Evaluation of a mode conversion dual-cell to measure the complex reflection coefficient at a solid-liquid interface

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Abstract.The measurement of the complex reflection coefficient, magnitude and phase, of shear waves reflecting from the solid-liquid interface is an interesting way to determine the viscosity of liquids at high frequency. The cell that uses mode conversion in order to generate the shear waves was developed by the authors in previous works. It was proven to be accurate, but the complex measurement process turns industrial applications infeasible, because a previous calibration measurement at the solid-air interface requires a slow cleaning process between each test. In this work, with the objective of improving the measurement cell, a new one with separated solid-air and solid-liquid interfaces was developed and its performance was evaluated. The cell uses two transducers that were multiplexed by means of a simple relay based circuit in order to use a single pulser/receiver. The solid material in contact with liquid samples was acrylic and the mode conversion happens at a solid-water interface in the oblique face of an aluminum prism. The interface defined by the acrylic buffer and the aluminum prism is used for normalization in order to reduce the undesired effects induced by temperature variations and instabilities in the electronic equipment. Glycerin at $22.5 \pm 0.1°C$ with dynamic viscosity of 1.07 Pa.s was tested at 1.0 MHz. Results showed difficulty in measuring the phase, strongly affected by the small temperature variations in the thermostatic bath. As for magnitude measurement, more precise results were obtained leading to coherent values of viscosity.

Keywords: ultrasound, shear waves, reflection coefficient, mode conversion, viscosity.

1. INTRODUCTION

The measurement of the shear wave reflection coefficient at a solid-liquid interface has been applied to study the visco-elastic properties of liquids at high frequency. The method was originally proposed by Mason et al. (Mason et al., 1949), and it was used by many authors to characterize lubricant oils (Buiochi, Adamowski and Furukawa, 1998, Franco et al., 2008), polymeric resins and solutions (Longin, Verdier, and Piau, 1998), polymeric films (Alig et al., 1997), liquid crystals (Mukai et al., 1997), slurries (Greenwood and Bamberger, 2002), and some products used in the food industry such as honey (Kulmyrzaev and McClements, 2000), xanthan gum (Saggin and Coupland, 2004) and edible oils (Saggin and Coupland, 2001, Franco et al., 2008). The method was also used for monitoring the resin cure process (Cohen-Tenoudji et al., 1987, Dixon and Lanyon, 2005) and the viscosity of molten materials (Balasubramaniam et al., 1999). The main advantage of the method is the possibility of carrying out on-line viscosity measurement, as required by the highly automatized industries these days (Buiochi et al., 2006).

The shear wave reflection coefficient can be expressed in terms of its magnitude and phase. Magnitude and phase are related, respectively, to amplitude and phase variations, when compared to a previous calibration measurement made in air. The phase highly depends on temperature, so it is difficult to be measured out of a laboratory. However, some authors have used a simplified method that uses only the magnitude of the reflection coefficient. This method is valid when the liquid behavior is predominantly Newtonian (Cohen-Tenoudji et al., 1987).

The necessity of a previous calibration measurement at the solid-air interface is the main disadvantage of the method, because a long cleaning process is required between each test with liquid. In this work, a novel design with an extra interface to obtain the calibration measurement was tested. For this purpose, a measurement cell with mode conversion, developed by the authors in a previous work (Franco et al., 2008), was duplicated: two transducers, working in pulse-echo mode, were multiplexed through a relay-based system controlled by the same computer used for the signals acquisition.

2. THEORETICAL BACKGROUND

The complex reflection coefficient (R^*) of plane shear waves striking the solid-liquid interface is defined in terms of the acoustic impedance of the media, as follows (Kino, 1987):

$$R^* = \frac{Z_L^* - Z_S}{Z_L^* + Z_S} = re^{j(\pi-\theta)}, \tag{1}$$

where Z_L^* and Z_S are the shear acoustic impedance of the liquid and the solid, respectively, r and θ are the magnitude and phase shift of the reflection coefficient, respectively, and $j = \sqrt{-1}$ is the imaginary unit. The acoustic impedance in the solid is considered a real value, because its attenuation is very small when compared with the attenuation in the liquid.

$Z_S = \rho_S c_S$ where ρ_S and c_S are the density and the wave propagation velocity, respectively.

The solution of the 1-D wave equation in a simultaneously elastic and viscous medium provides the following relation for the shear impedance in liquids (Whorlow, 1992):

$$Z_L^* = \sqrt{\rho_L G^*}, \qquad (2)$$

where ρ_L is the liquid density and G^* is the complex shear modulus, defined as $G^* = G' + jG''$. G' is the storage modulus and represents the elastic part of the stress in phase with the strain. $G'' = \omega\eta$ is the loss modulus and represents the viscous part of the stress in quadrature with the strain, where η is the viscosity and ω the angular frequency (Whorlow, 1992). Assuming a Newtonian medium, storage modulus must be zero and Eq. (2) is reduced to (Dixon and Lanyon, 2005):

$$Z_L^* = \sqrt{j\omega\rho_L\eta}. \qquad (3)$$

Substituting Eqs. (2) and (3) into Eq. (1), a relationship for R^* is obtained. From the imaginary part, an expression for viscosity is obtained as a function of r, θ and the known physical properties of the media (Shah and Balasubramaniam, 2000):

$$\eta = \frac{G''}{\omega} = \frac{Z_S^2}{\rho_L\omega}\left[\frac{4r(1-r^2)\sin\theta}{(1+r^2+2r\cos\theta)^2}\right]. \qquad (4)$$

From the real part, a direct relation between r and θ is obtained:

$$1 - 2r^2(1 + 2\sin^2\theta) + r^4 = 0. \qquad (5)$$

This relationship is independent of the material properties of the solids or the liquids (Dixon and Lanyon, 2005). Manipulating the equation above, an expression for phase shift as a function of the magnitude can be obtained:

$$\theta(r) = \frac{1}{2}\cos^{-1}\left[1 - \frac{(1-r^2)^2}{2r^2}\right]. \qquad (6)$$

Finally, the substitution of Eq. (6) into Eq. (4) leads to a relationship that provides the liquid viscosity using only the magnitude of the reflection coefficient:

$$\eta = \frac{2Z_S^2}{\rho_L\omega}\left[\frac{1-r^2}{1+r^2+\sqrt{r^2(6-r^2)-1}}\right]^2. \qquad (7)$$
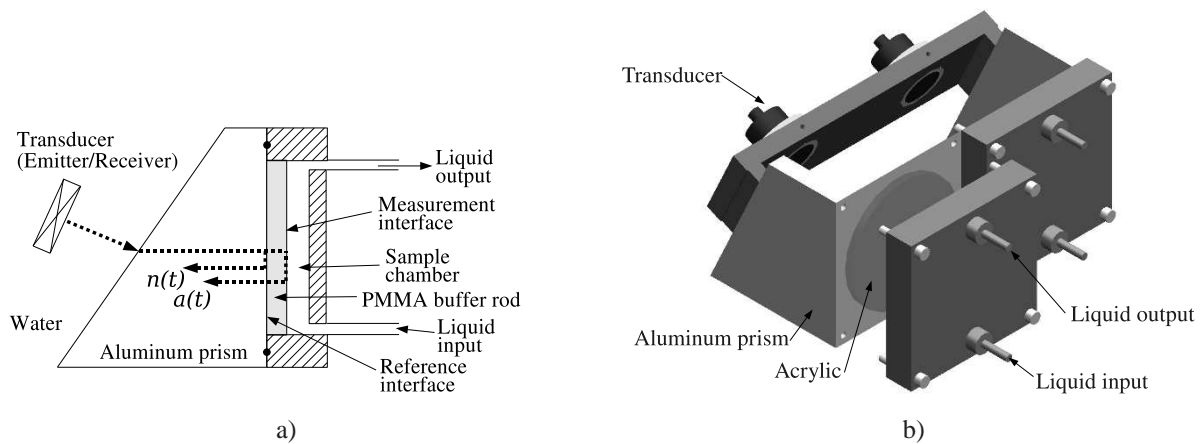
3. EXPERIMENTAL

3.1 Measurement cell

Figure 1. a) schematic representation and b) CAD image of the measurement dual-cell

The measurement cell uses mode conversion in order to generate the shear waves. This device, initially proposed by Buiochi et al. (Buiochi, Adamowski and Furukawa, 1998), is more accurate, because the undesired longitudinal waves generated by the shear transducer are reduced. However, others problems arise: the path through which the waves travel is longer and the operating temperature range is narrower. A schematic diagram of the measurement cell is shown in Fig. 1a. It 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 that travels through the water buffer, reaches the prism oblique face and is converted into a shear wave. The shear wave propagates through the prism 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 ($a(t)$). The PMMA was a 5.73-mm-thick buffer rod.

In order to obtain the calibration measurement using air in an extra interface, the cell described above was duplicated. The image of the new device is shown in Fig. 1b. The two transducers were multiplexed by means of a simple relay-based circuit controlled by the computer via serial port. An acquisition system at 571.5 MHz sampling rate and vertical resolution of 12 bits in averaged mode (Agilent DSO-6052A Oscilloscope) and a pulser/receiver (Panametrics 5072PR) were used. The acquisition and analysis of the signal were made using Matlab. The viscometer used to measure the low frequency viscosity was the Rheotest 2.1 by MLW (Germany).

3.2 Experimental determination of the reflection coefficient

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. In the PMMA-air interface case, a total reflection is obtained, i.e., the magnitude and phase shift of the reflection coefficient are 1 and 0° (see Eq. (1)), 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:

$$r = \frac{|A_{liq}(f_c)||N_{liq}(f_c)|^{-1}}{|A_{air}(f_c)||N_{air}(f_c)|^{-1}}, \tag{8}$$

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 f_c is a single frequency, usually the center frequency of the pulse returned from the solid-liquid interface. The phase shift is obtained by:

$$\theta = |\varphi_{liq}(f_i) - \psi_{liq}(f_i) - \varphi_{air}(f_i) + \psi_{air}(f_i)| \tag{9}$$

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 interfaces with and without liquid sample, respectively.

3.3 Methodology

The water temperature in the bath, containing the dual-cell, was monitored all time and its value remained at $22.5 \pm 0.1°$C. A previous calibration process was made by means of a measurement with air in both interfaces. Results of the normalized amplitudes, $|A(f_c)||N(f_c)|^{-1}$, and phases, $|\phi(f_c) - \psi(f_c)|$, for each interface as a function of the acquisition time are shown in Fig. 2a and 2b, respectively. A slight difference in the amplitudes of the signals returned from the interfaces was observed, in contrast to the big difference in the phase. This result was expected due to the small geometrical differences in the fabrication of the device. From these results, two compensation values can be obtained, one for the amplitude and another for the phase. The temporal mean of the amplitude and phase values from each interface were used.
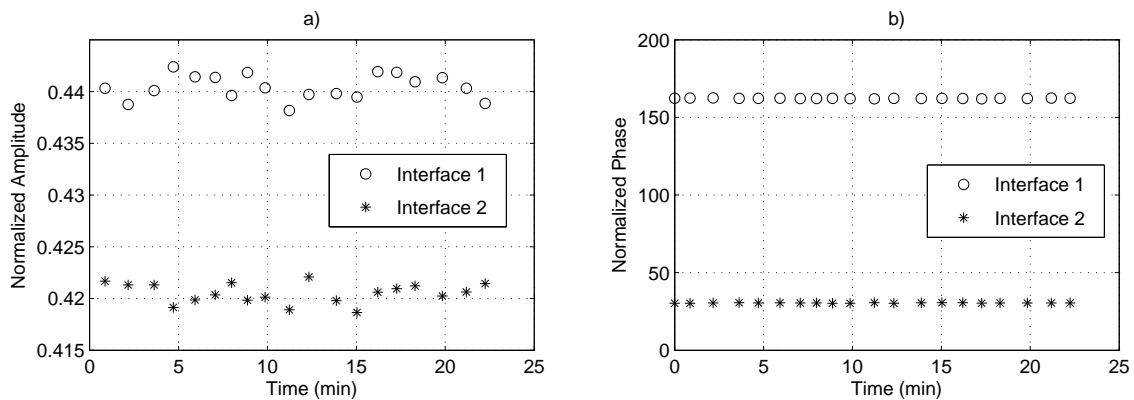

Figure 2. a) normalized amplitude and b) normalized phase in degrees obtained in the first calibration test

In order to test the stability of the measured quantities, two new measurements with air in both interfaces were made approximately 2 hours later. Then the magnitude and phase shift of the reflection coefficient were calculated using the

compensation values provided by the first test (Fig. 2). In these conditions, the magnitude and phase values must be close to 1 and 0, respectively. Results are shown in Fig 3., where more stability was obtained in the magnitude values, when compared to the phase shift values. The mean magnitude values of the tests were $\tilde{r}_1 = 1.0000 \pm 0.0037$ and $\tilde{r}_2 = 0.9997 \pm 0.0039$, and the mean phase shift values were $\tilde{f}_1 = 0.3281 \pm 0.1840$ and $\tilde{f}_2 = 0.3392 \pm 0.1538$.
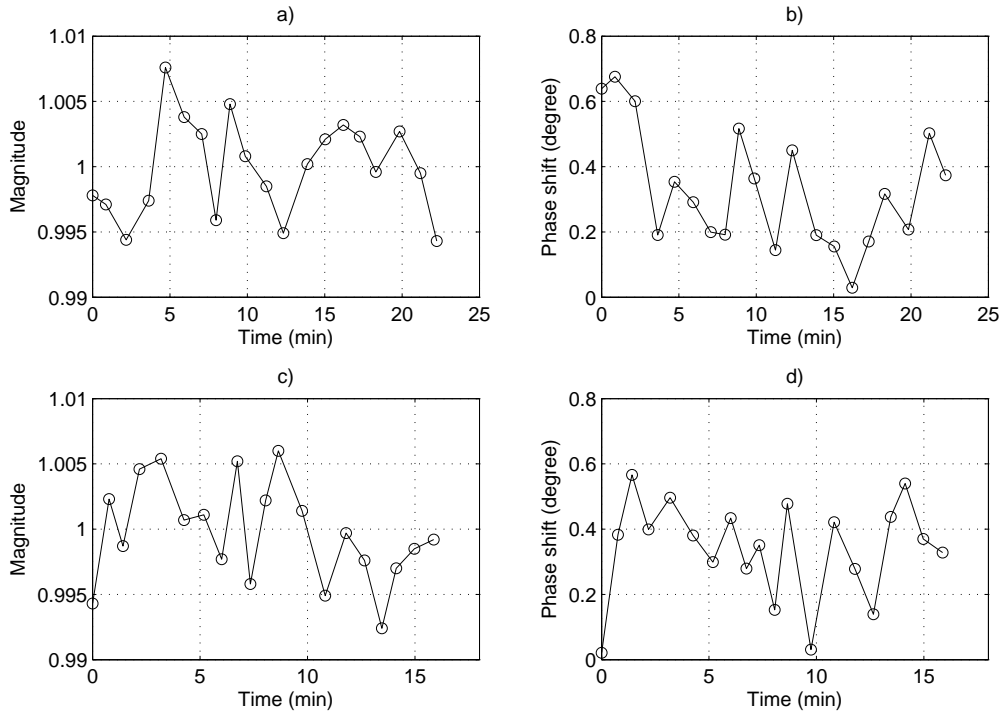

Figure 3. a) schematic representation and b) CAD image of the measurement cell

Above obtained results show the difficulty on the phase shift measurement, that leads to a high error in the viscosity measurement. In this case, the method that uses only the magnitude is a better choice. The simplified method can be used under Newtonian behavior. According to some authors (Harrison and Barlow, 1981, Kulmyrzaev and McClements, 2000) there is a simple relation between viscosity (η) and the Maxwell relaxation (τ_m) time for many liquids: τ_m(in ns) $= \eta$(in Pa.s). Then, as the viscosity of the glycerin at such temperature is 1.07 Pa.s, the Maxwel relaxation time is approximately 1 ns. As the working frequency is 1.0 MHz, the period of the wave ($1/f$) is approximately 1000 times the relaxation time. That should be enough to assure Newtonian behavior. Therefore, the viscosity was obtained using Eq. (7).

Two measurements were made with glycerin in one interface and air in the other. The first measurement was made injecting the liquid in the sample chamber 5 minutes after the acquisition had begun. That was made to check the variation of the measurement quantities in the presence of the liquid. The liquid was maintained in the chamber and, one hour later, the second measurement was made.

4. RESULTS AND DISCUSSION

Figure 4a shows the results of the reflection coefficient magnitude obtained with glycerin at $22.5 \pm 0.1°$. In the first measurement, the magnitude is very close to 1 at the beginning of the acquisition and then, when liquid is injected in the sample chamber, it drops to $\tilde{r} = 0.927$. In the second measurement, a very similar value of $\tilde{r} = 0.928$ was obtained. For the first measurement, $\tilde{r}$ is the mean of the last 14 acquisition and for the second measurement, $\tilde{r}$ is the mean of all acquisitions.

Figure 4b shows the viscosity results obtained in both measurements compared to the viscosity value obtained with the rotational viscometer (solid line). In the first test, it can be seen how the viscosity value rises from a value very close to 0 to $\tilde{\eta} = 1.04 \pm 0.08$, where the mean value was calculated with the last 14 acquisitions. In the second test, the mean value, using all acquisitions, was $\tilde{\eta} = 1.03 \pm 0.08$. Those results are in good agreement with the $\eta = 1.07$ Pa.s obtained with the rotational viscometer, and the error percentage is less than 4%.

In a new test, carried out with a temperature $1.5°$C higher, the initial calibration was lost. Consequently, it is required a new calibration process when the working temperature changes. This problem lies in the mode conversion approach used in this work, that causes the cell is suitable for use in a narrow range of temperature. That happens because the refraction angle of the mode conversion is a function of temperature. This problem must be smaller in devices that use directly shear
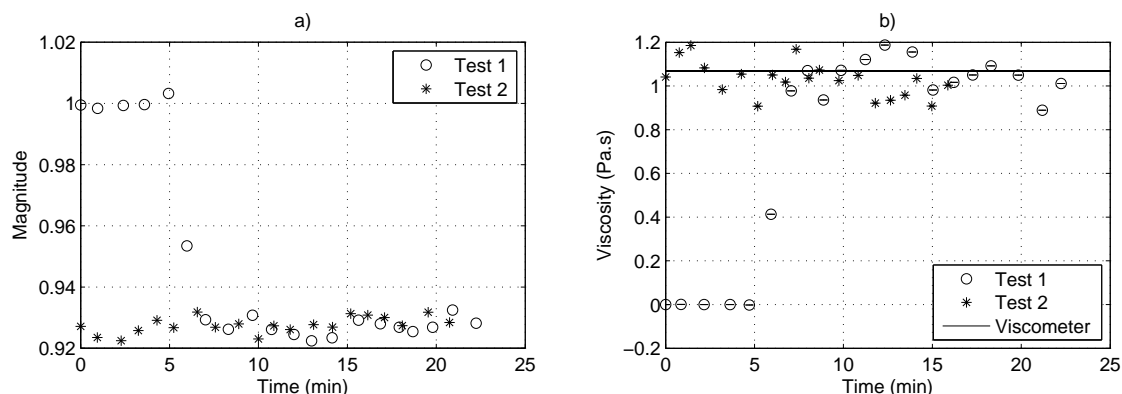
Figure 4. a) magnitude of the reflection coefficient and b) viscosity measured in the tests with glycerin

transducers.

5. CONCLUSIONS

A dual measurement cell with mode conversion was developed and its functionality was evaluated using glycerin. As expected, the magnitudes and phase shifts reflected from the two interfaces were different. Therefore, a previous calibration process with air in both interfaces was necessary. The tests were made in the temperature range between 22.4 and 22.6°C.

The instable behavior of the phase shift was verified; consequently, the simplified method that uses only the magnitude of the reflection coefficient is a better approach.

The developed dual-cell measured the glycerin viscosity successfully during the acquisition time. The relative error of the mean value, when compared to the value obtained with the rotational viscometer, was very small, showing the suitability of the method.

In a last test carried out at a 1.5°C-higher working temperature, the initial calibration could not be used. Then, a more detailed study of the cell behavior as a function of temperature is required.

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