1.1 Introduction

Thermodynamic and physico chemical properties of binary mixtures are studied to emphasize intermolecular interactions present in the liquid phase. Mixed solvents rather than pure solvents find practical application in most chemical, industrial, engineering and technological processes. The thermodynamic properties obtained by experimental data are the essential basis for the development of empirical, semi-empirical or theoretical models employed to represent and predict the behavior of fluids. The volumetric properties of solutions have proven to be a very useful tool in elucidating the structural interactions because they provide an indirect insight into the conformational feature of the components in solution. Precise measurements of densities of solutions have immense significance in chemical engineering design, process simulation, solution theory and molecular dynamics [1, 2].

1.1.1 Types of interactions in liquid mixtures

Liquid-liquid mixtures have attracted considerable attention due to their unusual behavior which depend upon the intermolecular interactions between the unlike molecules. In general there are solute-solute, solute-solvent, solvent-solvent types of interactions between components in liquid mixtures, which are responsible for non ideal behavior of these mixtures. These interactions reflect deviations / excess values of several parameters of the liquid mixtures. The strength and type of these interactions are evaluated by measuring and analyzing various acoustic and thermodynamic parameters of the liquid mixtures. Excess functions are favorable for understanding the types of interactions between components of mixtures.

There are many types of attractive or repulsive electrostatic interactions normally observed between the molecules of same or different solvents when they are mixed. They are

1. Ion-ion interactions
2. Ion-dipole interactions
3. Dipole-dipole interactions (Keesom forces)
4. Dipole-induced dipole interactions (Debye forces)
5. Induced dipole-induced dipole interactions (London dispersion forces)
6. Hydrogen bonding

Ion-ion interactions exist between point charges which are considered to be strongest. Ion-dipole interaction is a force of attraction between a point charge and dipolar molecule. Dipole-dipole interactions (keesom forces) result when two dipolar molecules interact with each other through space. When this occurs, the partially negative portion of one of the polar molecules is attracted to the partially positive portion of the second polar molecule. Dipole-induced dipole interactions are due to the presence of the partial charges of the polar molecule which causes a polarization or distortion of the electron distribution of the other molecule. As a result of this distortion, the second molecule acquires regions of partial positive and partial negative charge, and thus it becomes polar. Hence, a molecule may induce a dipole on neighbouring molecule and causes mutual attraction. The London dispersion force is the weakest and a temporary attractive force that results when the electrons in two adjacent atoms occupy positions that make the atoms form temporary dipoles. This force is sometimes called an induced dipole-induced dipole attraction. Ion-dipole, dipole-dipole, dipole-induced dipole, induced-induced dipole interactions are collectively known as Van der Waals interactions. Van der Waals interactions are the weakest of all intermolecular attractions between molecules.

Hydrogen bond plays an important role in chemical, physical and biological processes. Hydrogen bond is the electrostatic attractive force between the hydrogen attached to an electronegative atom of one molecule and an electronegative atom of another molecule. Usually the electronegative atom is oxygen, nitrogen, or fluorine, which has a partial negative charge. When the hydrogen bond, is between two atoms of the same molecule, it is intramolecular hydrogen bonding and if two atoms from different molecules form a hydrogen bond, it is called intermolecular hydrogen bonding. Obviously, one of the atoms in hydrogen bonds has to be a H atom.

A hydrogen atom has only one ‘s’ orbital, which becomes saturated after the formation of one covalent bond thus making it incapable of forming a second covalent bond. However, the positive hydrogen can polarize the lone pair orbital of another atom, delocalize the lone pair of electrons and thereby form a weak long covalent bond. Thus hydrogen bond is a simple case of dipole-dipole (or ion-dipole) attraction. Hydrogen bonds have also been found to exist between a carbon atom and an electronegative atom provided there are some electronegative atoms attached to the

carbon atom to activate the hydrogen atom (eg.$CHCl_3$). Hydrogen bond formation between the molecules of a liquid mixture, influences the thermodynamic properties of solutions to a greater extent.

Hydrogen bonds have the following special features.

1. These are weak in nature, for example, they have energy approximately 2-10 Kcal mol^{-1} where as an ordinary covalent bond has energy of about 80-100 Kcal $mol^{-1.}$
2. Increase in electro-negativity of an atom increases its power of forming hydrogen bonds.
3. Hydrogen ion in a hydrogen bond is not centrally situated but is nearer to one of the two adjacent electronegative atoms.

The hydrogen bond is a weak bond but it has great significance in determining the properties of substances because of its small bond energy and the small activation energy which are majorly accountable in its formation and dissociation. The physical properties of a substance or a mixture of substances are determined by the strength of these intermolecular interactions. The strength of these interactions and the forces among constituent particles can be analyzed by their thermodynamic and kinetic behavior [3-7].

To understand the type of interactions present between the molecules of two different solvents, systematic study of excess thermodynamic parameters like molar volumes, isentropic compressibility, Gibb's free energies, free lengths, viscosities are very much useful. For these, ultrasonic velosity, density and viscosity measurements are required. The study of ultrasonic velocities finds many applications in the fields of medicine, biology, chemistry and physics.

Determination of excess molar volumes is very useful to predict the ideality or non ideality of mixture of solvents. Since the molar volumes of mixtures reveal the type of forces (weak or strong) acting between the particles, we can predict the type of interactions operating between the components in the mixture. The measurement of densities is required for the determination of excess molar volumes. These calculations have much significance in designing biological and chemical engineering processes [8-11]. Not only excess molar volume but also information of other thermodynamic properties of solutions may be used to observe the association

between the internal structure of the system, nature of intermolecular interactions and the physical properties of solvents [12-20].

1.1.2 Thermo acoustic studies of molecular interactions:

Thermo acoustic studies of inter molecular interactions are very useful in chemical and food processing, material testing etc. The phenomena of ultrasonic vibrations are commonly used in mechanical machinery applications [21], preparation of colloids, imaging of biological tissues, pre-germination of seeds. From the knowledge of ultrasonic velocity, density, and viscosity of a liquid various acoustic parameters such as intermolecular free length, isentropic compressibility, internal pressure etc., can be obtained.

Plenty of research data is available in reviews by Hertzfeld and Litovitz [22], Nozdrev [23] and also in the volumes edited by Flugge [24], Bergmann [25], Mason [26], Beyer and Letcher [27]. Ultrasonic studies are useful in characterizing physico chemical behavior of liquid mixtures. Fort and Moore [28], Tumikoski and Nurmi [29], Flory and coworkers [30] have studied the non-ideal behavior of binary liquid mixtures. Ultrasonic studies have been carried out to find out ion-solvent interactions in electrolytic solutions containing different electrolytes and found that ultrasonic speed in these solutions depend on the size of the ion, polarity of the solvent and to some extent on the electronic configuration of transition metal ions [31].

The excess thermodynamic functions introduced by Scatchard in the year 1931, provided a way to represent directly the deviation of solution from ideal behavior. The difference between the thermodynamic function of mixtures for a real system and the value corresponding to a perfect solution at the same temperature, pressure and composition is called the excess thermodynamic parameter denoted by super script E.

Thus any excess parameter Y^E is given by

$$Y^E = Y^M{}_{real} - Y^M{}_{ideal}$$

Many theories for ideal solutions were developed to predict the properties of liquid mixtures and independently observable properties of pure components. These theories are formulated to account for the departure of a real solution from the ideal behavior. Theories concerning excess volumes of binary liquid mixtures were thoroughly reviewed and discussed by Rowlingson [32-34], Flory [35], Scott and

Fenby [36], Baattino[37], Kehiaian [38], Handa and Benson [39] Hijmens and Holleman [40].

The theories of binary liquid mixtures were proposed by Van der Waals [41] and Van Laar [42]. These theories successfully explained certain excess properties in critical region of liquid mixtures. In an attempt to improve Van Laar's theory, Hildebrand and Scott [43] and Scatchard [44-45] used Hildebrand's [46] concept of regular solutions to formulate a relation for excess volume. Eyring [47] developed the cell model, which was extensively used by Lennard-Jones and Devonshire [48-49] and Prigogine *et al.* [50]. This model relates the thermodynamic properties of liquid mixtures to the intermolecular energy parameters. Prigogine and coworkers have used the cell model to extend the theory of corresponding states to chain molecules. In this approach the chain molecules were considered as a series of quasi-spherical segments. Redlich and Kister [51] proposed an empirical equation to predict Y^E values for binary mixtures.

$$Y^E = X_1X_2[A_0 + A_1(X_1 - X_2) + A_2(X_1 - X_2)]$$

Where A_0, A_1 and A_2 are constants and X_1 and X_2 are the mole fractions of the two components. Extensive studies of the ultrasonic velocity in liquids and liquid mixtures and their interpretation in the light of molecular structure were also made by several investigators [52]. The relation between the molecular structure and the ultrasonic values was studied by different researchers [54-56]. Various studies on ultrasonic velocity measurements in a large number of organic liquids, the following general conclusions were drawn about the dependence on molecular structure.

1. Liquids having higher density give lower ultrasonic velocity but not necessarily in proportion.
2. Aromatic compounds have usually higher velocities than the aliphatic compounds even though the density of aromatic compounds is higher.
3. A decrease in velocity is observed by substitution of a heavier atom in place of lighter atom.
4. A double bond of unstauration in liquids is found to be resulting in low velocity.
5. Long molecules generally give rise to higher velocity even though their density is higher.

6. Polar molecules have higher velocities in alcohols, ketones, acetic anhydride, nitrobenzene, aniline, acetophenone, cyclo hexanol and water.
7. In non-polar groups, the velocity of amines and alcohols will not differ much and increase with increase in chain length.
8. In isomeric amines, the branched amines exhibit greater velocity than those of the straight chains.

1.1.3 Volumetric studies of specific interactions:

Density is an important volumetric property. The volumetric properties obtained from the measurements of density with components suggest qualitative assessment of the behavior of liquid mixtures. It can be used for the evaluation of the excess thermodynamic properties of the binary liquid mixtures, which provide information on the extent of specific intermolecular interactions between the components in binary systems.Volumetric properties offer significant information for the characterization of molecular interactions. Thermodynamic studies of liquid mixtures are very important for the design of separation processes of liquids. The nature and relative strength of molecular interactions operating among the components of liquid mixtures have been successfully predicted by computing their thermodynamic properties like excess molar volumes and partial molar volumes. Partial molar properties of dilute solutions provide information about the molecular interactions between solute – solute and solute – solvent molecules and can be adopted for the development of molecular models for describing the dynamic behaviour of solutions. The results of excess molar volume can be substantiated by studying partial molar volumes.

1.1.4 Viscometric studies of liquid mixtures:

Viscosity is a transport property, which depends on the transport of momentum that moving molecules offer to the neighboring molecules in the flow process of a fluid. Knowledge of viscosity of liquid mixtures at ambient and higher or lower temperatures is required in several industrial computations and in analytical sciences. Determination of flow in pipelines and capillaries, heat transfer and mass transfer operations, liquid and semi solid pharmaceutical formulation processes, developing separation methods like HPLC and capillary electrophoresis could be considered as the example applications of viscosity information. In the

pharmaceutical area, the sedimentation rate of suspensions, creaming of emulsions and drug release from aqueous solutions, are the important processes, which could be affected by the viscosity. The dynamic viscosities of the liquid mixtures can be evaluated using semi empirical formulae given by Grunberg-Nissan, Katti-chowdary, Heric Brewer, HIind *et al.* and McAllister fourbody model. They provide strong evidence to experimental values.

1.1.5 FT-IR spectroscopic studies:

FT-IR spectroscopy has been extensively used to study the intermolecular hydrogen bonding interactions between the binary liquid mixture. FT-IR spectroscopy is a successful method to explore the molecular structure of association effects among molecules. This technique offers the advantages to measure the association properties and hydrogen bonding capability to assess interactions between molecules by analyzing the band shape and shifts. The position of the peak or the band not only confirms the presence of a particular group but gives a good idea about the environment affecting the group and shapes. Greatest use of IR has been in interpreting the both intramolecular and intermolecular hydrogen bonding. The FT-IR spectroscopy gives additional evidence for observed strong interaction through acoustic study. Various acoustic, thermodynamic and transport properties, theoretical velocities, theoretical viscosities and spectroscopic investigations are carried out to pertain the molecular interactions among the molecules.

1.2 Review of Literature

A brief review of literature on the liquid- liquid mixtures is presented below. Many researchers studied the molecular interaction of binary liquid mixtures.

J.V. Srinivasu *et al.* [57] measured speeds of sound and densities of binary mixtures of 1,4-butanediol (1,4-BD) with methylpyridine isomers (α-picoline and β-picoline) and measured over the entire range of composition at temperatures T=(303.15,308.15,313.15 and 318.15) K and at atmospheric pressure**.** Ultrasonic velocity, density and viscosity have been measured in the binary liquid mixture of 1,4-dioxane with n-hexane or n-heptane or n-octane at 308.15,313.15, and 318.15 K over the entire composition range by Sk. Md. Nayeem [58]. C. Narasimha Rao *et al.* [59] worked on excess thermodynamic and spectral properties of ternary mixtures containing N-methylcyclohexylamine, bromobenzene and 1-alkanols. Gyanendra Sharma *et al.* [60] measured the apparent molar properties of aqueous protic ionic

liquid solutions namely diethylethanolammonium propionate at T = (293.15 to 328.15) K. L. Venkatramana [61] studied the acoustic, volumetric, transport and thermal properties of hydroxyl ammonium based ionic liquids namely, N-butyl-(N-hydroxyethyl) ammonium trifluoroacetate and N-butyl-(N-hydroxyethyl) ammonium nitrate.Vickramjeet Singh *et al.* [62] measured speed of sound and apparent molar isentropic compression of 1-butyl-3-methylimidazolium bromide in aqueous monosaccharide solutions. M. Srinivasa Reddy *et al.* [63] assessed molecular interactions in binary liquid mixtures of [Emim][BF4] with 2-methoxyethanol using thermo acoustic, volumetric and optical properties.

R.Ezhil Pavaia *et al.* [64] conducted molecular interaction studies by the measurement of velocity,density and viscosity for two ternary liquid mixtures of aniline+ toluene+ methyl isobutyl ketone and N-methylaniline + toluene+ methyl isobutyl ketone at (303, 308 and 313) K. Ultrasonic velocities, densities and viscosities of the binary liquid mixtures of butan-2-one with benzene and with carbon tetrachloride at 308 K by 2 MHz frequency over the entire mole fraction range were reported by G.S.Gayathri *et al.* [65]. Rahul Singh [66] studied volumetric and acoustic properties of binary liquid mixtures of acetonitrile with 2-ethoxyethanol and 2-butoxyethanol at different temperatures. R.D. Singh [67] reported optical, volumetric and viscometric studies of binary liquid mixtures of dimethyl sulfoxide (DMSO) with formamide and N,N-dimethylformamide over the entire composition range at (293, 303 and 313) K. D. Ubagaramary *et al.* [68] evaluated molecular interactions in binary liquid mixtures of 4 -methyl-2-pentanone with butan-2-one, furfuraldehyde, cyclohexanone at 308 K.

M. Srilatha *et al.* [69, 70] studied the molecular interactions of ethyloleate with m-toluidine, and aniline using ultrasonic velocity and viscosity measurements at (303.15, 308.15, 313.15 and 318.15) K. T.Srinivasa Krishna *et.al* [71] measured the densities, ultrasonic speeds and refractive indices of binary mixtures of imidazolium with pyrrolidin-2-one at temperatures from (298.15 to 323.15) K.

Babavali, Sk. Fakruddin *et al.* [72] reported excess thermo acoustical parameters in the study of molecular interactions in binary liquid mixtures containing quinoline with arenes (benzene and toluene) at temperatures T = (303.15, 308.15, 313.15 and 318.15) K. T. Krishna *et al.* [73] studied volumetric, ultrasonic and spectral parameters of molecular interactions in binary mixtures of 1-butyl-3-

methylimidazolium hexafluorophosphate with 2-propoxyethanol at temperatures from (298.15 to 323.15) K.

G.R. Satyanarayana *et al.* [74] studied the intermolecular interactions in the binary liquid mixtures of o-chlorophenol with alkoxyethanols through ultrasonic, transport and FT-IR spectral studies at different temperatures. Volumetric and viscometric properties of binary and ternary liquid mixtures of 1-butyl-3-methylimidazolium tetrafluoroborate, monoethanolamine and water are reported by Yin Yaranet [75]. Sharma *et al.* [76] reported the volumetric and viscometric properties of binary mixtures of 1, 8-cineole with o-, m- and p-cresol at (303.15, 308.15 and 313.15) K. Gyan Prakash Dubey *et al.* [77] investigated the molecular interactions in binary liquid mixtures containing tri-n-butylamine with 2-pentanone, 3-pentanone and 4-methyl-2-pentanone.

D. Shukla *et al.* [78] measured density and acoustic velocity for binary liquid mixtures of formamide, N-methylacetamide, dimethyl foramide and dimethylacetamide with acetonitrile at atmospheric pressure and (293.15 K, 298.15 K, 303.15 K, 308.15 K, or 313.15 K). M. Menachi [79] reported molecular interaction of methyl acetate with o/p/m-xylene by the ultrasonic measurements. Sk. Nayeem *et al.* [80] investigated the molecular interactions in binary mixture of benzyl benzoate + ethyl acetate at T = (308.15, 313.15, and 318.15) K.

H. Iloukhani *et al.* [81] made theoretical and experimental study on volumetric properties of binary systems consisting of 1,2-dichloroethane with benzene and its derivatives at T = (293.15 to 333.15) K. G. Arivazhagan *et al.* [82] reported the molecular interactions in the mixtures of benzene + methylacrylate / butyl acrylate through dielectric and spectroscopic studies. K. Saravanakumar *et al.* [83] measured density, viscosity and speed of sound of benzaldehyde with benzene at (303.15, 308.15, and 313.15) K.

Dalai Biswajit *et al.* [84] investigated ultrasonic and ^{31}P NMR spectra of an acidic nuclear extractant with mono substituted benzenes. V.D. Mane *et al.* [85] reported ultrasonic properties of some synthesized pyrazolines at different concentration in 70% of 1, 4-dioxane-water mixture**.** Kaur *et al.* [86] studied molecular interactions in the binary mixture of chloroform and methanol by using ultrasonic technique. Zahid *et al.* [87] made volumetric and viscometric study of binary systems of alcohols with alkane at T = (298.15, 303.15, 308.15, 313.15 and

318.15) K and at atmospheric pressure. L. Venkatramana *et al.* [88] worked on the thermophysical and spectroscopic properties of pure N-methylcyclohexyl ammonium based ionic liquids.

Vickramjeet Singh *et al.* [89] studied the volumetric properties of 1-butyl-3-methylimidazolium bromide in aqueous solutions of d (−)-ribose and d (−)-arabinose at different temperatures. L. Venkatramana *et al.* [90] studied the volumetric, acoustic and FT-IR spectral properties of ternary and constituent binary mixtures containing N-methylcyclo hexylamine, nitro -benzene and 1-alkanols at 303.15 K. V. Syamala [91] studied the effect of various substituents on benzene ring and their impact on volumetric, acoustic and transport properties of binary liquid mixtures with dimethylacetamide. L .Venkatramana *et al.* [92] examined the FT-IR spectra and excess thermodynamic properties of binary liquid mixtures of p-xylene with 1-alkanols at 303.15 K.

Gyan Prakash Dubey *et al.* [93] made thermodynamic and spectral studies of molecular interactions in binary liquid mixtures of 1-butoxy-2-propanol with 1-alcohols. V.P. Pawar *et al.* [94] studied the dielectric and thermodynamic properties in a binary liquid mixture of dimethylene chloride with formamide. Rafael Alcalde *et al.* [95] studied intermolecular interactions in formamide + 1, 2-alkanediol binary liquid system.

Vallabh Vasudevan *et al.* [96] studied force field parameters for N,N-dimethyl formamide (DMF) revisited the improved prediction of bulk properties and complete miscibility in water. Gyan Prakash Dubey *et al.* [97] made acoustic, volumetric, transport and spectral studies of binary mixtures of 1-tert-butoxy-2-propanol with alcohols at different temperatures.

P. Maragathavel *et al.* [98] measured ultrasonic velocity of binary liquid mixtures of n alkanols with carbon tetra chloride at 303.15 K. Ultrasonic, spectrophotometric and theoretical studies of sigma and pi-interactions of iodine with substituted benzene were done by Adaikala Baskar *et al.* [99]. Indra Neela *et al.* [100] investigated anion (F-, Cl- and cation (Na+) interactions with substituted benzene [C_6H_6-nYn (Y = -F, -CN, -NO_2; n = 1-6)].

P. Vasundhara *et al.* [101] measured thermodynamic properties of binary mixtures of aniline with halogenated aromatic hydrocarbons. Vinay Sanguri *et al.*

[102] studied applications of statistical mechanical theories for estimating thermodynamic properties of ternary and binary solutions using ultrasonic velocity and density data. M.R. Rao *et al.* [103] reported ultrasonic velocity in aqueous solutions of acetone.

Density and speed of sound were reported by Mishra Sujata *et al.* [104] in ethyl-hexyl phosphoric acid, n-butyl bromide and their mixtures. M. Gowri Shankar *et al.* [105] studied the ultrasonic velocity, densities and viscosities of methyl isobutyl ketone, diethyl ketone, cyclopentanone, cyclohexanone and 2-methyl cyclo hexonone with N-methy aniline.

C.M. Saxena *et al.* [106] studied the ultrasonic velocity, density and viscosity of ethyl amine and benzyl alcohol at 30, 35, 40^{o} C. D. Bala Karuna kumar *et al.* [107] reported densities, velocities and viscosities of binary liquid mixtures of NMP (N-methyl pyrrolidine).

Sangeetha Sharma *et al.* [108] reported molecular interactions in the binary liquid mixtures of esters and oleic acid. Excess molar volume, density and ultrasonic velocity have been reported by Harish Kumar [109] in water and DMSO system. M.P.Ramya Rajan [110] studied solute-solvent interactions in the binary mixtures of 1,4 diazo bicycle 2,2,2- octane at different temperatures. D. Ubangaramary *et al.* [111] measured the density, viscosity and velocities of isobutyl methyl ketone with cyclohexanone, methyl salicylate, acetophenone and chloro benzene at 308 K. The velocity, viscosity and density of zinc stearate and acetone, calcium stearate and acetone at 303K were measured by R. Kavita *et al.*[112]. Kathrina *et al.* [113] computed the excess molar volumes of binary liquid mixtures of N, N- diethylethanol ammonium chloride- glycerol. Excess isentropic compressions were estimated for binary liquid mixtures of ethoxy propane 1 – amine by Lidia M.V. Pinheiro [114]. Lenka Moravkova *et al.* [115] studied the excess isentropic compressbilities of ternary liquid mixtures of 2,2,4 – trimethyl penane + methyl benzene + butanol.

Rayapa Reddy *et al.* [116] reported ultrasonic studies of binary liquid mixtures of N-methyl pyrrolidine with cyclic compounds. D. Wankhede *et al.* [117] studied the binary liquid mixtures of propylene carbonate at different temperatures. S.K. Pradhan *et al.* [118] reported the thermo dynamic properties of diethyl ether with CCl_4. S.K. Fakrudhin *et al.* [119] studied various thermodynamic parameters of binary liquid mixtures of quinoline and o-xylene at (303.15 to 318.15) K. F.M

Sannanigannavar [120] reported the properities of binary liquid mixtures of ethylene glycol at 298.15 K. Harish Kumar and Savitha Chahal [121] studied the thermo dynamic properties of the binary liquid mixtures of acylonitrile with aromatic ketones. Y Sreedevi *et al.* [122] studied the thermodynamic behavior of the binary liquid mixtures of anisole with aniline for the entire composition range. Xin Xue *et al.* [123] studied the thermo acoustic properties of binary liquid mixtures of water and ethers.

Sravan Kumar *et al.* [124] studied intermolecular interactions in the binary liquid mixtures of acetone by measuring density, velocity and viscosity at different temperatures. R. Ahluvalia *et al.* [125] reported the properties of binary liquid systems of ethanoic acid, propanaic acid, butanonic acid with o-cresol, m-cresol and p-cresol at (298.15, 308.15, 318.15) K respectively.

M. Kondaiah *et al.* [126] measured the excess thermodynamic properties of the binary liquid mixtures of propanoic acid. Dimple *et al.* [127] studied the thermodynamic and acoustic properties of the binary liquid mixtures of 1-methyl -2 pyrrolidine-2-ones. Ezekiel Dixon Dikio *et al.* [128] studied thermodynamic properties of the binary liquid mixtures of pyridine and (m-xylene/ p-xylene/ o-xylene). From the measured values of density, velocity and viscosity Sahu *et al.* [129] studied the molecular interactions in diacetone alcohol and chlorobenzen. N. Jayamadhuri *et al.* [130] reported the ultrasonic and thermodynamic properties of binary liquid mixtures of benzoate with aceto nitrile and benzo nitrile. Abel Zuniga – Morena *et al.* [131] studied the binary liquid mixtures of nonane with decane.

K. Rajagopal *et al.* [132] studied the binary liquid mixtures of toulene with some aliphatic nitriles at different temperatures. Ivan Alonso *et al.* [133] reported the molecular interactions in the binary liquid mixtures of 2-haptanone with aniline, N-methyl aniline and pyridine. Leonardo Hadlich *et al.* [134] studied the thermo acoustic, volumetric properties of binary liquid mixtures of dibenzo thiophene, 4-methyl – dibenzo thiophene and 4, 6-dimethyl dibenzothiophene with alkanes. Ling Ling Zhang *et al.* [135] studied the thermodynamic and acoustic properties of binary mixtures of exo-tetrahydro dicyclo pentadiene with nonane, n-dodecane n-tridecane at temperatures (293.15 to 313.1)5 K. S.L. Oswal *et al.* [136] evaluated the excess thermodynamic and acoustic properties of tetrahydrofuran with aniline viz. N-methyl aniline and N-ethyl aniline at temperatures 303.15 to 323.15 K.

Chunping Li *et al.* [137] measured the ultrasonic velocity, density and viscosity of binary liquid mixtures of 1, 2 – diamino ethane + 1, 2 – ethanediol at temperatures (298.15 to 308.15) K. Jalal B. Parsaa and Mahboobeh Faraji [138] studied the properties of ternary liquid mixtures of 2-pyrrolidine 1, 2 – propanediol + water over the entire range of composition. Malyaba A. Abu-Daabes and Akl M. Awwad [139] worked on liquid mixture of N – (2 hydroxy ethyl) morphiline from the measured values of velocity and viscosity. Sundaramma *et al.* [140] measured the ultrasonic and thermodynamic prosperities of binary liquid mixtures of lithium formate and lead acetate. Gupta *et al.* [141] studied thermodynamic and thermo acoustic properties of liquid mixtures of o-cresol with ethyl methyl ketone acetone, acetophonone and ethyl acetate at 303.15 K. Roghayeh Majdan *et al.* [142] studied solute–solvent interactions in 1-butyl-1-methylpyrrolidinium trifluoro methanesulfonate + acetonitrile from solvent activity, density, speed of sound, viscosity, and electrical conductivity and refractive index measurements. Jagadish G. Baragi *et al.* [143] measured density, viscosity, refractive index, and speed of sound for binary mixtures of 1,4-dioxane with different organic liquids at T=(298.15,303.15, and 308.15) K.

J.M. Monzon *et al.* [144] measured thermodynamic properties of organic oxygen compounds excess enthalpies for (n-hexane + a diester). S.L. Oswal, D.B. Gheewala *et al.* [145] determined speeds of sound, isentropic compressibilities, viscosities and excess molar volumes of binary mixtures of alkanoates with tetra- and trichloromethanes at 303.15 K. S.K. Dash and B. Swain [146] investigated the molecular interaction in binary liquid mixtures of tri-n-butyl phosphate with both polar and non polar liquids using viscosity and ultrasonic parameters. C.M. Saxena *et al.* [147] measured the velocities and densities of binary liquid mixture of toluene with 1,2-dichloroethane at 303.15 K. Jose Tojo *et al.* [148] determined the densities and excess molar properties of dimethyl carbonate with alkanes (C_6 to C_{10}) at 101.3 k Pa. A. K. Dash *et al.* [149] reported ultrasonic properties and molecular interaction in ternary liquid mixture of dimethyl acetamide at different frequencies. F. Nabi *et al.* [150] studied excess molar volumes by using density data for the binary liquid mixtures of styrene with dimethylsulphoxide, acetone chlorobenzene and ethanol.

A.J. Kemeakegha *et al.* [151] assessed the interactions of ethyl acetoacetate with some (C_4–C_9) aliphatic ketones at 298.15 K. K. Saravanakumar *et al.* [152]

measured the thermophysical properties of acetophenone with N,N-dimethylethanolamine or with N,N-diethylethanolamine at temperatures of (303.15, 313.15 and 323.15) K and pressure of 0.1 M Pa. Jagadish G. Baragi *et al.* [153] reported the excess molar volumes and refractive indices of binary liquid mixtures of acetyl acetone with n-nonane, n-decane and n-dodecane at (298.15, 303.15, and 308.15) K.

S. Padma *et al.* [154] studied molecular interactions of 3, 4-dihydroxy-5-nitrobenzaldehyde in ethanol using ultrasonic interferometer. Spectroscopic and ultrasonic studies on the hydrogen bonded complexes of aromatic aldehydes with phenol in hexane medium are made by R. Kumar *et al.* [155]. Ultrasonic and spectroscopic investigations of hydrogen bonded complexes of aromatic aldehydes with aniline in n-hexane at 303.15 K was carried out by B.S. Santhi *et al.* [156]. Zhenyu Bao *et al.* [157] studied the densities, viscosities and volumetric properties of BF_3 anisole and BF_3 phenetole complexes at T = (283.15 to 303.15) K. T. Srinivasa Krishna *et al.* [158] measured the acoustic, volumetric, and optical properties of binary mixture of 1-butyl-3-methylimidazolium tetrafluoroborate with propylene glycols at T = (298.15 to 323.15) K. J. Abolghasem *et al.* [159] evaluated the viscosity of binary liquids at various temperatures using Jouyban–Acree model.

Shipra Baluja *et al.* [160] measured the thermodynamic and acoustical studies of binary mixtures of diethyl malonate with 1,4 dioxane, dimethyl formamide, tetra hydro furan and hexane at 308.15K. Yihe Wang *et al.* [161] worked on excess molar enthalpies of diethyl malonate+ (1-butanol, 2-methyl-1-propanol,1-pentanol, n-heptane, and ethyl acetate) at T= (288.2, 298.2, 313.2, 328.2, 338.2,and 348.2 K) and p = 101.3 kPa. Manapragada V. Rathnam *et al.* [162] studied the volumetric, viscometric and optical properties of molecular interactions in binary mixtures of diethyl malonate with ketones at (303.15, 308.15 and 313.15) K. K. Gruner *et al.* [163] studied on shear viscosity and mass density near the liquid-liquid critical point of polystyrene in diethylmalonate. M. Sethu Raman *et al.* [164] worked on ultrasonic, DFT and FTIR studies on hydrogen bonding interactions in aqueous solutions of diethylene glycol. H. Iloukhani *et al.* [165] studied volumetric properties of dimethyl carbonate with chloroethanes or chloroethenes at 298.15 K.

Balasaheb R. Arbad *et al.* [166] studied the viscosities, ultrasonic velocities at (288.15 and 298.15) K, and refractive indices at (298.15) K. of binary mixtures of 2,

4, 6-trimethyl-1, 3, 5-trioxane with dimethyl carbonate and propylene carbonate. Ren *et al.* [167] determined the density, excess molar volume and conductivity of binary mixtures of the ionic liquid 1, 2-dimethyl-3-hexylimidazolium bis (tri fluoro methyl sulfonyl) imide and dimethyl carbonate.

Amalendu Pal *et al.* [168] worked on excess molar volumes and viscosities of diethylene glycol diethyl ether with dimethyl carbonate, diethyl carbonate, and propylene carbonate at (298.15, 308.15, and 318.15) K. Romolo Francesconi *et al.* [169, 170] evaluated the excess molar enthalpies and excess molar volumes of binary mixtures containing dialkyl carbonates + anisole or phenetole 2,2,4_trimethylpentane at (288.15 and 313.15) K.

Shipra Baluja and Nikunj Kachhadia [171] measured density, viscosity and speed of sound in solutions of some imidazolinone derivatives in DMSO at 308.15 K. S. Ramanaiah *et al.* [172] studied on excess volumes and excess isentropic compressibilities of binary liquid mixtures of trichloroethylene with esters at 303.15 K. Manapragada V. Rathnam *et al.* [173] studied the molecular interactions in binary mixtures of ethyl benzoate + ethers at (303.15, 308.15 and 313.15) K. Yogesh S. Joshi *et al.* [174] reported molecular interactions in some ethylene glycol ethers with 1,4 dioxane through dielectric and volumetric properties. P. Paul Divakar *et al.* [175] predicted the ultrasonic velocities in liquid mixtures of cyclohexanone with di-hydroxy glycols at various temperatures.

P. Subrahmanyam Naidu *et al.* [176] studied the molecular interactions in binary liquid mixtures of dimethyl sulphoxide + 1 piperidine ethanol, 4 hydroxy 1-methoxy piperidine in the temperature range of (30 – 50^0) C. Fabio Comelli *et al.* [177] measured the isothermal vapor-liquid equilibria, densities, refractive indices, excess molar volumes, and excess molar enthalpies of dimethyl carbonate + 1,2-dichloroethane and + 1,1,1-trichloroethane. Romolo Francesconi *et al.* [178] studied the vapor-liquid equilibria, excess molar enthalpies, and excess molar volumes of dialkyl carbonates + methyl *tert*-butyl ether at 298.15 K. Pei-Jung Lien *et al.* [179] worked on excess molar enthalpies for dimethyl carbonate with *o*-xylene, *m*-xylene, *p*-xylene, ethylbenzene, or ethyl benzoate at 298.15 k and 10.2 M Pa. Stefano Ottani *et al.* [180] determined the densities, viscosities, and refractive indices of mixtures of poly (ethylene glycols) + dialkyl carbonates at 313.15 K.

Fabio Comelli *et al.* [181] studied the excess properties of binary mixtures of esters of carbonic acid + three aryl alcohols at 308.15 K. Hiroyuki Matsuda [182] evaluated excess molar enthalpies of the binary carbon dioxide + dimethyl carbonate system at temperatures of (298.15 to 308.15) K and pressures of (5.0 to 7.5) M Pa. Fabio Comelli *et al.* [183] worked on excess molar enthalpies, molar heat capacities, densities, viscosities, and refractive indices of dimethyl sulfoxide + esters of carbonic acid at 308.15 K. Meijun Huang *et al.* [184] measured liquid-liquid coexistence curves of dimethyl carbonate + *n*-decane and dimethyl carbonate + *n*-tetradecane in the critical region. Fang Han *et al.* [185] measured density, viscosity, and excess properties for aqueous poly (ethylene glycol) solutions from 298.15 to 323.15 K. Senthamil Selvi *et al.* [186] reported acoustical properties of ternary mixtures of crotonaldehyde with iodine in hexane at different temperatures. Nandhibatla V. Sastry *et al.* [187] determined the densities, excess molar and partial molar volumes for water +1-butyl- or, 1-hexyl- or, 1-octyl-3-methylimidazolium halide at room temperature.

V.K. Sharma *et al.* [188] investigated the thermodynamic properties of 1-ethyl-3-methyl imidazolium tetraflorate, aniline and methyl aniline mixtures by computing excess molar volume andexcess isentropic compressibility. Vasim R. Shaikh *et al.* [189] worked on thermodynamic studies of ionic interactions in aqueous solutions of N-butyl-pyridinium bromide at 298.15 K. Guanglai Zhu *et al.* [190] worked on thermodynamic properties and microstructures of the mixture of N-butylpyridinium tetrafluoroborate with acetonitrile. Dielectric studies of molecular interactions of 1-octanol and *p*-chlorophenol with amyl acetate and isoamyl acetate in carbon tetrachloride system were carried out by P. Krishnamurthi *et al.* [191]. S. Srinivas *et al.* [192] measured densities, viscosities and ultrasonic speeds of binary mixtures containing isopropyl alcohol and ketones at different temperatures. Rupali Talegaonkar *et al.* [193] evaluated acoustic properties of substituted thiazolyl schiff's bases in binary solvent mixtures at 30±0.10c. Apurba M. Ghosh *et al.* [194] studied the effect of temperature on the molecular dynamics of binary liquid mixtures of tetrahydrofuran in n-hexane by ultrasonic method.

Excess properties and spectroscopic studies for the binary system 1, 2-ethanediamine + polyethylene glycol at T = (293.15, 298.15, 303.15, 308.15, 313.15, and 318.15) K were reported by Tianxiang Zhao *et al.* [195]. A.Gayathri *et al.* [196]

made comparative analysis of ultrasound velocity in binary liquid systems of poly propelene glycol by mathematical and experimental methods. S.B. Mishra *et al.* [197] worked on ultrasonic studies of Cd $(NO_3)_2.2H_2O$ in dioxane + water solvent at 303.15 K. R.R.Zoting *et al.* [198] made thermodynamical investigation in binary liquid mixtures of electrolytes and non electrolytes namely carbon disulphides KBr and NaBr at different temperatures. A.S.Burghate *et al.* [199] reported the acoustic properties of substituted chalcogens in binary solvent mixtures of diaxane with water, acetone-water. Manisha Gupta *et al.* [200] worked on thermoacoustical response of 1, 4-dioxane with carboxylic acids. Debarun Dhar Purkayastha *et al.* [201] predicted the interactions in water–THF binary mixture by contact angle, FTIR and dielectric studies. Oleg Prezhdo *et al.* [202] studied on viscosity of selected normal and associated liquids.

Singh Ranvir *et al.* [203] assessed molecular interactions in binary mixtures of methyl amine with xylene. Topological studies of molecular interactions in binary and ternary liquid mixtures containing lactams and isomeric chlorotoluenes were done by V.K. Sharma [204]. Shekaari *et al.* [205] studied the thermodynamic behavior of thiophene with octane, 1-hexyl-3-methylimidazolium bromide, or 1-octyl-3-methylimidazolium bromide in dilute region at T = (288.15 to 303.15) K. Sudha *et al.* [206] investigated molecular interaction of 2-chloro-4-methoxy-3-nitro benzilic acid with various solvents at different concentrations using ultrasonic technique. Fattepur *et al.* **[**207**]** studied the molecular interactions in binary liquid mixtures of acetylacetone with n-hexane and n-heptane using excess acoustic parameters calculated from measured values of velosity and density. Fakruddin *et al.* [208] evaluated the excess Gibb's free energy function values at different temperatures in binary liquid mixture of quinolone and m-xylene for the study of molecular interactions. Acoustic and volumetric investigations were done by Sk. Nayeem *et al.* [209] in aromatic, cyclic and aliphatic ketones with dimethyl sulphoxide at 308.15 K.

Chadha *et al.* [210] measured the interionic interactions of glycine, L-alanine, glycylglycine and phenylalanine in aqueous 1-hexyl-3-methylimidazolium chloride ionic liquid solutions at T = (288.15 to 308.15) K. S.K. Sharma *et al.* [211] studied the effect of temperatureon viscometric properties of aliphatic amino acids (glycine/L-alanine/ L-valine) in aqueous solutions of tetraethylammonium iodide. Fan *et al.* [212] determined the densities and viscosities of binary liquid mixtures of 1-ethyl-3-

methylimidazolium tetrafluoroborate with acetone, methyl ethyl ketone, and N-methyl-2-pyrrolidone.

Rayapa Reddy *et al.* [213] studied the thermo physical properties and IR spectra of 2-chloroethanol and 2-phenylethanol in N-methyl-2-pyrrolidone at different temperatures and 101.3 kPa**.** Pal A Gaba [214] measured the speeds of sound and isentropic functions of binary liquid mixtures of n-alkoxypropanol with 1-butanol and 2-butanol at 298.15 K. Volumetric, ultrasonic and spectroscopic FT-IR studies for the binary mixtures of imidazolium based ILs with 1, 2-propanediol were studied by Amalendu Pal [215]. M. Chandra Sekhar *et al.* [216] reported the excess thermodynamic and FT-IR spectroscopic studies in binary liquid mixtures of 2-chloro aniline with isomeric butanols at T = (303.15 to 318.15) K. C. Rambabu and his co-workers investigated the thermo physical and spectroscopic properties of various binary mixtures [217-242].

1.3 Nature and Scope of the present work

Thermodynamic and physicochemical properties of binary mixtures are studied to provide information about intermolecular interactions and internal structure present in the liquid phase. Above review of literature is a condensation of research activities developed in recent years as a result of a better understanding of interactions prevailing in various organic liquid mixtures.

A meticulous literature survey on the ultrasonic studies indicates that a lot of work has been done in binary and ternary liquid mixtures of weak and strong interacting systems and very few studies are reported with diethyl malonate as main component. Diethyl malonate (DEM) is a diethyl ester of malonic acid. The methylene group (CH_{2}-) in the middle of the malonic part of the diethyl malonate is neiboured by two carbonyl groups (C=O) the hydrogen atom on the carbon adjacent to the carbonyl group is more acidic. The acidic hydrogen groups actively participate in the reactions and they are used as reaction intermediates in many chemical processes. Moreover, thermodynamic properties of these liquid mixtures are of interest for different branches of science and engineering and also play important role in technological processes, biological process of living organisms and in nature. This fact has encouraged the author to carry out a series of systematic investigations on the solvent properties of this solvent in binary liquid mixtures.

The systematic study of thermodynamic properties has immense significance to acquire the information about the molecular interactions among the constituents of mixtures. For the appropriate design of industrial processes, the knowledge of thermodynamic properties is crucial. Keeping this objective in view, the author has chosen, diethylmalonate as common component, 1-alkanols, 2-alkoxyethanols, amides and benzene and monosubstituted benzenes as components for binary liquid mixtures in the temperature range of (303.15, 308.15, 313.15 and 318.15) K over the entire range of composition. The author has attempted to measure the variations of the ultrasonic velocity (U), density (ρ) and viscosity (η) of all the binary liquid mixtures at different temperatures in the entire composition range. The following binary systems have been selected for studying interactions.

I. Diethyl malonate (DEM) + 1-Butanol (BUT)
Diethyl malonate (DEM) +1-Pentanol (PENT)
Diethyl malonate (DEM) + 1-Hexanol (HEX)

II. Diethyl malonate (DEM) + 2-Methoxyethanol (MOE)
Diethyl malonate (DEM) + 2-Ethoxyethanol (EOE)
Diethyl malonate (DEM) + 2-Butoxyethanol (BOE)

III. Diethyl malonate (DEM) + Formamide(FA)
Diethyl malonate (DEM) + N,N-Dimethylforamide (DMF)
Diethyl malonate (DEM) + N,N-Dimethylacetamide (DMA)

IV. Diethyl malonate (DEM) + Nitrobenzene (NB)
Diethyl malonate (DEM) + Toulene (TOL)
Diethyl malonate (DEM) + Benzene (BEN)

The present work is focused on the study of volumetric viscometric and acoustic properties. The following thermodynamic and transport parameters, which are useful for understanding the nature of interaction in the binary mixtures are evaluated.

Molar volume (V), Excess molar volume (V^E),

Adiabatic compressibility (β_{ad}), Deviation in adiabatic compressibility ($\Delta\beta_{ad}$),

Inter molecular free length (L_f), Excess intermolecular free length (L_f^E),

Acoustic impedance (Z), Excess acoustic impedance (Z^E) ,

Enthalpy (H), Excess enthalpy (H^E),

Excess velocity (U^E) Deviation in viscosity ($\Delta\eta$),

Excess Gibb's free energy of activation (G^E)

It is aimed to study the molecular interactions existing between the unlike molecules of the binary mixtures, with reference to hydrogen bonding dipole-dipole interactions. Further the experimental data of binary mixtures is used to test the applicability of empirical relations of Nomoto's relation, Van Dael ideal mixing relation, impedance relation, Rao's specific velocity relation and Junjie's theory. Viscosity data is used to test the applicability of Grunberg -Nissan, Katti - Chaudhri, Heric – Brewer, Hind *et al*. and McAllister (four- body model) theories. The molar volumes of the systems and specific interactions existing are further substantiated by partial molar volumes, apparent molar volumes and reduced molar volumes. To explore the molecular structure of association effects among molecules, FT-IR spectroscopy is a successful method. The FT-IR technique offers the advantages to measure the association properties, to assess interactions by analyzing band shifts, shape and hydrogen bonding capability. FT-IR spectroscopic analysis has also been carried out to study the existence of hydrogen bonding and dipole-dipole interactions between the molecules in the chosen systems.

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