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Transport Properties of Binary Liquid Mixtures of γ–butyrolactone + aliphatic Alcohols Studied in the Temperature Range of (303.15 to 313.15) K

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***Abstract*— Densities (ρ) and viscosities (η) have been measured as a function of mole fraction for the mixtures of γ–butyrolactone (GBL) with 1-propanol (1-pro), 2-propanol (2-pro), 1-butanol (1-but) and 2-butanol (2-but) at the temperatures of 303.15, 308.15, and 313.15 K. From these measurements, deviation in viscosity (Δη) was calculated. The computed property has been fitted to a Redlich-Kister type polynomial equation to derive binary coefficients and standard deviations.**

Keywords*— *γ–butyrolactone; aliphatic alcohol; deviation in viscosity;binary mixture; binary coefficient.

I. INTRODUCTION

γ–butyrolactone is an important industrial solvent which has several advantageous physico-chemical properties such as a broad liquid range (–45 ºC to 205 ºC), medium relative permittivity (41.65 at 25 ºC) and high viscosity (1.76 m. Pas at 25 ºC) [1]. It is an excellent electrolytic solvent used in lithium batteries [2, 3] Generally, alcohols are self-associated liquids and exist in a three-dimensional network of H-bonding in the liquid state. Hence the study of binary mixtures of GBL with alcohols have been made to understand how the solution structure has been varying due to H-bonding interactions between hydroxyl groups of alcohol in presence of external polar solvent.

As part of our current research programme, to determine the different thermodynamic properties of binary liquid mixtures [4-10], we report here with the deviation in viscosity (Δη) of the binary systems: γ–butyrolactone (GBL) with homologous series of aliphatic alcohols (1-propanol, 2-propanol, 1-butanol and 2-butanol). The experimental results have been fitted to Redlich-Kister type polynomial equation using multi parametric non-linear regression analysis [11] to derive the binary coefficients and to estimate the standard deviation between experimental and calculated data.

II. EXPERIMENTAL

A. *Material and Methods*

γ–butyrolactone (Aldrich, >99 mol %) was purified by doing distillation under reduced pressure as reported by Aurbach and Gottlieb [12] 1-propanol, 2-propanol, 1-butanol and 2-butanol were obtained from Merk (>99%) India and used after single distillation. The isomeric alcohols were stored over type 3A x 1.5 nm molecular sieves. The purities of the samples were confirmed by comparing their physical properties with the reported data. The densities and viscosities of pure substances and their comparison with literature values are listed in table 1.

B. *General Procedure*

Binary mixtures were prepared by mass in air tight bottles and the mass measurements were performed on a Dhona 100 DS, India, single pan analytical balance with a resolution of ± $0.01.10^{-6}$ kg. The required properties of the mixture were measured on the same day. The uncertainty in mole fraction was estimated to be less than ± 1.10^{-4}. Density of pure liquids and their mixtures were determined by using a 1.10^{-5} m^3 double arm pycnometer [10]. The uncertainty in density values was found to be ± 4.10^{-5} $g.cm^{-3}$ and 1.10^{-3} cm^3 $.mol^{-1}$.

Ubbelohde viscometer [13] having a capacity of about 15ml and the capillary having a length of about 90 mm and 0.5 mm internal diameter has been used to measure the flow times of pure liquids and liquid mixtures and it was calibrated with benzene and doubly distilled water (water conductivity less than 1.10^{-6} $ohm^{-1}.cm^{-1}$) as their density 0.87381 and 0.87341 $g.cm^{-3}$ at 298.15 and 308.15 K and 0.9970 and 0.9940 $g.cm^{-3}$ at 298.15 and 308.15 K respectively. The detailed experimental procedure of viscometer was discussed earlier [8]. Viscosity values (η) of pure liquids and mixtures are calculated using the relation:

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$$\eta = (at - b/t)\rho \qquad \ldots.(1)$$

where **a** and **b** are the characteristic constants of the viscometer, **ρ** is the density and **t** represents the flow time. The flow time of pure liquids and liquid mixtures were repeated for 5 times. The uncertainty of viscosity was $\pm 0.005 \times 10^{-3}$ m Pas.

III. RESULTS AND DISCUSSION

Before The values of density (ρ), viscosity (η) and deviation in viscosity (Δη) for the binary mixtures of γ–butyrolactone (1) with 1-propanol (2), 2-propanol (2), 1-butanol (2) and 2-butanol (2) at temperatures T= 303.15, 308.15, and 313.15 K along with the mole fraction are listed in table 2.

The deviation in viscosity is calculated using the relation

$$\Delta\eta = \eta_m - (x_1\eta_1 + x_2\eta_2) \qquad \ldots.(2)$$

where η_m, η_1, η_2, x_1, and x_2 are viscosity of the liquid mixture, viscosity and mole fractions of pure liquids respectively.

The deviation in viscosity ΔY was fitted by the method of non linear least squares to a Redlich-Kister type polynomial[14]

$$\Delta Y = x_1x_2 \Sigma A_i (x_1 - x_2)_i \qquad \ldots.(3)$$

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:

$$\sigma(\Delta Y) = [\Sigma (\Delta Y_{obs} - \Delta Y_{cal}) / (n - m)]^{1/2} \qquad \ldots.(4)$$

where 'n' represents the number of experimental points and 'm' is the number of coefficients. It is found that for the solution of the fifth degree polynomial, the agreement between the experimental values and the calculated ones is satisfactory. The derived parameters (A_i) and the estimated standard deviation (σ) for Δη are given in table 3.

A perusal of table 2 shows that the value of viscosity deviation, Δη are negative over the entire composition range for all the binary liquid mixtures at T=303.15 to 313.15 K and falls in the order and the same has been depicted in the figures 1-3.

2-But > 1-But > 2-Pro > 1-Pro > 0

It is well known that alkanols are strongly self associated and the degree of association depending on the factors such as chain length, the position of the –OH group, temperature and dilution by other solvents. The strength of H-bonding in alkanols is expected to decrease with increase in their chain length and with the addition of dissimilar molecules which disrupt the H-bonding network. GBL structure is stabilized by dipolar-dipole interactions. Thus, both GBL and alkanols are self-associated liquids; mixing of these solvents leads to negative molar volumes V^E as reported earlier [15] and positive Δη values for the binary mixtures. However, it is observed that Δη values are negative for all the systems and become more negative with an increase in chain length. This indicates the presence of dispersion forces [16] which are dominating than dipolar interactions between the mixing components in these mixtures to be more negative.

It is noticed that many binary liquid mixtures have no simple correlation between the strength of the interactions and the observed properties. Rastogi [17] has suggested that the observed excess property is a combination of an interaction part and a non interaction part. An inter comparison of V^E and Δη supports the contention of Kaulgud [18] that two properties are determined by different factors may lead to anomaly in the observed properties.

The temperature dependence of deviation in viscosity for the binary mixtures of γ–butyrolactone with four alcohols was studied in the temperature interval 303.15 to 313.15 K. The values of Δη are in decreasing trend with increase in temperature indicating that the molecular interactions decreasing with temperature between dissimilar molecules.

IV. CONCLUSIONS

This paper reports experimental data for deviation in viscosity (Δη) at T = 303.15, 308.15 and 313.15 K for four systems of GBL+1-pro, +2-pro, +1-but, +2-but. The results were analyzed to explain the intermolecular interactions between mixing components. Position of hydroxyl group with alkyl chain length has shown significant effect on thermodynamic properties of the investigated systems. Fair correlations were obtained from calculated deviation in viscosity (Δη) using the Redlich-Kister type polynomial equation.

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International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 6 Issue 07, July - 2017

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TABLE I

Experimental densities (ρ) and viscosities (η) of pure liquids at T = 303.15 K.

	$\rho x10^{-3}/(kg{\cdot}m^{-3})$		$\eta x10^{3}/\ kg\ m^{-1}\ s^{-1}$	
Component	exptl.	lit.	exptl.	lit.
γ–Butyrolactone	1.12032	1.11883[19]	1.62298	1.612[20]
1-Propanol	0.80801#	0.79959[21]	1.96860	-----
2-Propanol	0.79560#	0.78100[22]	2.12583#	2.079[22]
1-Butanol	0.80654#	0.8060[23]	2.29730	-----
2-Butanol	0.79883	0.79891[24]	2.57166	2.492[24]

values at T=298.15 K

TABLE II

The values of density (ρ), viscosity (η), and deviation in viscosity (Δη) for the binary liquid mixtures at various temperatures.

x_1	ρ x 10^{-3} kg·m^{-3}	η x10^{3} kg m^{-1} s^{-1}	Δη x10^{3} kg m^{-1} s^{-1}
γ–Butyrolactone (1) + 1-Propanol (2)			
T = 303.15 K			
0.0000	0.8233	1.9686	0.0000
0.0207	0.8309	1.9522	-0.0087
0.1409	0.8705	1.7664	-0.1498
0.2623	0.9094	1.6378	-0.2332
0.3852	0.9474	1.5141	-0.3112
0.4913	0.9794	1.4614	-0.3245
0.6003	1.0112	1.4300	-0.3154
0.7242	1.0462	1.4110	-0.2883
0.8512	1.0811	1.4846	-0.1674
0.9776	1.1149	1.5757	-0.0294
1.0000	1.1203	1.5967	0.0000
T = 308.15 K			
0.0000	0.8180	1.7128	0.0000
0.0207	0.8253	1.6826	-0.0251
0.1409	0.8654	1.5716	-0.1068
0.2623	0.9041	1.4671	-0.1817
0.3852	0.9424	1.3561	-0.2627
0.4913	0.9746	1.3054	-0.2875
0.6003	1.0064	1.2896	-0.2767
0.7242	1.0418	1.2642	-0.2719
0.8512	1.0766	1.3084	-0.1967
0.9776	1.1095	1.4492	-0.0251
1.0000	1.1141	1.4688	0.0000
T = 313.15 K			
0.0000	0.8144	1.5120	0.0000
0.0207	0.8226	1.4995	-0.0096
0.1409	0.8628	1.3931	-0.0988
0.2623	0.9015	1.2880	-0.1865
0.3852	0.9395	1.2354	-0.2215
0.4913	0.9715	1.2030	-0.2387
0.6003	1.0033	1.2116	-0.2145
0.7242	1.0383	1.2080	-0.2003
0.8512	1.0727	1.2605	-0.1296
0.9776	1.1054	1.3621	-0.0099
1.0000	1.1096	1.3688	0.0000
γ–Butyrolactone (1) + 1-Butanol (2)			
T = 303.15 K			
0.0000	0.8038	2.2973	0.0000
0.0214	0.8111	2.2053	-0.0770
0.1441	0.8509	1.8610	-0.3354
0.2658	0.8914	1.6584	-0.4527
0.3914	0.9326	1.5190	-0.5041
0.4966	0.9670	1.4449	-0.5045
0.6264	1.0085	1.4320	-0.4265
0.7621	1.0522	1.4582	-0.3052
0.8728	1.0865	1.4930	-0.1928
0.9824	1.1152	1.5761	-0.0329
1.0000	1.1203	1.5967	0.0000
T = 308.15 K			
0.0000	0.7991	2.0147	0.0000
0.0214	0.8070	1.9230	-0.0800
0.1441	0.8464	1.6487	-0.2873
0.2658	0.8866	1.4701	-0.3995
0.3914	0.9276	1.3627	-0.4383
0.4966	0.9622	1.3031	-0.4405
0.6264	1.0026	1.2857	-0.3871
0.7621	1.0455	1.3373	-0.2614
0.8728	1.0785	1.3912	-0.1471
0.9824	1.1095	1.4446	-0.0338
1.0000	1.1141	1.4688	0.0000
T = 313.15 K			
0.0000	0.7959	1.7924	0.0000
0.0214	0.8026	1.7267	-0.0566
0.1441	0.8420	1.5054	-0.2259
0.2658	0.8837	1.3851	-0.2947
0.3914	0.9256	1.2746	-0.3520
0.4966	0.9596	1.2313	-0.3508
0.6264	0.9984	1.2487	-0.2784
0.7621	1.0364	1.2663	-0.2033
0.8728	1.0699	1.3268	-0.0959
0.9824	1.1043	1.3596	-0.0167
1.0000	1.1096	1.3688	0.0000
γ–Butyrolactone (1) + 2-Propanol (2)			
T = 303.15 K			
0.0000	0.7913	1.8492	0.0000
0.0222	0.7988	1.7758	-0.0678
0.1463	0.8412	1.5588	-0.2534
0.2695	0.8829	1.4318	-0.3493
0.3947	0.9251	1.3518	-0.3978
0.4999	0.9600	1.3147	-0.4083
0.6003	0.9930	1.3049	-0.3928
0.7253	1.0335	1.3277	-0.3384
0.8510	1.0735	1.4170	-0.2174
0.9758	1.1128	1.5504	-0.0525
1.0000	1.1203	1.5967	0.0000
T = 308.15 K			
0.0000	0.7857	1.5831	0.0000

International Journal of Engineering Research & Technology (IJERT)
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Vol. 6 Issue 07, July - 2017

0.0222	0.7934	1.5290	-0.0515
0.1463	0.8359	1.3626	-0.2038
0.2695	0.8775	1.2650	-0.2873
0.3947	0.9194	1.2110	-0.3270
0.4999	0.9541	1.1889	-0.3370
0.6003	0.9867	1.2063	-0.3082
0.7253	1.0270	1.2161	-0.2841
0.8510	1.0672	1.2998	-0.1861
0.9758	1.1066	1.4297	-0.0419
1.0000	1.1141	1.4688	0.0000
T = 313.15 K			
0.0000	0.7817	1.3826	0.0000
0.0222	0.7893	1.3416	-0.0407
0.1463	0.8316	1.2181	-0.1624
0.2695	0.8734	1.1390	-0.2398
0.3947	0.9155	1.0996	-0.2775
0.4999	0.9504	1.0848	-0.2909
0.6003	0.9834	1.1001	-0.2742
0.7253	1.0239	1.1297	-0.2429
0.8510	1.0635	1.2390	-0.1319
0.9758	1.1024	1.3351	-0.0341
1.0000	1.1096	1.3688	0.0000
γ–Butyrolactone (1) + 2-Butanol (2)			
T = 303.15 K			
0.0000	0.7988	2.5717	0.0000
0.0279	0.8067	2.3187	-0.2258
0.1719	0.8466	1.9003	-0.5038
0.3115	0.8872	1.6602	-0.6078
0.4437	0.9275	1.4892	-0.6499
0.5500	0.9615	1.3833	-0.6521
0.6486	0.9941	1.3408	-0.5985
0.7642	1.0340	1.3639	-0.4627
0.8748	1.0737	1.4210	-0.2978
0.9814	1.1135	1.5837	-0.0312
1.0000	1.1203	1.5967	0.0000
T = 308.15 K			
0.0000	0.7936	2.1270	0.0000
0.0279	0.8015	1.9447	-0.1639
0.1719	0.8417	1.5710	-0.4428
0.3115	0.8824	1.3771	-0.5448
0.4437	0.9228	1.2830	-0.5520
0.5500	0.9567	1.2115	-0.5535
0.6486	0.9891	1.2044	-0.4957
0.7642	1.0286	1.2309	-0.3931
0.8748	1.0681	1.2955	-0.2557
0.9814	1.1073	1.4422	-0.0389
1.0000	1.1141	1.4688	0.0000
T = 313.15 K			
0.0000	0.7899	1.8097	0.0000
0.0279	0.7981	1.6773	-0.1201
0.1719	0.8383	1.3696	-0.3643
0.3115	0.8791	1.2411	-0.4313
0.4437	0.9196	1.1781	-0.4360
0.5500	0.9535	1.1513	-0.4159
0.6486	0.9856	1.1500	-0.3737
0.7642	1.0247	1.1655	-0.3073
0.8748	1.0639	1.2175	-0.2065
0.9814	1.1029	1.3365	-0.0405
1.0000	1.1096	1.3688	0.0000

TABLE III

Computed binary coefficients A_i and corresponding standard deviation (σ) for deviation in viscosity.

T / K	A_0	A_1	A_2	A_3	A_4	σ
γ–Butyrolactone (1) + 1-Propanol (2)						
303.15	-1.3	-0.17	-0.296	0.074	0.7	0.01
308.15	-1.14	-0.33	-0.12	-0.23	-0.04	0.012
313.15	-0.92	0.04	-0.51	-0.303	1.02	0.007
γ–Butyrolactone (1) + 2-Propanol (2)						
303.15	-1.64	0.005	-0.19	0.4	-0.7	0.006
308.15	-1.33	0.06	-0.37	0.1	-0.26	0.008
313.15	-1.16	-0.04	-0.08	0.399	-0.09	0.009
γ–Butyrolactone (1) + 1-Butanol (2)						
303.15	-2.001	0.61	0.25	0.22	-1.31	0.003
308.15	-1.77	0.42	0.42	0.61	-1.23	0.008
313.15	-1.38	0.38	0.52	0.48	-0.99	0.01
γ–Butyrolactone (1) + 2-Butanol (2)						
303.15	-2.66	-0.18	0.45	2.48	-3.65	0.024
308.15	-2.25	0.195	-0.24	1.33	-2.001	0.014
313.15	-1.72	0.36	-0.52	0.64	-1.34	0.006

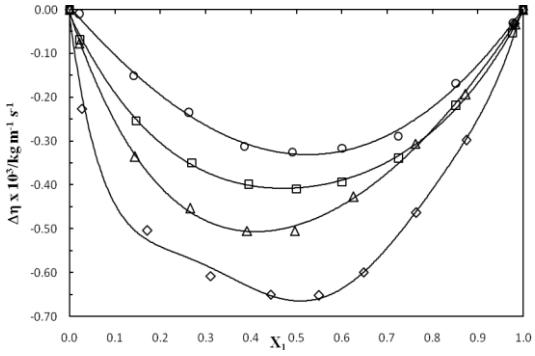

Figure 1: Variation of deviation in viscosity (Δη) vs mole fraction for the binary mixtures of γ–butyrolactone (1) + o; 1-propanol (2), □; 2-propanol (2), Δ; 1-butanol (2) and ◊; 2-butanol (2) at 303.15 K.

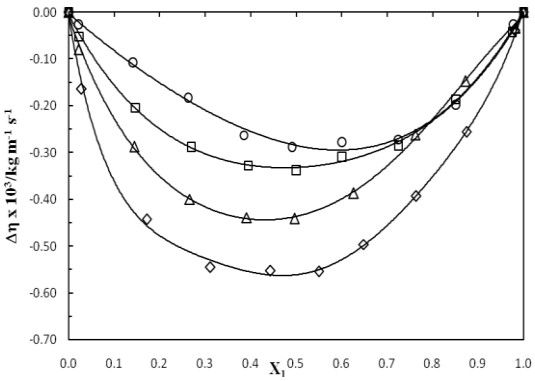

Figure 2: Variation of deviation in viscosity (Δη) vs mole fraction for the binary mixtures of γ–butyrolactone (1) + o; 1-propanol (2), □; 2-propanol (2), Δ; 1-butanol (2) and ◊; 2-butanol (2) at 308.15 K.

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 6 Issue 07, July - 2017

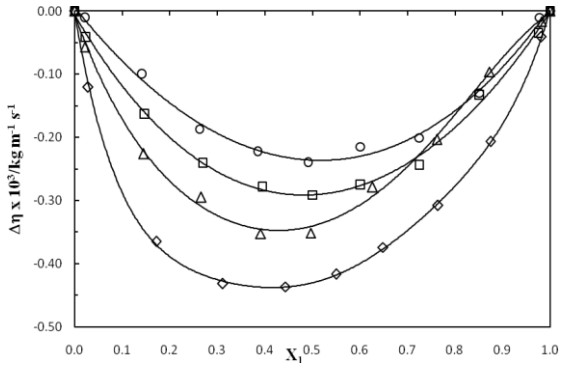

Figure 3: Variation of deviation in viscosity (Δη) vs mole fraction for the binary mixtures of γ–butyrolactone (1) + ο; 1-propanol (2), □; 2-propanol (2), Δ; 1-butanol (2) and ◊; 2-butanol (2) at 313.15 K.