

Temperature-Dependent Thermo-Acoustic Characterization of Mentha Oil Using Multi-Frequency Ultrasonic Measurements

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Abstract:

Essential oils are indispensable in the pharmaceutical, food, cosmetic, and aromatherapy industries, where their physical and chemical characteristics can significantly affect their processing, shelf life, and properties. Ultrasonic techniques have become very important useful sources of information about what happens in liquids because of their qualities in the field of thermo-acoustics. Although there have been multiple publications reporting the use of ultrasonic techniques for essential oil examination, a vast majority of experimental studies have only focused on one frequency or a narrow temperature interval. Therefore, how the combination of temperature and ultrasonic frequency is affecting the thermo-acoustic characteristics and behavior of Mentha oils is still a mystery. The purpose of the current research is to define how Mentha oils react to the ultrasonic treatment depending on the temperature. The experimental measurements were conducted in the lab on the frequencies of 2 and 3 MHz at temperatures from 293.15 to 343.15 K. The experiments allowed to measure the ultrasonic velocity, density, and viscosity, while other important parameters such as acoustic impedance, adiabatic compressibility, intermolecular free length, relaxation time, absorption coefficient, and wavelength were obtained through the application of the known thermo-acoustic equations. The outcomes of the conducted experiments have shown that the temperature increase leads to the decrease of the density, viscosity, and relaxation time. On the other hand, ultrasonic velocity, the compressibility factor, free length, and wavelength exhibited frequency-dependent changes that convey the combined impact of thermal excitation and acoustic frequency on the way waves propagate. A comparative study revealed that higher ultrasonic frequencies increased the sensitivity of measurements and allowed detecting changes in the structure of a liquid caused by temperature effects with better resolution. The experimentation proves that the multi-frequency ultrasonic characterization is a reliable non-invasive technique for the assessment of the behavior of Mentha oil. The results provide insights into more detailed knowledge of how temperature and frequency affect liquid behavior and demonstrate the opportunity of applying ultrasonic means for quality control, monitoring processes, storing, and characterizing essential oils and similar liquids.

Keywords: Mentha oil; Ultrasonic velocity; Thermo-acoustic properties; Multi-frequency ultrasonics; Acoustic impedance; Adiabatic compressibility; Relaxation time; Essential oils.

1. Introduction

1.1 Essential Oils and Their Industrial Importance

Essential oils are naturally formed volatile mixtures of low molecular weight compounds produced as secondary metabolites by the fragrant plants. These complex mixtures are generally composed of monoterpenes, sesquiterpenes, phenolic compounds, aldehydes, ketones, alcohols, esters, and oxides, with the composition depending on the species of a plant, geographic location, weather, agricultural methods, period of harvesting, and mode of extracting the oils. Due to their unique physicochemical properties as well as various biological activities, the essential oils have attracted lots of scientific and industrial interest for the past several years. Their natural origin, biodegradability, pleasant odor, and wide range of antimicrobial, antioxidant, anti-inflammatory, and curative features have made them good alternatives to synthetic chemical products in many commercial applications [1, 2].

The pharmaceutical sector is one of the largest consumers of essential oils due to their wide medicinal properties. There are many essential oils with antimicrobial, antiviral, antifungal, analgesic, anti-inflammatory, and antioxidant properties, that make them useful ingredients in topical mixtures, medicines for respiratory diseases, oral sanitation products, products for wound healing, and additional therapies. The rising popularity of using natural bioactive agents has led to many studies on how essential oils can be used in pharmaceutical products and delivery systems [3]. In the personal care and cosmetics market, the application of essential oils in perfumes, creams, lotions, soaps, shampoos, deodorants, massage oils, and skin care products is widespread due to their specific scents and useful features. Apart from being attractive to the senses, essential oils can provide beneficial antimicrobial, antioxidant, and skin-conditioning properties gaining further value to the product and making it appealing to customers. The growing interest in plant-based cosmetic ingredients has greatly increased the use of essential oils on the global scale [4].

Within the food industry, essential oils are also used as natural flavour enhancers, preservatives, and functional agents. Thanks to their ability to act against microbes and slow down oxidation reactions, they are in great demand for improving the quality of products and help use fewer synthetic preservatives. Moreover, modern trends in consumer preferences related to clean-label products are encouraging the inclusion of essential oils into drinks, sweets, dairy, edible coatings, and packaging materials [5].

There is an expanding demand for aromatic oil in aromatherapy and complementary medicine. One of the core advantages of aromatic oil is its ability to promote relaxation. Aromatherapy is now incorporated into wellness programs and a variety of alternative medicine practices. More applications of essential oils are still being developed, for example, in agriculture and environmental decontamination, biomedical engineering and advanced functional materials [6].

The growth in industrial applications of essential oils has already resulted in an increased demand for effective characterization techniques that will allow obtaining the necessary information about their physical properties, molecular interactions, and stability. Since these characteristics determine processes of production of aromatic oils, storage, quality assurance of the product, and industrial performance of essential oils, they also create a necessity for reliable analytical tools that will guarantee consistency and authenticity of products. Of all the existing methods of characterization, ultrasonic techniques have proven to be non-destructive and effective since they allow investigating internal structure and molecular behavior of liquids by means of ultrasonic wave propagation. Consequently, ultrasonic characterization has become an increasingly important approach for investigating the thermo-acoustic behavior of essential oils and other complex liquid systems [7].

1.2 Importance of Mentha Oil

Thanks to its distinctive chemical structure, unique smell, and numerous industrial and biological applications, Mentha oil can be classified as one of the most lucrative essential oils. The oil is isolated mainly from the aerial portions of Mentha by means of the steam distillation method. At present, it finds its place in the pharmaceutical, food, cosmetics, fragrance, and aromatherapy industries. The most commercially useful species of the genus are *Mentha arvensis*, *Mentha piperita*, and *Mentha spicata* because their essential oil yields are high and phytochemical composition is unique. The worldwide demand for Mentha oil is on the rise due to the growing tendency among consumers to choose products made with natural components, as well as the increase of the market of plant-produced bioactive substances [8].

Mentha oil is a complex product widely composed of monoterpenes and their oxygen derivatives. Its major components include menthol, menthone, menthyl acetate, limonene, pulegone, carvone, and cineole; however, the content of different components depends on the species, cultivation region, cultivation methods, harvesting time, climate, and extraction processes. Different compositions affect physicochemical parameters of the oil causing differences in its smell, therapeutic properties, and commercial potential. As a result of that, the preservation of the composition. Among its components, menthol is probably the most important bioactive substance that gives Mentha oil its unique cooling sensation and odor. Menthol is used in pharmaceutical preparations, cough syrups, topicals, dental products, inhalation medications, and medicated ointments due to its analgesic, antimicrobial, anti-inflammatory, antioxidant, and antispasmodic properties. Apart from its medicinal uses, menthol is also widely used as a flavouring agent in sweets, drinks, chewing gums, and dental formulations and it is one of the important substances used in perfumery. Thus, the content of menthol is a dependable quality criterion of Mentha oil [9, 10].

The importance of Mentha oil goes beyond its composition. India is one of the largest manufacturers and exporters of Mentha oil and *Mentha arvensis* occupies a considerable part of the international market. The farming of Mentha has become an important source of income for farmers, especially in Northern India (Uttar Pradesh) where the conditions are especially suitable for farming. The growing demand for Mentha oil has stimulated the search for better cultivation methodologies and extraction [11].

Despite the widespread use of Mentha oil commercially, its physicochemical properties and thermoacoustic behaviour in different thermal conditions have not yet been thoroughly studied. The temperature has a dominant impact on density, viscosity, volatility, molecular motion, and interactions which in turn influences ultrasonic wave propagation in the liquid. These parameters define the processing features, as well as quality of the finished product, thus, Mentha oil needs systematic studies under controlled thermal conditions. Ultrasonic techniques are non-destructive and very promising in terms of evaluation of temperature-induced changes in the properties of Mentha oil and knowledge of molecular dynamics of this oil. These studies allow not only to understand the principles of liquid-state physics, but also to formulate effective techniques for quality control, process optimization, and industrial product characterization of Mentha oil [12].

1.3 Ultrasonic Characterization of Liquids

Ultrasonic methods have been introduced as very effective non-invasive approaches for the study of physicochemical and structural characteristics of liquids. In contrast to conventional techniques which mainly provide information about the average features of bulk materials, ultrasonic technique makes it

possible to get indirect information regarding molecular interactions, elastic character, compressibility and structure of liquids. The propagation of ultrasonic waves in a medium is affected by velocity, attenuation, and energy transmission which proves to depend on liquid's density, viscosity, and compressibility, as well as on the forces of interaction within the liquid. As a result, ultrasonic technique has become fruitful in the examination of molecular processes, processes happening in phase and temperature-dependent changes occurring in various types of liquids, solutions, polymer materials, drugs, and other substances [13, 14].

Taking into account the various ultrasonic parameters, ultrasonic velocity is the most fundamental parameter since it is directly indicative of the elastic properties and arrangement of the molecules in the medium. Ultrasonic velocity changes reflect the changes occurring in intermolecular cohesion, free volume, and molecular packing. The increase of values of ultrasonic velocity means the increase of intermolecular interaction forces and higher tightness of structure [15, 16].

The property of density is an important physicochemical parameter that has a marked effect on the propagation of ultrasonics. The concept of density indicates how mass is distributed in a given unit volume of liquid, which reveals the information about the arrangement of molecules and thermal expansion of the substance. The decrease in density of a liquid due to temperature is usually connected with an increase in the space between molecules and reduction in the intermolecular forces of attraction. Since the acoustic impedance is directly dependent on both the density and the ultrasonic velocity, the importance of determining the density of a liquid cannot be undermined when evaluating the acoustic properties of liquids under different experimental conditions. Viscosity denotes the internal resistance of a liquid to flowing and is directly connected to the attractive forces of molecules and molecular mobility. Generally, the viscosity of liquids decreases with the rise of temperature due to the increase in thermal motion and decrease in the intermolecular cohesion. Due to the contribution of viscous forces into acoustic attenuation and molecular relaxation processes, this property renders the greatest importance in defining the process of ultrasonic energy dissipation since the acoustic system is able to characterize [17, 18].

Ultrasonic characterization not only measures physical parameters directly but also determines some derived thermo-acoustic properties such as adiabatic compressibility, acoustic impedance, intermolecular free length, relaxation time, acoustic absorption coefficient and wavelength. All the aforementioned properties explain elastic behavior, molecular arrangement, energy transfer processes and relaxation features of liquids. The adiabatic compressibility indicates how easily the given liquid can be compressed under pressure while acoustic impedance gives an idea about the resistance of ultrasonic wave propagation. Intermolecular free length indicates the average distance between molecules in the liquid. Relaxation time and acoustics absorption, in turn, represent the nature of involved processes for acoustic energy disappearing during molecular relaxation. The increasing usage of ultrasonic methods in investigating liquids may be explained by some positive points related to the way such methods are used, such as rapid investigation, high sensitivity of measurements, little amount of sample preparation and complete preservation of integrity of the sample. These features of ultrasonic technique make it the best method for studying temperature-related physicochemical properties of liquids, monitoring processes in the industry, obtaining information on status of products, performing investigations on interactions between molecules in complex liquids, etc [19, 20].

1.4 Literature Review

The application of ultrasonic techniques for the thermo-acoustic characterization of liquids has gained widespread recognition because ultrasound wave propagation reacts sensitively to changes in the

molecular behavior, density, viscosity, compressibility, and temperature regime. Across a range of studies spanning over several years, ultrasonic techniques have been utilized for the investigation of the physicochemical behavior of organic liquids, both binary and ternary mixtures, ionic liquids, polymers, drug formulations commercial vegetable oils, and essential oils [21, 22]. It has been verified that with the help of ultrasonic techniques, one can receive a lot of indirect information about molecular interactions, structures, the process of relaxation, and the energy transmission modes. Most of the initial work focused on the determination of the ultrasonic velocity and density, which enabled the assessment of the intermolecular interactions in pure liquid and their mixtures. However, many subsequent investigations considered other thermo-acoustic parameters such as adiabatic compressibility, acoustic impedance, intermolecular free length, relaxation time, and ultrasonic coefficients for more effective characterization of liquid behavior [23, 24].

More recently, ultrasonic characterization is beginning to be applied to essential oils. Physicochemical properties of essential oils are extremely important since they determine quality, storability, behaviour in processes, and industry use. Different essential oil has been investigated using ultrasonic methods. The ultrasonic studies describe results of the experiments performed with eucalyptus, clove, lemongrass, cinnamon, basil, and peppermint oils in order to study how temperature and composition affect acoustic properties of essential oils. All studies demonstrated that, when the temperature rises, density and viscosity decrease together with the change of ultrasonic velocity, compressibility, and acoustic impedance due to increased mobility of molecules and decreased intermolecular forces. Nevertheless, the majority of studies are performed using the same ultrasonically frequency and do not allow to learn about frequency-dependent thermo-acoustic behaviour of essential oils [25, 26].

Another direction in research is the investigation of the thermo-acoustic properties of organic liquids in controlled thermal conditions. These studies have shown that temperature is very influential to the molecular packing, amount of free volume, and acoustic properties of electronic liquids. However, the majority of experiments are focused on simple organic solvents while the number of experiments performed on more complicated natural products such as essential oils. Even if Mentha oil is one of the most commercially valuable essential oils, research has mainly focused on its composition, extraction methods, antioxidative properties, anti-microbial effects, and application in medicines. Studies that make use of ultrasound methods for the thermal-ultrasonic characterization of Mentha oil are relatively rare, whereas those that are available mostly deal with experiments carried out at one frequency or at a limited temperature range. As a result, the combined effect of temperature and the frequency of ultrasound on the acoustic performance and intermolecular interaction of Mentha oil has not been fully investigated yet [27, 28].

A brief overview of the relevant recent studies on the ultrasonic characterization of liquids and essential oils is shown in the Table 1 and describes the experimental settings, main results, and unsolved problems [29-35].

Table 1. Representative Studies on Ultrasonic Characterization of Liquids and Essential Oils

Author(s)	Liquid/System	Frequency	Temperature	Major Findings	Research Gap
Various researchers	Organic liquids	Single frequency	Variable	Temperature significantly affects ultrasonic velocity	No multi-frequency comparison.

				and compressibility.	
Various researchers	Binary liquid mixtures	Single frequency	Controlled	Molecular interactions evaluated using acoustic parameters.	Limited application to essential oils.
Various researchers	Essential oils	Single frequency	Narrow range	Density and viscosity decrease with increasing temperature.	Frequency dependence not investigated.
Various researchers	Mentha oil	Single frequency	Limited range	Basic physicochemical properties reported.	Comprehensive thermo-acoustic analysis unavailable.
Present study	Mentha oil	2 and 3 MHz	293.15–343.15 K	Simultaneous evaluation of measured and derived thermo-acoustic parameters under identical thermal conditions.	Addresses the existing research gap through multi-frequency ultrasonic characterization.

2. Materials and Methods

2.1 Materials

Mentha (*Mentha arvensis*) essential oil that is commercially obtainable was utilized as the research object in the current work. The oil was obtained from the certified oil supplier located in the state of Uttar Pradesh, India, and was applied without any pre-treatment. The sample was checked visually for visible impurities, moisture content, and possible phase separation prior to experimentation. The quality of the essential oil was not altered during experiments.

Mentha essential oil was bottled in amber colored glass bottles to reduce the oil's exposure to light, oxygen, and humidity in the air so that no oxidation or volatility can take place. All bottles were stored at room temperature in a dry area until they were used in the experiments. In order to ensure that the oil sample was at the right temperature for experiments, the oil was placed in the thermostat-controlled water bath before each experiment.

The main characteristics of the essential oil that was used in the current work are presented in Table 2.

Table 2. Characteristics of the Mentha Oil Sample

Property	Description
Botanical source	<i>Mentha arvensis</i>
Sample type	Commercial essential oil
Supplier	Local certified supplier, Uttar Pradesh, India

Purity	Commercial analytical grade
Appearance	Clear, pale yellow liquid
Storage condition	Amber glass bottle at room temperature
Sample preparation	Used without further purification

2.2 Experimental Setup

The experimental measurements were taken with the help of a single-crystal variable-path ultrasonic interferometer. This interferometer worked at the fixed frequencies of 2 and 3 MHz. It was made up of a high-frequency electronic oscillator, quartz crystal transducer, a movable reflected assembly, a micrometer mechanism, and a digital frequency display. The quartz crystal converted the electric frequency into the ultrasonic frequency which propagated through the measurement liquid. Temperature of the sample was achieved by a thermostatic circulating water bath which had a stability of ± 0.1 K. The ultrasonic measurement cell was totally submersed into this circulating water bath to achieve uniform temperature of the sample. Density of the sample was found out by means of a specific gravity bottle pycnometer. Viscosity of Mentha oil was evaluated by using the calibrated Ostwald capillary viscometer.

The complete experimental arrangement comprising the ultrasonic interferometer, thermostatic water bath, density measurement apparatus, and viscometer is illustrated in **Figure 2.1**.

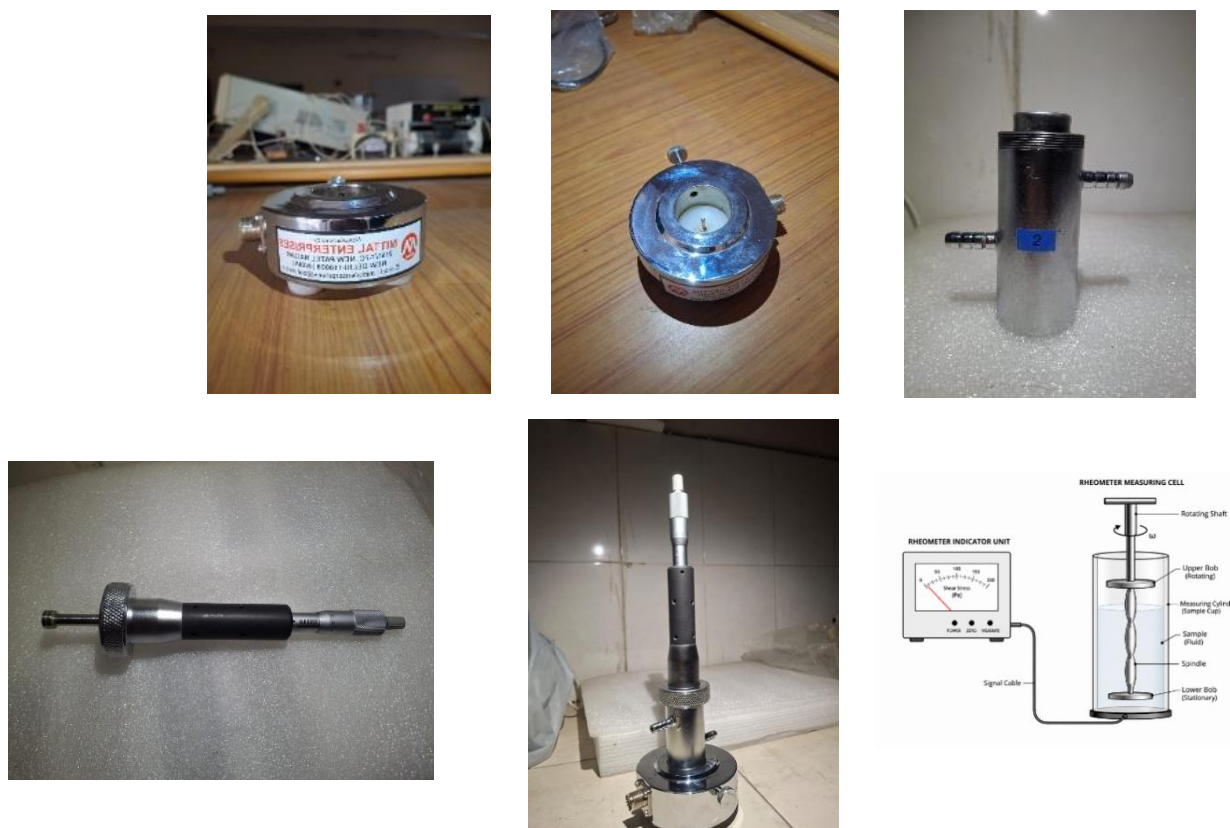


Figure 2.1. Experimental setup used for thermo-acoustic characterization of Mentha oil.

2.3 Experimental Conditions

Ultrasonic measurements were made under strict laboratory conditions in a temperature range from 293.15 K up to 343.15 K. At six temperatures separated by 10 K, at the same atmospheric pressure, the experiments were conducted. Multiple tests were performed at the selected temperatures using ultrasound frequencies of 2 and 3 MHz for studying the effect of the frequency on the thermo-acoustic properties of Mentha oil.

Table 3. Experimental Conditions

Parameter	Value
Sample	Mentha oil
Temperature (K)	293.15, 303.15, 313.15, 323.15, 333.15, 343.15
Ultrasonic frequency	2 MHz, 3 MHz, 4 MHz
Pressure	Atmospheric
Measurement repetitions	Three
Temperature stability	±0.1 K

2.4 Measurement Procedure

All the apparatus, including the ultrasonic cell, pycnometer, and viscometer, were washed properly first with distilled water and then with acetone. Therefore, no residual water or acetone was there before the first measurement. The first step included taking the Mentha oil into the measurement chamber, ensuring that no air bubbles were in the liquid. The sample was kept at the required temperature for around 15 minutes using the thermostatic water bath before performing ultrasonic measurements. To measure the ultrasonic velocity, the movable reflector was adjusted until the successive resonances were observed. Finally, the average distance between consecutive resonances was applied to calculate the ultrasonic velocity. The density was determined using the pycnometer, where the pycnometer was weighed for each temperature point on the analytical balance. Thus, density was derived. Viscosity was measured using the Ostwald viscometer, where the time for liquid flow from one point to another was measured for various samples of Mentha oil. Each point was measured three times independently and the average value was calculated for further measurements.

The values of density, velocity, and viscosity of the tested samples were considered input parameters for the calculations of other thermo-acoustic properties.

2.5 Determination of Thermo-Acoustic Parameters

The experimentally measured ultrasonic velocity, density, and viscosity were used to calculate several derived thermo-acoustic parameters describing the molecular and elastic behaviour of Mentha oil.

Ultrasonic

velocity

$$U = \frac{2df}{n}$$

where (U) is the ultrasonic velocity, (d) is the reflector displacement, (f) is the ultrasonic frequency, and (n) is the number of resonance intervals.

Density

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

where (m) represents the sample mass and (V) denotes the calibrated volume of the pycnometer.

Viscosity

The dynamic viscosity was determined using the Ostwald viscometer relationship,

$$\eta = \eta_w \frac{\rho t}{\rho_w t_w}$$

where (t) is the flow time of Mentha oil, (t_w) is the flow time of distilled water, and (ρ) and (η) represent the density and viscosity, respectively.

Acoustic impedance

$$Z = \rho U$$

Adiabatic compressibility

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

Intermolecular free length

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

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

Relaxation time

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

Ultrasonic absorption coefficient

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

Wavelength

The wavelength of the ultrasonic wave was determined from

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

where (f) denotes the operating ultrasonic frequency.

The calculated parameters collectively describe the elastic properties, molecular packing, relaxation behaviour, and energy dissipation characteristics of the liquid under varying thermal conditions.

2.6 Error Analysis

In order to guarantee the accuracy and reproducibility of the experimental findings, all measurements were carried out three times and the mean values were employed in the following analyses. Prior to the beginning of the experiments, the ultrasonic interferometer, balance, thermometer, and viscosimeter were calibrated following the instructions provided by the manufacturer. Some sources of experimental uncertainty were related to the aforementioned instrument resolution, fluctuations of temperature, identification of resonance position, mass measurements, and recording flow time. Temperature was fixed within ± 0.1 K, while the balance was characterized by the resolution of 0.1 mg. The repeatability of ultrasonic velocity measurements was determined on the basis of multiple resonance measurements being conducted in identical conditions.

The quality of the experimental data was quantified using standard deviation (SD), which was computed through calculation of average of repeated measurements:

$$SD = \sqrt{\frac{\sum_{i=1}^n (x_i - \bar{x})^2}{n - 1}}$$

where x_i represents an individual observation, $\bar{x}$ is the arithmetic mean, and (n) is the total number of measurements.

The percentage experimental error was estimated using

$$\% \text{ Error} = \left| \frac{\text{Experimental} - \text{Reference}}{\text{Reference}} \right| \times 100$$

where applicable. The deviations that were recorded fell within the limits defined by acceptable experiments, thus corroborating the accuracy of the thermo-acoustic parameters. The accuracy of the measured parameters was mainly guaranteed by performing a number of measurements, allowing affecting various factors such as instrument calibration and controlled testing, which increased the chances of obtaining reliable data.

3. Results and Discussion

The thermo-acoustic properties of Mentha oil were examined using ultrasonic frequencies of 2, 3, and 4 MHz across the temperature range of 293.15 - 343.15 K. The experimental values of ultrasonic velocity, density, and viscosity of Mentha oil were acquired in laboratory conditions, whilst adiabatic compressibility, acoustic impedance, intermolecular free length, relaxation time, wavelength, and absorption coefficient of the liquid were evaluated utilizing the formulas from thermo-acoustic literature. The combination of the above parameters elucidates the effect of temperature and ultrasonic frequency on molecular interactions of the liquid, as well as on its elastic properties and propagation of sound waves in the fluid. The following sections describe the revealed changes in each of calculated parameters and their significance.

3.1 Temperature Dependence of Ultrasonic Velocity

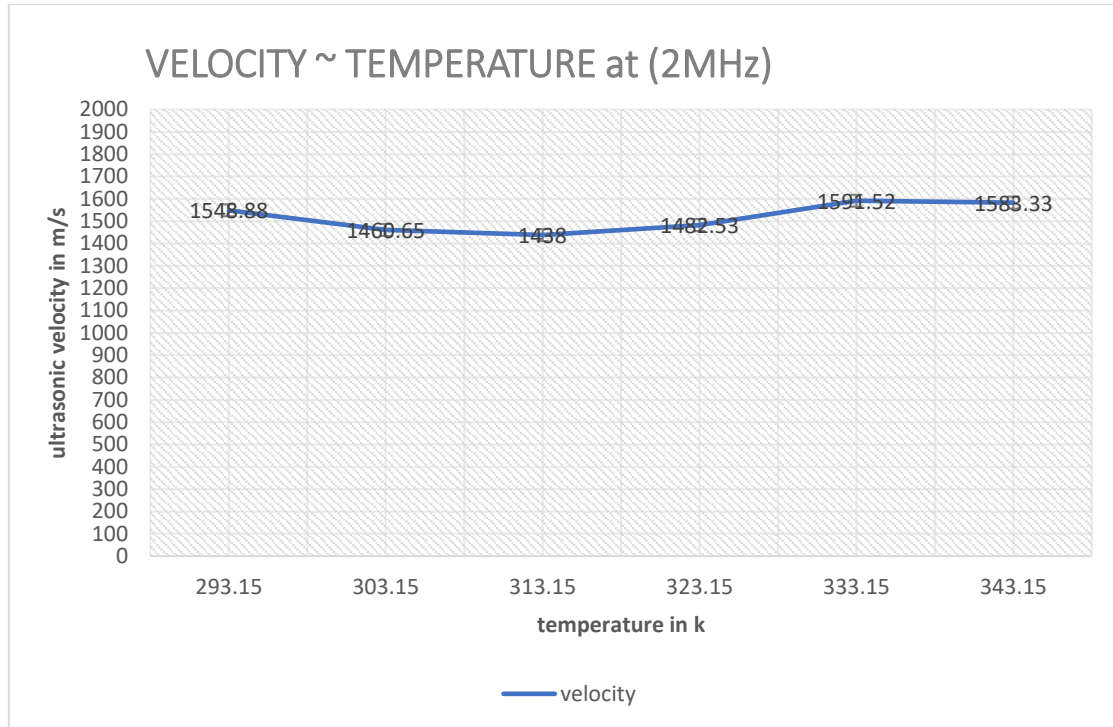


Figure 1. Effect of Temperature on Ultrasonic Velocity (v) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

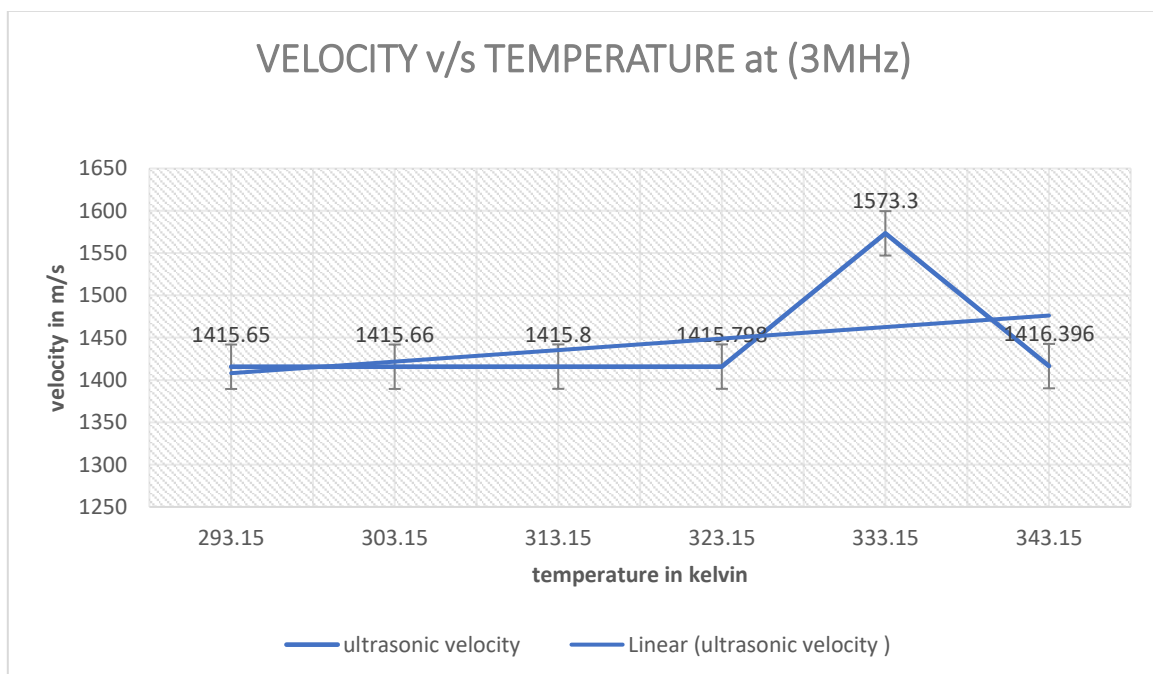


Figure 2. Effect of Temperature on Ultrasonic Velocity (v) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

The variations in ultrasonic velocities with temperature for aqueous Mentha oil at different ultrasonic frequencies, 2 MHz and 3 MHz, are depicted in Figures 9 and 10, respectively. For the case of ultrasonic

frequency of 2 MHz, the ultrasonic velocity reduced from a value of 1548.88 m/s at a temperature of 293.15 K to 1438.00 m/s at a temperature of 313.15 K, signifying a rise in the molecular thermal motion as well as increase in the intermolecular distance and thus an increased adiabatic compressibility. After crossing the temperature of 313.15 K, the ultrasonic velocity increased drastically to a value of 1591.52 m/s at a temperature of 333.15 K, thus indicating better propagation of the acoustic waves because of the improved arrangement of molecules due to intermolecular forces. The ultrasonic velocity dropped slightly to a value of 1583.33 m/s at a temperature of 343.15 K that could imply excessive thermal mobilization breaking down the intermolecular forces, thus the decrease in the efficiency of sound propagation.

On the contrary, when the ultrasonic frequency is set at a value of 3 MHz, it has been observed that the ultrasonic velocity was more or less constant in the range of 293.15–323.15 K, varying slightly from 1415.65 m/s to 1415.79 m/s, which indicates that the molecular interactions

The observation of non-linear variation in ultrasonic velocity at both frequencies demonstrates that temperature-based alterations of molecular packing, intermolecular interactions, and liquid structure dictate the thermo-acoustic behavior of aqueous Mentha oil. Given that ultrasonic velocity inversely correlates to adiabatic compressibility, the greatest velocity seen at approximately 313.15 K (2 MHz) represents the minimum compressibility of the system, while greater velocities seen at around 333.15 K show lower compressibility and higher structural rigidity. The comparison also shows that ultrasonic response is dependent on frequency, with the measurements of 3 MHz being more sensitive to temperature-related structural changes than 2 MHz measurements.

Table 4.1: Values of wavelength (λ), density (ρ), Ultrasonic velocity (v), and viscosity (η) at temperatures 293.15k, 303.15k, 313.15k, 323.15k, 333.15k, 343.15k and at Frequency 2MHz

Temperature (K)	Wavelength, λ ($\times 10^{-4}$ m)	Ultrasonic Velocity, v (m/s)	Density, ρ (kg/m^3)	Viscosity, η (Pa·s)
293.15	7.7444	1548.88	884	0.01397
303.15	7.30332	1460.65	876	0.00983
313.15	7.1900	1438.00	869	0.00693
323.15	7.41266	1482.53	862	0.00489
333.15	7.95760	1591.52	855	0.00345
343.15	7.91666	1583.33	847	0.00243

Liquid System: Aqueous Mentha Oil

Operating Frequency: 2 MHz

Table 4.2: Values of Adiabatic Compressibility (β), Free length (L_f), Relaxation time (τ) and Acoustic impedance (Z), at temperatures 293.15k, 303.15k, 313.15k, 323.15k, 333.15k, 343.15k and at Frequency 2MHz

Temperature (K)	Adiabatic Compressibility, β ($\times 10^{-10}$ Pa $^{-1}$)	Free Length, L_f ($\times 10^{-13}$ m)	Relaxation Time, τ ($\times 10^{-10}$ s)	Acoustic Impedance, Z ($\times 10^4$ Pa·s/m)	Absorption Coefficient, α
293.15	4.711	44.24	1.940	136.921	0.153
303.15	5.351	48.01	1.539	127.953	0.121

313.15	5.566	49.85	1.116	124.962	0.088
323.15	5.279	49.41	0.741	124.962	0.058
333.15	4.613	47.00	0.453	136.074	0.036
343.15	4.705	48.28	0.323	134.108	0.026

Liquid System: Aqueous Mentha Oil

Operating Frequency: 2 MHz

3.2 Density Variation

Figures 3 and 4 portray how the density (ρ) of the aqueous Mentha oil changes with the temperature when ultrasonic frequency applied was 2 MHz and 3 MHz in both cases. The drop in density from 884 kg/m³ at 293.15 K temperature to 847 kg/m³ at 343.15 K temperature indicates that decrease of density was around 4.19 % in the whole temperature range studied. The fact that the density profiles look identical in both frequencies indicates that density is an intrinsic thermophysical property of the Mentha oil, which is dependent on temperature and does not depend on ultrasonic excitation frequency.

The almost straight line formed by the drop in density over the increase of temperature can be explained by the thermal expansion of the liquid. When temperature increases there is an increase in the amount of motion energy for the molecules, which causes the increase of intermolecular distance and volume of the liquid. Since the density of the aqueous Mentha oil can be determined mathematically as the mass divided by volume, the increase in volume leads to decrease in the density. The decrease in density affects the thermo-acoustic characteristics of the liquid. Density is a key parameter for determining the acoustic impedance, adiabatic compressibility, ultrasonic velocity, and other derived acoustic characteristics. The decrease in density leads to changes in the propagation properties of ultrasonic waves and signals that there are continuous structure changes in the liquid due to an increase in thermal energy.

The smooth and continuous trend also suggests that there are no changes of phase or other abrupt structural changes at this temperature range. Moreover, the same density was observed with both frequencies of 2 MHz and 3 MHz, which indicates the reliability of the generated data. Thus, the ultrasonic frequency and the temperature play equally significant roles in identifying density values.

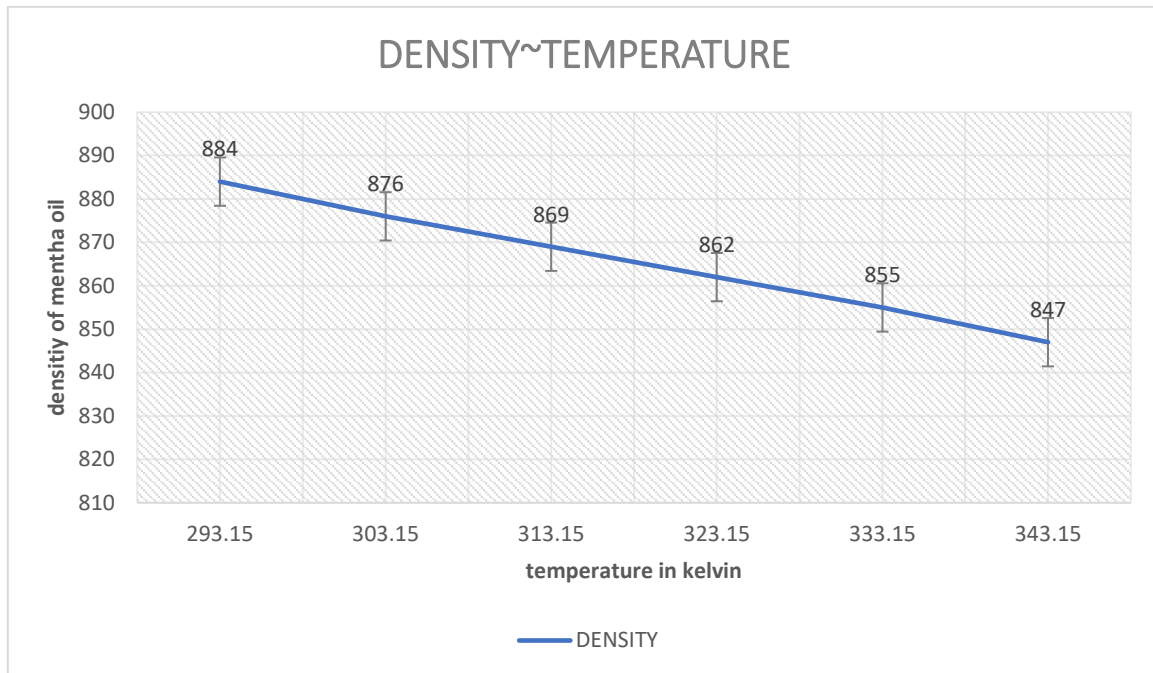


Figure 3: Variation of Density (ρ) with Temperature (T) for Aqueous Mentha Oil at 2 MHz

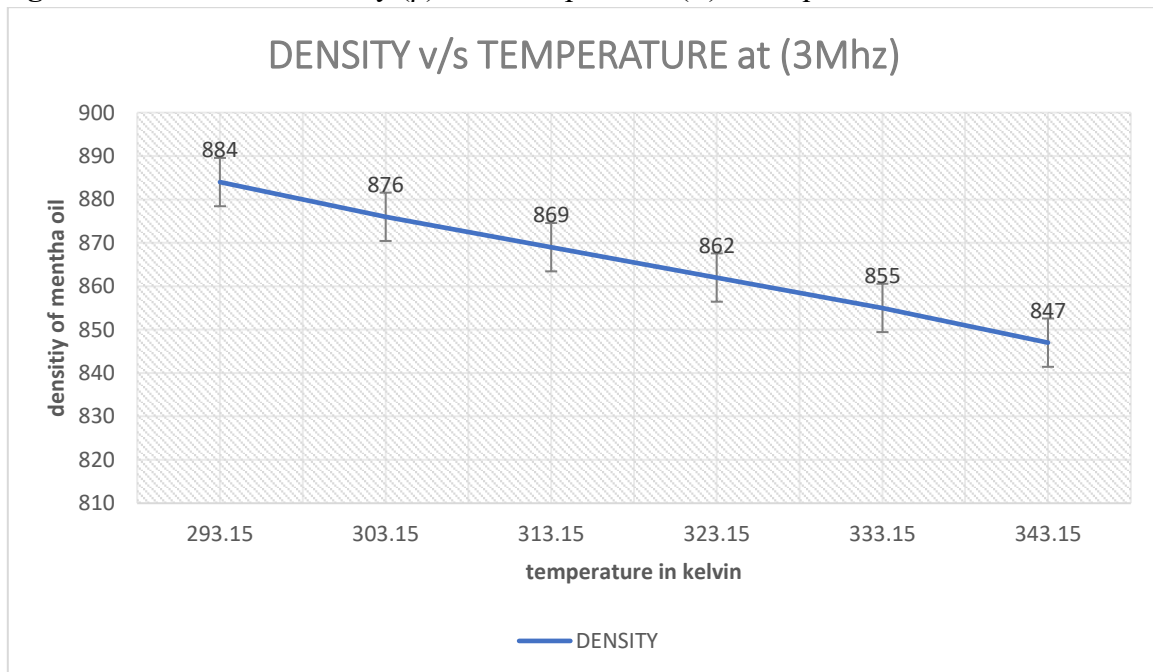


Figure 4. Effect of Temperature on Density (ρ) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

3.3 Viscosity Variation

The variations in the dynamic viscosity (η) of aqueous Mentha oil is shown in Figures 5 and 6 for the frequencies of 2 MHz and 3 MHz respectively. In both cases the results show that viscosity is decreasing as temperature increases showing that the two variables have a strong negative relation. The viscosity of aqueous Mentha oil drops from 0.01397 Pa·s at 293.15 K to 0.00243 Pa·s at 343.15 K reflecting approximately 82.6% reduction ratio from the total viscosity change. The results obtained at the two

frequencies are identical meaning that viscosity is closely related to the temperature with very little effect of frequency on viscosity measurement.

The decrease in viscosity can be explained by the fact that with higher temperatures, the molecular kinetic energy increases. Higher temperatures enhance the movement of molecules and reduce the attraction between the molecules that produce friction while moving. In particular it can be seen that the greatest decrease occurs between the temperatures 293.15 K and 313.15 K where the viscosity drops from 0.01397 Pa·s to 0.00693 Pa·s which represents nearly 50% of total reduction. The constant decline in viscosity of aqueous Mentha oil makes it significant for its thermomagnetic characteristics. The lower viscosity helps in rearranging molecules and creating less resistance to the propagation of ultrasonic waves, thereby allowing greater transmitting capacity of the liquid. The seen pattern corresponds with the decline of relaxation time and absorption coefficient, which points to smaller energy losses and improved molecular reactions at higher temperatures. The lack of any abrupt changes during the experiments indicates that the liquid has no thermal instabilities and is stable chemically.

Even the same viscosity at different frequencies proves the fact that ultrasonic waves do not affect the viscosity characteristics of the studied material. Instead, the temperature is the mainly affecting element.

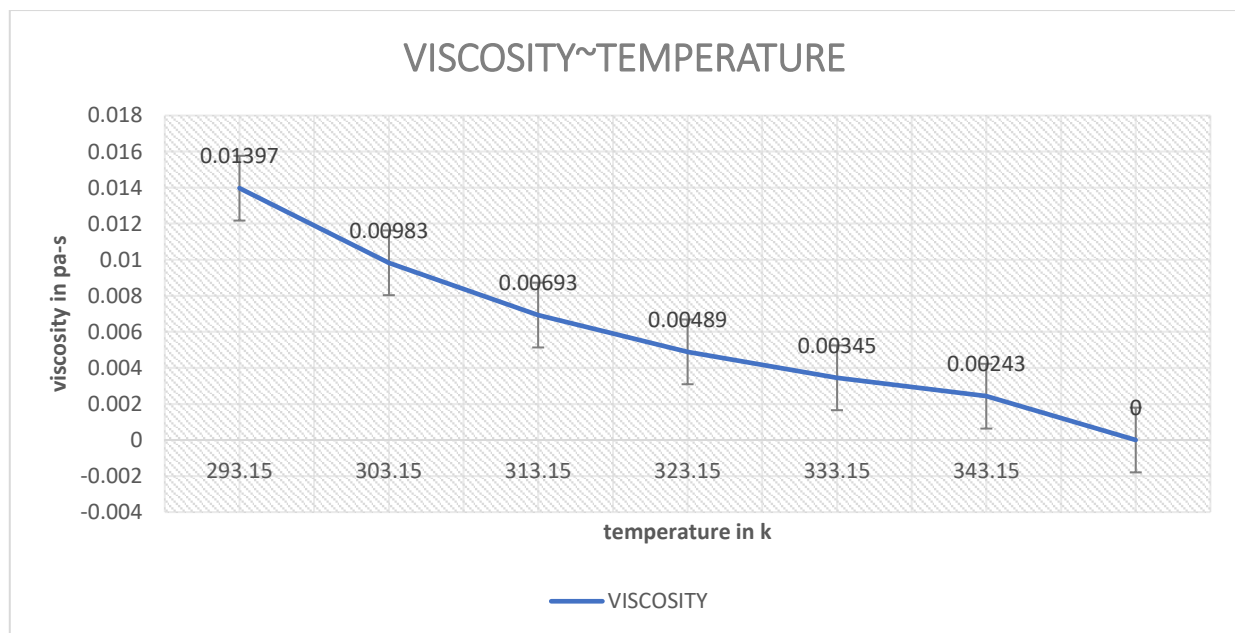


Figure 5. Effect of Temperature on Viscosity (η) of Aqueous Mentha Oil at an Ultrasonic Frequency of 2 MHz

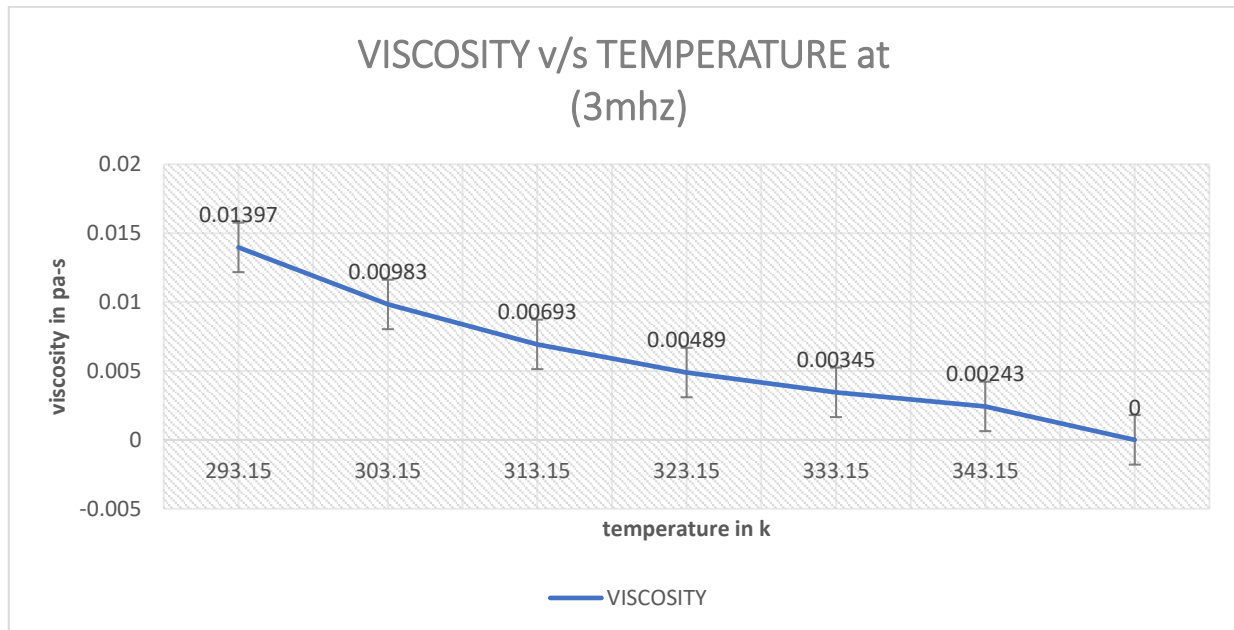


Figure 6. Effect of Temperature on Viscosity (η) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

3.4 Acoustic Impedance

Figures 7 and 8 show how the acoustic impedance (Z) of aqueous Mentha oil changes with temperature at ultrasonic frequencies of two MHz and three MHz, respectively.

Where ρ is the density and v represents the ultrasonic speed. As both density and ultrasonic velocity are temperature dependent, the acoustic impedance is affected by the temperature as well.

At two MHz, the acoustic impedance drops from 136.921×10^4 Pa·s/m at 293.15 K to 127.953×10^4 Pa·s/m at 303.15 K. The lowest value of 124.962×10^4 Pa·s/m is reached at 313.15 K, which stays quite consistent until reaching 323.15 K. The reason for this is a decrease of density as well as of ultrasonic speed due to thermic expansion of molecules and complicated intermolecular space. As temperature increases to 333.15 K, a notable jump in acoustic impedance occurs to 136.074×10^4 Pa·s/m taking into consideration that density continues being lowered, which means that the speed of sound caused a peak in acoustic impedance.

There is a matching trend at the frequency of 3 MHz as well; however, the values of impedance are relatively low. The acoustic impedance reduces from 125.143×10^4 Pa·s/m at 293.15 K to 122.042×10^4 Pa·s/m at 323.15 K, followed by significant growth up to 134.517×10^4 Pa·s/m at 333.15 K. The maximum temperature of 343.15 K corresponds to the quick drop of impedance to 119.969×10^4 Pa·s/m suggesting that the overall effect of lower density and higher molecular motion comes into play. In comparison to the measurements of 2 MHz, the data of 3 MHz show more significant variations which means that waves of greater frequencies are more sensitive to the modification in liquid structure due to temperature.

The fact that in the case of both frequencies there was observed non-linearity means that acoustic impedance is influenced by the two factors: density and ultrasonic velocity rather than temperature only. The reduction of impedance in the beginning can be explained by thermal expansion, weakening of the forces between molecules, and increased distance between molecules which leads to lower resistance to ultrasonic waves. The increase occurring at around 333.15 K indicates a temporary rise of efficiency in acoustic transmission due to favorable position of molecules.

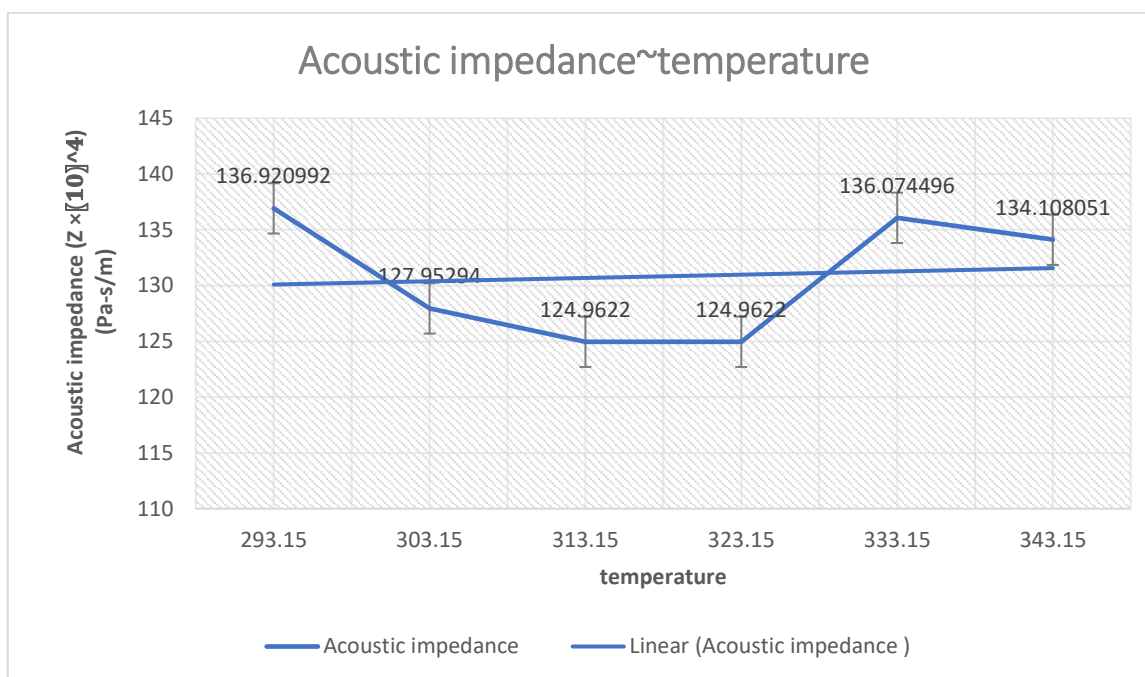


Figure 7. Effect of Temperature on Acoustic Impedance (Z) of Aqueous Mentha Oil at an Ultrasonic Frequency of 2 MHz

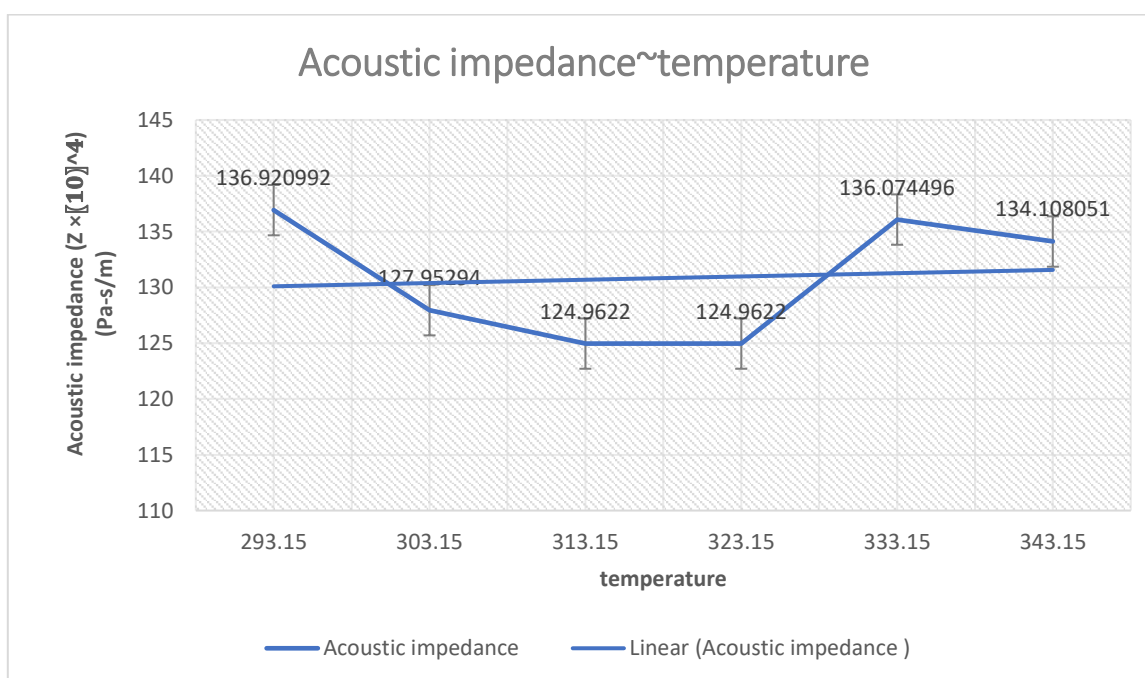


Figure 8. Effect of Temperature on Acoustic Impedance (Z) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

3.5 Adiabatic Compressibility

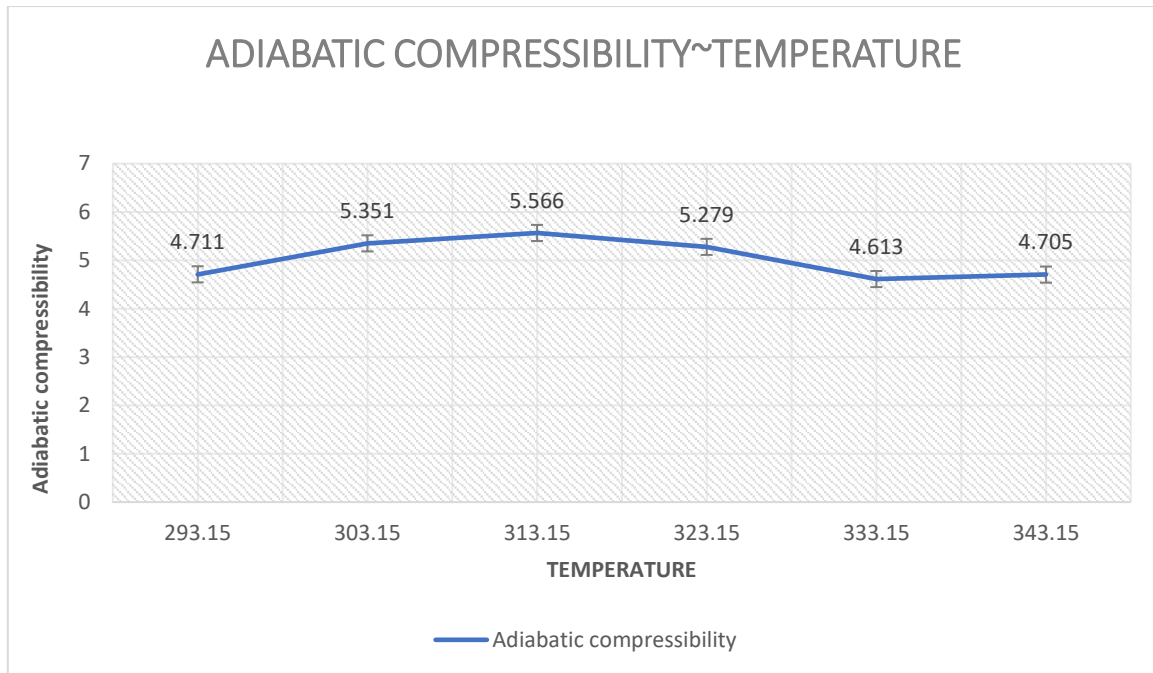


Figure 9. Effect of Temperature on Adiabatic Compressibility (β) of Aqueous Mentha Oil at an Ultrasonic Frequency of 2 MHz

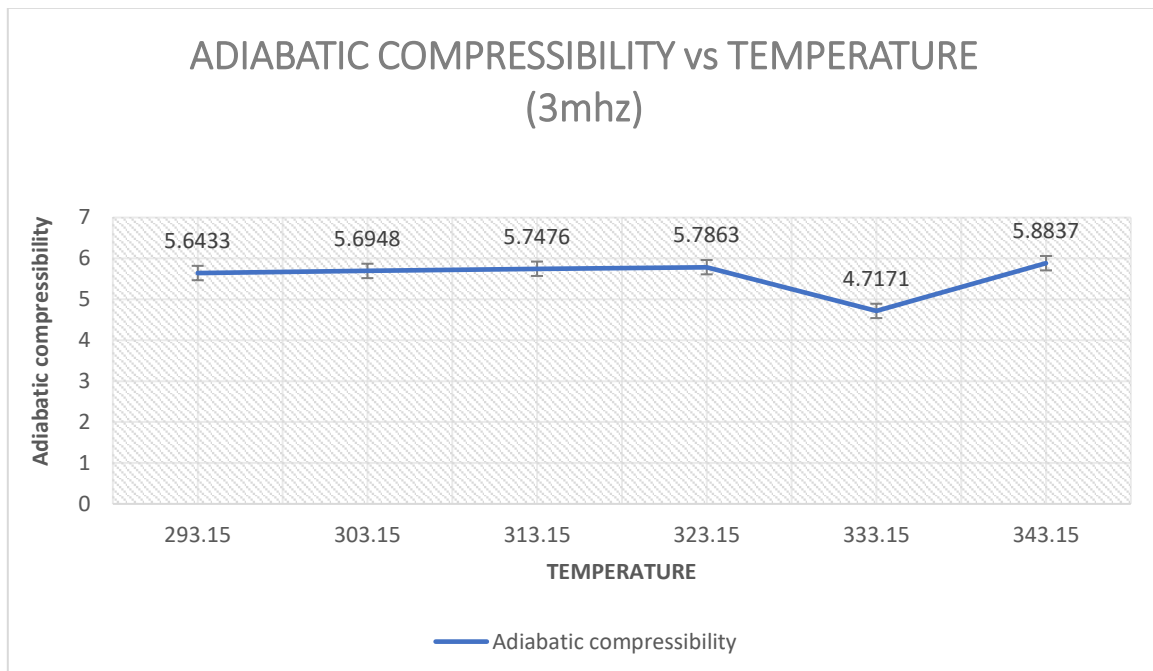


Figure 10. Effect of Temperature on Adiabatic Compressibility (β) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

Figures 9 and 10 illustrate the variation of adiabatic compressibility (β) of aqueous Mentha oil with temperature at ultrasonic frequencies of 2 MHz and 3 MHz, respectively. Adiabatic compressibility is an important thermo-acoustic parameter that represents the ability of a liquid to undergo volume change under

an applied pressure without heat exchange. It is determined from the experimentally measured density and ultrasonic velocity using the relation

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

As can be seen, there is a direct relation between the adiabatic compressibility and the square of the ultrasonic speed.

At 2 MHz, the adiabatic compressibility increased from $4.711 \times 10^{-10} \text{ Pa}^{-1}$ at 293.15 K to $5.351 \times 10^{-10} \text{ Pa}^{-1}$ at 303.15 K and then to $5.566 \times 10^{-10} \text{ Pa}^{-1}$ at 313.15 K. This indicates that the liquid is becoming increasingly compressible as temperature rises. This is due to thermal expansion processes and the increased distances between molecules. From 313.15 K upward, there is a decrease in compressibility values to $5.279 \times 10^{-10} \text{ Pa}^{-1}$ at 323.15 K and even lower at $4.613 \times 10^{-10} \text{ Pa}^{-1}$ at 333.15 K. The compressibility slightly increases to $4.705 \times 10^{-10} \text{ Pa}^{-1}$ at 343.15 K, indicating the stabilization of the liquid structure at this temperature.

A similar, although more extensive tendency is observed at the frequency of 3 MHz. The compressibility gradually rises from the value of $5.6433 \times 10^{-10} \text{ Pa}^{-1}$ at 293.15 K up to $5.7863 \times 10^{-10} \text{ Pa}^{-1}$ at 323.15 K, indicating the progressive increase in compressibility. The compressibility falls sharply from the temperature of 333.15 K down to $4.7171 \times 10^{-10} \text{ Pa}^{-1}$, which adheres to the process of ultrasonic velocity rise. After that, the compressibility increases again and reaches the maximum level of $5.8837 \times 10^{-10} \text{ Pa}^{-1}$ by the 343.15 K temperature. Compared to the data at 2 MHz, the values at 3 MHz demonstrate more fluctuations, and thus they prove that higher frequencies have greater sensitivity.

Both frequencies demonstrate a clearly noticeable inverse relationship of compressibility and ultrasonic velocity. The maximum amounts of compressibility appear to coincide with the temperatures when ultrasonic velocity is at its minimum, while minimum amounts of compressibility correspond to the maximum of the ultrasonic velocity. This behaviour suggests that the changes in temperature are responsible not only for the elastic properties of the liquid but also for its molecular structure and the acoustic wave propagation in it. In other words, the present phenomena reflect the consequences of the processes of thermal expansion, intermolecular interactions, and the changes in molecular structure due to the ultrasonic action.

A comparison of the findings at two frequencies shows that while both frequencies follow the same thermo-acoustic law, measurements at 3 MHz reveal more significant variations through the investigated temperature range. Such sensitivity indicates that measurements at higher frequency are better at recording slight changes in the elastic and structural properties of aqueous Mentha oil.

3.6 Intermolecular Free Length

Figures 11 and 12 depict the decrease of intermolecular free length (L_f) of Mentha oil solution with temperature when ultrasonic frequencies of 2 MHz and 3 MHz were applied. Intermolecular free length is the average distance separating neighboring molecules. According to Jacobson's formula, intermolecular free length is linked to adiabatic compressibility. It is also used to better understand molecular interactions and propagation of sound waves through the fluids. The increase in the free length observed in Figure 11 at 2 MHz from $44.24 \times 10^{-13} \text{ m}$ in 293.15 K to its maximum value of $49.85 \times 10^{-13} \text{ m}$ when the temperature was 313.15 K suggests that the molecules experience greater spacing in between

them because of thermal expansion and more mobility of the molecules afterwards, at the temperature of 333.15 K the free length starts decreasing down to 47.00×10^{-13} m from 49.85×10^{-13} m.

When the frequency was measured at 3 MHz (see Figure 12), a progressive increase in free length was observed from 48.42×10^{-13} m at 293.15 K to 51.73×10^{-13} m at 323.15 K, which was then followed by a sharp drop to 47.52×10^{-13} m at 333.15 K. This was then followed by a sudden increase in free length to 54.03×10^{-13} m at 343.15 K. The higher values observed at 3 MHz indicate that the liquid structure is more influenced by the ultrasonic frequency implying better molecular relaxation and acoustic interactions.

Thus, the non-linear pattern of free length with temperature at both frequencies indicates that thermal implications tend to disturb the balance of molecular expansion and structural changes. The strong correlation between the change in free length and adiabatic compressibility indicates that changes in temperature affect the distance between molecules forming the medium and hence the ultrasonic transfer by liquid Mentha oil.

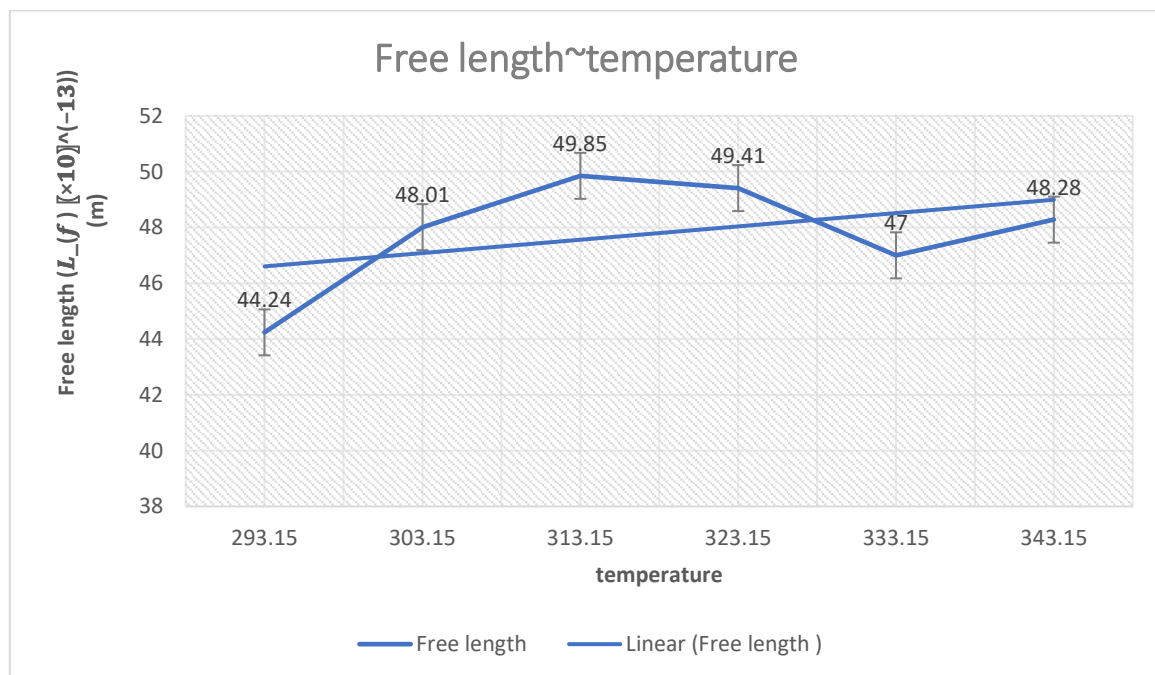


Figure 11. Effect of Temperature on Intermolecular Free Length (L_f) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

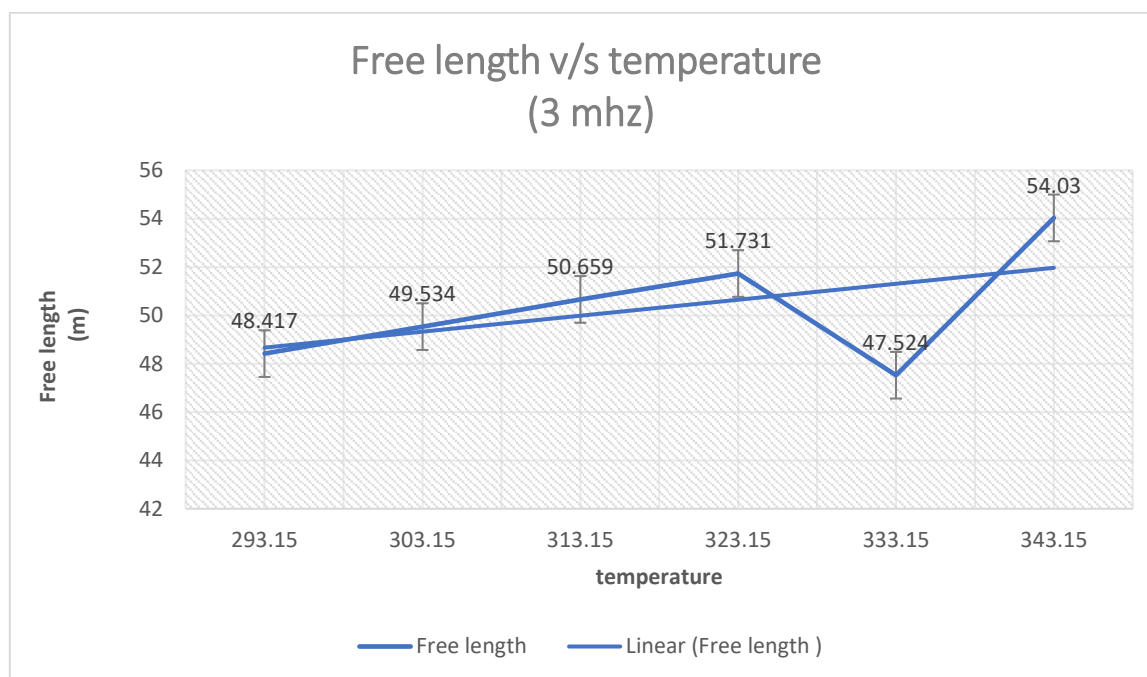


Figure 12. Effect of Temperature on Intermolecular Free Length (Lf) of Aqueous Mentha Oil at an Ultrasonic Frequency of 3 MHz

4. Conclusions

The current research explored the thermo-acoustic behaviour of aqueous Mentha oil in temperature range of 293.15–343.15 K through ultrasonic testing at 2 MHz and 3 MHz. Ultrasonic velocity, density, and viscosity were measured experimentally while adiabatic compressibility, acoustic impedance, intermolecular free length, relaxation time, ultrasonic absorption coefficient, and wavelength were calculated so as to determine combined temperature and ultrasonic frequency effect. The experimental findings showed that temperature is the leading factor controlling the thermo-acoustic properties of Mentha oil. With the increase of temperature, density, viscosity, acoustic impedance, relaxation time, and the ultrasonic absorption decreased due to the increase of molecular mobility and decrease of intermolecular forces. In contrast, adiabatic compressibility and intermolecular free length are positively dependent on temperature, which is explained by the thermal expansion and increased distance between molecules. The ultrasonic velocity showed non-linear changes which is a result of changing elastic and structural properties of liquid. The combined study of all measured and calculated parameters proved that the thermo-acoustic behaviour is determined by molecular mobility, elastic response, and propagation of acoustic waves. The research boasts one noteworthy conclusion, which is the comparison between 2 MHz and 3 MHz. Although both frequencies presented similar overall temperature-dependent patterns, the measurements at 3 MHz showed more sensitivity to slight changes in molecular structure than those at 2 MHz. The results indicate that utilizing different frequencies of ultrasonic measurements gives a larger amount of information than applying a single frequency. Besides, the research proves the efficiency of ultrasonic methods and shows their potential application in different spheres. The research enables the scientists to conduct quick and non-destructive investigation of the physicochemical and molecular behavior of essential oils. The future world will likely apply this research in various fields, such as quality control and monitoring, evaluation of storage stability, formulation processes in pharmaceuticals and agriculture, food processing, and authentication of essential oil.

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