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Densities, Viscosities, Refractive Indices and Excess properties of Binary liquid mixtures of Ethyl acetate with Alkoxyethanols at 308.15K

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Abstract
Densities(ρ), viscosities(η), refractive indices(n_D) and excess properties such as excess molar volume(V^E), excess viscosity(η^E), deviation of refractive index(Δn_D), excess molar refraction(R_m^E), excess Gibbs free energy(ΔG^{*E}) were calculated for the binary mixtures of ethyl acetate(EA) with alkoxyethanols like 2-methoxyethanol(2-ME), 2-ethoxyethanol(2-EE) and 2-butoxyethanol(2-BE) at 308.15K. Theoretical mixing rules for refractive index were calculated for the above said binary liquid systems. The values were fitted with Redlich – Kister Polynomial equation. The results were discussed on the basis of nature of molecular interactions.

Keywords: ethyl acetate, alkoxyethanols, density, viscosity, refractive index, excess parameters

1. Introduction
Knowledge about the density and viscosity of binary liquid mixtures are important for understanding the behavior of liquids. Volumetric properties of liquids are powerful tool in understanding the nature and physico-chemical behavior of molecular systems [1]. Esters play a vital role in medicinal, industrial, chemical and biological areas. Due to their biodegradability, they are important constituent of marine engine oils, drugs, inks, cosmetics, hydraulic fluids etc. Moreover esters are widely useful in food industry for their aroma and flavors. They also impart desired thermo plasticity to polymers. Many researchers have described the role of thermo dynamic excess properties with the pattern of molecular interaction between molecules of esters with n-alkanols [2-10].

Alkoxyethanols are used as a solvent for making varnishes, dyes, and resins. Hence the binary mixtures of ethyl acetate (EA) with 2-methoxyethanol (2-ME), 2-ethoxyethanol (2-EE) and 2-butoxyethanol (2-BE) were taken for the study to characterize the physicochemical behavior of these liquid mixtures at 308.15K.

2. Materials and methods
2.1 Materials
Ethyl acetate (Sigma-Aldrich product, AR-grade), 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 of refractive indices and densities.

2.2 Density Measurement
The solutions were taken in airtight closed glass bottles. The weightings were done by using the digital electronic balance (Anamed). It 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.

2.3 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.

2.4 Refractive Index Measurement
The refractive indices of liquids were taken by the Abbe's refracto meter. It is calibrated by distilled water and ethanol, the uncertainty of refractive index is ± 0.0001.

2.5 Calculations
Following are the excess or deviation of the parameters [11].
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

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

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 theoretical 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 points and 'p' refer to the number of parameters, X_{exp} indicate experimental properties and X_{cal} point out calculated properties.

3. Result and Discussions

Table-1 shows the literature and experimental values of ethyl acetate and alkoxyethanols at the temperature 308.15K [12-20]. Table-2 Shows, The variations of mole fractions of ethyl acetate with increasing the values of viscosities, densities and refractive indices.

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

Liquids	Experimental			Literature		
	ρ g. cm^{-3}	η mPa.s	n_D	ρ g. cm^{-3}	η mPa.s	n_D
Ethyl acetate	0.8882	0.3911	1.3695	0.8827 [12] 0.8811 [13]	0.387 [12,13]	1.3654 [12] 1.3647 [13]
2-Methoxyethanol	0.9510	1.2562	1.3960	0.9511 [14, 15]	1.2491 [14]	1.3949 [16]
2-Ethoxyethanol	0.9160	1.4870	1.4020	0.9163 [17]	1.480 [18]	1.4018 [17]
2-Butoxyethanol	0.8870	2.2871	1.4130	0.8889 [19]	2.2880 [20]	1.4132 [19]

The refractive index is measured in optical region and it must not contribute polarizability of the liquids in orientation effects [11]. Table-3 contains excess and deviation parameters such as V^E, η^E, Δn_D, $R_m{}^E$, and ΔG^{*E} of ethyl acetate and alkoxyethanols at the temperature 308.15K. The Excess molar volume (V^E) values are positive and excess viscosity (η^E) values are negative in all the systems studied. Further the curves in Fig.1 shows that the excess molar volume (V^E) has more positive values comparatively as the chain length of alkoxyethanols increases.

Fig.1 shows the excess molar volumes (V^E) has high positive peak values for all the systems studied and are located between 0.4 and 0.8 mole fractions of ethyl acetate. The highest positive peak is observed in the system EA+2-BE and lowest positive peak is observed in the EA+2– ME system.

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

Mole fraction of Ethyl 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}
Ethyl acetate + 2-Methoxyethanol					
0.0000	0.8882	0.3911	1.3695	22.4104	0.8887
0.1247	0.8902	0.4547	1.3740	22.2199	0.8812
0.2395	0.8933	0.5139	1.3785	22.0229	0.8734
0.3497	0.8973	0.5847	1.3825	21.7890	0.8641
0.4528	0.9024	0.6622	1.3860	21.5190	0.8534
0.5539	0.9081	0.7458	1.3890	21.2156	0.8413
0.6495	0.9148	0.8378	1.3910	20.8572	0.8271
0.7417	0.9225	0.9369	1.3925	20.4689	0.8117
0.8312	0.9309	1.0493	1.3935	20.0524	0.7952
0.9220	0.9398	1.1586	1.3945	19.6305	0.7785
1.0000	0.9510	1.2562	1.3960	19.2275	0.7625
Ethyl acetate + 2-Ethoxyethanol					
0.0000	0.8882	0.3911	1.3695	22.4104	0.8887
0.1049	0.8906	0.4758	1.3760	22.7563	0.9024

0.2079	0.8929	0.5609	1.3810	23.0189	0.9129
0.3079	0.8953	0.6517	1.3855	23.2520	0.9221
0.4067	0.8977	0.7483	1.3895	23.4568	0.9302
0.5056	0.9000	0.8507	1.3930	23.6343	0.9373
0.6067	0.9028	0.9646	1.3960	23.7755	0.9429
0.7036	0.9055	1.0845	1.3980	23.8610	0.9463
0.8026	0.9089	1.2165	1.3995	23.9054	0.9480
0.8946	0.9122	1.3442	1.4005	23.9196	0.9486
1.0000	0.9160	1.4870	1.4020	23.9571	0.9501
Ethyl acetate + 2-Butoxyethanol					
0.0000	0.8882	0.3911	1.3695	22.4104	0.8887
0.1512	0.8870	0.6244	1.3815	24.2813	0.9629
0.1574	0.8868	0.6352	1.3820	24.3635	0.9662
0.2427	0.8862	0.7752	1.3875	25.3740	1.0063
0.3304	0.8857	0.9255	1.3925	26.3915	1.0466
0.4245	0.8853	1.0914	1.3970	27.4410	1.0882
0.5726	0.8851	1.3717	1.4030	29.0413	1.1517
0.6339	0.8851	1.4912	1.4050	29.6783	1.1770
0.7374	0.8853	1.7046	1.4080	30.7327	1.2188
0.8682	0.8860	1.9870	1.4110	32.0069	1.2693
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 ethyl acetate + alkoxyethanols binary mixtures at 308.15 K

Molefraction of Ethyl 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}**
Ethyl acetate + 2-Methoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1247	0.0161	-0.0229	0.0012	0.1278	31.3144
0.2395	0.0471	-0.0501	0.0027	0.2491	15.5644
0.3497	0.0719	-0.0659	0.0037	0.3335	25.6484
0.4528	0.0854	-0.0749	0.0045	0.3818	40.0393
0.5539	0.0923	-0.0778	0.0048	0.3966	43.9645
0.6495	0.0946	-0.0723	0.0043	0.3566	53.1339
0.7417	0.0858	-0.0598	0.0033	0.2865	57.6692
0.8312	0.0673	-0.0338	0.0020	0.1884	70.7314
0.9220	0.0346	-0.0111	0.0006	0.0847	43.7275
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Ethyl acetate + 2-Ethoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1049	0.0588	-0.0303	0.0031	0.1837	140.8737
0.2079	0.1119	-0.0580	0.0047	0.2869	209.7489
0.3079	0.1560	-0.0768	0.0060	0.3653	252.0475
0.4067	0.1965	-0.0885	0.0068	0.4174	268.6744
0.5056	0.2376	-0.0945	0.0071	0.4419	260.0609
0.6067	0.2425	-0.0914	0.0068	0.4268	236.7337
0.7036	0.2350	-0.0776	0.0056	0.3624	206.1325
0.8026	0.1704	-0.0541	0.0039	0.2536	161.1764
0.8946	0.0857	-0.0273	0.0019	0.1255	101.3714
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Ethyl acetate + 2-Butoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1512	0.1121	-0.0581	0.0054	0.2359	499.4828
0.1574	0.1345	-0.0647	0.0057	0.2517	493.7776
0.2427	0.1956	-0.0916	0.0074	0.3397	612.2855
0.3304	0.2604	-0.1181	0.0086	0.4088	652.5715
0.4245	0.3035	-0.1306	0.0090	0.4408	663.0484
0.5726	0.3205	-0.1310	0.0086	0.4399	592.6645
0.6339	0.3170	-0.1225	0.0079	0.4149	542.1272
0.7374	0.2835	-0.1001	0.0064	0.3503	426.6010
0.8682	0.1604	-0.0502	0.0037	0.2095	244.2978
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

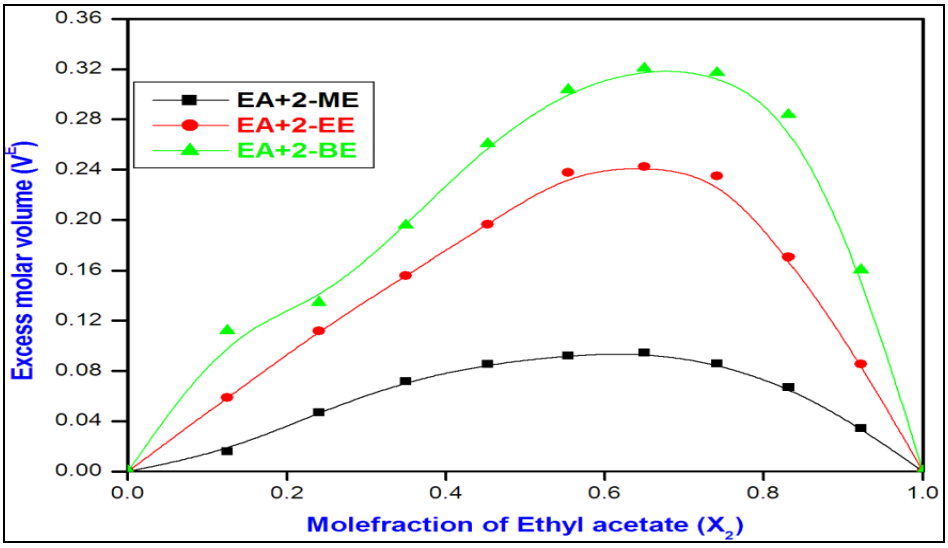

Fig 1: Excess molar volume (V^E) vs Mole fraction (X_2) of the ethyl acetate with alkoxyethanols

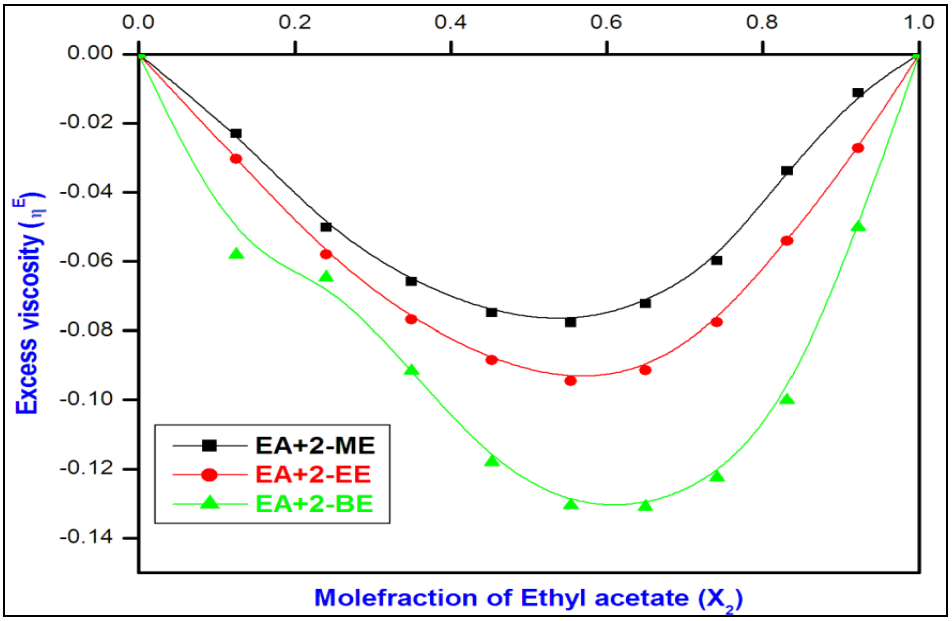

Fig 2: Excess viscosity (η^E) vs Mole fraction (X_2) of the ethyl acetate with alkoxyethanols

Fig.2 shows the variation of excess viscosity (η^E) with the mole fraction of ethyl acetate (EA). As the chain length of alkoxyethanols increases (η^E) values gets increases in the negative region. The excess viscosity (η^E) values are negative; and are located between 0.4 to 0.8 mole fractions of Ethyl acetate. The excess viscosity (η^E) values are found to be opposite to the sign of excess molar volume (V^E) for the three binary mixtures. A correlation between the (η^E) and (V^E) has been observed for a number of binary systems. The excess viscosity (η^E) is highest negative in EA+2-BE system and lower negative in EA+2-ME system.

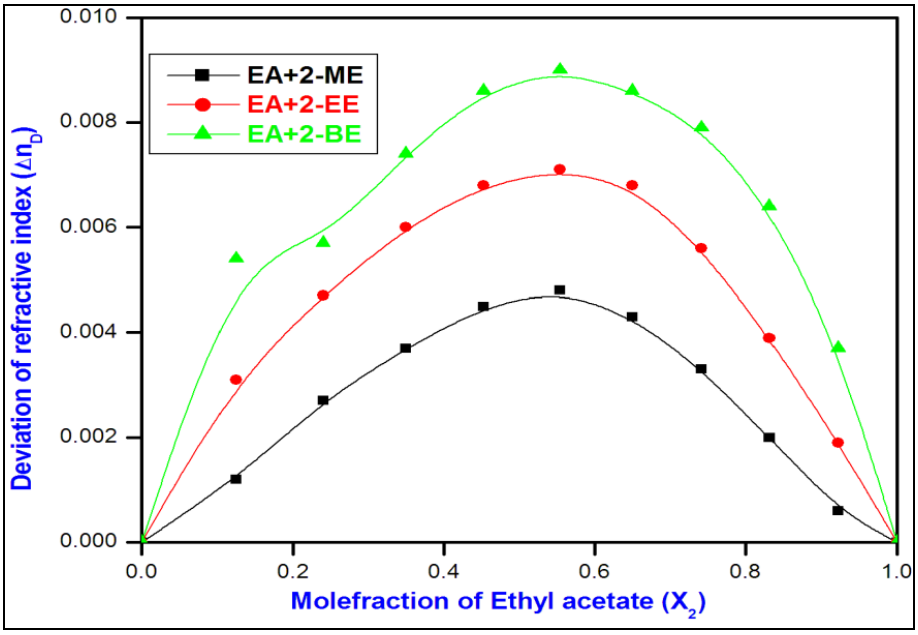

Fig 3: Deviation of refractive index (Δn_D) vs mole fraction(X_2) of the ethyl acetate with alkoxyethanols

Fig.3 Shows deviations of refractive index (Δn_D) values are positive for all the system taken for the study. The positive devotion is may be due to the strong specific forces between the constituent molecules.

Table-3 shows the variation of the excess molar refraction (R_m^E) with mole fraction of Ethyl acetate. All the values obtained are positive. The Excess Gibbs free energy (ΔG^{*E}) deals with the molecular interactions of fluid mixtures. The positive values are indicates the strong interaction between the molecules. The Excess Gibbs free energy (ΔG^{*E}) values are positive for all the systems taken for study (i.e.) EA+2-ME, EA+2-EE and EA+2-BE. The highest positive values of (ΔG^{*E}) are obtained in EA+2-BE system and lowest in EA+2-ME. Hence the molecular interaction is naturally high in EA+2-BE system which is complemented by the property of (η^E). Table-4 shows the values for adjustable parameters and standard deviations of Redlich–Kister polynomial equations. 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 [21].

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

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

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

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

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

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

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

The standard deviation is reported in Table-5. Theoretical mixing rules of Weiner and Lorentz–Lorenz(L–L) has the very low deviation when compared to the other mixing rules. Oster, Newton, Heller, Arago–Biot(A-B) and Gladstone–Dale(G-D) having the maximum deviations as compared with 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 predicted by nature of mixing composition.

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 ethyl 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	
Ethyl acetate + 2-Methoxyethanol								
V^E (cm^3 mol^{-1})	0.3700	-0.0520	-0.2440	-0.8230	0.9200	0.8710	-1.0430	2.3530
η^E (mPa s)	-0.3080	0.0620	0.0120	-0.1800	0.2760	0.1180	0.0180	0.0926
R_m^E (cm^3 mol^{-1})	1.6040	-0.1070	-1.4770	-0.1080	3.7250	0.2050	-3.8460	1.2188
Δn_D	0.0180	-0.0050	-0.0100	0.0140	0.0000	-0.0080	-0.0070	0.0079
ΔG^{*E} (J mol^{-1})	187.5000	-162.0000	-730.1000	-416.4000	4104.0000	578.1000	-3560.0000	181.9236
Ethyl acetate + 2-Ethoxyethanol								
V^E (cm^3 mol^{-1})	0.9340	0.5960	-0.3110	0.7910	1.1870	-0.1950	-1.8090	1.6893
η^E (mPa s)	-0.3810	0.0480	0.2030	-0.2170	-0.6330	0.1690	0.8110	0.3128
R_m^E (cm^3 mol^{-1})	1.8020	-0.4290	-1.4580	2.6020	5.5880	-2.1650	-5.9300	0.6934
Δn_D	0.0290	-0.0050	-0.0250	0.0440	0.0960	-0.0380	-0.1000	0.0051
ΔG^{*E} (J mol^{-1})	1079.0000	163.1000	-930.9000	612.6000	5518.0000	-771.8000	-5665.0000	64.4161
Ethyl acetate + 2-Butoxyethanol								
V^E (cm^3 mol^{-1})	1.2790	-0.2670	-0.0110	-0.6780	0.5270	0.9450	-1.7950	2.1157
η^E (mPa s)	-0.5390	0.0020	0.1590	-0.0160	-0.3730	0.0130	0.7520	0.1159
R_m^E (cm^3 mol^{-1})	1.8170	0.0960	-0.4750	-0.3060	3.5480	0.2090	-4.8900	0.6069
Δn_D	0.0360	0.0060	-0.0100	0.0040	0.0790	-0.0110	-0.1040	0.0061
ΔG^{*E} (J mol^{-1})	2584.0000	746.1000	-701.9000	2394.0000	7926.0000	-3138.0000	-9809.0000	227.4875

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

Standard deviation (σ)							
Systems	**L–L**	**Wiener**	**G–D**	**A–B**	**Heller**	**Newton**	**Oster**
Ethyl acetate + 2-Methoxyethanol	0.0023	0.0019	0.0042	0.0042	0.0031	0.0115	0.0263
Ethyl acetate + 2-Ethoxyethanol	0.0028	0.0022	0.0052	0.0052	0.0038	0.0141	0.0322
Ethyl acetate + 2-Butoxyethanol	0.0025	0.0017	0.0044	0.0044	0.0033	0.0120	0.0276

L–L: Lorentz–Lorentz; G–D: Gladstone–Dale; A–B: Arago–Biot

4. Conclusion

Density**,** viscosity, refractive index and the excess properties such as excess molar volume, excess viscosity, deviations of refractive index, excess molar refraction and excess Gibbs free energy were computed for Ethyl acetate with 2-methoxyethanol, 2-ethoxyethanol and 2-butoxyethanol systems. The order of molecular interactions concluded from the parameters V^E, η^E and Δn_D are EA+2-ME > EA+2-EE > EA+2-BE.

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