter d^1 is positive if it is negative they show weak interactions. On this basis, the positive d^1 values in this study substantiate the presence of strong interactions present in these liquid mixtures.

In conclusion, it may be said that the observed variation of all the parameters (V^E, η^E, G^{*E} and d^1) studied as the function of X_{TEA}, suggest that a new phase in solutions is emerging which can be attributed to the hydrogen bonded intermolecular complex formation between unlike molecules. The strength of bonding is expected to increase as the chain length of the alkoxy group of carbitol increases, and the results of this study corroborate this fact.

4. CONCLUSION

The densities and viscosities for TEA with cellosolves and carbitols of binary mixtures have been measured, and the values of V^E, η^E, G^{*E} and d^1 were calculated. The V^E values were found negative for all the mixtures whereas η^E, G^{*E} and d^1 values were found positive for the presence of specific interactions between TEA with cellosolves and carbitols. The magnitude of the interactions is shown in the following order.

Tiethylamine + cellosolves

TEA+MEL<TEA+EEL<TEA+BEL

Tiethylamine + carbitols

TEA+BC>TEA+EC>TEA+MC

In conclusion, it may be said that the observed variation of all the parameters (V^E, η^E, G^{*E} and d^1) of the mixtures studied supports the view that the interactions between unlike molecules predominate over dissociation effects in individual components. It is evident that the presence of strong interactions between unlike molecules is predominant and characterized by the negative V^E and positive η^E, G^{*E} and d^1 values. It also concluded for both the systems TEA + cellosolves and TEA + carbitols the strength of bonding is expected to increase as the chain length of the alkoxy group of cellosolves and carbitol increases, and the results of this study conforms this fact.

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Bibliographical Sketch

Dr. S. Bahadur Alisha did his B.Sc., from Andhra University, Vizag, India. M.Sc. Chemistry from Agra University, Agra, India. He did his M.Phil. and Ph.D. from Sri Krishnadevaraya University, India. He completed his Ph.D through Faculty Improvement Programme (FIP) of University Grants Commission, India. His major field of research is thermodynamic properties of binary liquid mixtures. He joined as lecturer at Department of Chemistry, S.L.S. Degree College, Pullareddypeta, 516175, India in 1984 and promoted as Reader in Chemistry in the year 2001. He is currently working as Associate Professor in the Department of Chemistry, S.L.S. Degree College, Pullareddypeta-516175, Andhra Pradesh, India. He is Vice-Principal of the college since 2008 onwards.