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Temperature Dependence of Bulk Viscosity in Edible Oils using Acoustic Spectroscopy

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Abstract

When ultrasound waves are applied to a compressible Newtonian fluid, bulk viscosity plays an important parameter to cause attenuation. Ultrasound spectroscopy is an important technique to characterise and determine the physico-chemical properties of many food components because it is a non-invasive, non-destructive, easy and accurate technique. The aim of this study was to find the bulk viscosity of three brands of sunflower and extra-virgin olive oil by using the Navier's-Stoke equation across a temperature range of 5°C to 40°C and to test the hypothesis that there is a significant difference in the value of bulk viscosity between the different brands of sunflower and olive oil used. The value of bulk viscosity was not found to be constant over the operating frequency range of 12-100 MHz, which suggested edible oils are non-Newtonian fluids. Also, no significant statistical difference of bulk viscosity values was found between different brands of the same oil ($p \geq 0.05$). This shows bulk viscosity is not affected by small compositional variations. Acoustic spectroscopy is increasingly being used to characterise food materials. More studies on bulk viscosity must be employed in order to be able to utilise this technology to its full strength.

Keywords: Bulk viscosity; Acoustic spectroscopy; Attenuation

Introduction

Edible oils occupy an important position in the human diet because of its nutritive value and also because of its organoleptic and rheological properties [1]. Acoustic spectroscopy is increasingly being used to characterise oils and fats due to its many advantages. Ultrasound waves are longitudinal sound waves of frequency of 20 KHz or more [2]. Ultrasonic analysis is a useful technique to characterize and determine many physico-chemical properties of oil and other food component mainly because it is a non-invasive, non-destructive, easy and accurate technique. It can be used 'on-line or off-line' and also works for opaque food objects where characterising food on visual methods can be difficult [1].

Compressible fluids are fluids whose density changes when high-pressure gradient is applied. When force is applied to these fluids, they flow in the form of transverse-pressure waves. The velocity of propagation of these pressure waves in compressible fluid is known as velocity of sound [3]. Newtonian fluids are ones whose shear stress is proportional to shear strain. Compressible Newtonian liquids exhibit two types of viscosity: shear and bulk viscosity. Shear viscosity is the resistance to the change in shape under shear stress and the bulk or volume viscosity is the resistance to change in volume under an applied pressure [4]. Bulk viscosity is also termed as volume viscosity, second viscosity coefficient, expansion coefficient of viscosity and coefficient of bulk viscosity. Bulk viscosity is significant if the compression or expansion in the fluids proceeds so rapidly that it takes longer time than the duration of change in volume to restore the thermodynamic equilibrium like with the absorption or dispersion of sound waves [5]. The experimental values obtained for ultrasonic absorption is often found to be much larger than the values obtained from classical Stroke's equation where only shear viscosity is considered [4]. This increase in the absorption can be attributed to the bulk viscosity and thermal conduction. Fluid molecules have translation, rotational and vibrational degrees of freedom. The translational motion is due to the dynamic viscosity and the rotational and vibrational motion is due to bulk viscosity. Therefore, to know the effect of vibrational and rotational energy on fluids obtaining the bulk viscosity data is very important [5,6]. There is also a certain bulk viscosity effect in fluid flow for fluids with large Reynold's number when the ratio of bulk to shear viscosity is of the order of the square root of Reynold's number [7]. However, given the importance of bulk viscosity little research has been conducted to characterise it and for many fluids it is unknown or inaccurately known particularly across different temperatures. Bulk viscosity is observed when sound particularly ultrasonic waves travels through fluids. Hence, ultrasound waves can also be used to measure the bulk viscosity of fluid.

In a study by Dukhin and Goetz [6], three methods were used to find the bulk viscosity: Brillouin spectroscopy, laser gradient spectroscopy and acoustic spectroscopy. It was found that the acoustic spectroscopy gave the most precise results for bulk viscosity; as with Brillouin spectroscopy there were 'high errors due to the difficulty in measuring the Brillouin linewidth' and with laser gradient spectroscopy complications arrived due to the fitting of the laser gradient with five adjustable parameters. Acoustic spectroscopy gives value for the speed of sound which can be used to measure compressibility and it is also the only method where multi-frequency measurements, in the range of 1-100 MHz can be taken. This is important to find the nature of the fluid, if it's a Newtonian or non-Newtonian fluid. If the fluid is Newtonian than the calculated bulk viscosity will be independent of the frequency changes [6].

The Navier-Stokes equation is important to study physical fluid dynamics. The general Navier-Stokes equation is written as:

$$\frac{\partial v}{\partial t}+(\mathbf{v}\nabla)\mathbf{v}=-\frac{1}{\rho}\nabla P+\eta\nabla^{2}\mathbf{v}+\frac{1}{\rho}\mathrm{F} \quad (1)$$

Where ρ ($kg.m^{-3}$) is the density, t (s) is time, **v** is the velocity vector,

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P (Pa) is pressure, η (Pa.s) is shear viscosity and *F* (N) is the body force term as such forces act on the volume of a fluid particle.

Bulk viscosity is an important term in the Navier-Stokes equation for a Newtonian compressible liquid [6].

$$\rho\left[\frac{\partial v}{\partial t}+\left(\mathbf{v}\nabla\right)\mathbf{v}\right]=-grad\ P+\eta\Delta\mathbf{v}+\left(\mu+\frac{4}{3}\eta\right)grad\ div\ \mathbf{v} \quad (2)$$

μ (Pa.s) is the bulk viscosity. For an incompressible liquid, the last term on the right hand side may be neglected as this term accounts for compressibility.

grad div **v**=0

Thus, the bulk viscosity term has no contribution for incompressible fluids. Therefore, for incompressible fluids the Navier-Stokes equation can be written as:

$$\rho\left[\frac{\partial v}{\partial t}+\left(\mathbf{v}\nabla\right)\mathbf{v}\right]=-grad\ P+\eta\Delta\mathbf{v} \quad (3)$$

Thus the effect of bulk viscosity is not very significant for incompressible fluids and for ideal monoatomic gas for which μ=0 [5].

When a wave propagates through a viscous and thermally non-conductive fluid then the general solution obtained from the Navier-Stokes equation with respect to attenuation is [6]:

$$2\left(\frac{\alpha_{long}v}{\omega}\right)^2=\frac{1}{\sqrt{1+t^2\omega^2}}-\frac{1}{\sqrt{1-t^2\omega^2}} \quad (4)$$

Where α_{long} (Np.m^{-1}) is ultrasound attenuation coefficient, v (ms^{-1}) is the velocity of sound, ω is the ultrasound frequency, t (s) is the viscous relaxation time and takes into account both bulk and shear viscosity and is given by:

$$t=\frac{1}{\rho v^2}\left(\frac{4}{3}\eta+\mu\right) \quad (5)$$

If attenuation is plotted as a function of frequency a normal distribution curve is obtained and at the critical frequency the maximum value obtained is approximately equal to the viscous relaxation time. The critical frequency is around 1000 GHz around, this high ultrasound range is difficult to achieve in real instruments but for low frequency this is achievable. The low frequency asymptotic function is given by [8]:

$$\alpha_{long}=\frac{\eta\omega^2}{2\rho v^3}\left[\frac{4}{3}+\frac{\mu}{\eta}+\frac{(\gamma-1)\tau}{\eta C_p}\right] \quad (6)$$

Where γ is the ratio of specific heats, τ (w.m^{-1}.K^{-1}) is the thermal conductivity and C_p (J.K^{-1}) is the specific heat at constant pressure, v (m.s^{-1}) is velocity, T(°C) is temperature, β (K^{-1}) is the bulk compressibility and ω is the angular frequency $\omega=2\pi$f, f (Hz) is the frequency of acoustic wave and i is the imaginary number. From equation (6) can be expressed as equation (7) to calculate bulk viscosity.

$$\mu=\frac{2\alpha\rho v^3}{\omega^2}-\frac{4\eta}{3}-\frac{(\gamma-1)\tau}{C_p} \quad (7)$$

α (Np.m) is the attenuation coefficient. The contribution of thermal conduction to bulk viscosity is dependent on (γ-1) · For liquids the ratio of specific heats is close to one and for gases it is greater than one as liquids are less compressible as compared to gases. Therefore, there is not much contribution to the bulk viscosity from the thermal properties of the material and the thermal term can be neglected [6].

Hence equation (7) can be re-written as:

$$\mu=\frac{2\alpha\rho v^3}{\omega^2}-\frac{4\eta}{3} \quad (8)$$

The temperature dependence of physical parameters of edible oils is demonstrated by the following model equations [9]:

$$c=c_0+c_1T \quad (9)$$

$$\rho=\rho_o+\rho_1T \quad (10)$$

$$\eta=\eta_o\exp\left[-\frac{\eta_1}{k\left(T+273.13\right)}\right] \quad (11)$$

Where c (m.s^{-1}) is velocity, ρ (kg.m^{-3}) is density, η (kj.mol^{-1}) is viscosity, k is Boltzmann, T is temperature (°C). The subscripted terms are constants. For sunflower oil, the values of the constants have been reported as: ρ_0 (kg.m^{-3}) is 933.76, ρ_1 (kg.m^{-3}, °C^{-1}) is -0.61 for 20 to 80°C; ln η_o is -13.83, η_1 (kJ.mol^{-1}) is 27.17 for 25°C to 50°C; c_0 (m.s^{-1}) is 1538 and c_1 (m.s^{-1}, °C^{-1}) is -3.28 for 5°C to 70°C. For olive oil c_0 (m.s^{-1}) is 1528.9 and c_1 (m.s^{-1}.°C^{-1}) is -3.23 for 20°C to 70°C. All the three parameters velocity, density and shear viscosity are temperature dependent (equations 9-11) and plays an important role in finding bulk viscosity. Also, Coupland and McClements [9] emphasised that these bulk properties of oils depend upon their chemical composition. Therefore, there is expected to be a dependence of bulk viscosity on temperature for edible oils which will be discussed in this work (Figure 1).

The aim of this work was to calculate the bulk viscosity of sunflower oil and extra-virgin olive oil across a temperature range of 5°C to 40°C and to test the hypothesis that there is a significant difference in the value of bulk viscosity between the different brands of sunflower (Tesco, Morrisons, Floras) and extra-virgin olive (Tesco, Morrisons, Sierra mágina) oil. Even for the same type of oil there are differences in the physico-chemical properties as the composition of food oil varies significantly depending on the geographical source, processing parameters (like distillation), storage time (as crystallisation or oxidation might take place) [9]. The length of fatty acid chain has an effect on the viscosity of the oil [10]. Hence, three different brands were investigated to find if these differences have any significant effect on bulk viscosity. The experimental procedure of conducting the study is mentioned in the next section. The results obtained from this work is illustrated in the Results and Discussion part. First, the justification of using the frequency squared equation (8) for finding the bulk viscosity was given using the graph of log attenuation v/s log frequency (Figure 2). Second, all the bulk properties like velocity, density, shear viscosity and bulk viscosity was tabulated in Table 1 and the dependence of bulk viscosity on the frequency was studied (Figures 3 and 4). Third,

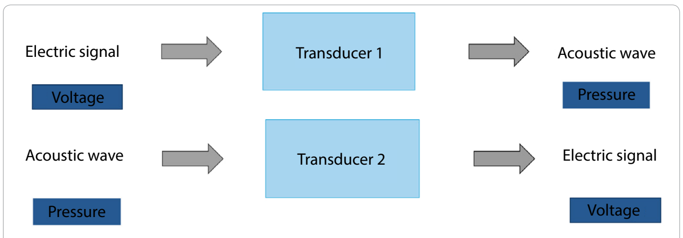

Figure 1: The first type of transducer emits ultrasound wave on application of electrical voltage which is picked up by the sample. The second type of transducer detects the ultrasound wave emitted by the transducer and converts it into electrical voltage which is measured as attenuation.

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a comparative study between the bulk and shear viscosity was made (Table 2 and Figure 5). Fourth, the hypothesis was tested to find out if

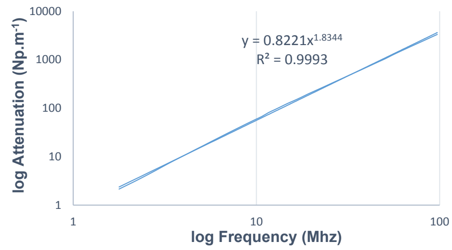

Figure 2: Plot of log Attenuation v/s log Frequency of Tesco sunflower oil at 25°C showing a best fit polynomial.

Temperature	Density	Velocity	Shear viscosity	Bulk viscosity		
T (°C)	ρ (kg m⁻³)	v (m s⁻¹)	η × 10⁻² Pa.s	μ × 10⁻² Pa.s		CV
				Mean*	SD	
6	928.71	1515.69	5.22	5.79	1.89	0.33
10	926.01	1501.93	4.75	4.51	1.59	0.35
15	922.57	1485.51	4.15	3.57	1.43	0.22
20	919.15	1469.02	3.55	2.91	1.21	0.41
25	915.74	1452.04	2.37	3.33	0.99	0.3
30	912.35	1435.36	2.1	2.77	0.86	0.31
35	908.97	1419.2	2.06	2.07	0.71	0.2
40	905.6	1403.98	1.42	2.36	0.6	0.25

Standard Deviation (SD)
Coefficient of Variation (CV)
*Mean was taken for the bulk viscosity in the frequency range of 12 MHz-100MHz
Ultrasound velocity readings was obtained from the Ultrasizer. Density was measured by Anton Paar DMA 4500 M density-meter and shear viscosity by Anton Paar MCR 302 rheometer.

Table 1: Density, velocity, shear viscosity and mean bulk viscosity of tesco sunflower oil at the selected temperatures.

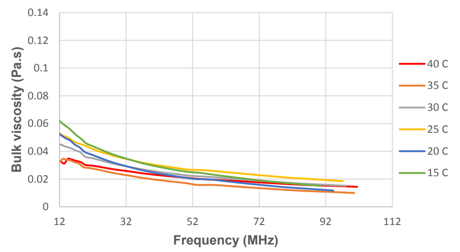

Figure 3: Plot of bulk viscosity v/s frequency of Tesco sunflower oil at each selected temperature. A decrease in the value of bulk viscosity is seen with the increase in temperature, showing frequency dependence and possible non-Newtonian behaviour.

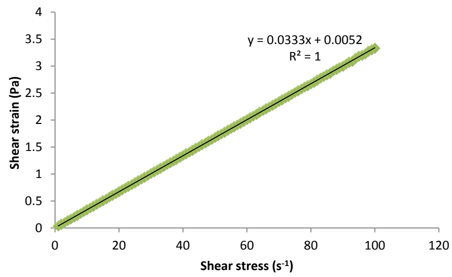

Figure 4: Plot of shear stress v/s shear strain, giving a linear relationship between stress and strain suggesting Newtonian behaviour of edible oils.

Temperature	Bulk viscosity		Shear viscosity	
T°C	μ × 10⁻² Pa.s		η × 10⁻² Pa.s	Ratio ($\frac{\mu}{\eta}$)
	Mean	SD		
6	5.79	1.89	1.89	1.11
10	4.51	1.59	1.59	0.95
15	3.57	1.43	1.43	0.86
20	2.91	1.21	1.21	0.82
25	3.33	0.99	0.99	1.41
30	2.77	0.86	0.86	1.32
35	2.07	0.71	0.71	1
40	2.36	0.6	0.6	1.66

Table 2: The bulk and shear viscosity values of Tesco sunflower oil at the selected temperatures and the ratio between bulk viscosity to shear viscosity.

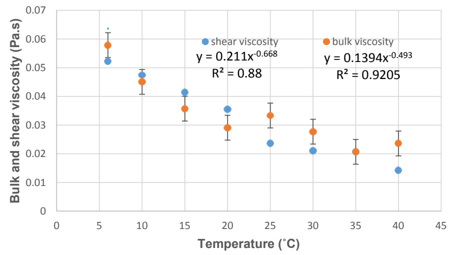

Figure 5: Plot of bulk and shear viscosity v/s temperature of Tesco sunflower oil showing a decrease in the values with the increase in the temperature.

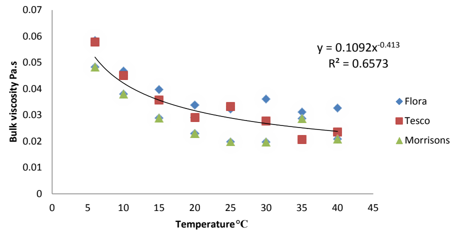

Figure 6: A plot of bulk viscosity v/s temperature of all the three brands (Tesco, Morrisons, Flora) of sunflower oil. As there is no significant difference between the brands an averaged polynomial best fit is obtained from this plot to know the temperature dependence of bulk viscosity for sunflower oil.

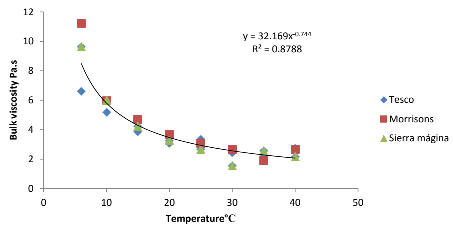

Figure 7: A plot of bulk viscosity v/s temperature of all the three brands (Tesco, Morrisons, Sierra magina) of extra-virgin olive oil. As there is no significant difference between the brands an averaged polynomial best fit is obtained from this plot to know the temperature dependence of bulk viscosity for extra-virgin olive oil.

there is a significant difference in the bulk viscosity values between the different brands of edible oils. Lastly, the temperature dependence of bulk viscosity was investigated (Figures 6 and 7).

Materials and Methods

Three brands of sunflower oil and three brands of extra-virgin olive oil have been used to evaluate potential variance of bulk viscosity with different brands of the same oil. The samples used for sunflower oil were Morrisons sunflower oil, Tesco sunflower oil and Floras sunflower oil. The samples used for olive oil was Morrisons extra virgin olive oil, Tesco extra virgin olive oil and Sierra mágina extra virgin olive oil. All the samples were locally purchased from Leeds supermarkets during July, 2015. Ultrasonic waves have been employed in this work to find the bulk viscosity. The attenuation coefficient, velocity of the sound wave after passing through the material across a range of frequencies is obtained from Ultrasizer MSV by Malvern Ltd. Density of the samples was measured by Anton Paar DMA 4500 M Density-meter. Anton Paar MCR 302 (Modular Compact Rheometer) was used to find the shear viscosity of the samples. All these parameters were measured to calculate the bulk viscosity using equation (8).

Ultrasizer MSV by Malvern Ltd was used to determine attenuation coefficient and velocity of sound as it passes through the oil samples at different selected temperatures. This is an acoustic spectroscopy instrument for liquids and emulsions operating in the frequency range of 1-100 MHz. Transducers are devices that convert energy from one form to other. This device makes use of two such transducers where one emits ultrasound waves on the application of voltage into the sample while the other detects it and converts into the corresponding voltage. Two pairs of such transducers are used. One pair operates in the low frequency range and the other in the high frequency range. On each run 50 measurements are taken covering the frequency range of 1 to 100 MHz. As sound waves travels through a medium attenuation is caused due to dissipation of energy in the form of shear viscosity, bulk viscosity, thermal conductivity and molecular relaxations. Ultrasizer measures this attenuation. This instrument needs 500 ml of sample to measure which is a measure drawback for limited sample volumes. It is connected to an external Huber Ministat temperature control unit which operates in the range of 5°C to 50°C. While conducting the experiment a stirrer constantly agitated the sample in order to reduce the thermal variation in the bulk sample, the speed of the stirrer can be adjusted. Care was taken so that no air bubbles were formed as these bubbles causes excess attenuation [11]. 10 repeat measurements were taken at each selected frequency and the mean was calculated to take into consideration any uncertainties or variations due to measurement.

The density measurements for the samples were accomplished by using Anton Paar DMA 4500 M Density-meter across a temperature range of 5°C to 40°C. The measurements by this instrument is based on the oscillating U-tube method. The thermal control is provided by two integrated Pt100 platinum thermometers together with Peltier elements. Viscosity related errors are automatically corrected over the full range of sample viscosities by measuring the damping effect of the viscous sample followed by a mathematical correction of the density value. Error while measuring the shear viscosity may arise due to 'sample under filling, uncertainties in the gap size, viscous heating effects, wall-slip errors, edge failure and radial migration.'

Anton Paar MCR 302 was used to take the shear viscosity readings. This rheometer is driven by air bearing supported EC (Electrically Commutated) motor technology. This ensures accuracy over a wide viscosity range. It is a digital instrument using digital signal processing technology. It makes use of patented normal force sensor. It also makes use of some patented features for convenience and to increase the efficiency. The measuring system used for our measurements was CP50-2 (Conical plate with diameter 50 mm and angle 2°). The temperature is controlled by a water bath. The shear viscosity values from 25°C to 40°C were measured and values outside the measurement range were extrapolated.

Thermal properties of the materials to be utilised in calculating the bulk viscosity were obtained from literature [6]. The ratio of specific heats, $\gamma=C_p/C_V$ was found to be almost equal to unity. Hence, the term due to the thermal property term in equation (9) was neglected. The bulk viscosity value was calculated from equation (10) using the values of mean attenuation, density, shear viscosity and the velocity of ultrasound as measured in experimental procedures.

Statistical analysis

The mean, standard deviation and coefficient of variance of the bulk viscosity were calculated at each temperature of the selected temperature range. T-test of two samples assuming equal variances between each brand and also single factor Annova was performed for both sunflower and olive oil to evaluate the significant difference in the value of bulk viscosity between different brands of sunflower oil and olive oil. Microsoft Excel (XLS) was used for the statistical analysis performed.

Results

Tesco sunflower oil has been taken as an example to show all the calculations and graphs. The experimental calculations for the other samples have been provided in the appendix for clarity and brevity.

A graph of log attenuation v/s log frequency has been plotted as shown in Figure 2 and the best fit polynomial was obtained in the frequency of the form f^{δ}, (where δ is the exponent). δ is found to be almost equal to 2 (δ=1.8344). A linear relationship between log attenuation and log frequency was seen (as regression coefficient > 0.999) for all the samples at each temperature.

The bulk parameters (density, shear viscosity, velocity) measured and the bulk viscosity calculated for Tesco sunflower oil across a temperature range of 6°C to 40°C is summarised in Table 1. The mean bulk viscosity and standard deviation has been calculated across the range of 12 MHz-100 MHz. A graph of bulk viscosity against frequency was plotted to illustrate the dependence of bulk viscosity on the frequency (Figure 3). Frequency below 12 MHz has not been included, as at this frequency range the attenuation value is too small due to molecular relaxations. The attenuation values obtained for 10 repeats of the same sample were averaged at each frequency. Repeated measurements of the same sample have been taken to increase the confidence level of calculating an accurate averaged value as exact value is not attainable at each time. These attenuation values were put into equation (10) along with the other parameters to calculate the bulk viscosity at each frequency. The standard deviation (SD) and coefficient of variation (CV) which is the ratio of the standard deviation by the mean were calculated. To find out about the nature of the fluid (Newtonian or non-Newtonian), a shear stress against strain diagram was obtained for Tesco sunflower oil (Figure 4). This shows an excellent linear relationship (R^2=1) between stress and strain for all the samples at the selected temperatures.

The ratio of bulk viscosity to shear viscosity was calculated in order to compare their values (Table 2). A graph of bulk viscosity and shear viscosity has been plotted to find their dependence on temperature (Figure 5). Statistical analysis performed on the different brands to find the existence of significant difference in the bulk viscosity value has been summarised in Table 3. The p-value obtained from single factor Anova between the different brands were 0.81($p \geq 0.05$) for olive oil, 0.17($p \geq 0.05$) for sunflower oil. This suggests there is no statistical significant difference in the bulk viscosity value between the different brands of the same oil. This is further supported by the p-values ($\geq$ 0.05) obtained from performing t-test of two samples assuming equal

Sunflower oil	**Samples**	**T-M**	**T-F**	**M-F**
	p-value	0.33	0.05	0.40
Olive oil	**Samples**	**T-M**	**T-S**	**M-S**
	p-value	0.51	0.77	0.74
T: Tesco; M: Morrisons; F: Flora; S: Sierra mágina				

Table 3: p-values from t-test of two samples assuming equal variances between each brand of sunflower oil and extra virgin olive oil.

Temperature (°C)	Tesco		Morrisons		Flora	
	Mean × 10^{-2} Pa.s	SD × 10^{-2} Pa.s	Mean × 10^{-2} Pa.s	SD × 10^{-2} Pa.s	Mean × 10^{-2} Pa.s	SD × 10^{-2} Pa.s
6	5.79	1.89	4.83	1.87	5.84	1.83
10	4.51	1.59	3.8	1.65	4.67	1.61
15	3.57	1.43	2.89	1.44	3.97	1.42
20	2.91	1.21	2.3	1.18	3.39	1.19
25	3.33	0.99	1.98	0.98	3.22	0.98
30	2.77	0.86	1.97	0.87	3.62	0.83
35	2.07	0.71	2.87	0.78	3.12	0.72
40	2.36	0.6	2.07	0.6	3.26	0.61

*Standard deviation SD
*Mean was taken for the bulk viscosity in the frequency range of 12 MHz-100MHz. the lowest temperature is decided by the operating limit of the instrument

Table 4: The bulk viscosity values of three different brands of sunflower oil at the selected temperature range.

Temperature (°C)	Tesco		Morrisons		Flora	
	Mean × 10^{-2} Pa.s	SD × 10^{-2} Pa.s	Mean × 10^{-2} Pa.s	SD × 10^{-2} Pa.s	Mean × 10^{-2} Pa.s	SD × 10^{-2} Pa.s
8	6.62	2.25	11.22	3.8	9.64	3.35
10	5.17	1.9	5.98	1.82	6.01	2.07
15	3.87	1.54	4.72	1.48	4.22	1.63
20	3.09	1.35	3.69	1.34	3.24	1.44
25	3.34	1.23	3.07	1.19	2.68	1.22
30	2.44	1.01	2.67	1.02	1.56	0.88
35	2.2	0.85	1.88	0.84	2.58	0.85
40	2.74	0.73	2.66	0.71	2.16	0.74

Standard deviation SD
*Mean was taken for the bulk viscosity in the frequency range of 12 MHz-100MHz. The lowest temperature is decided by the operating limit of the instrument

Table 5: The bulk viscosity values of three different brands of olive oil at the selected temperature range.

Temperature T(°C)	Density ρ (kg m^{-3})	Velocity *v* (m s^{-1})	Shear viscosity (η × 10^{-2} Pa.s)
6	928.96	1516.2	5.82
10	926.29	1501.93	5.29
15	922.84	1485.02	4.64
20	919.41	1469.02	3.99
25	915.82	1451.75	3.35
30	912.51	1434.15	2.7
35	909.16	1451.75	1.77
40	905.82	1403.29	1.61

Table 6: Measured parameters: density, velocity, shear viscosity of Morrisons sunflower oil at the selected temperatures.

Temperature T(°C)	Density ρ (kg m^{-3})	Velocity *v* (m s^{-1})	Shear viscosity (η × 10^{-2} Pa.s)
6	927.92	1513.26	4.93
10	925.9	1502.03	4.53
15	922.45	1485.02	3.87
20	919.03	1467.22	3.21
25	915.59	1451.6	2.48
30	912.22	1435.59	1.49
35	908.84	1419.59	1.33
40	905.48	1403.52	0.76

Table 7: Measured parameters: density, velocity, shear viscosity of Flora's sunflower oil at the selected temperatures.

Temperature T (°C)	Density ρ (kg m^{-3})	Velocity *v* (m s^{-1})	Shear viscosity (η × 10^{-2} Pa.s)
8	922.41	1505.02	6.26
10	920.48	1495.16	5.83
15	917.11	1477.55	5.12
20	913.2	1462.53	4.41
25	910.33	1445.98	3.26
30	906.95	1429.56	3
35	903.58	1413.53	2.53
40	900.21	1397.39	1.56

Table 8: Measured parameters: density, velocity, shear viscosity of Tesco extra-virgin olive oil at the selected temperatures.

Temperature T(°C)	Density ρ (kg m^{-3})	Velocity *v* (m s^{-1})	Shear viscosity (η × 10^{-2} Pa.s)
6	920.88	1502.07	5.26
10	919.52	1492.69	5.04
15	916.07	1479.57	4.5
20	912.65	1462.53	3.95
25	909.23	1444.25	3.35
30	905.84	1429.1	2.86
35	902.44	1412.98	2.76
40	899.06	1397.4	1.61

Table 9: Measured parameters: density, velocity, shear viscosity of Morrisons extra-virgin olive oil at the selected temperatures.

variances between each brand (Table 3). As there is no significant difference in bulk viscosity between the brands, a generalised temperature dependence model of bulk viscosity was established by taking the best fit from the average plot of the three brands. The bulk viscosity values calculated for all the samples have been listed in Tables 4 and 5.

Discussion

Justification of the use of frequency squared equation

Since the exponent term δ was almost equal to 2 (δ=1.8344) (Figure 2), hence the use of frequency squared equation (equation 10) to find the bulk viscosity was justified. Attenuation is the result of both classical mechanisms (shear and bulk viscosity, thermal contributions) as well as due to molecular relaxations. The reason for δ being less than 2 is due to the occurrence of molecular relaxations which has not been accounted for in the equation (Tables 6-9). One of the most important reasons for molecular relaxations maybe due to the molecular rearrangements that occur during the compression of oil in the ultrasonic field [12]. There might be some error as excess attenuation due to the formation of air bubbles in the sample inside the Ultrasizer. Also, any thermal fluctuations during the measurement might result in some error as attenuation, density, velocity are dependent on temperature as explained earlier.

Dependence of bulk viscosity on frequency

The higher the value of CV the more dispersed is the data. CV for the mean bulk viscosity across the frequency range was found to be ≥ 0.05 at each temperature (Table 1). This indicates that across the frequency range there is a variation in the bulk viscosity value which cannot be ignored. This suggests that the bulk viscosity values calculated for all

Temperature T (°C)	Density ρ (kg m^{-3})	Velocity v (m s^{-1})	Shear viscosity (η × 10^{-2} Pa.s)
6	922.41	1506.8	6.05
10	920.48	1494.07	5.67
15	917.11	1479.5	5.03
20	913.2	1461.05	4.4
25	910.33	1445.72	3.77
30	906.95	1429.1	3.2
35	903.58	1412.98	2.27
40	900.21	1397.4	2.05

Table 10: Measured parameters: density, velocity, shear viscosity of Sienna mágina extra-virgin olive oil at the selected temperatures.

the selected temperatures are frequency dependent and not constant. Also, from Figure 3 it is seen that the bulk viscosity decreases with the increase in frequency for all temperatures, showing bulk viscosity is frequency dependent. This indicates the non-Newtonian behaviour for sunflower oil and olive oil over the selected temperature and frequency range, as for a fluid to be Newtonian the bulk viscosity must be constant for a selected frequency range [6]. However, from the stress and strain diagram obtained from Figure 4, edible oils are showing Newtonian nature as the shear stress is proportional to the shear strain.

Comparison of bulk and shear viscosity

The ratio of bulk viscosity to shear viscosity was found to be around 1 at all the selected temperatures (Table 2). This shows for edible oils the bulk viscosity is almost equal to its shear viscosity. However, the contribution of bulk viscosity to sound propagation due to non-Newtonian fluid seems to be quite less than due to Newtonian fluid as the bulk viscosity of water (a Newtonian fluid) was reported to be almost three times larger than its shear viscosity [13]. A plot of bulk and shear viscosity against temperature (Figure 5) shows there is a decrease in both the values with the increase in temperature indicating both are temperature dependent.

Test of hypothesis

The hypothesis tested that there is a significant difference between the different brands of the same edible oil is rejected based on the results obtained from the statistical analysis ($p \geq 0.05$) (Table 3). This shows bulk viscosity of edible oils do not seem to be much affected by small compositional differences. This is an important finding for future bulk viscosity studies as only one brand can be used to represent a class of oil, saving both time and resources (Table 10).

Temperature dependence of bulk viscosity

A decrease in the bulk viscosity is seen as the temperature increases (Figures 6 and 7).

The temperature dependent bulk viscosity model has been established as:

$$\mu = \mu_1[\mathrm{T}\exp(\mathrm{A})]$$

Where μ (Pa.s) is the bulk viscosity, μ_1 (Pa.s.°C^{-1}) T is temperature (°C), A is constant, μ_1 is 0.1092 and A is -0.413 for sunflower oil (Figure 6) and μ_1 is 32.169 and A is -0.744 for extra-virgin olive oil (Figure 7). The findings from this study cannot be generalised for edible oils as the sample size was not adequate as only two types of oil: sunflower oil and extra-virgin olive oil was investigated and only three brands from each were taken into consideration. Further studies should be conducted with larger sample size like with other commonly used edible oils. For the different brands simple random sampling must be undertaken so that the samples well represents the entire class.

Conclusion

The temperature dependence of bulk viscosity in edible oil using acoustic spectroscopy was established using a model equation. The mean bulk viscosity decreases with the increase in temperature. The value of bulk viscosity is not constant over the frequency range, it decreases with increase in frequency. Therefore, in terms of bulk viscosity edible oils are non-Newtonian fluids. There is no significant statistical difference of bulk viscosity value between different brands of the same oil. This suggests that future studies with only one variety of oil will be enough. Even though other physical properties of edible oils have been extensively studied, little research has been done on bulk viscosity. More research should be undertaken to check the reproducibility of these results and validate the data.

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