REVIEW PAPER

A review of molecular interactions in organic binary mixtures

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Abstract−The intention of this review article is to review the knowledge about interactions in organic binary liquid mixtures. Molecular interactions in organic binary liquid mixtures are interesting due to their extensive use in many fields of solution chemistry. The thermodynamics of component molecules present in various systems interacting are particularly interesting because they display fantastic results. Studies of different organic liquid mixtures represent the different modes of interactions prevailing in the component molecules. The number of parameters required describing the properties of a given class of mixtures increases sharply with the number of segment types involved. In recent years, the theoretical and experimental investigations of interactions between unlike molecules have been conveniently carried out using excess thermodynamic functions. The properties of liquid mixtures depend on the forces between molecules and on the nature and volume of these molecules, and change with the composition of the mixtures. This change, in turn, is reflected in the thermodynamic properties of the mixtures. The influence of significant contributions of a chemical, physical and geometrical nature that change excess thermodynamic properties is considered and explained in detail.

Keywords: Thermodynamic Parameters, Organic Mixtures, Intermolecular Interactions, Volumetric Properties, Viscometric Studies, Ultrasonic Properties

INTRODUCTION

The precious sources of information that may be used to observe the association between the internal structure of the system, nature of intermolecular interactions and the physical properties of the solute, solvent studied are diverse studies of thermodynamic properties of solution [1-12]. By means of experimentation, the thermodynamic properties acquired are the essential basis for the development of empirical, semi-empirical or theoretical models employed to represent and predict the behavior of fluids [13]. The volumetric properties of solutions have proven to be a very useful tool in elucidating the structural interactions (i.e., solute-solvent, solute-solute, and solvent-solvent) occurring in solution, because they provide an indirect insight into the conformational feature of the components in solution. Precise calculations of densities and refractive indices of solutions have immense significance in designing engineering processes in chemical and biological industries [14-16]. The density, refractive index and thermodynamic parameters have been widely utilized to examine different kinds of association, extent of molecular interactions and molecular packing in solutions. In addition, these properties are also used to check the applicability of differential data and give the information regarding the nature and extent of molecular interactions in solution. Apparent molar volumes and limiting apparent molar volumes of dilute solutions are useful parameters for the development of molecular models to describe the thermodynamic behavior of solutions.

The appropriate design and development of separation processes in the chemical industry are accustomed by a sufficient knowledge of physico-chemical properties of liquid mixtures. An examination of the literature [17] demonstrates few available measurements of physical properties for binary mixtures of DMF with 1-alkanols at single temperature (lower 1-alkanols [18-22] and higher 1-alkanols [23-27]) and even less at numerous temperatures [28-31]. The excess properties of binary mixtures of N,N-dimethylformamide with 1-octanol, 1-nonanol and 1-decanol using experimental data have been reported [32]. Ali and co-workers are actively involved in the studies of thermodynamic, physicochemical and acoustical properties of molecular interactions in binary, ternary, industrially important monomers and solute-solvent interactions in solutions containing aromatic hydrocarbons/monomers/esters/amines/cycloalkanes [33-42]. The excess properties of binary liquid solutions are essential for designing the industrial equipment and the interpretation of the liquid state, mainly when polar-polar or polar-non polar liquids are the components of the mixtures. The thermodynamic and physicochemical properties of binary mixtures are studied for many reasons, the most important of which is to provide information about intermolecular interactions and internal structure present in the liquid phase. This review 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. The use of binary solvent mixtures is a highly versatile and very powerful means of altering (increasing or decreasing) the solubility of a solute. Binary solvent mixtures can alter the solubility of an extremely wide variety of solutes. In some cases, solubility can be improved by several orders of magnitude in solvent mixtures [43-46]. Considerable efforts have been made to advance the scientific knowledge through observations made to classify the behaviour based on the physiochemical properties of binary liquid mixtures over the last two decades. The physiochemical properties of pure components and of binary or even high-order constituent interactions are needed in a wide variety of

multicomponent condensed phase scientific and engineering models [47-50]. It was van't Hoff [51], after 1887, who first applied the powerful methods of thermodynamics to solutions in a systematic manner.

One of the classical challenges of the physical chemistry of liquids is to understand the relation between microscopic molecular interactions and macroscopic thermodynamic properties. In particular, the problem of the microstructure and molecular dynamics in aqueous solutions has attracted much experimental and theoretical attention [52-55]. Emphasis is placed on the advantages for interpretation inherent in the contemporaneous observation of all these properties. Hydrogen bonding plays an important role in the intermolecular interaction, recognition, and conformations of both small and large molecules. Thus, scientific interest is because many questions regarding the properties of hydrogen bonded systems have not yet been completely answered. Binary liquid mixtures rather than pure liquids, find practical applications in most chemical processes as their properties are less known.

THERMODYNAMIC STUDY OF ORGANIC BINARY MIXTURES

The methodical 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 vital. The nature and relative strength of molecular interactions operating among the components of liquid mixtures have been successfully predicted by measuring their thermodynamic properties like excess molar volumes, excess molar enthalpies, excess Gibbs free energies of mixing and isentropic compressibilities changes of mixing and analyzing them in terms of topology of constituents of mixtures. Industry requires consistent and accessible reference data on the thermodynamic properties like excess molar volumes, excess molar enthalpies, and excess isentropic compressibilities of a large variety of liquid mixtures. These properties not only provide consistent data and empirical rules for science and technology, but also enhance the understanding of the behavior of liquid mixtures. In recent studies the topology of the constituents of mixtures has been utilized to (i) extract information about the state of components in pure and mixed state along with nature and extent of interactions operating among them, and (ii) predict thermodynamic properties of binary and ternary mixtures [56-59].

Thermodynamic properties of liquid mixtures can be utilized to extract information about the existence of intermolecular interactions among the constituents of mixtures. The knowledge of thermodynamic properties is essential for the proper design of industrial processes, chemical engineering etc. and for optimizing thermodynamic model/theories development. The thermodynamic and acoustic properties are very essential for understanding the physicochemical behavior of the binary and multi-component liquid mixtures. Interest in the thermodynamic properties of binary liquid mixtures extended over most of the 19th century. Excess properties of liquid systems, such as molar volumes, are required for testing the theories of solutions, development of separation techniques and equipment, and for other industrial applications. Therefore, industrial development demands reliable and accessible reference data on the physi-

cal and chemical properties of pure components and mixtures. The data are required in the development of models for process design, energy efficiency and evaluation of environmental impacts. For these reasons the collection of experimental data not only increases the empirical knowledge by creating a database, but it is also useful for better generalization of models of the solvent, and binary (or more complex) systems. These are interesting challenges for chemists, physicists, engineers, biotechnologists and many other researchers working in different fields. The composition and temperature dependences of the volumetric properties of multi-component liquid mixtures have proved to be a useful indicator of the existence of significant effects resulting from intermolecular interactions [60-64].

Liquid-liquid mixtures due to their unusual behavior have attracted considerable attention [65]. The physico-chemical properties (density, viscosity, refractive index or speed of sound and the thermodynamic behavior of binary mixtures) have been studied for various reasons. One of the most important of which is that these properties provide information about molecular interactions. Many engineering problems require quantitative data of the viscosity and density of liquid mixtures. Such data find extensive application in solution theory and molecular dynamics [66]. Furthermore, these properties are used for the interpretation of data obtained from biochemical and kinetic studies [67]. As the excess thermodynamic functions are sensitively dependent on different in intermolecular forces and size of the molecules, so these properties have been widely used to study the intermolecular interactions between the various species present in the liquid mixtures.

Also, excess and deviation functions are conducive for understanding the types of interactions between components of mixtures. Binary liquid mixtures are of considerable interest [68-78] of their own right. The entire studies require the complete knowledge of a set of properties of the studied mixtures involving bulk properties. Nevertheless, chemists use binary liquid mixtures as solvents for chemical equilibria and for media in which to carry out chemical reactions between solutes. This is because the experimental information available in the literature concerns the properties of solutes in binary aqueous mixtures. Indeed, the properties of such mixed solvent systems are of great importance in helping to understand the nature of molecular aggregation that exists in binary mixtures. It is thus not surprising that various theoretical studies have been carried out to understand the nature of the component molecules in liquid mixtures and they depend on two aspects. One of these is the randomness associated with dispersion of the two species, or of their constituent elements in the case of complex molecules. The other aspect relates to the interactions between neighbouring molecules and, in particular, to the difference in the interactions between unlike and like neighbour pairs [79-81]. However, intermolecular interactions are the result of a combination of several energetic contributions [82].

Thermodynamic properties are essential since the change in properties caused by distinction of temperature, pressure and composition will determined without any reference to assumption, models or hypothesis [83]. The data produced from these properties is necessary for an understanding of the relations between the structure and reactivity of molecules; this justifies the importance of the study of key compounds from several classes of compounds because such data permit classification of the chemical behavior of the correspond-

ing species. The number of parameters required describing the properties of a given class of mixtures increases sharply with the number of segment types involved. Moreover, steric, inductive and intermolecular effects are known [84] to change considerably the values of these parameters for individual components of the mixtures. There are two major objectives of thermodynamic properties of binary liquid mixtures. One of these is to describe the properties of component molecules when it exists in what is called an equilibrium state, a condition in which its properties show no tendency to change. The other objective is to describe processes in which the properties of a system undergo changes, and to relate these changes to the energy transfers in the form of interactions, which accompany them [85]. Recent developments in the investigation of weak association complexes in solution [86,87] have shown a need for improved approximations for the thermochemical properties of a solute or solutes in binary systems, to allow compensation for the effects of solution non-ideality. To provide a firm thermodynamic basis for these approximations, much similar systems must be studied, establishing the qualitative and, if possible, the quantitative trends of behaviour of solutes in binary solvent systems of nonspecific (or physical) interactions [88].

Intermolecular interactions are universal in nature and are present in gas, solid and in liquid phase. It is an interesting and challenging problem to theoreticians and experimentalists when investigating the liquid state. In liquid state the molecules cannot be studied either as free molecules in gases or as the structurally fixed crystalline arrangement of solids. Understanding of liquids is largely empirical, since liquid has solid-like behavior in many aspects. Liquids are said to have a short-range order and long-range disorder. A certain range of disorder gives liquid the characteristic property of fluidity. The concept of cell theory of the liquid state [89,90] implies some amount of organization of structure in it. However, many experimental techniques like differential scanning calorimetry, nuclear magnetic resonance, Raman spectroscopy, infrared spectroscopy, ultraviolet spectroscopy are used to get detailed information at molecular level in the liquids.

Liquid mixtures frequently appear in chemical processes. For proper design of the chemical process and to test theories of solutions, there is a constant need for the thermodynamic excess property data. In addition, excess properties provide information about the intermolecular interactions and macroscopic behavior of liquid mixtures and can be used to test the predictive capability of thermodynamic models and methods. In recent years, there has been a considerable upsurge in the theoretical and experimental investigations of the excess thermodynamic properties of binary and ternary liquid mixtures [3]. Thermodynamic properties of liquids and liquid mixtures can be interpreted successfully by treating the molecules as consisting of various groups or segments and to determine the single set of parameters, which characterize the molecular structure and group interactions. These properties generally show deviation from a rectilinear dependence on composition [91,92] and there is evidence [93,94] that the sign and magnitude of such deviations depend on the strength of interactions between the component molecules in the mixtures. Such deviations from the ideal behaviour are expressed by the excess thermodynamic properties. There are two possible contributions towards the total excess properties of mixing: the positive effect caused by breaking up of the associated structures into fragments or molecules (such as breaking up of self-association), and the negative one, due to the physical interactions (for example hetero-association) or geometrical fitting or cooperative accommodation into each other's structure due to difference in size and shape of the component molecules [95,96]. The interactions leading to positive and negative excess functions may compete; the sign and magnitude of such functions depend on the relative strength of these interactions. The deviation of the physicochemical properties of mixtures from their additive values is maximum near the equimolar composition. This makes reasonable analysis of the predictive power of the calculation methods either for the composition corresponding to the maximum deviation from additivity, or for all the systems at the equimolar composition. From the above description, it is understood that the acoustic, thermodynamic and physicochemical properties provide useful and interesting information about the intermolecular interactions in the binary liquid mixtures.

The study of excess thermodynamic properties of mixtures offers a convenient means to understand the inter-relationship between the observed macroscopic properties of the mixtures and the microscopic interactions among like and unlike molecules. The advantage of in-depth and wide study of this inter-relationship is twofold: first, it provides experimental background to develop, test and improve thermodynamical models for calculating and predicting fluid phase equilibria; second, it offers a wide range of possibilities for continuous adjustment of physical properties of a given solvent [97]. The equilibrium properties of liquids are strongly dependent on structure, often expressed in terms such as packing density, free volume, or more exactly in terms of the radial distribution function. The properties of liquid mixtures depend on the forces between molecules and on the nature and volume of these molecules, and change with the composition of the mixtures. This change, in turn, is reflected in the thermodynamic properties of the mixtures. Either contributions of this nature have been ignored all together, or corrections to a state of null volume change on mixing have been adopted as a means of compensating for the effects referred to [98]. This device alters the various properties of the mixtures caused by the changes they would sustain if the volume were adjusted to the value linearly interpolated between the volumes of the pure components. It will be apparent, however, that adjustment of one thermodynamic quantity (e.g., volume) in this manner will not, in general, affect a simultaneous correction of others to their linearly interpolated values. The choice of volume as the property to be "conserved" is arbitrary, and there is no assurance that nullity of volume change obviates consideration of other characteristic properties of the liquid.

Measurements of physicochemical properties including density, viscosity, ultrasonic speed, and refractive index of some organic binary liquid mixtures are important and useful tools for the elucidation of intermolecular interactions. Volumetric, viscometric, refractive index and ultrasonic speed and their derived and excess functions provide a convenient tool, not easily obtained by other means, for the study of intercomponent interaction in mixed solvents [99]. The precise data of pure components as a function of temperature and composition are very important for the successful interpretation of binary systems and application of the modern solution theories. Considerable interest has been given on theoretical and experimental investigations to explain the molecular interactions. In principle, the interactions between the molecules can be established from the

study of the deviations from the ideal behavior of physical properties such as molar volume, compressibility, viscosity and molar refraction etc. The negative or positive deviations of a physical property from ideal value depend on the type and extent of the interactions between the unlike molecules, as well as on the temperature. For example, in the case of excess molar volume, when the interactions between the molecules of the mixed components are weaker than in the pure component, the excess molar volume will be positive, whereas when the association between the mixed components predominates, the excess molar volume will be negative. It has been pointed out [100,101] that the physical interactions, which involve mainly dispersion forces, lead to positive deviations of the excess molar volumes from the ideal value, whereas the chemical or specific interactions, which involve hydrogen bond formation or charge - transfer type interactions, lead to negative deviations. The variation of the compressibility is analogous of that of the excess molar volume, whereas the change in molar refraction and refractive index tends to show an inverse trend with composition of the mixtures.

From these thermodynamic studies, an assessment of the molecular interactions between the components has been obtained [102, 103]. The problem becomes more interesting when the interactions between like and unlike molecules in liquid mixtures are probed by using these properties. Apart from the experimental studies, many of the thermodynamic and acoustic properties of mixtures have been theoretically derived [104,105] from those of the pure components by assuming that the dissimilar molecules are non-interacting. This case exists in mixtures where both the components are non-polar. In the binary mixtures where one component is polar and the other is non-polar, appreciable interaction has been observed. Even if the interaction between polar and non-polar molecules is weak, there will be considerable change in the molecular environment. In such a case the physicochemical and thermodynamic properties are likely to be affected by the intermolecular hydrogen bonding, dipole-dipole, dipole-induced-dipole and charge transfer interaction between the unlike molecules [106,107].

The study of intermolecular interactions in organic binary liquid in the light of physicochemical properties have been a matter of great interest during the last few decades [108-111]. Several workers [112-113] have made a correlation of the strength of interaction with thermodynamic parameters.

Here, we have studied the physicochemical and thermodynamic properties of suitably chosen binary mixtures with a view to understand the strength and nature of intermolecular interactions. The interactions sometimes lead to formation of new species depending upon the nature and magnitude of their specific behavior. In the past few years, substantial attempts have been made on the measurement, analysis and interpretation of basic thermodynamic properties such as excess volume and density, viscosity, excess volume and enthalpy, and excess enthalpy of binary mixtures containing alkoxyethanols [114-123].

ELUCIDATION OF INTERACTIONS BY VOLUMETRIC STUDIES

Volumetric properties provide significant information for the characterization of molecular interactions, which result from an inter-

play of solute-solute, solvent-solvent, solute-solvent and structural effects [124,125]. Volumetric properties and other thermodynamic studies of liquid mixtures are very important for the design of separation processes of liquids. The knowledge of these properties is a potential source of information about the molecular structure of mixtures. In binary liquid mixtures excess molar volume of mixing V^E, has been used as a qualitative guide to study the intermolecular interaction and complex formation between the component molecules. It has been considered as a tool for the characterization of intermolecular forces that are present in solutions and liquid mixtures [126, 127]. The molecular interactions in these systems are complex, and mixing volume data certainly provide valuable information for understanding their molecular packing. The volume data are also needed for testing of theories of solutions. The mixing of different compounds give rise to solutions that generally do not behave ideally. The deviation from ideality is expressed by many thermodynamic variables particularly by excess properties. Excess thermodynamic properties of mixtures correspond to the difference between the actual property and the property if the system behaves, ideally, and thus are useful in the study of molecular interactions and arrangements [128]. Among the excess thermodynamic properties, excess molar volume is considered as a first-order thermodynamic quantity and it is very sensitive to change the structure (i.e., order) or randomness during the mixing processes. Thus, order creation and order destruction processes in solution can be determined through experimental data of excess molar volumes. In fact, volume changes for a mixed system result from changes in free volume of liquids, since the bond lengths and bond distances in the molecules themselves do not change. The optimum packing condition in a system is directly related to differences in molecular size and intermolecular interactions, in particular when hydrogen bonding occurs between unlike molecules creating association complexes, as well as being affected by the breaking of interactions between the molecules.

Excess molar volumes represent the first derivative of the excess Gibbs function with respect to the pressure, $V^E = (\partial G^E/\partial P)_{T,n}$, the pertinent partial volume corresponds to the variation of the chemical potential with pressure $V_i^E = (\partial \mu_i/\partial P)_{T,nj}$. Excess molar volumes may also be used to test different mixing rules for equations of state of gases as well as the liquid state. In general, large and positive values of excess molar volume V^E, represent an intercalation packing effect, that is, an expansion of the mixture, while negative values of excess molar volume V^E indicate strong interaction among the components of the system or when unlike interactions prevail over self-association. V^E values are positive and increase with increase in chain length of acrylic esters for binary mixtures of benzene with acrylic esters and for binary mixtures of acrylic esters with alkanols [129,130]. These workers explained the increase in V^E values based on non-specific interactions between the component molecules. The excess molar volumes of vinyl acetate with cyclohexane, n-alkanes were obtained, and V^E values were found to be positive, which increase slightly with the chain length of n-alkanes [131]. Similar study for the binary mixtures of acetyl acetone with aliphatic alcohols and for binary mixtures of 2, 2, 2-trifluoroethanol with 1-alcohols was carried out by other research groups [132,133]. This is because as the chain length increases the strength of hydrogen bonding of alcohol molecules decreases. It is reported that interaction between N, N-dimethylacetamide (DMA) and alkanols becomes increasingly weak as

we move from methanol to 1-propanol [134] and more negative for 1-octanol+DMA [135]. The V^E values were well explained for the binary mixtures of cyclohexanone+aromatic hydrocarbons and for the acrylic esters+aliphatic hydrocarbon mixtures [136-138]. For the former systems, the results were explained based on the bulkiness of aromatic hydrocarbons, which increases the values of V^E, and becomes more and more positive. Bulky groups are not able to fit themselves properly in voids created by other components in liquid mixtures, resulting in increase in V^E values while in case of later systems the results were controlled by dispersion interactions. The compared V^E values of tetrachloroethylene+branched alcohols with tetrachloroethylene+normal alcohols showed that the V^E values are more positive for former systems than the later [139,140]. This indicates that normal alcohol interact more strongly with tetrachloroethylene than branched alcohols. This may be attributed to the steric hindrance of the methyl groups in branched alcohols. It is observed that V^E values are positive for the binary mixtures of methylmetacrylate+cyclohexane [141]. Similar behavior of V^E was also reported for the binary mixtures of acrylic esters+alkyl aromatic hydrocarbons [142].

The observed V^E values can be discussed in terms of several effects which may be divided into physical, chemical and structural contributions [143]. Physical contribution comprises dispersion forces and non-specific physical interactions that add positive terms to V^E, chemical contributions consider the breaking up of the H-bonding structure, which tends to increase V^E and specific interactions such as the formation of hydrogen bonds and charge transfer complex between unlike molecules, resulting in a decrease in volume. Structural contributions arising from geometrical fitting of one component into the voids created by the other are due to differences in molar volume and free volume between components and lead to negative contribution to V^E.

The study on mixtures of amides with water analyzed the volume changes based on molecular size differences [144]. According to them, the ideal binary mixing without volume change is most frequently found in mixtures of molecules of similar size. Molecules of very different sizes exhibit a variety of volume changes. The free volume of liquids is a substantial fraction of the molar volume and does not vary greatly for many liquids with temperature. When solute of small molecules is dissolved in a fluid of large molecules, the molecules of former liquid are fitted into voids of the later. For instance, the molar volumes of dimethysulphoxide, acetone and styrene at 298.15 K are 7.1319×10^{-5}, 7.3857×10^{-5} and 11.5338×10^{-5} m^3 mol^{-1}, respectively, which might allow the components to fit into each other's structure [145], thereby reducing the volume of the mixture. The free volume, pertaining to a large molecule in its own environment, is being reduced, since the centers of the small molecules can approach large molecules more closely than other large molecules would. If the large molecules were embedded in a continuum, their corresponding void volumes would disappear completely and a large volume decrease would be observed in dilute solutions even in the absence of specific interactions in the solvent. The negative values of V^E for the system DMF+benzene, toluene and ethylbenzene [146], DMSO+tetrahydrofuran and methyltetrahydrofuran [147], methylcyclopentane+ketones (acetone, butanone and 2-pentanone) [148], 1-alkanol+alkylamines [149], 1-butanol+1, 3-butanediol [150], 1-butanol+glycerol [151] not only arise due to

specific interactions between the unlike molecules, but also due to sizes and shapes of the component molecules in the mixtures. Similar explanation is given for DMF+toluene, bromobenzene [152], DMF+water [153], binary mixtures.

It has been observed that excess molar volumes V^E are negative for the mixtures of 3-pentanone+ethylbenzene and o-xylene [154]. Negative V^E values for both systems indicate the predominance of dipolar interactions and the absence of specific interactions between the component molecules. It has also been shown that the experimental values of V^E for the mixtures of diisopropylether with several alkanols are negative [155]. Excess molar volumes are also negative due to specific interactions. The probability of complexation between diisopropyl ether and the alkanols must be lower. This is due to the influence of the hydrocarbon chain on the charge distribution, which produces these negative trends in V^E. However, when the complex DIPE-alkanol is formed, the ether can polarize the alkanol increasing the energy of the interaction and diminishing the volume of the complex.

The observed experimental values of V^E for equimolar mixtures of benzene+toluene are slightly positive, that for benzene+chlorobenzene is almost zero, and for benzene+bromobenzene is negative [156]. Since this change in the magnitude of V^E is brought by the introduction of -CH$_3$, -Cl, or -Br groups into the aromatic ring, this behavior can be due to the size effect [157] of the molecules. Thus, the excess molar volume may be given as the sum of the two terms as $V^E = V^E_{size} + V^E_{interaction}$. Since the sizes of aromatic hydrocarbons like toluene, chlorobenzene, bromobenzene and that of cyclohexane are all nearly equal, the V^E size effect for benzene+toluene, benzene+chlorobenzne, benzene+bromobenzene is almost same. On the other hand, for toluene+bromobenzene and toluene+chlorobenzene, V^E_{size} has been put equal to V^E_{expt}. $V^E_{interaction}$ so calculated for different mixtures at equimolar composition shows a systematic trend with the increasing π-electron density of the aromatic hydrocarbon in first three mixtures. Thus, the excess volumes are influenced by specific group interactions. The bulky groups also play an important role in contributing towards the positive value of excess volume, as the compounds with bulky groups are not able to orient themselves properly in the other component liquid mixtures. For instance, greater V^E values for DMF+2-methyl-1-propanol binary mixtures were observed [158] and similar behavior was observed for benzene+methyl acrylate/ethyl acrylate/butyl acrylate and styrene binary mixtures [159]. V^E values for N, N-dimethylmethanesulphonamide+aliphatic alcohols were observed to be greater [160] than those for N, N-dimethylacetamide+aliphatic alcohols [161] binary mixtures. This is because of the presence of more bulky groups in amide molecule in the former mixtures than that in the later one, which causes steric repulsion of appreciable magnitude between the component molecules. Thus, the excess molar volume can provide the evidence of complex formation between unlike molecules even in the cases where the U. V. and N. M. R spectroscopy fail completely to detect the presence of charge transfer interactions. This is because if the complex formation is due to the intermolecular bonding of the donar-acceptor type of the molecules, the strength of such type of bonding would be extremely weak [162]. However, if the complex formation is due to intermolecular hydrogen bonding between the component molecules, the strength of such bonding would be appreciably strong in magnitude. For instance, Benson et al. [163] observed the formation

of hydrogen bonds between acetone and alcohol molecules leading to negative V^E values for acetone+cyclopentanol binary mixture and Clara et al. [164] observed positive V^E values for binary mixtures of 1-pentanol+R-(+)-limonene. This would indicate that interactions between unlike molecules are weaker than interactions between like molecules in the pure liquids and that dispersion forces dominate the behaviour of the solution. An expansion in free volume is considered to occur, making the mixtures less compressible than the ideal mixture, which ultimately leads to positive values of V^E.

The positive V^E values for N, N-dimethylformamide+higher aliphatic alcohols (C$_7$-C$_{10}$) observed suggested strong dipolar interactions, which decrease with increase in the carbon chain-length of alcohols, introducing a steric effect between the molecules [165]. In binary mixtures of nitromethane with {2-methoxyethanol+2-butoxyethanol} systems, the V^E values were found to be positive [166]. The variation of positive V^E values is easily explicable, that is, an expansion in volume (which is due to the breaking of hydrogen bonds during the mixing process) makes the mixture more compressible than the ideal mixture, which ultimately culminates in positive values of V^E [166]. The large volume expansion in trifluoroethanol+1-alcohol over the whole composition range with positive excess molar volume, where the V^E increases, as the chain length of alcohol increases has been well documented [167]. This positive deviation results due to the formation of new hydrogen bonding OH-----F and breaking of hydrogen bonding of self-association in trifluoroethanol. The experimental values of V^E for the binary mixtures of chloroform+alkanols have also been observed [168]. V^E values were more positive for chloroform+propan-1-ol than for butan-1-ol. Since the change in V^E is brought by the self association of alkanols which decreases as the chain length increases so that the breaking of intermolecular interactions in butan-1-ol is much easier than in propan-1-ol. Other workers [160,169-171] observed similar behavior of interactions in binary systems.

The excess molar volumes acquired are positive for mixtures of 2-alkanols+1,2-dichloethane [172]. The positive V^E values can be ascribed to dominance of H-bonds between 2-alkanols and physical interactions over dipole-dipole interactions between 2-alkanols and 1,2-dichloroethane molecule. V^E (>0) increases from 2-propanol up to 2-octanol as the length of the 2-alkanol chain increases. The increase in V^E in this case, may be attributed to a weak interaction of type dipole-dipole between 2-alkanols and 1,2-dichloroethane, probably due to the decrease of the polarizability of 2-alkanols with the alkyl chain length, as was suggested by authors [173,174]. As the temperature of the mixtures increases, dissociation of the associated spaces takes place, and thus V^E increases.

ULTRASONIC SPEED PROPERTIES OF BINARY MIXTURES

Lagemann and Dunbar [175] were the first to point out the sound velocity approach for qualitative determination of the degree of association in liquids. Intermolecular interactions play an important role in liquid mixtures. They influence the arrangement, orientation and conformations of the molecules in solutions. Multicomponent liquid mixtures are frequently used as media for many chemical, biological and industrial processes, because they provide a wide range of properties [176-179]. During the last two decades, ultra-sonic study of liquid mixtures has gained much importance in assessing the nature of molecular interactions present in the mixtures. The deviation from linearity in the values of ultrasound velocity when studied as a function of concentration exhibits interesting variations in the case of liquid mixtures. Ultrasonic technique has become a powerful tool for studying the molecular behavior of liquid mixtures [180]. This is because of its ability of characterizing physico-chemical behavior of liquid medium. Binary liquid mixtures due to their unusual behavior have attracted considerable attention [181, 182]. The propagation of ultrasonic waves is essentially related to the physico chemical behavior of mixtures, such as molecular association or dissociation. Many investigations are concerned with theoretical aspects of the acoustic properties of solutions and mixtures. The developments and applications of ultrasonic techniques have been adequately employed in understanding the molecular interactions in pure liquids, and their binary mixtures [183-185]. This technique has been used by various workers with a great success for characterizing physicochemical behaviour of pure liquids, mixtures, gels etc. [90,186,187].

Nowadays, the interest for industrial multicomponent process is increasing. Thus, the recent developments made it possible to use ultrasonic speed in medicine, engineering, agriculture and chemical industry. It has been found useful in studying the chemical processes and play major role in the synthesis of various chemical substances. Another area where ultrasonics is nowadays used is for obtaining information about microstructure. The technique involves measurement of ultrasonic velocity and attenuation through the metal. Furthermore, the ultrasonic techniques have yielded useful results in estimating the crack density changes and identifying the crack nucleation mechanisms, besides providing supporting evidences of microstructural changes in the study of other seismic precursors such as acoustic emissions, electrical resistivity and electromagnetic emissions at laboratory scale. Use of ultrasonics in chemical synthesis is a highly active research area. The chemical effects of ultrasound energy are quite distinct from other conventional sources.

The application of ultrasonic waves in chemistry was viewed as a convenient technique [188]. Many studies have been carried out and it is well documented that the advantages of ultrasound procedure are good yield, short reaction time and mild conditions [188, 189]. It has been found that ultrasound affects both homogeneous and heterogeneous chemical systems. It can allow the use of cruder chemicals than the expensive purer ones. It can reduce the number of steps in a multistep chemical synthesis or in some cases completely switch the reaction pathway.

Ultrasonic study has been found to be a very powerful tool for investigating the departure of real liquid mixtures from the ideality in terms of certain excess thermodynamic properties. These properties provide a wealth of information about the nature and strength of intermolecular interactions in binary liquid mixtures. An ultrasonic wave is a pressure wave that can be treated as a series of compressions and rarefactions traveling along a material such that molecular planes are displaced from their mean positions. It is assumed that the compressions and rarefactions are both reversible and adiabatic [190].

A considerable advancement has been taken place in the last few years in the theoretical and experimental investigations of the excess thermodynamic properties of liquids and solutions. The most sig-

nificant reasons are, first, the study of solutions, which show different behavior from the pure liquids, and second, they provide a way for studying the forces acting between two molecules of different species. The most interesting of these are the new types of phase equilibra, which arise from the extra degrees of freedom, which are introduced by the variation in the proportion of pure components. Due to the importance of multicomponent liquid systems in product formulation in chemical industries, some predictive methods for the evaluation of properties in liquid solutions have gained enormous interest [191,192].

The mixing of liquids is accompanied by structural and thermodynamic variations, which are reflected in the acoustical properties of the mixtures. Correlation between structure of liquid, its thermodynamic properties and acoustic parameters is the main thrust area nowadays [193]. Studies on these properties play a vital role role in understanding the nature and physicochemical behavior of intermolecular interactions [191,194]. This is achieved through evaluation of derived properties, the most important of which is the isothermal compressibility, $k_T[=-(1/V)(\partial V/\partial P)_T]$. Accurate data are frequently required in physics, chemistry and chemical engineering for this thermodynamic property. Unfortunately, the conventional methods for measuring k_T either tend to be of low precisions or are excessively tedious. Fortunately, determination of the isentropic compressibility $k_s[=-(1/V)(\partial V/\partial P)_s]$ provides an indirect but for more accurate and convenient means of calculating k_T. The prime object of sound velocity measurements in liquid systems is to estimate the value of isentropic compressibility, which cannot be done by any other method [195]. Isentropic compressibility has been widely used to study the molecular interactions through its excess functions. On the other hand apart from k_T, it can also be used to deduce other useful thermodynamic properties, e.g., heat capacity ratio, internal pressure (π_i), cohesive energy density (CED), non-linearity parameter (B/A) etc. Isothermal compressibility and heat capacities ratio are two key parameters in molecular thermodynamics of fluid phase equilibria [196-201].

Investigation of interactions occuring in liquid phase is still under development. It is a generally accepted fact that ultrasonic speed is very promising in this area, as this quantity can be measured with very high accuracy. The effect of intermolecular interactions on the sound velocity was analyzed using very simple assumptions, like those of Jocobson [202], Schaaffs [203] and others [204,205], but also using advanced numerical techniques [206]. Very recently, ultrasonic speed has acquired increased importance as a useful physical property for the study of molecular interactions in liquid and solutions [167,170,207-215]. Recently the measurement of physical properties such as density, ultrasonic speed and viscosity of binary mixtures of N-methyl-2-pyrrolidone with methyl acetate, ethyl acetate and butyl acetate over the complete composition range at different temperatures has been investigated [216]. Various excess quantities such as excess molar volume, excess isobaric thermal expansion coefficient, excess isentropic compressibility and excess ultrasonic speed are calculated from the experimental data to get insight into the intermolecular interactions that are present in these mixtures [216]. Ultrasonic speeds and densities and of difurylmethane+(methanol or ethanol or propan-1-ol or butan-1-ol or pentan-1-ol or hexan-1-ol) binary mixtures have been measured over the entire composition range at T=298.15 K and atmospheric pressure [117]. By utiliz-

ing the measured ultrasonic speeds and density data, excess isentropic compressibility, excess ultrasonic speed, and excess intermolecular free length have been calculated for each of the binary systems over the entire composition range. In a recent study ultrasonic speeds in formamide+1-butanol or 2-methyl-1-propanol or 2-methyl-2-propanol mixtures have been measured at 298.15 K and 308.15 K [218]. The measured ultrasonic speed data were correlated by correlations like Nomoto's relation, Van Dael's mixing relation and impedance dependence relation. Excess isentropic compressibility was calculated from experimental ultrasonic velocity and excess volume data. The ultrasonic speed in binary and multicomponent systems was figured out theoretically by various workers [219-225] using empirical and semi-empirical relations, and therefore it is also worth to mention the efforts made by other research groups in this field [154, 224-235]. Pandey et al. [236,237] applied Flory theory [238,239] and the hard sphere model of liquid to deduce ultrasonic speed of pure liquids. Several workers [219,222,240] to compute ultrasonic speed in binary liquid mixtures have used Flory theory.

Various experimental and theoretical investigations have revealed that a representation in terms of derived thermodynamic parameters from ultrasonic speed is intermolecular free length (L_f), provides a better view in order to understand the intermolecular interactions in binary liquid mixtures. The variation of ultrasonic speed in solution depends upon the increase or decrease of intermolecular free length after mixing the components according to a model proposed by Eyring and Kincaid [230]. Hence, it is obvious that intermolecular free length is a predominating factor in determining the nature and variation of ultrasonic speed. Several attempts have been made in the past [241,242] to discuss the interactions in solutions in terms of intermolecular free length. Dependence of intermolecular free length on temperature has also been examined [241,242]. Jacobson [243] gave an empirical relation correlating intermolecular free length with isentropic compressibility for pure liquids. Kaulgud [244] has extended the concept of free length to binary solutions. The free length can be evaluated through ultrasonic speed [245] and thermodynamic [246] methods.

It was assumed during early studies of wave propagation that the amplitude of the propagating wave was infinitely small, and because of this, the characteristics of both the medium and ultrasonic wave propagating through it should not be affected by one another. However, for all practical purposes it is observed that the intensity of sound waves coming out from transducer is finite. However, measurements of ultrasonic propagation constants become unreliable [247,248] when sound waves of finite amplitude are made to propagate in liquids, because nonlinear effects like acoustics streaming and distortion of wave front occur due to greater attenuation of high frequency constant compared to low frequency ones. For most liquids, the velocity of sound is 5 to 10 times as great as the average kinetic velocity of gas molecule.

TRANSPORT PROPERTIES VARIATION AND DEGREE OF INTERACTION IN BINARY MIXTURES

Viscosity is a transport property, and depends on the transport of momentum that moving molecules offer to the neighboring molecules in the flow process of a fluid. It is called the resistance produced by moving molecules for the flow of the fluid. Fluidity is defined

as the reciprocal value of viscosity. 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 [249]. Viscosity is a characteristic property of fluids. The viscosity of binary liquid mixtures has been studied by many investigators to predict the effect of its variation on the type and degree of interaction between the component molecules. Viscosity deviation is related to the resistance of a mixture to flow; therefore, negative values indicate that the resistance to flow of the mixture is less than the resistance to flow of the pure compounds. In contrast, positive deviations of viscosity values for the mixtures are a consequence of a higher resistance to flow than those of pure compounds. If the viscosity data for the mixtures are obtained as a function of composition, the existence or non-existence of specific interaction between component molecules may be studied by estimating the strength of these interactions. Rowlinson et al. [70] reported the positive deviations from the linear dependence of viscosities on mole fraction or volume fraction and the occurrence of maxima where the component molecules interact more strongly. Negative deviations occur where London forces or dispersion forces are responsible for the interactions [250]. The occurrence of viscosity maxima or minima in viscosity versus composition curves have been attributed to the complex formation between unlike molecules. For instance, the well-defined maximum in viscosity against composition curves for amide+water binary mixtures has been observed [251]. The maxima are large for N, N-disubstituted amides whereas the extent of maxima was found to decrease for N-monosubstituted amides. For such systems, on dilution with water, there occured mearly an interchange of hydrogen bonding between donor - acceptor molecules. For N, N-disubstituted amides the maxima reflect an interaction of strong association with water through multiple hydrogen bonding greater in number than pure disubstituted amides.

The measurement of viscosity provides information about liquid systems, which are of considerable interest because of their extensive applications in many industrial processes. The interest shown by many workers in this field led to the formulation of a large number of methods for correlating or predicting the viscosity-composition data of liquid mixtures [252-257]. Among all the physical properties of fluids needed to optimize the industrial process design, viscosity is one of the most important property. In the chemical industry, knowledge of viscosity is necessary for hydraulic calculations, fluid transport through pipes and pores surfaces, and in many mass and energy transfers. The viscosity of n-decane has also been investigated at high temperatures and pressures [258].

Deviation in viscosity $\Delta\eta$ is a useful parameter, which helps us in deduction of intermolecular interactions in binary mixtures. The deviation in viscosity was reported to be negative for benzene+cyclohexane and benzene+ethylene dichloride mixtures [106]. In these systems, complexes are not formed or complexes of low stability are formed. Postigo et al. [259] reported negative values for hexane+

diethylamine system, indicating small interactions between the component molecules, which are close to ideal behavior and in hexane +1-decanol and 1-decanol+diethylamine mixtures. $\Delta\eta$ values were reported to be positive because of the change in the associated structure of alkanols. Negative values were also reported for DMSO+ methanol mixtures, which may be due to the difference in molecular size between the component molecules [260]. The binary mixtures of benzene+cyclic ethers have been investigated and $\Delta\eta$ values were found to be negative, suggesting weakening of dipole-dipole interactions [261]. Positive $\Delta\eta$ values for toluene+cyclic ethers have also been reported, indicating the formation of new specific interactions in binary mixtures. Negative $\Delta\eta$ values for 3-pentanone+ ethylbenzene and o-xylene binary mixtures have also been reported [152]. Negative trends reported for the both systems indicate the predominance of dipolar forces and the absence of specific interactions. Negative values of $\Delta\eta$ were also reported for binary mixtures in which both the components were polar and one or both components were self-associated. For instance, the negative values for N, N-dimethylacetamide+aliphatic alcohols and benzyl alcohol+ isoamyl alcohol, isopropyl alcohol [262] binary mixtures have been well-documented [134]. It was explained that these trends of $\Delta\eta$ are not only due to size differences but also due to the association between component molecules through hydrogen bonding. Different research groups [39,263-272] also reported a similar type of association between unlike molecules.

The viscosity of mixtures strongly depends on the entropy of the mixtures, which is related to the liquid structure and its enthalpy (and consequently with molecular interactions between the components of the mixture) [273]. So, the viscosity deviations are considered as the function of molecular interactions between different molecules. Positive viscosity deviations indicate strong interaction, whereas negative $\Delta\eta$ values suggest the absence of strong specific interactions between the unlike molecules in the mixture. The interactions that make the flow easier (i.e., the breaking of the intermolecular associates, because of the motion of individual species is less difficult than that of big molecular complexes) are negative contributions to $\Delta\eta$, while the interstitial arrangement and interactions that produce the formation of intermolecular associates hinder the flow, so the sign of their contribution to viscosity properties is positive. There is evidence for the weak complex formation between benzene and CCl_4, a system involving weak dipole interaction [274]. The $\Delta\eta$ is positive for mixtures in which maxima in the viscosity vs. composition curve is observed, indicating the formation of stable complexes. For example, stable complexes are formed in the DMSO +water [275], DMSO+acetic acid, acetone+water [106], DMSO +tri-n-butyl phosphate [276], N-methyl-2-pyrrolidone+substituted benzenes [277], diethylene glycol monoethyl ether+water [278], glycol monomethyl ether+water [279] binary mixtures.

The zero values for $\Delta\eta$ of non-electrolytes (ideal solution) binary mixture can be theoretically predicted only in homologous non-associating binary mixtures where the component molecules have about equal molar volumes [280]. However, in some cases, $\Delta\eta$ is effectively zero; for instance, it is zero for benzene+chloroform and benzene+carbon tetrachloride except at high concentration of benzene [106]. Interaction between benzene and chloroform is said to involve a weak bond intermediate between a hydrogen bond and formation of an electron-transfer complex [281]. A negative trend

in $\Delta\eta$ values for 2-pentanol with alkanes was observed, which reveals that as the temperature increases, $\Delta\eta$ values tend towards ideality (become less negative) [282]. In the study halo hydrocarbons with 2-methyl-1-propanol, negative trends for deviation in viscosity for all systems were observed [283]. The halogenated compounds show weak dipole-dipole interactions in pure state. Mixtures of these compounds with chlorinated derivatives show negative values of $\Delta\eta$, lower than those with brominated compounds. This is due to the larger size of brominated derivatives, which allow a better interstitial accommodation than chlorinated derivatives do, so the flow would become more difficult for the systems containing bromobenzene and bromocyclohexane. Similar behavior has also been observed in other systems involving haloalkanes and butanols [284,285].

In the viscosity results of hydrogen bonded liquids mixtures (diol+ water or alcohol), negative value of $\Delta\eta$ for 1, 2- and 1, 4-butanediol in pentanol have been found [286-290]. A negative deviation in viscosity corresponds to the destruction of the intermolecular structures existing in pure components. Less structured entities created in the solution make the flowing of 1, 2- and 1, 4-butanediol+alcohol mixtures easier than that of the pure diols. The investigated viscosity for 1-alkanol+cyclohexane, 2-alkanol+cyclohexane and 1-alkanol +methyl cyclohexane revealed $\Delta\eta$ to be negative for all binary mixtures [291]. The magnitude of negative deviations increases with the increase in the chain length of 1-alkanol molecules. Similar negative deviations have also been reported for ethanol+cyclohexane and 1-hexanol+cyclohexane [292,293]. In recent studies, negative deviations of $\Delta\eta$ in various binary mixtures have been observed [164,166,294-297]. A positive deviation in viscosity of dimethylsulphoxide and acetic acid system has been observed, which indicates that the solutions are highly non-ideal and the interaction between the components of the mixture is quite strong [298].

CONCLUSIONS

The composition and temperature dependence of excess properties of binary liquid mixtures provides extensive information regarding the intermolecular interactions between the component molecules. These thermodynamic properties at different temperatures and concentrations of pure chemicals and their binary liquid mixtures over the whole composition range are useful for practical chemical engineering purposes and many biological systems. Clearly, over the years, there has been considerable advancement in the experimental investigation of the excess thermodynamic properties of liquid mixtures in the chemical industry. Studies on different physicochemical and thermodynamic properties of liquid binary mixtures within wide ranges of compositions and temperatures are important sources of information that may be used to examine the relationship between the internal structure of the system, nature of intermolecular interactions, and the physical properties of the mixed solvent studied.

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