

THERMOPHYSICAL STUDIES OF BINARY LIQUID MIXTURE OF METHYL ACETATE AND ALKOXYETHANOLS BY THE TEMPERATURE 308.15K

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ABSTRACT

The Thermophysical parameters of densities(ρ), viscosities(η), refractive indices(n_D) and the 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 Methyl acetate($C_3H_6O_2$) and Alkoxyethanols at 308.15K. The Alkoxyethanols were selected 2-methoxyethanol($C_2H_8O_2$), 2-ethoxyethanol($C_4H_{10}O_2$) and 2-butoxyethanol($C_6H_{14}O_2$). The comparative study of theoretical mixing rule relations for the refractive index of liquids has been carried out. The data obtained from the binary mixture were calculated and the interaction parameters were fitted with Redlich – Kister Polynomial equation. The computed parameters were discussed on the basis of nature of molecular interactions.

Keywords: Methyl acetate, Alkoxyethanols, Thermochemical and Excess properties,

Redlich–Kister Polynomial equation, Molecular interaction.

1. Introduction

The Information on the volumetric properties of non-electrolyte solutions are necessary for the study of heat transfer, mass transfer, chemical separation, and fluid flow. The Thermophysical properties are vital tools as knowing to the liquid state and also the bond between the liquid structures [1-5]. Carbon acts major parts in the modernization of our science society. Our study is focused in esters and alcohols, Alcohols having wide applications in medicine, chemical industry, and biology. They are strongly self-associated liquids. The complexes of alcohol symbolize an essential class of hydrogen-bonded solvents. Ester group is an investigating element for interested bulk molecular connections in mixtures originating esters as one of the components beside with either non-polar or polar solvents [6]. Esters and Alcohols are involved in trans-esterification reactions to convert their lower alkyl chain to longer alkyl chain. The accessibility of free electrons in carbonyl group to do something as a proton acceptor with alcohols has involved the attention of experimental investigations [7].

Methyl acetate is a flammable liquid occasionally used as a solvent because of the less toxicity and quick vanishing speed. It is valuable as the fast-evaporating component in high and low solvent systems of polymers and resins. In order to obtain more valuable of compound, huge amount of by-product methyl acetate is frequently hydrolyzed to methanol and acetic acid in trade polyvinyl alcohol plants [8-9].

Alkoxyethanols like 2-methoxyethanol, 2-ethoxyethanol and 2-butoxyethanol are used as a solvent in making dyes, resins and varnishes. It is an extremely attractive group of solvent getting ether (i.e) hydroxyl and oxygen group is mentioned for it is accepting and donating ability. Moreover, the viscosity(η) is compulsory liquid move from beginning to end cavity

surfaces, pipes, and for hydraulic computing in energy and mass relocate, in chemical industry. The refractive index is appraised of standard electromagnetic energy. Excess viscosities(η^E), Excess molar volumes(V^E) are frequently applied to investigate molecular interactions in solutions, which assist us to know those actual performances and expand hypothetical models for those explanations on top of the imitation processes. The modern history, we have been the great curiosity in hypothetical and trial investigations of the thermophysical properties of solutions [10]. The Redlich–Kister polynomial equation is applied to recognize the standard deviation in hypothetical and untried values of the liquid solutions. The different theoretical mixing rules are applied in these readings. After during investigation, the interaction behavior of Methyl acetate(MA) with Alkoxyethanols like 2-methoxyethanol(2-ME), 2-ethoxyethanol(2-EE) and 2-butoxyethanol(2-BE) at 308.15K were studied.

2. Materials and Methods

2.1 Materials

Chemicals like Methyl acetate, 2-methoxyethanol, 2-ethoxyethanol and 2-butoxyethanol were provided by Loba products (99% Purity), which are utilized without further purification. The purity of chemicals were tested with their literature values such as 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). Its accuracy is ± 0.0001 g. Weightings were done at least five times for accuracy of the measurements. The density of pure liquid solutions were utilized

by double armed Pycnometer. 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 $\pm 0.01s$. 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 $\pm 0.01K$.

2.4 Refractive Index Measurement

The refractive index 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 .

3. Calculations

Following are the excess or deviation of the parameters [11-16].

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

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

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

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

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

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 hypothetical values. The standard deviation data were resumed the equation,

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

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

4. Result and Discussions

Here [Table 1](#) reveals the literature and experimental values of Methyl acetate and Alkoxyethanols at the temperature 308.15K [17-28]. [Table 2](#) Shows, the variations of mole

fractions of methyl acetate with increasing the values of viscosities, densities, refractive indices, molar refraction and polarizability.

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
Methyl acetate	0.9150	0.3551	1.3550	0.9152 [21]	0.351 [21]	1.3550 [24]
				0.9152 [24]	0.355 [24]	1.3550 [25]
				0.9152 [25]	0.355 [25]	1.3528 [26]
				0.9122 [26]	0.352 [26]	
2-Methoxy ethanol	0.9510	1.2562	1.3960	0.9511 [23]	1.246 [19]	1.3949 [26]
				0.9510 [17]	1.2491 [20]	
				0.9510 [18]	1.257 [22]	
				0.9511 [19]	1.390 [26]	
				0.9511 [20]		
				0.9511 [21]		
				0.9507 [26]		
0.9530 [27]						
2-Ethoxy ethanol	0.9160	1.4870	1.4020	0.9164 [20]	1.4823 [20]	1.4017 [27]
				0.9164 [22]	1.480 [22]	1.4018 [28]
				0.9174 [27]	1.480 [28]	
				0.9163 [28]		
2-Butoxy ethanol	0.8870	2.2871	1.4130	0.8878 [19]	2.079 [19]	1.4132 [27]
				0.8889 [27]		

The refractive index is measure in optical region and it must not contribute polarizability of the liquids in orientational effects [10]. Here Table 3 Contains excess and deviation of parameters such as V^E , η^E , Δn_D , R_m^E , and ΔG^{*E} of Methyl acetate and Alkoxyethanols at the temperature 308.15K.

Table 2

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

Mole fraction of Methyl 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 $\alpha \times 10^{-26}$ $cm\ mol^{-1}$
Methyl acetate + 2-Methoxyethanol					
0.0000	0.9150	0.3551	1.3550	17.6453	0.6998
0.1015	0.9173	0.3770	1.3605	17.8942	0.7096
0.2022	0.9197	0.4049	1.3660	18.1418	0.7194
0.3008	0.9225	0.4332	1.3715	18.3798	0.7289
0.4022	0.9254	0.4889	1.3770	18.6141	0.7382
0.4999	0.9288	0.5725	1.3815	18.7936	0.7453
0.6044	0.9327	0.6899	1.3855	18.9419	0.7512
0.6987	0.9368	0.8250	1.3885	19.0367	0.7549
0.7998	0.9414	0.9671	1.3910	19.1049	0.7576
0.9002	0.9461	1.1108	1.3935	19.1684	0.7602
1.0000	0.9510	1.2562	1.3960	19.2275	0.7625
Methyl acetate + 2-Ethoxyethanol					
0.0000	0.9150	0.3551	1.3550	17.6453	0.6998
0.0862	0.9140	0.4024	1.3635	18.3808	0.7289
0.1745	0.9130	0.4610	1.3725	19.1608	0.7599
0.2634	0.9120	0.5408	1.3815	19.9577	0.7915
0.3567	0.9112	0.6312	1.3890	20.7122	0.8214
0.4538	0.9110	0.7381	1.3945	21.3858	0.8481
0.5562	0.9114	0.8561	1.3980	21.9794	0.8716
0.6584	0.9122	0.9800	1.3995	22.4685	0.8910
0.7657	0.9134	1.1207	1.4005	22.9465	0.9100
0.8798	0.9146	1.2886	1.4010	23.4282	0.9291
1.0000	0.9160	1.4870	1.4020	23.9571	0.9501
Methyl acetate + 2-Butoxyethanol					
0.0000	0.9150	0.3551	1.3550	17.6453	0.6998
0.0645	0.9106	0.4705	1.3670	18.9672	0.7522
0.1321	0.9065	0.5907	1.3780	20.3221	0.8059
0.2070	0.9026	0.7206	1.3890	21.8033	0.8647
0.2883	0.8988	0.8654	1.3975	23.2797	0.9232
0.3743	0.8959	1.0256	1.4040	24.7286	0.9807
0.4764	0.8931	1.2217	1.4085	26.2940	1.0427
0.5864	0.8912	1.4439	1.4100	27.7857	1.1019
0.7029	0.8898	1.6820	1.4115	29.3536	1.1641
0.8396	0.8884	1.9610	1.4120	31.1186	1.2341
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(ΔG^{*E}) for Methyl acetate + alkoxyethanol binary mixtures at 308.15K.

Mole fraction of Methyl acetate X_2	Excess molar volume V^E $\text{cm}^3 \text{mol}^{-1}$	Excess viscosity η^E mPa s	Deviation of refractive index Δn_D	Excess molar Refraction R_m^E $\text{cm}^3 \text{mol}^{-1}$	Excess Gibbs free energy ΔG^{*E} J mol^{-1}
Methyl acetate + 2-Methoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.1015	0.1093	-0.0682	0.0013	0.0907	-167.0286
0.2022	0.2167	-0.1303	0.0027	0.1801	-302.4908
0.3008	0.2850	-0.1922	0.0042	0.2598	-442.8505
0.4022	0.3423	-0.2266	0.0055	0.3360	-458.8931
0.4999	0.3574	-0.2331	0.0060	0.3573	-372.8867
0.6044	0.3367	-0.2058	0.0057	0.3473	-237.3464
0.6987	0.2781	-0.1609	0.0049	0.2839	-90.5715
0.7998	0.2018	-0.1088	0.0032	0.1939	-15.1157
0.9002	0.1088	-0.0552	0.0016	0.0992	11.2529
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Methyl acetate + 2-Ethoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0862	0.0979	-0.0502	0.0044	0.1911	11.2040
0.1745	0.1996	-0.0916	0.0093	0.4141	40.6776
0.2634	0.3049	-0.1124	0.0141	0.6500	126.8510
0.3567	0.3957	-0.1275	0.0172	0.8156	183.4152
0.4538	0.4322	-0.1307	0.0182	0.8760	227.8973
0.5562	0.4115	-0.1286	0.0169	0.8232	231.0476
0.6584	0.3491	-0.1203	0.0136	0.6676	200.2735
0.7657	0.2437	-0.1011	0.0095	0.4682	146.2341
0.8798	0.1346	-0.0623	0.0047	0.2299	80.5798
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Methyl acetate + 2-Butoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0645	0.1374	-0.0091	0.0083	0.3179	428.1197
0.1321	0.2777	-0.0196	0.0153	0.6190	703.8040
0.2070	0.4071	-0.0343	0.0220	0.9343	870.6448
0.2883	0.5299	-0.0467	0.0258	1.1436	963.8689
0.3743	0.5866	-0.0526	0.0273	1.2533	996.0973
0.4764	0.6032	-0.0539	0.0259	1.2274	959.6640
0.5864	0.5325	-0.0441	0.0210	1.0060	858.5491
0.7029	0.3910	-0.0311	0.0157	0.7590	682.0116
0.8396	0.2149	-0.0162	0.0083	0.3952	403.0799
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

The Excess molar volume(V^E) values are positive and excess viscosity(η^E) values are negative in all systems. Further the curves in (Fig.1) shows that the excess molar volume(V^E) increases as the chain length of alkoxyethanols increases.

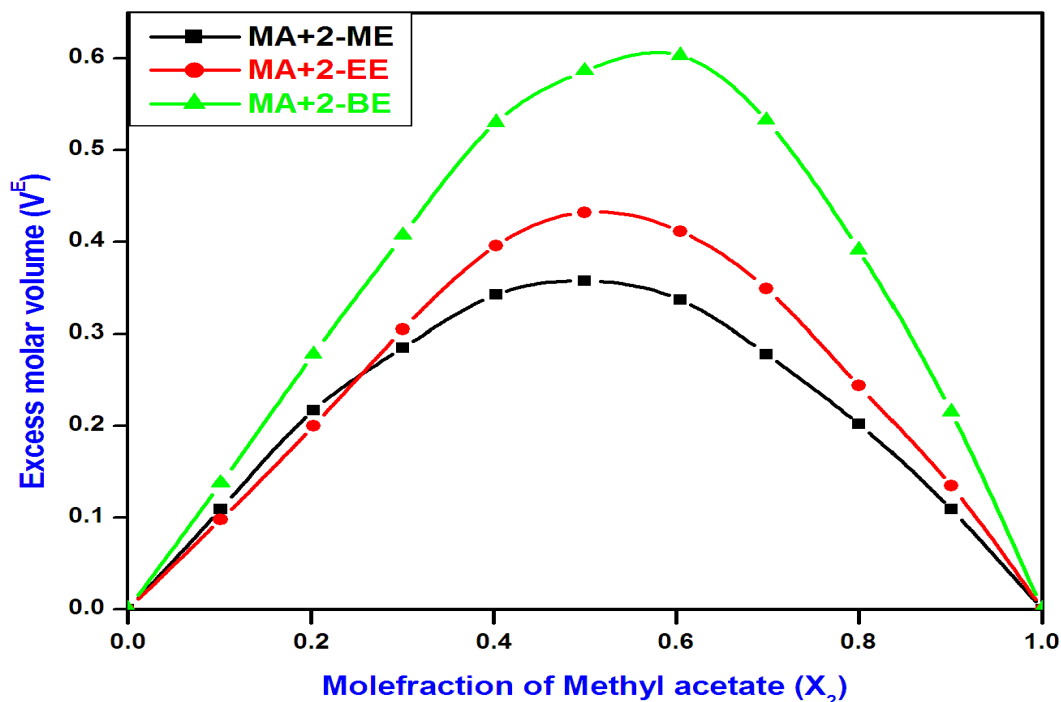


Fig.1. Excess molar volume(V^E) vs Mole fraction(X_2) of the Methyl acetate with alkoxyethanols.

The excess molar volume(V^E) has high positive readings which are located between 0.4 and 0.6 Mole fractions of Methyl Acetate. The highest positive readings of the whole range of the systems are present in MA+2-BE. It may be attributed to supremacy of molecular dissociation [29].

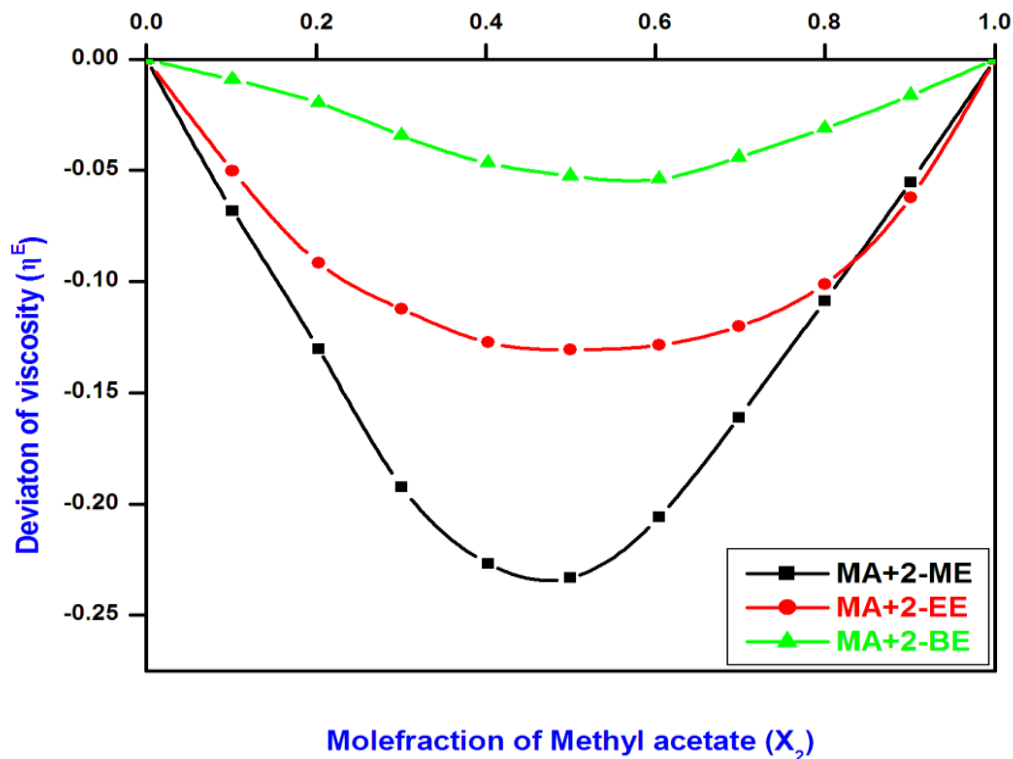


Fig.2. Excess viscosity (η^E) vs Mole fraction (X_2) of the Methyl acetate with alkoxyethanols.

The (Fig.2) shows that excess viscosity(η^E) increasing as the chain length of alkoxyethanols increases. These excess viscosity readings are negative; where as the highest negative readings are located between 0.4 to 0.6 mole fractions. The excess viscosity(η^E) trend is 2-ME > 2-EE > 2-BE. The excess viscosity(η^E) readings are found to be opposite to the sign of excess molar volume V^E for the three binary mixtures, which is in agreement with the views proposed by Brocos et al., [30, 31]. A correlation between the sign (η^E) and (V^E) has been observed for a number of binary systems; (i.e.) (η^E) negative when (V^E) is positive and vice-versa [32, 33].

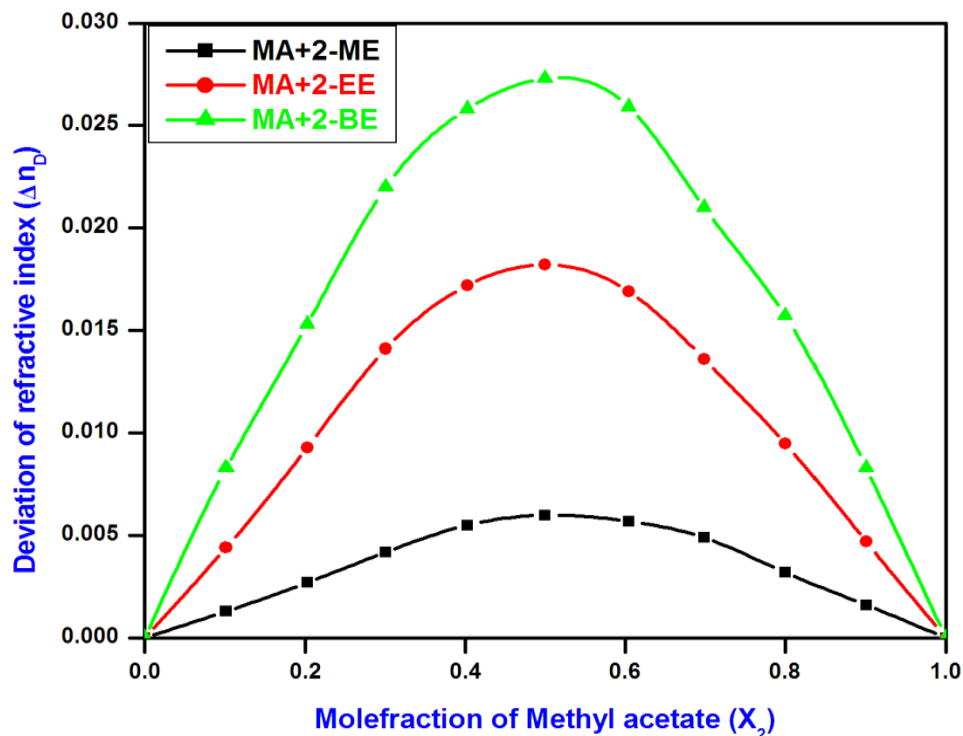


Fig.3. Deviation of refractive index(Δn_D) vs mole fraction(X_2) of the Methyl acetate with alkoxyethanols.

The (Fig.3.) shows deviations of refractive index(Δn_D) readings. Which are positive for the whole system. These positive readings are due to the strong specific forces between the participated molecules, such as hydrogen bonding between the constituent molecules.

In the (Table 3), the excess molar refraction(R_m^E) shows the highest positive readings. Which are located between 0.4 and 0.6 mole fractions of methyl acetate and the order of magnitude and change of sign indicates the size variation between one and the same molecules. The Excess Gibbs free energy(ΔG^{*E}) deals with the molecular interactions of fluid mixtures. The positive readings are indicates the strong interaction between the molecules and negative reading shows the dispersion forces between the molecules. The Excess Gibbs free energy(ΔG^{*E}) readings are negative in MA+2-ME and positive for MA+2-EE and MA+2-BE of the whole

composition series of systems. From (Table 4), we can views the readings of adjustable parameters and standard deviations of Redlich–Kister polynomial equations [10]. The readings of standard deviations are giving the acceptable outcome for the experimental readings. The experimental refractive index readings are examined by seven theoretical mixing rules [34 – 36].

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

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

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

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

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

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

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

Table 4

Values of adjustable parameters(B_k) and the corresponding standard deviations(σ), for excess molar volumes(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})for Methyl acetate + alkoxyethanol binary mixtures at 308.15K.

Parameters	B_k							σ
	B_0	B_1	B_2	B_3	B_4	B_5	B_6	
Methyl acetate + 2-Methoxyethanol								
V^E (cm ³ mol ⁻¹)	1.4540	0.0880	-1.3810	-0.1960	4.3660	0.1050	-4.4380	2.5452
η^E (mPa s)	-0.9470	-0.1970	1.2170	0.1960	-2.9240	0.0000	2.6530	0.1514
R_m^E (cm ³ mol ⁻¹)	1.4730	-0.1430	-1.8560	0.1020	4.4040	0.0390	-4.0190	1.0965
Δn_D	0.0240	-0.0030	-0.0310	0.0010	0.0670	0.0020	-0.0590	0.0076
ΔG^{*E} (J mol ⁻¹)	-1547.0000	-2071.0000	2682.0000	471.0000	-4745.0000	1584.0000	3608.0000	405.7533
Methyl acetate + 2-Ethoxyethanol								
V^E (cm ³ mol ⁻¹)	1.7610	0.2800	-2.2580	-0.5710	5.3820	0.2960	-4.8800	1.8100
η^E (mPa s)	-0.5360	0.0020	0.2230	-0.2140	-2.0190	0.2090	2.3300	0.3296
R_m^E (cm ³ mol ⁻¹)	3.5160	0.9040	-3.6840	-1.1290	7.2850	0.2370	-7.1080	0.8797
Δn_D	3.5160	0.9040	-3.6840	-1.1290	7.2850	0.2370	-7.1080	0.8797
ΔG^{*E} (J mol ⁻¹)	946.3000	7.2280	-1295.0000	-1534.0000	1475.0000	1528.0000	-1123.0000	27.3726
Methyl acetate + 2-Butoxyethanol								
V^E (cm ³ mol ⁻¹)	2.4640	0.7310	-3.5740	0.5900	10.4200	-1.2900	-9.2840	2.2624
η^E (mPa s)	-0.2130	-0.1160	0.2720	0.1450	-0.5830	-0.0300	0.5210	0.0724
R_m^E (cm ³ mol ⁻¹)	4.9480	2.2840	-6.6040	1.3400	21.1100	-3.5400	-19.3800	0.8107
Δn_D	0.1050	0.0520	-0.1530	0.0720	0.5710	-0.1220	-0.5210	0.0123
ΔG^{*E} (J mol ⁻¹)	4152.0000	500.5000	-8996.0000	8151.0000	37616.0000	-8521.0000	-32660.0000	364.3747

Table 5

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

Standard deviation σ							
Systems	L-L	Wiener	G-D	A-B	Heller	Newton	Oster
Methyl acetate + 2-Methoxyethanol	0.0058	0.0046	0.0106	0.0106	0.0079	0.0291	0.0665
Methyl acetate + 2-Ethoxyethanol	0.0076	0.0039	0.0091	0.0091	0.0068	0.0250	0.0572
Methyl acetate + 2-Butoxyethanol	0.0085	0.0058	0.0138	0.0138	0.0103	0.0379	0.0865
L-L: Lorentz-Lorentz; G-D: Gladstone-Dale; A-B: Arago-Biot							

The standard deviation is computed and reported in (Table 5). Theoretical mixing rules like Lorentz-Lorenz(L-L), Arago-Biot(A-B), Newton, Gladstone-Dale(G-D), Heller(H), Wiener(W) and Oster deals with predicting the refractive index of a fluid. It has been accomplished and validity for the liquid binary mixtures. The presentation of the theoretical mixing rules of Wiener(W) has small deviation while Oster and Newton's has comparatively higher deviation values. The deviations indicate that values of the experimental and theoretical are almost nearby. The deviations are less than 2% for the calculated binary systems [34].

5. Conclusion

Thermophysical parameters such as viscosity (η), density (ρ) and the refractive index (n_D) were calculated from Methyl acetate with alkoxyethanols solutions at 308.15K. 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}) readings were calculated and fitted with Redlich–Kister polynomial equations. The order of molecular interactions are (MA+2-ME > MA+2-EE > MA+2-BE). Hence from the presentations of theoretical mixing rules of Oster and Newton's readings were comparatively maximum deviations than that of other theoretical mixing rules.

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