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The study of thermo physical properties of binary liquid mixtures of 2-(2-butoxy ethoxy) ethanol (butyl carbitol) with n-butyl amine, sec.-butyl amine, tert-butyl amine, n-hexyl amine, n-octyl amine and cyclo hexyl amine at 308.15 K

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ABSTRACT

Densities and viscosities of binary liquid mixtures of 2 - (2-Butoxy ethoxy) ethanol (Butyl Carbitol) (BC) with n-butyl amine (NBA), sec-butyl amine (SBA). ter-butylamine (TBA), n-hexylamine (NHA), n-octylamine (NOA) and cyclohexylamine (CHA) have been measured at 308.15 K. 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 interaction parameter d^1 of the Grunberg and Nissan has been calculated. 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), (G^{*E}) and d^1 values. The excess volume, excess Viscosity and excess molar Gibb's free energy of the activation of viscous flow have been fitted to Redlich – Kister equation to derive the coefficients and standard deviations.

Key words: Binary liquid mixtures, Butyl carbitol, Amines, excess volumes, Redlich – Kister equation, molecular interactions.

INTRODUCTION

The Study of excess thermodynamic functions is useful tools in understanding the nature of molecular interactions in binary liquid mixtures [1]. In chemical industry, knowledge of the properties of binary liquid mixtures is essential in designs involving chemical separations, heat transfer, mass transfer, and fluid flow [2–7].The mixing of two solvents has been reported to result in specific interactions such as hydrogen bonding, dipole–dipole, and charge transfer reactions[8]. Strong and weak interactions between unlike molecules in mixtures lead to deviations from ideality. Deviations from ideality in binary liquid mixtures are usually associated with synergism and are attributed to differences in the chemistry and molecular architecture of the mixing solvents [9–14] as well as the experimental conditions such as temperature and the mixing ratios of the binary liquids.

In chemical industry there exists a continuing need for reliable thermodynamic data of binary systems. A survey of the literature shows that very few attempts [15-17] have been made to study excess properties for mixtures containing alkoxy alcohols (carbitols). Amines and alkoxy alcohols [18-19] in their pure state exhibit self-association through hydrogen bonding. Amines are better electron donors, which allow them to have specific interactions. Further, amines form water insoluble compounds of medicinal importance. The molecular interaction study of alkoxy alcohols (carbitols) is of interest because of investigating the effect of simultaneous presence of ether and alcoholic functional groups in the same molecule. The presence of ethereal oxygen enhances the ability of the -OH group of the same molecule to form hydrogen bonds with other organic molecules [20, 21]. These characteristics of amines and alkoxy alcohols (carbitols) make them interesting for our study. In continuation of our physico-chemical property studies of binary liquid mixtures [22-25] and survey of the literature it is noticed that no attempts have been made to study the excess volumes and viscosity of binary liquid mixtures of 2-(2-Butoxy ethoxy) ethanol (Butyl carbitol) with a series of amines which include n-butyl amine (NBA), sec-butyl amine (SBA), ter-butylamine(TBA), n-hexylamine (NHA), N-octylamine (NOA), and cyclohexylamine (CHA) at 308.15 K. These quantities have been fitted to the Redlich – Kister equation to obtain the binary coefficients a_i and standard errors 'σ' between the calculated and the experimental parameters. Aiming for a wider understanding of the intermolecular interactions involved in these binary liquid mixtures and the results are presented here.

MATERIALS AND METHODS

2-(2-Butoxy ethoxy) ethanol (Butyl carbitol) (BC), n-butyl amine (NBA), sec-butyl amine (SBA), ter-butylamine (TBA), n-hexylamine (NHA). N-Octylamine (NOA), and cyclohexylamine (CHA) were purchased from Merck, Mumbai, India and used as purchased. Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of closed system. The weighing were done by using Mettler balance with the precision of ± 0.1 mg. The uncertainty in the mole fraction was estimated to be less than ± $1x10^{-4}$. Mixtures were allowed to stand for some time before every measurement so as to avoid air bubbles.

The densities of pure liquids and their binary mixtures were measured by using a single-capillary pycnometer (made of Borosil glass) having a bulb capacity of ≈ 10 mL. The capillary, with graduated marks, had a uniform bore and could be closed by a well-fitting glass cap. The marks on the capillary were calibrated by using triple distilled water. The uncertainty in density measurements was within ± 2×10^{-4} kg m^{-3}.

Viscosity measurements were carried out with a precision of ± 0.2% using calibrated Schott-Gerate AVS 400 viscometer (U.S.A), whose flow time for double distilled water was found to be 375 seconds at 25^0C. 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. The purities of the liquids were checked by comparing the values of densities and viscosities with literature data (Table.1) and found in good agreement in general.

Table: 1: Comparison of experimental density and viscosity of pure liquids with literature values at 308.15 K

Liquid	ρ x 10^{-3} Kg m^{-3}		η x 10^{3} Kg m^{-1} s^{-1}	
	Expt.	Lit.	Expt.	Lit.
2-(2-Butoxy ethoxy) ethanol	0.9399	0.9456[a]	0.3481	0.4323[a]
n-Butyl amine	0.7241	0.7239[b]	0.4248	0.4249[b]
Sec-butyl amine	0.7084	0.7083[b]	0.3994	0.3996[b]
Tert- butyl amine	0.6809	0.6812[b]	0.4122	0.4126[b]
n-Hexyl amine	0.7522	0.7522[b]	0.6001	0.6000[b]
n-Octyl amine	0.7704	0.7702[b]	0.9263	0.9267[b]
Cyclo hexyl amine	0.8525	0.8527[b]	1.3245	1.3249[b]

a Reference [40]
b Reference [41]

RESULTS AND DISCUSSION

The experimental results of the measurement of densities (ρ) and Viscosities (η) of binary liquid mixtures are presented in Table. 2. The molar volume (V), excess 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) were calculated from the measured data using the following equations 1 to 5 respectively.

$$V = (X_1M_1 + X_2M_2) / \rho \quad \ldots \quad (1)$$

$$V^E = V - (X_1 V_1 + X_2 V_2) \quad \ldots \quad (2)$$

$$\eta^E = \eta - (X_1 \eta_1 + X_2 \eta_2) \quad \ldots \quad (3)$$

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

$$d^1 = \ln \eta / [X_1 \ln \eta_1 + X_2 \ln \eta_2 + X_1 X_2] \quad \ldots \quad (5)$$

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 2-(2-Butoxy ethoxy) ethanol (BC) and amines respectively, d^1 is a constant [26, 27] and X_1 and X_2 are the mole fraction of 2-(2-Butoxy ethoxy) ethanol (BC) and corresponding amines respectively.

The above calculated values are also included in Table.2.

Table:2:Values of Density (ρ), Viscosity (η), Excess viscosity (η^E),, Excess molar volume (V^E), Excess Gibbs free energy of activation of viscous flow (G^{*E}) and Grunberg-Nissan interaction parameter (d^1) for the binary liquid mixtures of Butyl carbitol (BC) + N-Butyl amine (NBA) at 308.15 K

Mole fraction of Butyl carbitol X_{BC}	ρ x 10^{-3} Kg m^{-3}	η x 10^3 Kg m^{-1} s^{-1}	η^E x 10^3 Kg m^{-1} s^{-1}	V^E x10^5 m^3 mol^{-1}	G^{*E} x 10^3 N mol^{-1}	d^1
Butyl carbitol (BC) + N-Butyl amine (NBA)						
0	0.7241	0.4248	0.0000	0.0000	0.0000	---
0.0611	0.7457	0.7304	0.1189	-0.0011	259.1301	7.2084
0.1276	0.7673	1.036	0.2212	-0.0079	392.7322	5.5985
0.2005	0.7889	1.3416	0.3040	-0.0105	461.5525	4.5439
0.2807	0.8105	1.6472	0.3645	-0.0113	487.3444	3.7884
0.3692	0.8321	1.9528	0.3996	-0.0156	479.7355	3.2158
0.4675	0.8537	2.2584	0.4048	-0.0192	442.7938	2.7618
0.5773	0.8753	2.564	0.3748	-0.0227	377.8210	2.3910
0.7007	0.8969	2.8696	0.3033	-0.0273	284.1864	2.0812
0.8405	0.9185	3.1752	0.1816	-0.0304	159.4479	1.8169
1	0.9399	3.4811	0.0000	0.0000	0.0000	---
Butyl carbitol (BC) + Sec-Butyl amine (SBA)						
0	0.7084	0.3994	0	0	0	---
0.0623	0.7316	0.7076	0.1162	-0.0090	273.1110	7.4823
0.1301	0.7548	1.0156	0.2153	-0.0154	409.3522	5.7584
0.2041	0.778	1.324	0.2956	-0.0217	477.6902	4.6581
0.2851	0.8012	1.6322	0.3542	-0.0320	501.5363	3.8789
0.3743	0.8244	1.9404	0.3875	-0.0403	491.1298	3.2896
0.473	0.8476	2.2486	0.3916	-0.0471	451.0657	2.8247
0.5826	0.8708	2.5572	0.3624	-0.0589	383.2591	2.4484
0.7053	0.894	2.8654	0.2925	-0.0677	286.8578	2.1338
0.8434	0.9172	3.1736	0.1751	-0.0786	160.1982	1.8673
1	0.9399	3.4811	0	0	0	---
Butyl carbitol (BC) + Tert-Butyl amine (TBA)						
0	0.6809	0.4122	0	0	0	---
0.0647	0.7068	0.7191	0.1083	0.0008	261.0791	6.9162
0.1347	0.7327	1.026	0.2004	0.0024	391.4984	5.3591
0.2106	0.7586	1.3329	0.2744	0.0007	456.0710	4.3573
0.2932	0.7845	1.6398	0.3278	-0.0033	477.6654	3.6451
0.3836	0.8104	1.9467	0.3573	-0.0021	466.2363	3.1045
0.4828	0.8363	2.2536	0.3597	-0.0020	426.6532	2.6784
0.5922	0.8622	2.5605	0.3309	-0.0012	360.7833	2.3314
0.7135	0.8881	2.8674	0.2655	0.0018	268.6851	2.0419
0.8485	0.914	3.1743	0.1581	-0.0010	149.3790	1.7972
1	0.9399	3.4811	0	0	0	---

Butyl carbitol (BC) + N-Hexyl amine (NHA)						
0	0.7522	0.6001	0	0	0	---
0.0797	0.771	0.8881	0.0584	-0.0058	155.7336	3.4346
0.1631	0.7898	1.1761	0.1061	-0.0106	239.1864	2.8293
0.2504	0.8086	1.4642	0.1427	-0.0169	280.3280	2.4073
0.342	0.8274	1.7523	0.1669	-0.0204	292.3672	2.0905
0.4381	0.8462	2.0404	0.1781	-0.0253	282.4380	1.8431
0.539	0.865	2.3285	0.1755	-0.0330	254.6279	1.6436
0.6453	0.8838	2.6166	0.1574	-0.0364	211.1655	1.4774
0.7572	0.9026	2.9047	0.1231	-0.0426	153.7422	1.3374
0.8752	0.9214	3.1928	0.0712	-0.0505	83.1920	1.2174
1	0.9399	3.4811	0	0	0	---
Butyl carbitol (BC) + N-Octyl amine (NOA)						
0	0.7704	0.9263	0	0	0	---
0.0975	0.7873	1.1818	0.0064	0.0121	70.2027	1.3016
0.1955	0.8042	1.4373	0.0115	0.0219	110.6690	1.1478
0.294	0.8211	1.6928	0.0154	0.0294	131.0501	1.0298
0.3932	0.838	1.9483	0.0175	0.0414	136.7612	0.9346
0.4929	0.8549	2.2038	0.0182	0.0508	131.4276	0.8571
0.5931	0.8718	2.4593	0.0177	0.0576	117.3859	0.7925
0.694	0.8887	2.7148	0.0155	0.0682	96.1385	0.7370
0.7954	0.9056	2.9703	0.0119	0.0759	69.0211	0.6895
0.8974	0.9225	3.2258	0.0068	0.0838	36.8641	0.6481
1	0.9399	3.4811	0	0	0	---
Butyl carbitol (BC) + Cyclohexyl amine (CHA)						
0	0.8525	1.3245	0	0	0	---
0.0697	0.8612	1.5402	0.0654	0.0059	54.6496	1.2884
0.1442	0.8699	1.7559	0.1204	0.0112	93.8406	1.1558
0.2241	0.8786	1.9716	0.1638	0.0170	119.9692	1.0426
0.3101	0.8873	2.1873	0.1940	0.0248	134.4747	0.9443
0.4027	0.896	2.403	0.2100	0.0316	138.3242	0.8589
0.5028	0.9047	2.6187	0.2099	0.0388	131.8963	0.7833
0.6113	0.9134	2.8344	0.1916	0.0458	115.2891	0.7160
0.7295	0.9221	3.0501	0.1524	0.0551	88.1691	0.6549
0.8585	0.9308	3.2658	0.0899	0.0637	50.1426	0.6001
1	0.9399	3.4811	0	0	0	---

The dependence of η^E, V^E and G^{*E} on the mole fraction of 2-(2-Butoxy ethoxy) ethanol (BC) X_{BC} for all the six systems were fitted to the following Redlich-Kister equation by the least-squares method and the values are given in Table.3.

$$Y^E = x(1-x)\sum_{i=1}^{4} A_i (2x-1)^I \quad \text{…. (6)}$$

Where Y^E is η^E, V^E and G^{*E} parameters.

The parameters A_i, obtained by a non-linear least squares polynomial fitting procedure, are also given in Table.3 together with the standard deviation (σ) values.

The variation of the parameters η, V^E, η^E and G^{*E} with mole fraction of 2-(2-Butoxy ethoxy) ethanol (X_{BC}) for the systems under study are shown graphically in Figs. 1 to 4 respectively.

Table 3: The Co-efficients of a_i of the Redlich-Kister type polynomial equation (6) and the corresponding Standard deviations σ (Y^E) for the binary mixtures of BC+ Amines at 308.15 K

Excess Property	A_0	A_1	A_2	A_3	A_4	σ
Butyl carbitol (BC) + N- Butyl amine (NBA)						
$\eta^E \times 10^3$ (Kg m^{-1} s^{-1})	0.00038	2.13936	-3.45663	1.77913	-0.4621	0.00054
$V^E \times 10^6$ (m^3 mol^{-1})	0.00168	-0.11544	0.43405	-0.83539	0.51449	0.00211
$G^{*E} \times 10^3$ (N mol^{-1})	0.00368	6082.45	-39420.93	185595.34	-634873.79	0.27222
Butyl carbitol (BC) + Sec-Butyl amine (SBA)						
$\eta^E \times 10^3$ (Kg m^{-1} s^{-1})	0.00039	2.0467	-3.25294	1.62655	-0.42085	0.00046
$V^E \times 10^6$ (m^3 mol^{-1})	0.00246	-0.26125	0.99776	-1.99642	1.25542	0.00558
$G^{*E} \times 10^3$ (N mol^{-1})	0.00414	6395.48	-42763.92	206780.59	-717715.08	0.30901
Butyl carbitol (BC) + Tert-Butyl amine (TBA)						
$\eta^E \times 10^3$ (Kg m^{-1} s^{-1})	0.00019	1.84459	-2.8349	1.30484	-0.31478	0.00021
$V^E \times 10^6$ (m^3 mol^{-1})	0.00053	-0.01388	0.12164	-0.22765	0.12096	0.00172
$G^{*E} \times 10^3$ (N mol^{-1})	0.0025	5866.37	-37410.27	173438.22	-584378.18	0.22165
Butyl carbitol (BC) + N-Hexyl amine (NHA)						
$\eta^E \times 10^3$ (Kg m^{-1} s^{-1})	-0.000025	0.81246	-1.0339	0.26479	-0.04338	0.000082
$V^E \times 10^6$ (m^3 mol^{-1})	0.00225	-0.18836	0.79397	-1.53883	0.92809	0.00544
$G^{*E} \times 10^3$ (N mol^{-1})	0.00017	2694.098	-11858.09	39812.63	-110679.94	0.08982
Butyl carbitol (BC) + N-Octyl amine (NOA)						
$\eta^E \times 10^3$ (Kg m^{-1} s^{-1})	0.000038	0.07419	-0.07662	0.00399	-0.0015	0.000094
$V^E \times 10^6$ (m^3 mol^{-1})	-0.00348	0.32717	-1.39639	2.69207	-1.61225	0.01039
$G^{*E} \times 10^3$ (N mol^{-1})	0.0000009	935.1578	-2709.08	6598.76	-18191.67	0.01579
Butyl carbitol (BC) + Cyclohexyl amine (CHA)						
$\eta^E \times 10^3$ (Kg m^{-1} s^{-1})	0.000059	1.03905	-1.50728	0.60891	-0.14078	0.00013
$V^E \times 10^6$ (m^3 mol^{-1})	-0.0026	0.20608	-0.83939	1.70709	-1.06873	0.00555
$G^{*E} \times 10^3$ (N mol^{-1})	0.00021	940.313	-2519.27	4477.44	-7976.19	0.02398

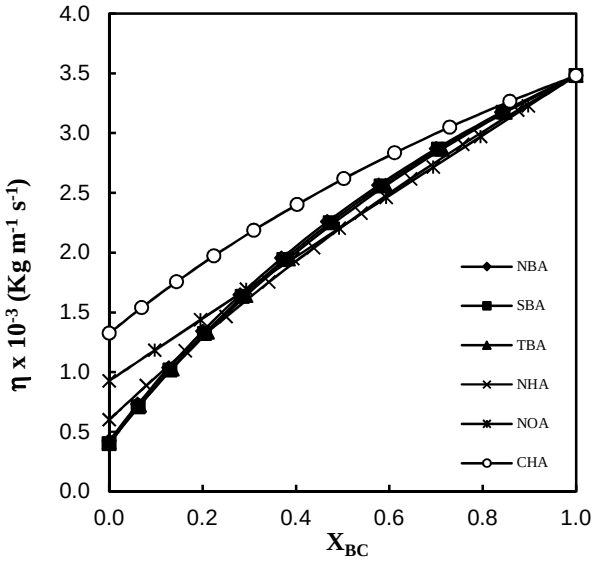


Fig.1: Plots of Viscosities (η) for various amines Vs mole fractions of Butyl Carbitol (X_{BC}) at 308.15 K

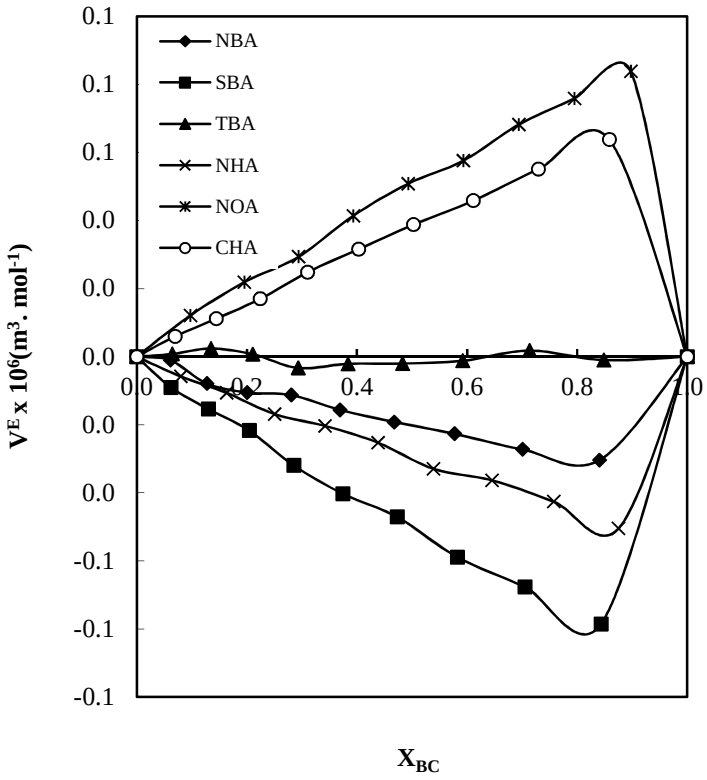


Fig.2: Plots of excess volumes (V^E) for various amines vs mole fractions of Butyl carbitol (X_{BC}) at 308.15 K

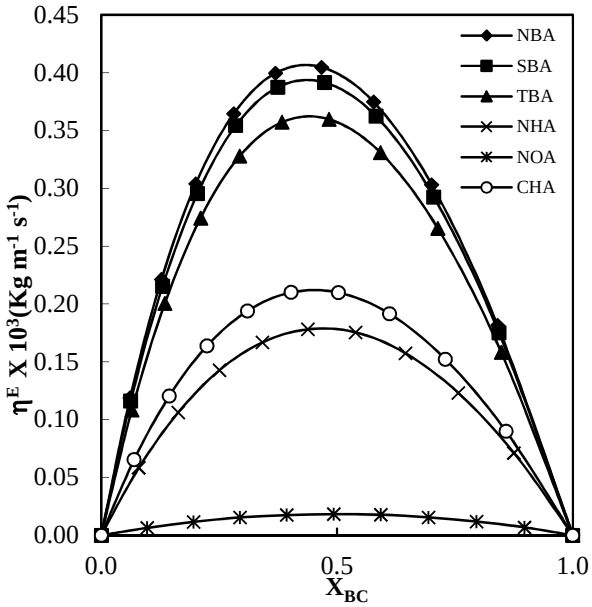


Fig.3: Plots of excess viscosities (η^E) for various Amines vs. mole fraction of Butyl Carbitol (X_{BC}) at 308.15 K

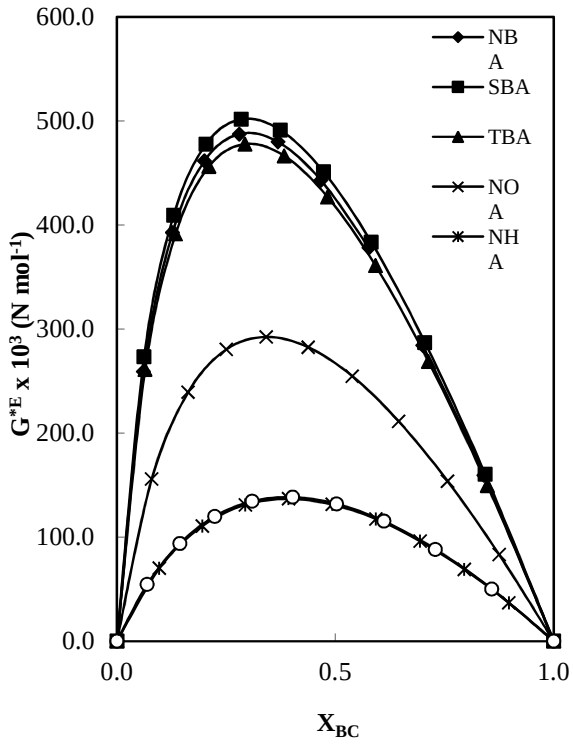


Fig. 4: Plots of excess Gibbs free energy of activation of viscous flow (G^{*E}) for various amines vs. mole fraction of Butyl Carbitol X_{BC} at 308.15 K

From Table.1and Fig.1, it is observed that the viscosity of binary liquid mixtures under study varied non-linearly with the mole fraction of 2-(2-Butoxy ethoxy) ethanol (X_{BC}). This suggests the presence of intermolecular interactions between unlike molecules of these mixtures. A similar observation was made by Narayana Swamy et.al [24] from the viscosity studies of binary liquid mixtures of BC + amines.

It is clear from the Fig.2 that the negative V^E values are obtained over the entire composition range for all these systems except BC+NOA and BC+ CHA which indicates the presence of strong molecular interactions between the unlike components of the mixtures. It is also observed from Fig.2 and Table.2 that the V^E values fall in the sequence.

BC + NOA > BC + CHA > BC + TBA > BC + NBA > BC +NHA > BC + SBA.

From Fig.2, it is further observed that the negative/ Positive V^E Vs X_{BC} plots were found to be large and symmetrical showing a maximum between 0.5 to 0.8 mole fractions of BC (X_{BC})

According to Subha et al., [28] several effects may contribute to the sign and values of V^E and the following three effects may be considered as being important.

1. Break up of hydrogen bonds and dipolar interactions in BC and intermolecular Interactions in amines.
2. The possible intermolecular interactions like hydrogen bonding or electron donor-acceptor interactions between unlike molecules.
3. Interstitial accommodation of one component molecules into the other unlike Component molecules due to their differences in size and shape.

The overall signs of these excess functions depend on the combined effects of these three effects. From the Fig. 2 it is noticed that in the case of BC + NOA and BC + CHA positive V^E values are observed. An interpretation to this

behavior can be given using the experience made with the quantitative evaluation of alcohol + amine mixtures by the ERAS model proposed by the earlier workers [29-33]. The first effect leads to positive excess volume and the latter two effects leads to negative excess volume. The actual volume change would depend upon the relative strengths of these three effects. In the present study the observed negative values of V^E show that the main contribution to V^E is due to hydrogen bond formation between hydroxyl & ethereal groups of BC and amino groups of amines and the difference in size of the unlike molecules. Moreover, the negative values of V^E may also be partly due to the specific acid-base interactions between BC and amine molecules by considering BC as Lewis acid and amines as Lewis bases.

Very recently NamTram [34] emphasized the importance of acid-base interactions between tert-butyl alcohol and N, N dimethyl formamide / N, N-dimethyl acetamide in order to evaluate the interaction energy of alcohol-amine systems. From the examination of the results in Table.1and Fig.2.

It is observed that the magnitude values of V^E fall in the sequence :
BC +NOA > BC + CHA > BC+TBA > BC + NBA > BC + NHA > BC + SBA.

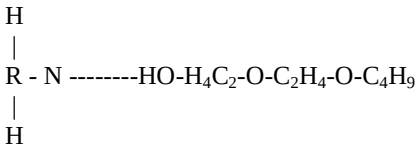
From the Fig .2, it is also observed that the negative excess volumes of normal amines with BC increase with increase in chain length of amines. This can be explained by considering amines as proton acceptors and BC as proton donor. As the chain length of normal amines increases proton accepting ability of these amines increase and electron density will be more and more on nitrogen atom of NH_2 group due to inductive effect. So, as the chain length of the amines increase, the interaction ability (hydrogen bonding ability) of amines increase with increase in BC component. In the case of branched amines, negative V^E values increase with increase in branching. This is very clear that as the amines become more and more branched, the proton accepting ability increases due to the increase in $–CH_3$ groups on the carbon atom attached to amine group. However, the slight positive V^E values are observed at higher BC mole fraction in case of NBA & CHA indicating lesser specific interactions at these mole fractions. A similar observation was made by Haraschita. et.al. [35] from the V^E studies of γ- Picoline + sec-butanol and + tert-butanol.

In the case of cyclohexylamine, V,^{E} values are less positive than the corresponding normal hexyl amine, which may be due to steric and other effects because of cyclic nature of cyclohexylamine.

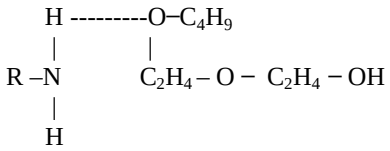
From the Fig.3, it is noticed that shows that of η^E values are positive over the whole composition range for all the systems under study. A correlation between the signs of η^E and V^E has been observed for a number of binary systems [36, 37] where η^E being Positive and V^E is negative and vice-versa. In general, for 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 thereby resulting in positive η^E values. The positive values of η^E for the mixtures of BC + amines fall in the order.

BC + NBA > + BC + SBA > BC + TBA > + > + BC + CHA > + NHA > + BC +NOA

It can be predicted that in view of the strong proton donating ability of BC and strong proton accepting ability of amines, the overall negative V^E values and overall positive η^E values in all these systems may be regarded as an evidence for the formation of two sets of hydrogen bonds between BC and amines. Among them the first set of hydrogen bond formation is between N atom of amino group of amine and H atom of – OH group of 2-(2-Butoxy ethoxy) ethanol



and second set of hydrogen bond formation is between H atom of amino group of amines and oxygen atom of etheric group of 2-(2-Butoxy ethoxy) ethanol (BC). Among the two the first set of hydrogen bond formation predominates.



From the Table.1& Fig.4, it is also observed that the values of G^{*E} are found to be positive which is an indication of the presence of strong specific interactions between the unlike molecules and are in the following order

BC+ SBA > + BC + NBA > BC + TBA > BC + NOA > + BC + CHA > + BC + NHA

Fort and Moore [38] and Ramana moorthy [39] reported that for any binary liquid mixture, the positive value of d^1 indicates the presence of strong interactions and the negative value of ^{d1} indicates the presence of weak interactions between the components. On this basis the d^1 values in the present study for all the systems confirm the presence of strong interactions between the component molecules. A similar observation was made by Subha et.al. [24] from the d^1 values of the binary liquid mixtures of propionic acid with alcohols.

CONCLUSION

The present study reports the volumetric and viscometric properties of 2-(2-Butoxy ethoxy) ethanol (BC) with n-butyl amine, sec- butyl amine, tert- butyl amine, n-hexyl amine, n – Octyl amine and cyclo hexyl amine over the entire composition range at 305.15 K. From the results, it is observed that the variation of the properties of the mixtures studied supports the view 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 ,G^{*E} and d^1 values.

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