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Molecular Interaction Studies on Physiochemical and Derived Properties of Binary Mixtures at Atmospheric Pressure

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Abstract: The physiochemical properties like refractive indices, densities, and static dielectric constants of binary mixtures were measured over the entire concentration range. Refractive index and densities were measured at 293.15 K, and dielectric constants at (293.15, 298.15, and 303.15 K) temperatures for the n-propanol-formamide binary system. From dielectric data, the related and excess properties were calculated and reported in this work. From experimental refractive indices and densities, various related and excess properties were estimated and reported in this study. Static dielectric constants, densities, and refractive index of the binary system increase with the increase in the mole fraction of formamide. The results obtained were discussed in terms of intermolecular interactions, structural effects, hydrogen bonding, and other possible interactions between like and unlike molecules of the binary system.

Keywords: Physiochemical properties; Related properties; Excess properties; Binary system.

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1. INTRODUCTION

The physiochemical properties like refractive index, densities, and dielectric constants of pure liquids and their mixtures are very important for accurate design and proper development in many industrial processes (1). Variation in these physical properties with temperature, composition, and frequency gives important information about molecular structure, intermolecular interactions, hydrogen bonding, molecular associations, charge transfer, dipole-dipole, and dipole-induced dipole interactions (2).

Formamide is an amide derived from formic acid, which is used as a solvent in various chemical processes. It is used in the agrochemical and pharmaceutical industries and as a solvent in the polymer and resin industry. N-propanol is a primary alcohol and a clear, colorless, transparent liquid soluble in water. It is used as a solvent in the manufacturing of the pharmaceutical industry and as a chemical intermediate. The knowledge of the physiochemical properties of the corresponding liquids and their mixtures plays an important role in analytical sciences and pharmaceutical processes like synthesis, design, extraction, purification, and the processes involved (3). Limited information is available on the physiochemical properties of n-propanol with formamide. Only some reports are available on binary interaction studies of n-propanol and formamide with other polar liquids using different techniques and instruments. The dielectric study of formamide-butylene glycol at various temperatures was reported by Navarkhele et al. (4). Kabir et al. (5) reported densities and excess molar volumes of ethanol, methanol, and n-propanol with pure water at various temperatures. Static permittivity, refractive index, density, and related properties of pyridine with 1-propanol have been reported by Trivedi et al. (6).

In this study, authors reported refractive indices (n), densities (ρ) at 293.15 K, and static dielectric constants at 293.15, 298.15, and 303.15 K temperatures over the entire composition range. The related properties like excess dielectric constant (ε^E), effective Kirkwood correlation factor (g^{eff}), atomic polarization, electronic polarization, permittivity at a higher frequency, molar volume, molar refraction, polarizability, solvated radii, molar polarization, deviation in molar refractivity and the excess properties like excess density (d^E), excess refractive index (n^E), excess molar polarization $(P_m)^E$ and excess molar volume (V^E) were estimated from the experimental data using appropriate equations to confirm molecular interaction, hydrogen bonding, and other possible interactions between the components of mixtures.

2. EXPERIMENTAL SECTIONS

2.1. Chemicals

Chemicals formamide and n-propanol were obtained from a pharmaceutical company –Aurangabad – Maharashtra, with 99.9% purity and used without extra purification. The final compositions were prepared by adding formamide in n-propanol at eleven different stages. All mixtures were ready just before the experimental use.

2.2. Equipments

The refractive indices of the mixtures were measured using a digital pocket refractometer PAL-RI, made by Atago-Japan. The apparatus measures the refractive index in the range of 1.3306 to 1.5284. The refractive indices of pure liquids and their mixtures were measured only at 293.15 K temperature. The accuracy in the measurement of the refractive index given by the manufacturer is ± 0.0003 at 20 °C.

A wet sensor was used to measure static dielectric constants made by Delta-T Devices Ltd. UK that is based on the frequency domain reflectometry technique. Five to six readings were recorded for the mixture, and the average value of that was taken as a dielectric constant. The working detail of the wet sensor was explained in our earlier publication (7). The uncertainty in the dielectric constant given by the manufacturer is about ± 3%. The temperature controller arrangement with a water bath was used to sustain a constant temperature within the correctness limit of ± 1 °C.

Anton Paar oscillation U-tube densitometer (model DMA- 35, Austria) was used to measure the densities of the mixtures. The densitometer is calibrated with double-distilled water and air. The densities of pure liquids and their mixtures were measured only at 293.15 K temperature. The uncertainty in density given by the firm is ± 0.001 g·cm^{-3} at 20 °C.

2.3. Excess Dielectric Constants (ε^E)

The formation of a new structure in the binary mixtures can be confirmed from the knowledge of excess dielectric constants (ε^E). The excess dielectric constant is calculated using the equation (8):

$$\varepsilon^E = (\varepsilon_{sm}) - [\varepsilon_1 . \Phi_1 + \varepsilon_2 . \Phi_2] \quad (1)$$

Where (ε_{sm} is the static dielectric constants of mixtures, $\varepsilon_1, \varepsilon_2, \Phi_1$ and Φ_2 are the static dielectric constants and mole fractions of liquid 1 (n-propanol) and liquids 2 (formamide) respectively.

2.4. Kirkwood Factor (g)

The orientation of the electric dipoles in polar mixtures can be confirmed by the nature of Kirkwood correlation factor (9) values. The modified Kirkwood correlation factor, that is, effective Kirkwood correlation factor (g^{eff}) given by (10,11), is estimated by the equation:

$$\frac{4\Pi N}{9KT}\left(\frac{\mu_1^2 . \rho_1 . \Phi_1}{M_1} + \frac{\mu_2^2 . \rho_2 . \Phi_2}{M_2}\right) g^{eff} = \frac{(\varepsilon_{sm} - \varepsilon_{\infty m})(2\varepsilon_{sm} + \varepsilon_{\infty m})}{\varepsilon_{sm}(\varepsilon_{\infty m} + 2)^2} \quad (2)$$

where "g^{eff}" is the effective Kirkwood correlation factor for the binary mixtures.

In above equation, N, K, T, $\mu_1^2, \mu_2^2, \rho_1, \rho_2, M_1, M_2, \Phi_1, \Phi_2, \varepsilon_{sm}$ and $\varepsilon_{\infty m}$ represent Avogadro's number, Boltzmann constant, temperature, the squared dipole moment of liquid 1-2, the density of liquid 1-2, the molecular weight of liquid 1-2, the mole fraction of liquid 1-2, static dielectric constant of the mixture and dielectric constant at a higher frequency, respectively.

2.5. Molar Refraction, Solvated Radii, Polarizability, Molecular Polarization, Atomic Polarization, Permittivity at Higher Frequency, and Deviation in Molar Refraction

From experimental refractive indices, densities, and dielectric constants data of pure liquids and mixtures, the authors have estimated molar refraction (R_m), atomic polarization (P_A), polarizability (α), solvated radii (r) and molecular polarization (P_m) by the following equations (12-16):

$$R_m = \left(\frac{n^2 - 1}{n^2 + 2}\right) V_m = P_A + P_E = P_T = P_D \quad (3)$$

$$P_A = 1.05\, n^2 \quad (4)$$

$$\left(\frac{n^2 - 1}{n^2 + 2}\right) = \left(\frac{4}{3}\right) \Pi n' \alpha \quad (5)$$

$$V_m = \left(\frac{4}{3}\right) \Pi r^3 \quad (6)$$

$$P_m = V_m \left(\frac{\varepsilon_s - 1}{\varepsilon_s + 2}\right) \quad (7)$$

The right-hand side of equation (3) is equal to the summation of both atomic polarization (P_A) and electronic polarization (P_E), and that is equal to total polarization (P_T) or distortion polarization (P_D). In the above equations "n" is the refractive index, $n' = \frac{N}{V_m}$, N is Avogadro's number "ε_s" static dielectric constant and V_m = (M/d) is molar volume; in this, 'M' is molecular weight and 'd' is the density of the liquids, respectively.

The permittivity at higher frequency ε_∞ is the square of the refractive index, and the equation calculated it:

$$\varepsilon_\infty = n^2 \quad (8)$$

2.6. Excess Refractive Index (n^E), Excess Density (d^E), Excess Molar Polarization (P_m)^E Excess Molar Volume (V)^E and Deviation in Molar Refraction

By using experimental values of refractive indices and densities, the excess refractive index (n^E) and

excess density were determined by the following equations:

$$n^E = n_{mix} - \left(\Phi_1 . n_1 - \Phi_2 . n_2\right) \quad (9)$$

$$d^E = d_{mix} - \left(\Phi_1 . d_1 - \Phi_2 . d_2\right) \quad (10)$$

where n_{mix} and d_{mix} are the values of refractive indices and densities of the mixtures and Φ_1, Φ_2, n_1 , n_2, d_1, and d_2 are the mole fractions, refractive indices, and densities of the first and second liquids, respectively.

From calculated molar polarization, the $(P_m)^E$ of the mixtures was determined by the equation:

$$P_m^E = P_{mix} - \left[P_{m1} . \Phi_1 + P_{m2} . \Phi_2\right] \quad (11)$$

In the above equation, Pmix and φ_1, φ_2, P_{m1}, P_{m2}, are the polarization of mixtures and mole fraction, molar polarization of liquid 1 and 2, respectively.

Excess molar volume $(V)^E$ was determined using the following equation (17):

$$V^E = Vm_{mix} - \left(\Phi_1 . Vm_1 - \Phi_2 . Vm_2\right) \quad (12)$$

Where V_{mmix} is the molar volume of the mixtures $\Phi_1 . \Phi_2$ represents mole fraction, V_{m1} and V_{m2} are the molar volume of the components 1 and 2, respectively.

From the knowledge of refractive indices and densities, the deviation in molar refraction was determined by the well-known equation (18):

$$Rm^E / \Delta_R = Rm_{mix} - \left(\Phi_1 . Rm_1 - \Phi_2 . Rm_2\right) \quad (13)$$

In the above equation (Rm_{mix}) is the molar refractivity of the mixtures $\Phi_1 . \Phi_2$ represents mole fraction and Rm_1 and Rm_2 are the molar refractivity of pure liquids 1 and 2, respectively.

3. RESULTS AND DISCUSSION

3.1. Static Dielectric Constant

Table 1 lists the experimental dielectric constants of the studied binary system. The dielectric constants of the mixtures decrease with an increase in temperature. The decrease in dielectric constants with an increase in temperature is due to orientation polarization. When the thermal motion of the mixture is increased, there is a fall in orientation polarization that reduces the alignment of the permanent dipoles; therefore, there is a decrease in dielectric constants (19). From the same Table, it is also noticed that the dielectric constant increases with an increase in mole fraction. This may be due to a decrease in the size and shape of the complex molecules after hydrogen bonding. This may increase the number of dipoles in the binary mixtures and increase the mobility as well as the volume of the rotating molecules. The experimental results are in agreement with the earlier results given by (20,21).

Table 1: Values of static dielectric constants for binary mixture of n-propanol + formamide at various molar fractions at different temperatures.

Mole fraction of formamide	Static dielectric constant T=293.15K	Static dielectric constant T=298.15K	Static dielectric constant T=303.15K
0.00	21.55	19.88	18.53
0.07	29.92	28.22	26.75
0.15	37.77	35.91	34.58
0.23	45.60	43.56	42.15
0.32	54.01	51.58	50.32
0.41	63.68	60.94	59.12
0.51	74.02	71.15	69.24
0.62	84.18	81.28	79.30
0.73	93.80	90.78	88.76
0.86	103.37	100.17	98.19
1.00	112.79	109.52	107.35

Figure 1 illustrates the variation in static dielectric constants against mole fraction. From Figure 1, it is observed that the variation in¿ with mole fraction is non-linear. In polar mixtures, if the molecular association is happening, a non-linear variation in the dielectric constants with concentration is expected, and the same is noticed in the Figure. This shows that an intermolecular association is occurring in the studied binary system (22,23).

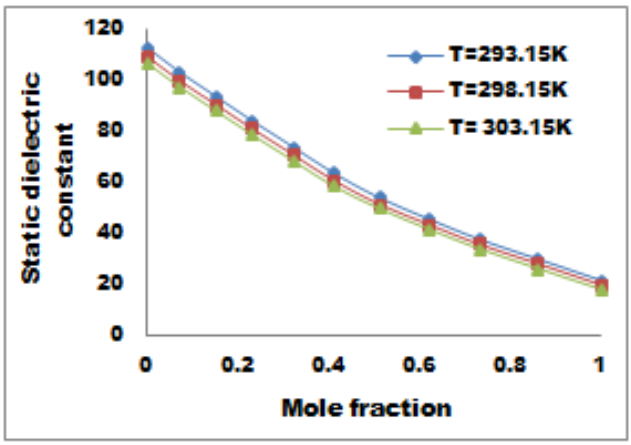

Figure 1: Variation in static dielectric constants (ε_s) against mole fraction at different temperatures.

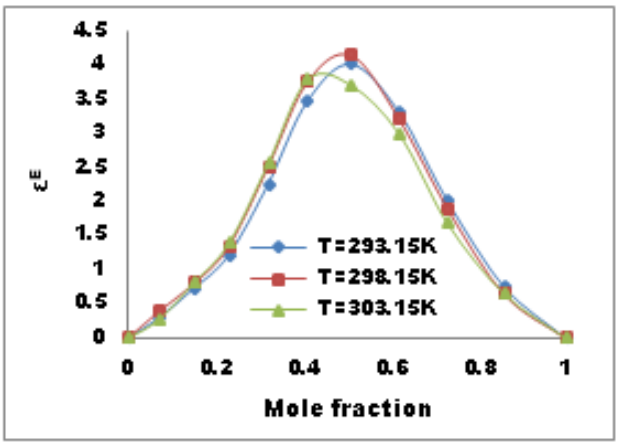

Figure 2: Variation in excess dielectric constant against mole fraction at different temperatures.

Table 2 gives experimental densities and refractive indices of the binary system at 293.15 K. The density values of the mixtures increase with an increase in the mole fraction of formamide. When the concentration of formamide increases, that may create intermolecular bonds, and new bonds may form between similar and dissimilar molecules that may decrease the volume of the mixtures, increasing density. Similar results have been recorded by Farid (12).

Table 2: Values of densities and refractive indices for a binary mixture of n-propanol + formamide at various mole fractions at 293.15 K temperature.

Mole fraction of formamide	Density ($g \cdot cm^{-3}$)	Refractive index
0.00	0.8032	1.3874
0.07	0.8362	1.3933
0.15	0.8690	1.3994
0.23	0.9021	1.4056
0.32	0.9350	1.4119
0.41	0.9680	1.4181
0.51	1.0011	1.4244
0.62	1.0341	1.4307
0.73	1.0671	1.4367
0.86	1.1001	1.4431
1.00	1.1333	1.4495

From Table 2, it is also noticed that refractive indices of the studied binary system increase with increasing mole fraction of formamide over the entire composition range.

3.2. Excess Dielectric Constant (ε^E)

The estimated (ε^E) values are presented in Figure 2. From Figure 2, it is observed that the excess dielectric constants are positive over the entire mole fraction range for the studied temperatures. This confirms that in the mixtures, the two liquids relate together in such a way that the effective dipole moment increases. In addition, the number of dipoles in the mixture may be more than the corresponding average number in the pure liquids, which may be due to the formation of a new structure leading to a higher macroscopic permittivity. Similar results are reported by Hosmani et al. and Navarkhele et al. (24,25).

3.3. Kirkwood Factor (g)

The orientation of electric dipoles in polar mixtures may be confirmed by Kirkwood factor (g). If the values of (g^{eff}) are greater than 1(g^{eff} >1), it shows parallel orientation, and if (g^{eff}) is less than 1 (g^{eff} < 1), it shows anti-parallel orientation of electrical dipoles. The effective Kirkwood correlation factor (g^{eff}) of the binary system was calculated using equation 2 and graphically illustrated in Figure 3. Figure 3 noted that the g^{eff} values are greater than 1 (g^{eff} >1) over the entire mole fraction range. This is a sign of the parallel orientation of electric dipoles in n-propanol molecules. From Figure 3, it is also observed that the variation in g^{eff} with mole fraction is non-linear at the studied temperatures, confirming the net increase in dipole ordering due to H-bond complexation. The results are in agreement with the earlier results of Trived et al. and Hosamani et al. (6,24).

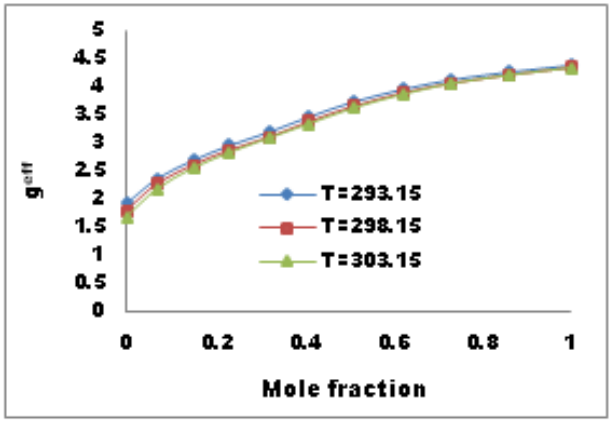

Figure 3: Variation in effective Kirkwood correlation factors against mole fraction at different temperatures.

3.4. Molar Volume (V_m), Molar Refractions (R_m), Polarizability (α), Solvated Radii (r), Molar Polarization (P_m), Atomic Polarization (P_A), Electronic Polarization (P_E) and Permittivity at Higher Frequency (ε_∞)

The estimated values of the molar volume (V_m), molar refractions (R_m), polarizability (α), solvated radii (r), molar polarization (P_m), atomic polarization (P_A), electronic polarization (P_E), and permittivity at

higher frequency (ε_∞) of the studied binary mixtures are illustrated in Table 3. From Table 3, it is noticed that all the above parameters are decreasing over the entire mole fraction range except (ε_∞) and (P_A). Since permittivity at higher frequency and atomic polarization depends on the refractive indices of the mixtures. Since the refractive index of the studied binary system increases, these parameters also increase with the mole fraction of formamide.

Table 3: Values of molar volume (V_m), molar refraction (R_m), polarizability (α), solvated radii (r), molar polarization (P_m), atomic polarization (P_A), electronic polarization (P_E), and permittivity at higher frequency (ε_∞) for a binary mixture of n-propanol + formamide at various mole fractions at 293.15 K temperature.

Mole fraction of formamide	V_m (cm³/mol)	R_m (cm³/mol)	α (10^{24}) cm/mole	(r) A⁰	(P_m) cm³/mole	(P_A)	(P_E)	ε_∞
0.00	77.292	18.204	7.22	2.65	67.444	2.02	19.19	1.93
0.07	72.222	17.248	6.83	2.58	65.432	2.03	15.21	1.94
0.15	67.525	16.348	6.48	2.52	62.432	2.05	14.29	1.95
0.23	63.158	15.500	6.14	2.47	59.177	2.07	13.42	1.97
0.32	59.112	14.705	5.83	2.41	55.946	2.09	12.61	1.99
0.41	55.335	13.947	5.53	2.36	52.808	2.11	11.83	2.01
0.51	51.803	13.229	5.24	2.31	49.758	2.13	11.09	2.02
0.62	45.501	12.546	4.97	2.26	46.812	2.14	10.39	2.04
0.73	45.407	11.889	4.71	2.21	43.985	2.16	9.72	2.06
0.86	42.491	11.264	4.46	2.16	41.281	2.18	9.07	2.08
1.00	39.744	10.667	4.24	2.12	38.708	2.21	8.47	2.11

3.5. Excess Density (d^E), Excess Refractive Index (n^E), Excess Molar Polarization (P_m)^E, Excess Molar Volume (V^E), and Deviation in Molar Refraction (Δ_R)

Table 4 gives estimated excess density (d^E), excess refractive index (n^E), excess molar polarization (P_m)^E, excess molar volume (V^E), and deviation in molar refraction with mole fraction of formamide. As Table 4 shows, the (d^E) values of the binary mixtures are negative over the entire composition range at the studied temperature. This may be due to a decrease in the molar volume of the binary mixtures.

Table 4: Values of excess density, excess refractive index, excess molar polarization, excess molar volume, and deviation in molar refractivity for binary mixture of n-propanol + formamide at various mole fractions at 293.15 K temperature.

Mole fraction of formamide	Excess density $(D)^E$	Excess Refractive index $(n)^E$	Excess molar polarization $(P_m)^E$	Excess molar volume (V^E)	Δ_R cm³.mol⁻¹
0.00	0.00000	0.00000	0.000	0.00	0.00
0.07	-0.00018	-0.00013	0.86	-1.25	-0.19
0.15	-0.00028	-0.00024	0.73	-2.12	-0.33
0.23	-0.00017	-0.00027	0.35	-2.69	-0.41
0.32	-0.00026	-0.00018	-0.0026	-2.96	-0.45
0.41	-0.00025	-0.00020	-0.26	-2.99	-0.46
0.51	-0.00014	-0.00012	-0.44	-2.77	-0.42
0.62	-0.00013	-0.00004	-0.51	-2.35	-0.35
0.73	-0.00023	-0.00026	-0.46	-1.74	-0.27
0.86	-0.00012	-0.00019	-0.29	-0.95	-0.15
1.00	0.00000	0.00000	0.000	0.00	0.00

From Table 4, it is also observed that the excess refractive indices values of the binary mixtures are negative over the entire composition range. The (n^E) give a signal of maximum solvent-solvent interaction that depends mainly on different physical properties of the solvents, such as dipole moment, dielectric constant, donor number, chemical structure, basicity, and polarizability (12).

The estimated values of $(P_m)^E$ of the binary mixture are given in Table 4. From the values, it is observed that the excess molar polarization is positive in the n-propanol-rich region and negative in the formamide-rich region at the studied temperature.

The (V^E) of the binary mixtures is given in Table 4 and graphically illustrated in Figure 4. From Figure 4, it is noticed that the excess molar volume values are negative over the entire mole fraction range at

the studied temperature. The negative values of excess molar volumes may be due to the volume contraction after mixing. Knowledge of (V^E) values can confirm the non-ideal behavior of real mixtures. Negative values of excess molar volumes show strong intermolecular interaction, hydrogen bonding, charge transfer complexes, and other complex-forming interactions, including strong dipole-dipole interactions between the components of molecules in the mixture (26).

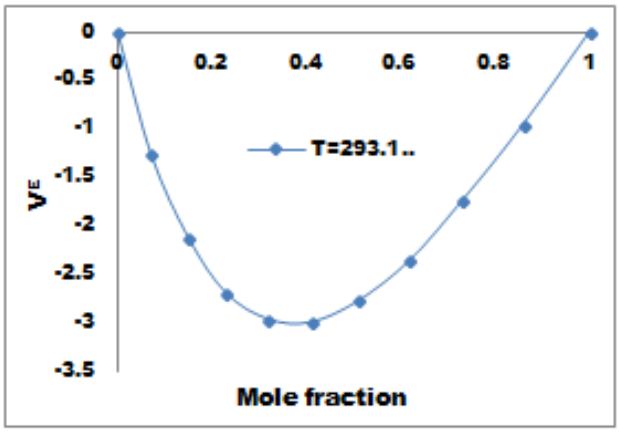

Figure 4: Variation in excess molar volume against molar fraction at 293.15 K temperature.

The deviation in molar refraction (Δ_R) is given in Table 4 and graphically illustrated in Figure 5. The (Δ_R) values are negative over the entire mole fraction range for the studied temperature. From Figure 5, it is seen that the variation in (Δ_R) with mole fraction exhibits a parabolic shape with a minimum value located between 0.3 and 0.4 mole fraction. The negative deviation in molar refraction signifies electronic perturbation due to the orbital mixing of the components of the mixtures. The negative (Δ_R) can be used as a measure of the strength of the interaction between the elements of the mixtures and is strongly dependent on composition and temperature (27,28). Negative values of (Δ_R) also point out greater dispersive forces in the mixtures than in pure components (29).

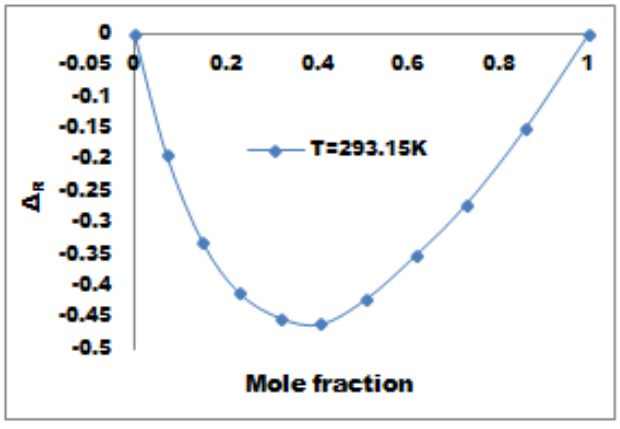

Figure 5: Variation in deviation in molar refractivity against mole fraction at 293.15 K temperature.

4. CONCLUSION

This study provides fundamental physiochemical properties and their related and excess properties of a binary system. Variation in these properties with mole fraction is explained in terms of molecular interactions, hydrogen bonding, and other possible interactions. It was found that the dielectric constants decrease with an increase in temperature and increase with the increase in the mole fraction of the solute. The densities and refractive indices of the binary mixtures increase with an increase in the mole fraction of solute at the studied temperature. For the studied temperatures, the dielectric data shows an excellent non-linear behavior supporting an intermolecular association and hydrogen bonding in the binary system.

The excess dielectric constants of the binary mixture are found to be positive and show a more polar structure, leading to higher macroscopic permittivity. The non-linear variation in (g^{eff}) values confirms the presence of molecular interactions, hydrogen bonding, and other possible interactions in the binary mixtures.

The estimated parameters of molar volume, molar refraction, polarizability, solvated radii, molar polarization, and electronic polarization of the binary system decrease, whereas permittivity at a higher frequency and atomic polarization increase with an increase in mole fraction.

The excess densities (d^E), excess refractive indices (n^E), excess molar volumes (V^E), and deviation in molar refractivity of the binary mixtures are found to be negative over the entire composition range and at the studied temperature. The negative excess molar volumes (V^E) confirm volume contraction after mixing of the binary mixtures. The values of (Δ_R) are negative and show greater dispersive forces in the mixtures than in pure components from a molecular interaction perspective; we suggest that the contributors to (V^E) are most likely from the hydrogen bonding formation interactions, charge transfer complexes and other complex-forming interactions in the binary system.

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6. AUTHOR CONTRIBUTIONS

The manuscript was written with contributions from all listed authors. All authors have approved of the manuscript.

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8. CONFLICT OF INTEREST

The authors declare no competing financial interest.

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