Internal Pressure and Refractive Index of 2- Methoxyethanol with Dimethylaminoethanol and Diethylaminoethanol at Different Temperatures Vrijesh Kumar Pandey^a, Vikash Verma^a, Anjali Awasthi^b, Rajeev Kumar Tripathi^c, S.K. Shukla^c, Aashees Awasthi^{a,*}

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Abstract: The density (ρ), refractive index (n_D) and internal pressure ($P_{int.}$) of binary mixtures of 2-methoxyethanol with dimethylaminoethanol (DMAE) and diethylaminoethanol (DEAE) have been recorded at different temperatures like 303.15, 313.15 and 323.15K respectively. The experimental values of density, refractive index and internal pressure ($P_{int.}$) are used to detect the molar refraction (R_m), reduced molar free volume (V_m/R_m), and molecular radii (r). The deviations of refractive index (Δn_D), molar refractions (ΔR_m), reduced molar free volumes $\Delta(V_m/R_m)$ and internal pressures ($\Delta P_{int.}$) have been also estimated. With composition and temperature, the variations of these parameters of the mixtures are discussed in terms of molecular interactions due to physical, chemical and structural effects between the unlike molecules. The applicability of different refractive index mixing rules is tested against the experimentally measured values, and good agreement has been obtained.

Keywords: DMAE, refractive index, density, internal pressures, 2-ME.

1. Introduction

Physico-chemical properties are very important factors in chemical and engineering processes, because of their power upon the usefulness of the operations. Mass and heat transfer processes and flow operations are evident examples of the importance of the knowledge for these properties [1, 2]. Experimental thermo-physical properties are used to obtain information about the molecular level structures of liquid mixtures, as well as about the intermolecular interactions

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and structural features leading to the behavior and macroscopic properties of fluids. Intermolecular interactions are a very composite issue, and thus, experimental results and theoretical models have to be combined to elucidate the fluid structure. Literature survey [3–9] indicates that refractive index and related properties have been used to study the solute-solvent interactions in the liquid mixtures. 2-Methoxyethanol is used as a solvent for many unlike purposes such as varnishes, dyes, and resins. In organo metallic chemistry it is commonly used for the synthesis of Vaska's complex. Amino-alcohols are bi-functional organic compounds having two kinds of polar groups, hydroxyl and amino groups. They have been widely used as absorbents for separating carbon dioxide from industrial gas mixtures and solvents for important organic reactions. The binary systems of 2-ME with DMAE and DEAE are of considerable interest for investigating the intra- and inter-molecular behavior of amino-alcoholic solvent systems. To the best of our knowledge, there has been no temperature-dependent study on these systems from the view point of their refractive index behavior. This fact allows us to plan some extensive studies in this research field in order to investigate the closest interactions between the unlike molecules by examining the optical and thermodynamic parameters. In this study, we have reported a detailed investigation on the refractive indices (n_D) of binary mixtures of 2-ME with DMAE and DEAE at 303.15, 313.15 and 323.15K covering the entire miscibility range ($0 \le x \le 1$). The experimental values of density and refractive index are used to calculate the molar refractions (R_m) , reduced molar free volumes (V_m/R_m) , molecular radii (r) and internal pressure (*Pint*). The deviations of refractive index (Δn_D), molar refractions (ΔR_m), reduced molar free volumes $\Delta(V_m/R_m)$ and internal pressures ($\Delta Pint$) have been also calculated. The applicability of nine different refractive index mixing rules is tested against the experimentally measured values.

2. Experimental section

The chemical used were of analytical grade, 2-ME, DMAE and DEAE. Mixtures were stored in glass stoppered flasks to avoid contamination and evaporation. All mixtures were prepared by mass using Sartorius Electronic balance, Model-CPA225D with a precision of \pm 0.01 mg. Density measurements were made using a single-capillary pycnometer made of Borosil glass having a bulb capacity of 13.5 cm³. The precision of density measurement was \pm 0.00001 g cm⁻³. The measurements were replicated atleast three times for each measurement, and the results reported are the average values. The refractive indices of pure liquids and their binary mixtures were measured using a thermostated Abbe's refractometer. The refractometer was calibrated by measuring refractive indices of triply distilled water and toluene at known temperatures. The refractive index values were obtained using sodium D-line accurate to ± 0.0001 units. The sample mixtures were directly injected into the prism assembly of the instrument by means of an air-tight hypodermic syringe. The refractometer containing the sample to be tested was allowed to stand for around 30 minutes in a thermostatic water bath so as to minimize any possible thermal fluctuations in the refractometer. A minimum of three independent readings were taken for each composition. The temperature was maintained by an electronically controlled thermo stated water bath supplied by Orbit. The purity of solvents and reliability of experimental measurements of density and refractive index data was ascertained by comparing the data of pure liquids with the corresponding values, which were available in the literature [10-16] at 303.15, 313.15 and 323.15 K.

Results and discussion

The values of density and refractive index of pure components are listed in *Table 1*. The experimentally measured values of refractive indices (n_D) of both the binary mixtures of 2-ME with DMAE and DEAE at different temperatures are shown in *Tables 2* and *3*, respectively. The

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deviations in refractive index (Δn_D) also known as synergy of refractive index has been calculated using the following relation:

$$\Delta n_D = D_{(mix)} - (\phi_1 n_{D1} + \phi_2 n_{D1})$$
(1)

where, ϕ_1 , ϕ_2 , n_{D1} and n_{D2} represents the volume fractions and refractive indices of components 1 and 2, respectively.

The molar refraction (R_m) gives information about the presence of specific intermolecular interactions in the binary liquid mixture. The experimental molar refractions (R_m) is obtained by rolation as:

the

$$R_m = \left(\begin{array}{cc} D \\ p \\ p \end{array}\right) \left(\begin{array}{cc} 2 & M_{1x1} + M_{2x2} \\ \rho_m \\ p \end{array}\right)$$
(2)

where, ρ_m is the density of the mixture, x_1 and x_2 are mole fractions and M_1 and M_2 are the molecular weights of component 1 and 2, respectively. Deviations in molar refractions have been calculated using the relation as [17]:

$$\Delta R_m = R_m - (x_1 R_{m1} + x_2 R_{m2}) \tag{3}$$

where, R_{m1} and R_{m2} are the molar refractions of pure components 1 and 2, respectively.

The deviations of reduced molar free volumes have been calculated using the relation [17]:

$$\Delta \left(\frac{Vm}{Rm} \right) = \left(\begin{array}{c} 2 \\ D \\ -1 \end{array} \right) - \left(\begin{array}{c} x_1V_1 + x_2V_2 \\ x_1Rm_1 + x_2Rm_2 \end{array} \right)$$
(4)

The theoretical estimation of molecular radii of pure liquids can be successfully done by using the values of refractive index as:

$$r = [\{(\begin{array}{ccc} 3 & 2 \\ & \\ \end{array}) (\begin{array}{c} D \\ \end{array}) \} V_m]$$

$$4\pi N_A \quad n^2 + 2$$

$$($$

Internal pressures of binary liquid mixture can be evaluated using the well-known Buchler -Hirschfelder – Curtiss equation of state given as [18]:

$$P_{int} = 2^{1/6} RT$$

$$2^{1/6} V_m - 2r N^{1/3} V^{2/3}$$

$$A m$$
(6)

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where, r is the molecular radius and V_m the molar volume of the mixture. N_A is the Avogadro's number, T is absolute temperature in Kelvin and R is the universal gas constant. Deviations of internal pressures of binary liquid mixture have been obtained by the relation as:

$$\Delta P_{int} = P_{int} - (x_1 P_{int,1} + x_2 P_{int,2})$$
(7)

where Pint, Pint, 1 and Pint, 2 are the internal pressures of binary mixture and pure components 1 and 2, respectively.

The values of molar refraction (R_m), reduced molar free volumes ($V_m R_m$), molecular radii (r_m) and internal pressure (P_{int}) of both the binary mixtures of 2-ME with DMAE and DEAE, at different temperatures, are presented in Tables 2 and 3, respectively.

Table: 1. Experimental values of density (ρ) and refractive index (n_D) of pure liquids along with the corresponding values available in the literature at various temperatures

Liquid	T/K	ρ	$(g \cdot m^{-3})$	n _D		
		Expt.	Lit.	Expt.	Lit.	
2-Methoxyethanol	303.15	0.9554	0.9558 [26]	1.398	1.398 [27]	
	313.15	0.9462	0.9462 [26]	1.394	1.394 [27]	
	323.15	0.9369	0.9368 [26]	1.390	1.390 [27]	
2-Dimethylethanolamine	303.15	0.8791	0.8792 [22]	1.421	1.420 [22]	
	313.15	0.8707	0.8707 [22]	1.419	1.419 [22]	
	323.15	0.8621	0.8614 [25]	1.415	1.415 [28]	
2-Diethylethanolamine	303.15	0.8788	0.8761 [23]	1.433	1.428 [22]	
	313.15	0.8691	0.8668 [23]	1.426	1.419 [22]	
	323.15	0.8570	0.8573 [23]	1.418	1.401 [28]	

It is observed from Tables 2 and 3 that the experimental refractive index of 2-ME + DMAEand 2-ME + DEAE systems decreases linearly with an increase in mole fractions of 2-ME in temperature increasing.

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Table: 2. Refractive indices (n_D) , molar refractions (R_m) , reduced molar free volumes (V_m/R_p)	m),
molecular radii (r) and internal pressure (P_{int}) for 2-ME (1) + DMAE (2).	

x ₁	φ1	n _D	R _m	V_m / R_m	r	Pint
			(cm ³ mol ⁻¹)		(A ⁰)	(M Pa)
303.15 K						
0.0000	0.0000	1.4210	25.71	3.9434	2.1686	82.80
0.0981	0.0787	1.4202	25.06	3.9499	2.1502	84.56
0.1880	0.1539	1.4196	24.50	3.9549	2.1339	86.18
0.2922	0.2449	1.4188	23.85	3.9615	2.1149	88.06
0.3912	0.3354	1.4177	23.24	3.9707	2.0966	89.86
0.4976	0.4375	1.4159	22.56	3.9857	2.0762	91.83
0.6039	0.5450	1.4135	21.88	4.0065	2.0550	93.82
0.7075	0.6552	1.4104	21.20	4.0327	2.0335	95.79
0.8017	0.7606	1.4070	20.57	4.0623	2.0132	97.62
0.9009	0.8771	1.4028	19.90	4.0997	1.9911	99.57
1.0000	1.0000	1.3980	19.22	4.1433	1.9682	101.53
313.15 K						
0.0000	0.0000	1.4195	25.88	3.9557	2.1733	84.50
0.0981	0.0787	1.4184	25.22	3.9648	2.1546	86.23
0.1880	0.1539	1.4174	24.63	3.9732	2.1377	87.83
0.2922	0.2449	1.4162	23.96	3.9832	2.1182	89.70
0.3912	0.3354	1.4149	23.33	3.9942	2.0995	91.51
0.4976	0.4375	1.4130	22.65	4.0103	2.0789	93.47
0.6039	0.5450	1.4103	21.95	4.0335	2.0572	95.46
0.7075	0.6552	1.4068	21.25	4.0641	2.0351	97.42
0.8017	0.7606	1.4031	20.60	4.0970	2.0142	99.24
0.9009	0.8771	1.3987	19.92	4.1369	1.9917	101.18
1.0000	1.0000	1.3940	19.24	4.1805	1.9687	103.19
323.15 K						
0.0000	0.0000	1.4154	25.91	3.9900	2.1742	85.77
0.0981	0.0787	1.4141	25.24	4.0010	2.1554	87.50
0.1880	0.1539	1.4129	24.65	4.0112	2.1382	89.10
0.2922	0.2449	1.4116	23.97	4.0223	2.1185	90.99
0.3912	0.3354	1.4102	23.34	4.0344	2.0997	92.82
0.4976	0.4375	1.4083	22.66	4.0509	2.0790	94.82
0.6039	0.5450	1.4056	21.95	4.0747	2.0573	96.83
0.7075	0.6552	1.4022	21.25	4.1051	2.0352	98.84
0.8017	0.7606	1.3985	20.60	4.1387	2.0142	100.69
0.9009	0.8771	1.3943	19.92	4.1777	1.9918	102.69
1.0000	1.0000	1.3900	19.25	4.2185	1.9692	104.75

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Table: 3. Refractive indices (n_D) , molar refractions (R_m) , reduced molar free volumes (V_m/R_m) , molecular radii (r) and internal pressure (P_{int}) for 2-ME (1) + DEAE (2).

x ₁	φ ₁	n _D	R _m (cm ³ mol ⁻¹)	V_m/R_m	r (A ⁰)	P _{int} (M Pa)
202 15 K						
0.0000	0.0000	1.4283	34.33	3.8845	2.3879	63.70
0.0988	0.0615	1.4276	32.81	3.8901	2.3522	66.08
0.1973	0.1280	1.4273	31.36	3.8925	2.3169	68.65
0.2850	0.1923	1.4265	30.05	3.8988	2.2842	71.12
0.3906	0.2768	1.4253	28.50	3.9084	2.2442	74.32
0.4916	0.3661	1.4231	26.99	3.9262	2.2039	77.64
0.5968	0.4693	1.4195	25.38	3.9557	2.1592	81.51
0.6996	0.5818	1.4151	23.79	3.9925	2.1132	85.76
0.8036	0.7096	1.4100	22.20	4.0361	2.0651	90.58
0.9022	0.8464	1.4043	20.70	4.0862	2.0174	95.73
1.0000	1.0000	1.3980	19.22	4.1433	1.9682	64.14
313.15 K	0.0000	1 4100	04.07	0.0574	0.001.0	66 6 A
0.0000	0.0000	1.4193	34.07	3.9574	2.3819	66.64
0.0988	0.0614	1.4185	32.54	3.9640	2.3458	69.31
0.1973	0.1278	1.4182	31.09	3.9665	2.3104	71.84
0.2850	0.1921	1.4178	29.82	3.9698	2.2784	75.13
0.3906	0.2766	1.4169	28.29	3.9774	2.2389	78.56
0.4916	0.3658	1.4156	26.84	3.9883	2.1999	82.55
0.5968	0.4689	1.4123	25.25	4.0163	2.1555	86.94
0.6996	0.5814	1.4083	23.68	4.0509	2.1100	91.92
0.8036	0.7093	1.4038	22.12	4.0907	2.0626	97.24
0.9022	0.8462	1.3990	20.66	4.1341	2.0160	103.19
1.0000	1.0000	1.3940	19.24	4.1805	1.9687	63.68
323 15 K						
0.0000	0.0000	1.4017	33.14	4.1096	2.3600	66.36
0.0988	0.0614	1.40148	31.67	4.1116	2.3247	69.15
0.1973	0.1278	1.4017	30.29	4.1100	2.2904	71.79
0.2850	0.1921	1.4018	29.09	4.1084	2.2597	75.23
0.3906	0.2765	1.4024	27.69	4.1031	2.2230	78.85
0.4916	0.3657	1.4025	26.35	4.1024	2.1864	83.05
0.5968	0.4689	1.4005	24.85	4.1204	2.1440	87.67
0.6996	0.5814	1.3980	23.38	4.1433	2.1009	92.91
0.8036	0.7093	1.3951	21.91	4.1702	2.0560	98.51
0.9022	0.8462	1.3926	20.56	4.1937	2.0129	104.75
1.0000	1.0000	1.3900	19.25	4.2185	1.9692	63.68

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Table 2 and *3* shows that the refractive index decreases linearly as a function of increasing on concentration of 2-ME of 2-ME + DMEA and 2-ME + DEEA systems, respectively and also decreases with increase in temperatures. These associated molecules are bulkier than 2-ME molecules; this may cause steric hindrance, resulting in the weakening of the intermolecular forces. This is probably the reason for the decrease in the velocity above 0.4976 and 0.4916 mole fractions for respective systems [23, 17]. The presence of intra-molecular hydrogen bonding in pure state of 2-ME and DMAE/DEAE molecules may reduce the possibility of intermolecular interaction with the unlike molecules, which occur only via weak dipolar interactions.

Figures 1 (a, b) exhibit that Δn_D values are positive and nonlinear over the entire composition range at all temperatures for both the systems. The Δn_D value shows maximum at $x_1 \sim 0.5726$ for (2-ME + DMAE) and $x_1 \sim 0.5054$ for (2-ME + DEAE) binary system. It is well known that the refractive index deviation depend on several energetic and structural effects [20, 21]. The positive deviations Δn_D (on volume fraction dependence basis) is considered due to the presence of significant interactions in the mixtures, whereas negative deviations Δn_D indicates weak interactions between the contributing components of the mixtures [8, 17, 22]. The observed variations of Δn_D values indicate the presence of significant interactions in these mixtures. The extent of the positive deviations Δn_D from linear dependence on composition is in order: 2-ME + DEAE > 2-ME + DMAE. Both, 2-ME and aminoalcohol molecules are polar and strongly selfassociated liquids having inter- and intra-molecular hydrogen bonding. In a reliable abstraction, a negative contribution to the Δn_D values should be expected. But contrary to our expectation, the positive trends are observed in Δn_D values for 2-ME + DMAE/DEAE mixtures. On this basis, a positive contribution to Δn_D should be expected on mixing the liquids when favorable interactions between groups occur. For 2-ME and DMAE/DEAE systems, the positive Δn_D values suggest that the aminoalcohols - 2-ME interaction predominate those between

aminoalcohols molecules. It is suggested that mixing of 2-ME with aminoalcohols would induce mutual dissociation of the hydrogen-bonded structures present in pure liquids with subsequent formation of (new) hydrogen bonds with proton acceptor oxygen atom of 2-ME molecules and hydrogen atom of aminoalcohols molecules. The sign and magnitude of Δn_D also varies with the structural characteristics of the liquid components arising from the geometrical fitting of one component into the structure of other component due to difference in the molecular size and shape of the components. If the unlike molecules have almost the same molar volumes, this effect should be insignificant. However, even if slight difference in the free volume between different species could facilitate the penetration of one component into the other, and as the difference of the free volumes of the two pure species increases, the more positive should be the contribution to Δn_D .

The values of refractive index (Tables 2 and 3) and its deviation (Figure 1) are found to be higher for 2-ME + DEAE system than 2-ME + DMAE system. The refractive index of a compound describes its ability to refract light as it moves from one medium to another and thus, the higher the refractive index of a compound, the more the light is refracted. As stated by Deetlefs et al. [10], the refractive index of a substance is higher when its molecules are more tightly packed or in general when the compound is denser. Hence, a perusal of Tables 2 and 3 reveals that refractive index is higher for 2-ME + DEAE than 2-ME + DMAE system indicating to the fact that these molecules are more tightly packed in the solution.

The strength of intermolecular interactions between 2-ME + DMAE/DEAE molecules is in order: 2-ME + DEAE > 2-ME + DMAE. The position of Δn_D maxima does not change with an increase in temperature.

Figure 2 (*a*, *b*) show the deviation in molar refraction (ΔR_m) with mole fractions (x₁) of 2-ME. The molar refraction is a sensitive function of wavelength, temperature and solution composition. A negative non-linear trend is observed in ΔR_m values, for both the binary liquid

mixtures, over the entire concentration range. The negative value of ΔR_m suggests the presence of intermolecular interactions between the components of the liquid mixtures. The values of ΔR_m slightly decrease with increase in the temperature [19].

Figure 3 (*a*, *b*) present the deviations of reduced molar free volume versus mole fraction of FA. The values of deviations of reduced molar free volume $\Delta(V_m/R_m)$ are negative and nonlinear over the whole concentration range, at all temperatures, for both the systems. Brocos et al. [17] stated that the molar refraction deviation function must be calculated on mole fraction basis and refractive index deviation function on volume fraction basis, which makes it directly interpretable as a sign-reversed measure of the deviations of reduced free volumes from ideality. We have evaluated all the deviation functions, Δn_D , ΔR_m and $\Delta(V_m/R_m)$ and found that deviations in refractive index Δn_D have the opposite sign to the deviation in reduced free molar volume, $\Delta(V_m/R_m)$, for the present binary system. Negative values of $\Delta(V_m/R_m)$ show the minima at the same concentrations where positive values of Δn_D maxima occur. Negative values of $\Delta(V_m/R_m)$ show strong molecular interactions between the components of the liquid mixtures [19].

Figure 4 (a, b) show the trends of deviation of internal pressure versus mole fraction of 2-ME, for both the liquid mixtures, respectively. The internal pressure is defined as the energy required vaporizing a unit volume of a substance. The values of P_{int} increase with increase in mole fractions of 2-ME. The internal pressure for pure alcohols decreases with increasing chain length [11]. The value of internal pressure is found to be greater for 2-ME + DEAE than 2-ME + DEAE system. The deviations of internal pressure (ΔP_{int}) are negative and non-linear in nature over the complete composition range. The effect of temperature is not very significant for ΔP_{int} values [19, 12].

Figure: 1(a, b). Plot of Δ*nb* as a function of mole fraction of FA (x1) at (0) 303.15 K, (·) 313.15 K, (▲) 323.15 K.



igure: 2(a, b). Plot of Δ*R*^{*m*} as a function of mole fraction of FA (x₁) at (o) 303.15K, (·) 313.15 K, (▲) 323.15 K.



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		1						1	.0
	х							0.6	0.8
	0.0	0.2	0.4	0.6	0.8	1.0	0.0	0.2	0.4
-0.4						-2.0			
-0.4									
-0.3							-1.5		
-0.3									
-0.2							-1.0		

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Figure: 3(a, b). Plot of Δ*P*_{int} as a function of mole fraction of FA (x1) at (0) 303.15 K, (·) 313.15 K, (▲) 323.15 K.



Figure: 4(a, b). Plot of $\Delta(V_m/R_m)$ as a function of mole fraction of FA (x1) at (o) 303.15 K, (·) 313.15 K, (**A**) 323.15 K.

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4. Conclusions

The refractive indices (n_D) of binary mixtures of 2-methoxyethanol with aminoalcohols have been measured over the whole composition range at 303.15, 313.15 and 323.15 K. The variations of refractive index and related parameters with composition and temperature of the mixtures are discussed in terms of molecular interactions due to physical, chemical and structural effects between the unlike molecules. The outcome were analyzed in terms of the molecular characteristics of the interacting molecules and found to exhibit an extensive hydrogen bonding between 2-methoxyethanol with aminoalcohol molecules in the binary liquid mixtures as in order 2-ME + DEAE > 2-ME + DMAE.

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