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Volumetric and Excess Properties of Binary Liquid Mixtures of Butyl Acetate with Alkoxyethanols At 308.15k

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Abstract*: Experimental values of density(ρ), viscosity(η) and refractive index(n_D) for the binary mixtures of butyl acetate(BA) ($C_6H_{12}O_2$) with alkoxyethanolssuch as 2-methoxyethanol(2-ME) ($C_2H_8O_2$),2-ethoxyethanol(2-EE) ($C_4H_{10}O_2$) and 2-butoxyethanol(2-BE) ($C_6H_{14}O_2$) were measured at 308.15K. From the above experimental values ofρ, η and n_D,the following interaction parameters are calculated. They are excess molar volume(V^E), excess viscosity(η^E), deviation of refractive index(Δn_D), excess molar refraction(R_m^E) and excess Gibbs free energy(ΔG^{*E}). The above results are fitted with Redlich-Kister polynomial equation. The values of all the calculated parameters of the binary mixtures and their deviations indicates weaker forces between the ester group (RCOOR) of Butyl acetate with hydroxyl group (-OH) of alkoxyethanols.

Keywords: Butyl acetate, alkoxyethanols, Excess properties, Redlich–Kister polynomial equation, Molecular interaction.

I. INTRODUCTION

Esters are used as plasticizing agents during polymer processing toimpart favorable thermo elastic properties [1]. The ester functional group is an excellent agent for probing bulk molecular interactions in binary mixtures consisting esters as one of the components along with either non-polar or polar solvents. The availability of free electrons in oxygen, the well known tendency of carbonyl group to act as proton acceptor (in hydrogen bonding), and the sufficient polarity within the functional group for the stable coexistence of the dipoles, could easilybe imagined as responsible factors that make interesting interactions at bulk level in the esters containing binary solutions [2].The butyl acetate (BA)is polar in nature and finds wide industrial applications [3].The Alkoxyethanols like 2-methoxyethanol(2-ME), 2-ethoxyethanol(2-EE) and 2-butoxyethanol(2-BE) are commonly used as a solvent in binary liquid mixtures. 2-methoxyethanol, commonly called as methyl cello solve is a widely used industrial solvent in view of its solvating properties and quasi-aprotic character. It is an ether alcohol showing physicochemical characteristics that are intermediate between protic and dipolar aprotic solvents [4].2-ethoxyethanol is widely used as complexes of solvents, coemulsifiers, dyes and lacquers [5]. 2-ethoxyethanol is a common solvent in laboratory practice and in the chemical industry and is able to form two types of hydrogen bonds, inter- and intra-molecular ones. This capacity keeps attracting the interest of many researchers for the theoretical and experimental study of various properties of systems of cellosolve [6]. The main use of 2–butoxyethanol is a solvent and it is used in paints and surface coatings, followed by cleaning products and inks. These compounds with a carbonyl group (C=O) are strongly hydrogen–bond acceptors [7].From the experimental data of densities, viscosities, refractive indices of the butyl acetate with alkoxyethanols like 2-methoxyethanol($C_2H_8O_2$), 2-ethoxyethanol($C_4H_{10}O_2$) and 2-butoxyethanol($C_6H_{14}O_2$) system, the following parameterssuch as molar volume(V^E), excess viscosity(η^E), deviation of refractive index(Δn_D), excess molar refraction(R_m^E) and excess Gibbs free energy(ΔG^{*E}) were calculated.The calculated values of above excess parameters values are fitted with Redlich – Kister polynomial equation. The values of a_i of polynomial equation were calculated by the least square fit method. The values of these parameters of each studied system are used to identify the standard deviation of the experimental and theoretical values. The results were analyzed in terms of the molecular interactions through hydrogen bonding between the hydrogen atom of (-OH) group of alkoxyethanol and oxygen atom of the (C=O) group of ester (i.e) butyl acetate in the liquid mixtures.

II. MATERIALS AND METHODS

A. Materials

Butyl acetate (99.5% Purity), 2-methoxyethanol, 2-ethoxyethanol and 2-butoxyethanol were provided by Loba products, AR-grade (99% Purity), which are utilized without further purification. The purity of chemicals were tested with their literature values such as refractive indices and densities.

B. Density Measurement

The solutions were taken in airtight closed glass bottles. The weightings were done by using the digital electronic balance (Anamed). Its accuracy is ±0.0001g. Weightings were done at least five times for accuracy of the measurements. The density of pure liquids was utilized by double armed Pycnometer (5ml). Pycnometer was calibrated with freshly prepared double distilled water.

C. Viscosity Measurement

The Viscosities of liquids were employing by Ostwald's Viscometer and calibrated with double distilled water. The rate of flow of liquid in the Viscometer was measured not less than five times for every solution for the better accuracy in measurements. The fluid flow was measured by the stopwatch (Edutek — 19671697), a correctness of accuracy is ±0.01s. The Viscometer was kept in the thermostat for maintaining the fixed temperature 308.15K. The heat was managed to pass around water bath, (supplied by M/s Sakti Scientific Instruments Company, India) and its accuracy is ±0.01K.

D. Refractive Index Measurement

The refractive index of liquids was taken by the Abbe's refracto meter. It is calibrated by distilled water and ethanol, the uncertainty of refractive index is ± 0.0001.

III. CALCULATIONS

Following are the excess or deviation of the parameters [8].

Excess molar volume $$V^E = \sum_{i=1}^{N} x_i M_i \left(\rho^{-1} - \rho_i^{-1}\right) \quad (1)$$

Excess viscosity $$\eta^E = \eta - \sum_{i=1}^{N} x_i \eta_i \quad (2)$$

Deviation of refractive index $$\Delta n_D = n_D - \sum_{i=1}^{N} x_i n_{Di} \quad (3)$$

Excess molar refraction $$R^E = R_m - \sum_{i=1}^{N} \phi_i R_{mi} \quad (4)$$

Excess Gibbs free energy $$\Delta G^{*E} = RT\left[\ln(V\eta) - \sum_{i=1}^{N} x_i \ln(V_i \eta)\right] \quad (5)$$

Where x_i is a symbol of the mole fraction and φ_i is the volume fraction of the pure factor i, correspondingly. ρ, η, n_D, V, and R_m are the density, viscosity, refractive index, molar volume, and molar refraction of the mixtures respectively, and ρ_i, V_i, η_i, n_{Di} and R_i the representational properties of the pure liquids.

$$A^E = x_1(1-x_1)\sum_{i=1}^{N} a_i (2x_2 - 1)^i \quad (6)$$

The Equation(6), excess functions were fitted to Redlich–Kister type polynomial equation and values of coefficient a_i were computed by the method of least squares fit and recognize standard deviation of data based experimental and hypothetical values. The standard deviation values are summarized by the following relation.

$$\sigma = \left[\frac{\sum (X_{\exp} - X_{cal})^2}{n-p}\right]^{1/2} \quad (7)$$

Here 'n' is the number of experimental data points and 'p' refer to the number of coefficients, X_{exp} indicate experimental properties, X_{cal}indicate calculated properties.

IV. RESULT AND DISCUSSIONS

Table (1) reveals the experimental and the literature values of butyl acetate and alkoxyethanols at the temperature 308.15K [9-17]. Table (2) Shows, the variations of mole fractions of Butyl acetate with increasing the values of viscosities, densities and refractive indices.

Table (1) Experimental and literature values of pure Liquids at 308.15K

Table.2 Experimental values of density (ρ), viscosity (η), refractive index (n_D), molar refraction (R_m) and polarizability (α) of Butyl acetate + alkoxyethanols binary mixtures at 308.15 K

Mole fraction of Butyl acetate (X_2)	Density (ρ) g cm^{-3}	Viscosity (η) mPa s	Refractive index (n_D)	Molar refraction(R_m) cm^3 mol^{-1}	Polarizability ($\alpha x10^{-26}$) cm mol^{-1}
Butyl acetate + 2-Methoxyethanol					
0.0000	0.8654	0.5842	1.3880	31.6741	1.2561
0.1617	0.8508	0.6094	1.3905	30.5932	1.2132
0.2996	0.8429	0.6334	1.3925	29.4570	1.1682
0.4193	0.8406	0.6711	1.3940	28.2754	1.1213
0.5284	0.8420	0.7266	1.3950	27.0483	1.0727
0.6235	0.8490	0.7975	1.3955	25.7757	1.0222
0.7130	0.8593	0.8777	1.3960	24.4946	0.9714
0.7935	0.8746	0.9704	1.3960	23.1786	0.9192
0.8669	0.8951	1.0667	1.3960	21.8593	0.8669
0.9347	0.9207	1.1621	1.3960	20.5428	0.8147
1.0000	0.9510	1.2562	1.3960	19.2275	0.7625
Butyl acetate + 2-Ethoxyethanol					
0.0000	0.8654	0.5842	1.3880	31.6741	1.2561
0.1373	0.8697	0.6944	1.3915	30.7907	1.2211
0.2600	0.8738	0.7900	1.3950	30.0111	1.1901
0.3749	0.8782	0.8867	1.3980	29.2415	1.1596
0.4793	0.8827	0.9794	1.4000	28.4742	1.1292
0.5779	0.8876	1.0682	1.4010	27.6764	1.0976
0.6732	0.8929	1.1585	1.4015	26.8660	1.0654
0.7626	0.8984	1.2448	1.4020	26.0996	1.0350
0.8438	0.9040	1.3267	1.4020	25.3711	1.0061
0.9230	0.9099	1.4101	1.4020	24.6545	0.9777
1.0000	0.9160	1.4870	1.4020	23.9571	0.9501
Butyl acetate + 2-Butoxyethanol					
0.0000	0.8654	0.5842	1.3880	31.6741	1.2561

Liquids	Experimental			Literature		
	ρ g. cm^{-3}	η mPa.s	n_D	ρ g. cm^{-3}	η mPa.s	n_D
Butyl acetate	0.8654	0.5842	1.3880	0.8654 [9]	0.593 [9] 0.584 [10]	1.3882 [9] 1.3882 [10]
2-Methoxyethanol	0.9510	1.2562	1.3960	0.9511 [11, 12]	1.2491[11]	1.3949 [13]
2-Ethoxyethanol	0.9160	1.4870	1.4020	0.9163 [14]	1.480 [15]	1.4018 [14]
2-Butoxyethanol	0.8870	2.2871	1.4130	0.8889 [16]	2.2880[17]	1.4132 [16]

0.1025	0.8673	0.7129	1.3910	31.8758	1.2641
0.2007	0.8692	0.8165	1.3940	32.0781	1.2721

0.3001	0.8711	0.9358	1.3970	32.2804	1.2801
0.4005	0.8730	1.0763	1.4000	32.4806	1.2881
0.4984	0.8750	1.2328	1.4025	32.6415	1.2945
0.5998	0.8772	1.4162	1.4050	32.7944	1.3005
0.6985	0.8796	1.6110	1.4070	32.9052	1.3049
0.7977	0.8820	1.8175	1.4090	33.0116	1.3091
0.8970	0.8845	2.0459	1.4110	33.1162	1.3133
1.0000	0.8870	2.2871	1.4130	33.2220	1.3175

Table.3 Excess molar volume (V^E), Excess viscosity (η^E), deviation of refractive index (Δn_D), excess molar refraction ($R_m{}^E$) and excess Gibbs free energy of activation for viscous flow (ΔG^{*E}) for Butyl acetate +alkoxyethanols binary mixtures at 308.15 K

Molefraction of Butyl acetate (X_2)	Excess molar volume (V^E) $cm^3\ mol^{-1}$	Excess viscosity (η^E) mPa s	Deviation of refractive index (Δn_D)	Excess molar refraction ($R_m{}^E$) $cm^3\ mol^{-1}$	Excess Gibbs free energy (ΔG^{*E}) J mol^{-1}
Butyl acetate + 2-Methoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1617	0.1029	-0.0421	0.0012	0.1637	-95.9291
0.2996	0.1749	-0.0852	0.0021	0.2722	-190.4452
0.4193	0.2390	-0.1147	0.0026	0.3353	-230.8807
0.5284	0.2762	-0.1264	0.0028	0.3528	-216.6132
0.6235	0.2685	-0.1227	0.0025	0.3249	-166.2282
0.7130	0.2375	-0.1096	0.0023	0.2884	-112.4885
0.7935	0.1813	-0.0841	0.0017	0.2171	-49.6662
0.8669	0.1117	-0.0551	0.0011	0.1425	-5.3362
0.9347	0.0532	-0.0269	0.0005	0.0706	11.2703
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Butyl acetate + 2-Ethoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1373	0.1420	-0.0138	0.0016	0.1763	126.7661
0.2600	0.2711	-0.0290	0.0034	0.3436	174.1462
0.3749	0.3650	-0.0360	0.0048	0.4607	201.9881
0.4793	0.4057	-0.0375	0.0053	0.4990	210.1476
0.5779	0.3957	-0.0377	0.0049	0.4618	197.0299
0.6732	0.3544	-0.0334	0.0041	0.3869	174.2681
0.7626	0.2900	-0.0279	0.0033	0.3104	139.4320
0.8438	0.2076	-0.0192	0.0022	0.2084	101.5608
0.9230	0.1035	-0.0073	0.0011	0.1030	59.0341
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Butyl acetate + 2-Butoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1025	0.0365	-0.0459	0.0004	0.0430	148.1298
0.2007	0.0743	-0.1096	0.0010	0.0933	152.8911
0.3001	0.1161	-0.1594	0.0015	0.1418	155.8201
0.4005	0.1527	-0.1900	0.0020	0.1865	163.3621

0.4984	0.1751	-0.2002	0.0020	0.1959	169.2630
0.5998	0.1704	-0.1895	0.0020	0.1918	169.8744
0.6985	0.1393	-0.1627	0.0015	0.1499	154.8655
0.7977	0.0928	-0.1252	0.0011	0.1027	116.9885
0.8970	0.0412	-0.0657	0.0006	0.0538	73.5651
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

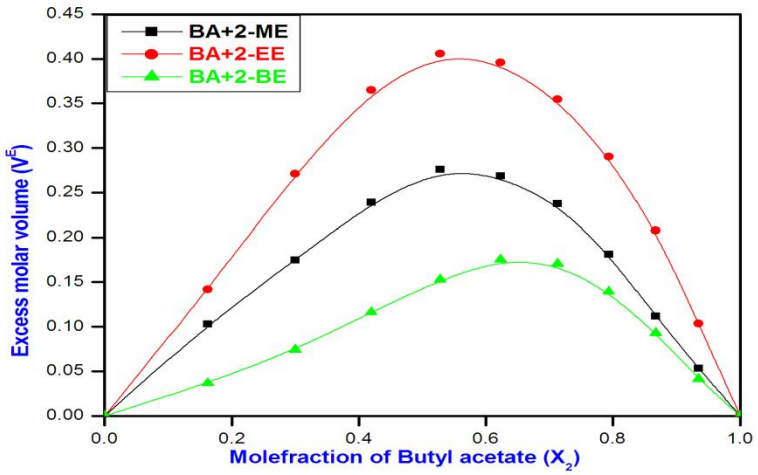

Fig.1. Excess molar volume(V^E) vs Mole fraction(X_2) of the Butyl acetate with alkoxyethanols.

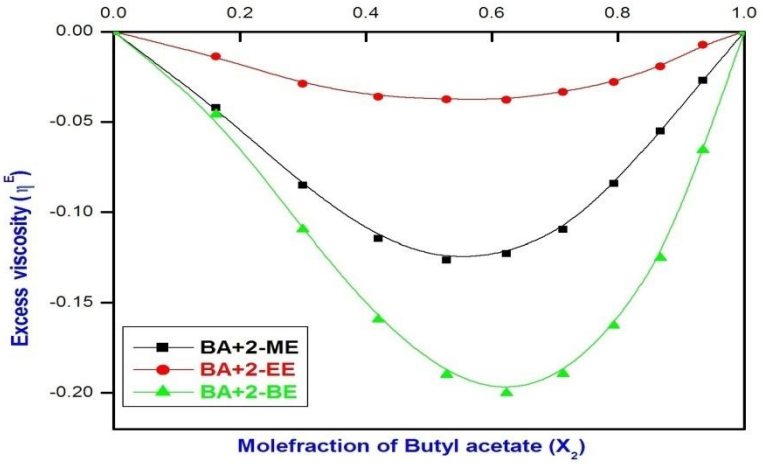

Fig.2.Excess viscosity (η^E) vs Mole fraction (X_2) of the Butyl acetatewith alkoxyethanols.

The refractive index is measure in optical region of the liquids [8]. Table (3) contains excess and deviation of parameters such as V^E, η^E, Δn_D, $R_m{}^E$, and ΔG^{*E} of Butyl acetate and alkoxyethanols at the temperature 308.15K. Further the curve in Fig.(1) shows that the excess molar volume(V^E) observed is positive. It indicate breakdown of one or both components in solution system. The breaking up (or) rupture of H-bonding of components held together by weaker physical forces such as dipole-dipole (or) dipole induced dipole interactions or by other Vander Waals forces. Among the weaker physical forces of BA+2-ME, BA+2-EE and BA+2-BE, The BA+2-BE is observed somewhat stronger when compare to the other two systems taken for the study.The excess molar volume (V^E) has high positive peak which are located between 0.4 and 0.8 mole fractions of Butyl acetate. The highest positive peak is observed in the systemofBA+2-EE and the lowest positive peak is observed in the BA+2-EE system.

Fig. (2) shows the variation of excess viscosity (η^E) with mole fraction of Butyl acetate. These excess viscosity values are all negative values for all the systems studied. The highest negative values obtain for BA+2-BE system and the lowest negative values

are obtained for BA+2-EE. The peak negative values are located between 0.4 and 0.8 mole fractions of all the systems studied. The excess viscosity (η^E) values are found to be negative which indicates that dispersion forces are dominant.

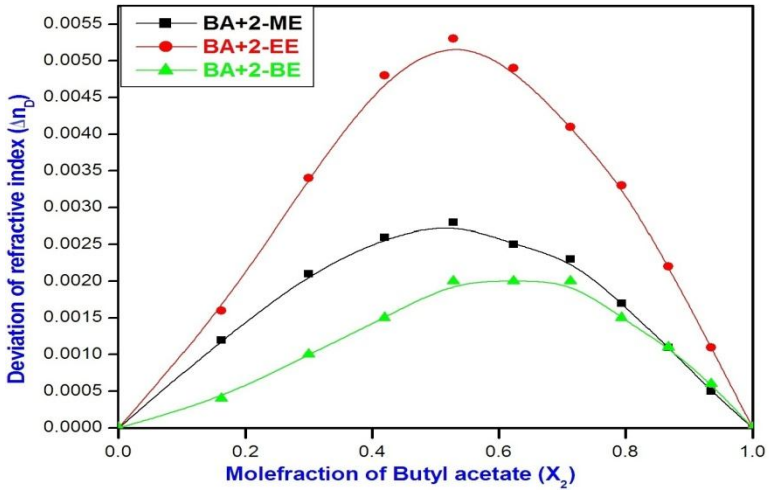

Fig.3. Deviation of refractive index(Δn_D) vs mole fraction(X_2) of the Butyl acetate with alkoxyethanols.

Fig. (3) shows deviations of refractive index(Δn_D) values with mole fraction of Butyl acetate which are positive for all the system taken for the study. Thepositive deviation is may be due to some forces between the participated molecules, such as hydrogen bonding, dipole-dipole or dipole induced dipole or Vander Waals forces between the constituent molecules.

Table (3)also shows the variation of the excess molar refraction (R_m^E) with mole fraction of butyl acetate. The magnitude and sign changein excess molar refraction indicates the size difference between the identical molecules. The size difference between the constituent molecules is the main contributing factor for volume additive of one into another. When two unlike molecules of unequal molar volumes are usually mixed, there is a possibility of accommodation of interstices position of one molecule into another. The positive values of (R_m^E) indicating the presence of weak intermolecular interaction.

The Excess Gibbs free energy (ΔG^{*E}) represent the interaction behavior of liquid mixtures through the sign of its values. The positive values are indicates the strong interaction between the molecules and negative values indicates the dispersion force between the molecules [8]. The Excess Gibbs free energy (ΔG^{*E}) values are positive for the systemsof BA+2-EE and BA+2-BE and negative values forBA+2-ME system. From Table (4), we can views the values of adjustable parameters and standard deviations by using the Redlich–Kister polynomial equations for the excess properties.

The values of standard deviations are giving the acceptable outcome for the experimental readings. The experimental refractive index values are calculated by seven theoretical mixing rules [18].

$$Lorentz - Lorentz : \frac{n_D^2 - 1}{n_D^2 + 2} = \left(\frac{n_{D1}^2 - 1}{n_{D1}^2 + 2}\right)\phi_1 + \left(\frac{n_{D2}^2 - 1}{n_{D2}^2 + 2}\right)\phi_2 \quad (8)$$

$$Arago - Biot : n_{D1}\phi_1 + n_{D2}\phi_2 \quad (9)$$

$$Newton : n_D^2 - 1 = (n_{D1}^2 - 1)\phi_1 + (n_{D2}^2 - 1)\phi_2 \quad (10)$$

$$Gladstone - Dale : n_D - 1 = (n_{D1} - 1)\phi_1 + (n_{D2} - 1)\phi_2 \quad (11)$$

$$Heller : \frac{n_D - n_{D1}}{n_{D1}} = \frac{3}{2}\left[\frac{(n_{D2}/n_{D1})^2 - 1}{(n_{D2}/n_{D1})^2 + 2}\right]\phi_2 \quad (12)$$

$$Weiner : \frac{n_D^2 - n_{D1}^2}{n_D^2 + 2n_{D2}^2} = \left[\frac{n_{D2}^2 - n_{D1}^2}{n_{D2}^2 + 2n_{D1}^2}\right]\phi_2 \quad (13)$$

$$Oster: \frac{(n_D^2-1)(2n_D^2+1)}{n_D^2} = \frac{(n_{D1}^2-1)(2n_{D1}^2+1)}{n_{D1}^2}\phi_1 + \frac{(n_{D2}^2-1)(2n_{D2}^2+1)}{n_{D2}^2}\phi_2 \quad (14)$$

The standard deviation values are reported in Table (5). Theoretical mixing rules of Weiner have the very low deviation when compared to the other mixing rules. The Gladstone–Dale(G–D) and Arago–Biot(A-B) relation gives identical values. From the values of standard deviation, the deviations are very low when compared to the theoretical values. The seven different mixing rules are applied successfully and the results are interpreted on the basis of nature of molecular interactions.

Table 5: Values of standard deviation for refractive index by different theoretical mixing rules for Butyl acetate + alkoxyethanols binary mixtures at 308.15 K

Standard deviation (σ)							
Systems	L–L	Wiener	G–D	A–B	Heller	Newton	Oster
Butyl acetate + 2-Methoxyethanol	0.0014	0.0012	0.0026	0.0026	0.0019	0.0074	0.0167
Butyl acetate + 2-Ethoxyethanol	0.0023	0.0020	0.0043	0.0043	0.0031	0.0119	0.0270
Butyl acetate + 2-Butoxyethanol	0.0008	0.0006	0.0014	0.0014	0.0011	0.0039	0.0089
L–L: Lorentz–Lorentz; G–D: Gladstone–Dale; A–B: Arago–Biot							

Table 4: Values of adjustable parameters (B_k) and the corresponding standard deviations (σ), for excess molar volumes, deviation of viscosity, deviation of refractive index, excess molar refraction and excess Gibbs free energy for Butyl acetate + alkoxyethanols binary mixtures at 308.15 K

Parameters	(B_k)							(σ)
	B_0	B_1	B_2	B_3	B_4	B_5	B_6	
Butyl acetate + 2-Methoxyethanol								
V^E (cm^3 mol^{-1})	1.1020	-0.4770	-1.3460	0.5820	3.6690	-0.1150	-3.4160	2.4193
η^E (mPa s)	-0.5150	0.1180	0.6210	0.2430	-1.5770	-0.3550	1.4660	0.1254
R_m^E (cm^3 mol^{-1})	1.4430	-0.0630	-1.5380	-0.1960	4.7550	0.2440	-4.6460	1.3006
Δn_D	0.2370	-0.1760	-0.1980	-0.4720	0.9620	0.6430	-0.9970	0.0078
ΔG^{*E} (J mol^{-1})	-874.4000	-371.6000	426.5000	-527.1000	1990.0000	892.5000	-1537.0000	321.5316
Butyl acetate + 2-Ethoxyethanol								
V^E (cm^3 mol^{-1})	1.6580	0.0170	-1.6160	-1.2320	4.7690	1.2050	-4.8040	1.7803
η^E (mPa s)	-0.1510	-0.0090	-0.0290	0.0870	0.0970	-0.0770	0.0830	0.2990
R_m^E (cm^3 mol^{-1})	2.0040	0.3960	-2.1370	-1.5730	5.1430	1.1690	-5.0050	0.9467
Δn_D	0.0210	0.0030	-0.0250	-0.0200	0.0530	0.0170	-0.0480	0.0057
ΔG^{*E} (J mol^{-1})	862.6000	114.6000	-1011.0000	291.9000	4662.0000	-413.1000	-4507.0000	30.3671
Butyl acetate + 2-Butoxyethanol								
V^E (cm^3 mol^{-1})	0.7050	-0.1960	-0.8640	0.3660	1.4860	-0.1690	-1.3260	1.9866
η^E (mPa s)	-0.8020	-0.0850	0.3600	0.7440	-0.9530	-0.6570	1.3950	0.1530
R_m^E (cm^3 mol^{-1})	0.8110	-0.0080	-1.0670	-0.2420	2.1600	0.2490	-1.9030	0.6153
Δn_D	0.0080	0.0000	-0.0110	-0.0040	0.0230	0.0040	-0.0200	0.0047
ΔG^{*E} (J mol^{-1})	733.1000	-449.2000	-1458.0000	3072.0000	8026.0000	-2613.0000	-7297.0000	32.0171

V. CONCLUSION

Physical parameters such as viscosity (η), density (ρ) and the refractive index (n_D) were calculated forButyl acetate with alkoxyethanols solutions. The excess properties such as excess molar volume(V^E), excess viscosity(η^E), deviations of refractive index(Δn_D), excess molar refraction(R_m^E) and excess Gibbs free energy(ΔG^{*E}) values were calculated and fitted with Redlich–Kister polynomial equations. The order of molecular interactions concluded from the results of V^E, $\eta^E$are BA+2-BE > BA+2-ME > BA+2-EE.

VI. ACKNOWLEDGEMENT

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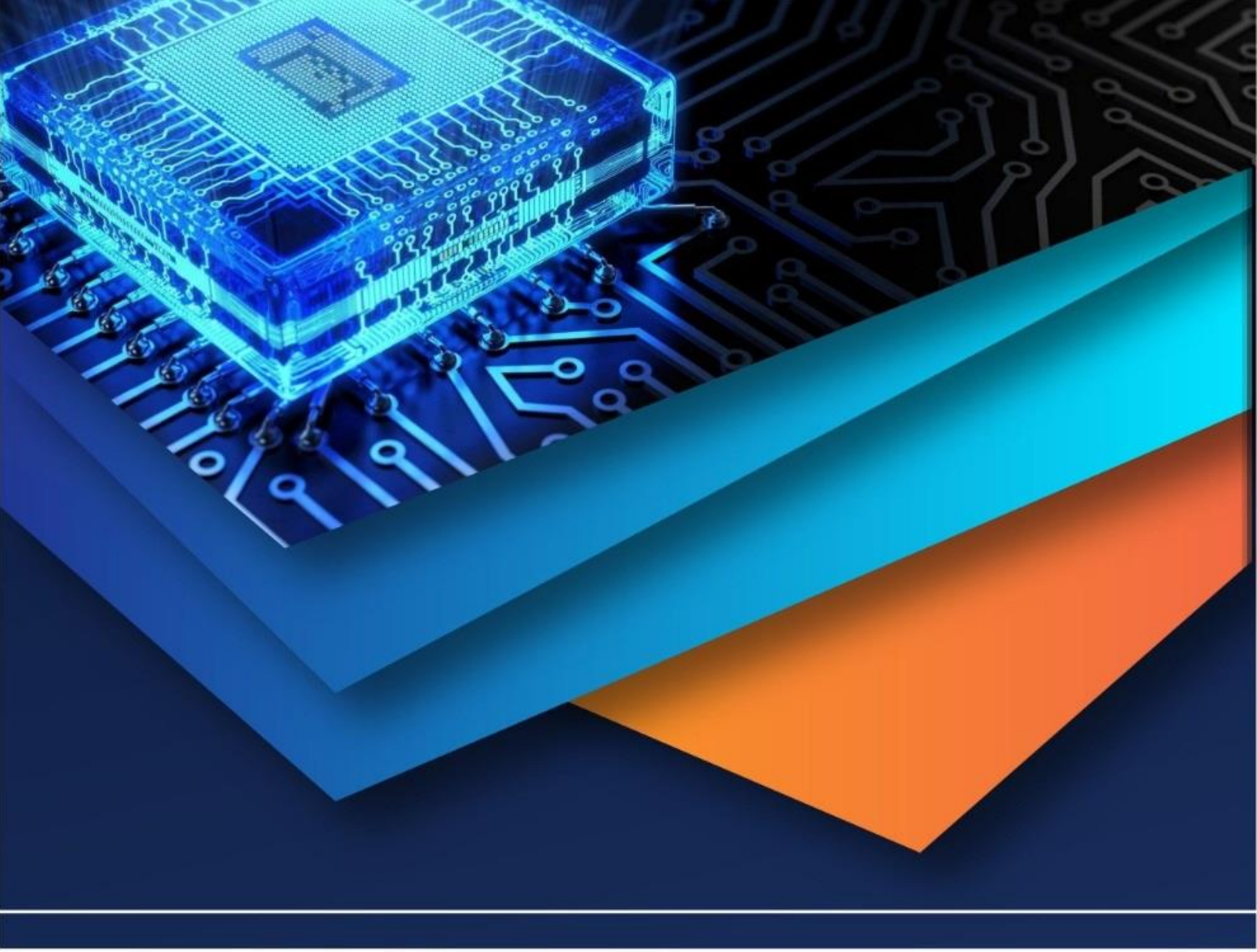

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