



Study of Molecular Interactions Through Density, Viscosity and Ultrasonic Measurements of Propylene Glycol and Heptanol Binary Mixtures

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ABSTRACT

Density (ρ), viscosity (η) and ultrasonic velocity (u) for the binary mixtures of propylene glycol and heptanol have been measured at temperatures $T=303K$, $313K$ and $323K$. This experimental data have been used to calculate acoustical parameters namely excess adiabatic compressibility (K_s^E), excess molar volume (V^E), excess intermolecular frelength (L_f^E), excess Gibb's energy (ΔG^*E), excess enthalpy (H^E) and excess internal pressure (π^E). The results were interpreted in terms of molecular interaction between the components of the mixtures. The observed negative and positive values of various excess and deviation parameters are attributed to the existence of strong molecular interactions. The computed values were fitted with the Redlich Kister polynomial equation to estimate the binary coefficients and standard deviation between experimental and calculated data at different temperatures and it is found that the error percentage is within the limits (2-3%)

Keywords:— excess Gibb's energy, excess internal pressure, free length, isentropic compressibility, hydrogen bonding.

I. INTRODUCTION

Grouping of solvents into species is often based on the nature of the inter-molecular forces because the manner where by solvent molecules are associated with every different combination brings about a marked impact at the resulting characteristics. After the creation of the concept of ionization strength of solvents effect on the rate equilibrium processes [1-2] due to the close connection among liquid structure and macroscopic characteristics, evaluation of density, viscosity and ultrasonic speeds is precious to learn about the liquids [3-4]. The results molecular interactions studies plays an important role in analyzing some of the applications such as drug synthesis, biological activities of the system, pharmaceutical and petro chemicals and cosmetics. The variant of these excess values with temperature and

composition for mixtures containing polar molecules and hydrogen bonded components may be complicated because of a lower or higher in hydrogen bonding interaction due to mixing, depending upon the type of the organic liquids whether they may be polar or non-polar, the signs and magnitudes of these excess values can throw light at the strength of interactions. Considerable work has been done on alcohols as one of the aspect in binary and ternary mixtures. The molecules containing –OH group easily forms associative groups due to hydrogen bonding. Now-a-days, propylene glycol is getting used for the synthesis of nanoparticles. Propylene glycol acts as reducing agent in addition to stabilizing medium. The prevailing research involves the measurements of ultrasonic speed, density and viscosity in certain binary mixtures. From the measured values, acoustic and thermodynamic parameters had been calculated to become aware of the thermodynamic stability inside the combinations. The excess parameters are computed to take a look at the energy of interactions among the molecules. Theoretical model velocities also are investigated for the binary systems to verify the type of interplay and to pick out the solute – solvent interactions. An attempt has been made to establish that the ultrasonic research is a successful approach to pick out the precise interactions in certain binary systems [5]. To examine of thermodynamic properties of multi factor liquid mixtures and records on the evaluation in terms of different models are needed for commercial and pharmaceutical utilization [6]. The excess thermodynamic capabilities [7] are sensitively dependent not most effective on the variations in intermolecular forces, but also at the differences within the length of the molecules. We under took investigations at the thermodynamic and transport properties of binary mixtures concerning propylene

glycol and heptanol in this article, we report the values of compressibility (K_s^E), excess molar volume (V^E), excess intermolecular frelength (L_f^E), excess Gibb's energy (ΔG^{*E}), excess enthalpy (H^E) and excess internal pressure (π^E) at $T = 303K, 313K,$ and $323K$ over the entire range of composition. The results of excess values have been fitted to the Redlich–Kister [8] polynomial equation. The intermolecular interactions had been estimated in the light of these excess parameters. Theoretical speed of sound and viscosity values have been evaluated using numerous empirical relations within the liquid mixture Propylene glycol + heptanol at $T/K = 303-323K$. This kind of evaluation of theoretical speed of sound values proves to be useful to affirm the applicability of numerous postulates of these theories of liquid mixtures and to arrive at a few useful inferences regarding the strength of molecular interactions among liquid mixtures in a few cases.

Chemical substances studied in this article are, propylene glycol and heptanol of AR grade received from Merck Co. Inc., Germany, with mass fraction purity extra than 0.99. All of the chemical compounds were similarly purified by way of standard methods [9] and most effective middle fractions have been accrued. The purity analysis of the chemicals used within the present study is given in Table 1 and purity of the compound was checked by using gas chromatographic analysis. The density and velocity of sound had been experimentally determined at different temperatures 303-323K and compared to literature values [10-17].

The equimolar aggregate of (propylene glycol+heptanol) is first prepared. The mass measurements have been completed on a digital electronic balance (Mettler Toledo AB a hundred thirty five, Switzerland) with an accuracy of ± 0.01 mg.

Table 1: The Purity Analysis of the Chemicals

Chemical name	Initial fraction	Purification method	Final fraction
1Heptanol	0.99	Distillation	0.99
Propylene Glycol	0.99	Distillation	0.99

The liquid mixtures have been prepared simply before the use. The uncertainty in mole fraction was expected to be much less than ± 0.0001 . The viscosities were measured with Ostwald viscometer. The viscometer became calibrated at different temperatures using redistilled water. The uncertainty in viscosity measurement is as much as 0.001 mPa s . The flow time has been measured after the attainment of bath temperature by each mixture. The float measurements have been made with a digital stop watch with a precision of 0.01 s . For all of the pure components and mixtures, three to 4 readings were taken and the common of these values had been used in all the calculations. The densities of the natural compounds and their mixtures have been determined accurately with the use of 10 mL specific gravity bottles in digital electronic balance (Mettler Toledo AB 135, Switzerland) with an accuracy of $\pm 0.01 \text{ mg}$. The average uncertainty in the measured density turned into $\pm 0.0001 \text{ kg m}^{-3}$. The velocity of sound changed into measured with a single crystal variable path

interferometer (Mittal establishments, New Delhi, India) running at a frequency of two MHz that had been calibrated with water and benzene. The uncertainty in the velocity of sound was observed to be $\pm 0.5 \text{ ms}^{-1}$. In all property measurements, the temperature turned into controlled within $\pm 0.1 \text{ K}$ using a regular temperature bath (M/s Sakti medical contraptions organization, India) via circulating water from the thermostat.

III. THEORY

Comparison of experimental densities (ρ), viscosities (η) and speed of sound (U) pure liquids with literature values are given in Table 2.

The experimental decided values of Ultrasonic speed, density, viscosity for the binary combination of propylene glycol + heptanol at $T=303 \text{ K}$, 313 K , and 323 K over the complete composition range is given in Table 3. In the present work, the authors calculated the extra values of isentropic compressibility and excess free length values to check the applicability of thermodynamical ideality (the perfect blending rules) to the components underneath study.

The excess values of isentropic compressibility K_s^E were calculated as follows

$$K_s^E = K_s - K_s^{\text{id}} \dots \dots \dots (1)$$

where K_s represent the calculated value of isentropic compressibility for the mixture

Table 2 Comparison of Experimental Densities (ρ), Viscosities(η) and Speed of Sound (U) Pure Liquids with Literature Values.

Liquid	T/K	$10^{-3} \rho / (\text{Kg.m}^{-3})$		$10^3 \eta / (\text{Kg m}^{-1} \text{ s}^{-1})$		$U / (\text{m.s}^{-1})$	
		Exp	Literature	Exp	Literature	Exp	Literature
1-Heptanol	303	0.8187	0.8149[30]	5.6331	6.188 [30]	1336	1312[30]
Propylene Glycol	303	1.0307	1.030[31]	23.19[31]	22.4	1604	1624[31]

$$K_s = \frac{1}{\rho U^2} \dots \dots \dots (2)$$

K_s^E is its excess value, K_s^{id} is the ideal isentropic compressibility value, ρ is the density and U represents the speed of sound. K_s^{id} for an ideal mixture was calculated from the relation recommended by Benson and Kiyohara [18, 19] and Douheret et al [20]

$$K_s^{id} = \sum \phi_i \left\{ K_{s,i}^0 + \frac{TV_i^0 (\alpha_i^0)^2}{C_{p,i}^0} \right\} - T \left(\sum x_i V_i^0 \right) \left(\frac{\sum \phi_i \alpha_i^{02}}{\sum x_i C_{p,i}^0} \right) \dots \dots \dots (3)$$

in which $K_{s,i}^0$, V_i^0 , α_i^0 , $C_{p,i}^0$ are the isentropic compressibility, molar volume, isobaric thermal expansion coefficient and molar isobaric heat capacity of pure component i , T represents temperature, f_i is the volume fraction and x_i represents the mole fraction of i in the mixture.

The density values have been used to calculate the excess volumes, V^E , using the following equation,

$$V^E = \frac{x_1 M_1 + x_2 M_2}{\rho} - \left(\frac{x_1 M_1}{\rho_1} + \frac{x_2 M_2}{\rho_2} \right) \dots \dots \dots (4)$$

where ρ is the density of the mixture and x_1 , M_1 , and ρ_1 and x_2 , M_2 , and ρ_2 are the mole fraction, molar mass, and density of pure components 1 and 2, respectively.

The excess values of free length (L_f^E), Gibbs free energy (DG^{*E}) and enthalpy (H^E) were calculated by using the expressions given in literature [21-29] as follows,

$$L_f^E = L_f - K_T (K_s^{id})^{1/2} \dots \dots \dots (5)$$

where L_f represents the calculated value for the mixture and K_T represent a temperature dependent constant whose value is $K_T = (91.368 + 0.3565T) \times 10^{-8}$.

Table 3: Experimental values of densities (ρ) a speed of sound (U) and viscosities (η)a for the liquid mixtures at T = 303,313,323K

$X1+X2$	$\rho / \text{Kg m}^{-3}$	$U / \text{m.s}^{-1}$	$\eta / 10^{-3} \text{Kg m}^{-1} \text{s}^{-1}$
Propylene Glycol(1)+Heptanol(2) at T=303K			
0.0000	0.8187	1336	5.3124
0.1703	0.8257	1354.6	6.6933
0.316	0.844	1378.2	7.8833
0.4419	0.8683	1394	9.0707
0.5519	0.8871	1411.2	10.6965
0.6488	0.9186	1420.6	12.2657
0.7348	0.9295	1454.2	14.2312
0.8117	0.9371	1490.7	16.6242
0.8808	0.9661	1531.5	18.5445
0.9432	0.9909	1572.7	21.0247
1	1.0307	1604	23.1899
Propylene Glycol(1)+Heptanol(2) at T=313K			
0	0.8102	1288.3	5.3124
0.1703	0.8203	1298.6	6.4413
0.316	0.8307	1322	7.6931
0.4419	0.8511	1333	9.0845
0.5519	0.8667	1351	10.3212
0.6488	0.8801	1366	11.8463
0.7348	0.8993	1389.1	14.0715
0.8117	0.9213	1411.6	15.7344
0.8808	0.9465	1437.4	17.434
0.9432	0.9886	1460.3	19.2147
1	1.0121	1498.6	20.1131
Propylene Glycol(1)+Heptanol(2) at T=323K			
0	0.8067	1239.3	4.3714
0.1703	0.8145	1259	5.2321
0.316	0.8293	1264.3	6.3024
0.4419	0.8404	1270	7.2817
0.5519	0.8529	1280.6	8.3422
0.6488	0.8067	1239.3	4.3714
0.7348	0.8864	1300	11.6365
0.8117	0.9094	1311	12.7834
0.8808	0.9273	1332	13.9373
0.9432	0.9529	1350.2	14.964
1	0.9924	1372.4	15.2435

Excess Gibbs free energy of activation ΔG^{*E} was calculated as follows,

$$\Delta G^{*E} = RT \left[\ln \left(\frac{\eta V}{\eta_2 V_2} \right) - x_1 \ln \left(\frac{\eta_1 V_1}{\eta_2 V_2} \right) \right] \dots\dots\dots(6)$$

Where R represents gas constant, T is absolute temperature, η is the viscosity of the mixture and η_1, η_2 are the viscosities of the pure compounds, V is the molar volume of mixture and V_1, V_2 are the molar volumes of the pure compounds,

Excess enthalpy H^E was calculated from usual relation.

$$H^E = H - (x_1 H_1 + x_2 H_2) \dots\dots\dots(7)$$

Where H represents the calculated value of enthalpy for the mixture and H_1, H_2 represent enthalpy of pure components 1 and 2, respectively.

Excess internal pressure was calculated by using the relation

$$\pi^E = (\pi_i - (x_1 \pi_1 + x_2 \pi_2)) \dots\dots\dots(8)$$

The excess molar volumes and deviations in viscosity were fitted to Redlich and Kister [8] equation of the type

$$Y = x_1 x_2 \sum_i^n a_i (x_1 - x_2)^i \dots\dots\dots(9)$$

where Y is $K_s^E, L_f^E, V^E, G^E, H^E, \pi^E$ and x_1, x_2 are mole fractions

In each case, the optimum number of coefficients is ascertained from an examination of the variation in standard deviation (σ).

σ was calculated using the relation

$$\sigma(Y^E) = \left[\sum (Y_{obs}^E - Y_{cal}^E)^2 / n - m \right]^{1/2} \dots\dots\dots(10)$$

where n represents the number of experimental points and m is the number of adjustable parameters.

IV. RESULT AND DISCUSSIONS

The experimental values of velocity of sound, density and viscosity in case of the binary liquid combinations of propylene glycol +heptanol below study over the entire variety of composition and at distinct temperatures, T = (303, 313 and 323) K are given in Table 3. From the records of velocity of sound, density and viscosity, the values of excess isentropic compressibility (K_s^E), excess molar volume (V^E), excess free length (L_f^E), excess Gibbs free energy (ΔG^{*E}), excess enthalpy (H^E) and excess internal pressure (π^E) were calculated. These excess parameters have been plotted against mole fractions one after the other over the entire range and at specific temperatures. The plots are shown in Fig 1-6.

The excess parameters of isentropic compressibility (K_s^E), excess molar volume (V^E), excess free length (L_f^E), excess Gibbs free

energy (ΔG^{*E}), excess enthalpy (H^E) and excess internal pressure (π^E) were fitted to the Redlich-Kister kind polynomial equation and the coefficients A_i evaluated with the aid of the technique of least squares, along with standard deviation (σ) are given in Table 4. The deviations found in the excess parameters indicate the strength of interactions present among the issue molecules of the liquid combinations under study [32]. The variations in these excess parameter values reflect the interactions among the integration species, depending upon the composition, molecular

shapes and sizes of the additives and temperature. The outcomes which have an effect on the values of excess thermodynamic functions may be the result of physical, chemical and structural contributions along with deviations in adiabatic compressibility can be explained in terms of contributions made by the following factors: (a) Increase in free length due to loss of dipolar association, difference in size and shape of the component molecule which leads to decrease in sound velocity and increase in adiabatic compressibility and (b) Decrease in free lengths as a result of dipole-dipole interactions or hydrogen bonded complex formation between unlike molecules which leads to decrease of sound velocity and increase of compressibility.

It is reported that negative deviations in compressibility indicates strong molecular interactions in liquid mixtures while positive sign is an indication of weak interactions due to dispersion forces. The positive deviation in compressibility for the system indicate the existence of weak interaction between the unlike molecules [33,34] As the alkanol chain increases the methylene groups increase, which produce greater steric hindrance to the formation of hydrogen bonds between. So, relatively weaker hydrogen bonding is observed in this mixture. The excess isentropic compressibility (K_s^E) for the liquid mixtures under study are as shown in Figure 1.

Table 4 : Coefficients of Redlich–Kister equation A_i , and standard deviations(σ), for adiabatic compressibility (K_s^E), excess molar volume (V^E), intermolecular frelength(L_f^E), excess Gibb's energy(G^E), excess enthalpy (H^E) and excess internal pressure(π^E)

Properties	A_0	A_1	A_2	A_3	A_4	σ
Propylene Glycol+heptanol at 303K						
$10^{-12} K_s^E / (m^2.N^{-1})$	186	-104.546	283.593	67.8	-330.104	1.79E-05
$10^6 V^E / (m.mol^{-1})$	20.3	-242.4	-549.1	-213	1204.6	0.004299
$L_f^E * 10^{-13} / m$	98.551	-55.5437	140.136	13.3047	-163.131	-4.14495E-06
$\Delta G^E / (J.mol^{-1})$	3350	-12010	-3214	-2040	102150	0.5554176
$H^E / (J.mol^{-1})$	-3530	-128450	-296370	-68070	682990	0.6327816
Π^E	-123600000	-803800000	-190250000	-82900000	4.17E+09	2155.6404
Properties	A_0	A_1	A_2	A_3	A_4	σ
Propylene Glycol+heptanol at 313K						
$10^{-12} K_s^E / (m^2.N^{-1})$	226.4949	-152.547	68.3169	185.4285	54.649	4.62E-06
$10^6 V^E / (m.mol^{-1})$	28.7	-284.2	-591.1	-129.4	1245.6	0.0070429
$L_f^E * 10^{-13} / m$	108.9074	-74.1852	38.3369	66.2571	17.2534	5.47482E-06
$\Delta G^E / (J.mol^{-1})$	4414	-1055	-26347	-7104	88251	0.0521508
$H^E / (J.mol^{-1})$	630	-116830	-260880	-114390	610380	0.5419871
Π^E	-122200000	-536400000	-1.44E+09	-785400000	3.35E+09	3527.447
Properties	A_0	A_1	A_2	A_3	A_4	σ
Propylene Glycol+heptanol at 323K						
$10^{-12} K_s^E / (m^2.N^{-1})$	273.8877	-93.2636	63.265	-41.6044	100.3869	4.18E-06
$10^6 V^E / (m.mol^{-1})$	33.5	-300.9	-692.9	-99	1464.5	0.0022876
$L_f^E * 10^{-13} / m$	116.0248	-46.4009	26.0484	-21.1147	52.6917	-5.77153E-06
$\Delta G^E / (J.mol^{-1})$	4834	-11904	-30120	-45520	96289	0.0524254
$H^E / (J.mol^{-1})$	18930	-138130	-328620	-90760	818300	0.4161192
Π^E	63600000	-709200000	-1.78E+09	-670400000	474390000	396.39346

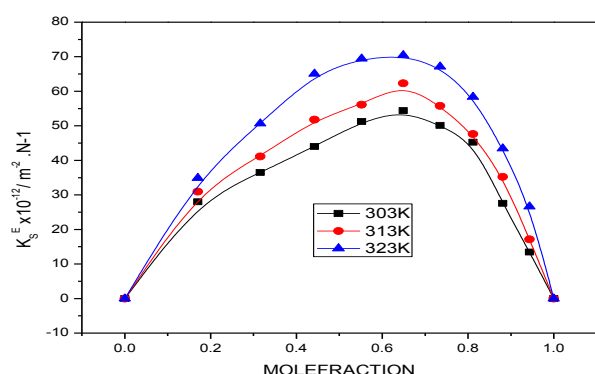


Figure 1: Temperature dependence of excess adiabatic compressibility (K_s^E)

From Figure 1. It can be observed that the K_s^E values are positive over the entire mole fraction range indicating the presence of weak interactions in these mixtures and also that the molecules are loosely packed in the mixtures due to their shape and size. The liquids having different molecular sizes and shape mix well there by reducing the volume which causes the values of K_s^E to be negative and if the liquids are of almost same size then they do not mix well and there is a lot of free space in between as suggested by Fort and Moore [35]. So, it can be said that the molecular interactions are weak in these liquid mixtures and that the medium is loosely packed. Similar results were observed by earlier workers [36]. The sign of excess volume (V^E) of a system depends on the relative magnitude of expansion/contraction on mixing of two liquids. If the factors causing expansion dominate the factors causing contraction, then V^E becomes positive. On the other hand, if the contractive factors dominate the expansive factors, then V^E becomes negative. The factors responsible for expansion in volume are as follows [37,38]:

1. Loss of dipolar association (i.e., rupturing of H-bonding of component by the other or breaking up of associates held by weaker forces, namely, dipole-dipole or dipole-induced dipole interactions or by Van der Waals forces.
2. The geometry of molecular structure, which does not allow

fitting of one component in to other component.

3. Steric hindrance, opposes the proximity of the constituent molecules. The negative V^E values arise due to the dominance of the following factors: (i) Chemical interaction between constituent molecules, such as hetero molecular associations through the formation of H-bond is known as strong specific interaction. (ii) Accommodation of molecules of one component into the other interstitials of molecules with each other component. (iii) Geometry of the molecular structure that favours fitting of the component molecules with each other. the sign and magnitude of the V^E values are sensitive to the carbon chain lengths of the 1-alkanol molecules.

From Figure 2, the V^E values are varying from negative to positive. This kind of behavior of V^E can attributed to the formation of H-bonds, disruption of alcohol self-associations and the structural characteristic geometrical fitting of one component into the other as a result of the increase in difference of size and shape of the component molecules [39]

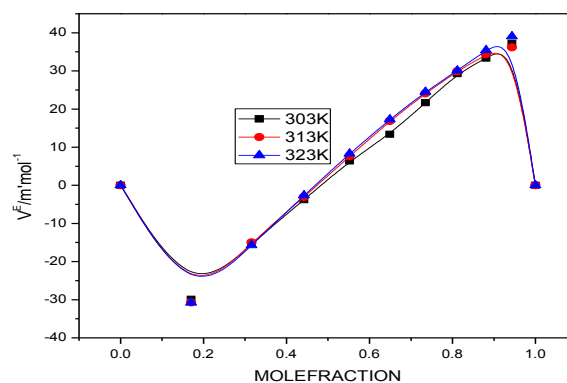


Figure 2: Temperature dependence of excess free volume (V_f^E)

From Figure 3. It could be discovered that the LfE values are positive which suggest that specific interactions are not present between unlike molecules in these systems [40].

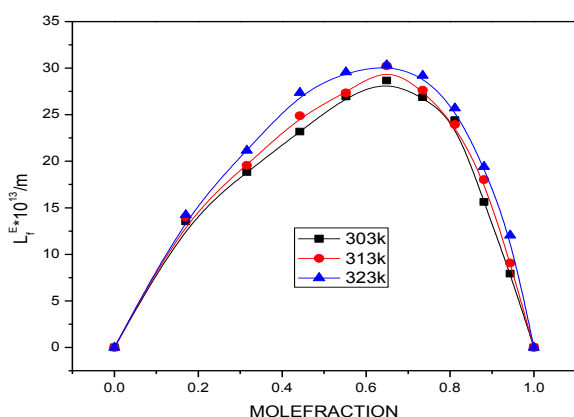


Figure 3: Temperature dependence of excess intermolecular frelength (L_f^E)

Figure 4, represent the excess Gibb's free energy of activation (ΔG^{*E}) with respect to mole fraction x_1 , over the entire composition range and at $T = (303, 313 \text{ and } 323) \text{ K}$.

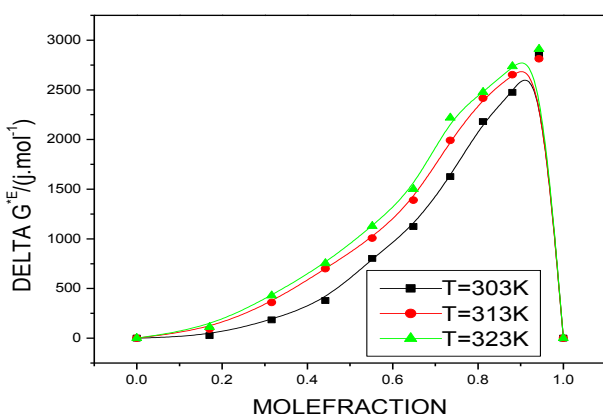


Figure 4: Temperature dependence of excess Gibb's free energy G^{*E}

It can be seen from Figure 4 that the majority ΔG^{*E} values are positive at all temperatures and over the entire range of mole fraction. These positive values indicate the existence of strong intermolecular interaction through hydrogen bonding between the component molecules of the liquid mixtures under study. The maximum deviation is observed for (Propylene glycol + heptanol) system. Similar results were observed by earlier workers [41].

From Fig 5 it's clear that the excess values of Enthalpy (H^E) are positive with respect to the mole fraction, x_1 , over the whole composition variety and at $T = 303, 313, 323 \text{ K}$.

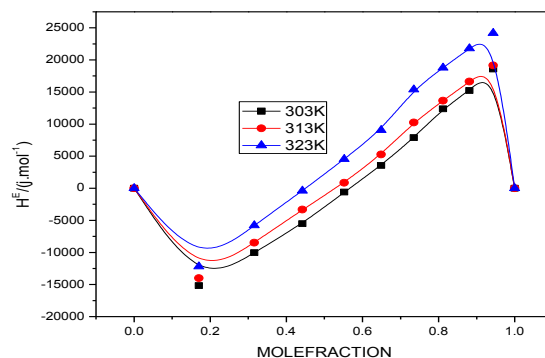


Figure 5: Temperature dependence of enthalpy H^E

The tremendous values of H^E insist the reality that there are strong unique interactions between unlike molecules in these liquid combinations [42]. The fine H^E values also recommend the lifestyles of inter molecular hydrogen bond and the breaking of related structures in case of Propylene glycol and heptanol.

From Figure 6 The values of unfastened volume inner strain increase with admire to growing the 1-heptanol awareness. Such as growing internal strain commonly indicates affiliation via hydrogen bonding [43]. So that you can constitute the presence of interaction a number of the liquid mixtures, it's miles crucial to take a look at the extra parameters.

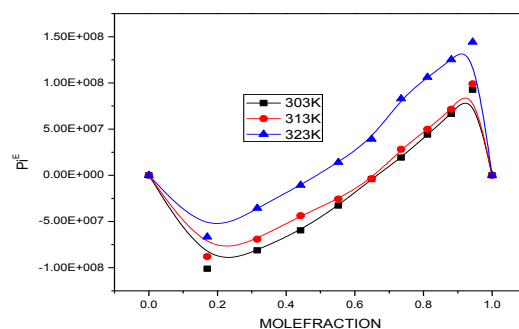


Figure 6: Temperature dependence of excess internal pressure (π^E).

IV. CONCLUSIONS

The values of K_s^E , V^E , L_f^E , ΔG^{*E} , H^E and π^E had been computed from the measured values of density, the velocity of sound and viscosity. Therefore by way of studying the variations in those parameters, the formation of hydrogen bond between the compounds is recognized. Wonderful values had been discovered within the case of excess isentropic compressibility, excess free length and excess Gibb's free energy, while values are varying from bad to high quality for excess Enthalpy, excess molar volume and excess internal pressure of activation. The presence of vulnerable bonding between the factor molecules may be showed with these values. Other than the above-stated values/reasons, the distinction in molar loads of the liquid molecules is likewise answerable for the non-existence particular interactions.

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