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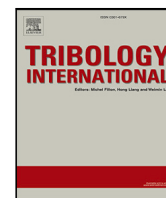
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In-situ ultrasonic viscometry of lubricants under temperature and shear

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ABSTRACT

Understanding the behaviour of engine and gear oils, especially the viscosity, under temperature and shear is important to improve machine operation. A novel viscometer using ultrasound is presented and used under a range of temperatures. A single cross-temperature (between 20 °C and 60 °C) ultrasonic calibration is shown to be sufficient. Next, the ultrasonic viscometer is compared to a conventional viscosity measurement technique for Newtonian and non-Newtonian lubricants. Newtonian viscosity standard fluids and shear-thinning engine oils are studied. Both viscosity measurement techniques match for Newtonian fluids but ultrasonic measurements are consistently lower for shear-thinning fluids. It suggests that the ultrasonic viscometer is similar to a high-shear viscometer with a shear rate of about 10^6 s^{-1} .

1. Introduction

The use of gears and engines in an industrial context implies extreme operating conditions: high temperatures, pressures, and shear rates. Contacts are lubricated to ensure proper operation of machines and longer life span. To increase efficiency and reduce costs, it is of interest to understand lubricant behaviour in the various lubrication regimes ranging from boundary to hydrodynamic lubrication conditions.

Liquid lubricants are studied here as they represent most of the consumption in industrial applications [1]. Most lubricants are made of a base oil, which represents at least 70% of their weight [1,2]. Chemical compounds called additives are added to improve the base oil properties.

Besides chemical and physicochemical interaction of lubricant additives with surfaces in tribocontacts, viscosity is the most important physical property of a lubricant to enable appropriate lubrication [1]. It is generally measured using conventional viscometers, such as rheometers, falling-body, and capillary viscometers. However, these viscometers are seldom able to measure viscosity on-line and cannot measure viscosity in the tribological contact. Viscosity significantly varies under temperature, pressure, and shear rate, which reach their maximum in the contact. This is where most premature failures happen due to fatigue and improper lubrication (e.g. spalling, smearing, fretting) [3,4]. Therefore, measuring viscosity in the contact is of great interest.

Shearing is the deformation of a material due to a force parallel to its surface. In lubrication, a fluid is mechanically sheared when trapped

between two solids in relative motion to each other. A propagating shear wave can also cause an oscillatory shear. Shear modifies lubricants at a molecular level and thus has an impact on the fluid properties such as viscosity. Low-shear has a temporary effect, but high-shear or repeated shearing can permanently affect the molecules and fluid properties [5].

Some fluids are considered Newtonian: their viscosity does not change on a given shear rate range. Some fluids are shear-thickening or shear-thinning: their viscosity increases or decreases with the shear rate, respectively (Fig. 1). Most lubricants are shear-thinning: they display a first Newtonian plateau, a transition zone, and a second Newtonian plateau.

From a fundamental research perspective, it is of interest to quantify fluid viscosity over a wide shear rate range. Moreover, high-shear viscosity has applications in industry as engine and gears operate at high shear rates. There is a need to assess the viscosity [6], the permanent viscosity loss [7], or the impact of additives [8] in oils under these conditions. There are also applications outside of mechanics, such as in the pharmaceutical [9], the food-processing [10], or other industries [11].

Depending on the application, fluids withstand varying operating conditions, especially inside contacts. The operating conditions are difficult to reproduce ex-situ, therefore in-situ measurements should be favoured. The purpose of ultrasonic viscometry development is to overcome the limits of conventional viscometers. The aim of the present work is to understand the impact of operating conditions on ultrasonic viscometry.

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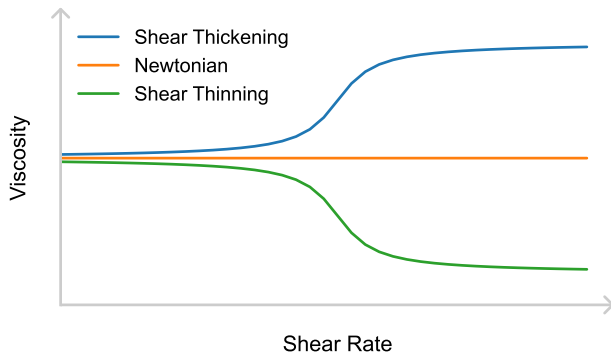


Fig. 1. Shear-thickening, Newtonian, and shear-thinning fluids have different viscosity behaviours under shear.

2. Ultrasonic viscometry

Ultrasonic reflectometry is an in-situ and non-destructive testing technique [12,13]. Ultrasonic transducers are relatively small and affordable. This is why the technique has been extensively studied since the 1950s, with applications in medicine, imaging, and engineering. Longitudinal ultrasonic waves have been used to measure several tribosystem properties, such as film thickness [14,15], wear [16], or load [17].

Mason [18] was the first to use shear acoustic waves to measure viscosity. He used vibrating crystals to apply a transverse excitation in fluids: the crystal resonance frequency shifted depending on the fluid viscosity. However, the technology required the crystal to be immersed in the fluid, which made viscosity measurement in contacts impossible.

Further works [19–21] also used shear waves but moved the position of the vibrating element outside the fluid and correlated resonance frequency or damping to viscosity. These techniques required an understanding of wave propagation through layers.

Reflectometry is based on wave transmission and reflection at interfaces [22,23]. The reflection coefficient is the portion of reflected wave compared with incident upon reaching an interface. It depends on the material properties from both sides of the interface, such as the acoustic impedance, which represents the ability of a wave to travel through a material. For a two-layer system, the reflection coefficient is described as $R = \frac{z_2 - z_1}{z_2 + z_1}$, with z_1 and z_2 the acoustic impedances of layer 1 and layer 2, respectively.

In the application described in the present paper, the first layer is a metal component. Its acoustic impedance is $z_1 = \rho c$, with ρ the metal density and c the shear wave velocity. The second layer is the studied fluid. Its acoustic impedance is $z_2 = \sqrt{\rho i \omega \eta}$ for Newtonian fluids, with ρ the fluid density, ω the angular frequency, and η the fluid viscosity. The orders of magnitudes are $z_1 = 10^7 \text{ kg s}^{-1} \text{ m}^{-2}$ and $\text{Re}(z_2) = 10^5 \text{ kg s}^{-1} \text{ m}^{-2}$. In this case, $z_1 \gg z_2$, thus $R \approx 1$. The measurement is not sensitive enough to viscosity.

The reflectometry technique was recently improved by Schirru [24] using a matching layer to increase the measurement sensitivity. Quarter-wave matching layers have been used since the 1950s and originally aimed at widening the bandwidth and increasing the efficiency of piezoelectric transducers [25–27]. Their name comes from their thickness that is chosen so it is a quarter of the transducer wavelength.

The novelty in Schirru's approach was to use the matching layer not in contact with the piezoelectric transducer but to acoustically match other layers. He thus formed a three-layer system and bonded shear ultrasonic sensors, generally between 1 MHz and 10 MHz, to the metal surface as shown in Fig. 2. The matching layer material was polyimide so its shear acoustic impedance was encompassed by the one of the metal and the one of the lubricant: $z_1 > z_2 > z_3$. Its thickness L was chosen according to the sensor frequency: $L = \frac{nc}{4f}$, with n an integer, c

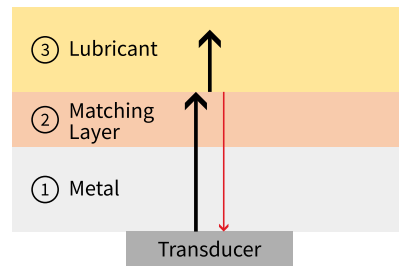


Fig. 2. A three-layer system comprising a matching layer to increase ultrasonic shear waves sensitivity to lubricant properties.

the shear velocity of the wave, and f the input frequency. This shear ultrasonic setup is used throughout the present work.

A shear ultrasonic wave is emitted by the transducer and travels through the three-layer system. Constructive interferences are created inside the matching layer, increasing the wave energy transmitted to the fluid. The wave reflected from the fluid is recorded by the transducer. It is compared with the one reflected from an air reference in order to compute the reflection coefficient R (Eq. (1)). A calibration curve is then used to link the reflection coefficient with the viscosity, as described in Section 4.3.

$$R = \frac{\text{lubricant signal}}{\text{reference signal}} \quad (1)$$

In this work, the three-layer system principle and shear ultrasonic waves were used to measure oil viscosity. First, the impact of temperature on the ultrasonic calibration was gauged. Second, the impact of shear rate was assessed, both on viscosity and on ultrasound. To do so, a comparison was made between an ultrasonic viscometer and a conventional viscometer.

3. Materials and equipment

3.1. Ultrasonic instrumentation

A parallel-plate rheometer was chosen as the conventional viscometer as it could measure low-viscosity fluids on a relatively wide range of shear rates. The Anton Paar MRC301 rheometer, housed at INSA Lyon, presented the following characteristics. The upper plate rotated while the bottom plate was fixed. The plate gap was controlled and could be reduced down to 0.1 mm, allowing the measurement of fluids with a dynamic viscosity as low as a few mPa s with an error lower than 5%. The upper plate rotated at a variable speed to control shear rates from 10^{-3} s^{-1} up to 18000 s^{-1} . Temperature could be set from -15°C to 150°C with a 0.01°C precision using a Peltier device and a passive insulated hood.

The flat metal plates with direct line of sight of the lubricant also made the rheometer easy to instrument with ultrasonic sensors. The lower plate was instrumented with a matching layer and a piezoelectric element (Fig. 3). The matching layer was $50 \mu\text{m}$ thick and the transducer was a 5 MHz shear element.

A computer was used to design waveforms through a LabVIEW interface. The voltage pulses were generated by a portable oscilloscope (PicoScope 5444b Series) and carried by coaxial cables. A piezoelectric transducer converted the electric signal into an ultrasonic wave. The reflected ultrasonic wave was received by the same transducer (technique known as pitch-catch) and converted into an electric signal. Acquired data was processed as described in Section 4.3.

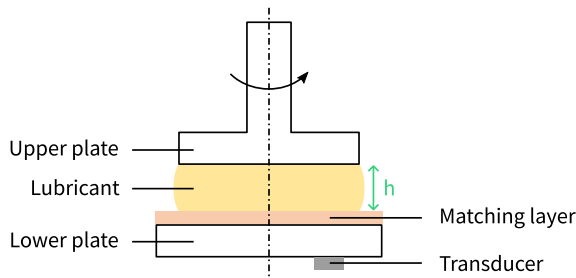


Fig. 3. The lower plate of the rheometer is instrumented with a matching layer and an ultrasonic transducer.

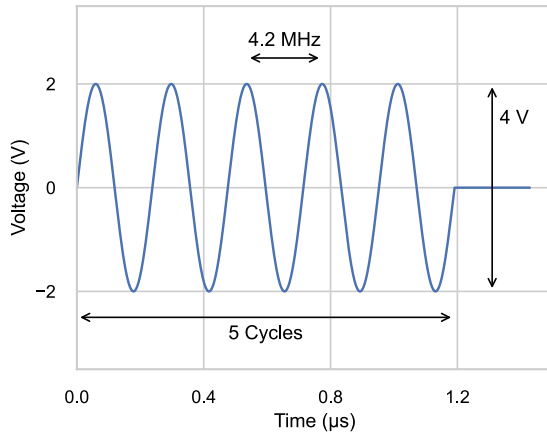


Fig. 4. The input wave is defined with its shape, frequency, number of cycles and voltage amplitude.

3.2. Test oils

Viscosity standard fluids from Cannon [28] and VWR [29] were used to calibrate the ultrasound viscometer. The viscosities were known at various temperatures between 20 °C and 100 °C, and ranged a few mPa s up to around 150 mPa s. They were mineral hydrocarbon base oils and presented a Newtonian-like behaviour, meaning their viscosity did not depend on the shear rate.

In Sections 6.1 and 6.4, the same fluids and the same mixture ratio as Bair [30] were used to compare the rheometer and the ultrasonic viscosity measurements. Squalane (SQL) was chosen as a low-viscosity base oil. It had a low molecular weight (422.81 g mol⁻¹) inducing a Newtonian behaviour. The additive was polyisoprene (PIP). This polymer had a very high molecular weight (≈ 40 000 g mol⁻¹) which induced shear-thinning. The mixture was made of 85% SQL and 15% PIP and was representative of low-viscosity gear and engine lubricants. It is denoted SQL+PIP in this work.

Some other fully-formulated fluids, including a 10W-30 engine oil, were also used in Section 6.1.

4. Experimental method

4.1. Rheometer methodology

During the temperature experiment, the rheometer was only used for its temperature regulation. The upper plate of the device was not in contact with the sample and did not shear it.

During the shear experiment, the lubricant was deposited on the lower plate of the rheometer in a sufficient quantity to fill the gap (less than 1 mL). The upper plate was lowered to reach a desired gap between 0.2 mm and 0.5 mm depending on the fluid viscosity. The rheometer applied a constant shear rate and measured the viscosity 20

times over the course of 1 min. The shear rates were 10 s⁻¹; 100 s⁻¹; 500 s⁻¹; 1000 s⁻¹; 5000 s⁻¹; 10000 s⁻¹. The experiments were carried out at three different temperatures: 20 °C, 40 °C, and 60 °C.

As the in-between plates gap h , the upper plate radius R , and the rotational speed ω were known, the shear rate $\dot{\gamma}$ could be computed using Eq. (2). Dynamic viscosity η was then computed using Newton's law (Eq. (3)), with τ the shear stress.

$$\dot{\gamma}_{mean} = \frac{2}{3} \times \frac{\omega R}{h} \quad (2)$$

$$\eta = \frac{\tau}{\dot{\gamma}} \quad (3)$$

4.2. Ultrasound methodology

The ultrasound input signal was a sine wave with input parameters presented in Fig. 4. 20 points were captured during the 1-minute test. Each point was made of 200 captures that were averaged to reduce noise.

The fluid was allowed 10 min to reach temperature equilibrium before each recording.

During the shear experiment, the ultrasonic and the rheometer measurements were performed simultaneously. The ultrasound acquisition was very short and at a very high rate, preventing an impact on the rheometer measurement, as evidenced in Section 4.4.

4.3. Ultrasound signal analysis

The signal processing is detailed in this section and depicted in Fig. 5.

The emitted and the reflected waves were recorded on the same time-domain signal called the A-Scan. The signal was cropped to only keep the first reflection. Zero-padding on the time-domain signal increased the frequency resolution of the Fast Fourier Transform output. The reflection coefficient was computed by dividing the frequency-domain signal amplitude of the lubricant by the amplitude of the reference (as shown in Eq. (1)). The lowest reflection coefficient of each fluid was selected. Because fluids of known viscosity were used, it was possible to link the reflection coefficient and the viscosity. A logarithm fit was then performed to build the calibration curve. This curve was specific to the sensor and the operating conditions.

Following the same process, reflection coefficients of unknown fluids were measured. Using the calibration curve, each reflection coefficient was converted to a viscosity.

4.4. Matching layer impact

Following the ultrasonic instrumentation, the lubricant was now in contact with a polymer instead of stainless steel from the original lower plate. The material properties had changed and the plate parallelism might have been different. This invasive instrumentation might have had an impact on the viscosities measured by the rheometer. To ensure this was not the case, two measurements were performed: one with the original lower plate and one with the instrumented lower plate.

In Fig. 6, the rheometer measurements of a Newtonian oil with and without matching layer are compared. They show a very good agreement with less than 1.4% difference at 100 s⁻¹. It confirms that the matching layer had a negligible impact on the rheometer measurements.

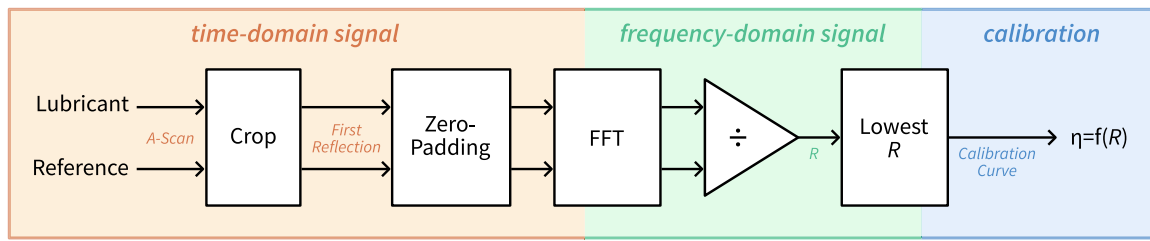


Fig. 5. The signal processing is separated in three steps: the time-domain signal, the frequency-domain signal, and the calibration. It correlates the reflection coefficient R to the viscosity η .

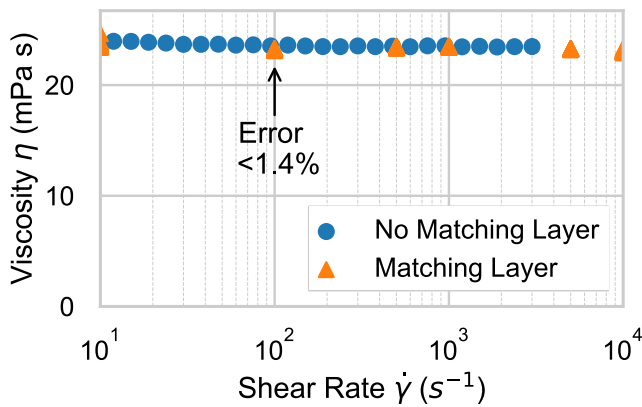


Fig. 6. Rheometer viscosity measurements with and without matching layer. The instrumentation of an invasive matching layer has negligible repercussions on the rheometer measurements.

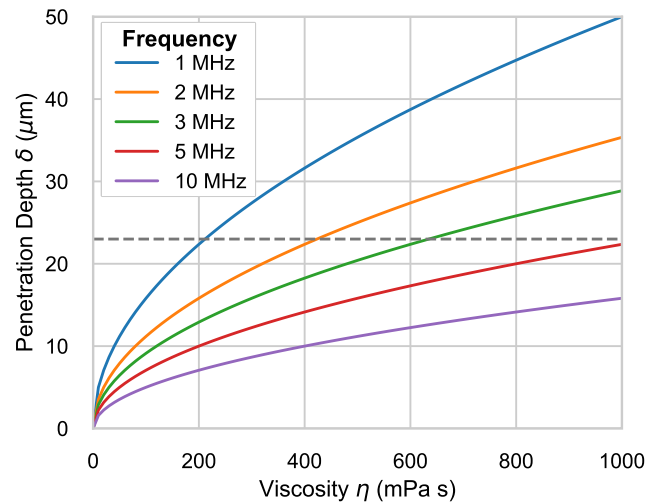


Fig. 8. Penetration depth of ultrasonic shear waves in liquids.

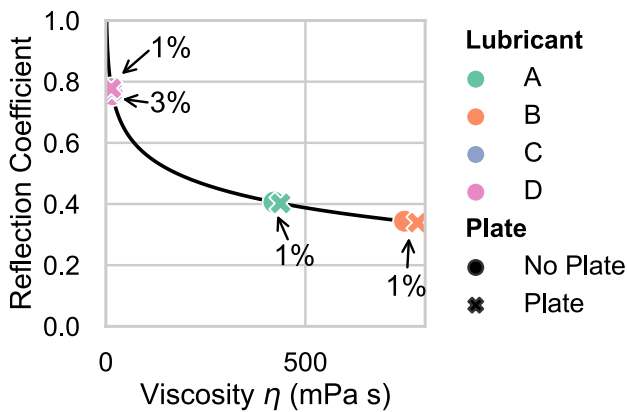


Fig. 7. Reflection coefficient measurements with and without upper plate in the rheometer. The contact of the upper plate with the sample has negligible repercussions on the ultrasonic measurements.

4.5. Higher plate impact

The ultrasonic wave travelled through the lower plate, the matching layer, and the lubricant. The presence of the rheometer upper plate could have had an impact on the wave by creating unwanted reflections and interferences. To ensure this was not the case, two static ultrasonic measurements were performed: one with the upper plate in contact with the fluid and one without it.

In Fig. 7, the ultrasound viscosity is compared with and without the upper plate for four Newtonian lubricants. The measured reflection coefficients have less than 3% difference. It confirms that the upper plate had a negligible impact on the ultrasonic measurement.

This is an expected result as shear waves propagate on very short distances in fluids; they are rapidly attenuated. This attenuation is

characterised through the penetration depth. It is the distance travelled by a wave before its amplitude drops to $1/e$ of its initial value. The penetration depth δ is described with Eq. (4), where η is the dynamic viscosity, ρ_L the liquid density, and ω the angular frequency.

As shown in Fig. 8, the penetration depth of a 5 MHz wave (red curve) in a 850 kg m^{-3} and 1000 mPa s fluid is at most $25 \mu m$. This penetration distance was much smaller than the 0.2 mm gap used in these experiments. As such, the wave did not reach the upper plate and there was no interference problem.

$$\delta = \sqrt{\frac{2\eta}{\rho_L \omega}} \quad (4)$$

4.6. Limitations and uncertainties

Several sources of uncertainty should be considered, some experimental and some from the data processing.

The largest experimental uncertainty is linked to the temperature-viscosity relationship. The Vogel-Cameron empirical law (Eq. (5)) describes the link between the dynamic viscosity η and the temperature T . It uses three numerical parameters a , b , and c , which are determined using fluid viscosities at three different temperatures. In Fig. 9, the Vogel-Cameron law is used to show the viscosity decrease on a temperature range for two viscosity calibration lubricants (N26 and N35). It highlights that viscosity varies more at low temperatures. It is thus important to precisely control the temperature, as an error on temperature regulation leads to an error on viscosity. Measuring at low temperatures leads to more uncertainty.

In practice, temperature errors are caused by a regulation latency due to heating when shearing a viscous fluid at a high shear rate. In this case, the viscosity drop should not be mistaken with the start of the

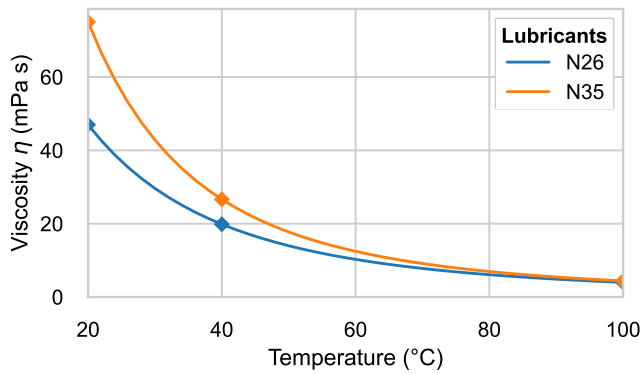


Fig. 9. Viscosity-behaviour of fluids from the Vogel-Cameron law. Viscosity varies more at low temperatures.

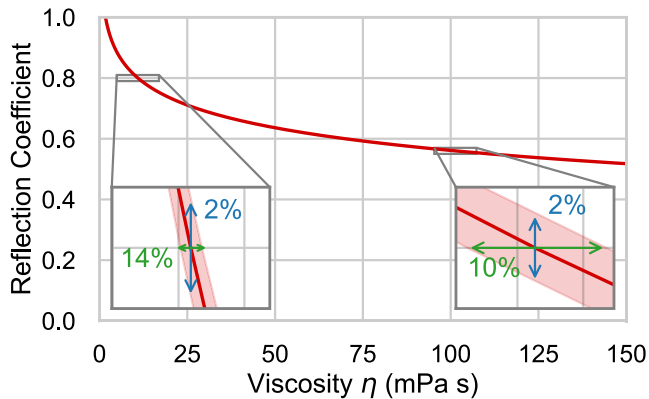


Fig. 10. Reflection coefficient uncertainty.

transition zone. As such, it is better to perform viscosity measurements at 60 °C than at 20 °C. However, high temperatures generate viscosities that might be too low to be measured with the parallel-plate rheometer. A compromise is thus required.

$$\eta(T) = a \times \exp\left(\frac{b}{T - c}\right) \quad (5)$$

The experimental uncertainty on the measurement of the reflection coefficient is assessed through repetitions. The relative error between the highest and the lowest reflection coefficients measured for a fluid does not exceed 2%.

However, this uncertainty is increased during the data processing. The logarithm shape of the calibration curve amplifies the error: a low relative error on the reflection coefficient leads to a larger error on the viscosity.

The calibration curve is shown in red in Fig. 10. In the boxed graphs, a 2% error on the reflection coefficient is added in light red and is emphasised with blue vertical arrows. The viscosity error is then represented with horizontal green arrows. For low viscosities around 10 mPa s, the viscosity error is 14%. For viscosities around 100 mPa s, the viscosity error is 10%.

It highlights the need of a very precise reflection coefficient measurement, especially at low viscosity.

To reduce the calibration curve uncertainty, a possibility is to increase the ultrasound measurement sensitivity by using aluminium instead of steel. Aluminium is softer than steel, its acoustic impedance is lower which reduces the acoustic mismatch with the fluid. This was however not recommended in this application as the steel hardness was needed to ensure the desired gap geometry (such as parallelism) for the rheometer measurements.

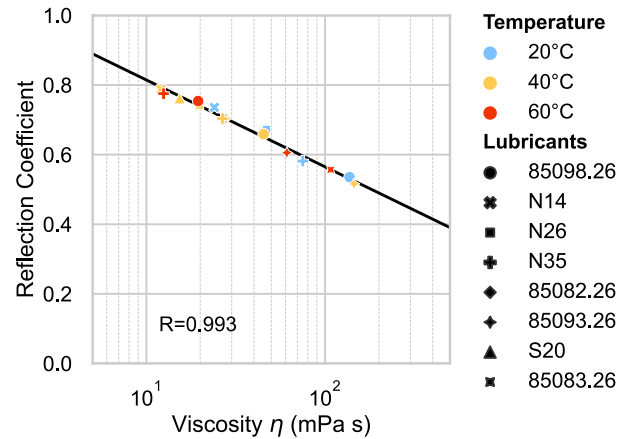


Fig. 11. The calibration curve is not temperature-dependent: a cross-temperature calibration curve can thus be used.

5. Temperature results

The impact of temperature on ultrasonic viscosity measurements should be assessed, more particularly the impact on the ultrasonic calibration process. To do so, the link between reflection coefficient and viscosity under a range of temperature was studied. The reflection coefficients from several Newtonian-like viscosity calibration fluids were measured at three different temperatures (20 °C, 40 °C, and 60 °C). The viscosities of viscosity calibration fluids are known over a wide range of temperatures. Each reflection coefficient could thus be linked to a viscosity. The calibration lubricants should be chosen so that their viscosities span a range wider than the unknown fluids to be measured.

The reflection coefficient versus viscosity points are plotted in Fig. 11. Each point represents a single fluid at a single temperature. Their colour (blue, yellow, or red) indicates the acquisition temperature (20 °C, 40 °C, or 60 °C, respectively). A logarithm curve is then fitted to the calibration points and shown in black. The x-axis of the graph is in log-scale so the logarithmic curve is represented as a straight line.

The built calibration curve is a good fit, as evidenced with a close to unity correlation coefficient. It proves a single cross-temperature calibration curve is sufficient to link the reflection coefficient and the viscosity of lubricants. This means fluids *travel* on the calibration curve as the temperature changes.

The calibration of an ultrasonic apparatus, albeit necessary, is a time and resource consuming step. The single cross-temperature calibration is a primordial finding to facilitate ultrasonic viscometry implementation at a larger scale. Several benefits can be drawn for future calibrations.

1. **Time savings.** A single calibration is performed instead of one per temperature.
2. **Larger viscosity range.** It can be difficult and expensive to have a wide viscosity range of calibration fluids. This is solved by using more temperatures. For example, a high viscosity fluid at 60 °C can be hard to procure, which is not the case of one at 20 °C.
3. **Higher accuracy.** Because it is easier to procure a wide range of viscosities, more points can be used to fit the calibration curve.

The cross-temperature calibration was used in subsequent measurements.

6. Shear results

6.1. Newtonian-like and non-Newtonian fluids

The impact of shear on viscosity should be assessed. SQL is chosen as the first Newtonian-like fluid to be studied. Its viscosity is compared

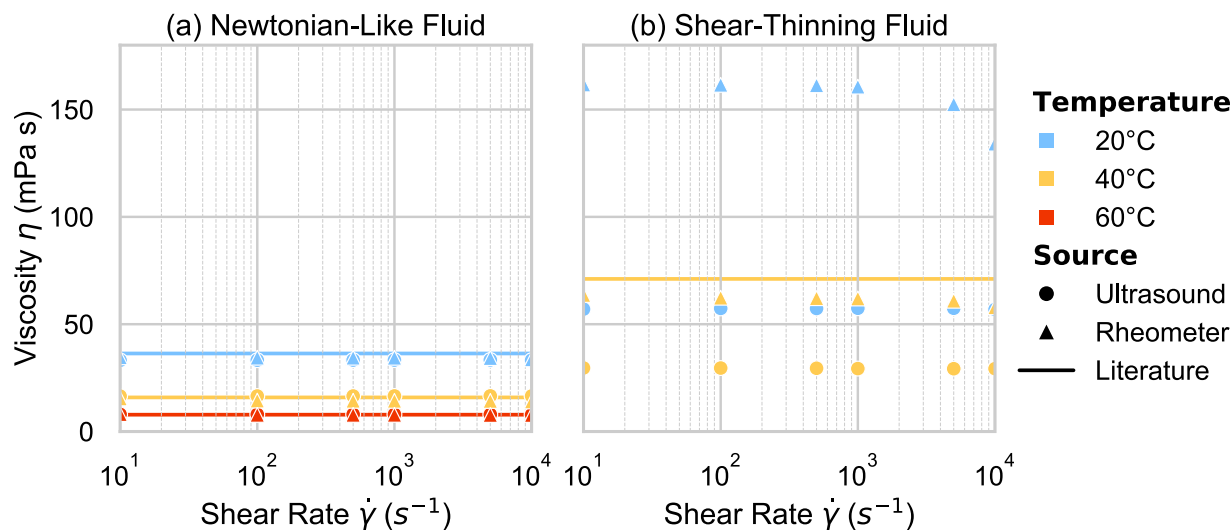


Fig. 12. Rheometer and ultrasonic viscosity measurements for (a) the Newtonian-like fluid SQL and for (b) its shear-thinning counterpart SQL+PIP. The literature data comes from Bair [30].

under three different temperatures and using three sources: the rheometer, the ultrasound, and low-shear literature data from Bair [30]. The results are plotted in Fig. 12a. As expected, the viscosity decreases as the temperature increases. For each temperature, the viscosity remains constant across the shear rate range, which is expected of a Newtonian-like fluid. Additionally, there is a very good agreement between the three viscosity sources.

More Newtonian-like fluids are then studied. In Fig. 13a, all data points lie on the $x = y$ line, meaning there is a strong agreement between the ultrasonic and the rheometer measurements. The correlation coefficient is close to 1. It validates the ultrasonic measurements for Newtonian-like fluids.

SQL+PIP, considered to be an engine oil (see Section 3.2), is then studied in Fig. 12b. At low temperatures, the rheometer viscosity is constant at low shear rates, then decreases at higher shear rates. This is expected of a shear-thinning fluid. The literature data [30] at 40°C was measured at low shear. The gap between the low-shear rheometer viscosity and the literature data is probably due to a small difference in the blend ratios of the sample used in Bair's work and in the present study. However, the ultrasonic viscosity is constant and lower than the rheometer viscosity.

The study of additional fully-formulated fluids show similar results (Fig. 13b). There is a gap between the ultrasonic and the rheometer data, with the rheometer viscosity being consistently higher.

Moreover, it is of interest to compare the results of SQL and its shear-thinning counterpart SQL+PIP. The viscosity of SQL+PIP is necessarily higher than that of SQL as PIP is a very viscous additive. The ultrasonic viscosity of SQL+PIP, although lower than its rheometer viscosity, is higher than the ultrasonic viscosity of SQL alone. This suggests that ultrasound is not sensitive solely to the base oil and that it measures the high-shear viscosity of lubricants.

6.2. Shear heating

Most viscosity measurement techniques rely on shearing the sample of interest at a certain rate. When the fluid is mechanically sheared, through rotational speed in the rheometer for example, heating happens.

In Fig. 12b, the rheometer viscosity of the shear-thinning fluid decreases as the shear rate increases. This is mostly visible at 20°C. It is known that some of the viscosity decrease could be due to shear heating but it is difficult to quantify how much and what is additionally caused by shear-thinning. The ultrasonic data was acquired simultaneously

with the rheometer data. It does show a slight decrease as the shear rate increases but tends to indicate that minimal shear heating is occurring. It is therefore reasonable to assume that the viscosity decrease is mostly due to shear-thinning.

6.3. Rheometer vs. ultrasound shear

The discrepancy between rheometer and ultrasound data for non-Newtonian fluids has been similarly observed by Shepard et al. in [31], by Rabani et al. in [32], or by Schirru et al. in [24].

Shepard et al. and Rabani et al. used an oscillatory probe to measure viscosity; it was successful on Newtonian-like fluids but showed a gap for non-Newtonian fluids. Rabani et al. used a 625 kHz frequency for their probe, which they estimated to apply a 5000 s^{-1} shear rate on water. They believed the probe shear rate did not have an impact as it was close to what the conventional viscometer applied. But the frequency was thought to have an impact.

Similarly to the present paper, Schirru et al. used ultrasound to measure viscosity.

Based on the cited papers and the current work, there are two factors that could explain the gap between ultrasonic and conventional viscosity measurements for non-Newtonian fluids.

The first factor is the difference in shear rates between the rheometer and the ultrasound. The rheometer shears at a maximum of 10^4 s^{-1} while the ultrasound is similar to a high-shear viscometer. This has no impact on Newtonian fluids as their viscosity is constant whatever the shear rate. But for shear-thinning fluids, the high-shear viscosity is lower than the low-shear viscosity (Fig. 1).

The second factor is the type of shear and how it affects the molecules. The molecular structure of lubricants is of interest. In Newtonian fluids, all molecules have similar sizes and behaviour. They are all similarly excited under shear. However, shear-thinning fluids are partly made of additives such as polymers. These are generally large molecules tangled at rest. Under mechanical shear, they untangle and the molecules align with the flow. At a macro level, the fluid opposes less resistance; its viscosity is lower. Under oscillatory shear, the molecules excitation depends on the relaxation time. As such, polymers do not get excited as much as the smaller molecules. Only a partial response of the fluid is recorded with ultrasound.

The differences between rheometer and ultrasonic shears are illustrated in Fig. 14. The x -axis is split to best represent both types of shear. The rheometer measures between 10 s^{-1} and 10^4 s^{-1} , while the ultrasound is between 10^6 Hz and 10^7 Hz . The orange line represents

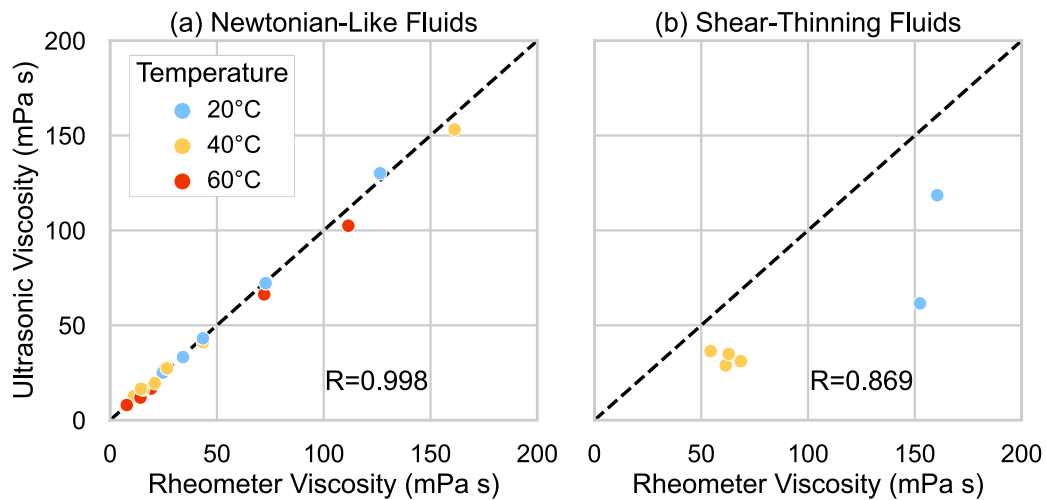


Fig. 13. Rheometer and ultrasonic viscosity measurements comparison for (a) Newtonian-like fluids (b) fully-formulated fluids.

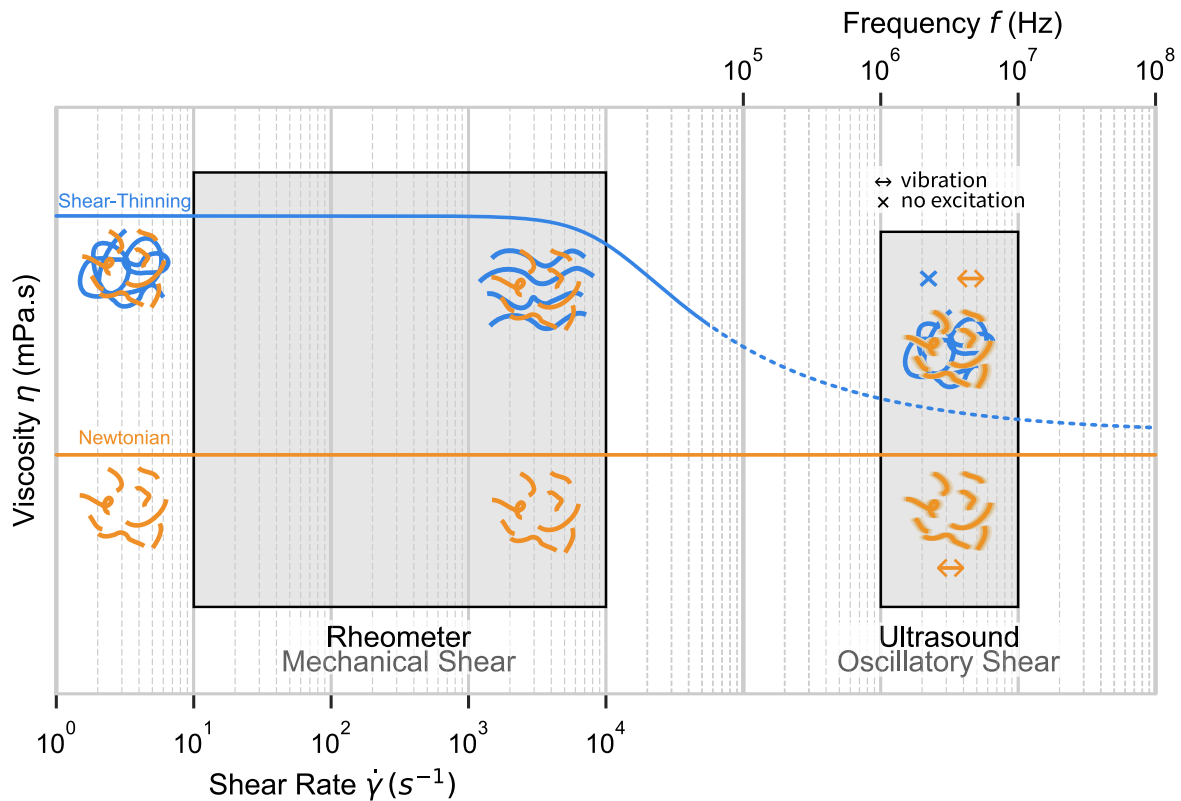


Fig. 14. Mechanical and oscillatory shear for Newtonian and non-Newtonian fluids.

Newtonian fluids. These are made of small and light molecules which are not impacted by shear. The blue curve represents shear-thinning fluids. These are made of a combination of small and large molecules. The reaction of large molecules to shear is the cause of shear-thinning behaviour.

As the rheometer and the ultrasonic shears are applied at different shear rates, have different types, and are undetected by the other measurement technique, they should be decoupled. This is investigated in the following section.

6.4. Decoupling rheometer & ultrasound shear

SQL+PIP is measured with the same rheometer as previously but using a Cone-Plate geometry to expand the shear rate range. This data

represents the first Newtonian plateau and the start of the transition zone.

The high-shear viscosity is measured with ultrasound. It produces a single point because it is performed at a single frequency. The conversion from frequency to shear rate is a subject of debate. The Cox-Merz law (Eq. (6)) [33] is widely accepted and is used here. In this equation, η is the dynamic viscosity, ω the angular frequency, and $\dot{\gamma}$ the shear rate. The original paper established the law for relatively low shear rates from 25 s^{-1} to 2000 s^{-1} and for angular frequency from 0.1 rad/s to 20 rad/s . Bair et al., in [34], showed the law only applied to low-molecular weight liquids. Given the lack of consensus on a Cox-Merz law replacement, it is used here. Thereby, the 4.2 MHz ultrasound

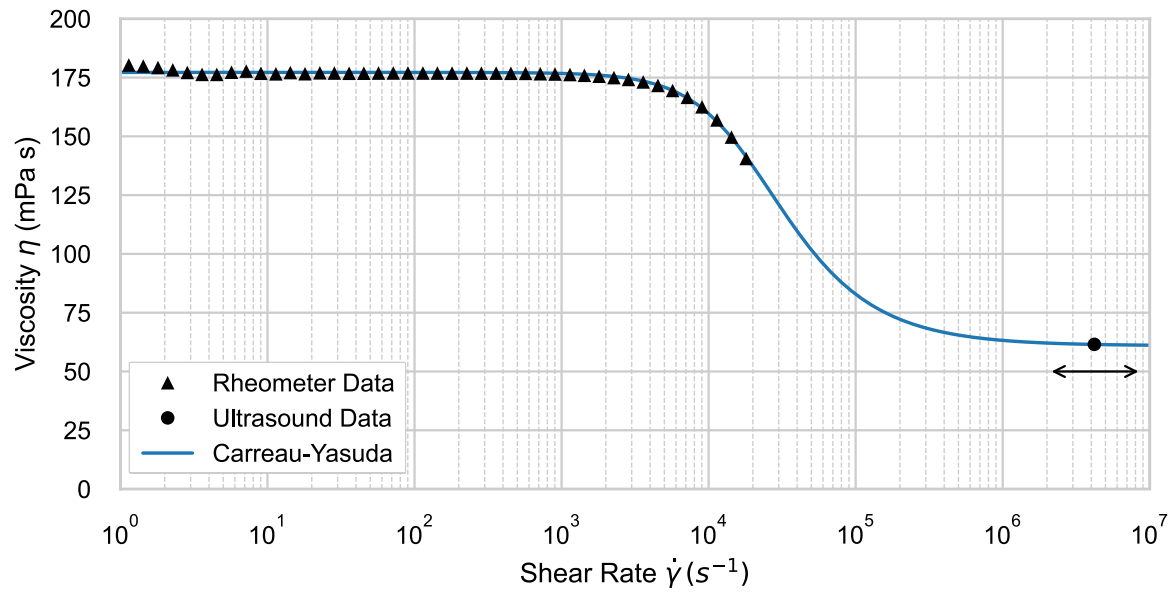


Fig. 15. Rheometry and Carreau-Yasuda Model of SQL+PIP, a shear-thinning engine lubricant.

frequency is considered to apply a $4.2 \cdot 10^6 \text{ s}^{-1}$ shear rate.

$$\eta(\omega) = \eta(\dot{\gamma})|_{\omega=\dot{\gamma}} \quad (6)$$

In order to better understand the viscosity across the whole shear rate range, a Carreau-Yasuda model (see Eq. (7)) [35] is fitted to the experimental data. In this equation, η_0 and η_∞ are the dynamic viscosities of the first and second Newtonian plateaux, respectively; τ is the shear stress, $\dot{\gamma}$ the shear rate, and n and a are numerical parameters. The output is $\eta(\dot{\gamma})$, the dynamic viscosity as a function of the shear rate. The results are presented in Fig. 15.

$$\eta(\dot{\gamma}) = \eta_\infty + (\eta_0 - \eta_\infty) \left[1 + (\tau \dot{\gamma})^a \right]^{\frac{n-1}{a}} \quad (7)$$

More points are needed at high shear to improve the fit and better understand the link between shear rate and frequency.

As shown by Taylor et al. [36], industrial applications such as engines shear fluids between 10^2 s^{-1} and 10^8 s^{-1} . It is thus primordial to understand the viscosity behaviour of lubricants under a wide range of shear rates. Conventional viscosity measurements at high shear rates ($> 10^4 \text{ s}^{-1}$) and very high shear rates ($> 10^6 \text{ s}^{-1}$) are limited by several factors: viscosity and temperature range, as well as time, cost, and equipment availability. Ultrasound could provide an alternative high-shear viscosity measurement, with wide viscosity and temperature ranges.

7. Conclusion

The present work deepens the understanding of ultrasonic viscometry by evaluating the impact of temperature and shear on measurements.

A first set of experiments studied the impact of temperature on the ultrasonic calibration. The apparatus calibration was a tedious process that needed to be performed at each temperature of interest. It is now simplified and shown that a single cross-temperature calibration was sufficient. The calibration step is now faster, covers a wider viscosity range, and is more accurate.

A second set of experiments compared the ultrasonic viscometer with a conventional shearing rheometer. First, a good agreement was found between conventional and ultrasonic viscosity measurement techniques for Newtonian-like fluids. This result validates the ultrasonic viscometer. Then, shear-thinning fluids were measured. The ultrasonic

viscosity was systematically lower than the rheometer viscosity. This finding is similar to previous works performed with several oscillating viscometers. These results suggest that the measurement conditions of the ultrasonic viscometer are similar to those of a high-shear viscometer.

Ultrasonic viscometry is a promising novel in-situ technique with many potential fields of application. To enable its use in industry, additional work is considered to better understand the impact of operating conditions on ultrasonic viscometry and to provide the relationship between frequency and shear rate.

CRedit authorship contribution statement

G. Peretti: Formal analysis, Investigation, Writing – original draft. **N. Bouscharain:** Investigation, Supervision, Writing – review & editing. **N. Dörr:** Funding acquisition, Supervision, Writing – review & editing. **F. Ville:** Funding acquisition, Supervision, Writing – review & editing. **R.S. Dwyer-Joyce:** Project administration, Supervision, Conceptualization, Funding acquisition, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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