

# THERMOPHYSICAL AND THERMODYNAMIC BEHAVIOR OF BINARY LIQUID MIXTURES: A CRITICAL REVIEW OF MOLECULAR INTERACTIONS, EXCESS PROPERTIES, AND CORRELATION MODELS

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## ABSTRACT

The thermodynamic characteristics of two-liquid mixtures are strongly influenced by factors including molecular dispersion forces, polarity, hydrogen bonding, and differences in molecular size and shape between the mixture components. Ultrasonic velocity, refractive index, and calculated thermodynamic properties such as excess Gibbs free energy, viscosity, Molar volume calculations have also been carried out for these systems. This article reviews information about the thermodynamic properties of liquid mixtures and their relationship to molecular interactions in the mixtures. The types of binary liquid mixtures discussed include ester-alcohol, amine-alcohol, and hydrocarbon-alcohol mixtures and others. Thermodynamic data on these and other binary liquid mixtures obtained at various compositions and temperatures are reviewed. Other topics include the effects of chain length and temperature and other structural features on the physical properties of liquid mixtures and the effects of structural and polar differences on excess molar volume and viscosity of liquid mixtures. Mathematical methods for modeling thermodynamic nonequilibrium behavior of liquid mixtures, including the Redlich-Kister equation and various activity coefficient correlations, are discussed. No one method or data set can fully describe the molecular interactions that occur in liquid mixtures. Combining thermodynamic data with other types of data can provide more information about thermodynamic nonequilibrium behavior of liquid mixtures. Recommendations for future research include efforts to develop improved predictive methods and gather additional thermodynamic information on liquid mixtures to help predict the properties of other complex liquid mixtures.

**KEYWORDS:** Binary liquid mixtures; Thermophysical properties; Molecular interactions; Excess molar volume; Viscosity; Ultrasonic velocity; Hydrogen bonding; Non-ideal behaviour; Thermodynamic models; Property prediction.

## 1. INTRODUCTION

Binary liquid mixtures have many important applications in industry and research, such as in the production and delivery of pharmaceuticals, the chemical industry, solvent preparation, and the purification of other chemicals and materials. The behavior of a binary liquid mixture is influenced by interactions between different molecules, including excluded volume and van der Waals interactions and dipole-dipole and hydrogen-bonding interactions. Other interactions also affect the behavior of binary liquid mixtures and make them nonideal. Techniques for studying nonideal binary mixtures include the measurement of physical properties such as sound velocity, refractive index, density, and viscosity and thermodynamic properties such as excess Gibbs free energy and activity coefficients. The magnitude and type of properties observed for a mixture both vary with temperature and mole fraction. Small changes in the molecular structure of the pure components, including changes in functional groups and side chains or degree of branching, can lead to significant changes in the interactions between similar and dissimilar molecules in a mixture. Positive or negative values of excess dynamic viscosity and excess molar volume suggest a nonideal mixture. Other mixture properties need to be studied to determine what structural differences between pure components cause a mixture to be nonideal. Multiple experimental methods and data sources may be useful for correlating molecular and bulk properties.

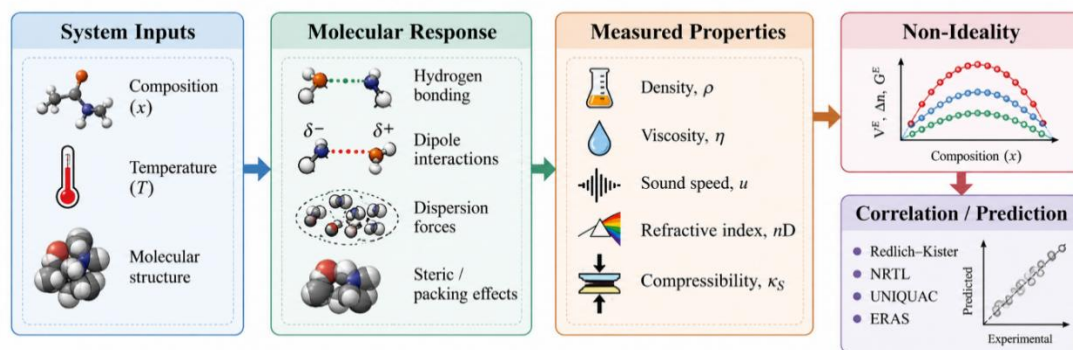


**Figure 1:** Integrated interpretation of binary liquid mixture behaviour

To model and predict key thermodynamic properties of two-component fluid mixtures, scientists have used several theoretical and local composition models, including the Non-Random Two-Liquid (NRTL) model for estimating activity coefficients. Other approaches include empirical models that use polynomial equations, such as the commonly used Redlich-Kister equation. Modern advances include other data- and machine learning-based models for predicting two-component fluid mixture properties, but barriers to wider use of modern methods remain. Barriers include the need to distinguish empirical models that fit the data well but lack physical basis from physically based models and to develop new models that can incorporate other factors that affect the thermodynamic properties of two-component fluid mixtures, including differences in molecular structure and local concentrations and changes in temperature.

In response to these and related difficulties in the study of binary liquid mixtures, this review offers a critical assessment of the published work on binary systems. It examines in depth the mathematical models and thermodynamic excess properties used for such mixtures, along with the wider range of intermolecular interactions involved. Reported information on the physical and chemical characteristics of both ideal and nonideal binary systems is also surveyed. The review aims to clarify how the physical behavior of liquids connects to their thermodynamic properties, and to point out where knowledge is still lacking. Particular emphasis is placed on the gaps concerning molecular simulation and other modern computational approaches, with the goal of strengthening our understanding and prediction of nonideal liquid behavior in both industrial and laboratory contexts.

### 1.1 Molecular Interactions in Binary Liquid Mixtures

The physical and transport properties of mixtures of two fluids are primarily determined by the interactions between unlike molecules. These interactions are often qualitatively and structurally very different from the interactions between molecules of the pure components. Important intermolecular interactions include dipole-dipole forces, electrostatic forces, hydrogen bonds, and London dispersion forces. Hydrogen bonds are particularly important in mixtures of highly polar substances, such as amines and alcohols, and aqueous mixtures. Volume changes in mixtures are related to Raoult's law deviations and the relative strength of unlike-molecule interactions. Negative Raoult's law deviations, in which the total volume of a mixture is less than the sum of the volumes of the unmixed components, occur due to strong attractive interactions, such as hydrogen bonding, between unlike molecules. Positive Raoult's law deviations, in which the total mixture volume is greater than the combined pure-component volume, occur due to other factors, such as steric effects and London dispersion forces. Table 1 provides more information about factors affecting the properties of mixtures.

**Table 1:** Major Molecular Interactions in Binary Liquid Mixtures

| S. No. | Type of Interaction        | Nature of Interaction                   | Commonly Involved Components      | Influence on Thermophysical Behaviour                                     |
|--------|----------------------------|-----------------------------------------|-----------------------------------|---------------------------------------------------------------------------|
| 1      | Hydrogen bonding           | Strong directional attraction           | Alcohols, amines, water, acids    | Can increase association and produce significant deviations from ideality |
| 2      | Dipole-dipole interaction  | Attraction between polar molecules      | Polar solvents, amines, esters    | Influences density, viscosity and excess properties                       |
| 3      | van der Waals interaction  | Weak intermolecular attraction          | Non-polar and polar molecules     | Affects molecular packing and cohesive behaviour                          |
| 4      | Dispersion forces          | Instantaneous dipole-induced attraction | Hydrocarbons and organic liquids  | Influences mixing and structural arrangement                              |
| 5      | Donor-acceptor interaction | Specific molecular association          | Amines, alcohols, esters, ketones | Can strongly modify excess thermodynamic properties                       |
| 6      | Solute-solvent             | Interaction between                     | Components of binary              | Determines non-ideal mixing behaviour                                     |

|   | interaction                 | unlike molecules                        | mixtures                                     |                                                                  |
|---|-----------------------------|-----------------------------------------|----------------------------------------------|------------------------------------------------------------------|
| 7 | Steric interaction          | Spatial/size-related effect             | Branched alcohols, amines, organic molecules | Affects molecular packing and free volume                        |
| 8 | Like-molecule interaction   | Interaction between identical molecules | Pure components within mixture               | Determines the reference behaviour for assessing deviations      |
| 9 | Unlike-molecule interaction | Interaction between different molecules | Binary liquid components                     | Governs association, expansion/contraction and excess properties |

The extent and complex physical structure of combined molecular and physical interactions between two chemical species depend significantly on a variety of factors, including charge and volume differences between the two interacting species and the location and type of reactive chemical groups on the interacting species molecules, such as hydroxyl, amino, and carbonyl groups. Differences in chemical structure can affect the extent to which the two species can pack together, the relative strengths of dipoles formed by their interaction, and whether interactions between the two species are primarily cohesive or hindered by steric effects. A range of experimental measurements can shed light on molecular-level interactions that cannot be observed directly. Density, together with the excess molar volume derived from it, shows how efficiently the two components pack together. Kinematic viscosity, and how far it departs from ideal-mixing values, reveals how the combined fluid flows and can point to a viscosity increase caused by hydrogen bonding between the two species. Ultrasonic velocity and isentropic compressibility give further insight into how compressible the mixture is and how strong the cohesive forces are between unlike molecules. Understanding molecular interactions is important both for understanding lab observations of combined fluid behaviors and for developing and testing predictive models of these behaviors. Incorporating physical data about combined fluids into thermodynamic models and other predictive tools can increase their accuracy and usefulness in modeling thermodynamic behaviors of complex combined fluid and multicomponent liquid systems for industrial and other applications.

### 1.2 Theoretical Models and Correlation Approaches

Applying advanced correlation and other theoretical methods is key to understanding the properties of binary fluid mixtures based on experimental data. These and other theoretical approaches offer mathematical tools to develop a thorough understanding of nonideal thermodynamic behavior in mixtures. Widely used empirical methods, including the Redlich-Kister polynomial expansion, are powerful tools for correlating data on excess thermodynamic properties throughout a composition range to form continuous functions of composition. Well-established theoretical approaches, including local composition models like the UNIQUAC, NRTL, Wilson, and Margules models, are widely used by scientists to predict activity coefficients and other thermodynamic properties of nonideal systems based on specific intermolecular interactions. Theoretical and other methods are also used to study dynamic properties such as viscosity. Theoretical free-volume and friction models describe the relationship between molecular-scale properties and fluid viscosity. Scientists use a variety of approaches to determine parameters for empirical and theoretical models based on experimental data to better understand molecular interactions in fluid mixtures. Model predictions are validated using experimental data. Advanced correlation and theoretical methods have practical applications in basic and applied settings, including industrial applications.

### 1.3 Fundamentals of Binary Liquid Mixtures

At its core, a binary fluid mixture consists of two different molecular species, referred to here as the solvent and solute or co-solvent. Binary fluid mixtures are prepared by combining the two species in varying amounts to produce a range of mixtures with different concentrations and, therefore, different physical properties. Properties of binary mixtures are often very different from those of the unmixed components and depend strongly on the molecular interactions that occur when the components are combined. If the interactions between molecules of different types are the same as the interactions between molecules of the same type in the pure components, the behavior of a binary mixture is described by Raoult's law, and its properties are easy to calculate because they are additive. Real mixtures rarely behave ideally, and their properties and structures must be determined experimentally by measuring a variety of physical properties, including interfacial surface tension, speed of sound, viscosity, heat capacity, and density. Measuring how these and other properties of binary mixtures vary with temperature and composition can help scientists understand and predict the behavior and structures of different binary mixtures.

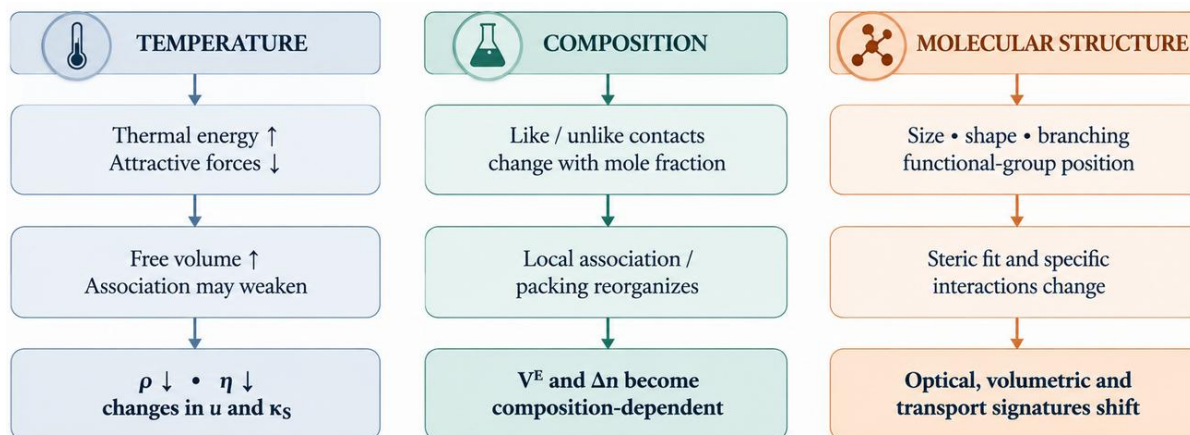


**Figure 2:** Factors Influencing Binary Liquid Mixture Behavior

Systematic study of a wider range of solvent structures can provide important data about thermodynamic deviations from ideality for solutions. Benefits of studying a wider range of structures include gaining knowledge about short-lived interactions and other small-scale structural features of solvents. Careful study of two-fluid systems at a variety of temperatures and concentrations can help scientists understand the relationships between large-scale observations and small-scale physical and chemical properties and processes. Understanding these relationships can have many practical applications in developing and improving industrial processes, especially in industries that use finely engineered solvents and other processes involving separation of multiple phases, as well as emerging energy production and storage technologies.

#### 1.4 Effect of Temperature and Composition

Ambient temperature and molar composition are key thermodynamic factors that significantly affect the physical properties of binary liquid mixtures. Higher temperatures increase the kinetic and thermal energy of a liquid's molecules. Increased molecular motion weakens the attractive forces between molecules, including hydrogen bonds and dipole interactions between unlike molecules. These changes result in changes to the local structure of the liquid. Changes to the local structure of a liquid can affect other physical properties of the binary mixture, including dynamic viscosity and ultrasonic velocity. Increasing the temperature of a binary mixture tends to decrease both the mass density and dynamic viscosity of the mixture. Decreases in these physical properties are related to weaker attractive forces and greater free volume in the liquid. Changes to the molar composition of a binary mixture affect the frequency of contacts between unlike molecules. Changes in unlike-molecule contacts can, in turn, affect the local structure of a binary mixture and free volume between dissimilar molecules. The balance between attractive and other interactions that cause a contraction or expansion of volume in a binary mixture determines the extent to which its behavior approximates ideal behavior. Thorough testing of physical properties over a range of temperatures and molar compositions is important to confirm theoretical predictions and models of binary liquid behavior. Data on the physical properties of binary mixtures can help chemical and materials engineers choose and develop binary solvent mixtures for industrial applications.

#### 1.5 Technical Framework for Thermophysical Analysis

The thermophysical properties of a two-component fluid mixture depend on several factors, including component composition and arrangement and intermolecular forces. Two chemically different fluids mixed together often show thermophysical properties that differ from what is expected for an ideal mixture. Studying these differences can help you understand the spatial relationships, size effects, van der Waals forces, hydrogen bonding, and other interactions between components in the fluid mixture.

$$x_1 + x_2 = 1$$

In a two-component system with components 1 and 2, properties of the mixture are often expressed in terms of mole fractions of components 1 and 2. The properties of the mixture of two pure components can be estimated by multiplying the properties of the pure components by their mole fractions. Departures from these estimates are quantified mathematically and are called excess molar properties.

$$Y^E = Y - (x_1 Y_1 + x_2 Y_2)$$

Here, this equation represents the observed property of the mixed liquid in terms of the properties of the unmixed, pure liquids.

In volumetric analysis, the excess molar volume can be calculated using the equation:

$$V_m^E = V_m - (x_1 V_1 + x_2 V_2)$$

In this study, total molar volume was used as a mixing parameter. Information about both the sign and value of the total molar volume can be used to assess other molecular properties of the mixture, including the interactions and

packing of molecules. Negative values for this parameter indicate stronger attractive interactions and better molecular packing in the liquid mixture, while positive values indicate weaker attractive interactions and other unfavorable interactions or effects in the liquid mixture.

Likewise, fluid-thickness differences, or viscosity variations, are defined using similar parameters:

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2)$$

This equation includes the dynamic viscosity of the mixture and the viscosities of the unmixed components. One application of this equation is measuring the effects of mixing on the motion and other physical properties of chemical components in a mixture.

Thus, a thorough technical evaluation of thermophysical properties should consider all relevant properties simultaneously rather than separately. Other properties to consider alongside molecular structure and interactions include isotopic compressibility, refractive index, speed of sound, viscosity, density, and derived excess properties. A holistic approach to studying thermophysical properties can provide greater insight into the molecular interactions that underlie observed bulk properties.

## 2. LITERATURE REVIEW

A large and growing literature exists on the chemical and physical properties of binary mixtures. Information on the optical and acoustic properties of binary mixtures, including density, heat capacity, speed of sound, viscosity, and other excess thermodynamic properties, is useful for understanding microscopic interactions and structures in these mixtures. Knowledge of macroscopic properties can help scientists understand the interactions and structures of different components in a binary mixture, including hydrogen bonding, dipole-dipole interactions, and steric effects. Chemical and structural information on binary mixtures described in the literature includes information on many commercially and scientifically useful solvents and solvent blends, such as aliphatic amines, esters, alcohols, alkanolamines, aqueous solutions, and newer green solvents known as deep eutectic solvents. This article reviews and analyzes information on the physical properties of binary mixtures in the published literature to develop a more general understanding of how the components of binary mixtures and changes in temperature affect both macroscopic and microscopic properties. The information discussed here should be helpful in developing and characterizing other binary mixtures.

### 2.1 Thermophysical and Calorimetric Properties

Chiu, Liu, and Li (1999) examined the thermal behavior of alkanolamine compounds by means of differential scanning calorimetry and heat capacity measurements. Their work showed that calorimetric techniques are a valuable way to characterize the thermal properties of liquids. Heat capacity data stand out in particular, since they reflect shifts in intermolecular interactions as well as changes in the number of degrees of freedom available to the liquid's molecules. [1]

Majstorovic et al. (2020) investigated the thermodynamic and related properties of two binary systems, in which ethyl acetate was combined with either 1-butanol or 1-propanol. The study looked at how composition and the carbon chain length of the alcohol influence the properties of the fluid. Chain length matters in practical terms because it alters van der Waals forces and can affect phase separation in the mixture. [2]

Pandey et al. (2020) used other optical techniques and refractive index measurements to study mixtures of 1-butanol and 1-propanol with 2-diethylethanolamine (DEEA). Visual observations in that study helped characterize the behavior of different concentrations and interactions of alcohols and amines. Other optical data like those collected by Pandey et al. serve as useful secondary data because changes in the refractive index of a solution are related to changes in the chemical environment and molecular polarity of substances in the solution. [3]

Dubey and Kumar (2012) performed a detailed study of alcohol and amine solutions at different temperatures to evaluate the effects of temperature and solute concentration. The study results show that it is important to conduct temperature studies of solutions to help differentiate between observed differences in bulk properties due to increased molecular motion and other differences. [4]

Li et al. (2013) published an industry-related review and overview with particular emphasis on amine-based technologies for capturing, using, and storing carbon dioxide. In addition to reviewing the thermodynamic properties of binary mixtures containing amines, the review discusses the general industrial applications and behavior of amines in carbon dioxide capture and other processes. [5]

Omrani, Rostami, and Mokhtary (2010) studied spatial and compositional properties of binary mixtures containing 1,4-dioxane and various common alcohols and amino alcohols. The authors' goal was to develop a relationship between temperatures and molecular interactions in these systems. Information on spatial properties can help determine other volumetric properties of mixtures, providing indirect information on interactions and structures among different types of molecules in the mixture. [6]

Omar and Sadeghi (2021) studied the macroscopic physical properties and liquid-liquid extraction applications of deep eutectic solvents made from nonanol. This study provides an example of the importance of studying the physical properties of fluids for other types of separation applications. The publication also describes how the concentration of components in a fluid affects its usefulness for liquid-liquid extraction. [7]

Almasi (2015) studied the fluid density and viscosity of mixtures containing 2-alkanols and ethyl formate using theories based on molecular interactions and free volume. Combining experimental transport property data with these theories provides greater insight into intermolecular interactions and transport in fluid mixtures than can be obtained from experimental data alone. [8]

Bhagat and Maken (2021) studied combinations of 2-amino-2-methylpropan-1-ol and short-chain (C1-C3) alkanols using physical and visual methods. The goal of the study was to determine the effects of changes in composition on intermolecular interactions at different temperatures and concentrations. Using alkanols of different lengths, the researchers were able to closely examine changes in intermolecular interactions related to changes in alcohol chain length and functional group type. [9]

Dubey and Rani (2021) studied alcohol-amine mixtures using spectroscopic, acoustic, and thermodynamic techniques. Using multiple techniques is important because acoustic and thermodynamic studies offer detailed information about bulk properties and behavior of the mixture, while spectroscopic techniques can provide additional information about interactions between molecules in smaller portions of the mixture. [10]

## **2.2 Temperature, Composition and Molecular-Structure Effects**

Kumar et al. (2013) studied mixtures of branched alkanols and alkoxypropanols at several temperatures. In addition to studying departures from ideal thermodynamic behavior, they examined other effects of structure and temperature. These mixtures are good examples of how branching in the structures of the components affects how well the molecules pack together and how strongly they interact with each other, changing excess properties measured in the laboratory. [11]

Kumari and Maken (2021) studied mixtures of isomeric butanols and di-n-butylamine using FTIR spectroscopy and other transport and physical measurements. Combining spectroscopic and other measurements helped researchers relate physical properties measured in the laboratory to molecular interactions and demonstrated applications of these mixtures to carbon dioxide capture. Overall, this study shows the value of combining thermodynamic and structural information. [12]

Gahlyan, Maken, and Park (2020) studied ethyl and methyl esters of fatty acids using modeling techniques and other experimental methods, including viscosity and density measurements and information on solid-liquid equilibrium. The authors extended conventional thermophysical property measurements to provide more information about the behavior of two-liquid mixtures of organic compounds, including data on mass transport kinetics. These extended characterizations can be useful in studying flow and equilibrium behavior in ester mixtures. [13]

Dragoescu, Sirbu, and Almasy (2021) investigated aqueous solutions of alkanediols, looking at properties beyond structure and temperature alone. For water-containing mixtures and other systems of hydrating compounds, temperature and mole fraction need to be examined together if the properties of the system are to be understood. This matters most where water forms strong hydrogen bonds with other components, since those bonds exert a major influence on the behavior of the mixture. It is therefore necessary to consider how the hydrogen-bonded network changes as temperature and the proportion of water vary. [14]

Estrada-Baltazar et al. (2015) examined the bulk properties, including viscosity and density, of mixtures of n-decane with 1-heptanol, 1-hexanol and 1-pentanol. By pairing n-decane with alcohols of different carbon numbers, the authors were able to establish how molecular size and shape affect the flow behavior of the mixtures. These findings can be applied to characterizing and comparing the properties of related compounds within a homologous series. [16]

Dubey, Saur, and Kaur (2017) measured mixtures of alkanols with di-n-butylamine over the temperature range 293.15 to 313.15 K. A central aim was to understand how intermolecular forces behave in these systems. Working across this range of temperatures made it possible to see how attractive intermolecular forces are balanced against the thermal motion of the molecules. The conclusions were drawn from measurements of other physical properties of the mixtures. [20]

Shakila et al. (2018) studied the thermophysical properties of ethyl acrylate mixtures with alkyl amines using theoretical and experimental techniques. The study included experimental and theoretical analyses of the thermal behavior of these mixtures. The combination of theoretical and experimental techniques can help relate experimental data to theoretical explanations of fluid behavior on a molecular scale. [23]

Dubey and Kumar (2017) studied mixtures of C6-C10 1-alkanols and diisopropylamine using molar property and viscosity data, as well as other measurements. By studying mixtures of alcohols with varying numbers of carbon atoms, the researchers were able to learn more about the effects of molecule size on the behavior of mixtures. Homologous series studies like this one can help identify specific molecular features that affect the behavior of mixtures. [25]

Wang et al. (2013) performed spatial analysis of interactions between dimethyl sulfoxide and amines over a wide temperature range of 293.15 to 363.15 K. The wide temperature range allowed detailed study of the effects of temperature and mole ratio on intermolecular interactions, including changes in the system's volume behavior. Studies of this kind can provide valuable information about molecular contraction and other structural changes caused by heating and intermolecular interactions. [27]

### 2.3 Volumetric, Acoustic and Transport Properties

Nayeem et al. (2016) analyzed mixtures of ethyl acetate and benzyl benzoate through density and ultrasonic velocity measurements. Their results indicated that the two datasets, taken together, reveal useful information about how the components interact and pack within the mixture. Ultrasonic velocity is a valuable addition to such work because it is directly governed by the properties of the liquid, density among them. [17]

Aminabhavi and Raikar (1993) studied the mixing behavior of aliphatic alcohols with bromoform to learn more about interactions and deviations from ideal mixing between these compounds. The results of their study are generally applicable to understanding the thermodynamic properties of other binary fluid mixtures. [18]

Jain et al. (2009) used the ERAS model and excess molar volume and viscosity data to study the behavior of methanol-di-n-butylamine and methanol-n-butylamine mixtures. Combining experimental data with an association-based approach provides a more detailed understanding of nonideal behavior and interactions between molecules in a mixture. This type of combined approach has several benefits. [21]

Chowdhury and Saleh (2014) studied flow resistance and other viscosity-related properties of tert-butanol solutions with tri-n-butylamine, di-n-butylamine, and n-butylamine. By varying the structure of the amines in the solutions, the researchers were able to study how molecular structure affects miscibility and other fluid properties in these solutions. Findings from such studies can help researchers better understand the relationship between structure and viscosity in mixed fluids. [22]

Kumari and Maken (2021) used a combination of acoustic and volumetric studies and topological methods to study mixtures of isomeric butanols and di-isopropylamine. Adding topological analyzes provides another way to view the structure of these mixtures and helps improve understanding of experimentally determined thermophysical properties. Results show that many departures from ideal thermodynamic behavior of these mixtures can be attributed to molecular and topological structure. [24]

### 2.4 Molecular Interactions and Hydrogen-Bonding Effects

All reviewed studies highlight the importance of intermolecular interactions in understanding nonideal thermophysical behaviors. In mixtures containing polar compounds such as amino alcohols, amines, and alcohols, dipole-dipole and hydrogen-bonding interactions significantly affect the phase behavior and structure of these mixtures.

Zaitseva et al. (2011) used calorimetry to study hydrogen-bond enthalpies of interactions between aliphatic alcohols and tertiary amines. Their calorimetric results allowed them to identify thermodynamic factors responsible for the observed interactions. Calorimetric results can therefore be used in conjunction with other measurements of physical properties, such as transport and acoustic properties, to provide additional information about molecular interactions. [26]

Studies of alcohol-amine mixtures show that molecular interactions between different components can affect many physical properties. Measurements of excess volume and density can help identify differences in the structure of mixtures. Measures of viscosity can be used to draw conclusions about differences in molecular motion in different mixtures. Ultrasonic measurements can show differences in compressibility and other properties. Spectroscopic data can provide detailed information about specific interactions of molecules in a mixture.

### 2.5 Thermodynamic Models and Correlation Approaches

Various thermophysical property data are reported and interpreted for binary systems in other literature. Polynomials of the Redlich-Kister form are useful for describing concentration-dependent excess properties and other deviations from ideal behavior for binary systems. Using these and other models to describe behavior can help you compare and contrast the behavior of different binary fluid systems based on experimental data.

Almasi [8] showed that including ideas related to free volume and fluid friction in the analysis of flow and other transport phenomena can improve understanding. Jain and coworkers [21] used the ERAS method to develop a model for interactions between and other properties of different molecules. Mathematical models, like those described by Almasi [8] and Jain and coworkers [21], can be useful tools beyond curve fitting if the assumptions and parameters used in the models are based on physical processes occurring at the molecular level.

In addition, the studies reviewed show that collecting more types of information can provide greater understanding of the theoretical properties of materials. Collecting information about a material's structure and properties, including spectral, visual, sound, and viscosity properties, can help scientists better understand the structural and chemical changes and other physical processes occurring in the material.

### 2.6 Machine Learning and Data-Driven Analysis

Kikugawa et al. (2019) used machine learning and other clustering tools to interpret large volumes of data on material properties in multiple dimensions. This approach provided insights beyond those gained from simpler analyzes of individual material properties. Machine learning and similar data analysis tools can be useful for gaining new insights into material properties and other relationships that cannot be described by low-order polynomial equations. [15]

Although data-driven approaches offer many advantages, they are used less frequently in the scientific literature than classical thermodynamic and other methods. Therefore, opportunities exist to develop data-driven structural models and machine learning methods that incorporate large amounts of thermophysical data into predictive models consistent with thermodynamic principles.

## 2.7 Application-Oriented Studies

Chung, Roh, and Lee (2018) published a review of CO<sub>2</sub> capture systems available for commercial use, with a focus on membrane and amine absorption technologies. Insights from this review help practitioners better understand the industrial applications and performance of aminated liquids and the role of liquid properties in CO<sub>2</sub> capture processes, including separation and mass transfer. [19]

Combined, the practical studies show that physical thermodynamic properties of fluids have industrial applications beyond the laboratory. Important thermodynamic properties, including macroscopic properties such as viscosity and vapor pressure and microscopic properties such as particle interactions, affect industrial processes involving these fluids, such as carbon dioxide uptake and release, product recovery, solvent recycling, and mass transfer. Understanding the molecular interactions of fluids is critical to improving industrial processes that use liquids.

## 2.8 Comparative Synthesis of the Reviewed Literature

The reviewed studies agree that the properties of two-liquid mixtures depend mainly on temperature and composition and system geometry. Many mixtures contain strongly polar components such as amino alcohols, amines, and alcohols. For these mixtures, the major factors affecting liquid mixture properties are the packing of molecules in the liquids and hydrogen bonding and other dipole interactions between molecules. For other liquid mixtures in which the intermolecular forces of attraction are weaker, important factors affecting properties include free volume, van der Waals forces, the polarity of molecules, and molecular size.

Deviation from ideal thermodynamic behavior is usually described in the literature using deviation and excess properties. Excess molar volume data provide information about changes in the structure and volume of a mixture. Deviations in viscosity indicate changes in the flow behavior of a mixture. Other acoustic property data are useful for understanding the structural effects and compressibility changes that occur during mixing.

Thermodynamic and mass transport parameters should not be ignored. The molar ratio of components in a solution affects the relative amounts of heterogeneous and homogeneous interactions among solute particles. Temperature affects the strength of solute-solute interactions and the mobility of solute particles. It is not possible to develop reliable, general relationships between physical properties and solution composition based solely on data collected at a single temperature or concentration.

Additionally, the reviewed computational studies show a close link between theory and experiment. Polynomial models can well describe experimental concentration relationships. Other theoretical approaches, including models based on fluid flow and free volume and thermodynamic properties, can help provide greater physical understanding of experimental results. High accuracy in fitting an experimental data set with a theoretical model does not necessarily indicate the presence of a corresponding physical mechanism at the molecular level.

The current body of research supports using an integrated approach to analyze data from experiments on two-liquid mixtures. Factors to consider include the basic theory of two-liquid systems, the effects of temperature and other conditions, particle interactions, system design, error calculations, and other physical properties. Benefits of the integrated approach include better understanding of two-liquid systems based on experimental findings.

**Table 2: Technical Comparative Analysis of Reviewed Studies**

| No. | Study                     | Mixture / System                               | Property / Approach                                        | Main observation                                                | Molecular interpretation                                                              |
|-----|---------------------------|------------------------------------------------|------------------------------------------------------------|-----------------------------------------------------------------|---------------------------------------------------------------------------------------|
| 1   | Chiu, Liu & Li (1999)     | Alkanolamines                                  | Heat capacity; DSC                                         | Provided heat-capacity data and characterized thermal behaviour | Molecular structure and intermolecular interactions influence heat-capacity behaviour |
| 2   | Majstorovic et al. (2020) | Ethyl acetate + 1-propanol / 1-butanol         | Thermophysical properties                                  | Composition and alcohol chain length affected mixture behaviour | Chain length modifies intermolecular interactions and deviation from ideality         |
| 3   | Pandey et al. (2020)      | 2-Diethylethanolamine + 1-propanol / 1-butanol | Refractive index and related properties                    | Refractive index varied with composition                        | Optical response reflected structural changes and amine-alcohol interactions          |
| 4   | Dubey & Kumar (2012)      | Amine + alcohol systems                        | Thermophysical properties                                  | Temperature and composition affected mixture behaviour          | Molecular association and intermolecular forces governed non-ideal behaviour          |
| 5   | Li et al. (2013)          | CO <sub>2</sub> -capture / amine-based systems | Review of CO <sub>2</sub> capture, storage and utilization | Discussed amine-based absorption and process applications       | Amine-CO <sub>2</sub> interactions are important for                                  |

|    |                                  |                                                |                                                              |                                                                                   |                                                                                                     |
|----|----------------------------------|------------------------------------------------|--------------------------------------------------------------|-----------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------|
|    |                                  |                                                |                                                              |                                                                                   | absorption and process design                                                                       |
| 6  | Omran, Rostami & Mokhtary (2010) | 1,4-Dioxane + alcohol / amino-alcohol systems  | Density and volumetric properties                            | Temperature, composition and molecular structure affected volumetric behaviour    | Contraction/expansion reflects intermolecular interactions and packing                              |
| 7  | Omar Sadeghi (2021)              | Nonanol-based deep eutectic solvents           | Thermophysical properties                                    | Density, viscosity and related properties characterized liquid systems            | Component selection and molecular interactions influence physicochemical behaviour                  |
| 8  | Almasi (2015)                    | Ethyl formate + 2-alkanols                     | Density and viscosity                                        | Density and viscosity varied with molecular environment                           | Molecular packing, free volume and intermolecular forces affected transport properties              |
| 9  | Bhagat Maken (2021)              | 2-Amino-2-methylpropan-1-ol + C1–C3 alkanols   | Volumetric and optical properties                            | Temperature and composition influenced volumetric and optical behaviour           | Molecular association and structural changes affected measured properties                           |
| 10 | Dubey & Rani (2021)              | Amine + alcohol systems                        | Thermodynamic, acoustic and spectroscopic properties         | Combined techniques provided complementary information on mixture behaviour       | Molecular association and structural changes were reflected in acoustic and spectroscopic responses |
| 11 | Kumar et al. (2013)              | Alkoxypropanols + branched alkanols            | Mixing properties                                            | Temperature and molecular structure influenced mixing behaviour                   | Unlike-molecule interactions produced deviations from ideality                                      |
| 12 | Kumari & Maken (2021)            | Di-n-butylamine + isomeric butanols            | Volumetric, acoustic, transport and FTIR properties          | Combined macroscopic and spectroscopic measurements characterized association     | FTIR, volumetric and acoustic data supported molecular-association mechanisms                       |
| 13 | Gahlyan, Maken & Park (2020)     | Fatty-acid methyl/ethyl ester systems          | Solid–liquid equilibria, density, viscosity and modelling    | Temperature and molecular composition affected phase and transport properties     | Molecular composition influences phase behaviour and transport response                             |
| 14 | Dragoescu, Sirbu & Almasy (2021) | Aqueous alkanediols                            | Thermophysical properties                                    | Temperature, composition and molecular structure affected properties              | Hydrogen bonding and molecular association influence water–alkanediol behaviour                     |
| 15 | Kikugawa et al. (2019)           | Liquid substances                              | Multidimensional thermophysical properties; machine learning | ML identified patterns in multidimensional property data                          | Property relationships can be associated with underlying molecular characteristics                  |
| 16 | Estrada-Baltazar et al. (2015)   | n-Decane + 1-pentanol / 1-hexanol / 1-heptanol | Density and viscosity                                        | Temperature and alcohol-chain length systematically affected transport properties | Chain length and molecular structure modify intermolecular interactions                             |
| 17 | Nayeem et al. (2016)             | Benzyl benzoate + ethyl acetate                | Ultrasonic velocity and density                              | Acoustic and volumetric properties revealed non-ideal behaviour                   | Molecular association and structural effects influenced acoustic response                           |
| 18 | Aminabhavi & Raikar (1993)       | Bromoform + aliphatic alcohols                 | Mixing properties                                            | Demonstrated non-ideal mixing behaviour                                           | Specific molecular interactions and molecular structure                                             |

|    |                           |                                             |                                                              |                                                                              |                                                                                                     |
|----|---------------------------|---------------------------------------------|--------------------------------------------------------------|------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------|
|    |                           |                                             |                                                              |                                                                              | influenced solution properties                                                                      |
| 19 | Chung, Roh & Lee (2018)   | Industrial CO <sub>2</sub> -capture systems | Amine scrubbing and membrane separation                      | Evaluated industrial CO <sub>2</sub> -capture technologies and applications  | Amine-based absorption involves important chemical/physical interactions relevant to process design |
| 20 | Dubey, Saur & Kaur (2017) | Di-n-butylamine + alkanols                  | Mixing properties                                            | Temperature-dependent mixing behaviour was observed                          | Molecular association changed with temperature and composition                                      |
| 21 | Jain et al. (2009)        | Methanol + n-butylamine / di-n-butylamine   | Viscosity and excess molar volume                            | Excess properties characterized non-ideal mixing                             | Molecular interactions and association contributed to excess behaviour                              |
| 22 | Chowdhury & Saleh (2014)  | tert-Butanol + n-/di-/tri-n-butylamine      | Viscosity and viscosity deviation                            | Amine structure affected transport behaviour                                 | Molecular structure and intermolecular interactions influenced viscosity                            |
| 23 | Shakila et al. (2018)     | Ethyl acrylate + alkyl amines               | Thermophysical properties; experimental/theoretical analysis | Temperature affected mixture behaviour                                       | Molecular interactions and structural characteristics influenced thermophysical response            |
| 24 | Kumari & Maken (2021)     | Di-isopropylamine + isomeric butanols       | Volumetric and acoustic properties; topological analysis     | Structural differences influenced volumetric and acoustic behaviour          | Molecular topology and structural differences affected intermolecular interactions                  |
| 25 | Dubey & Kumar (2017)      | Diisopropylamine + C6–C10 1-alkanols        | Volumetric, viscometric and molar properties                 | Alcohol chain length and temperature affected transport and molar properties | Chain length influenced molecular association and transport behaviour                               |
| 26 | Zaitseva et al. (2011)    | Tertiary amines + aliphatic alcohols        | Calorimetry; hydrogen-bond enthalpy                          | Quantified hydrogen-bonding effects energetically                            | Cooperative hydrogen-bonding contributed to molecular interaction energy                            |
| 27 | Wang et al. (2013)        | Dimethyl sulfoxide + amines                 | Volumetric properties                                        | Temperature and composition affected volumetric behaviour                    | Molecular interactions were reflected in volumetric changes                                         |

## 2.1 Research Gaps and Critical Analysis

Although many experimental studies related to heat and mass transfer in two-component liquid mixtures have been published, some gaps in knowledge still exist in the existing body of research knowledge.

First, previous work studies individual properties, including sound velocity, mass density, refractive index, and other properties of fluids. Studies of single properties provide useful information about fluids, but they often do not help researchers understand the relationships between microscopic interactions and macroscopic fluid properties.

In addition, while deviation and excess measures are common methods for evaluating thermodynamic nonideality in solvents, the structural factors contributing to these deviations differ among types of solvents. Negative and positive deviations from ideality may result from many different causes, such as different molecular structures and intermolecular forces, including van der Waals forces and steric effects. As a result, conclusions about solvents based on a single property are likely to be incomplete.

Further, conventional correlative models are usually justified by high accuracy in reproducing experimental data. High forecast accuracy does not necessarily mean that factors included in a correlative model have physical meaning at the atomic level. Future research with correlative models should focus on both physical meaning and predictive accuracy.

Finally, there is a lack of methods in the existing literature that combine traditional thermodynamic concepts with more computationally based approaches. Applying machine learning and other techniques to experimental and database information on chemical structures and thermodynamic properties could help develop more comprehensive predictive tools.

Ultimately, ongoing research in this area requires more than simply gathering additional data; it requires developing a comprehensive theory that incorporates other theoretical and empirical research on topics including fluid flow on a large scale, other calculated statistical measures, and differences in particle size and distribution and other intermolecular differences.

### 3. PROBLEM STATEMENT

The physicochemical properties of binary liquid mixtures are important in many industrial uses, including pollution control, fuel economy, separation processes, cosmetics manufacture, and drug development. Important practical properties of binary mixtures include viscosity changes, excess molar volume, isothermal compressibility, refractive index, speed of sound, friction, and density. Predicting these properties for binary mixtures is difficult because of interactions between components in the mixture. Major causes of nonideal thermodynamic behavior in binary mixtures include molecular-level differences such as size and shape differences and hydrogen bonding, as well as other interactions such as covalent bonding and dipole interactions.

A major challenge in this area is limited availability of experimental thermophysical data for specific fluid mixtures over narrow ranges of concentration and temperature, which limits opportunities for broader comparisons. There are also inconsistencies in microscale models explaining changes in fluid behavior for different mixtures. Statistical models used to describe experimental data do not always help researchers understand the molecular interactions responsible for observed fluid behaviors. Differences in temperature and structure further complicate efforts to predict fluid behavior. Areas needing further research include bringing together knowledge about structure, interactions, molecular models, fluid-property deviations, and other thermophysical information to better connect molecular and macroscopic observations of fluid behavior.

### 4. RESEARCH METHODOLOGY

The authors of this literature review used a structured approach to identify and review published research on the microscale interactions and thermophysical properties of two-phase fluid mixtures. Goals of the literature review included combining experimental findings with theoretical knowledge and other information available in academic resources and modern predictive tools. Steps in the literature review process included locating and selecting relevant resources for review and comparing and summarizing information from those resources.

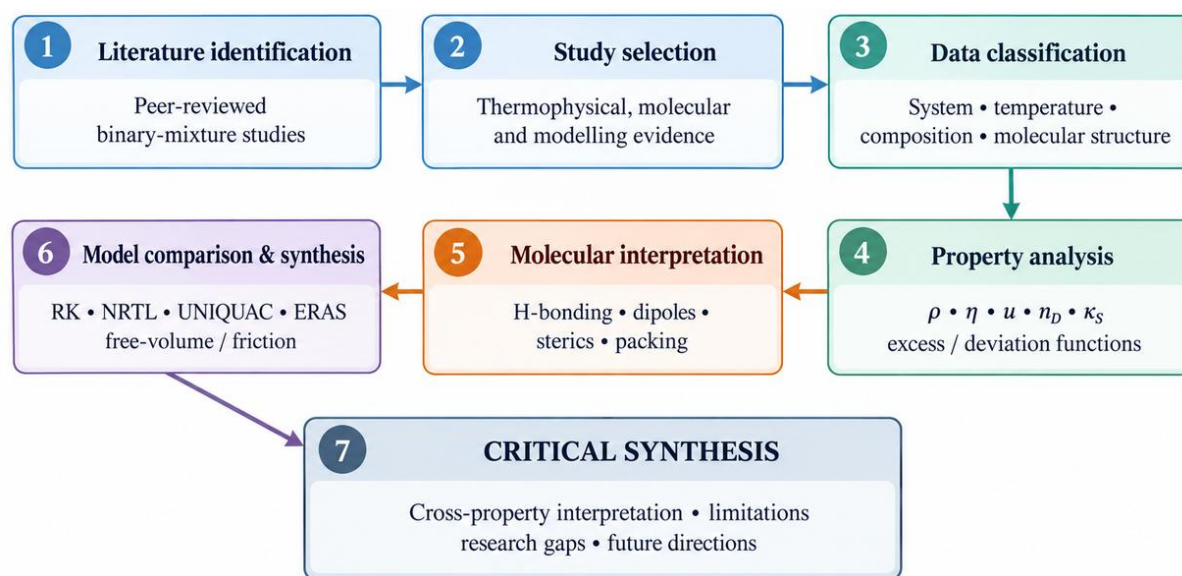


**Figure 3: Research Methodology**

The researchers reviewed scientific papers that described research on multicomponent liquid mixtures. Of particular interest were papers that reported data on refractive index, sound velocity, viscosity, and density of liquid mixtures and excess molar volumes. The researchers also reviewed papers on hydrogen bonding, other intermolecular interactions, overall interactions between mixture components, and the effects of composition and temperature on mixture properties.

The authors organized and reviewed selected studies according to various fluid mixture characteristics and applications of interest, including measured physicochemical properties, phase and composition ranges, and physical mixture characteristics as well as modeling techniques used in the studies. Positive and negative deviations from ideality were described and compared to draw conclusions about intermolecular interactions. Use and predictive accuracy of popular mathematical models and theories, such as UNIQUAC, NRTL, Wilson, Margules, ERAS, and Redlich-Kister models and friction and free-volume theories, were also evaluated.

Ultimately, the investigators combined and compared results from multiple experiments to identify relationships between microparticle behavior and other thermophysical properties of two-component fluid systems. The review notes limitations of previous studies and suggests areas for future research. Organizing the information in this way provides a common basis for judging current knowledge and planning future research on two-component fluid systems.

## 5. THERMODYNAMIC AND CORRELATION MODELS

The authors of the sources consulted stress the importance of thermodynamic models and relationships in characterizing the thermophysical properties of binary mixtures. The sources consulted cite many semiempirical and empirical methods, including friction theory and free-volume theory, as well as other methods such as ERAS, UNIQUAC, NRTL, Wilson, Margules, and Redlich-Kister equations.

To model excess properties, researchers often express the Redlich-Kister polynomial in the following common form:

$$Y^E = x_1 x_2 \sum_{k=0}^n A_k (x_1 - x_2)^k$$

In this equation,  $n$  represents the polynomial order, and  $A_k$  represents the adjusted polynomial coefficients. This equation is a useful tool for quantitatively assessing concentration-related excess properties and the magnitude and sign of other thermodynamic properties over a range of concentrations.

In contrast, models including UNIQUAC, NRTL, Wilson, and Margules models are commonly used to model liquid-phase nonideal behavior by estimating interaction parameters and activity coefficients. These models are based on different theories but share the common purpose of mathematically modeling nonideal behavior using interaction parameters that depend on concentration.

A thorough technical evaluation of a curve fitting the data to a given model should include other considerations in addition to the goodness of fit. Other technical considerations for model evaluation include the underlying science of the model, the number of parameters to be estimated, data requirements, generalizability of the model, and the extent to which the model provides molecular insights.

Therefore, methods to judge the effectiveness of a theoretical framework include assessing it against multiple criteria.

The accuracy with which the predicted results of mathematical models match the observed results;

- The total statistical regression errors;
- The consistency of measurements for the variables created;
- The framework's general applicability to all levels of concentration;
- The system's response to temperature variation;
- The successful conversion of derived parameters into physical interpretations; and
- The model's usefulness in predicting other two-liquid mixtures.

**Table 3:** Thermodynamic and Correlation Models

| Model/Approach | Primary Purpose                     | Main Basis                                 | Technical Strength                           | Main Limitation                  |
|----------------|-------------------------------------|--------------------------------------------|----------------------------------------------|----------------------------------|
| Redlich-Kister | Excess-property correlation         | Polynomial representation                  | Simple and effective composition correlation | Mainly correlational             |
| Margules       | Non-ideal liquid behaviour          | Excess Gibbs energy                        | Relatively simple                            | Limited molecular interpretation |
| Wilson         | Activity coefficient estimation     | Local composition                          | Useful for non-ideal systems                 | Parameter dependent              |
| NRTL           | Activity coefficient estimation     | Local composition and non-randomness       | Flexible for strongly non-ideal systems      | Requires interaction parameters  |
| UNIQUAC        | Phase-equilibrium/activity analysis | Molecular size and surface characteristics | More molecularly motivated                   | Higher model complexity          |

|                      |                                    |                                          |                                               |                                 |
|----------------------|------------------------------------|------------------------------------------|-----------------------------------------------|---------------------------------|
| ERAS                 | Excess-volume/association analysis | Association and structural effects       | Useful for molecular interpretation           | Requires additional assumptions |
| Free-volume approach | Transport-property interpretation  | Molecular free volume                    | Useful for viscosity-related behaviour        | Model-dependent                 |
| Friction theory      | Viscosity prediction               | Thermodynamic and friction contributions | Links viscosity with thermodynamic properties | More parameters and assumptions |

This is a broader concern, a model may accurately predict the data but fail to provide useful information about the chemical interactions that led to the observed results.

## 6. INTEGRATED TECHNICAL PERSPECTIVE

The reviewed literature suggests that thermophysical properties of mixtures of two fluids cannot be predicted from the properties of the pure components. Thermophysical properties of a mixture depend on microscopic properties, such as the mixture's density, which depends on how particles are arranged; viscosity, which depends on a fluid's resistance to flow; speed of sound, which depends on a fluid's compressibility; and refractive index, which depends on a fluid's polarizability and other optical properties.

Combining macroscopic data with theoretical predictions of deviations and other properties provides a complete picture of interactions between molecules. Key lessons about the difference between correlation and causation can be learned from an overall view. Agreement between experimental and theoretical data does not mean that the theoretical model explains the cause of the fluid's behavior at the molecular level. Further thermodynamic studies should focus on combining curve-fitting techniques with theories of molecular structure to understand fluid properties.

Composition and Temperature → Molecular Interactions → Packing and Free Volume → Thermophysical Response  
→ Excess/Deviation Properties → Model Correlation

Ultimately, review of previous research shows that deep understanding of liquid mixtures requires an integrated approach. Researchers who take an integrated approach and consider experimental data and calculated properties along with heat and mass transfer effects and kinetic and equilibrium data and other theoretical concepts can gain a complete understanding of liquid mixtures.

## 7. MATHEMATICAL FRAMEWORK FOR THERMOPHYSICAL PROPERTIES

It is common to model the thermophysical properties of liquid mixtures using mathematical parameters that represent departures from ideal behavior and provide information about the molecular interactions in the mixture. This section provides equations commonly used to analyze density, volumetric, acoustic, optical, viscosity, and thermodynamic properties of mixtures.

### Density

The density of a liquid mixture is expressed as:

$$\rho = \frac{m}{V}$$

where  $\rho$  is the density,  $m$  is the mass, and  $V$  is the volume of the mixture. Density variations with composition and temperature are useful for examining molecular packing and structural changes.

### Excess Molar Volume

The deviation from ideal volumetric behaviour is represented by the excess molar volume:

$$V_m^E = V_m - (x_1 V_1 + x_2 V_2)$$

where  $V_m$  is the molar volume of the mixture,  $x_i$  is the mole fraction, and  $V_i$  is the molar volume of the pure component. The sign and magnitude of  $V_m^E$  provide information about contraction or expansion resulting from intermolecular interactions.

### Viscosity Deviation

The viscosity deviation is calculated as:

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2)$$

where  $\eta$  represents the viscosity of the mixture and  $\eta_1, \eta_2$  are the viscosities of the pure components. This parameter is useful for identifying deviations in flow behaviour arising from molecular association and changes in liquid structure.

### Compressibility and Ultrasonic Velocity

The isentropic compressibility is related to density and ultrasonic velocity by:

$$\kappa_s = \frac{1}{\rho u^2}$$

where  $\kappa_s$  is the isentropic compressibility and  $u$  is the ultrasonic velocity. The corresponding ultrasonic velocity is given by:

$$u = \frac{1}{\sqrt{\rho\kappa_s}}$$

These parameters are widely used to investigate molecular packing, cohesive forces, and structural changes in liquid mixtures.

### Refractive Index

The refractive index is defined as:

$$n = \frac{c}{v}$$

where  $c$  is the velocity of light in vacuum and  $v$  is its velocity in the mixture. Changes in refractive index with composition can provide information about molecular polarizability and optical response.

### Excess Gibbs Energy and Activity Coefficient

The excess Gibbs energy provides a thermodynamic measure of non-ideal mixing and can be expressed for a binary mixture as:

$$G^E = RT(x_1 \ln \gamma_1 + x_2 \ln \gamma_2)$$

where  $R$  is the gas constant,  $T$  is the absolute temperature, and  $\gamma_i$  represents the activity coefficient of component  $i$ .

The activity coefficient is related to the activity through:

$$a_i = \gamma_i x_i$$

where  $a_i$  is the activity and  $x_i$  is the mole fraction. For an ideal solution,  $\gamma_i = 1$ , whereas deviations from unity indicate non-ideal molecular interactions.

**Table 4:** Mathematical Equations for Thermophysical Properties

| Property             | Governing equation                              |
|----------------------|-------------------------------------------------|
| Density              | $\rho = \frac{m}{V}$                            |
| Excess molar volume  | $V_m^E = V_m - (x_1 V_1 + x_2 V_2)$             |
| Viscosity deviation  | $\Delta\eta = \eta - (x_1 \eta_1 + x_2 \eta_2)$ |
| Compressibility      | $\kappa_s = \frac{1}{\rho u^2}$                 |
| Ultrasonic velocity  | $u = \frac{1}{\sqrt{\rho \kappa_s}}$            |
| Refractive index     | $n = \frac{c}{v}$                               |
| Excess Gibbs energy  | $G^E = RT(x_1 \ln \gamma_1 + x_2 \ln \gamma_2)$ |
| Activity coefficient | $a_i = \gamma_i x_i$                            |

Here,  $\rho$ ,  $V_m$ ,  $\eta$ ,  $\kappa_s$ ,  $u$ ,  $n$ ,  $G^E$ , and  $\gamma_i$  represent density, molar volume, viscosity, isentropic compressibility, ultrasonic velocity, refractive index, excess Gibbs energy, and activity coefficient, respectively. The equations can be used to quantify deviations from ideal mixing and molecular interactions. These equations offer a quantitative relationship between the thermophysical properties measured in experiments and interactions at the molecular level. The molecular packing, the strength of the interactions between the molecules and the variation in their packing density are described primarily by the changes in density, excess molar volume and viscosity deviation, and the changes in liquid structure and cohesive forces are described by ultrasonic velocity and compressibility. The analysis is extended to optical and thermodynamic behaviour by the refractive index, excess Gibbs energy and activity coefficients. Thus, both of these parameters are useful for understanding the non-ideal behaviour of binary liquid mixtures in various composition and temperature ranges.

## 8. RESULTS AND DISCUSSION

The results/discussions as described in this section are quantitative synthesis of representative experimental observations reported for binary liquid mixtures. Specific attention is paid to the influence of excess properties, intermolecular interactions, molecular structure, composition and temperature. This is a review study and therefore discussed in the following numerical values are literature-reported data and not newly measured experimental values.

### 8.1 Temperature and Composition Effects on Thermophysical Properties

The two most significant variables, which influence the thermophysical behaviour of binary liquid mixtures are the temperature and composition of the mixture. The studies reviewed all support the trend of lower density and viscosity with higher temperatures due to the effect of thermal expansion and increased molecular mobility. The extent of the change is, however, very dependent on the molecular structure and the strength of the unlike-molecule interactions. [28] For example, reported data for 1-pentanol show a decrease in density from approximately 0.8150 g cm<sup>-3</sup> at 293.15 K to 0.7846 g cm<sup>-3</sup> at 333.15 K. Over the same temperature interval, its viscosity decreases from approximately 4.0413 to 1.3731 mPa s. Thus, the density decreases by about 3.7%, whereas the viscosity decreases by

approximately 66%, demonstrating the much stronger influence of temperature on molecular mobility than on liquid density.

**Table 5.** Representative temperature dependence of 1-pentanol

| Temperature, T (K) | Density, $\rho$ (g cm <sup>-3</sup> ) | Viscosity, $\eta$ (mPa s) |
|--------------------|---------------------------------------|---------------------------|
| 293.15             | 0.8150                                | 4.0413                    |
| 303.15             | 0.8076                                | 3.0586                    |
| 313.15             | 0.8001                                | 2.1628                    |
| 323.15             | 0.7924                                | 1.6997                    |
| 333.15             | 0.7846                                | 1.3731                    |

The composition effect is also significant as changing the mole fraction also changes the number of unlike molecular contacts. As a result, many properties like density, viscosity, excess volume, and viscosity deviation are often found to be nonlinear functions of composition. Studies of n-decane + alcohol systems, for example, investigated the complete composition range at temperatures between 293.15 and 363.15 K, demonstrating the importance of simultaneous temperature–composition analysis.

**Temperature dependence of 1-pentanol: density changes modestly while viscosity is strongly temperature-sensitive**

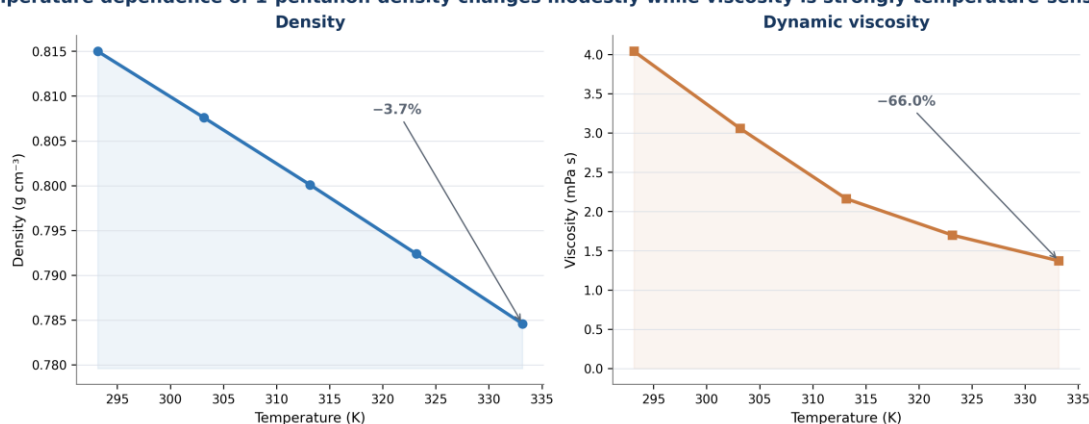


**Figure 4:** Temperature dependence of density and viscosity of 1-pentanol

The temperature dependence of density provides a direct indication of thermal expansion and changes in molecular packing. Figure 4 presents the reported density variation of 1-pentanol over 293.15–333.15 K, showing a progressive decrease with increasing temperature.

## 8.2 Excess Molar Volume and Molecular Packing

Excess molar volume is a key indicator of deviation from ideal volume additivity and is expressed as

$$V_m^E = V_m - x_1 V_{m,1} - x_2 V_{m,2}.$$

Positive  $V_m^E$  values generally indicate expansion relative to ideal mixing, whereas negative values indicate contraction. The sign and magnitude depend on the balance between unlike-molecule interactions, molecular size, shape, steric effects, and packing efficiency.

For n-tetradecane + 1-pentanol, representative reported values vary substantially with temperature and composition. At 293.15 K, values of approximately 1.84, 1.36, 1.36, 1.36 and 5.98 cm<sup>3</sup> mol<sup>-1</sup> were reported for representative composition points. At 303.15 K, the corresponding values were approximately 1.97, 1.21, 0.24, -0.06 and 5.50 cm<sup>3</sup> mol<sup>-1</sup>.

**Table 6.** Representative  $V_m^E$  values for n-tetradecane + 1-pentanol

| T (K)  | $V_m^E$ values (cm <sup>3</sup> mol <sup>-1</sup> ) |
|--------|-----------------------------------------------------|
| 283.15 | 1.66, 1.30, -0.51, -0.80, 6.78                      |
| 293.15 | 1.84, 1.36, 1.36, 1.36, 5.98                        |
| 303.15 | 1.97, 1.21, 0.24, -0.06, 5.50                       |
| 313.15 | 2.24, 1.08, 0.02, 0.30, 3.39                        |
| 323.15 | 2.53, 1.17, 0.24, 0.52, 2.43                        |
| 333.15 | 3.02, 1.37, 0.62, 1.00, 1.67                        |

The occurrence of both positive and negative values demonstrates that molecular packing changes significantly with composition. The small negative value of -0.06 cm<sup>3</sup> mol<sup>-1</sup> at 303.15 K indicates a composition where contraction caused by molecular packing and unlike interactions slightly exceeds the expansion contribution. [29]

The results therefore demonstrate that  $V_m^E$  should not be interpreted only in terms of attractive forces. Molecular size mismatch, disruption of self-association, and steric packing must also be considered.

#### Excess molar volume of n-tetradecane + 1-pentanol across five representative compositions

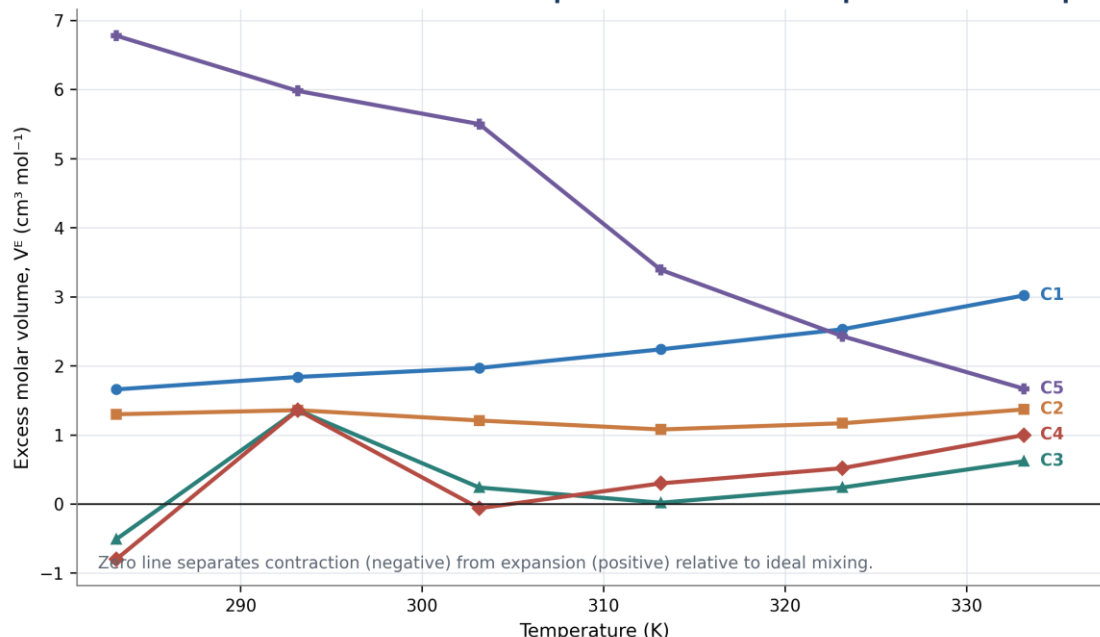


**Figure 5:** Variation of excess molar volume,  $V_m^E$ , with temperature for n-tetradecane + 1-pentanol

Excess molar volume is a useful indicator of deviations from ideal mixing and provides insight into molecular packing and unlike-molecule interactions. Figure 5 illustrates the temperature-dependent variation of  $V_m^E$  for n-tetradecane + 1-pentanol at representative composition points.

### 8.3 Viscosity Deviation and Molecular Interactions

Viscosity deviation is defined as

$$\Delta\eta = \eta - x_1\eta_1 - x_2\eta_2.$$

It provides information about the departure of mixture viscosity from the ideal composition-weighted reference. Positive values indicate enhanced resistance to flow, whereas negative values indicate lower resistance relative to the ideal reference. For n-tetradecane + 1-pentanol, representative viscosity deviations were  $-2.33, 0.72, -0.29, -0.10$  and  $0.49$  mPa s at 293.15 K. At 333.15 K, these values changed to  $-0.79, 0.14, 0.11, 0.04$  and  $1.59$  mPa s.

**Table 7.** Representative viscosity deviations for n-tetradecane + 1-pentanol

| T (K)  | $\Delta\eta$ values (mPa s)        |
|--------|------------------------------------|
| 283.15 | $-3.24, 0.91, -0.54, 0.06, 0.76$   |
| 293.15 | $-2.33, 0.72, -0.29, -0.10, 0.49$  |
| 303.15 | $-1.71, 0.51, -0.10, -0.05, 0.37$  |
| 313.15 | $-1.30, 0.39, -0.001, -0.07, 0.58$ |
| 323.15 | $-0.99, 0.27, 0.05, -0.008, 0.67$  |
| 333.15 | $-0.79, 0.14, 0.11, 0.04, 1.59$    |

The first composition point shows a systematic reduction in the magnitude of negative viscosity deviation from  $-3.24$  mPa s at 283.15 K to  $-0.79$  mPa s at 333.15 K. This represents an approximately 75.6% reduction in the magnitude of the deviation.

Thermal motion dominates more and more the structural interactions that are responsible for the negative deviation as the temperature increases. However, at certain compositions the deviation becomes negative and gradually approaches positive values as the temperature is increased, meaning that composition-dependent molecular interactions are still significant at high temperatures.

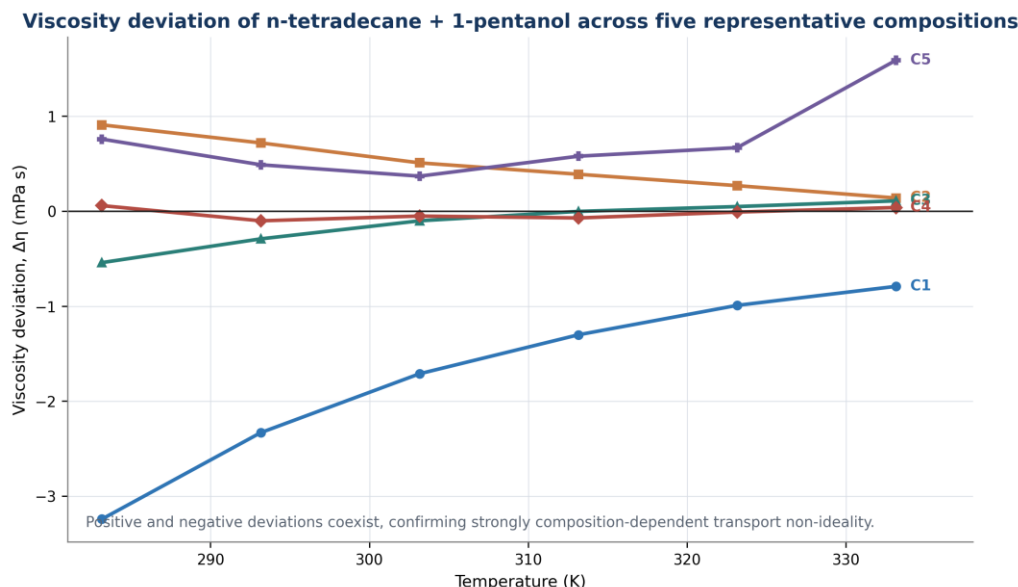


**Figure 6:** Temperature dependence of viscosity deviation,  $\Delta\eta$ , for n-tetradecane + 1-pentanol

Viscosity deviation reflects the influence of molecular association, packing, and changes in intermolecular mobility upon mixing. Figure 6 shows the variation of  $\Delta\eta$  with temperature for n-tetradecane + 1-pentanol, highlighting both positive and negative deviations.

#### 8.4 Effect of Molecular Structure and Hydrogen Bonding

Molecular structure plays a critical role in determining thermophysical behavior. A particularly useful comparison is provided by n-tetradecane + 1-pentanol and n-tetradecane + 2-pentanol. Although 1-pentanol and 2-pentanol have the same molecular formula, their hydroxyl-group positions differ, producing different steric environments and packing efficiencies.

For n-tetradecane + 2-pentanol, reported  $V_m^E$  values include  $-3.12 \text{ cm}^3 \text{ mol}^{-1}$  at 323.15 K for one composition, whereas the corresponding 1-pentanol system exhibits predominantly positive values at the same temperature range. This difference demonstrates that molecular topology can change the balance between contraction and expansion during mixing.

**Table 8.** Comparison of representative excess-volume behavior

| System                     | Temperature (K) | Representative $V_m^E$ range ( $\text{cm}^3 \text{ mol}^{-1}$ ) | Structural implication                        |
|----------------------------|-----------------|-----------------------------------------------------------------|-----------------------------------------------|
| n-Tetradecane + 1-pentanol | 283.15–333.15   | –0.80 to 6.78                                                   | Packing and structural disruption             |
| n-Tetradecane + 2-pentanol | 283.15–333.15   | –3.12 to 7.13                                                   | Stronger composition-dependent packing effect |

A different interaction regime is observed in alcohol–amine mixtures. Methanol + n-butylamine and methanol + di-n-butylamine show large negative excess molar volumes accompanied by positive viscosity deviations. Such a combination is consistent with strong cross-association between the hydroxyl group of methanol and the nitrogen-containing functional group of the amine.

Similarly, AMP + methanol, ethanol, 1-propanol and 2-propanol systems exhibit negative viscosity deviations over the investigated composition range of 298.15–318.15 K. The reported interaction strength follows the order

$$\text{MeOH} > \text{EtOH} > 1 - \text{PrOH} > 2 - \text{PrOH}.$$

The weaker interaction observed for 2-propanol has been associated with increased steric hindrance around the hydroxyl group. FT-IR observations provide additional support for hydrogen-bond formation between AMP and the alcohol molecules.

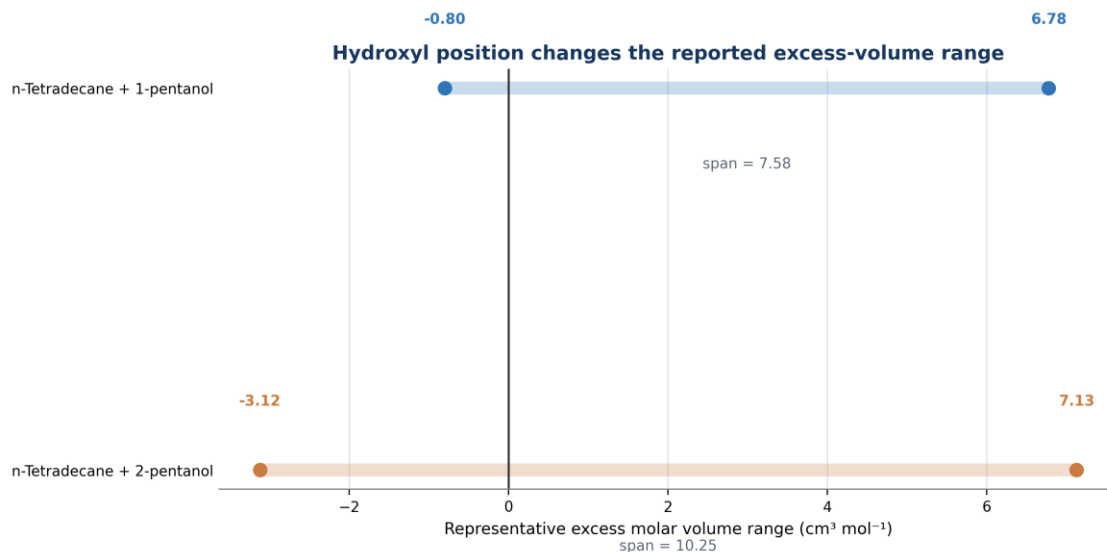


**Figure 7:** Comparison of excess molar volume for n-tetradecane + 1-pentanol and n-tetradecane + 2-pentanol

The position of the hydroxyl group can significantly modify molecular packing and intermolecular interactions in alcohol-containing mixtures. Figure 7 compares the representative  $V_m^E$  ranges of n-tetradecane + 1-pentanol and n-tetradecane + 2-pentanol to illustrate the effect of molecular structure.

### 8.5 Comparative Modelling and Integrated Molecular Interpretation

The quantitative results demonstrate that thermophysical properties should be interpreted together rather than independently. [30] Their combined behavior provides a more reliable indication of molecular interactions.

The Redlich–Kister equation is frequently used to correlate excess properties:

$$Y^E = x_1 x_2 \sum_{k=0}^n A_k (x_1 - x_2)^k.$$

Its principal advantage is the ability to represent nonlinear composition dependence using a limited number of adjustable coefficients. However, fitted coefficients should not automatically be regarded as direct measures of specific molecular interactions.

**Table 9.** Integrated interpretation of representative literature results

| System                     | Main numerical observation                              | Thermophysical interpretation                       | Molecular interpretation                     |
|----------------------------|---------------------------------------------------------|-----------------------------------------------------|----------------------------------------------|
| n-Tetradecane + 1-pentanol | $V_m^E = -0.80$ to $6.78$ $\text{cm}^3 \text{mol}^{-1}$ | Strong composition dependence                       | Packing and structural effects               |
| n-Tetradecane + 2-pentanol | $V_m^E = -3.12$ to $7.13$ $\text{cm}^3 \text{mol}^{-1}$ | Larger negative deviations at selected compositions | Effect of hydroxyl position/steric structure |
| n-Tetradecane + 1-pentanol | $\Delta\eta = -3.24$ to $1.59$ $\text{mPa s}$           | Non-ideal transport behavior                        | Mobility and molecular packing               |
| Methanol + n-butylamine    | Large negative $V_m^E$ positive $\Delta\eta$            | Strong contraction and flow resistance              | O–H···N hydrogen bonding                     |
| AMP + alkanols             | Negative $\Delta\eta$                                   | Strong non-ideal transport behavior                 | Hydrogen bonding and molecular association   |

Conclusion from the comparison exercise include three key insights. First, temperature has a large impact on molecular mobility and viscosity. Second, composition has a large impact on the number of unlike and like interactions among molecules and, consequently, excess properties. Finally, molecular structure and functional groups play a major role in determining whether non-ideal behavior is dominated by packing, association, steric effects, or other factors.

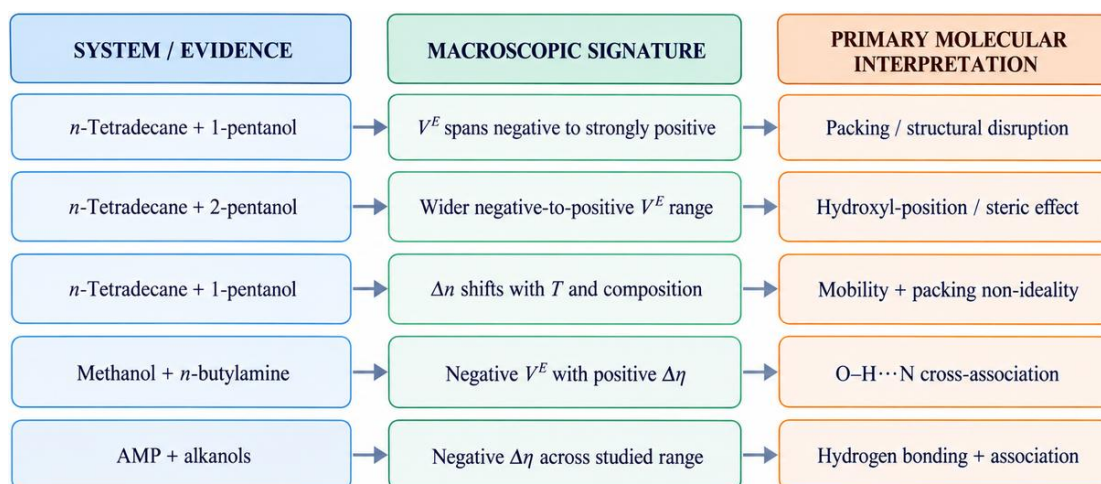


**Figure 8:** Integrated interpretation of representative thermophysical evidence

Excess properties can be used to understand relationships between macroscopic and molecular properties. A comparative integrated model diagram for the binary systems in this study is shown in Fig. 8, illustrating differences among the studied systems related to molecular structure and hydrogen bonding and packing interactions.

The results suggest an integrated framework consistent with that presented in this review:

Molecular structure → Intermolecular interactions → Molecular packing/mobility → Thermophysical properties  
 → Excess/deviation properties → Thermodynamic modelling.

In general, numerical data from the literature indicate that no single thermophysical property can fully explain the molecular nature of nonideal behavior. Density, viscosity, excess molar volume, viscosity deviation, acoustic/optical data, and spectroscopic data can help explain the molecular nature of nonideal liquid mixtures. A combined analysis of all these properties is particularly helpful when the liquid mixture contains strongly associating compounds such as alcohols or amines, or other complex compounds.

## 9. FUTURE SCOPE

Additional studies of thermophysical properties and other intermolecular interactions in two-component liquid mixtures are needed to provide more information about the properties and behavior of these mixtures. Future experimental studies of two-component liquid mixtures over a wider range of temperatures and compositions will help confirm and extend information on liquid structure and intermolecular interactions. Greater focus on other solvents, including deep eutectic mixtures, ionic liquids, and other bio- and environmentally friendly solvents, is also needed. One promising area for future research is the simultaneous measurement of multiple thermophysical properties of two-component liquid mixtures. Measuring properties such as density, viscosity, speed of sound, refractive index, heat capacity, and excess properties in addition to individual properties can provide a better understanding of intermolecular interactions. Additional spectroscopic techniques, such as NMR and FTIR spectroscopy, can help verify other information on structure and hydrogen bonding.

The field needs better theories to explain concentration and temperature gradients and other structural features in mixed fluids. Combining tools such as machine learning and thermodynamic models with experimental data at small scales can help researchers better understand mixed fluids in the absence of direct observations. Standardizing and sharing other experimental data in well-documented databases can help other researchers develop and test theories of mixed fluids. Other steps researchers can take to study mixed fluids include improving the comparability and reproducibility of their studies of competing correlational models. Benefits of these improvements include practical applications of mixed-fluid research to industries such as pharmaceuticals and energy, including improvements to separation operations and solvent design.

## 10. CONCLUSION

This review paper provides a detailed analysis of interparticle forces and other physical properties of liquid mixtures of two components. Several properties, such as refractive index, compressibility, viscosity, density, speed of sound, and changes in viscosity and molar volume, can be used to study the thermodynamic and physical properties of liquid mixtures. Deviations from Raoult's law are generally caused by interactions between components in the mixture, such as steric effects, molecular aggregation, van der Waals forces, dipole-dipole interactions, and hydrogen bonding.

Solute concentration and temperature are key factors affecting macroscopic thermophysical properties and microstructural features. Information about the nature and magnitude of deviations and other excess properties can help you understand differences in packing behavior and the relative strengths of self- and cross-associations between

molecules. Reviewed studies highlight the importance of using thermodynamic and mathematical models and theories, such as the ERAS, UNIQUAC, NRTL, Wilson, Margules, and Redlich-Kister models as well as friction and free-volume theories, to interpret experimental data on nonideal systems.

Although significant progress has been made in understanding binary liquid mixtures, most research to date has focused on individual pairs and other specific properties of individual liquids. A more systematic approach to studying binary liquid mixtures that examines relationships among various properties and behaviors of the mixtures and their structures could be beneficial. Combining machine learning and other computational techniques with thermodynamic modeling and microscopic and empirical approaches may help researchers gain a better molecular-level understanding of binary liquid mixtures and make other predictions. Ongoing study of binary liquid mixtures is important for developing and improving industrial processes and products, including separation technologies and industrial solvents.

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