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Experimental study of the complex reflection coefficient of shear waves from the solid-liquid interface

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Abstract

The determination of the reflection coefficient of shear waves reflected from a solid-liquid interface is an important method in order to study the viscoelastic properties of liquids at high frequency. The reflection coefficient is a complex number. While the magnitude measurement is relatively easy and precise, the phase measurement is very difficult due to its strong temperature dependence. For that reason, most authors choose a simplified method in order to obtain the viscoelastic properties of liquids from the measured coefficient. In this simplified method, inconsistent viscosity results are obtained because pure viscous behavior is assumed and the phase is not measured. This work deals with an effort to improve the experimental technique required to measure both the magnitude and phase of the reflection coefficient and it intends to report realistic values for oils in a wide range of viscosity (0.092 – 6.7 Pa.s). Moreover, a device calibration process is investigated in order to monitor the dynamic viscosity of the liquid.

Keywords: Reflection coefficient; shear waves; viscoelasticity; solid-liquid interface

1. Introduction

The complex reflection coefficient measurement of shear waves in the solid-liquid interface is a technique used to determine the viscoelastic properties of liquids at high frequency. This technique, introduced by Mason et al. [1] in the early 1949, provides information about the molecular relaxation phenomenon and has technological importance because it permits monitoring physicochemical properties, such as composition, viscosity [2] and elasticity [3]. The main advantages are the non-invasiveness and the small volume of liquid needed [4][5].

The complex reflection coefficient is obtained in the frequency domain by normalizing the solid-liquid sample echo with respect to the solid-air echo recorded in a previous experiment (calibration step). While magnitude is relatively easy to measure, the phase measurement is very difficult due to the strong temperature dependence on the wave propagation velocity. However, by normalizing the echo returned from the solid-liquid interface with another

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echo returning from a solid-solid interface or a discontinuity in the solid, as a notch, most non-desired effects can be reduced [6].

This work describes an experimental method developed in order to measure the reflection coefficient (magnitude and phase) and intends to report realistic values for five different liquids. These liquids, four automotive oils (SAE 40, 90, 140 and 250) and one edible oil (olive oil), covering a wide viscosity range (0.0926.7 Pa.s), were tested on three frequencies (1.0, 2.25 and 3.5 MHz). The reflection coefficient values obtained were compared to the theoretical values predicted by the Newtonian liquid model (zero elasticity model). Moreover, by fitting the magnitude of the reflection coefficient using a power-like function, it is explored an indirect method that permits the measurement of the dynamic viscosity of the liquids.

2. Theoretical background

The harmonic solution of the wave equation for an elastic medium with viscous losses provides the following relation between the liquid shear impedance (Z_L) and the complex shear modulus (G^*) [7]:

$$Z_L^* = \sqrt{\rho G^*}, \quad (1)$$

where ρ is the liquid density and $G^* = G' + jG''$, where G' is the storage modulus and G'' is the loss modulus. Assuming pure viscous behavior, the storage modulus must be null ($G' = 0$). As the loss modulus is related to the viscosity (η) by $G'' = \eta\omega$, the liquid shear impedance is [8, 9]:

$$Z_L^* = \sqrt{j\omega\rho\eta}. \quad (2)$$

When the shear wave propagates in a solid and strikes the solid-liquid interface, a small part of the energy is transmitted into the liquid and most of it is reflected. The relation between the incident (σ_i) and the reflected (σ_r) mechanical tension is called reflection coefficient (R^*):

$$R^* = \frac{\sigma_r}{\sigma_i} = re^{j(\pi-\theta)}, \quad (3)$$

where r and θ are the magnitude and phase shift of the reflection coefficient, respectively. The reflection coefficient is related to the acoustic impedance of the media by means the following expression [10]:

$$R^* = \frac{Z_L^* - Z_S}{Z_L^* + Z_S}, \quad (4)$$

where Z_S is the shear acoustic impedance of the solid medium. As the shear waves in the liquid are strongly attenuated Z_L^* is a complex quantity. On the other hand, as the attenuation in the solid is very small, when compared to the attenuation in the liquid, a real valued can be considered.

3. Experimental

The measurement cell developed for the ongoing research, as shown in Fig. 1 is composed of a piezoelectric ceramic transducer, a water buffer, an aluminum prism, a polymethyl-methacrylate (PMMA) buffer rod, and a sample chamber. One face of the PMMA buffer rod is in contact with the sample chamber, and the other is bonded to the aluminum prism opposite to its oblique face. The cell is immersed in water. The transducer emits a short longitudinal wave (pulsed excitation) that travels through the water buffer, reaches the prism oblique face and is converted into a shear wave. The angle of incidence of the longitudinal waves in the prism is 20° at 20°C . This angle is above the critical angle for the longitudinal waves in the water-aluminum interface. Consequently, there are only

shear waves propagating in the prism. However, as a consequence of the Poisson effect, a longitudinal wave is generated inside the prism, but its amplitude is small when compared to the shear wave amplitude. The shear wave inside the prism propagates and reaches the prism-PMMA interface (reference interface) with normal incidence. At this interface, the reflected wave ($n(t)$) returns to the receiver, and the transmitted wave travels through the PMMA buffer rod, is reflected at the PMMA-sample interface (measurement interface) and returns to the receiver ($a(t)$).

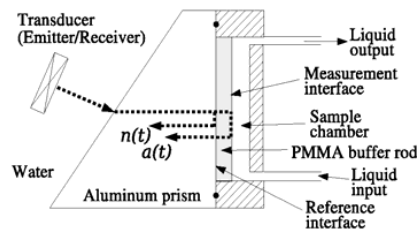


Fig.1 Schematic representation of the measurement cell.

The PMMA was a 5-mm-thick buffer rod, for the frequency range from 1 to 3.5 MHz. When frequencies are lower than 1 MHz there is superposition of the reflections from the reference and measurement interfaces, and, when frequencies are greater than 3.5 MHz there is high attenuation in the PMMA. The shear wave velocity in the PMMA is determined from the delays of the signals reflected at the reference and measurement interfaces, calculated by means of the correlation function and the Hilbert transform [11].

The complex reflection coefficient is obtained by referring, in the frequency domain, the signal from the PMMA-liquid interface to the one from the PMMA-air interface recorded in a previous experiment. When there is air in the sample chamber, total reflection is obtained and the magnitude and phase of the reflection coefficient are 1 and 180° , respectively. To eliminate the problems with equipment drift between these two measurements, the method employs normalization with respect to the reflected signal at the prism-PMMA interface. Then, the magnitude of the reflection coefficient is obtained by:

$$r = \frac{|A_{liq}(f_i)| |N_{air}(f_i)|}{|A_{air}(f_i)| |N_{liq}(f_i)|}, \quad (5)$$

where $N(f)$ and $A(f)$ denote the Fourier transforms of the pulses reflected from the reference and measurement interfaces ($n(t)$ and $a(t)$), respectively, and the phase shift is obtained by:

$$\theta = |\varphi_{liq}(f_i) - \psi_{liq}(f_i) - \varphi_{air}(f_i) + \psi_{air}(f_i)| \quad (6)$$

where $\varphi(f_i) = \arg[A(f_i)]$ and $\psi(f_i) = \arg[N(f_i)]$, and the subscripts *liq* and *air* make reference to the cases with and without liquid sample. f_i was the central frequency of the received pulse.

In this technique, the measurement of the phase must be carried out in controlled conditions, due to the strong temperature dependence. Experimental results with the measurement cell showed that small temperature changes, in the order of 0.1°C , can lead to phase variations of several degrees. For the viscosity and frequency ranges used in this work, the expected phase values are between 1 and 8 degrees. It means that the induced error in the measurement due to temperature instability can be very significant. Nevertheless, as temperature changes in a relatively long term, and the test is made in a small time interval, the errors caused by temperature instability can be considerably reduced (below the phase measurement precision).

Each test lasted five minutes and consisted of five acquisitions with air followed by other five acquisitions with the liquid sample. For each air-liquid pair, the reflection coefficient is calculated and the final result is the mean of

the five values obtained. Before each test, the cell and the 25 ml liquid sample were immersed in a temperature controlled bath for over an hour.

An acquisition system of 2 GHz sampling rate and 12 bit resolution in average mode (Agilent 54820A Oscilloscope) and a pulser/receiver (Panametrics 5072PR) were used. A schematic representation of the measurement setup is shown in Fig. 2. In order to obtain maximum amplitude of the reflected shear wave, the angle of incidence of the longitudinal wave in the water-prism interface can be adjusted.

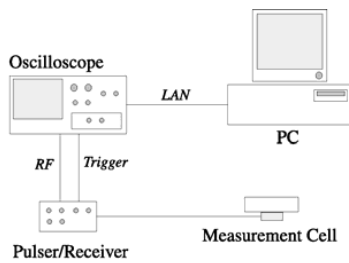


Fig.2 Schematic representation of the measurement setup.

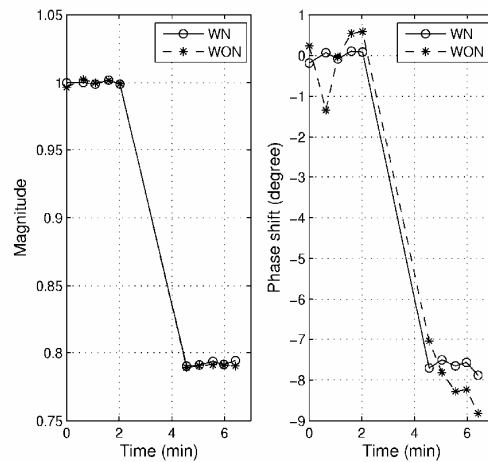


Fig.3 Effect of the normalization on the measured quantities (WO: with normalization, WON: without normalization).

Fig. 3 shows the magnitude and phase shift with SAE 250 at 3.5 MHz comparing the results obtained with (WN) and without (WON) normalization. The graph plots both the magnitude and phase shift of the reflection coefficient as a function of the acquisition time. The first five measurements were made with air in the sample chamber and the subsequent five measurements were made with liquid. Unlike the case of the magnitude, where a slight reduction in the standard deviation was obtained with normalization, a strong reduction in the standard deviation of the phase was obtained. These graphics were very useful because they permit to analyze the influence of temperature upon measured quantities.

Table 1. Physical properties of the tested oils.

Tested oil	ρ (kg/m ³)	η (Pa.s)	T (°C)
Olive Oil	881.6	0.092	15.0
SAE 40	857.2	0.3	22.5
SAE 90	871.2	0.6	22.5
SAE 140	876.6	1.22	22.5
SAE 250	913.8	6.7	22.5

It was tested an edible oil (olive oil) and four automotive oils (SAE 40, 90, 140 and 250) at three frequencies: 1.0, 2.25 and 3.5 MHz. The reported frequency is the central frequency of the pulse emitted by the ultrasonic transducer. However, received pulses present a reduction in the central frequency value due to attenuation. Table 1 shows the physical properties of the tested liquids. Density was measured by means of a pycnometer calibrated with distilled

water and viscosity was obtained by using the rotational viscometer Rheotest 2.1 (MLW, Mendingen, Germany) at the reported temperature (the same temperatures of the ultrasonic tests).

4. Results and discussion

Fig. 4 shows the magnitude of the reflection coefficient for all the oils as a function of frequency. The dashed line represents the values obtained with the Newtonian liquid model, i.e., by using (2) and (4) with the properties reported in Table 1. The Newtonian model fits the experimental data with reasonable success.

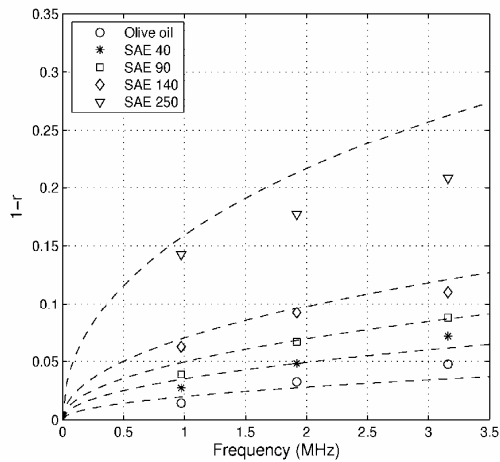


Fig.4 Magnitude of the reflection coefficient. Dashed lines represent the values predicted by the Newtonian liquid model

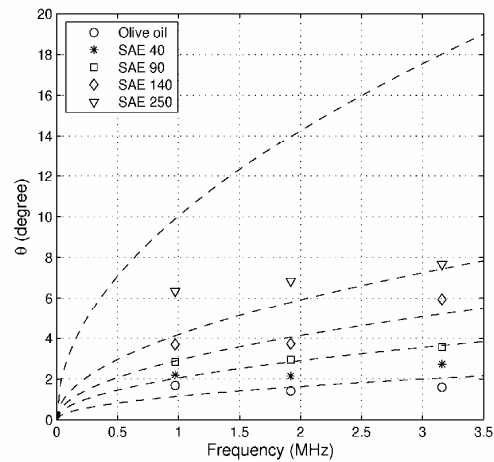


Fig.5 Phase shift of the reflection coefficient. Dashed lines represent the values predicted by the Newtonian liquid model.

Fig. 5 shows the experimental results of the phase shift as a function of frequency. There is a big difference between the measured values and those predicted by the Newtonian model.

As experimental difficulties induce a high degree of uncertainty in the phase measurement, four repetitions of the measurement with olive oil at 3.5 MHz and 15°C were carried out. Results are shown in Table 2, where the repeatability of the measurement is confirmed. The standard deviations of the magnitude and phase shift were approximately 0.1% and 8.0% of the mean value, respectively.

Table 2. Repeatability test with olive oil at 3.5 MHz and 15°C.

Experiment	r	θ (degree)
1	0.9851	1.8508
2	0.9859	1.6769
3	0.9869	1.5864
4	0.9875	1.8475
Result	0.9864 ± 0.0011	1.7404 ± 0.1309

Experimental values of the magnitude of the reflection coefficient were fitted using the following power-like function:

$$\eta_0 \rho = [P_1(1-r) + P_2]^{1/\alpha}, \quad (7)$$

where P_1 , P_2 and α are the fitting parameters. (7) is a modification of the power-law where an additional parameter (P_2) was included in order to improve the fitting. The obtained values of the parameter α were about 2.4 for all test frequencies. Thus the model parameter α was fixed at 2.4 and the fitting was carried once more. A least-square algorithm in Matlab was used. Fig. 6 shows the experimental results ($I-r$) plotted versus the viscosity-density product ($\eta\rho$), plotting fitted curves in dashed lines. Relative error between the experimental value of viscosity and the predicted value using (7), for all oils at all frequencies, were less than 11%. That shows the feasibility of using a simple calibration process with the measurement cell.

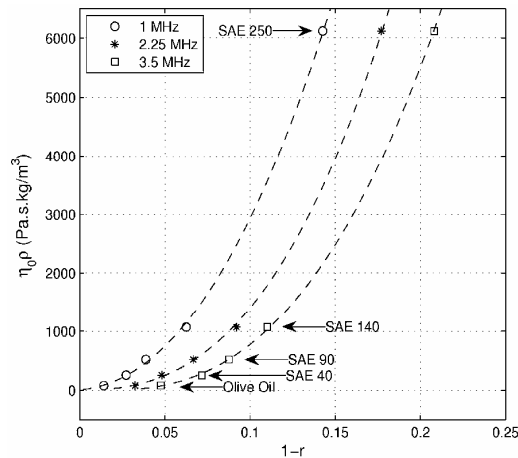


Fig.6 Fitting results ($\alpha=2.4$).

5. Conclusion

The experimental technique developed in this work allows the measurement of the reflection coefficient with reasonable success. As expected, the magnitude of the reflection coefficient can be measured more precisely than the phase.

Newtonian model cannot predict the reflection coefficient values, so more sophisticated models are required.

The on-line measurement of the dynamic viscosity (zero shear viscosity) has great importance in the industry. The calibration of the ultrasonic cell used in this work is an interesting possibility in order to fulfill this requirement.

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