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# Acoustic and Volumetric Studies of the Binary Mixtures of Furfuryl Alcohol with Ethylene-, Diethylene- and Triethylene-Glycols at 303.15K

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## ABSTRACT

*Ultrasonic velocities and densities of the binary mixtures of furfuryl alcohol and ethylene glycol, diethylene glycol and triethylene glycol are experimentally determined over the entire composition range at room temperature of 303.15K at local atmospheric pressure. Using this acoustic and volumetric data, various thermodynamic and thermoacoustic parameters viz., molar volume, isentropic compressibility and mean free length useful for interpreting the intermolecular interactions between the furfuryl alcohol with glycol molecules are computed using the theoretical relations. To account for the nature and strength of the interactions, the excess parameters of these quantities are also determined. In order to check the consistency of the computed excess parameters, these parameters are fitted to Redlich-Kister type polynomial. The obtained negative values of excess parameters indicate that the interactions between furfuryl alcohol and glycol molecules are strong due to the hydrogen bonding between the molecules of the component liquids. The trend in the excess values indicate that the interactions between the unlike molecules of the liquid mixtures under study at room temperature follow the order: Furfuryl Alcohol+ Ethylene Glycol>Furfuryl Alcohol+ Diethylene Glycol>Furfuryl Alcohol+ Triethylene Glycol.*

## Highlights

- Ultrasonic velocity and density of furfuryl alcohol with ethylene glycol, diethylene glycol and triethylene glycol mixtures are measured at 303.15 K.
- Thermodynamic and excess parameters are computed theoretically.
- Strong interactions exist between the molecules of furfuryl alcohol and glycols due to hydrogen bonding.
- Among the three mixtures studied, the interactions between furfuryl alcohol and ethylene glycol are the strongest and between the furfuryl alcohol and triethylene glycol are the weakest.

**Keywords:** Ultrasonics, Thermodynamic parameters, Excess parameters, Molecular interactions.

## INTRODUCTION

Ultrasonic technique has become a powerful tool for studying the molecular behavior of liquid mixtures. This is because of its ability of characterizing physico- chemical behavior of liquid medium. The measurement of ultrasonic velocity has been adequately employed in understanding the molecular interactions in liquid mixtures. Molecular interaction studies can be carried out by both spectroscopic and non-spectroscopic techniques. Ultrasonic velocity and density measurements have been widely used in the field of interactions and structural aspect evaluation studies [**1-3**]. The ultrasonic studies are extensively used to estimate the thermodynamic properties and predict the intermolecular interactions of binary mixtures. The sound velocity is one of those physical properties that help in understanding the nature of liquid state. Thermodynamic and transport properties of pure and liquid mixtures are extensively used to understand the nature of interactions between the molecules. The derived parameters offer convenient method for the study of thermo dynamic properties of liquid mixtures not easily obtained by other means [**4, 5**].These structural arrangements are influenced not only by the shape of the molecules but also by their mutual interactions. The negative excess functions suggest the occurrence of discrete groups of molecules arranged into specific geometric structures. The positive values in excess properties correspond mainly to the existence of dispersion forces [**6, 7**]. Binary liquid mixtures rather than single component liquid system are widely used in processing and product formulations in many industrial applications. Among the various properties considered in process and product design and optimization excess molar volume and excess isentropic compressibility are useful in understanding the structural changes due to intermolecular interactions.

Production of biofuels and biochemical's furfural has gained increasing attention as a potential chemical and it is the most widely used chemical among the furan based chemicals for the production of furfuryl alcohol (resin production), foundry sand, lubrication oil extraction, and a novel application is as a nematicide [**8-10**]. The most important derivative of furfural is furfuryl alcohol [**11**]. Furfural alcohol is very important in polymer industry [**12**]. The availability of thermodynamic property data for furfuryl alcohol is limited in the literature whereas the number of other compounds produced in furfural production and within further upgrading of furfuryl alcohol is numerous [**13**]. A detailed search in the literature shows that very few experimental studies have been conducted on the physical properties of binary mixtures of furfuryl alcohol has been reported [**14, 15**]. In recent years, there has been considerable interest in experimental investigations of excess thermodynamic properties of binary liquid mixtures containing glycols [**16**].

Glycols have the activated hydrogen atoms of the hydroxyl groups, which are available for interaction with proton acceptors for the formation of hydrogen bonds. In the pure state, the glycols are usually self-associated through intermolecular hydrogen bonds forming aggregates, while on mixing with proton acceptor solvents, the networks of the self-associated molecules are disrupted and hetero-association takes place [**16**]. Glycols or diols are chemical compounds containing two hydroxyl (–OH) groups. Among the glycols, ethylene glycol(EG), diethylene glycol(DEG) and triethylene glycol(TEG) are homologous series of diols commonly used. The glycols have relatively low values of relative permittivity and dipole moment, yet they are self-associated through hydrogen bonding. Due to the physicochemical properties of solutions of glycols and their extensive applications in food, biotechnology, petroleum, oil, pharmaceutical and cosmetic industry, the thermoacoustic, volumetric and thermodynamic study of binary mixtures of furfuryl alcohol and glycols has become an interesting area of research. The literature survey indicates that studies on the acoustic and thermodynamic data of the binary liquid mixtures of furfuryl alcohol with ethylene-, diethylene- and triethylene- glycols are very rare.

In the present investigation, the experimental values of ultrasonic velocity and density of the binary mixtures of furfuryl alcohol with ethylene-, diethylene- and triethylene- glycols at room temperature (303.15K) at local atmospheric pressure are reported. Also, the computed thermodynamic

parameters along with the excess parameters are presented and the interactions between the molecules of the mixtures are explained in terms of the computed excess parameters.

## MATERIALS AND METHODS

For the preparation of various compositions of the liquid mixtures, Shimadzu AY120, a single pan electronic balance with an uncertainty of ±0.01 milligram is used. The uncertainty in the mole fraction is of $1\times10^{-4}$ and the prepared mixtures are stored in air-tight glass bottles. For the determination of the ultrasonic velocity and density, the liquids are thoroughly mixed before transferring into the apparatus used. The required properties are measured within 24 h of the preparation of the mixture. Ultrasonic velocity is measured with the help of single crystal variable path fixed frequency interferometer, Model-F05 supplied by Mittal Enterprises, New Delhi at a fixed frequency of 2MHz with an uncertainty less than 0.1%. Temperature of the mixture is kept constant for the measurement of speeds of sound using a microprocessor assisted circulating water bath, (supplied by Mac, New Delhi) regulated to ±0.01K .The densities of the mixtures are measured using a vibrating-tube Rudolph Research Analytical densitymeter, model DDM-2911 automatically thermostated within ±0.01K. The uncertainty in density measurements is $\pm1\text{x}10^{-5}$ g $cm^{-3}$. The information of chemicals used their purity and purification techniques are given in table 1.

**Table 1.** Details of studied chemicals, CAS number, source, purification method, chemical structure and molar mass

| Chemical | CAS number | Supplier | Purification Technique (Purity% GC) | Molar mass $10^{-3}$ kg $mol^{-1}$ |
|---|---|---|---|---|
| Furfuryl Alcohol | 98-00-0 | S.D Fine | Fractional distillation (99.8) | 98.10 |
| Ethylene Glycol | 107-21-1 | Merck | Fractional distillation (99.7) | 62.07 |
| Diethylene Glycol | 111-46-6 | Merck | Fractional distillation (99.5) | 106.12 |
| Triethylene Glycol | 112–27–6 | Merck | Fractional distillation (99.5) | 150.17 |

## RESULTS AND DISCUSSION

The data on some of the properties associated with the liquids and liquid mixtures like density and ultrasonic velocity find extensive applications in solution theory and molecular dynamics. Such results are necessary for interpretation of data obtained from thermochemical, electrochemical, biochemical and kinetic studies.

The experimentally measured ultrasonic velocity and density of the pure liquids and liquid mixtures of furfuryl alcohol with ethylene glycol, diethylene glycol and triethylene glycol at 303.15 K temperature are presented in Table 2. From the measured values of ultrasonic velocity and density, certain thermoacoustic and thermodynamic parameters viz., molar volume ($V_m$), isentropic compressibility ($\kappa_s$) and mean free length ($L_f$) useful for understanding the nature of the interactions between the furfuryl alcohol and glycol molecules are computed using the relations available in literature [**17**] and are reported in Table 2. It may be noted here that the thermodynamic parameters are computed using Benson-Kiyohara approach [**17**]. In order to explain the strength of the interactions between the component molecules, the excess parameters viz., excess molar volume ($V_m^E$), isentropic compressibility ($\Delta\kappa_s$) and mean free length ($L_f^E$) are also computed using the relation:

$$Y^E = Y_{mix} - (x_1Y_1 + x_2Y_2)$$

Where, Y is the parameter, $x_1$ and $x_2$ are the mole fractions of the components.

The above excess parameters ($Y^E$) are fitted to Redlich–Kister [17] type polynomial equation:

$$Y_{cal}{}^{E} = x_1 x_2 \sum A_{j-1}(x_2 - x_1)^{j-1}$$

Where, $A_{j-1}$ is adjustable parameters. The values of coefficient $A_{j-1}$ are evaluated by the method of least squares with all points weighted equally and the standard deviations are calculated using [17]:

$$\sigma_{Y^E} = \sum (Y_{\exp}{}^{E} - Y_{cal}{}^{E})^2 / (m-n)^{1/2}$$

Where, 'm' is the number of experimental data points and 'n' is the number of coefficients considered (n = 5 in the present study). The fitting coefficients of the Redlich–Kister polynomial ($A_{j-1}$) along with the standard deviations (σ) are presented in table 3.

**Table 2.** Experimental ultrasonic velocity (U/ms$^{-1}$), density (ρ/kgm$^{-3}$), and computed molar volume ($V_m$/10$^{-6}$ m$^3$mol$^{-1}$), isentropic compressibility ($\kappa_s$/10$^{-10}$ m$^2$N$^{-1}$), mean free length ($L_f$/10$^{-11}$ m),and corresponding excess thermodynamic parameters ($V_m{}^E$, $K_s{}^E$and $L_f{}^E$) for the binary mixtures of furfuryl alcohol+ethylene glycol, furfuryl alcohol+diethylene glycol and furfuryl alcohol+ triethylene glycol at a temperature 303.15 K

| $x_1$ | U | ρ | $V_m$ | $\kappa_s$ | $L_f$ | $V_m{}^E$ | $K_s{}^E$ | $L_f{}^E$ |
|---|---|---|---|---|---|---|---|---|
| **Furfuryl Alcohol (1) + Ethylene Glycol (2)** | | | | | | | | |
| 0.0000 | 1643.75 | 1106.88 | 56.0765 | 3.3437 | 3.7953 | 0.0000 | 0.0000 | 0.0000 |
| 0.0666 | 1620.89 | 1109.38 | 58.1129 | 3.4309 | 3.8445 | -0.0452 | -0.0122 | -0.0069 |
| 0.1383 | 1598.61 | 1111.79 | 60.3114 | 3.5196 | 3.8939 | -0.0889 | -0.0231 | -0.0127 |
| 0.2158 | 1576.83 | 1114.08 | 62.6929 | 3.6100 | 3.9436 | -0.1294 | -0.0321 | -0.0175 |
| 0.2997 | 1555.48 | 1116.23 | 65.2820 | 3.7027 | 3.9939 | -0.1645 | -0.0389 | -0.0209 |
| 0.3910 | 1534.50 | 1118.19 | 68.1083 | 3.7979 | 4.0449 | -0.1913 | -0.0431 | -0.0229 |
| 0.4906 | 1513.81 | 1119.93 | 71.2070 | 3.8964 | 4.0970 | -0.2059 | -0.0441 | -0.0231 |
| 0.5997 | 1493.35 | 1121.37 | 74.6206 | 3.9988 | 4.1505 | -0.2029 | -0.0412 | -0.0213 |
| 0.7198 | 1473.04 | 1122.47 | 78.4012 | 4.1058 | 4.2057 | -0.1749 | -0.0337 | -0.0172 |
| 0.8525 | 1452.79 | 1123.12 | 82.6132 | 4.2186 | 4.2630 | -0.1119 | -0.0203 | -0.0103 |
| 1.0000 | 1432.51 | 1123.24 | 87.3366 | 4.3384 | 4.3232 | 0.0000 | 0.0000 | 0.0000 |
| **Furfuryl Alcohol (1) + Diethylene Glycol (2)** | | | | | | | | |
| 0.0000 | 1568.73 | 1109.39 | 95.6562 | 3.6629 | 3.9723 | 0.0000 | 0.0000 | 0.0000 |
| 0.1085 | 1556.29 | 1111.48 | 94.6934 | 3.7146 | 4.0003 | -0.0602 | -0.0158 | -0.0085 |
| 0.2150 | 1542.90 | 1113.50 | 93.7552 | 3.7725 | 4.0314 | -0.1127 | -0.0254 | -0.0136 |
| 0.3194 | 1528.95 | 1115.35 | 92.8477 | 3.8353 | 4.0648 | -0.1508 | -0.0302 | -0.0160 |
| 0.4220 | 1514.73 | 1117.02 | 91.9728 | 3.9018 | 4.0999 | -0.1723 | -0.0313 | -0.0164 |
| 0.5227 | 1500.47 | 1118.49 | 91.1300 | 3.9711 | 4.1361 | -0.1773 | -0.0295 | -0.0153 |
| 0.6216 | 1486.31 | 1119.76 | 90.3178 | 4.0426 | 4.1731 | -0.1667 | -0.0256 | -0.0132 |
| 0.7188 | 1472.37 | 1120.86 | 89.5344 | 4.1154 | 4.2106 | -0.1421 | -0.0203 | -0.0104 |
| 0.8142 | 1458.72 | 1121.80 | 88.7777 | 4.1893 | 4.2482 | -0.1051 | -0.0140 | -0.0071 |
| 0.9079 | 1445.43 | 1122.58 | 88.0457 | 4.2637 | 4.2858 | -0.0572 | -0.0071 | -0.0036 |
| 1.0000 | 1432.51 | 1123.24 | 87.3366 | 4.3384 | 4.3232 | 0.0000 | 0.0000 | 0.0000 |
| **Furfuryl Alcohol (1) + Triethylene Glycol (2)** | | | | | | | | |
| 0.0000 | 1597.30 | 1116.15 | 134.5428 | 3.5116 | 3.8894 | 0.0000 | 0.0000 | 0.0000 |
| 0.1462 | 1580.14 | 1117.34 | 127.5887 | 3.5844 | 3.9296 | -0.0549 | -0.0098 | -0.0054 |
| 0.2780 | 1562.62 | 1118.55 | 121.3110 | 3.6613 | 3.9715 | -0.1064 | -0.0156 | -0.0085 |
| 0.3977 | 1545.07 | 1119.61 | 115.6323 | 3.7414 | 4.0147 | -0.1381 | -0.0182 | -0.0098 |
| 0.5067 | 1527.70 | 1120.50 | 110.4754 | 3.8239 | 4.0587 | -0.1499 | -0.0184 | -0.0098 |
| 0.6064 | 1510.66 | 1121.23 | 105.7727 | 3.9081 | 4.1032 | -0.1453 | -0.0169 | -0.0088 |
| 0.6980 | 1494.04 | 1121.82 | 101.4664 | 3.9935 | 4.1477 | -0.1287 | -0.0142 | -0.0074 |
| 0.7823 | 1477.89 | 1122.30 | 97.5077 | 4.0795 | 4.1922 | -0.1034 | -0.0109 | -0.0056 |
| 0.8604 | 1462.25 | 1122.69 | 93.8556 | 4.1658 | 4.2363 | -0.0722 | -0.0072 | -0.0037 |
| 0.9327 | 1447.12 | 1122.99 | 90.4751 | 4.2522 | 4.2800 | -0.0373 | -0.0035 | -0.0018 |
| 1.0000 | 1432.51 | 1123.24 | 87.3366 | 4.3384 | 4.3232 | 0.0000 | 0.0000 | 0.0000 |

The variation of experimental ultrasonic velocity and the density of the three binary liquid mixtures under study viz., furfuryl alcohol + ethylene glycol, furfuryl alcohol + diethylene glycol and furfuryl alcohol + triethylene glycol at room temperature 303.15 K with the mole fraction of furfuryl alcohol is pictorially shown in figures 1 and 2 respectively. If there were no interactions between the unlike molecules then the ultrasonic velocity in the mixture changes linearly. However, the observed nonlinear variation in ultrasonic velocity and density of the three liquid mixtures under study with the mole fraction of furfuryl alcohol suggest that some type of interactions exist between the molecules of furfuryl alcohol and glycols. The variation of ultrasonic velocity in a solution depends upon the increase or decrease of intermolecular free length, after mixing the components on the basis of a model for propagation proposed by Eyring and Kincaid [**18**], ultrasonic velocity should increase if the intermolecular free length decreases as a result of mixing of components. However, the nature and strength of the interactions cannot be established from the variation of these experimentally determined parameters. Molar volume ($V_m$), isentropic compressibility ($\kappa_s$) and mean free length ($L_f$) of the pure liquids (Table 2) are observed to increase with the increase in the concentration of furfuryl alcohol indicating intermolecular interactions between component molecules.

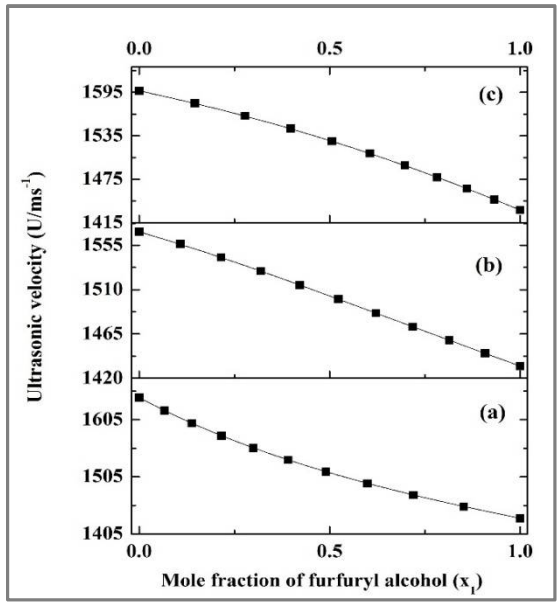


**Figure 1.** Variation of ultrasonic velocity (U/ms$^{-1}$) of (a) furfuryl alcohol+ethylene glycol, (b) furfuryl alcohol+ diethylene glycol and (c) furfuryl alcohol+triethylene glycol at 303.15K with the mole fraction of furfuryl alcohol.

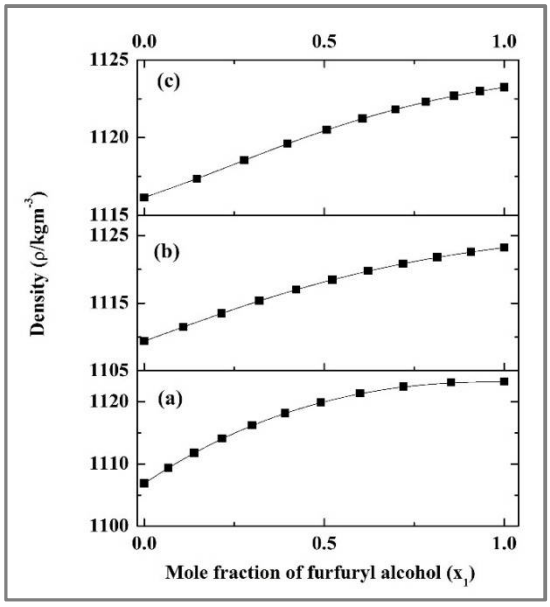


**Figure 2.** Variation of density (ρ/kgm$^{-3}$) of (a) furfuryl alcohol+ethylene glycol, (b) furfuryl alcohol+diethylene glycol and (c) furfuryl alcohol+triethylene glycol at 303.15 K with the mole fraction of furfuryl alcohol.

**Table 3.** Redlich-Kister coefficients and corresponding standard deviations of the excess parameters ($V_m^E$, $K_s^E$ and $L_f^E$) for the binary mixtures of furfuryl alcohol+ ethylene glycol (I),furfuryl alcohol+ diethylene glycol(II) and furfuryl alcohol+ triethylene glycol (III) at a temperature 303.15K

| System | $V_m^E$ | | | $\Delta K_s$ | | | $L_f^E$ | | |
|---|---|---|---|---|---|---|---|---|---|
| | I | II | III | I | II | III | I | II | III |
| $A_0$ | -0.8258 | -0.7104 | -0.5987 | -0.1761 | -0.1204 | -0.0738 | -0.0922 | -0.0627 | -0.0391 |
| $A_1$ | 0.0992 | 0.0102 | 0.0752 | -0.0223 | -0.0487 | -0.0129 | -0.0173 | -0.0281 | -0.0089 |
| $A_2$ | 0.0125 | 0.0799 | 0.1427 | -0.0031 | -0.0077 | 0.0077 | -0.0038 | -0.0051 | 0.0041 |
| $A_3$ | 0.0022 | 0.0500 | 0.0781 | 0.0003 | 0.0002 | -0.0001 | -0.0006 | -0.0001 | 0.0002 |
| $A_4$ | 0.0021 | 0.0194 | 0.0242 | 0.0008 | 0.0007 | 0.0010 | 0.0001 | 0.0004 | 0.0004 |
| σ | 0.0008 | 0.0003 | 0.0004 | 0.0002 | 0.0002 | 0.0003 | 0.0007 | 0.0001 | 0.0003 |

Figure 3 represents the variation of (a) excess molar volume ($V_m^E$) and (b) deviation in isentropic compressibility ($\Delta\kappa_s$) of the binary mixtures of furfuryl alcohol + ethylene glycol, + diethylene glycol

and + triethylene glycol with the mole fraction of furfuryl alcohol at 303.15K. From figure 3, it is observed that all the three excess parameters under investigation show a negative trend with the increase in the mole fraction of the furfuryl alcohol. It is well known that the values of the excess parameters would depend upon the relative strengths of several opposing effects that include physical, chemical and structural contributions [**19**]. The physical contribution consists of dispersion forces or weak dipole–dipole interactions that lead to positive contribution towards the excess parameters. Chemical contribution includes specific interactions such as the formation of hydrogen bonds, formation of charge transfer complexes and other complex forming interactions including strong dipole– dipole interaction between components molecules result in negative excess parameters.

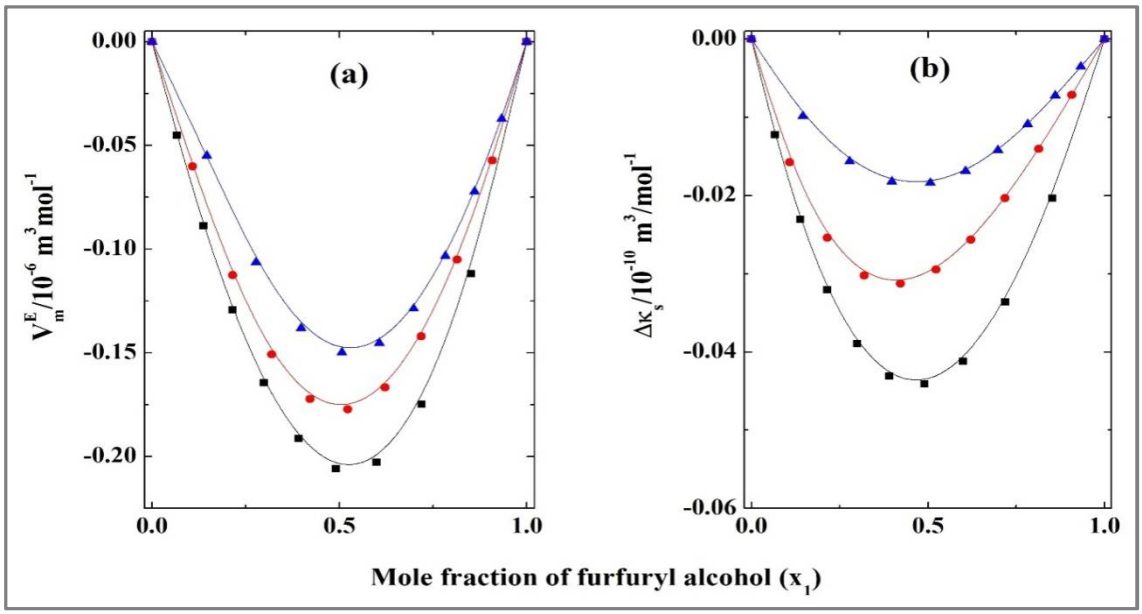


**Figure 3.** Variation of (a) excess molar volume ($V_m^E$) and (b) deviation in isentropic compressibility ($\Delta\kappa_s$) of the binary mixtures of furfuryl alcohol+ethylene glycol (■), +diethylene glycol (●) and +triethylene glycol (▲) with the mole fraction of furfuryl alcohol at 303.15 K.

The observed negative $V_m^E$ values (Figure 3) for all the three binary mixtures under study, indicate that the mixture is less compressible than the corresponding ideal mixture, suggesting significant interactions between the unlike component molecules, i.e., the structure of the component FFA has been disrupted by the addition of glycol. This is probably due to (i) the breakdown of self-associated structures of FFA and glycol and (ii) intramolecular hydrogen bond formation in FFA and glycol molecules. Also, the absolute value of $V_m^E$ is maximum around $0.49x_1$ for FFA+EG, around $0.47x_1$ for FFA+DEG and around $0.51x_1$ for FFA+TEG mixtures showing the dependence of the maxima on the size of the second component (glycol) in these binary mixtures. The observed values of $V_m^E$ can be explained in terms of dipole–dipole interactions and geometrical contribution between unlike molecules which leads to increase of speed of sound and decrease of isentropic compressibility as observed in the present investigation. This fact is further substantiated by the observed negative values of $\Delta\kappa_S$ (Figure 3) and $L_f^E$ (Table 2) with composition of with the composition of FFA in all these mixtures. Similar results are reported by Pal *et al*. [**20**] for EG, DEG and TEG with 2-butoxyethanol. The observed trend in the excess values indicates that the interactions between the unlike molecules of the liquid mixtures under study at room temperature follow the order: Furfuryl Alcohol + Ethylene Glycol > Furfuryl Alcohol + Diethylene Glycol > Furfuryl Alcohol + Triethylene Glycol.

## APPLICATION

Thermodynamic and acoustic properties of pure liquids and liquid mixtures of FFA and glycols are useful in the engineering applications concerning heat transfer, mass transfer and fluid flow. In chemical process industries, materials are normally handled in fluid form and as a consequence, the physical, chemical and transport properties of fluids, assume importance.

## CONCLUSION

In the present study, the data of experimental ultrasonic velocity and density is reported for binary mixtures of furfuryl alcohol+ethylene glycol, furfuryl alcohol+diethylene glycol and furfuryl alcohol+triethylene glycol at 303.15K at local atmospheric pressure. The theoretically computed molar volume, isentropic compressibility and mean free length along with their excess parameters are useful in interpreting the nature and strength of molecular interactions between FFA and glycol molecules. Finally, it can be concluded that strong interactions due to the hydrogen bonding exist between the FFA and glycol molecules.

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