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Experimental and Theoretical Insights to Physicochemical Properties of Aqueous Solutions of 1, 2-Ethanediol and 1, 2, 3-Propanetriol at Different Temperatures

Sanghamitra Pradhan [1], Soumya Ranjan Mishra [1], Sujata Mishra [1*]

[1] Department of Chemistry, Institute of Technical Education and Research, Siksha'O'Anusandhan Deemed to be University, Khandagiri square, Bhubaneswar, 751030, Odisha, India
* Correspondence: drsujatamishra97@gmail.com; sujatamishra@soa.ac.in;

Scopus Author ID 57203777707
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Abstract: Water is designated as "Universal solvent" due to its physical and chemical attributes. Water becomes attracted to different types of molecules due to the polar arrangement of oxygen and hydrogen atoms having partial negative and positive charges. The density (ρ), ultrasonic velocity (U), viscosity (η) and refractive index (n_D) of the aqueous solutions of 1, 2-ethanediol / 1, 2, 3-propanetriol have been determined at three different temperatures. Derived properties such molar volume (V), isentropic compressibility (β_s), acoustic impedance (Z) and free length (L_f) have been calculated using density, ultrasonic velocity and refractive index values at the measured temperatures. To evaluate various, inter and intra-molecular associations present in the systems, the excess properties (V^E, βs^E, Z^E, $L_f{}^E$, η^E and Δn_D) are estimated. The excess values obtained experimentally have been fitted to the Redlich -Kister polynomial equation. Multiple linear regression and mixing rules have been adopted to calculate the viscosity and refractive index for the aqueous solutions at 298.15, 308.15 and 318.15K.

Keywords: Water; 1, 2-ethanediol; 1,2,3-propanetriol; Ultrasonic velocity; Viscosity; Refractive index.

1. Introduction

Studies on physicochemical properties of aqueous solutions of hydrogen bonded compounds are vital since they are instrumental in depicting molecular interactions present in the systems [1]. With the purpose of having clear insight into the physicochemical properties and nature of molecular interactions between the components, investigations on density, viscosity, refractive index is essential [2]. Excess properties are also of interest in understanding the nature of molecular interactions present between the unlike components [3-8]. These outcomes find direct applications in the chemical and biochemical industries [9]. Normally in the reaction systems, methanol, ethanol, 1,2-ethanediol and 1,2,3-propanetriol are used which are highly soluble in water and this is so because hydrogen bonds are formed between water molecules and the OH group of these substrates. The role of systems containing hydrogen bonding is fascinating due to their importance in various biological, physical and chemical processes [10]. Due to the significant role of water in biological systems, an improved understanding of water structuring is advantageous [11]. In this respect, the complex system dynamics like hydrogen-bonded liquids and their mixtures is these days one of the most active areas of research. 1,2,3-propanetriol a recognized cryo-protectant operates through stabilizing

living cells and tissues were lower than cooling to subzero temperatures, as well as hold back the creation of ice. The thermal performance of 1, 2, 3-propanetriol is due to the existence of an extended hydrogen-bond network. In comparison to other solvents 1, 2-ethanediol is considered to be safe because it has a high flash point and low flammability limit range relative to methanol [12]. It is used in the internal combustion engine of the automobiles as coolant. To examine the role of methanol as modifier in extraction systems, physicochemical properties such as volume, viscosity and acoustic properties have been studied [13]. Mixture of 1,2,3-propanetriol and water is also commonly used for storing materials that have biomedical applications [14]. The speed of sound and viscosities of poly ethylene glycol solutions have been studied by Pal and Singh [15]. Vural et al. have studied the excess molar volume and refractive index of binary mixtures of glycerol + methanol and glycerol + water at 298.15 K and 303.15K [16]. Thermo physical behaviour of water + 1,2-ethanediol at 283.15K, 293.15K, 303.15K and 313.25K has been studied by Tsierkezos and Molinou[17]. Dehydration of 1-propanol by extractive distillation method with 1,2-ethanediol has been carried out by Pla-Franco et al [18]. The dielectric behaviour of 1,2-ethanediol and propylene 1,2-ethanediol has also been reported [19]. Chaube et al. have investigated the dielectric relaxation studies of 1,2-ethanediol mono phenyl ether and methanol with the help of time domain reflectometry [20]. The dielectric studies of 1,2-ethanediol mono phenyl ether with 1-butanol have also been carried out at different temperatures by Chaube and Rana [21]. Kinart et al. have studied the thermodynamic and physicochemical properties of binary mixtures of sulfone with 1,2-ethanediol s at 303.15K [22]. Delgado et al have investigated the solubility of sulfamethazine in aqueous propylene 1,2-ethanediol mixtures at varied temperatures [23].

Because of the practical significance of these components, the present work has been designed. In this paper, measured density, viscosity, refractive index and acoustic properties for aqueous solutions of 1, 2-ethanediol /1, 2, 3-propanetriol at 298.15K, 308.15K and 318.15K have been investigated. The excess values for these properties have been calculated from the experimental data and they provide important information for analyzing the type of intermolecular organization in the systems. By using this information, the influence of carbon atom numbers as well as hydroxyl group on the physicochemical properties of aqueous solutions diol and triol can be examined.

2. Materials and Methods

The chemical reagents used in this study are 1, 2-ethanediol ($C_2H_6O_2$) / 1, 2, 3-propanetriol ($C_3H_8O_3$) are chosen as solute and are purchased from Merck having purity more than 99.9 percent. Double distilled water (H_2O) is taken as a solvent for this investigation. The loss of volume of the reagents has been checked by storing them in stoppered bottles. The purity has been checked by comparing the experimental density and ultrasonic velocity values of 1, 2-ethanediol, 1,2,3-propanetriol and water with those available in the literature and are given in Table 1.

Appropriate quantities of 1, 2-ethanediol and 1, 2, 3-propanetriol are taken to prepare the aqueous binary mixture. By means of digital density and ultrasonic velocity meter (DSA 5000 M, Anton Paar), the density, ultrasonic and viscosity measurements have been carried out. The accuracy rate for density and ultrasonic measurements is ±0.00005 kgm^{-3} and ± 0.5 m/s respectively. The instrument operates at 2MHz frequency and possesses thermostat built inside to maintain the temperature with the variation of ±0.01 K. The refractive indices of the binary mixtures as well as pure components have been recorded with the help of Abbe's

refractometer (MA-02) at the mentioned temperatures. The desired temperature has been retained with the help of water circulated thermostat. The refractometer maintains an accuracy of ±0.001 and operates at 589 nm.

2.1. Methodology preamble.

The molar volume (V) values of the mixtures of water + 1,2-ethanediol /1,2,3-propanetriol are estimated from the density (ρ) values using Equation 1.

$$V = (M_1X_1 + M_2X_2)/\rho \quad (1)$$

Where, X_1 and X_2 are the mole fractions and M_1, M_2 are the molar masses of H_2O and 1, 2-ethanediol/1, 2, 3-propanetriol, respectively.

Different thermo-acoustic parameters like isentropic compressibility (β_s), intermolecular free length (L_f) and acoustic impedance (Z) are determined using Equations 2 to 4.

$$\beta_s = \frac{1}{\rho U^2} \quad (2)$$

$$Z = \rho U \quad (3)$$

$$L_f = K\beta_s^{1/2} \quad (4)$$

Where K represents Jacobson's constant and has a value of 93.875 +0.375 T) $* 10^{-8}$.

To elucidate molecular interactions in the above considered aqueous solutions, excess values of different investigated parameters are calculated using Equation 5.

$$\Delta A/_{A^E} = A - (X_1A_1 + X_2A_2) \quad (5)$$

Here $\Delta A/A^E$ symbolize the excess properties (V^E, $\beta_s{}^E$, Z^E, $L_f{}^E$, η^E and Δn_D).A specifies the properties of the aqueous solutions and A_1 and A_2 state the properties of those of pure constituents. The theoretical values of excess properties have been calculated by fitting to Redlich-Kister polynomial (Equation 6). With the help of MATLAB R2016b, the coefficients, A_i have been estimated.

$$\Delta A/_{A^E} = X_1X_2 \sum A_i(2X_2 - 1)^i \quad (6)$$

The standard deviations, σ (A^E) have been calculated using Equation 7.

$$\sigma(A^E) = \sum(A^E_{exp} - A^E_{cal})^2/(n-p)]^{\frac{1}{2}} \quad (7)$$

Where p is the number of coefficients and n gives the number of observations.

In order to check the effectiveness of different mixing rules adopted for viscosity and refractive index, the average absolute deviation (AAD) and average relative deviation (ARD) for the binary mixtures have been calculated using following equations.

$$AAD = \frac{\sum_{i=1}^{n}(A_{exp} - A_{cal})}{n} \quad (8)$$

$$ARD = \frac{\sum_{i=1}^{n}[(A_{exp} - A_{cal})/A_{cal}]}{n} \quad (9)$$

2.1.1. Mixing rules.

2.1.1.1. Refractive index.

The refractive indices for (1,2-ethanediol /1,2,3-propanetriol + water) systems have been evaluated at different temperatures using refractive index mixing rules such as Lorentz-Lorenz (L-L) and Gladstone-Dale (G-D) relations [24].

Lorentz-Lorenz (L-L)

$$\frac{n_D^2-1}{n_D^2+2} = \emptyset_1 \frac{n_{D1}^2-1}{n_{D1}^2+2} + \emptyset_2 \frac{n_{D2}^2-1}{n_{D2}^2+2} \quad (10)$$

Gladstone-Dale (G-D)

$$n_D\text{-}1 = (n_{D1}\text{-}1)\,\emptyset_1 + (n_{D2}\text{-}1)\,\emptyset_2 \quad (11)$$

Where n_{D1}, $n_{D2,}$ n_D represent the refractive indices of water, 1,2-ethanediol /1,2,3-propanetriol and mixture respectively. $\emptyset_1$ and $\emptyset_2$ represent the volume fractions of pure solute and solvent.

2.1.1.2. Viscosity.

There are several mixing rules to predict the viscosity in the binary mixture comprising of unlike components. The mixing rules proposed by Hind (Hi) and Grunberg and Nissan (G-N) are represented below [25].

Hind (Hi)

$$\eta = X_1^2\eta_1 + X_2^2\eta_2 + 2\,X_1X_2H \quad (12)$$

Where η is the viscosity of the aqueous solutions, X_1, X_2 and η_1, η_2 represent the mole fractions and viscosities of solvent (water) and solute (1,2-ethanediol /1,2,3-propanetriol), respectively. H indicates the cross pair interactions and is calculated with the help of equation 13 given below.

$$H = \frac{\eta - \eta_{cal}}{2X_1X_2} \quad (13)$$

Where η_{cal} is obtained from equation 14.

$$\eta_{cal} = X_1^2\eta_1 + X_2^2\eta_2 \quad (14)$$

Grunberg- Nissan (G-N)

$$ln\eta = X_1 ln\eta_1 + X_2 ln\eta_2 + X_1X_2G \quad (15)$$

Where G refers to the strength of interaction between the unlike components in the binary mixture and can be calculated as follows:

$$G = \ln\left(\frac{\eta}{\eta_{cal}}\right)/X_1X_2 \quad (16)$$

Where

$$\eta_{cal} = \eta_1^{X_1}\eta_2^{X_2} \quad (17)$$

3. Results and Discussion

The values of density (ρ), ultrasonic velocity (U), viscosity (η) and refractive index (n_D) of aqueous solutions of 1, 2-ethanediol /1, 2, 3 -propanetriol measured at T = 298.15, 308.15 and 318.15K are presented in Table 2 and 3. It is observed from Table 2 and 3 that in solution the values of above mentioned properties increase with an increase in mole fractions of 1,2-ethanediol and 1,2,3-propanetriol and agrees well the literature values [17].

4.1. Multiple linear regression analysis.

To elucidate the connection of the dependent variable with two or more independent variables this analysis is performed. In the present investigation, multiple regression analysis have been applied to calculate the viscosities and refractive indices of the aqueous solutions of 1,2-ethanediol /1,2,3-propanetriol at T= 298.15, 308.15 and 313.15K. The precision of the multiple linear regressions has been verified from the obtained regression statistics listed in Table 4 and 5.

4.2. Mixing rules.

The average absolute deviation and average relative deviation (AAD) and (APD) have been calculated to evaluate their rationality and presented in Table 6. Table 7 represents the AAD and APD calculated for viscosity in 1, 2-ethanediol /1, 2, 3-propanetriol + water binary mixtures at different temperatures.

4.3. Excess properties.

The alteration of mixture properties from the ideal behaviour is termed as excess properties. These are found to be significant as they help to envisage the nature of molecular interactions exists in a binary mixture.

4.3.1. Excess molar volume.

Effects such as chemical, physical and structural contribute to V^E. Non-specific associations mainly arise from physical effects that give rise to positive V^E. Structure forming interactions arising due to chemical or specific effects contribute to negative value of V^E. The excess molar volumes for 1, 2-ethanediol /1, 2, 3-propanetriol and water binary mixture over the entire mole fraction of 1, 2-ethanediol are represented in (Fig. 1 and 2). For 1, 2-ethanediol + water solutions studied at different temperatures, the V^E values are negative over the whole mole fraction range of 1, 2-ethanediol [17]. The negative V^E values show minima at $X_2 = 0.6$ for (298.15K, 308.15K and 318.15K) and then gradually decreases as the mole fraction of 1, 2-ethanediol increases. The negative V^E values predict specific interactions arising due to the formation of 1, 2-ethanediol + water complex, or 1,2-ethanediol interacting among them in the binary mixture. As the temperature increases from 298.15K – 318.15K, the negative V^E values decreases. This indicates there is weak interaction between the components at high temperature due to the rotation of individual groups and movement of atoms resulting in the disruption of a stable structure. But in case of 1, 2, 3-propanetriol and water binary mixture at the measured temperature, the values of V^E tend to remain positive. Above 0.6 mole fraction the values of V^E gradually become negative indicating specific interaction between the unlike components in the 1, 2, 3-propanetriol rich region. Even though both 1, 2, 3-propanetriol and water are polar, the physical forces such as hydrogen bonding dominate over chemical forces. The positive V^E region indicates the presence of hydrogen bonding between the 1, 2, 3-propanetriol molecules due to more number of carbon atoms and -OH groups [16]. Several conformations of 1, 2, 3-propanetriol molecules exist simultaneously in the mixture and get stabilised by inter- and intra-molecular hydrogen bonds [27].

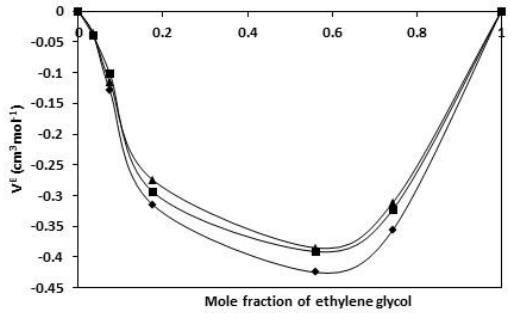

Figure 1. Excess molar volume for aqueous solutions of 1,2-ethanediol at T= (♦298.15, ■ 308.15 and ▲318.5K).

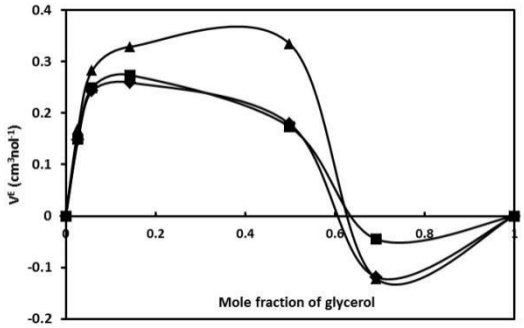

Figure 2. Excess molar volume for aqueous solutions of 1,2,3-propanetriol at T= (♦298.15, ■ 308.15 and ▲318.5K).

4.3.2. Deviation in viscosity.

Figures 3 and 4 represent the deviation in viscosity (η^E) for aqueous solutions of 1, 2-ethanediol /1, 2, 3-propanetriol.Viscosity is a significant property of liquid mixtures in exemplifying its flow. The curves in (Fig. 3 and 4) indicate that values of η^E are negative over the whole composition range. In the case of 1,2,3-propanetriol + water mixture, the negative trend of the curves decreases with rise in temperature which may be due to the disruption of interaction between the unlike constituents of the liquid mixture at high temperature and found as coherent with the V^E values. The deviations in viscosity for 1, 2, 3-propanetriol and water represented in figure 4 also exhibit a negative trend. Both the systems exhibit the minima at X_2 = 0.3. The large negative deviation is due to the H- bonding between the components as well as dipole – dipole interaction between two constituents. The viscosity of any substance varies with temperature. Water is less viscous as compared to the other two liquids. It has been stated by Pal and Sharma that the presence of dispersion and dipolar forces between the two components of a mixture may be accountable for observed negative deviation in viscosity [26]. Structural dissimilarities between the two unlike components can also be a reason for negative contribution. Aqueous solutions of 1, 2, 3-propanetriol exhibit unusual thermal behaviour of viscosity due to the presence of three hydrophilic alcoholic groups that participate in intra and inter-molecular hydrogen bonding [].

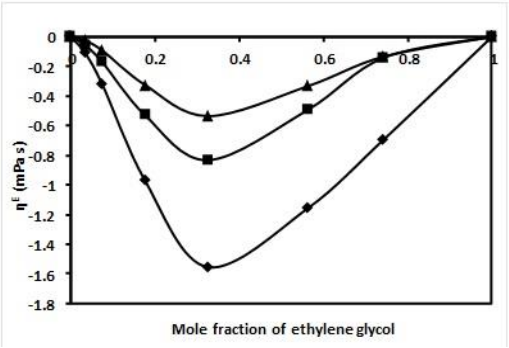

Figure 3. Excess η^E for aqueous solutions of 1,2-ethanediol at T= (♦298.15, ■ 308.15 and ▲318.5K).

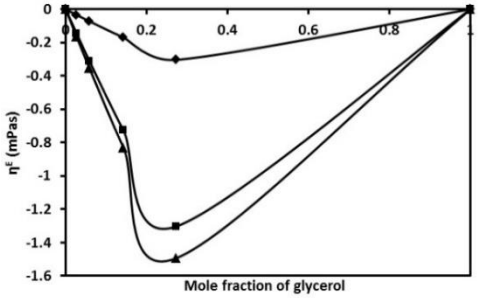

Figure 4. Excess η^E for aqueous solutions of 1, 2, 3-propanetriol at T= (♦298.15, ■ 308.15 and ▲318.5K).

4.3.3. Deviation in refractive index.

The variation in the change in refractive index with values of mole fraction of 1, 2-ethanediol and 1, 2, 3-propanetriol in the aqueous solutions have been measured at different temperatures and are represented in (Fig. 5 and 6) respectively. It has been observed that the changes in refractive index of aqueous solutions of 1, 2-ethanediol and 1, 2, 3-propanetriol are positive throughout the entire composition range. The positive deviations can be attributed to the breaking of hydrogen bonding of self - associated diol and triol molecules and weak bonding between the, unlike molecules. Another important factor that affects the excess properties of the binary mixtures is the influence of electron donating groups such as - CH_2CHCH_2 group. It has been reported in the earlier investigations that $\Delta\eta$ and Δn_D normally increase with an increase in size of alkyl group [27].

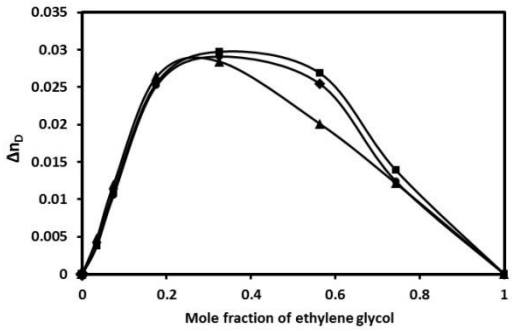

Figure 5. Deviation in refractive index for aqueous solutions of 1,2-ethanediol + at T= (♦298.15, ■ 308.15 and ▲318.5K).

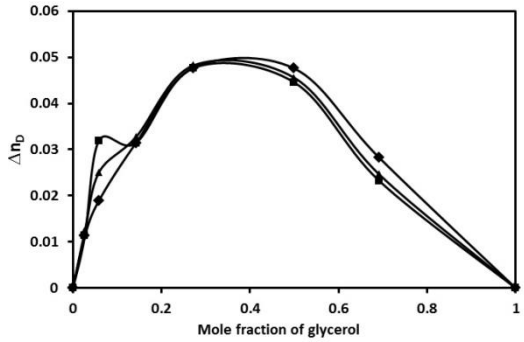

Figure 6. Deviation in refractive index for aqueous solutions of 1,2,3-propanetriol at T= (♦298.15, ■ 308.15 and ▲318.5K).

4.3.4. Excess isentropic compressibility (β^E), acoustic impedance (Z^E) and free length (L_f^E).

The plots of β^E versus mole fraction of 1, 2-ethanediol and 1, 2, 3-propanetriol are represented in (Fig. 7 and 8). It has been found that for 1, 2-ethanediol /1, 2, 3-propanetriol and water mixtures, the β^E values are negative over the entire composition range at all temperatures.

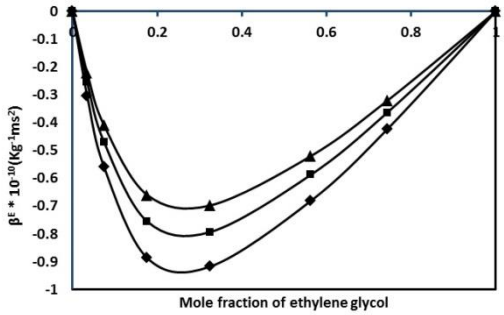

Figure 7. Plot of β^E versus mole fraction of 1,2-ethanediol at T= (♦298.15, ■ 308.15 and ▲318.5K).

The observed negative values of β^E indicate physical interactions between 1, 2-ethanediol /1, 2, 3-propanetriol molecules in the mixture through hydrogen bonding. The interaction among alcohol molecules is stronger when compared to the association with the water due to the presence of a greater number of OH groups. But with a rise in temperature the negative trend of compressibility decreases suggesting expansion in volume. As a result of temperature rise, the strength of hydrogen bonds gets weakened due to vibrations and the structure breaking effect dominates on the structure making effect.

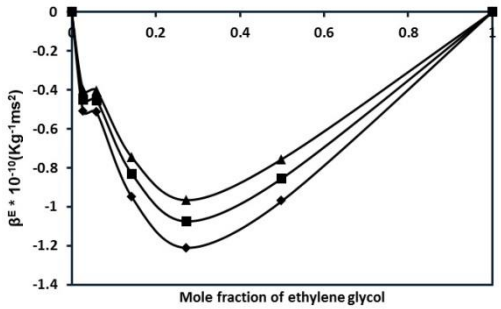

Figure 8. Plot of β^E versus mole fraction of 1,2,3-propanetriol at T= (♦298.15, ■ 308.15 and ▲318.5K).

The Z^E determines the opposition that a system offers to the flow and reveals that the dispersion is noticeable. There is a possibility of weak dipole –dipole type of forces acting between the components in aqueous solutions of 1, 2-ethanediol /1, 2, 3-propanetriol at three measured temperatures (Fig. 9 and 10). The positive deviations for 1, 2-ethanediol and water mixture arising due to weak hydrogen bonding have a maximum at X_2 =0.35. The negative magnitude of Z^E is good agreement with the positive values of V^E of 1, 2, 3-propanetriol and water system under investigation. This is so because 1, 2, 3-propanetriol molecules in the liquid state exist in different stable conformations having strong intra-molecular arrangement and vary with external conditions [14].

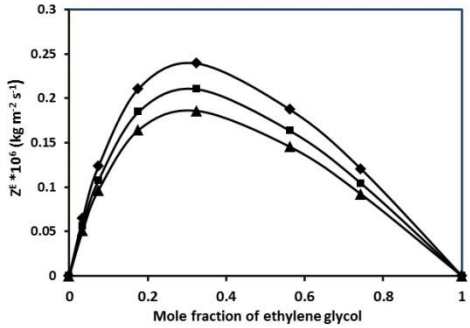

Figure 9. Plot of Z^E versus mole fraction of 1, 2-ethanediol at T= (♦298.15, ■ 308.15 and ▲318.5K).

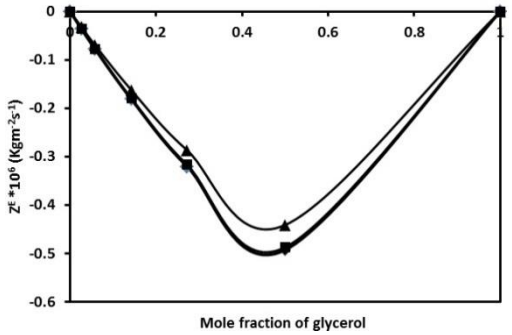

Figure 10. Plot of Z^E versus mole fraction of 1, 2, 3-propanetriol at T= (♦298.15, ■ 308.15 and ▲318.5K).

The positive deviation in excess intermolecular free length (L_f^E) in (Fig. 11 and 12) and analogous behaviour by β^E for 1, 2-ethanediol /1, 2, 3-propanetriol with water have been found.

The less magnitude of L_f^E suggests that there is a probability of weak interactive forces with dispersion forces and the existence of different conformational isomers in the binary systems.

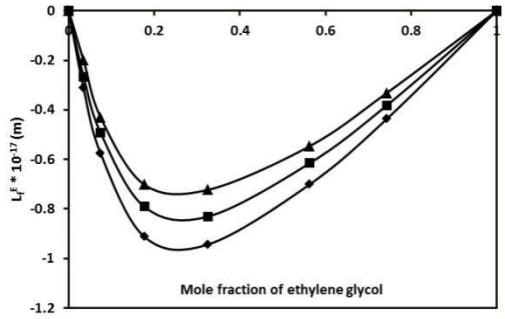

Figure 11. Plot of excess L_f versus mole fraction of 1,2-ethanediol at T= (♦298.15, ■ 308.15 and ▲318.5K).

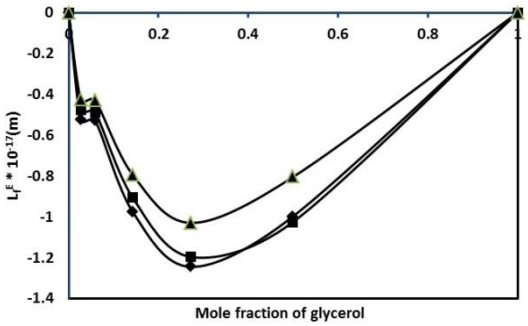

Figure 12. Plot of excess L_f versus mole fraction of 1,2,3-propanetriol at T= (♦298.15, ■ 308.15 and ▲318.5K).

The fitting coefficients for all the investigated excess properties at measured temperatures together with the values of standard deviation are enlisted in Tables 8 and 9.

4.3.5. Excess value of Gibb's energy of activation of flow.

The trend of excess Gibb's free energy of activation of flow (G^E) exhibits the H-bond heterogeneous interactions between the components of unlike constituents. It has been reported that 1, 2-ethanediol has –OH groups present at both the ends with gauche conformations that facilitates intra as well as inter-molecular H-bonding. In the present investigation, the negative η^E values and positive values of G^E of aqueous solutions of 1,2-ethanediol /1,2,3-propanetriol are obtained (Fig. 13 and 14) which suggest weak molecular interaction between the unlike components in comparison to diols and triols, where the hydroxyl groups interact with one another through hydrogen bonding and links the molecules together [19].

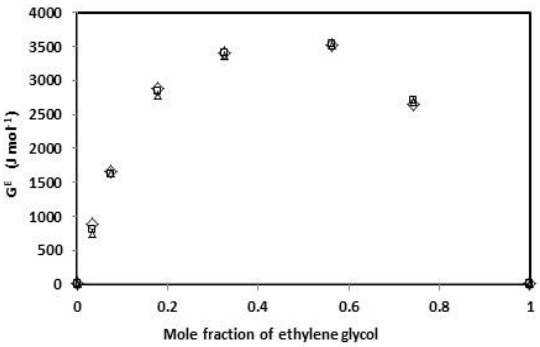

Figure 13. Excess Gibb's energy for aqueous solutions of 1,2-ethanediol at T= (◊298.15, □ 308.15 and Δ 318.5K).

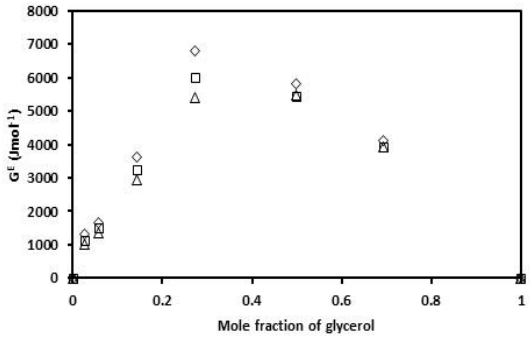

Figure 14. Excess Gibb's energy for aqueous solutions of 1,2,3-propanetriol at T= (◊ 298.15, □ 308.15 and Δ 318.5K).

Table 1. List of experimental and literature values of densities, ultrasonic velocity and refractive index of water, 1, 2-ethanediol and 1, 2, 3-propanetriol.

Liquids	Temperature (K)	Density(g/cm^3) (ρ)		Ultrasonic velocity (m/s) (U)	
		Experimental	Literature	Experimental	Literature
	298.15 K	0.99709	0.99730[15]	1496.59	1509.12[15]
	308.15 K	0.99377	0.99403[15]	1520.54	1519.80[15]
Water	318.15 K	0.9901	0.99022[16]	1536.33	
1,2-ethanediol	298.15 K	1.1047	1.10635[15]	1646.48	1644.2[15]
	308.15 K	1.09824	1.10290[15]	1624.48	-
	318.15 K	1.09114	-	1601.06	
1,2,3-	298.15 K	1.27196	1.2600[16]	1920.0	1964.00[16]
propanetriol	308.15 K	1.2646	-	-	-
	318.15 K	1.263	-	-	-

Table 2. Representation of experimental ρ, U, η and n_D values for aqueous solutions of 1,2-ethanediol at T =298.15, 308.15 and 318.15K.

X_2	**ρ (kg/m^3)**	**U (m/s)**	**η (mPa.s)**	**n_D**
298.15K				
0	0.9971	1496.59	0.881	1.333
0.0344	1.0098	1542.21	1.1589	1.340
0.0744	1.0249	1593.75	1.5559	1.351
0.1765	1.0535	1673.48	2.6695	1.375
0.3253	1.0728	1704.56	4.1212	1.393
0.5626	1.0949	1699.55	8.2599	1.412
0.7432	1.1024	1679.24	11.433	1.416
1	1.1047	1646.48	16.100	1.428
308.15K				
0	0.9937	1520.54	0.7367	1.332
0.0344	1.0063	1557.22	0.9259	1.339
0.0744	1.0196	1601.61	1.2217	1.350
0.1765	1.0480	1666.22	2.0194	1.374
0.3253	1.0665	1689.45	3.0532	1.370
0.5626	1.0881	1678.10	5.8542	1.411
0.7432	1.0954	1656.34	7.9872	1.415
1	1.0982	1624.48	10.743	1.425
318.15K				
0	0.9901	1536.33	0.6267	1.330
0.0344	1.0021	1566.3	0.7577	1.338
0.0744	1.0159	1604.54	1.0175	1.349
0.1765	1.0421	1656.69	1.5802	1.373
0.3253	1.0599	1672.71	2.3388	1.363
0.5626	1.0814	1656.46	4.2905	1.398
0.7432	1.0883	1633.19	5.7632	1.412
1	1.0911	1601.06	7.7645	1.424

Table 3. Representation of experimental ρ, U, η and n_D values for aqueous solutions of 1,2,3-propanetriol at T =298.15, 308.15 and 318.15K.

X_2	ρ (kg/m^3)	U (m/s)	η (mPa.s)	n_D
		298.15K		
0	0.9970	1496.59	0.8810	1.333
0.0269	1.0354	1572.92	1.3964	1.348
0.0587	1.0434	1582.73	1.5022	1.36
0.1426	1.0977	1688.81	3.2149	1.384
0.2723	1.1640	1808.06	13.611	1.418
0.4995	1.2149	1877.27	63.671	1.449
0.6919	1.2496	1890.91	398.85	1.456
1	1.2719	1920.00	1411.11	1.470
		308.15K		
0	0.9937	1520.54	0.7413	1.332
0.0269	1.0319	1587.69	1.0967	1.347
0.0587	1.0389	1597.79	1.1938	1.372
0.1426	1.0926	1689.32	2.3938	1.383
0.2723	1.1581	1796.28	8.589	1.417
0.4995	1.2087	1858.58	59.371	1.445
0.6919	1.2411	1878.10	388.95	1.45
1	1.2646	-	-	1.469
		318.15K		
0	0.9901	1536.33	0.633	1.33
0.0269	1.0275	1595.98	0.8876	1.346
0.0587	1.0339	1605.88	0.9687	1.363
0.1426	1.0875	1687.4	1.8372	1.382
0.2723	1.1519	1782.45	6.0436	1.415
0.4995	1.2023	1839.17	56.471	1.443
0.6919	1.2409	1861.23	370.35	1.448
1	1.2630	-	-	1.465

Table 4. Representation of regression statistics of η obtained from multiple linear regression analysis for aqueous solutions of 1, 2-ethanediol /1,2,3-propanetriol at T = 298.15K, 308.15K and 313.15K.

Regression statistics	η (mPa.s)		
	T =298.15K	T =308.15K	T =313.15K
	1,2-ethanediol +water		
R-square value	0.988	0.992	0.994
Standard error	0.648	0.339	0.220
Standard deviation	0.486	0.658	0.659
	1,2,3-propanetriol +water		
R-square value	0.758	0.999	0.991
Standard error	3.919	0.073	0.885
Standard deviation	2.771	0.499	0.478

Table 5. Representation of regression statistics of n_D obtained from multiple linear regression analysis for aqueous solutions of 1,2-ethanediol /1,2,3-propanetriol at T = 298.15K, 308.15K and 313.15K.

Regression statistics	n_D		
	T =298.15K	T =308.15K	T =313.15K
	1,2-ethanediol +water		
R-square value	0.921	0.986	0.904
Standard error	0.013	0.013	0.011
Standard deviation	0.613	0.367	0.389
	1,2,3-propanetriol +water		
R-square value	0.872	0.868	0.867
Standard error	0.020	0.019	0.019
Standard deviation	0.351	0.346	0.345

Table 6. Average absolute deviations (AAD) and average relative deviation (ARD) in L-L and G-D relations for refractive index in aqueous solutions of (a)1,2-ethanediol / (b)1,2,3-propanetriol at different temperatures.

Temperature(K)	(L-L) (AAD)	(L-L) (ARD)	(G-D) (AAD)	(G-D) (ARD)
298.15	0.0064	0.0047	0.0070	0.0059
308.15	-0.0037	-0.0026	-0.0052	-0.0036

318.15	0.0032	0.0024	0.0037	0.0029
Temperature(K)	(L-L) (AAD)	(L-L) (ARD)	(G-D) (AAD)	(G-D) (ARD)
298.15	0.0275	0.0207	0.0139	0.0103
308.15	0.0283	0.0212	0.0145	0.0108
318.15	0.0301	0.0226	0.0177	0.0130

Table 7. Average absolute deviations (AAD) and average relative deviation (ARD) in Hind (Hi) and Grunberg-Nissan (G-N) relations for viscosity in aqueous solutions of 1,2-ethanediol /1,2,3-propanetriol at different temperatures.

1,2-ethanediol + water	Hi (AAD)	Hi (ARD)	G-N (AAD)	G-N (ARD)
T = 298.15K	1.261	0.5568	1.404	0.4462
T = 308.15K	0.9800	0.5717	0.9605	0.3980
T = 318.15K	0.7365	0.5635	0.6573	0.3493
1,2,3-propanetriol + water	Hi (AAD)	Hi (ARD)	G-N (AAD)	G-N (ARD)
T = 298.15K	-85.652	-0.496	36.421	0.541
T = 308.15K	-88.173	-0.529	35.323	0.439
T = 318.15K	-91.256	-0.555	33.460	0.374

Table 8. Coefficients of Redlich - Kister polynomial and values of standard deviation for V^E, η^E, βs^E, $L_f{}^E$, Z^E and Δn_D for aqueous solutions of 1,2-ethanediol.

Properties	A_0	A_1	A_2	A_3	σ
			298.15 K		
V^E	-1.7063	-0.4399	-0.7943	1.0026	0.0108
η^E	-5.4281	5.9072	0.0353	-9.4284	0.0091
β^E	-3.015	2.2072	-2.384	2.4457	0.0001
L_f	-3.100	2.2705	-2.4515	2.515	0.0001
Z^E	0.8252	-0.5687	0.4633	-0.2332	0.00003
Δn_D	0.1198	-0.0813	-0.0136	-0.0089	0.00003
			308.15 K		
V^E	-1.563	-0.3621	-0.6694	0.7977	0.0168
η^E	-2.5726	4.4902	1.0428	-5.1684	0.00427
β^E	-2.6087	1.9709	-1.9751	1.803	0.0001
Lf	-2.7316	2.0638	-2.0684	1.888	0.0002
Z^E	0.722	-0.5139	0.4006	-0.1786	0.0002
Δn_D	0.1211	-0.0763	-0.0136	-0.0198	0.00002
			318.15 K		
V^E	-1.5275	-0.5887	-0.472	1.5068	0.0014
η^E	-1.698	2.7525	0.456	-3.884	0.0022
β^E	-2.3079	1.72	-1.6671	1.5698	0.0002
Lf	-2.4592	1.8342	-1.7757	1.6737	0.0002
Z^E	0.6411	-0.4432	0.3451	-0.2147	0.0002
Δn_D	0.0772	-0.1374	0.1492	0.1841	0.00002

Table 9. Coefficients of Redlich - Kister polynomial and values of standard deviation for V^E, η^E, βs^E, $L_f{}^E$, Z^E and Δn_D for aqueous solutions of 1,2,3-propanetriol.

Properties	A_0	A_1	A_2	A_3	σ
		298.15K			
V^E	0.7161	0.2682	-4.9776	-11.3415	0.0232
η^E	-1.913	-1.0388	-0.4871	-0.0973	0.9890
β^E	0.8271	0.3916	0.1970	0.3885	0.8699
L_f	0.8508	0.4026	0.2034	0.0399	0.4599
Z^E	-1.9727	-0.9796	-0.4809	-0.0948	0.4574
Δn_D	0.1863	0.1863	0.1863	0.1863	0.0002
		308.15K			
V^E	0.6938	0.5427	-3.5867	-1.0318	0.0240
η^E	-0.8200	-0.44201	-0.2068	-0.4137	0.2966
β^E	0.76639	0.3640	0.1830	0.3605	0.7452
L_f	0.8011	0.3803	0.1916	0.3764	0.4036
Z^E	-1.9512	-0.9672	-0.4750	-0.0937	0.3759
Δn_D	0.174	-0.1357	-0.0906	-0.1069	0.0086
		318.15K			
V^E	1.338	-0.2034	-7.2520	-13.4494	0.0243

Properties	A_0	A_1	A_2	A_3	σ
η^E	-9.4665	-5.2496	-2.4547	-0.4909	0.9137
β^E	0.6778	0.3220	0.1620	0.3192	0.5829
L_f	0.8238	0.4180	0.2069	4.0776	0.4253
Z^E	-1.7665	-0.8756	-0.4303	-0.0848	0.2972
Δn_D	-0.3717	-1.3836	-1.0332	-0.4523	0.0002

4. Conclusions

From the ultrasonic velocities and density values, different acoustic properties have been calculated. The sign and magnitude of different excess functions obtained from the measured properties at different temperatures reflect the nature of molecular interaction existing between the species present in the solutions. In the studied aqueous alcohol mixtures, specific attention has been paid to in vigil physical forces such as intra and intermolecular hydrogen bonding. It has been found that for the studied binary mixtures V^E values are found to be negative and positive suggesting weak interaction between unlike components and strong specific interaction arising from intra-molecular hydrogen bonding between the alcohol molecules. This interaction gets enhanced with increased number of OH groups and carbon atoms. The trend of derived excess acoustic properties compliments the V^E values. The sign of Δn_D and η^E values relate well with the earlier investigations reported in the literature. The presence of self-association as well as hydrogen bonding interaction in the aqueous solutions 1,2-ethanediol / water and 1,2,3-propanetriol gives rise to non-ideality. The deviations in the observed physico-chemical properties are developed due to this. The mixing rules adopted for calculating the theoretical values of n_D and η values exhibit fair agreement with the experimental values.

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Conflicts of Interest

The authors declare no conflict of interest.

References

1. Parthipan, G.; Thenappan, T. Dielectric and thermodynamic behavior of binary mixture of anisole with morpholine and aniline at different temperatures. *Journal of Molecular Liquids* **2008**, *138*, 20-25, https://doi.org/10.1016/j.molliq.2007.06.010.
2. Boodida, S.; Bachu, R.; Tangeda, S.; Satyanarayana, N. Excess molar volumes and viscosity deviations of binary liquid mixtures of N-methylacetamide with ethyl acetate, ethyl chloroacetate and ethyl cyanoacetate. *Indian Journal of Chemistry - Section A Inorganic, Physical, Theoretical and Analytical Chemistry* **2008**, *47*, 66-70.
3. Acree, W.E. Comments on "Density, dynamic viscosity, excess properties and intermolecular interaction of triethylene glycol + N,N-dimethylformamide binary mixture". *Journal of Molecular Liquids* **2019**, *275*, 441-442, https://doi.org/10.1016/j.molliq.2018.11.098.

4. Audiseshaiah, O.; Raveendra, M.; Kiran Babu, L.; Sivakumar, K.; Rami Reddy, Y.V. Physicochemical properties and FT-IR spectral studies of binary liquid mixtures containing anisole with isomeric cresols. *Physics and Chemistry of Liquids* **2019**, *0*, 1-17, https://doi.org/10.1080/00319104.2019.1706174.
5. Duereh, A.; Sato, Y.; Smith, R.L.; Inomata, H. Application of the Preferential Solvation Viscosity Model to Binary Liquid Mixtures: Aqueous, Nonaqueous, Ionic Liquid, and Deep Eutectic Solvent Systems. *Industrial & Engineering Chemistry Research* **2019**, *58*, 14991-15002, https://doi.org/10.1021/acs.iecr.9b01179.
6. Haarmann, N.; Sosa, A.; Ortega, J.; Sadowski, G. Measurement and Prediction of Excess Properties of Binary Mixtures Methyl Decanoate + an Even-Numbered n-Alkane (C6–C16) at 298.15 K. *Journal of Chemical & Engineering Data* **2019**, *64*, 2816-2825, https://doi.org/10.1021/acs.jced.9b00185.
7. Luning Prak, D.J.; Morrow, B.H.; Cowart, J.S.; Trulove, P.C.; Harrison, J.A. Thermophysical Properties of Binary Mixtures of n-Dodecane with n-Alkylcyclohexanes: Experimental Measurements and Molecular Dynamics Simulations. *Journal of Chemical & Engineering Data* **2019**, *64*, 1550-1568, https://doi.org/10.1021/acs.jced.8b01135.
8. Singh, K.P.; Singh, S. Thermodynamic and acoustic properties of binary mixtures of PEGDME 250 with 1-propanol and 1-butanol at 293, 303 and 313°K. *Physics and Chemistry of Liquids* **2019**, *57*, 658-678, https://doi.org/10.1080/00319104.2018.1510930.
9. Popoola, L.T.; Yusuff, A.S.; Aderibigbe, T.A. Assessment of natural groundwater physico-chemical properties in major industrial and residential locations of Lagos metropolis. *Applied Water Science* **2019**, *9*, 1–10, https://doi.org/10.1007/s13201-019-1073-y.
10. Zorębski, E.; Lubowiecka-Kostka, B. Thermodynamic and transport properties of (1,2-ethanediol+1-nonanol) at temperatures from (298.15 to 313.15)K. *The Journal of Chemical Thermodynamics* **2009**, *41*, 197-204, https://doi.org/10.1016/j.jct.2008.09.018.
11. Mohamed, S.; El-Sakhawy, M.; Kamel, S. Enhancement of water resistance and antimicrobial properties of paper sheets by coating with shellac. *Letters in Applied Nanobioscience*, **2019**, *8*, 637-642. https://doi.org/10.33263/LIANBS83.637642.
12. Yang, C.; Liu, Z.; Lai, H.; Ma, P. Excess Molar Volumes and Viscosities of Binary Mixtures of p-Cresol with Ethylene Glycol and Methanol at Different Temperature and Atmospheric Pressure. *Journal of Chemical & Engineering Data* **2006**, *51*, 457-461, https://doi.org/10.1021/je050376y.
13. Dalai, B.; Kar, P.; Kuanar, B.; Dash, S.K.; Singh, S.K. Physico-chemical properties of binary systems involving modifier (methanol or propionic acid) and an acidic extractant di (2-ethylhexyl) phosphoric acid (DEHPA): volumetric, acoustic and viscometric routes**.** *Biointerface Research in Applied Chemistry***, 2020,** *10***,** 5880-5885. https://doi.org/10.33263/BRIAC104.880885.
14. Egorov, G.I.; Makarov, D.M.; Kolker, A.M. Volume properties of liquid mixture of water+glycerol over the temperature range from 278.15 to 348.15K at atmospheric pressure. *Thermochimica Acta* **2013**, *570*, 16-26, https://doi.org/10.1016/j.tca.2013.07.012.
15. Pal, A.; Singh, W. Speeds of Sound and Viscosities in Aqueous Poly(ethylene glycol) Solutions at 303.15 and 308.15 K. *Journal of Chemical & Engineering Data* **1997**, *42*, 234-237, https://doi.org/10.1021/je960173x.
16. Vural, U.S.; Muradoglu, V.; Vural, S. Excess molar volumes and refractive index of binary mixtures of 1,2,3-propanetriol + methanol and 1,2,3-propanetriol + water at 298.15K and 303.15K. *Bull. Chem. Soc. Ethiop.* **2011,** *25*, 111-118, https://doi.org/10.4314/bcse.v25i1.63374.
17. Tsierkezos, N.G.; Molinou, I.E. Thermodynamic Properties of Water + Ethylene Glycol at 283.15, 293.15, 303.15, and 313.15 K. *Journal of Chemical & Engineering Data* **1998,** *43*, 989-993, https://doi.org/10.1021/je9800914.
18. Pla-Franco, J.; Lladosa, E.; Loras, S.; Monton, J.B. Approach to the 1-propanol dehydration using an extractive distillation process with 1,2-ethanediol. *Chem. Eng. Process* **2015,** 7-3, https://doi.10.1016/j.cep.2015.03.007.
19. Sengwa, R.J. A comparative dielectric study of ethylene glycol and propylene glycol at different temperatures. *Journal of Molecular Liquids* **2003**, *108*, 47-60, https://doi.org/10.1016/S0167-7322(03)00173-9.
20. Chaube, H.A.; Rana, V.A.; Hudge, P. Dielectric relaxation studies of binary mixture of 1,2-ethanediol mono phenyl ether and methanol by time domain reflectometry. *J. Mol. Liq.* **2014,** *193*, 29-36, https://doi.org/10.1016/j.molliq.2015.07.020.
21. Chaube, H.A.; Rana, V.A. Dielectric studies of binary mixtures of ethylene glycol mono phenyl ether with 1-butanol at different temperatures. *Journal of Advanced Dielectrics* **2017**, *07*, https://doi.org/10.1142/S2010135X17500230.
22. Kinart, C.M.; Ćwiklińska, A.; Maj, M.; Kinart, W.J. Thermodynamic and physicochemical properties of binary mixtures of sulfolane with ethylene glycol, diethylene glycol, triethylene glycol, and tetraethylene glycol systems at 303.15K. *Fluid Phase Equilibria* **2007**, *262*, 244-250, https://doi.org/10.1016/j.fluid.2007.09.009.
23. Delgado, D.R.; Romdhani, A.; Martínez, F. Solubility of sulfamethizole in some propylene glycol+water mixtures at several temperatures. *Fluid Phase Equilibria* **2012**, *322-323*, 113-119, https://doi.org/10.1016/j.fluid.2012.03.014.

24. Mehra, R. Application of refractive index mixing rules in binary systems of hexadecane and heptadecane withn-alkanols at different temperatures. *Journal of Chemical Sciences* **2003**, *115*, 147-154, https://doi.org/10.1007/BF02716982.
25. Tariq, M.; Altamash, T.; Salavera, D.; Coronas, A.; Rebelo, L.P.N.; Lopes, J.N.C. Viscosity Mixing Rules for Binary Systems Containing One Ionic Liquid. *ChemPhysChem* **2013**, *14*, 1956-1968, https://doi.org/10.1002/cphc.201300086.
26. Pal, A.; Sharma, S. Excess Molar Volumes and Viscosities of 1-Propanol + Ethylene Glycol, + Ethylene Glycol Monomethyl, + Ethylene Glycol Dimethyl, + Diethylene Glycol Dimethyl, + Triethylene Glycol Dimethyl, + Diethylene Glycol Diethyl, and + Diethylene Glycol Dibutyl Ethers at 298.15 K. *Journal of Chemical & Engineering Data* **1998**, *43*, 532-536, https://doi.org/10.1021/je9702955.
27. Koohyar, F.; Kiani, F.; Sharifi, S.; Sharifirad, M.; Rahmanpour, S. Study on the Change of Refractive Index on Mixing, Excess Molar Volume and Viscosity Deviation for Aqueous Solution of Methanol, Ethanol, Ethylene Glycol, 1-Propanol and 1, 2, 3-Propantriol at T = 292.15 K and Atmospheric Pressure. *Research Journal of Applied Sciences, Engineering and Technology* **2012**, *4*, 3095-3101.