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RESEARCH ARTICLE **OPEN ACCESS**

PREDICTION OF REFRACTIVE INDICES OF SOME BINARY LIQUID MIXTURES OFCONTAINING 1,3-DIOXOLANE WITH ALKANOLS (C5-C10) AT 298.15K

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ABSTRACT

Density (ρ), viscosity (η) and refractive indices (n) of the binary mixture between 1,3-Dioxolane with pentanol. hexanol, heptanol, octanol, nonanol and decanol for the entire concentration range have been measured at 298.15K, furthermore, deviation in refractive indies from ideal mixture for this mixture has also been evaluated.The determination of light refractive index is an important step in the characterization of liquid mixtures with different industrial applications. Through it, the purity of their substances can be assessed and, together with density andultrasonic velocities, it has a significant role in characterizing and understanding the thermodynamic properties of the liquids.The results are presented in terms of excess refractive indices from the experimental values.The deviations from ideality of the refractive indies are explained on the basis of molecular interaction between the component molecules in these binary liquid mixtures.

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INTRODUCTION

The refractive index of light is a useful parameter for characterizing liquids that are widely applied in the industry, such as oils, waxes, sugar syrups, etc. It is a fundamental physical property that measures the speed of light in a material and characterizes its optical properties [1]. It has been used for years for accurate identification and characterization of pure liquids and mixtures [2]. A number of authors [3, 4] have noted that the refractive index, density and viscosity are essential for characterizing and understanding the thermodynamic properties of liquids. The molecular interaction in a fluid mixture can also be estimated from the refractive index and the density of its pure components [5, 6]. The refractive index is useful in the indirect measurement of density and salinity and in the detection of structural properties of liquid-liquid mixtures. Its application has also led to the development of alternatives to fuel substitutes, additives and oil treatment with chemicals [7]. By using mixing rules [8], the composition of an unknown mixture as well as the presence of molecular interactions in binary mixtures can be determined. The refractive indices of pure, binary and multicomponent fluids can readily be measured directly using refractometers. However, there are times when the experimental values are not available and it is desirable to estimate the refractive index of binary or multicomponent liquids from the pure components by usingmixing rules.Refractive index is useful in the indirect measurement of density and salinity and in the detection of structural properties of liquid-liquid mixtures. Its application has also led to the development of alternatives in fuel substitutes, additives, and treatment of oils with chemicals used mixing rule to determine the composition of an unknown mixture and the presence of molecular interactions in binary mixtures. Several researchers have noted that deviation between theoretical and experimental values of the refractive indices of mixtures can be reduced by considering the concept of excess volume. Refractive index is an important property that is often used to characterize materials, which indicates the ratio of the velocity of light in vacuum to the velocity of light in the material [9]. It is a thermodynamic property, which depends on temperature, pressure, and wavelength [10]. Compared to other bulk properties, the measurement of refractive index is relatively fast and convenient; therefore, it is often used to gain molecular insights between the component molecules. The study of molecular interaction has attracted the attention of many workers. In recent paper, ultrasonic technique has become a powerful tool in providing valuable information regarding the molecular behavior of liquids. Excess properties are the measure the different type of attractions. The various type of molecular interaction that may operate between molecules of different type are dispersion forces, charge transfer, hydrogen bonding, dipole-dipole and dipole-induced dipole interaction. In any given system more than one type of molecular interaction present. The interaction of alkanols with 1,3-Dioxolaneis interesting due to the acidic nature. The O-H bonds in alcohols are polar and allow the release of hydrogen atom as proton. The order of acidity in alcohols is:

Primary alcohol > Secondary alcohol > Tertiary alcohol

Keeping this in view, four binary liquid mixturespentanol. hexanol, heptanol, octanol, nonanol and decanol with 1,3-Dioxolane (Cyclic ether) were selected to study their molecular interactions through their acoustical behavior.

EXPERIMENTAL PROCEDURE

Chemicals: The source and purity of the chemical compound are shown in table-1.The substances density, refractive index and ultrasonic velocity is compared with the literature data (Table-2) to ascertain the purity, and a good agreement between the experimental data and literature data [11-22] was observed.

Apparatus and procedure: All six binary liquid mixtures were prepared by weighing appropriate amounts of pure liquids on a digital electronic balance (Citizen Scale (I) PVT. LTD. Mumbai, India.) with a precision ± 0.1. The experimental uncertainty in mole fractions did not exceed ± 0.0005. All the solutions were prepared by mass ratios and stored in the air-tight stopper measuring flasks.

Refractive Indices

Refractive indices of pure liquids and liquid mixtures were measure using white light by an Abbe refractometer (Model R- 8 M/S Mittal Enterprises, New Delhi). Refractometer was calibrated with kept constant at 298.15 ± 0.03 K by circulating water of the thermostate with the help of pump through both the prism boxes of the refractometer. Refractive indices of liquid were measured after attainment of constant temperature. An average five measurements was made for each sample. The measured values of refractive indices of pure 1,3-dioxolane with pentanol, hexanol, heptanol, octanol, nonanol and decanol at 298.15K were 1.402, 1.4042, 1.4155, 1.4267, 1.4357, 1.4368, 1.4398 respectively, which compare well with the corresponding literature values.

Table 1. CAS Registry Number, Mass Fraction Purity of the chemicals

Component	Formula	CAS Reg. No.	Supplier	Mass Fraction Purity (%)	Method Purity analysis method
1,3-Dioxolane	$C_3H_6O_2$	646-06-0	CDH Delhi	99.7	Chromatography by the supplier
Pentanol	$C_5H_{12}O$	71-41-0	CDH Delhi	99.7	Chromatography by the supplier
Hexanol	$C_6H_{14}O$	111-27-3	CDH Delhi	99.5	Chromatography by the supplier
Heptanol	$C_7H_{16}O$	111-70-6	CDH Delhi	99	Chromatography by the supplier
Octanol	$C_8H_{18}O$	111-87-5	CDH Delhi	99.7	Chromatography by the supplier
Nonanol	$C_9H_{20}O$	143-08-8	CDH Delhi	99	Chromatography by the supplier
Decanol	$C_{10}H_{22}O$	112-30-1	CDH Delhi	99	Chromatography by the supplier

Table 2. Comparison of Experimental and Literature density (ρ), Viscosity (η) and refractive index (*n*) of pure Components with Available Literature Values at T = 298.15K

Compound	ρ (g.cm^{-3})		η (mPa s)		n	
	This work	**Literature**	**This work**	**Literature**	**This work**	**Literature**
1,3-Dioxolane	1.0616	1.0577[15]	0.5885	0.5878[16]	1.402	1.3905[22]
		1.0586[17]		0.5873[16]		1.3979[22]
Pentanol	0.8124	0.8108[11]	3.3978	3.5411[14]	1.4042	1.4053[20]
		0.8107[11]		3.5424[14]		1.4081[20]
Hexanol	0.8176	0.8187[11]	4.6091	4.5924[14]	1.4155	1.4154[21]
		0.8152[13]		4.5932[14]		1.4161[21]
Heptanol	0.8196	0.8187[11]	5.9066	5.9943[14]	1.4267	1.4226[20]
		0.8197[17]		5.9943[19]		1.4224[21]
Octanol	0.8236	0.8216[11]	7.1508	7.6605[14]	1.4357	1.4283[20]
		0.8218[11]		7.5981[14]		1.4264[20]
Nonanol	0.8248	0.8244[13]	8.9258	8.965[15]	1.4368	1.4319[21]
		0.824224[13]		9.715[18]		1.4318[20]
Decanol	0.8292	0.8267[13]	11.8027	11.825[15]	1.4398	1.4346[23]
		0.8264[17]		11.817[16]		1.4358[21]

Measurements

Density: Densities of pure components and liquid-liquid mixtures were measured with a 25-ml specific gravity bottle by relative measurement method with an accuracy of ± 0.01 kg.m^{-3}. The specific gravity bottle with the experimental mixture was immersed in the temperature-controlled water bath (MSI Goyal scientific, Meerut, U.P. India.), operating in the temperature range of -10^{0}C to 85^{0}C with an accuracy ± 0.1^{0}C. Double distilled water used for the calibration of the specific gravity bottle. At least three times for each composition in experimental were generally repeated and the results were treatment.

Viscosity: The viscosities of pure liquids and their binary mixtures were measured by using a Ostawald's viscometer. The viscometer was calibrated with doubly distilled water and benzene, liquid was allowed to stand for about 30 minutes in a thermostatic water bath so that the thermal fluctuations in viscometer were minimized. The accuracy in viscosity data was ± 0.0005 mPa.s. The flow time of pure liquids and liquid mixtures were repeated for five times. The efflux Time was measured with an electronic stopwatch (Racer) with a time resolution (± 0.015), and an average of at least five flow time readings was taken. Glass stopper was placed at the opening of the viscometer to prevent the loss due to evaporation during measurements. The measured values of viscosities of pure 1,3-dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol compare well with the corresponding literature values.

The mixtures were prepared by mixing known volumes of the pure liquids in air tight stoppered bottles. The weights were taken digital electronic balance (Citizen Scale (I) PVT. LTD. Mumbai, India) with a precision ± 0.1.

Theoretical : The excess viscosity (η^E) is evaluated from the experimental values of viscosity (η) for component liquid and their binary mixtures by

$$\eta^E = \eta_{1,2} - \eta_{1,} X_1 + \eta_2 \; X_2 \qquad (1)$$

Where $\eta_{1,2}$ is viscosity in the mixture and $\eta_{1,}$ $\eta_{2,}$ $X_{1,}$ X_2 are the viscosity and mole fractions respectively of the component liquid 1 and 2.

The experimental values of refractive indices (n_{mix}) and density (ρ_{mix}) at 298.15 K with mole fraction of second components for all binary mixtures of 1,3-Dioxolane and second components are reported in Table 3. Values of deviation in refractive indices (Δn) or (n^E) for all the systems studied are evaluated using experimental values of refractive indices for pure liquids and liquid mixtures employing following equation.

$$\Delta n = n_{\text{mix}} - n_{\text{ideal}} \qquad (2)$$

where n_{mix} and n_{ideal} are refractive indices of actual binary mixture and ideal binary mixtures. The refractive index deviation, Δn, has been calculated on a mole fraction basis and n_{ideal} has been evaluated by Equation (3).

$$n_{ideal} = X_1 n_1 + X_2 n_2 \quad (3)$$

where X_1, X_2 and n_1, n_2 are the mole fraction and refractive indices of the components 1 and 2 respectively. Values of deviation in refractive indices, Δn, with compositions of the binary mixtures studied at experimental temperature are also recorded in Table 3.

The excess value of viscosity, refractive index related parameters has been calculated by using the following relation

$$A^E = A_{exp.} - (X_1 A_1 + X_2 A_2) \quad (4)$$

Where A represents the parameter such as intermolecular free length, molar volume, isentropic compressibility, viscosity and internal pressure and X_1 $and X_2$ is the mole fractions of components whose parameters.

RESULTS AND DISCUSSION

Electromagnetic theory of light is the basis of these mixing rules of refractive index which treats the molecules as dipoles or assemblies of dipoles by an external field. In the present work, an attempt has been made to study the validity of four mixing rules for predicting the refractivity of eight binary mixtures comprising 1,3-dioxolane as the first component and pentanol, hexanol, heptanol, octanol, nonanol and decanol as the second component, over the entire mole fraction range of 1,3-dioxolane in their respective systems. The experimental values of densities (ρ), viscosity (η) and refractive indices (n) at 298.15K with mole fraction of second components are reported in table-3.

Excess Viscosity (η^E): The excess viscosity (η^E) data of all the binary mixtures of 1,3-Dioxolane with pentanol, hexanol, heptanol, octanol, nonanol and decanol are graphically presented Figures 1 at 298.15 K. An examination of curves in Figure 1 shows that the values of excess viscosity (η^E) data for 1,3-Dioxolane with pentanol, hexanol, heptanol, octanol, nonanol and decanol are negative over the entire composition range at 298.15 K**.** The measurement of viscosity in binary liquid mixture provides some reliable information in the study of molecular interaction. Table- 3 shows that the viscosity decrease with increase in concentration of 1,3-Dioxolane molecule. More insight about molecular interaction [24] can be obtained by excess viscosity (η^E) values.

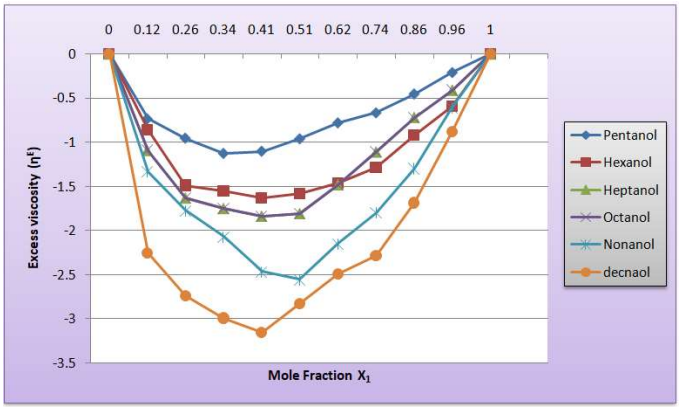

Figure 1. Curves of excess viscosity η^E against the mole fraction of 1,3-dioxolane x_1, for the binary mixture (1,4-dioxane (1) + Alkanols(2)) at 298.15K. The solid lines represent the values calculated from the Redlich−Kister equation

According to Fort and Moore, [25-26] the excess viscosity gives the strength of the molecular interaction between the interacting molecules. The excess value of viscosity at the six binary mixtures 1,3-Dioxolane + Pentanol, 1,3-Dioxolane + Hexanol, 1,3-Dioxolane + Heptanol, 1,3-Dioxolane + Octanol, 1,3-Dioxolane + Nonanol and 1,3-Dioxolane + Decanol at the 298.15 K are reported in Table- 3. The Figure -1 represents the variation of excess viscosity (η^E) is found to be negative for all six binary liquid mixtures over the entire composition range at the 298.15 K. Which suggest the presence of weak intermolecular interactions. For systems where dispersion, induction and dipolar forces are operating, the values of excess viscosity are found to be negative, whereas the existence of specific interaction leading to the formation of complexes in mixtures tends to make positive. The excess viscosity is negative through the whole range of concentration in all the studied systems. The large negative values of excess viscosity for all systems can be attributed to the presence of dispersion, induction and dipolar forces between the components.

The negative excess viscosity (η^E) for all the six binary liquid mixtures (1,3-Dioxolane + Pentanol, 1,3-Dioxolane + Hexanol, 1,3-Dioxolane + Heptanol, 1,3-Dioxolane + Octanol, 1,3-Dioxolane + Nonanol and 1,3-Dioxolane + Decanol) studied are indicative of the predominance of dispersion forces and further their magnitudes increase from pentanol to decanol (C_5-C_{10}), hence suggesting an increase in dispersion forces in the same order . Alcohols are good solvent that can dissolve both the polar and non-polar components. The hydrophilic –OH group of alcohols can dissolve the polar whereas the short hydrophobic hydrocarbon group can dissolve the non-polar. Alcohols are strongly self-associated liquids with a three dimensional network of hydrogen bonds and can be associated with any other group having some degree of polar attraction. The associative alcohols molecule act as proton donor enabling hydrogen bonding with 1,3-Dioxolane molecule. In the system studied, the complex formation is likely to occur between $H^{\delta+}$ of alcohol and $O^{\delta-}$ of ether group of 1,3-Dioxolane. Hence in the present study there is existence of solute-solvent interactions. The algebraic values of excess viscosity for binary mixtures of 1,3-Dioxolane with Pentanol, Hexanol, Heptanol, Octanol, Nonanol and Decanol fall in the order:

Decanol < Nonanol < Octanol < Heptanol < Hexanol < Pentanol

In the alkanol mixture, the 1,3-Dioxolane is completely dissolved and so no changes of hydrogen bond raptures and only the interaction with the 1,3-Dioxolane ring and the active group of alkanols, which are mostly dispersive in nature. The increase in mole fraction of 1,3-Dioxolane increase the net dispersive interaction and hence the velocity continuously increases as observed. As the mole fraction of 1,3-Dioxolane increases, the hydrogen bond repture of the boat form is of considerable extent and they leads to additional dipole type interaction. 1,3-Dioxolane being non-polar the predominant dispersive type interactions with temporary dipolar type are existing as a net result of intermolecular forces in all systems.

Excess Refractive Indices*:* Experimental results for refractive indices, sound velocity and density at the temperature 298.15K are summarized in table-3. For each mixture, the refractive indices were fitted with a Redlich-Kister equation. The excess refractive indices (n^E) are plotted as a function of the mole fraction of 1,3-Dioxolane in figure-2. Figure-2 shows that for binary mixtures (1,3-dioxolane + pentanol, 1,3-dioxolane + hexanol, 1,3-dioxolane + heptanol, 1,3-dioxolane + octanol, 1,3-dioxolane + nonanol and 1,3-dioxolane + decanol) excess refractive indices (n^E) is negative over the whole range of mole fractions and becomes. Results of excess and deviation properties provide insights into the negative deviation from the ideal mixture, indicating which interactions are prevalent in the systems under study. When mixing pure components deviations from the ideal mixture can be smaller or larger depending on the formation of different types of intermolecular bonds and interactions and the packing ability. Dipole-dipole [26-27] and H-bond interactions between hetero molecules in the mixture lead to negative excess molar volumes, as well as structural effects such as favorable interstitial accommodation and efficient packing.

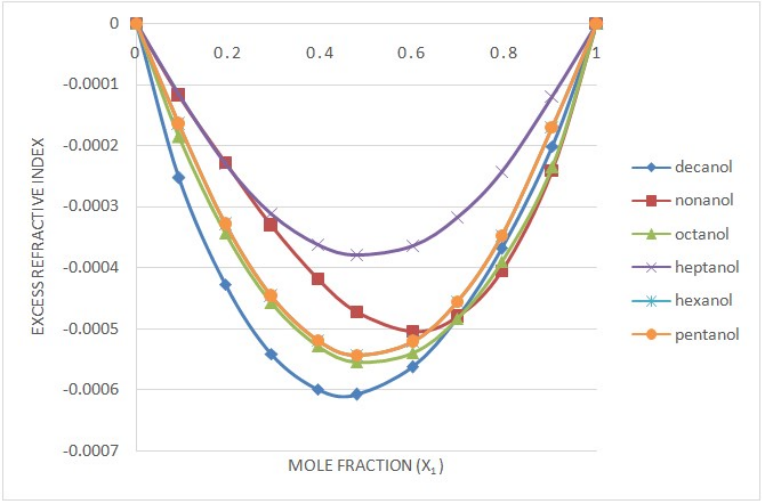

Figure 2. Curves of excess refractive indices (n^E) against the mole fraction of 1,3-dioxolane x_1, for the binary mixture (1,3-Dioxolane (1) + Alkanols(2)) at 298.15K. The solid lines represent the values calculated from the Redlich−Kister equation.

Table 3. Experimental Values of density (ρ), viscosity (η) and refractive indices (n) derived parameters excess viscosity (η^E) and refractive indices (n^E) for the binary mixtures of 1,3-Dioxolane (1) + Alkanols (2) at 298.15K

Mole fraction 1,3-Dioxolane (x_1)	Density (ρ) $g.cm^{-3}$	Viscosity (η) mPa.s	Refractive indices (n)	Excess viscosity (η^E) mPa.s	Excess refractive indices (n^E)
1,3-Dioxolane + Pentanol					
0	0.8124	3.3978	1.40421	-	-
0.0939	0.8276	2.3973	1.3994	-0.7367	-0.0035
0.1942	0.8436	1.8970	1.3983	-0.9552	-0.0032
0.2941	0.8640	1.4437	1.3971	-1.1280	-0.0031
0.3942	0.8836	1.1866	1.3963	-1.1038	-0.0025
0.4787	0.9068	1.0904	1.3953	-0.9627	-0.0023
0.5999	0.9316	0.9311	1.3940	-0.7815	-0.0020
0.6972	0.9596	0.7717	1.3928	-0.6675	-0.0018
0.7928	0.9876	0.7171	1.3917	-0.4535	-0.0016
0.9035	1.0260	0.6489	1.3909	-0.2108	-0.0009
1.0000	1.0616	0.5885	1.3905	-	-
1,3-Dioxolane + Hexanol					
0	0.8176	4.6091	1.4155	-	-
0.0912	0.8252	3.3826	1.4128	-0.8597	-0.0004
0.1955	0.8432	2.3306	1.4101	-1.4925	-0.0005
0.2923	0.8584	1.9839	1.4076	-1.4500	-0.0006
0.3982	0.8792	1.5720	1.4049	-1.4361	-0.0006
0.4942	0.8992	1.3059	1.4025	-1.3162	-0.0006
0.6059	0.9264	1.0343	1.3997	-1.1387	-0.0007
0.6976	0.9508	0.9131	1.3973	-0.8912	-0.0008
0.8018	0.9836	0.7680	1.3946	-0.6174	-0.0009
0.8914	1.0168	0.7304	1.3923	-0.2947	-0.0009
1.0000	1.0616	0.5885	1.3905	-	-
1,3-Dioxolane + Heptanol					
0	0.8196	5.9066	1.4267	-	-
0.0928	0.8304	4.3181	1.4232	-1.0951	-0.0001
0.1905	0.8412	3.2577	1.4196	-1.6358	-0.0002
0.2939	0.8592	2.5895	1.4158	-1.7541	-0.0003
0.3894	0.8740	1.9926	1.4123	-1.8431	-0.0003
0.4818	0.8916	1.5315	1.4089	-1.8128	-0.0004
0.6021	0.9184	1.2190	1.4045	-1.4856	-0.0004
0.6952	0.9420	1.0959	1.4011	-1.1136	-0.0004
0.7892	0.9756	0.9903	1.3976	-0.7193	-0.0005
0.9006	1.0156	0.7057	1.3935	-0.4114	-0.0006
1.0000	1.0616	0.5885	1.3905	-	-
1,3-Dioxolane + Octanol					
0	0.8296	7.1508	1.4357	-	-
0.0885	0.8296	5.6095	1.4315	-1.0951	-0.0002
0.1967	0.8464	3.9321	1.4266	-1.6358	-0.0002
0.2998	0.8560	3.2616	1.4219	-1.7541	-0.0002
0.3902	0.8712	2.4284	1.4178	-1.8431	-0.0003
0.4963	0.8876	1.9058	1.4130	-1.8128	-0.0003
0.6008	0.9140	1.3631	1.4082	-1.4856	-0.0003
0.6925	0.9340	1.1376	1.4040	-1.1136	-0.0004
0.7975	0.9676	0.9141	1.3992	-0.7193	-0.0005
0.8940	1.0104	0.7652	1.3948	-0.4114	-0.0005
1.0000	1.0616	0.5885	1.3905	-	-

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1,3-Dioxolane + Nonanol					
0	0.8248	8.9258	1.4368	-	-
0.0876	0.8336	6.8601	1.4326	-1.3354	-0.0001
0.1913	0.8404	5.8531	1.4277	-1.4778	-0.0002
0.2942	0.8504	4.4022	1.4229	-2.0708	-0.0003
0.3963	0.8692	3.1558	1.4181	-2.4659	-0.0004
0.4959	0.8844	2.3340	1.4134	-2.4573	-0.0004
0.6050	0.9092	1.7321	1.4083	-2.1496	-0.0005
0.6947	0.9332	1.3334	1.4041	-1.8005	-0.0005
0.7993	0.9648	0.9642	1.3992	-1.2976	-0.0006
0.9013	1.0084	0.8031	1.3944	-0.6083	-0.0007
1	1.0616	0.5885	1.3905	-	-
1,3-Dioxolane + Decanol					
0	0.8292	11.8027	1.4398	-	-
0.0881	0.8364	8.5615	1.4299	-2.2532	-0.0307
0.191	0.8396	7.8207	1.4219	-1.8401	-0.0238
0.2921	0.8560	5.5340	1.4144	-2.9930	-0.0176
0.3937	0.8672	4.2319	1.4082	-3.1558	-0.0123
0.4956	0.8824	3.4173	1.4032	-2.8276	-0.0083
0.604	0.9076	2.5370	1.3987	-2.4923	-0.0052
0.7129	0.9308	1.5262	1.3952	-2.2819	-0.0029
0.7983	0.9616	1.1637	1.3926	-1.6867	-0.0015
0.8971	1.0040	0.8623	1.3904	-0.8801	-0.0001
1	1.0616	0.5885	1.3905	-	-

On the other hand, disruption of dipole-dipole interactions and intermolecular hydrogen bonds between molecules of the same substance when put in a mixture. Alcohols are organic compounds with a highly polar hydroxyl group, which enables interconnection of molecules with strong hydrogen bonds and construction of associated liquids. In mixtures with another organic component, alcohol molecules tend to dissociate from the aggregates and form hydrogen bonds with molecules of another kind. The degree of dissociation from the aggregate depends on the affinity towards the proton of a functional group of another molecule, a proton acceptor. 1,3-dioxolane is also a polar compound, which acts as H-bond acceptor, and therefore there is a possibility of intermolecular interactions in mixtures with alcohols. Depending on the strength of these two types of interactions, disruption of bonds between molecules of alcohol and formation of new ones between molecules in the mixture, negative deviation [28-31] from ideal behaviour can be noticed. Another possibility to interact are dipole-dipole interactions. Since the dipole moment of 1,3-dioxolane is higher than the ones for investigated alcohols, it can be concluded that these interactions are stronger between molecules of acetate than those with alcohol molecules. On the basis of the above, it seems that molecular interactions play an important role in governing the deviation in refractive indices for the binary systems studied in the present investigation. It may also be concluded that there is a gradual change in molecular interactions between the components of binary mixtures.

CONCLUSIONS

The present investigations show that the interactions resulting in the interstitial accommodation of 1,3-dioxolane in to alcohols are the predominant factor over dipole-dipole and dipole induced-dipole interaction between the unlike molecules. The existence of molecular interaction in the mixture is in the order:

Pentanol < Hexanol < Heptanol < Octanol < Nonanol < Decanol

It may also be concluded that there is a gradual change in molecular interactions between the components of binary mixtures. The values of excess refractive indices for all the systems studied are negative.

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