

# Thermodynamic Properties of Organic Liquid Mixtures Related to Molecular Interactions between the Components

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**Abstract-** In the present paper, thermodynamic properties of organic liquid mixtures related to molecular interactions between the components have been analysed intensively. In addition to that, the present paper also includes a brief discussion on comparative study between the experimental and theoretically calculated values of refractive indices at three temperatures 293, 303 and 313K. The results have been discussed in terms of average percentage deviation (APD).

## 1. INTRODUCTION

The knowledge of the thermodynamic properties of organic liquid mixtures is of immense importance for understanding the molecular interactions between the components. This also helps to evolve theoretical models and is useful in industrial applications [1 - 4]. Excess properties of liquid systems, such as excess molar volume, deviation in molar refraction are needed for the design of separation equipment and to test the theories of the solution [5]. In addition, excess properties provide information about the molecular interactions and macroscopic behaviour of fluid mixtures and can be used to test and to improve thermodynamical models for calculating and predicting the fluid phase equilibria. The knowledge of excess Gibb's free energy of activation for viscous flow helps in understanding the molecular orientation and to study the extent of intermolecular interaction between component molecules of the liquid mixtures. In recent years, there has been considerable upsurge in the theoretical and experimental investigation of the excess thermodynamic properties of binary liquid mixtures [6, 7].

In order to examine molecular interactions in the mixture of butylamine with alcohols through excess properties viz.

excess molar volume ( $V_m^E$ ), molar refraction deviation ( $\Delta R_m$ ), viscosity deviation ( $\Delta \eta$ ) and excess Gibb's free energy of activation for viscous flow ( $\Delta G^{*E}$ ), this chapter reports here the density ( $\rho_m$ ), refractive index ( $n_m$ ) and viscosity ( $\eta_m$ ) for binary mixtures of butylamine with 1-butanol and *tert*-butanol over the entire composition range at temperatures 293, 303 and 313K. Further, seven mixing rules for refractive index proposed by Lorentz-Lorenz (L-L), Gladstone-Dale (G-D), Weiner (W), Heller (H), Arago- Biot (A-B), Newton (N) and Eykmen (EyK) for the prediction of refractive index theoretically have been applied to these binary mixtures. A comparative study has been made between the experimental and theoretically calculated values of refractive indices at all the three temperatures. The results have been discussed in terms of average percentage deviation (APD).

## 2. RESULTS

The experimental values of density ( $\rho_m$ ), refractive index ( $n_m$ ) and viscosity ( $\eta_m$ ) at three temperatures viz. 293, 303 and 313K along with the calculated values of excess molar volume ( $V_m^E$ ), molar refraction deviation ( $\Delta R_m$ ), viscosity deviation ( $\Delta \eta$ ) and excess Gibb's free energy of activation for viscous flow ( $\Delta G^{*E}$ ) are given in tables

5.1 and 5.2 for butylamine + 1-butanol and butylamine + *tert*-butanol mixtures respectively. Adjustable parameters  $a_i$  with the standard deviations  $\sigma(V_m^E)$  for both the binary mixtures at all the three temperatures 293, 303 and 313K are given in *table 5.3*. Average percentage deviations (APD) of theoretically calculated refractive indices from different semi-empirical relations are given in *table 5.4*. All the excess parameters viz.  $V_m^E$ ,  $\Delta R_m$ ,  $\Delta\eta$  and  $\Delta G^{*E}$  plotted against the mole fraction of butylamine ( $X_1$ ) are shown in *table 5.1 to 5.4* respectively.

### 3. DISCUSSION

According to Fort and Moore [8], a negative excess molar volume ( $V_m^E$ ) is an indication of strong hetero-molecular interaction in the liquid mixtures and is attributed to change-transfer, dipole-dipole interaction and hydrogen bonding between the unlike molecules, while a positive sign indicates a weak interaction and is attributed to dispersive forces. The magnitudes of the contribution will vary with the components and composition of the mixtures. In the present investigation, both the mixtures gave negative magnitude of  $V_m^E$ . This depicts the presence of hetero-molecular interaction, which supports the Fort and Moore [8] observation. Wen-Lu Weng [9] and Papaioannou *et al.* [10] have also observed negative values of  $V_m^E$  in the case of butylamine + 1-butanol mixture. The negative  $V_m^E$  values of butylamine + 1-butanol observed in the present investigation have been found to be in good agreement with the values of  $V_m^E$  reported earlier by Wen-Lu Weng [9] and Papaioannou *et al.* [10]. The negative values of  $V_m^E$  indicate that volume contraction takes place upon mixing of butylamine with 1-butanol/*tert*-butanol due to cross association between these dissimilar molecules [11]. The magnitude of the volume contraction follows the sequence of 1-butanol > *tert*-butanol. This is in agreement with the results reported by Ali and Nain [11] on binary mixtures of THF with 1-butanol and *tert*-butanol which suggests that the excess values follow the same sequence, due to the steric hindrance produced by the presence of three methyl groups at 3<sup>o</sup> carbon atom in *tert*-butanol, as compared to one butyl group in 1-butanol. Molar refraction deviation ( $\Delta R_m$ ) represents the electronic perturbation due to orbital mixing of two components [12].  $\Delta R_m$  gives

the strength of the interaction in mixture and is a sensitive function of wavelength, temperature and mixture composition. *Table 5.2* shows that  $\Delta R_m$  values are negative for both the mixtures at all the temperatures. The values of  $V_m^E$  and  $\Delta R_m$  support each other. The  $V_m^E$  and  $\Delta R_m$  values become more negative at higher temperature for both the mixtures, suggesting an increase in interaction between unlike molecules.

*Table 5.3* shows that viscosity deviation ( $\Delta\eta$ ) values are positive for butylamine + 1-butanol and negative for butylamine + *tert*-butanol mixtures. The positive values of  $\Delta\eta$  in butylamine + 1-butanol mixture can be explained on the basis of complex formation between unlike molecules through hydrogen bonding. The negative values of  $\Delta\eta$  for butylamine + *tert*-butanol may be attributed to the presence of weak dipole-dipole interaction due to predominance rupture of dipolar association of *tert*-butanol and steric hindrance produced by three methyl group over the hydrogen bond formed between unlike molecules. Our positive values of  $\Delta\eta$  in the case of butylamine + 1-butanol do not corroborate with the values of  $\Delta\eta$  on the same mixtures reported earlier by Wen-Lu Weng [9] and Dominguez *et al.* [13]. However all the excess parameters  $V_m^E$ ,  $\Delta R_m$  and  $\Delta\eta$  reported here, are in conformity with each other.  $\Delta\eta$  values are found to decrease with increasing temperature in the positive and negative directions in the case of butylamine + 1-butanol and butylamine + *tert*-butanol mixtures respectively.

*Table 5.4* shows that,  $\Delta G^{*E}$  values are positive for butylamine + 1-butanol and negative for butylamine + *tert*-butanol mixtures respectively. However, a smaller positive trend has been observed at temperature 313K in butylamine rich region. The positive values of  $\Delta G^{*E}$  indicates the presence of strong interaction in butylamine + 1-butanol mixture, whereas a negative  $\Delta G^{*E}$  values for butylamine + *tert*-butanol mixture may be assigned to the predominance of dispersive forces. This is due to the fact that C-O bond is weaker in the case of tertiary alcohols due to +I (electron repelling) effect of alkyl groups while O-H bond is weaker in primary alcohols as electron density increases between O-H bond and hydrogen tends to separate as a proton. During O-H cleavage, proton is given out showing that alcohols are acidic in nature. Acidic nature increases from tertiary to secondary and from secondary to

primary as +I effect decreases [14]. Gupta *et al.* [15] and Mialkowski *et al.* [16] have also reported similar variations in the  $\Delta G^{*E}$  values for binary mixtures of ether +alcohols and  $\gamma$ -butyrolactone + dimethylcarbonate.

Further, Seven empirical relations were used for the prediction of refractive indices. The refractive index data correlated with these semi-empirical equations for both the binary mixtures show an excellent agreement with the respective experimental values of refractive index. It can be seen from *table 5.4* that the correlation of these equations showed the suitability of all the seven relations for representing the mixing refractive indices of the binary mixtures of butylamine + 1-butanol/*tert*-butanol. The applicability of these semi-empirical relations for predicting refractive index has also been emphasized by others [17, 18].

#### 4. CONCLUSION

The measured values of refractive index and viscosity along with the estimated values of various developed parameters suggest the existence of hetero-molecular association through hydrogen bonding in butylamine + 1-butanol mixture while behaviour of butylamine + *tert*-butanol mixture indicates the presence of long range dispersive forces among the components. The behaviour of all the excess parameters studied here supports each other. The various semi-empirical relations for representing the refractive index data theoretically give reasonably good results.

**Table 5.1**

Density ( $\rho_m$ ), refractive index ( $n_m$ ), viscosity ( $\eta_m$ ), excess molar volume ( $V_m^E$ ), molar refraction deviation ( $\Delta R_m$ ), viscosity deviation ( $\Delta\eta$ ) and excess Gibb's free energy of activation for viscous flow ( $\Delta G^{*E}$ ) for butylamine + 1-butanol mixture with mole fraction of butylamine ( $x_1$ ) at T= 293, 303 and 313K.

$x_1$	$\rho_m$ (g.cm <sup>-3</sup> )	$n_m$	$\eta_m$ (mPa.s)	$V_m^E$ (cm <sup>3</sup> .mol <sup>-1</sup> )	$\Delta R_m$	$\Delta\eta$ (mPa.s)	$\Delta G^{*E}$ (kJ.mol <sup>-1</sup> )
<b>T=293K</b>							
0.0000	0.8098	1.399	2.8250	0.0000	0.0000	0.0000	0.0000
0.1011	0.8029	1.399	2.7285	-0.0600	-0.0496	0.1268	0.2895
0.2025	0.7967	1.400	2.6276	-0.1870	-0.0879	0.2500	0.5713
0.3028	0.7915	1.401	2.5339	-0.4110	-0.1217	0.3779	0.8496
0.4032	0.7848	1.401	2.3650	-0.4630	-0.1357	0.4307	1.0530
0.5033	0.7780	1.401	2.1510	-0.4780	-0.1416	0.4379	1.1932
0.6032	0.7707	1.402	1.9208	-0.4200	-0.1205	0.4284	1.2897
0.7030	0.7632	1.402	1.6457	-0.3160	-0.1047	0.3737	1.2859
0.8020	0.7558	1.402	1.3375	-0.2070	-0.0894	0.2842	1.1506
0.9000	0.7484	1.403	0.9978	-0.0760	-0.0391	0.1611	0.8034
1.0000	0.7414	1.403	0.6158	0.0000	0.0000	0.0000	0.0000
<b>T=303K</b>							
0.0000	0.8017	1.392	2.2970	0.0000	0.0000	0.0000	0.0000
0.1011	0.7953	1.392	2.2045	-0.1280	-0.0610	0.0855	0.2644
0.2025	0.7893	1.393	2.1168	-0.3000	-0.1047	0.1765	0.5299
0.3028	0.7835	1.393	2.0382	-0.4800	-0.1473	0.2744	0.7979
0.4032	0.7769	1.393	1.8997	-0.5460	-0.1846	0.3128	0.9874
0.5033	0.7700	1.393	1.7283	-0.5680	-0.1874	0.3177	1.1159
0.6032	0.7628	1.394	1.5458	-0.5300	-0.1719	0.3112	1.2021
0.7030	0.7554	1.394	1.3303	-0.4570	-0.1545	0.2714	1.1916
0.8020	0.7480	1.394	1.0910	-0.3561	-0.1215	0.2064	1.0572
0.9000	0.7405	1.395	0.8289	-0.2200	-0.0547	0.1170	0.7276
1.0000	0.7324	1.395	0.5358	0.0000	0.0000	0.0000	0.0000
<b>T=313K</b>							

0.0000	0.7934	1.389	1.8880	0.0000	0.0000	0.0000	0.0000
0.1011	0.7872	1.389	1.8014	-0.1770	-0.0740	0.0569	0.2404
0.2025	0.7813	1.389	1.7252	-0.3840	-0.1524	0.1247	0.4906
0.3028	0.7752	1.390	1.6587	-0.5570	-0.1940	0.2006	0.7479
0.4032	0.7684	1.390	1.5443	-0.6200	-0.2055	0.2286	0.9246
0.5033	0.7614	1.390	1.4071	-0.6510	-0.2108	0.2335	1.0450
0.6032	0.7540	1.391	1.2591	-0.6200	-0.1970	0.2274	1.1191
0.7030	0.7466	1.391	1.0884	-0.5540	-0.1813	0.1984	1.1036
0.8020	0.7390	1.391	0.9004	-0.4480	-0.1520	0.1509	0.9716
0.9000	0.7314	1.392	0.6960	-0.3200	-0.0810	0.0855	0.6596
1.0000	0.7227	1.392	0.4685	0.0000	0.0000	0.0000	0.0000

Table 5.2

Density ( $\rho_m$ ), refractive index ( $n_m$ ), viscosity ( $\eta_m$ ), excess molar volume ( $V_m^E$ ), molar refraction deviation ( $\Delta R_m$ ), viscosity deviation ( $\Delta\eta$ ) and excess Gibb's free energy of activation for viscous flow ( $\Delta G^{*E}$ ) for butylamine + *tert*-butanol mixture with mole fraction of butylamine ( $x_1$ ) at T= 293, 303 and 313K.

$x_1$	$\rho_m$ (g.cm <sup>-3</sup> )	$n_m$	$\eta_m$ (mPa.s)	$V_m^E$ (cm <sup>3</sup> .mol <sup>-1</sup> )	$\Delta R_m$	$\Delta\eta$ (mPa.s)	$\Delta G^{*E}$ (kJ.mol <sup>-1</sup> )
<b>T=293K</b>							
0.0000	0.8018	1.396	6.6250	0.0000	0.0000	0.0000	0.0000
0.1012	0.7953	1.397	3.3710	-0.0003	-0.0058	-2.6459	-1.0598
0.2015	0.7890	1.398	2.4141	-0.0006	-0.0138	-3.0000	-1.2923
0.3026	0.7827	1.398	1.8566	-0.0008	-0.0192	-2.9500	-1.3466
0.4031	0.7765	1.399	1.4827	-0.0009	-0.0221	-2.7200	-1.3127
0.5032	0.7704	1.400	1.2412	-0.0009	-0.0225	-2.3600	-1.1665
0.6029	0.7645	1.400	1.0820	-0.0009	-0.0205	-1.9200	-0.9238
0.7026	0.7586	1.401	0.9192	-0.0008	-0.0161	-1.4837	-0.7443
0.7980	0.7530	1.402	0.8050	-0.0006	-0.0097	-1.0246	-0.5155
0.9012	0.7470	1.403	0.7173	-0.0003	-0.0021	-0.4922	-0.1998
1.0000	0.7414	1.403	0.6158	0.0000	0.0000	0.0000	0.0000
<b>T=303K</b>							
0.0000	0.7944	1.389	5.5260	0.0000	0.0000	0.0000	0.0000
0.1012	0.7877	1.390	3.0631	-0.0003	-0.0068	-1.9639	-0.8910

0.2015	0.7812	1.390	2.2105	-0.0005	-0.0157	-2.3206	-1.1228
0.3026	0.7748	1.391	1.6560	-0.0006	-0.0220	-2.3740	-1.2558
0.4031	0.7684	1.392	1.4045	-0.0007	-0.0256	-2.1260	-1.0799
0.5032	0.7622	1.392	1.2752	-0.0007	-0.0265	-1.7563	-0.7345
0.6029	0.7561	1.393	1.1065	-0.0007	-0.0249	-1.4268	-0.5060
0.7026	0.7500	1.393	0.9783	-0.0006	-0.0209	-1.0555	-0.2304
0.7980	0.7443	1.394	0.8325	-0.0005	-0.0147	-0.7221	-0.0765
0.9012	0.7383	1.395	0.6678	-0.0003	-0.0057	-0.3670	-0.0254
1.0000	0.7325	1.395	0.5358	0.0000	0.0000	0.0000	0.0000
<b>T=313K</b>							
0.0000	0.7871	1.385	4.7960	0.0000	0.0000	0.0000	0.0000
0.1012	0.7802	1.386	2.9805	-0.0004	-0.0086	-1.3513	-0.6247
0.2015	0.7734	1.387	2.2640	-0.0007	-0.0189	-1.6139	-0.7326
0.3026	0.7666	1.387	1.6765	-0.0009	-0.0261	-1.7499	-0.9022
0.4031	0.7600	1.388	1.5416	-0.0010	-0.0302	-1.4419	-0.5120
0.5032	0.7535	1.389	1.3784	-0.0011	-0.0313	-1.1697	-0.1972
0.6029	0.7472	1.389	1.2170	-0.0010	-0.0295	-0.9031	0.0820
0.7026	0.7409	1.390	0.9894	-0.0009	-0.0248	-0.7080	0.1467
0.7980	0.7350	1.391	0.8258	-0.0007	-0.0177	-0.4724	0.2532
0.9012	0.7287	1.391	0.6088	-0.0004	-0.0073	-0.2629	0.0841
1.0000	0.7227	1.392	0.4685	0.0000	0.0000	0.0000	0.0000

Table 5.3

Adjustable parameters  $a_i$  with the standard deviations  $\sigma(Y^E)$  for excess molar volume ( $V_m^E$ ), molar refraction deviation ( $\Delta R_m$ ), viscosity deviation ( $\Delta \eta$ ) and excess Gibb's free energy of activation for viscous flow ( $\Delta G^{*E}$ ) for both the binary mixtures at varying temperatures.

Parameters	Temp. (K)	$a_1$	$a_2$	$a_3$	$a_4$	$a_5$	$\sigma^E$
<b>butylamine +1-butanol</b>							
$V_m^E$ (cm <sup>3</sup> .mol <sup>-1</sup> )	293	-2.735	0.109	5.788	0.010	-4.235	0.0185
	303	-2.242	0.118	0.715	1.120	-0.361	0.0088
	313	-2.648	-0.316	0.403	1.869	0.635	0.0079
$\Delta R_m$	293	-0.308	0.117	-1.220	-0.289	1.463	0.0026
	303	-0.752	-0.608	1.434	0.892	2.580	0.0031
	313	-0.845	-0.930	-1.454	6.293	8.202	0.0416
$\Delta \eta$ (mPa.s)	293	1.759	1.350	-2.887	-2.490	4.127	0.0062
	303	1.276	1.094	-2.322	-2.056	3.266	0.0051
	313	0.935	0.845	-1.808	-1.635	2.478	0.0040
$\Delta G^{*E}$ (kJ.mol <sup>-1</sup> )	293	4.766	-1.248	-0.844	-3.643	4.487	0.0062
	303	4.455	-0.861	-1.408	-3.710	4.768	0.0067
	313	4.170	-0.717	-1.539	-3.459	4.419	0.0062
<b>butylamine +tert-butanol</b>							
$V_m^E$ (cm <sup>3</sup> .mol <sup>-1</sup> )	293	-0.004	0.001	0.004	-0.001	-0.004	0.0001
	303	-0.003	-0.002	0.003	0.001	-0.002	0.0001
	313	-0.004	-0.006	0.005	0.006	-0.009	0.0001
$\Delta R_m$	293	-0.093	0.007	-0.006	-0.049	0.129	0.0001
	303	-0.109	0.015	-0.013	-0.033	0.117	0.0001
	313	-0.125	0.003	-0.012	-0.016	0.107	0.0001
$\Delta \eta$ (mPa.s)	293	-9.343	-8.271	-9.493	-10.330	-4.951	0.0307
	303	-7.115	-13.362	10.931	3.690	-31.198	0.0363
	313	-4.430	-4.576	-3.854	-4.474	-4.922	0.0569
$\Delta G^{*E}$ (kJ.mol <sup>-1</sup> )	293	-4.560	-3.564	-4.555	-3.668	1.240	0.0182
	303	-3.547	-5.248	1.824	-1.055	-6.496	0.0384
	313	-0.782	-6.330	-0.259	2.387	-4.870	0.0736

Table 5.4

Average percentage deviation (APD) of theoretically calculated refractive index from different semi-empirical relations at all the three temperatures.

Temp.(K)	L-L	G-D	W	H	A-B	N	Eyk
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**butylamine +1-butanol**

<b>293</b>	-0.07860	0.00278	0.00280	0.00285	0.00279	0.00272	-0.07384
<b>303</b>	-0.11447	-0.00617	-0.00615	-0.00613	-0.00617	-0.06620	-0.10786
<b>313</b>	-0.14150	-0.01283	-0.01282	-0.01279	-0.01283	-0.01287	-0.13357

**butylamine +tert-butanol**

<b>293</b>	0.00799	0.00800	0.00806	0.00820	0.00800	0.00781	0.00788
<b>303</b>	0.00587	0.00588	0.00593	0.00604	0.00589	0.00574	0.00579
<b>313</b>	0.00589	0.00591	0.00598	0.00612	0.00591	0.00592	0.00578

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In the course of his distinguished professional career, Dr. Maurya has been appointed as **Head Examiner** for UG and PG Programs from leading Indian Universities - **U.P. Technical University, Lucknow during 2005-06 and Chhatrapati Shahu Ji Maharaj University, Kanpur for three terms during 2000-2004**. On the basis and recognition of his knowledge and significant research contributions in diversified fields of Mathematical and

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