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Molecular interaction studies of ketones with 2–methoxyethanol: Densities, viscosities, refractive indices and excess properties at 303 K

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ABSTRACT

Densities, viscosities and refractive indices have been measured as a function of composition for the binary mixtures of ketones with 2–methoxyethanol at 303 K. Molar refraction and polarizability values are calculated by using measured refractive indices. The excess properties such as excess molar volume (V^E*), deviations in viscosities (*η^E*), deviation in refractive indices (*Δn_D*), excess molar refraction (*R_m^E*) and excess Gibbs free energy (*ΔG^{*E}*) values are calculated by using the experimental values. The excess properties values are correlated with Redlich–Kister polynomial equation to obtain their coefficients and standard deviations. The experimental refractive index values are analyzed by different theoretical mixing rules and the standard deviations are predicted. A negative deviation of excess molar volume and positive deviation of excess Gibbs free energy values indicate that specific types of interaction occurred between unlike molecules. The specific force may be cited in the form of hydrogen bonding between the unlike molecules (C=O…H–O).*

Keywords: Density, Viscosity, Excess property, Ketone, 2-methoxyethanol, Molecular interaction.

INTRODUCTION

Volumetric and viscometric properties of liquid mixtures are most important in the field of molecular modeling and drugs designing. Physico–chemical and thermodynamic properties of non–electrolyte solutions are essential in the chemical separations, heat transfers, mass transfers and fluid forces. The determination of density (ρ), viscosity (η) and refractive index (n_D) are valuable tools for determining the liquid state and liquid structure of the consistent species. Excess molar volume (V^E) and deviation in viscosity (η^E) are the most important factors due to elucidate the interaction forces between the molecules and the relation between the compounds. According to reviewers, reports, the valuable information regarding the molecular complexes are based on the structural and macroscopic studies of the individual molecule [1–5].

In our present investigation we have to summarize the interaction behaviors of some ketones like dimethylketone (DMK), ethylmethylketone (EMK) and diethylketone (DEK) with 2–methoxyethanol (2–ME) at 303 K. Ketones are the simplest molecules that contain a common carbonyl group (C=O). 2–alkoxyethanol is very interesting class of solvents having both (i.e.) oxygen (–O–) and hydroxyl (–OH–) groups and for its donating and accepting ability. These concentrations of binary mixtures were chosen to study the relative strength of intermolecular interactions between the C=O…..H–O molecules.

MATERIALS AND METHODS

All compounds used in this work were supplied by Loba (purity >> 99%) chemicals which were used without further purification. The purity of chemicals was checked by comparing their densities and refractive indices with literature values. The binary mixtures were prepared using airtight stoppered bottles (due to evaporation), and the weight of the empty bottle was measured by digital electronic balance (Anamed, M–300DR) with accuracy ±0.001g. The weights were measured at least three times for accuracy of the composition of mixtures. The density values of liquid and liquid mixtures were measured using a double armed pycnometer which has a bulb volume of 10 ml. The pycnometer was calibrated with freshly prepared double distilled water. A digital electronic balance was used in the density measurements.

The viscosities of pure and binary liquid mixtures were measured using an Ostwald's viscometer. It was calibrated with double distilled water. The measurement of flow time of the solution between the two points of the viscometer was performed at least five times for each solution and the result was averaged. The time flow was measured using digital stop watch with an accuracy of ±0.01sec. The viscometer was fitted vertically in the thermostat at a constant temperature. For all the measurements, temperature was controlled by circulating water bath through an ultra thermostat (Concord) with an accuracy of ± 0.15 K. The uncertainty in density, viscosity and refractive index measurements was within ± 0.0001 g cm^{-3}, ± 2×10^{-4} mPa s and ± 0.0001.

The excess properties like excess molar volume, deviation of viscosity, deviation of refractive index, excess molar refraction and excess Gibbs free energy were determined from the experimental data according to the following relations [6–9],

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

$$\eta^E = \eta - \sum_{i=1}^{N} x_i \eta_i \tag{2}$$

$$\Delta n_D = n_D - \sum_{i=1}^{N} x_i n_{Di} \tag{3}$$

$$R^E = R_m - \sum_{i=1}^{N} \phi_i R_{mi} \tag{4}$$

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

where, x_i and φ_i represent the mole fraction, volume fraction of the pure component i, respectively. V, ρ, η, n_D and R_m are the molar volume, density, viscosity, refractive index and molar refraction of the mixtures, respectively, and V_i, ρ_i, η_i, n_{Di} and R_{mi} the corresponding properties of the pure components. The calculated values of excess properties are excess molar volume, deviation of viscosity, deviation of refractive index, excess molar refraction and excess Gibbs free energy values are fitted with Redlich-Kister polynomial equation [10],

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

The values of a_i of equ (6) were calculated by the method of least square fit. The values of these parameters of each studied system are used to identify the standard deviation of the experimental and theoretical values. The standard deviation values are summarized by the following relation,

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

Where, n is the number of experimental points, p is the number of parameters, X_{exp} and X_{cal} are the experimental and calculated properties.

RESULTS AND DISCUSSION

Table 1 contains the experimental and literature values of pure compounds at temperature 303 K. The experimental values of density (ρ), viscosity (η) and refractive index (n_D) were listed as shown in Table 2 respectively, as a function of mole fraction of solute (ketones). As the concentration of the solute increases the values of density, viscosity and refractive index values are decreases.

The molar refraction values are calculated by using Lorentz–Lorenz relation and are reported in Table 2 respectively. The magnitude of variation of molar refraction was decreasing for DMK + 2–ME and increasing for EMK + 2-ME and DEK + 2-ME. The molar refraction is related to the strength of dispersion forces which leads to the dipolar rotation of the molecule [16]. Fig. 1 shows the variation of excess molar volume of dimethylketone, ethylmethylketone and diethylketone with 2–methoxyethanol at 303 K.

Table 1. Experimental and literature values of pure components at 303 K**.**

Compounds	**Experimental**		**Literature**	
	ρ (g cm^{-3})	**n_D**	**ρ (g cm^{-3})**	**n_D**
DMK	0.7847	1.3605	0.7854[11]	1.3576[12]
EMK	0.8081	1.3905	0.7945[13]	1.3769[13]
DEK	0.8022	1.3775	0.8046[13]	1.3900[13]
2–ME	0.9553	1.4005	0.9602[14]	1.3983[15]

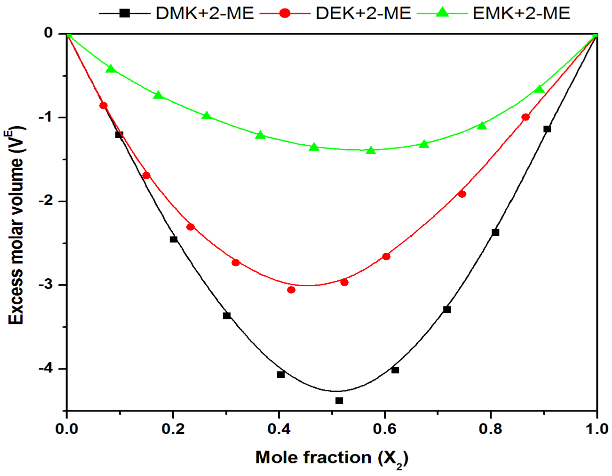


Fig 1. Excess molar volume (V^E) of ketones (■) DMK + 2–ME, (●) DEK + 2–ME and (▲) EMK+2–ME *vs.* mole fraction of the solute (X_2).

The excess molar volume value is negative for the whole composition range and maximum value around at ~ 0.5 mole fraction of ketones in alkoxyethanol. Sreenivasulu and Naidu [17] have reported the maximum value of V^E, ethylmethylketone (–3.943 cm^3 mol^{-1}), methyl n–propylketone (–3.509 cm^3 mol^{-1}) and diethylketone (–2.409 cm^3 mol^{-1}) at 303 K. Compared with literature, the maximum value of V^E is obtained for ketones with alcohols at 303 K. It may be due to the aliphatic nature of both molecules and interacts easily. In the case of ketone + 2–ME systems, the maximum value of V^E obtained for DMK + 2–ME (–4.3769 $m^3 mol^{-1}$) EMK + 2–ME (–3.0587 $m^3 mol^{-1}$) and DEK + 2–ME (–1.4033 $m^3 mol^{-1}$).

Table 2. Experimental values of density (ρ), viscosity (η), refractive index (n_D), molar refraction (R_m) and polarizability (α) of ketones (DMK, EMK and DEK) + 2–methoxyethanol binary mixtures at 303 K.

Mole fraction X_2	**Density ρ $g\ cm^{-3}$**	**Viscosity η mPa s**	**Refractive index n_D**	**Molar refraction R_m $cm^3\ mol^{-1}$**	**Polarizability α x 10^{-26} $cm\ mol^{-1}$**
Dimethylketone + 2–methoxyethanol					
0.0000	0.9553	1.9678	1.4005	19.3314	0.7666
0.0981	0.9542	1.8925	1.3960	18.7146	0.7422
0.2007	0.9527	1.7745	1.3915	18.0947	0.7176
0.3013	0.9473	1.6363	1.3870	17.5620	0.6965
0.4039	0.9385	1.4877	1.3825	17.0848	0.6775
0.5134	0.9225	1.3291	1.3785	16.7243	0.6632
0.6196	0.8986	1.1553	1.3751	16.5440	0.6561
0.7175	0.8724	0.9711	1.3717	16.4455	0.6522
0.8089	0.8454	0.7807	1.3681	16.3841	0.6497
0.9060	0.8139	0.5775	1.3644	16.3859	0.6498
1.0000	0.7847	0.3726	1.3605	16.3545	0.6486
Ethylmethylketone + 2–methoxyethanol					
0.0000	0.9553	1.9678	1.4005	19.3314	0.7666
0.0829	0.9461	1.8764	1.3980	19.3265	0.7664
0.1718	0.9347	1.7799	1.3955	19.3617	0.7678
0.2629	0.9224	1.6714	1.3928	19.4089	0.7697
0.3640	0.9085	1.5368	1.3900	19.4762	0.7724
0.4660	0.8937	1.3866	1.3875	19.5778	0.7764
0.5731	0.8775	1.2213	1.3853	19.7262	0.7823
0.6738	0.8614	1.0575	1.3835	19.9028	0.7893
0.7828	0.8431	0.8694	1.3815	20.1209	0.7979
0.8910	0.8234	0.6705	1.3795	20.3841	0.8084
1.0000	0.8022	0.4638	1.3775	20.6996	0.8209
Diethylketone + 2–methoxyethanol					
0.0000	0.9553	1.9678	1.4005	19.3314	0.7666
0.0687	0.9520	1.9196	1.3990	19.5078	0.7736
0.1492	0.9464	1.8435	1.3975	19.7638	0.7838
0.2325	0.9380	1.7495	1.3960	20.0899	0.7967
0.3179	0.9275	1.6384	1.3948	20.4846	0.8124
0.4230	0.9131	1.4928	1.3935	21.0232	0.8337
0.5232	0.8961	1.3467	1.3925	21.6401	0.8582
0.6024	0.8812	1.2244	1.3920	22.1967	0.8803
0.7455	0.8544	0.9864	1.3915	23.2665	0.9227
0.8649	0.8315	0.7725	1.3910	24.2219	0.9606
1.0000	0.8081	0.5180	1.3905	25.2931	1.0030

Table 3. Excess molar volume V^E, deviation of viscosity η^E, deviation of refractive index Δn_D, excess molar refraction Rm^E and excess Gibbs free energy of activation of viscous flow ΔG^{*E} of ketones (DMK, EMK and DEK) + 2–methoxyethanol binary mixtures at 303 K.

Mole fraction X_2	**Excess molar volume V^E $cm^3\ mol^{-1}$**	**Deviation of 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} $J\ mol^{-1}$**
Dimethylketone + 2–methoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0981	-1.2027	0.0842	-0.0005	-0.3191	271.9744
0.2007	-2.4520	0.1258	-0.0010	-0.6414	497.0665
0.3013	-3.3655	0.1470	-0.0015	-0.8764	681.9973
0.4039	-4.0694	0.1579	-0.0020	-1.0559	846.3488
0.5134	-4.3769	0.1589	-0.0020	-1.1187	1010.1225
0.6196	-4.0146	0.1446	-0.0014	-1.0013	1113.9417
0.7175	-3.2939	0.1200	-0.0008	-0.8021	1110.2436
0.8089	-2.3687	0.0890	-0.0004	-0.5658	974.2525
0.9060	-1.1348	0.0453	-0.0001	-0.2663	665.9721
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Ethylmethylketone + 2–methoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0687	-0.8553	0.0513	-0.0008	-0.2330	147.7584
0.1492	-1.6928	0.0920	-0.0015	-0.4572	297.3349
0.2325	-2.3030	0.1187	-0.0022	-0.6276	432.6981
0.3179	-2.7311	0.1315	-0.0025	-0.7422	546.5914
0.4230	-3.0557	0.1383	-0.0028	-0.8300	660.0397
0.5232	-2.9663	0.1373	-0.0028	-0.8102	741.4612
0.6024	-2.6610	0.1300	-0.0025	-0.7263	776.5124
0.7455	-1.9109	0.0995	-0.0015	-0.5097	730.2504
0.8649	-0.9914	0.0586	-0.0009	-0.2656	534.3959
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Diethylketone + 2–methoxyethanol					
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0829	-0.4274	0.0333	-0.0006	-0.1184	168.4377
0.1718	-0.7427	0.0705	-0.0010	-0.2049	348.8168
0.2629	-0.9858	0.0990	-0.0017	-0.2822	513.9841
0.3640	-1.2189	0.1165	-0.0021	-0.3532	662.9998
0.4660	-1.3609	0.1195	-0.0023	-0.3911	770.2903
0.5731	-1.4033	0.1154	-0.0020	-0.3893	839.0320
0.6738	-1.3270	0.1031	-0.0015	-0.3505	845.1908
0.7828	-1.1073	0.0790	-0.0010	-0.2816	755.6508
0.8910	-0.6710	0.0428	-0.0005	-0.1665	509.1087
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000

The negative deviation of V^E indicates that a specific type of interaction occurred between the unlike molecules. Many authors have reported the excess molar volume of ketone binary mixtures with various additives [18–21]. The high negative deviation from ideality in the three binary mixtures were due to the strong intermolecular force such as chemical (or) specific interaction of the molecules, like hydrogen bonding, dipole–dipole interaction between the molecules.

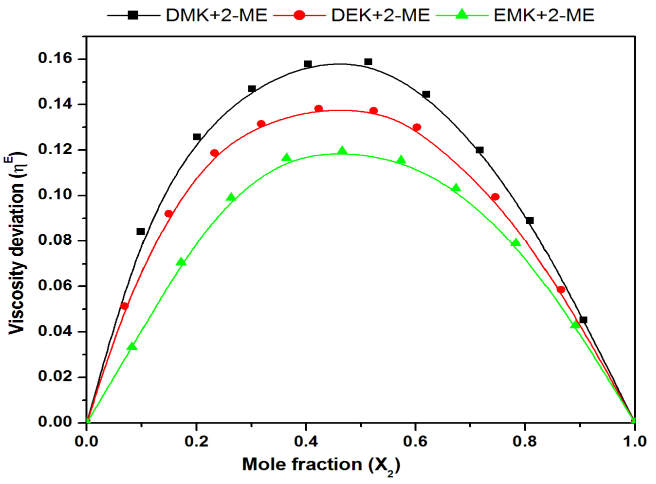


Fig 2. Deviation of viscosity (η^E) of ketones (■) DMK + 2–ME, (●) DEK + 2–ME and (▲) EMK + 2–ME *vs.* mole fraction of the solute (X_2).

In the case of Fig 2 deviations in viscosity values are positive for all the studied systems. Because of these factors while mixing the two components the molecules are interacted by the dipoles. The deviations of viscosity values suggest the typical results to the ideality [22–24]. The η^E values are positive therefore the molecules are attracted by the specific force in between the molecules. Especially the positive contribution of η^E arises shown in Fig 2 the molecules are aggregated by hydrogen bonding.The deviation of refractive index values of DMK, EMK, and DEK with 2–methoxyethanol binary mixtures are reported in Table 3. The (Δn_D) values are negative for 2–methoxyethanol. The deviation in refractive index values, indicates that the specific type of interaction among the unlike molecules and its insights depend upon its composition [25].

The proper deviation of refractive index (Δn_D) is a significant one, while refractive index values are obtained by the optical region and it is related to the strength of dispersion forces which leads to dipolar rotation of the molecules. The calculated (Δn_D) values are negative for 2–methoxyethanol. The positive contribution reveals that the nature of dipolar rotation relates to the strong correlation between the unlike molecules. The negative to positive deviation of (Δn_D) values are diminishing the chain length of alkoxyethanols. The excess molar refraction values are reported in the Table 3. The excess molar refraction values are negative for the whole composition range for all the studied systems. This behavior implies that the H–bond formed in the distinct species. The variation in molar volume is also a capability parameter that helps in the volume contraction and expansion. The volume contraction depends upon the size and shape of the molecules [26].

The magnitude and sign change in excess molar refraction indicate the size difference between the identical molecules. If we mix the two unlike molecules of unequal molar volumes there is a possibility of acceleration of one molecule in the intestinal position of other. The 2–methoxyethanol molar volume is ($79.654\text{x}10^{-6}$) so, the possibility of bond formation between the unlike molecules such as ketones with 2-ME.

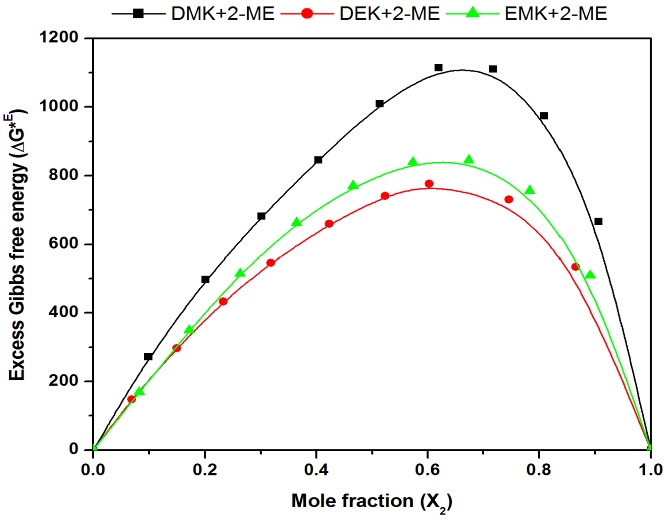
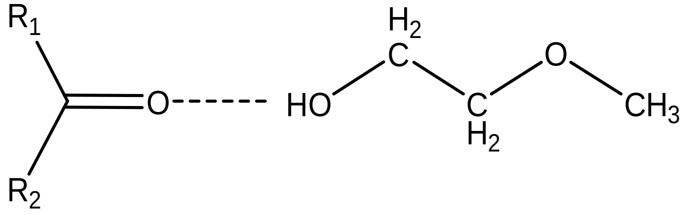


Fig 3. Excess Gibbs free energy (ΔG^{*E}) of ketones (■) DMK + 2–ME, (●) DEK + 2–ME and (▲) EMK + 2–ME *vs.* mole fraction of the solute (X_2).

Fig. 3 show the variation of excess Gibbs free energy with mole fraction of ketones. The excess Gibbs free energy values are positive for the whole composition range for all the studied systems. The contributing factor was the interaction between the components. The positive deviation of (ΔG^{*E}) is detected in binary mixtures, where strong specific interaction between unlike molecules. The negative value of (ΔG^{*E}) indicate dispersing forces among the unlike molecules [23]. From our studied systems the (ΔG^{*E}) values are positive and high for DMK + 2–methoxyethanol as compared to other systems. This is a considerable interest in the interaction behavior of C=O in ketone and (–OH) in alkoxyethanol for one example given in Fig. 4, DMK with 2-ME.



$R_1, R_2 = CH_3; CH_3, CH_3CH_2; CH_3CH_2, CH_3CH_2$

Fig 4. Hydrogen bond formation of ketones with 2–methoxyethanol.

Table 4. Values of adjustable parameters (A_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 of ketones (DMK, EMK and DEK) + 2–methoxyethanol binary mixtures at 303 K.

Parameters	**Adjustable parameters (A_k)**							**S.D σ**
	A_0	A_1	A_2	A_3	A_4	A_5	A_6	
				Ketones + 2–methoxyethanol				
V^E cm^3 mol^{-1}	-17.610	0.830	16.010	-2.429	-45.040	1.606	46.620	0.021
	-5.736	1.457	5.630	-0.574	-27.920	-0.912	28.000	0.027
	-12.380	-0.978	15.840	-12.690	-54.270	13.480	50.680	0.087
η^Em Pa s	0.657	-0.040	-0.649	1.114	3.383	-1.070	-3.389	0.003
	0.487	0.057	-0.193	-0.061	1.069	0.005	-1.361	0.001

	0.578	-0.034	-0.701	0.920	3.364	-0.874	-3.232	0.010
R_m^E cm^3 mol^{-1}	-4.509	-0.067	4.536	-0.952	-12.100	1.017	12.070	0.004
	-1.632	0.218	2.003	-0.162	-7.984	-0.066	7.606	0.007
	-3.380	-0.297	4.574	-3.449	-15.260	3.698	14.060	1.513
ΔG^{E*} J mol^{-1}	4167.000	-1360.000	-4224.000	-8252.000	26486.000	9559.000	-26408.000	19.104
	3279.000	-856.000	-1954.000	-5128.000	13736.000	5973.000	-15056.000	6.186
	2980.000	-1173.000	-1759.000	-2263.000	12803.000	3456.000	14007.000	10.212

The values of adjustable parameters (a_i), standard deviation (σ) values of ketones with 2–ME are listed in Table 4. The standard deviation of the liquid mixtures gives a complementary approach to the experimental values. The experimental refractive index values are analyzed by seven different theoretical mixing rules (Lorentz–Lorenz, Weiner, Heller, Gladstone, Newton, Oster and Edwards) [27–30] and the average standard deviations are calculated and are reported in Table 5.

Table 5. Values of average standard deviation for different theoretical mixing rules.

Average standard deviation (σ)							
Compounds	**L–L**	**Wiener**	**G–D**	**Heller**	**Newton**	**Oster**	**Edwards**
2–methoxyethanol							
DMK	0.0006	0.0009	0.0011	0.0005	0.0008	0.0035	0.0011
EMK	0.0004	0.0005	0.0011	0.0004	0.0006	0.0025	0.0008
DEK	0.0007	0.0006	0.0013	0.0006	0.0009	0.0037	0.0013

The different theoretical mixing rules follow the equation as given below,

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

$$Weiner : \frac{n_D^2 - n_{D1}^2}{n_D^2 + 2n_{D2}^2} = \left[\frac{n_{D2}^2 - n_{D1}^2}{n_{D2}^2 + 2n_{D1}^2}\right]\phi_2 \tag{9}$$

$$Heller : \frac{n_D - n_{D1}}{n_{D1}} = \frac{3}{2}\left[\frac{(n_{D2}/n_{D1})^2 - 1}{(n_{D2}/n_{D1})^2 + 2}\right]\phi_2 \tag{10}$$

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

$$Newton : n_D^2 - 1 = (n_{D1}^2 - 1)\phi_1 + (n_{D2}^2 - 1)\phi_2 \tag{12}$$

$$Oster : \frac{(n_D^2 - 1)(2n_D^2 + 1)}{n_D^2} = \frac{(n_{D1}^2 - 1)(2n_{D1}^2 + 1)}{n_{D1}^2}\phi_1 + \frac{(n_{D2}^2 - 1)(2n_{D2}^2 + 1)}{n_{D2}^2}\phi_2 \tag{13}$$

$$Edwards : \frac{n_D - 1}{n_D} = \frac{n_{D1} - 1}{n_{D1}}\phi_1 + \frac{n_{D2} - 1}{n_{D2}}\phi_2 \tag{14}$$

From the theoretical mixing rules Lorentz–Lorenz (L–L) has a minimum deviation and Oster has the maximum deviation for all the studied systems compared to the other rules. The predicted values of ASD (Average Standard Deviation) give a complementary approach and the experimental values are well fitted with theoretical values.

APPLICATIONS

Ketones are used as solvents for insecticides, fungicides and as intermediates in the synthesis of pharmaceuticals. 2–methoxyethanol is mainly used as solvents in varnishes, dyes and resins. The heterogeneous type of interaction (C=O…H–O) is identified in the studied systems.

CONCLUSIONS

The experimental values of density (ρ), viscosity (η), refractive index (n_D) are measured at 303 K and are reported. From the experimental values the molar refraction (R_m) are calculated and interpreted. The excess properties like excess molar volume, deviation of viscosity, deviation of refractive index, excess molar refraction and excess Gibbs free energy values are calculated and reported. The excess properties are correlated with Redlich–Kister polynomial equation and the standard deviation values are calculated. From the above parameters, an interaction occurs in the form of H–bonding through the unlike molecules such as ketones (C=O) and alkoxyethanol (–OH). The interaction behavior of DMK + 2–ME is greater than other two systems. Finally it may be concluded that the heterogeneous type of interaction (C=O…H–O) is identified for all the studied systems.

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