

Molecular Interaction and Intermolecular Dynamics of Mentha Oil Investigated by Multi-Frequency Ultrasonic Techniques

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Abstract:

Understanding of the molecular connections along with the intermolecular dynamics of essential oils is of importance due to the improvement of quality, stability, processing, and applications of essential oils. Among all methods, ultrasonic methods are known for being cheap, quick, and sensitive for the study of liquids' internal structures and the properties. Despite the fact that studies on essential oils have been conducted so far, most of them were restricted by single-frequency measurements or studies on thermophysical properties on the macro level, which did not provide a significant number of information regarding molecular mechanisms of the structure changes with temperature. In particular, large studies that would allow clarifying the influence of temperature and ultrasonic frequency on the intermolecular dynamics of Mentha oil are missing. We conducted the study on intermolecular dynamics and interactions of the aqueous solution of Mentha oil within the temperature range from 293.15 K to 343.15 K while conducting multi-frequency ultrasonic testing at frequencies 2 and 3 MHz. We measured ultrasonic velocity, density, and viscosity and evaluated parameters such as acoustic impedance, adiabatic compressibility, intermolecular free length, relaxation time, absorption coefficient, and wavelength using thermodynamic equations. According to the results of the experiments, increasing temperature decreases the ability of molecules to interact and function together. Also, increasing temperature influences different aspects like density, viscosity, acoustic impedance, relaxation time, and ultrasonic absorption. In addition, the research shows differences in adiabatic compressibility, intermolecular free length, and ultrasonic velocity which result in a better movement of molecules, increase in the distance between them, and signify the existence of changes ongoing in the liquid. Another observation made in the course of this research is that 3 MHz frequency can be considered to be more efficient than 2 MHz frequency for recognizing more slight changes that occur due to the temperature changes. Thus, it allows revealing definite processes ongoing in the molecules. Such findings prove the efficiency of the multi-frequency ultrasonic characterization method and show the possibilities of this method to studies the influence of thermal excitation. What is more, the research reveals more information on the relationship between the impact of heating, intermolecular forces, and acoustic response when it comes to Mentha oil. The study can be considered useful since it contributes to the understanding of liquid systems in terms of temperature-dependent processes of molecules interaction.

Keywords: Mentha oil; Molecular interactions; Multi-frequency ultrasonics; Thermo-acoustic properties; Ultrasonic velocity; Acoustic impedance; Adiabatic compressibility; Intermolecular free length; Relaxation time; Liquid-state dynamics.

1. Introduction

Essential oils are intricate compositions of volatile organic compounds derived from aromatic plants and are commonly used in pharmaceutical, cosmetic, food, and perfume manufacture due to their biological activity and specific smell along with therapeutic properties. The chemical and physical properties of these oils depend on the types of molecules and physical forces existing between them [1]. Temperature and outside factors may alter the molecular structure, affecting such properties as density, viscosity, compressibility, acoustic transmission, and thermal stability. Thus, the insight into molecular interactions and structure of essential oils is crucial in order to improve the quality of the final product and optimize the production process [2-4].

Among commercially useful essential oils, Mentha oil stands out because of high menthol content and extensive use of Mentha oil in the pharmaceutical and confectionary industry, cosmetics and oral hygiene products, and aromatherapy [5]. Though the specific chemical composition of Mentha oil makes it effective for many purposes, its efficacy is defined by molecular arrangement and behaviors depending on the temperature. Heat alters intermolecular forces, molecular mobility and structure of the medium, which influence the speed of sound waves in the medium[6-7].

Ultrasonic methods represent a modern approach to non-destructive analysis of liquids. These techniques supply indirect information about molecular interactions, elastic properties, cohesive forces, and structural relaxation. Unlike traditional physicochemical methods, which deal with general properties, ultrasonic parameters are extremely sensitive to changes in molecular order and intermolecular interactions [8-10]. Various parameters such as the velocity of ultrasound, acoustic impedance, compressibility, intermolecular free length, relaxation time, and ultrasound absorption enable the thorough study of liquid dynamics [11]. The ultrasonic methods were widely applied for the study of essential oil and liquid mixtures, but the majority of studies were limited to the measurements with a single ultrasonic frequency and research of temperature dependence of individual thermo-acoustic parameters. In those studies, only sparse attention was given on the analysis of those parameters in terms of molecular interactions, intermolecular dynamics, and structural reorganization. The influence of ultrasonic frequency on the sensitivity of molecular characterization has not been analyzed [12-15].

This study explored the molecular interactions and the intermolecular dynamics of aqueous Mentha oil using ultrasonic measurements conducted at both 2 MHz and 3 MHz frequencies and the temperature range ranging from 293.15 to 343.15 K. By measuring ultrasonic velocity, density, and viscosity, and deriving thermo-acoustic parameters like acoustic impedance, adiabatic compressibility, intermolecular free length, relaxation time, absorption coefficient, and wavelength, the study evaluated temperature-dependent changes in molecular packing, intermolecular forces, elastic behavior, and acoustic energy transfer [16-18]. The integration of thermal and multi-frequency measurements allowed for the thorough understanding of Mentha oil behavior and successful demonstration of ultrasonic techniques for obtaining information about internal structures of essential oils. The way in which molecules act in liquids is determined by the constant interaction between attractive intermolecular forces, molecular movement, and thermal energy. Unlike regular solids, liquids have only short-range molecular order, so their structure is very flexible and can change depending on external factors [19-20]. Temperature changes affect the

relationship between adhesive and disruptive forces, which leads to different arrangements of molecules and varying amounts of free space in a volume, as well as energy transmission. Properties at a microscopic level are reflected in macroscopic features, including bulk density, viscosity, compressibility, and sound speed. Therefore, studying the interconnection between molecular interactions and the properties of acoustic waves can be particularly helpful in revealing the structure and behavior of liquid substances [21-23]. Ultrasonic techniques have proved to be important in investigating the main characteristics of liquids since they allow us to study the reaction of the liquid to ultrasonic waves. The propagation of sound waves through the medium is determined by the density, compressibility, and viscoelasticity of the liquid, which depend on molecular forces. As soon as the wave moves through the liquid, it causes periodic movement of its particles. The way the molecules behave in response to this perturbation conveys the strength of intermolecular forces, molecular arrangement and relaxation processes taking place in the given liquid. Therefore, ultrasound measurements give indirect but very sensitive information about molecular arrangement in the system without changing the sample [24-27].

A range of thermoacoustic parameters extracted from the ultrasonic measurements can be treated as critical indicators of liquid structure and molecular dynamics. Ultrasonic sound velocity reveals the elasticity of the studied liquid and its resistance to deformation during the acoustic sound wave passage. Acoustic impedance serves to indicate resistance faced by the sound waves and depends on the sound velocity and mass density. The adiabatic compressibility indicates how the volume of the liquid changes when the pressure is applied and is directly related to intermolecular coherence. Intermolecular free length, i.e. the average distance between neighbouring molecules, gives the data concerning the packing of molecules in the liquid. Temperature is a major factor in the alterations of the mentioned thermo-acoustic properties since temperature affects molecular motion and the interactions between molecules constantly. With the increase of thermal energy, the molecules obtain more kinetic energy which lessens the attractive forces and induces thermal expansion. The change in the intermolecular distance leads to the decrease of the liquid density and viscosity, as well as influences the compressibility, the acoustic impedance, and the relaxation behaviour of the liquid [28]. All of these temperature-driven changes affect the transmission of ultrasonic waves in the medium and make the ultrasonic measurements appropriate for the study of thermal effect on the liquid microstructure. It is crucial to find or understand these connections since they will help in prediction of the functioning of essential oils, stability and processing. Although lots of studies have used the ultrasonic techniques to study binary liquid mixtures, pharmaceuticals, vegetable oils, and essential oils, most of them have investigated only one parameter of thermo-acoustic properties or the dependence of one thermo-acoustic parameter on the temperature at one ultrasonic frequency. This paper adds to the increasing use of ultrasonic techniques in exploring the properties of liquids and making an in-depth interpretation of molecular interactions rather than providing just an account of changes in thermo-acoustic parameters. The results proved to be beneficial for future applications of ultrasonic methodologies in the field of essential oils as well as other complex liquid systems.

2. Materials and Methods

2.1 Materials

Mentha oil that is commercially available was used as the sample in this study. The oil was purchased from a legitimate supplier and remains unpurified to maintain its original properties. The oil was kept in airtight brown glass bottles placed at room temperature to avoid oxidation and evaporation of the oil. Prior

to each measurement, the sample was allowed to reach the temperature at which the measurements were conducted.

2.2 Experimental Approach

The thermo-acoustic experiments were realized using a single crystal ultrasonic interferometer with frequencies of 2 and 3 MHz. The interferometer was equipped with a quartz crystal transducer, which allowed for the generation and detection of the ultrasonic waves accurately. The temperature was managed with a thermostatic constant temperature circulating water bath with precision of ± 0.1 K. To measure density, a pycnometer was used, while the dynamic viscosity of the sample was measured using an Ostwald viscometer under the same thermal conditions. Prior to the experiments, all measuring instruments had been calibrated to avoid systematic errors. The measurements were repeated three times at each temperature, and the average values were taken.

2.3 Experimental Situation

The experiments were carried out at 293.15–343.15 K with the temperature increasing by 10 K in each step. Ultrasonic measurements were done in the same condition but only at 2 and 3 kHz. The details of experimental settings are given in Table 1.

Table 1. Experimental conditions used for ultrasonic measurements.

Parameter	Value
Sample	Aqueous Mentha oil
Temperature range	293.15–343.15 K
Temperature interval	10 K
Ultrasonic frequencies	2 MHz and 3 MHz
Measured parameters	Ultrasonic velocity, density, viscosity
Derived parameters	Acoustic impedance, adiabatic compressibility, intermolecular free length, relaxation time, absorption coefficient, wavelength

2.4 Experimental Procedure

The sample was kept in the thermostatic bath until it reached thermal equilibrium at the particular temperature. The ultrasonic velocity was obtained through the ultrasonic interferometer by examining the wave pattern between the two successive maxima. The density value was obtained from the calibrated pycnometer, while the viscosity was obtained through the Ostwald viscometer. This procedure for determining the ultrasonic velocities was repeated for both ultrasonic frequencies so that the data obtained will be consistent and comparable. The values of all aforementioned parameters obtained were used to evaluate the rest of the thermo-acoustic parameters mentioned above.

2.5 Evaluation of Thermo-Acoustic Parameters

The experimentally measured ultrasonic velocity, density, and viscosity were used to calculate the thermo-acoustic parameters according to standard relationships:

- **Acoustic Impedance**

$$Z = \rho v$$

- **Adiabatic Compressibility**

$$\beta = \frac{1}{\rho v^2}$$

- **Intermolecular Free Length (Jacobson's relation)**

$$L_f = K\sqrt{\beta}$$

where (K) is Jacobson's temperature-dependent constant.

- **Relaxation Time**

$$\tau = \frac{4\eta}{3\rho v^2}$$

- **Ultrasonic Absorption Coefficient**

$$\alpha = \frac{8\pi^2 f^2 \eta}{3\rho v^3}$$

- **Wavelength**

$$\lambda = \frac{v}{f}$$

These parameters collectively provide valuable information regarding molecular interactions, liquid elasticity, intermolecular spacing, structural relaxation, and acoustic energy dissipation within the liquid medium.

3. Results and Discussion

3.1 Temperature-Dependent Molecular Interactions

The behavior of ultrasonic waves propagating in a liquid depends on molecular structure, cohesion, and elasticity of the liquid. Thus differences in ultrasonic speed and density and viscosity of the liquid can show how molecules function at the determined temperature. The complex study of these parameters suggests some conclusions about molecular arrangement and intermolecular distance of Mentha oil. The obtained results indicate that the temperature has a huge impact on Mentha oil. When the temperature increases, the kinetic energy of the molecules rises as a result of which interaction forces are not effective and coherent interactions become weaker. Thus, the possibility of molecular movements is enhanced and distance between molecules changes. The results of the density measurements show that the increase in

temperature results in thermal expansion, which means that the effective volume of the molecules is larger due to constant mass. Moreover, the decrease in viscosity reveals a decrease in internal friction, which indicates that molecules are moving more freely due to thermal motions that exceed cohesive forces.

Ultrasonic speed showed a non-linear dependence on temperature at both 2 MHz and 3 MHz, indicating that the propagation of sound is determined by density, and elastic properties of the liquid. These observations indicate that the temperature causes constant molecular rearrangement in the liquid, leading to local modification of intermolecular interaction and structural organization. Because the ultrasonic speed is closely related to the elastic modulus of the medium, its changes can indicate the changes in the molecular packing and liquid stiffness. Moreover, a comparison of measurements made at 2 MHz and 3 MHz showed that the ultrasonic frequency at 3 MHz has higher sensitivity for the changes in the structure caused by the temperature effect. The measurements performed at both frequencies showed similar trends, although the measurements made at 3 MHz clearly showed slight variations in the organization of the liquid molecules. The general behavior of ultrasonic speed and density confirms that the thermo-acoustic behavior of Mentha oil is a result of the continuous change in the molecular mobility, intermolecular cohesion, and liquid structure.

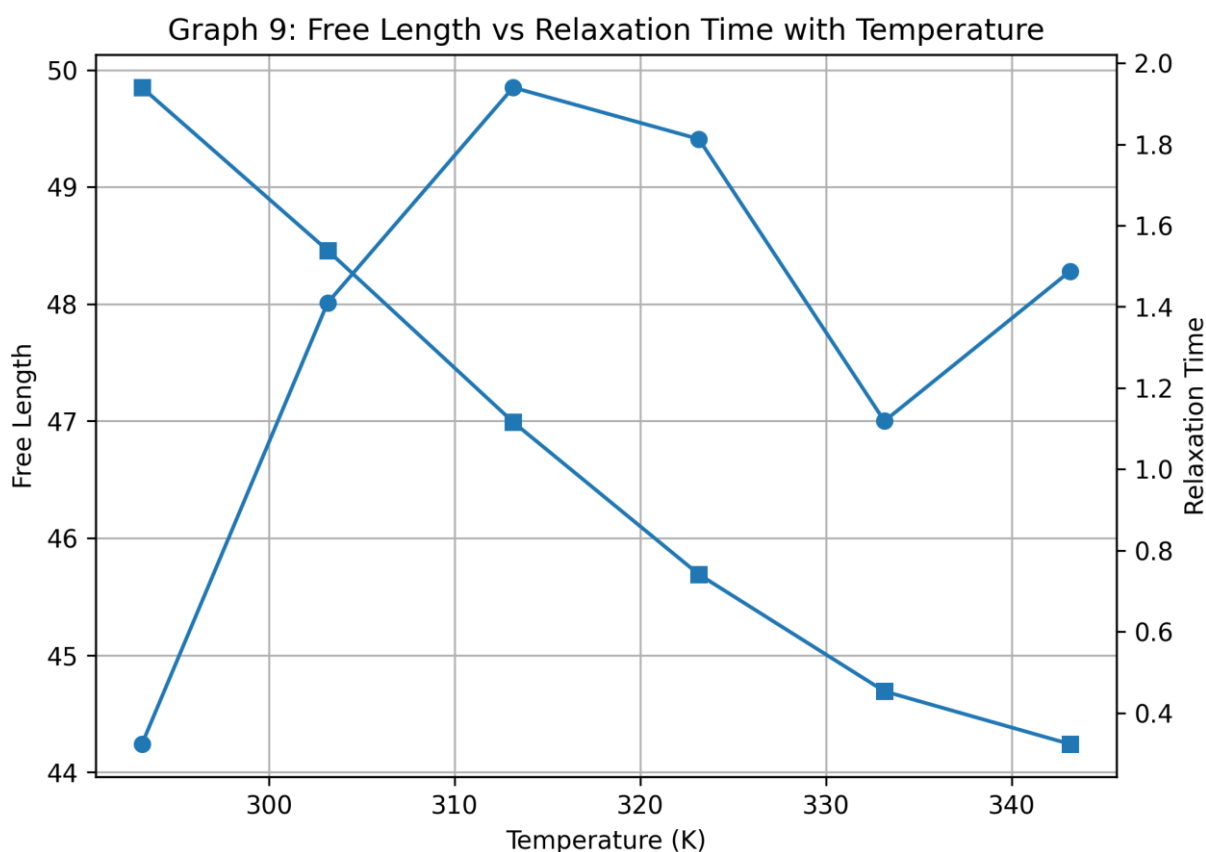


Figure 1: Free Length and Relaxation Time vs Temperature

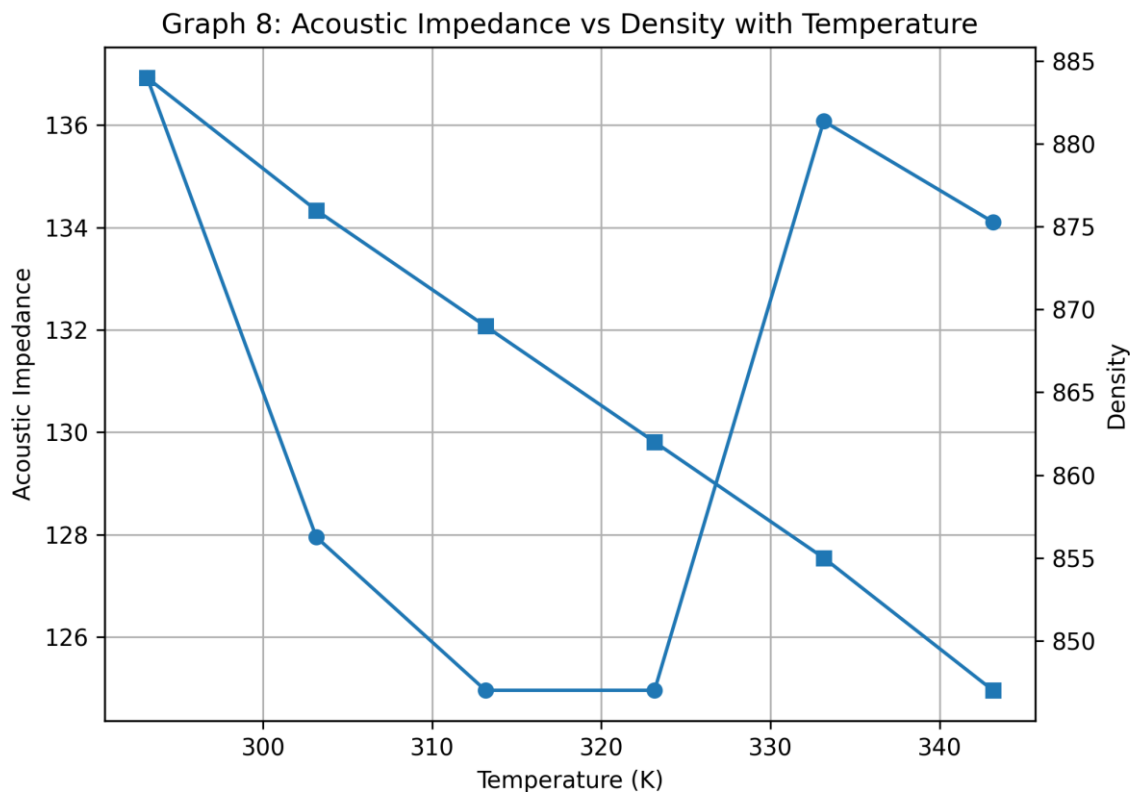


Figure 2: Acoustic Impedance and Density vs Temperature

3.2 Elastic Behaviour and Intermolecular Dynamics

The elastic nature of a liquid depends on the strength of the intermolecular forces as well as the ability of the molecules themselves to react to an outer pressure. When acoustic studies are carried out, some of the important components, which give a complete understanding of the elastic property of the liquid as well as its internal structure, can be Z (acoustic impedance), β (adiabatic compressibility), and L_f (intermolecular free length). Application of the above parameters in tandem helps to understand impact of temperature on molecular processes and their structural changes in Mentha oil. The results of the experiments show that the impact of temperature on the elastic properties of Mentha oil comes out to be rather significant. It has been established that acoustic impedance starts decreasing with growth of temperature due to simultaneous decrease of density along with temperature dependence of ultrasonic speed. The decreasing acoustic impedance proves that there is a minor increase in the mobility of molecules, which is confirmed by an increase of their thermal excitation level.

The discrepancies in compressibility indicate that the elastic modulus of Mentha oil declines when molecular motion increases. Furthermore, the increase in free length of intermolecules confirms this notion. Free length is the average effective distance of adjacent molecules, which changes with thermal expansion. At higher temperatures, molecules have a greater kinetic energy which results in the widening of space in between them and hence increase in free length of the mixture. The comparison of data collected at 2 MHz and 3 MHz frequencies shows similar trend but the analysis of 3 MHz data makes it possible to observe stronger variations of the acoustic impedance value, compressibility, and free length because it is more sensitive to changes in the molecular ordering of the liquid. By looking at the behaviour of acoustic impedance, adiabatic compressibility, and intermolecular free length together, we understand

that thermal excitation is changing the ratio between the cohesion forces and mobility of molecules in continuous manner. So, it can be stated that these phenomena show that acoustic response detected is caused by alteration in many properties of liquid. Thus, multi-frequency ultrasound technique provides a unique way to study the internal structure of Mentha oil.

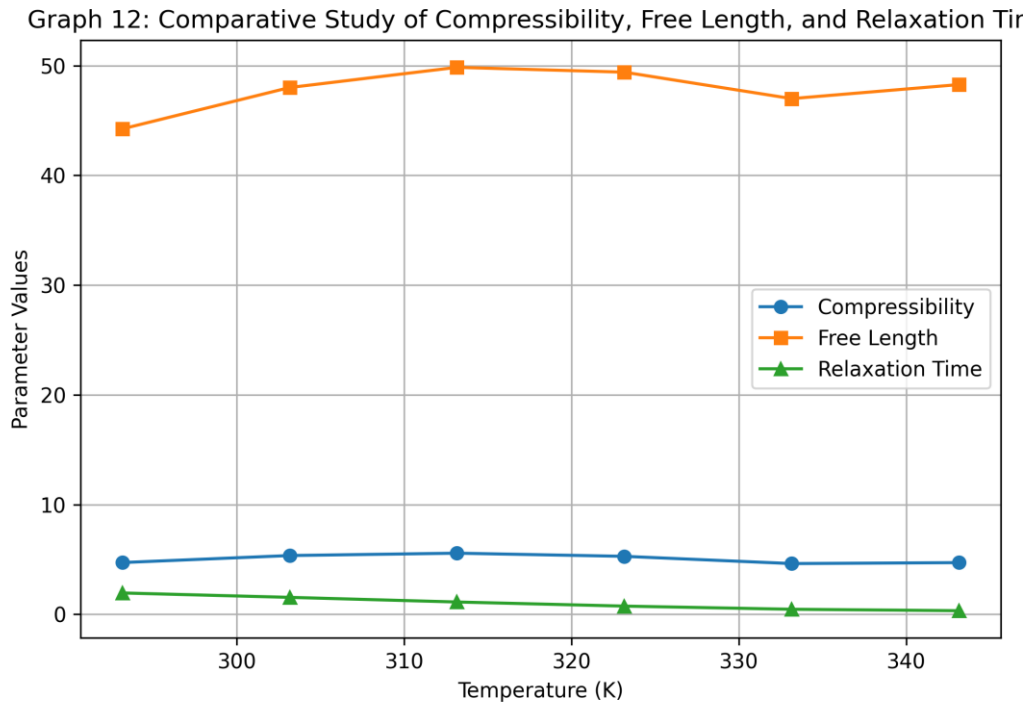


Figure 3: Compressibility, Free Length, and Relaxation Time vs Temperature

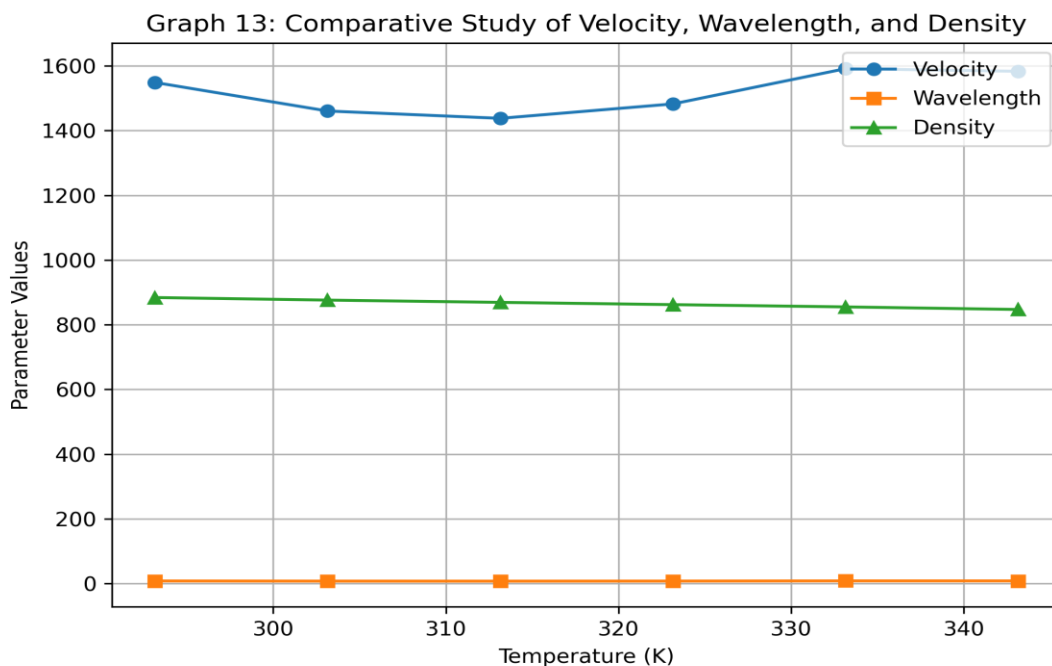


Figure 4: Velocity, Wavelength, and Density vs Temperature

3.3 Structural Relaxation and Acoustic Energy Dissipation

When ultrasonic waves propagate through the liquid, this process involves acoustic energy transfer and dissipation, which is greatly affected by the molecular relaxation processes. Relaxation time, ultrasonic absorption coefficient and wavelength reflect the molecular behavior with respect to the ultrasonic excitation and provide important information about energy dissipation, structural rearrangement, and acoustic transmission in the liquid. All these parameters are connected to the molecular mobility and viscoelasticity of the Mentha oil.

The results obtained from experiments show that relaxation time decreases with increasing temperature for both frequencies of ultrasonic waves: 2 MHz and 3 MHz, which proves that molecular rearrangement processes take place at a higher pace at elevated temperature conditions. Moreover, with the increase in the molecules' kinetic energy, intermolecular attractions forces become weaker, which allows the molecules to be more sensitive to the fluctuations in pressure generated by ultrasonic energy. Thus, the decrease in relaxation time may be interpreted as the improved molecular mobility and increased pace of the structural changes. The same temperature-dependent behaviour is exhibited by ultrasonic absorption coefficient. The decrease in absorption with the increase in temperature indicates that ultrasonic energy waves are absorbed with more difficulty and the energy dissipation is lower in the liquid. In summary, ultrasonic waves travel more easily through liquid when the temperature goes up.

The difference concerning the wavelength also proves that temperature has a certain effect on the acoustics. Since the wavelength can directly influence the ultrasonic speed, its change gives evidence of interaction of two factors - thermal expanding and elastic property changes. Based on the data received, it is possible to say the temperature changes acoustics with regard to molecular structure and elastic properties of Mentha oil.

The comparative analysis of the application of two ultrasonic frequencies shows that the overall tendency is the same, but the measurements at 3 MHz frequency proved to be more sensitive to temperature changes in relaxation process and attenuation process. The reason is that higher frequency improves ability to detect minor changes in relation to molecular behavior in liquid.

The overall joint interpretation of relaxation time, acoustics absorption coefficient and wavelength indicates that the temperature makes molecules move more actively, decreases losses of energy and improves efficiency during the process. It can be concluded that the reason for dynamic reaction of Mentha oil is the combination of three determining factors: molecular relaxation, viscous dissipation and arrangement of structure.

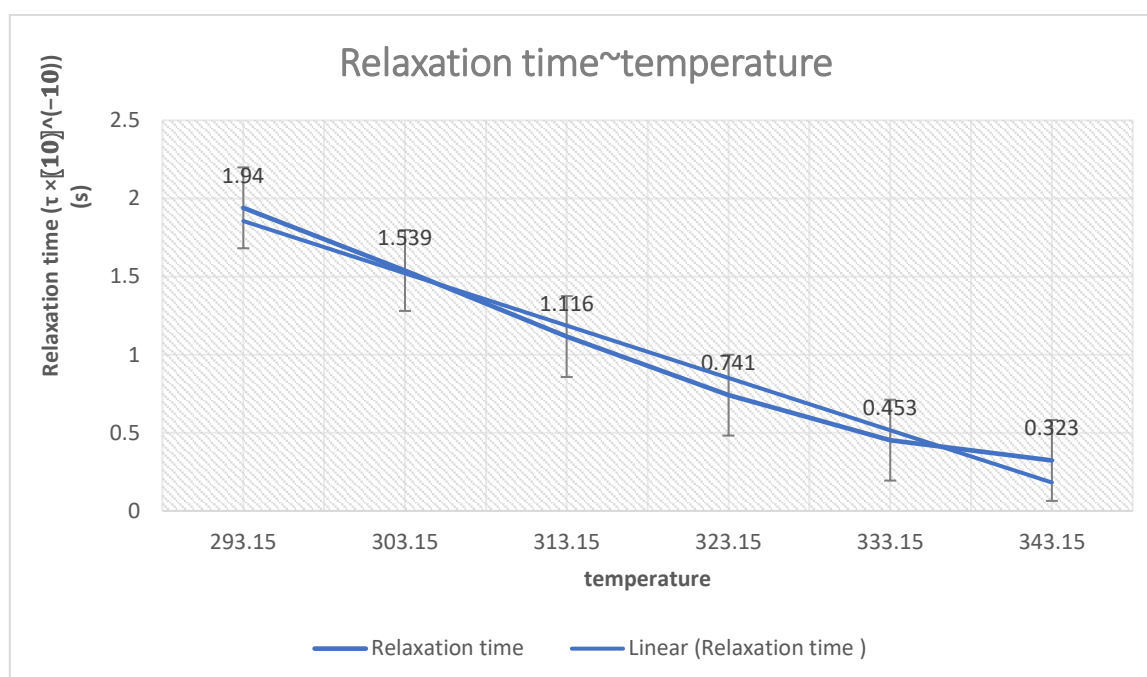


Figure 5. Effect of Temperature on Relaxation Time (τ) of Aqueous Mentha Oil at an Ultrasonic Frequency of 2 MHz

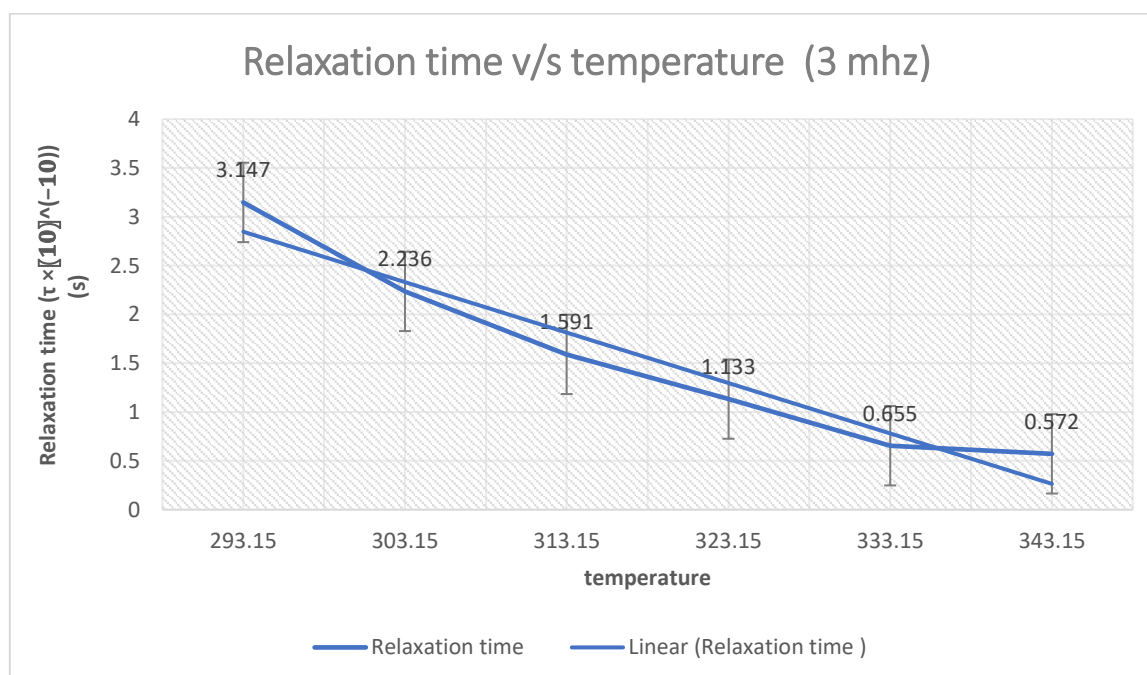


Figure 6. Effect of Temperature on Relaxation Time (τ) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

3.4 Multi-Frequency Analysis of Molecular Response

One of the main purposes of this research was to analyze the effects of ultrasonic frequency on molecular interaction characterization in Mentha oil. Even though temperature influences the overall thermo-acoustic behaviour of a liquid, the ultrasonic waves will also be influenced by the frequency of excitation. A comparative study of two frequencies of 2 and 3 MHz provides useful insights regarding the sensitivity of

both frequencies in detecting the structural changes caused by temperature rise. The results present that both frequencies have the same temperature dependences reflecting the similarity in the molecular processes involved. However, different frequencies demonstrate differences in the amount of changes as well as sensitivity to these changes. The experimental results received using 3 MHz frequency showed a more significant variation in the indices describing ultrasonic velocity, acoustic impedance, compressibility, free length, relaxation time and absorption coefficient. Higher sensitivity of 3 MHz frequency can be explained by a shorter wavelength of ultrasonic waves that results in stronger interaction with microstructural elements of liquids. As temperature increases, molecular motions and thermal expansion lead to changes in intermolecular distances and interaction forces.

The changes in structure can be detected even more vividly at higher frequencies, which makes it easier to distinguish the structural alterations and the elastic character of the material. On the contrary, at 2 MHz, the readings show a smoother response compared to the lower frequencies, as they mostly indicate the thermophysical properties of the liquid.

The comparison further shows that the technique at ultrasonic frequency is an important aspect in molecular characterization optimization. Lower frequencies are more applicable when measuring the general physicochemical behavior of materials while higher ones are better when measuring intermolecular interactions, relaxation, and energy transmission. Therefore, several frequencies can give additional information that cannot be obtained from high constant frequency data. In the end, the multiparametric analysis shows that the use of temperature and ultrasonic frequency leads to better understanding of the behavior of Mentha oil than the investigation of any of the parameters alone.

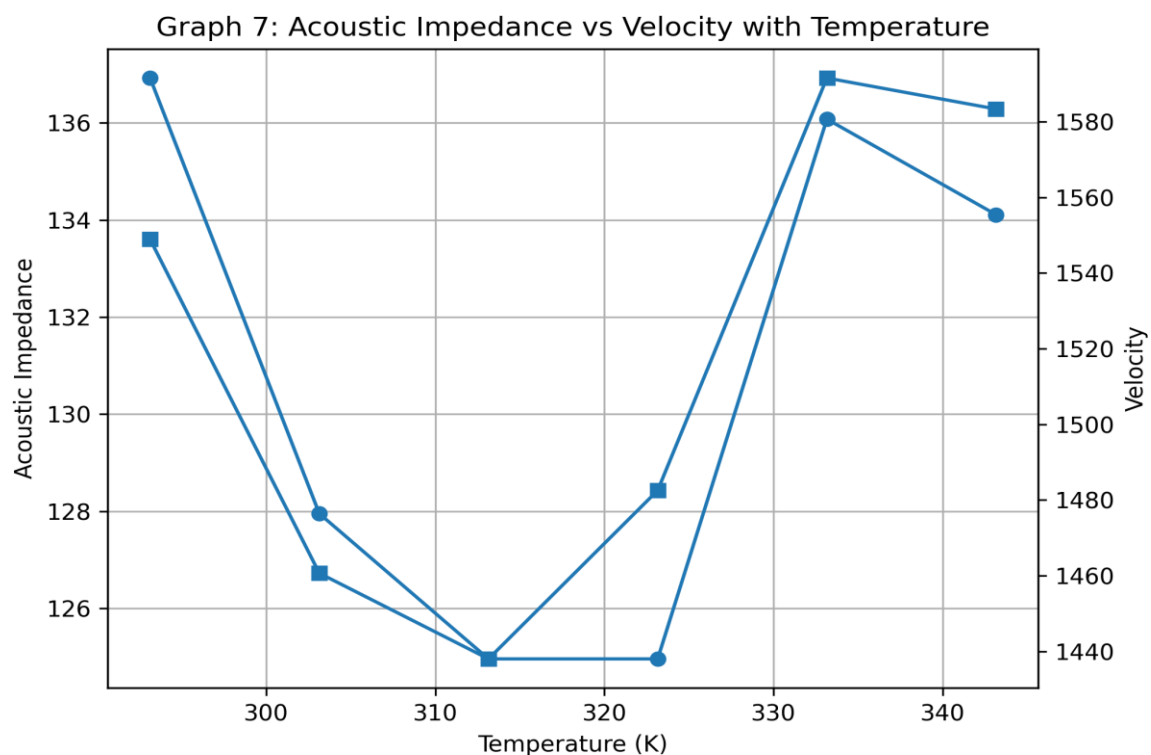


Figure 7: Acoustic Impedance and Velocity vs Temperature

3.5 Proposed Molecular Interaction Mechanism

By looking at how thermo-acoustic parameters were computed and measured through observations, we gain insight into the molecular interactions characterized by the Mentha oil in ultrasonic activation. Any changes such as in ultrasonic speed, density, viscosity, acoustic impedance, adiabatic compressibility, intermolecular free length, relaxation time, and absorption coefficient react not individually but as part of the chain of transformations caused by temperature. A set of thermo-acoustic parameters shapes the subsequent process associated with thermal-induced changes in molecular interactions.

At low temperatures, Mentha molecules possess strong intermolecular forces, keeping them in compact spatial position. This tight arrangement translates into higher density, viscosity, acoustic impedance, and relaxation time, as greater resistance to movement is being shown. Under such conditions the ultrasonic wave penetrates through stable medium with limited space for the molecules to move. Upon increasing the temperature, the heat supplied to the liquid leads to increased molecular motion and thus overcoming intermolecular pull. Consequently, the increase in inter-molecular motion results in molecular expansion and higher spacing in between the molecules and, thus, a decrease in density and viscosity accordingly.

Changes in structure are associated with the improvement of adiabatic compressibility and intermolecular free length values, which suggests a decrease in the rigidity of the liquid structure. Transformation of molecular organization has a significant effect on the propagation of ultrasonic waves. For example, variations of ultrasonic velocity and acoustic impedance are evidence of changing elastic properties of the medium; decreases in relaxation time and ultrasonic absorption coefficient of the liquid suggest that the molecules become faster in responding to the action of ultrasonic waves and dissipate less energy.

The data analysis shows that ultrasonic frequency impacts the degree of sensitivity of the process in question, although mechanisms are the same at both frequencies, it can be seen that measurements made at 3 MHz provide more accurate information about the changes in structure.

Increase in Temperature → Higher Molecular Kinetic Energy → Weakening of Intermolecular Cohesive Forces → Increased Molecular Mobility → Greater Intermolecular Free Length → Reduced Density and Viscosity → Modified Elastic Properties → Changes in Ultrasonic Propagation, Relaxation, and Energy Dissipation.

This mechanistic interpretation provides a unified explanation for the experimental observations and demonstrates that the thermo-acoustic behaviour of Mentha oil is governed by continuous molecular restructuring rather than isolated variations in individual physicochemical properties. The proposed model enhances the understanding of temperature-dependent intermolecular dynamics and highlights the capability of multi-frequency ultrasonic techniques to investigate liquid microstructure in a rapid, non-destructive, and highly sensitive manner.

4. Conclusions

The study aimed to look into the interactions and motion of pairs of water-based Mentha oils. This research was carried out using ultrasonic techniques in frequency ranges of 2-3 MHz and at temperatures between 293.15 K and 343.15 K. Measurements of ultrasonic velocity, density, viscosity and evaluation of acoustic impedance, adiabatic compressibility etc. were performed to study how temperature affects the molecular structure of water-based Mentha oil.

The obtained results indicate that the increasing temperature of the sample leads to the growth of the molecular motion and a decrease in intermolecular forces between molecules; the above leads to density, viscosity, acoustic impedance, relaxation time and absorption coefficient reduction. In addition, the changes of the adiabatic compressibility parameters indicate the influence of molecular structure of Mentha oil on its acoustics and elastics. The major result of this research was the broad frequency comparison of ultrasonic studies. Although both frequencies of 2 MHz and 3 MHz had similar temperature-dependent behavior, the 3 MHz frequency showed higher sensitivity in the detection of slight variations in molecular organization and structure dynamics. These results confirm that ultrasonic frequencies are essential for enhancing molecular characterization resolution; thus, they should be taken into consideration during ultrasonic studies of complicated liquid systems. The studies show that ultrasonic methods are rapid, dependable, and non-destructive methods for examination of the internal structure of essential oils. The present study uses the combination of experimental and calculated values of thermodynamic acoustics parameters to present a comprehensive molecular interpretation of structure changes in Mentha oil due to temperature variations. The scope of future investigations may go beyond using this study by examining different ultrasonic frequencies as well as varied temperature and pressure ranges and various species of Mentha or essential oils with different chemical compositions and properties. Combining ultrasonic measurements with the analytical techniques of GC-MS, FTIR, Raman spectroscopy, and chemometric analysis would shed light on the connections between chemical composition and molecular interactions. Research of this type will enhance how ultrasonic techniques are used as sophisticated tools for characterizing liquids and conducting molecular diagnostics.

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