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Measuring Newtonian viscosity from the phase of reflected ultrasonic shear wave

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Abstract

In this paper, an acoustic shear impedance model is employed to obtain a relation between the viscosity of a Newtonian fluid and phase characteristics of ultrasonic shear wave reflection from a solid–fluid interface. The phase and magnitude of the reflection coefficient can be decoupled in this model. The decoupling allows an independent relation between the acoustic shear impedance (viscosity–density product) and phase of the reflection coefficient. The model was experimentally verified for different fluid–solid combinations. Comparison of the results with the commonly used absolute reflection coefficient method demonstrates that phase measurement provides improved measurements.

Keywords: Ultrasound; Viscosity; Shear wave; Phase difference; Reflection

1. Introduction

Sound waves produce a thin Stokes layer at a solid–liquid interface. This Stokes layer depends on the kinematic viscosity–density product (also called the shear impedance) of the fluid. By determining the reflection coefficient of the plane wave reflected from a solid–liquid interface, the shear impedance of the liquid can be measured. If the density of the fluid is known or measured using some other technique, then the viscosity of the fluid can be obtained.

Mason [1,2] introduced the concept of non-invasive ultrasound viscometry, using the reflections from a quartz crystal, to determine the viscosity response of fluids. Since then several distinct ultrasound-based viscometric devices have been developed for non-invasive, on-line property and process monitoring [3–8].

Viscosity measurements have been carried out using ultrasound as well as other acoustical methods [3,9]. Specifically, ultrasound shear wave propagation at a solid–fluid interface has been used to characterize the viscosity of the fluid at the interface in many different ways. Parameters like amplitude change [10], phase change [11] and frequency shifts [12] have been correlated with the viscous and viscoelastic properties of the fluid.

The ultrasonic reflection coefficient depends on the mechanical properties of the solid and the fluid. For a well-characterized solid, the longitudinal wave reflection coefficient is a function of the longitudinal impedance (bulk modulus–density product) of the fluid. Similarly, the shear wave reflection depends on the shear impedance (shear modulus–density product) of the fluid. Shear modulus of a Newtonian fluid can be defined as the product of viscosity and circular frequency.

Viscosity affects both the amplitude and the phase of the incident ultrasound wave. High-resolution amplitude measurements are inherently problematic because of the material noise, the overlying electronic instrumentation noise and the errors introduced by limitations in the vertical resolution due to digitization. Phase measurements, on the other hand, can be measured more accurately due to the availability of high sampling rate digitizers and steady triggering mechanisms (to reduce jitter problems) available today. A method is presented in the following paragraphs that uses the phase of the reflected shear wave at a solid–fluid interface to measure the acoustic shear impedance (viscosity–density product) of the fluid.

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2. The acoustic shear impedance model

Consider a plane horizontally polarized shear wave incident (Fig. 1) on the solid–fluid interface at an incident angle (θ_s). The nomenclature for the symbols used in this paper is provided in Appendix A. Stress equality at the contact surface, between the Newtonian viscous fluid and the solid substrate, forms a basis for shear reflectance [2]. Using this equality, the complex reflection coefficient is obtained as

$$R = r\mathrm{e}^{\mathrm{i}(\pi-\phi)} = \frac{Z_l \cos\theta_s - Z_s \cos\theta_l}{Z_l \cos\theta_s + Z_s \cos\theta_l}. \quad (1)$$

The complex impedance of the fluid, in terms of its viscosity and density [11], is given as

$$Z_l = \sqrt{\mathrm{i}\rho\eta\omega}. \quad (2)$$

In Eq. (2), the medium is assumed to be a perfectly viscous fluid (purely Newtonian) and hence has only the imaginary term. Therefore, the elastic component of the shear modulus of the liquid media is neglected. In contrast, the impedance of a lossless elastic solid substrate is real and is given by

$$Z_s = \rho_s C_s. \quad (3)$$

Substituting the expressions for the impedances from Eqs. (2) and (3) into Eq. (1), we obtain a complex expression for the reflection coefficient R.

From the real part of the complex reflection coefficient, a direct relationship between the phase ϕ and the magnitude r of reflection coefficient is obtained as

$$1 - 2r^2(1 + 2\sin^2(\phi)) + r^4 = 0. \quad (4)$$

It must be noted that this relationship is independent of the material properties of the solid or the liquid and also of the angle of incidence.

The imaginary part of Eq. (1) gives a relation among viscosity, phase and magnitude of reflection coefficient:

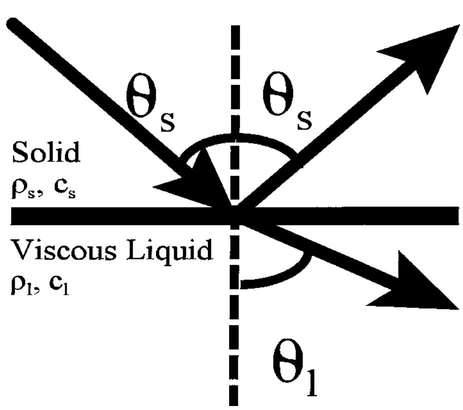

Fig. 1. Schematic of a plane polarized shear horizontal wave incident at a solid/fluid interface.

$$\eta = \frac{2\rho_s^2 c_s^2}{\pi\rho f} \frac{\cos^2(\theta_l)}{\cos^2(\theta_s)} \frac{r\sin\phi(1-r^2)}{(1+r^2+2r\cos\phi)^2}, \quad (5)$$

where Snell's law defines the relation between the angle of incidence (in the solid) and the angle of refraction in the liquid as

$$\theta_l = \sin^{-1}\left(\frac{c_l}{c_s}\sin\theta_s\right). \quad (6)$$

For normal incidence, $\theta_s = \theta_l = 0°$. The phase can be obtained from known reflection coefficient amplitude r by reducing Eq. (4) to

$$\phi(\omega) = \frac{1}{2}\cos^{-1}\left(1 - \frac{(1-r(\omega)^2)^2}{2r(\omega)^2}\right). \quad (7)$$

Similarly, the amplitude r can also be obtained from known phase using Eq. (4):

$$r(\omega) = \pm\left(1 - 2\sin^2(\phi(\omega)) \pm \sin(\phi(\omega))\sqrt{1+\sin^2(\phi(\omega))}\right)^{1/2}. \quad (8)$$

Note that the amplitude of the reflection coefficient is a positive real number. The reflection coefficient constitutes the amount of energy reflected back, and hence $r \leqslant 1$. Four roots are obtained for r as shown in Eq. (8), three of which are eliminated by the above arguments that r is bounded in (0,1].

Hence, it is clear that Eq. (4) allows for decoupling of the amplitude and the phase of the reflection coefficient. Once the phase $\phi(\omega)$ is measured at a particular frequency, $r(\omega)$ is calculated using Eq. (8). Then, the viscosity can be computed using Eq. (4). Since small changes in phase can be measured more accurately than amplitude [13], phase difference based method has the potential for increased sensitivity to the measurement of subtle viscosity or density variations.

3. Analysis of the model

From Eq. (5), it is evident that the sensitivity of the measurement depends on the angle of incidence of a shear wave at the solid–fluid interface. Fig. 2 illustrates this relationship for three fluids with different shear impedances. This result was obtained using the properties of Plexiglas (Table 1) as the solid. It is apparent from this result that the maximum sensitivity is obtained at normal incidence (i.e. when the angle is 0°) and the sensitivity decreases gradually from 0° to 30° and then at a more rapid rate beyond 30°. This was expected since the shear wave modes have maximum sensitivity to shear coupling at the interface when the particle motion is tangential to the interface. At higher angles, the mode conversion will lead to the generation of longitudinal

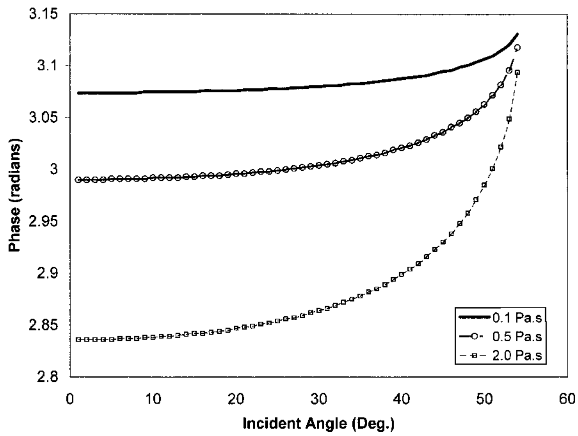

Fig. 2. Sensitivity of phase to the angle of incidence demonstrating that the normal incidence of shear wave is the optimum. Here, the solid was Plexiglas and the density of the fluid was 880 kg m^{-3} and longitudinal velocity was 1600 m s^{-1} and the frequency was 10 MHz.

Table 1
Measured properties of solids used as substrates

Substrate	Shear wave velocity (m s^{-1})	Density (kg m^{-3})
Plexiglas	1300	1200
Graphite-fine grained	1689	1728
Aluminum	3100	2710

mode into the fluid that is not influenced by the shear impedance of the fluid. In this paper, only case studies involving normal incidence of shear wave at the solid–liquid interface will be further discussed.

The acoustic property of the solid substrate plays an important role in determining the sensitivity and range of the acoustical impedance measurements. The reflection factor depends on the shear impedance of the solid. For instance, the higher the shear impedance of the solid, the smaller the range of angles with sensitivity. Sensitivity also increases with increasing incident wave frequency. In Fig. 3, the amplitude of the reflection coefficient is plotted as a function of fluid viscosity for a fixed fluid density (880 kg m^{-3}) for two different solid substrates (Plexiglas and fine grained graphite). Plexiglas has acoustic impedance closer to that of the calibration fluids compared with graphite. Graphite was considered because of its potential in high temperature applications. The sensitivity of the viscosity measurements is proportional to the slope of the viscosity–reflection coefficient amplitude curve. As the impedance match between the solid and fluid improves, the gradient becomes steeper. Hence, for Plexiglas, the sensitivity is very high at low viscosities but at the expense of a reduced dynamic range in viscosity measurement. The graphite shows a wider measurement range, but at reduced sensitivity.

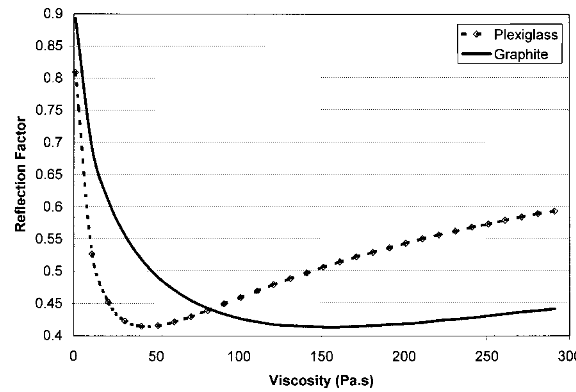

Fig. 3. Reflection factor for normal shear wave as a function of viscosity demonstrating the minima of 0.4142 for a fluid with a density of 880 kg m^{-3} and longitudinal velocity of 1600 m s^{-1} at a frequency of 10 MHz.

The impedance model does not consider the effect of relaxation processes in fluids, but the range of frequencies used in this study, and many common ultrasonic measurements, are well below this relaxation frequency. The molecular time scale for the relaxation phenomena for water, e.g. 10^{-13} s, thus, represents a relaxation frequency of 10^7 MHz. Also, as the viscosity of the fluid increases, this value will decrease. Hence, at very high frequencies and for highly viscous fluids, this may not be physically true [6,14]. However, for the case studies considered, the relaxation behavior was not anticipated.

An interesting phenomenon is the presence of a minima in the results presented in Fig. 3. The reflection coefficient amplitude increases at higher viscosities beyond this minima. Hence, the reflection coefficient response to frequency is not a monotonic decreasing function. The minima is present for all fluids, when the solid/liquid shear impedance ratio reaches a particular value. For all Newtonian liquids, this ratio is 2.41 and the minimum of the reflection coefficient is 0.4142 at this point. The range of measurement using the amplitude of the reflection coefficient is reduced by the presence of the minima. As shown in Fig. 3, for a frequency of 10 MHz, the range of evaluation of viscosity using Plexiglas as the solid substrate is approximately between 0 and 25 Pa s. This range will increase at lower frequencies as well as for materials, such as graphite and aluminum, which have higher shear impedance. A more detailed analysis on the behavior of the amplitude of the reflection and an interpretation using various models for viscous fluid cases has been discussed by Sheen et al. [15].

In Fig. 4, the phase (ϕ) of the reflection coefficient is plotted as a function of viscosity for the same solid–liquid combinations as in Fig. 3. It can be observed that the total phase change is within 0–2π range and that the decrease is monotonic, unlike the amplitude of the reflection coefficient r. This phenomenon is important in applications, such as monitoring the melting or

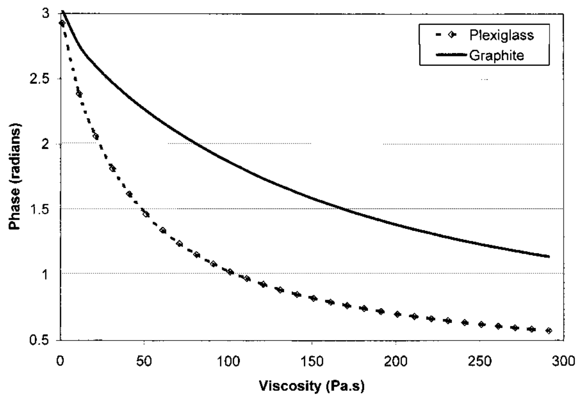

Fig. 4. The phase of the reflection factor for normal shear wave as a function of viscosity demonstrating the minima of 0.4142 for a fluid with density of 880 kg m^{-3} and longitudinal velocity of 1600 m s^{-1} at a frequency of 10 MHz.

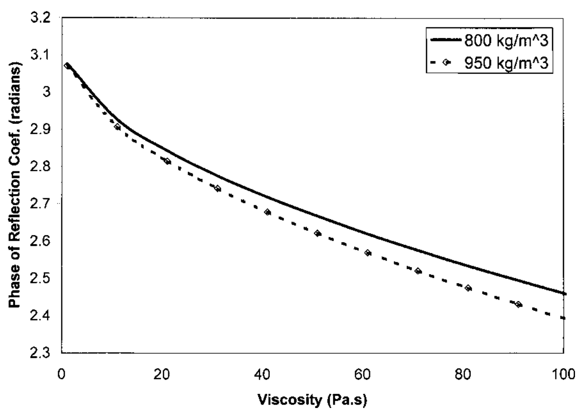

Fig. 5. Effect of change in density on the phase of the reflection coefficient as function of viscosity.

solidification processes where very large viscosity range may be encountered. Hence, the phase method may allow for a wider range of measurement when compared to the absolute reflection factor amplitude method.

The variation in density of a fluid with temperature is relatively insignificant compared with the viscosity variation with the same temperature change. In Fig. 5, the density of the fluid is varied from 800 to 950 kg m^{-3}, which is an extreme variation in fluid density for most cases. For this change in the density, the maximum reflection coefficient variation is close to 3%. An extreme case where this argument is not valid could be material phase change, such as near a solidification or a melting point.

4. Experimental setup

Three solid substrates (Plexiglas, graphite and aluminum) were used (Table 1) in conjunction with various NIST traceable viscosity standards (hydrocarbon base) procured from Cannon Instruments, PA (Table 2).

Table 2
Fluid properties of calibration standards obtained from Cannon, PA

Fluid type	Viscosity (η) at 25°C (Pa s)	Density (ρ) at 25°C (kg m^{-3})	Impedance ($\sqrt{\eta\rho}$) at 25°C (kg m^{-2} s$^{-1/2}$)
N100	0.1965	862.4	169.4616
S200	0.4009	873.2	350.06588
N350	0.7496	880.9	660.32264
S600	1.334	888.2	1184.8588

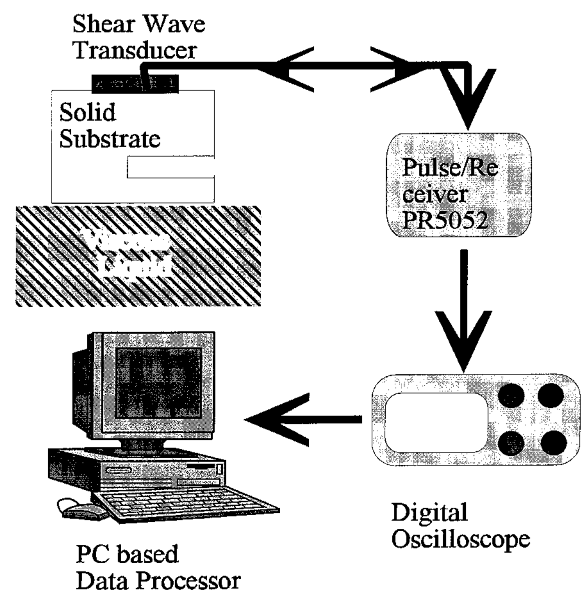

Fig. 6. Schematic of the experimental setup using pre-calibrated viscosity fluids from Cannon which is marked as Newtonian fluids.

A method of computing reflection coefficients suggested by Mak [16] was adopted. In this method, the reference signal, A_{ref}, is obtained using a stress free interface (air) as a reference. Then, the interface is loaded with the fluid and the signal reflected from the solid–fluid interface, A_{fluid}, is recorded.

Shear waves were generated and received (Fig. 6) by a broad band piezoelectric transducer (Panametrics) with a resonant frequency of 1 MHz. The signals were generated using a pulser/receiver (Panametrics 5052PR) and digitized by an 8 bit A/D converter (Tektronix TDS 320). The signals were recorded in a personal computer using a standard GPIB interface.

Pressure on the transducers must be uniformly maintained for all the experiments. As many measurements were to be conducted using the same shear transducer in contact with different solid substrates, a spring loaded clamp was designed to fit on the back of the substrate to hold the transducer in place and apply consistent uniform pressure during data collection. The piezoelectric transducers were coupled to the substrate by an epoxy resin (Epolite 9921A), without any hardener. This epoxy was preferred because of its property to harden under pressure and transmit shear waves. However, the hardening led to change in properties of the couplant with time and change in temperatures and pressure. To avoid the effects of the changing nature of couplant and temporal variations in the signals generated by the pulser/receiver, a notch was milled in the substrate to reflect a part of the shear waves. The notch gave a constant reference (C_{ref}, C_{fluid}) for either of the two measurements (A_{ref}, A_{fluid}). The reference signals (C_{ref}, C_{fluid}) were used to compensate for any variations in the input impulse and the generation mechanism of the transducer. At the frequency of operation, the reflection of the ultrasonic waves from the notch and the surface behaved similar to a flat surface reflector. Effect due to diffraction from the notch edge was not observed in the experiments. This can be explained because of the use of two reference signals, C_{ref} and C_{fluid} which offset any edge effects during the measurement. The fluids were maintained at a constant temperature in a water bath. A maximum fluid temperature variation was estimated at ±0.5°.

Phase measurements are sensitive to jitter in trigger time. Hence, a Stanford Research System DG35 time delay trigger was used to generate a steady trigger. The averaged jitter error in this measurement was very small compared to the pulser synchronization trigger and the threshold trigger in the oscilloscope.

Two methods for computing the viscosity of the fluid (with known density) are compared in this paper. The reference and compensation time domain (RF) signals were obtained first as $A_{ref}(t)$, and $C_{ref}(t)$ using air as the fluid. Then, signals were obtained, after introducing the viscosity standard fluids at the interface, as $A_{fluid}(t)$ and $C_{fluid}(t)$. Any DC offset was removed from these signals, through post-processing of the data. The digital signals from the reference notch and the solid–fluid interface were gated using a Boxcar window. Frequency spectrums of these signals were obtained using a standard fast Fourier transform algorithm. To stay in a region of good signal-to-noise ratio, a −6 dB bandwidth was considered for the viscosity spectrum. The phase difference spectrum was computed as

$$\phi(\omega) = \phi_{ref}(\omega) - \phi_{fluid}(\omega) + \psi_{fluid}(\omega) - \psi_{ref}(\omega). \quad (9)$$

The phase at the central frequency of the transducer (in this case $f = 1$ MHz) was used for computing the shear impedance of the fluid. Substituting the phase of the reflection coefficient (at 1 MHz) into Eq. (5) and using the expression given in Eq. (7), the viscosity–density product was computed. The densities of the calibration fluids are available from the manufacturer of the standards and were used to compute the viscosity from the acoustic shear impedance relation (using Eq. (2)).

The viscosity obtained from the phase method was compared with the viscosity obtained using time domain based absolute reflection factor based amplitude method [10]. Here, digital signals were processed by a standard Hilbert transform [17] to obtain the modulation envelope information of the signal. The absolute amplitude coefficient is obtained as

$$r = \left|\frac{B_{fluid}(t)}{B_{ref}(t)}\right|_{max}\left|\frac{D_{ref}(t)}{D_{fluid}(t)}\right|_{max}. \quad (10)$$

Neglecting the phase change ($\phi = 0$), Eq. (4) was used to compute viscosity.

5. Results and discussion

In this study, the difference in phases of the two Fourier transformed shear wave signals, obtained by processing reflected RF signals, is used to predict the shear impedance. The phase difference was computed using Eq. (10) at a frequency of 1 MHz, which was the central frequency of the transducer used in the case studies because the signal-to-noise ratio is maximum at this point.

The results of experiments conducted using the three substrates and the calibration standards are shown in Figs. 7–9. In the plots, the phase method is compared with the amplitude method and the solid line represents the expected results, assuming that the viscosity values provided by Cannon Instruments is accurate. It must be noted that the viscosity values measured using the phase and the amplitude methods are both obtained directly from the ultrasonic data using the model, and not through any calibration process.

In experiments with the Plexiglas substrate (Fig. 7), both the phase- and amplitude-based results match well with the viscosity values provided by the manufacturer at very low viscosities. With increase in the shear impedance, the viscosities measured using the phase method correlates better than the amplitude method. The probable reason for improved measurement of the

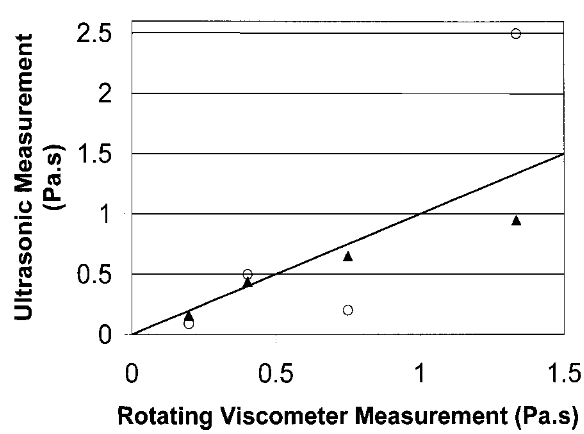

Fig. 7. Comparison between the ultrasonic measurement, using the phase method (▲) and the absolute reflection factor method (○), and the calibration rotating viscometer reading for Plexiglas substrate.

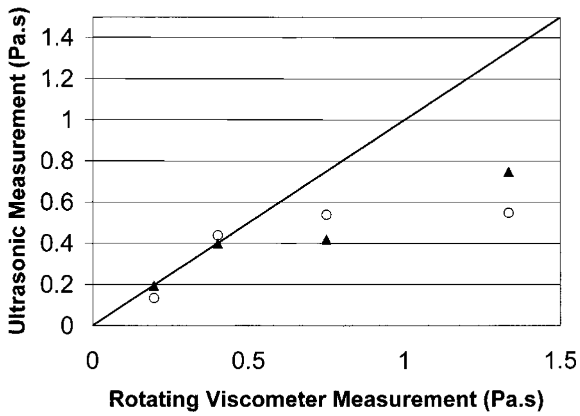

Fig. 8. Comparison between the ultrasonic measurement, using the phase method (▲) and the absolute reflection factor method (○), and the calibration rotating viscometer reading for graphite substrate.

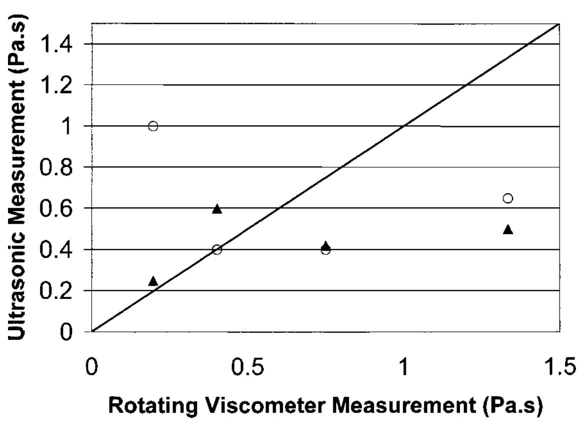

Fig. 9. Comparison between the ultrasonic measurement, using the phase method (▲) and the absolute reflection factor method (○), and the calibration rotating viscometer reading for aluminum substrate.

phase method are (a) the amplitude measurement may be averaging over the bandwidth of the transducer used, and (b) the imprecision in the amplitude measurements due to limitations in the vertical resolution during digitization (bits used by the analog to digital converter) as well as any noise present in the signal during measurement. Trigger jitter was a known issue with phase measurements and was resolved by using precise trigger electronics, as discussed before. In addition, the use of the phase difference between a reference notch and the interface signal (instead of absolute phase of the interface signal) further reduced any jitter effects.

For the graphite (Fig. 8) and the aluminum (Fig. 9) substrates, however, both the amplitude and the phase methods performed similarly. The accuracy of measurements was lower when compared to the Plexiglas results, particularly at the higher viscosities. This is attributed to the larger shear impedance mismatch between the solid and the fluid, which reduces the sensitivity of the measurements.

The discrepancy between the phase measured viscosity and the calibrated value was found to increase as the shear impedance of the fluid increased [18]. This has been confirmed by measurements, on the same fluid standards, using the amplitude method explained by Sheen et al. [15]. They also showed that the Newtonian fluid assumption, for computing the reflection coefficient at a solid–fluid model, is only valid for the fluid standards with impedance lower than 1 $kg\,m^{-2}\,s^{-1/2}$. For fluid standards with higher shear impedance values, they have also reported wide discrepancies. They demonstrated that a non-Newtonian model using a combination of Maxwell's, Voigt's, and power-law theories does provide a satisfactory approach for fluid standards with higher shear impedances. These fluid standards, labeled as Newtonian fluids by the manufacturer, were calibrated using conventional rotating type and capillary type viscometer which operate at very low shearing frequencies (10–300 Hz). In comparison, the ultrasonic measurements in this study were at frequencies exceeding 1 MHz. Hence, it may be probable that the fluid standards are indeed Newtonian at lower shearing rates, but become non-Newtonian at higher frequencies. Further work in this regard is a suggested topic of future research.

6. Summary

Phase information in the shear wave reflected from a solid–fluid interface was used to determine the shear impedance of viscous fluids based on an impedance model. The phase and the amplitude of the reflection coefficient decouple and their relationship is independent of the material properties. Also, the monotonic relationship between reflection factor and the phase allows for an increase in the range of viscosity measurements compared to the amplitude based methods. Various NIST traceable viscosity standards were used in experiments to verify the model. These experiments demonstrated that, for low impedance solids such as Plexiglas, the phase method improved the accuracy of the measurement, when compared to an amplitude method. Applications based on this method require a judicious choice of substrate properties for reliable measurements.

Acknowledgements

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Appendix A. Nomenclature

A_{ref}, A_{fluid} reflected shear wave signal received from contact surface for solid–air and solid–fluid interfaces
B_{ref}, B_{fluid} peak of Hilbert transformed shear wave signal envelope received from contact surface for solid–air and solid–fluid interfaces
C_{ref}, C_{fluid} received shear wave signal from compensation notch for solid–air and solid–fluid interfaces
D_{ref}, D_{fluid} peak of Hilbert transformed shear wave signal envelope received from compensation notch for solid–air and solid–fluid interfaces
R complex reflection coefficient
Z_l, Z_s acoustic shear impedance of fluid and solid $kg\,m^{-2}\,s^{-1}$
c_l, c_s acoustic shear velocity in fluid and solid in $m\,s^{-1}$
f frequency in Hz
r magnitude of the reflection coefficient
ψ_{ref}, ψ_{fluid} phase of the shear waves reflected from the compensation notch for solid–air and solid–fluid interfaces in radians
ϕ_{ref}, ϕ_{fluid} phase of the shear waves reflected from the contact surface for solid–air and solid–fluid interfaces in radians
η viscosity of the fluid in Pa s
θ_l, θ_s angle of transmission and reflection
ρ_l, ρ_s density of the fluid and solid in $kg\,m^{-3}$
ϕ phase of the reflection coefficient in radians
ω circular frequency ($2\pi f$)

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