Measurements of the Material Properties of Liquids Using Normal Acoustic Plate Waves

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Abstract
The new density/viscosity sensor has been described. The sensor has used the normal acoustic plate waves, namely, the lowest symmetrical Lamb and horizontal-polarized waves generated and received with 2 wedge transducers. The theory describing connection of the sensor output and the liquid parameter has been discussed.

Keywords: Ultrasonic measurement, symmetrical Lamb plate wave, horizontal-polarized normal plate wave, liquid density, viscosity.

1. Introduction

The demand to control the different technological processes leads to an extensive need to measure physical and chemical parameters of liquids in-line. Thereby measurements have to be often carried out in harsh environments. In these cases the use of ultrasonic measurement technologies has found advantageous applications to determine such important parameters of technological liquid as the compressibility, the density $\rho$, and shear viscosity $\eta_s$, since they can be implemented without direct contact between a sensor and investigated mediums. However there are no ultrasonic techniques to measure directly one of called parameters. The results of ultrasonic measurements are the longitudinal $Z^L_i = \rho c^L_i$ or shear $Z^L_s = (1 + i) \sqrt{\rho \omega \eta_s / 2}$ impedances ($\omega$ is the radial frequency, $i = \sqrt{-1}$) and the ultrasonic velocity $c_i$ of liquids. Hence, in order to find a numerical value the compressibility, the density $\rho$ or shear viscosity $\eta_s$ in-line, it is necessary to measure all ultrasonic characteristics of liquids $Z^L_i, Z^L_s, c^L_i$ at the same time. In other words, measurement methods, which can be applied for the development of the devices of technological process control, have to be reciprocally compatible and to realize a high accuracy of measurement. Ultrasonic control methods based on measurements of the propagation velocity of acoustical waves or the attenuation coefficient have been good studied and repeatedly discussed [1]. The advantages and disadvantages of conventional acoustical impedance measurements have been good considered [1-5]. They have based on measurements of either the wave reflection coefficient from the solid-liquid interface or the velocity and/or the attenuation of the fundamental longitudinal/torsional modes in cylindrical/ rectangular waveguides immersed in a liquid. This discussion has showed that the compatibility and the verification of the necessary accuracy and sensibility of different methods have met definite difficulties. So, the wedge-fluid interface has weakly influenced the change of the reflection coefficient since the numerical values of both impedances are typically different. As for measurements of the velocity and/or the attenuation of the fundamental longitudinal and torsional modes in cylindrical/ rectangular waveguides, they have permitted to realize higher sensible and precise measurements. The interaction between the wave and the liquid takes place along a large
waveguide part contacting with the liquid and embedded by wave. Frequency range of such measurements is limited by frequencies of 0.1—0.5MHz. The alternative approach has been suggested in [6]. The waveguide has presented a strip delay line in the form of a rectangular plate. Piezoelectric shear transducer has been mounted on one end surface of the waveguide. It has generated and received a shear wave that has propagated along the strip length. Displacements caused with the wave have been parallel to the strip surface. If a part of the waveguide has been immersed in a liquid, the wave has been attenuated and the propagation velocity or phase velocity has been decreased slightly. In general this approach is analogical to the torsional mode method but its advantage consists in the possibility to increase measurement frequencies essentially. However, placing of the transducer on the strip end surface has very limited the sensibility of the method. Really, it can be showed [6] that the attenuation of ultrasonic signal amplitude due the shear liquid viscosity is $\propto \exp(l/d)$, where $l$ and $d$ are the length of the slip immersed in the liquid and the slip thickness. As the typical cross-sizes of piezoelectrical plate are 3—4mm, it is very difficult to realize the relation $l/d$ above 4—5.

2. The sensor’ construction

The aim of the research presented here has been to develop a sensor in which the interaction of the ultrasonic wave with an investigated liquid leads to large amplitude change of a received wave. Its physical base has been the use of normal acoustic plate waves, namely, the lowest symmetrical Lamb or horizontal-polarized waves. These waves have been generated and have received with two wedge transducers (see figure 1). They have propagated in the waveguide and contact with a liquid along the immersed strip part analogously to the torsion wave case. The efficiency of this interaction has been higher considerably thanks to more high frequency of used. The waveguide thickness can be chosen by a user, to obtain a desirable sensibility of measurements.

![Figure 1. The sensor’ construction](image)

1- the container for a fluid  
2-the plate, $R \gg \lambda$,  
$\lambda$ - the wave length,  
$R$ - the bend radius  
3and 4 – the wedge transducers
Each waveguide/plate wave has been generated and received with the own wedge transducer after the reflection from the free plate end. Electrical channels of waves (generators and amplifies the electrical impulses) have been independent. The informative source is the change of the wave attenuation and time propagation due to the interaction between ultrasonic wave and investigated fluid in comparison with the free waveguide. These changes have been calculated from the measured ratio \( r \) of two signals. One of them has propagated through the plate interacting with the liquid on the part way \( l \). Another signal is as the reference signal and presents the ultrasonic signal reflected on the interface between the wedge and the waveguide toward the piezoelectric plate (figure 1). The sensor functional capability has been examined with measurements in the glycerol-water solution. The measurements have been performed in the frequency range of 2–5MHz, using the aluminium waveguide whose thickness and the length has been 0.2 and 200mm, correspondently \((l/d=1000)\). Except the generator and the amplifier there is the calibrated attenuator that has been joined between the transducer and the receiver entrance. The signal amplitude has been maintained constant with this attenuator. The accuracy of attenuator gradation has been 1.0 ± 0.1dB. After that the signal has been observed with the digital oscilloscope.

3. Experiments

The measurement possibilities of the ultrasonic sensor have mainly depended on the accuracy measurements of ultrasonic attenuation coefficient (the amplitude change on the length unity) and/or the propagation velocity due to the waveguide-liquid interface. For this aim it is necessary to investigate experimentally and theoretically the influence of liquid physical properties on the signal amplitude and propagation time. In our experiments typical measured signal changes due to waveguide immersing in a liquid has been 10–30dB relative to the free waveguide when the length of the waveguide-liquid interface has been 40–100mm. It has depended on such liquid properties as shear viscosity, the density and the sound velocity. The error of the amplitude measurements can be estimated \( \pm 5 \% \). The both parameters of the signal propagation have depended on diffraction effects and losses in the waveguide as well. The influences of these contributions on the experimental results have been investigated with the following measurements for both the horizontal-polarized waves and the Lamb waves. At first a short waveguide part has been immersed in a liquid. The waveguide rest length has been in the air. The amplitude and propagation time of the signal reflected from the waveguide end had been measured. Then the length of immersed waveguide part has been consecutively increased and the measurements have been repeated. Consequently, the way that has crossed the observed ultrasonic signal has always remained constant and has changed the length of the liquid-waveguide interface. As the contact liquid has been used glycerol-water mixtures in which glycerol concentrations have been varied in the range of 0 - 100%. It has given the possibility to change the physical properties of the contacting liquid in a wide range, especially, the viscosity (from 1cP up to 1P) and the sound velocity (up to 1.5 time). The length of immersed waveguide parts have been choose 40–100mm in the dependency of the liquid influence grade on the signal attenuation. These experiments have shown that changes of the signal amplitude and the propagation time due the influence of the waveguide-liquid interface and calculated on one length unity have not depended on the interface length for the both wave types. In other words, in the described geometry the influence the waveguide-liquid interface on diffraction effects has not improved the above-called measurement errors for the both waves. The new sensor has been ensured the high accuracy that has been only limited by device error of the amplitude measurements. For
example, the amplitude decrease due to the waveguide-water interface has amounted to 20–30 dB in comparison to the air. This result has been good agreed with the measurements of the propagation velocity changes calculated from the propagation time changes. The sound velocity/the wave length changes because of the liquid contact have not improved 0.1–0.4% relatively to the air for all investigated mixture up to the concentration of 100% glycerol. Gotten experimental results have shown that the influence of the diffraction and the energy losses in the waveguide on the attenuation coefficient is to be excepted if to measure the ratio of signal amplitude with the waveguide-liquid interface to the amplitude with the air interface.

4. Theoretical estimations and discussion

In order to calculate the fluid parameters on the base of the sensor’ data, the dependence of the ratio \( r \) on the fluid parameters has been evaluated theoretically for both wave types. It is known [7] that the symmetrical horizontal-polarized zero-order wave \( s_{sh}^0 \) in the solid free plate presents the pure shear wave. In this case mechanical deformations generated by the wave on the waveguide surface are pure tangential. If the surface of the waveguide has an acoustical contact with a liquid, the boundary conditions change in comparison with the free waveguide. The tangential component of the stress tensor of the waveguide should be equal to that of the liquid and the tangential component of the particle velocity should also be continuous.

The shear impedance of liquids \( Z_s^L \) is much less than the shear impedance of solid \( Z_s^W \) for the investigated frequency range (for example, if the shear viscosity is \( \eta_s = 10 \text{P} \) and the density is \( \rho_s = 1 \text{g/cm}^3 \), the relation is \( Z_s^L / Z_s^W \approx 1.5\% \) and the frequency is \( f = 2 \text{MHz} \). Furthermore, one can propose that the solution of the wave equation for the wave propagated in the waveguide with the liquid interface should be written down analogously to one for the free waveguide.

Then one can look for the solution in this case as the attenuating shear wave

\[
 u_y^w = A_y^w \cos(\beta z) \exp(i(\gamma x - \omega t)),
\]

where \( u_y^w \) is the \( y \)-component of the particle velocity, \( x \) is the propagation direction, \( z \) is the perpendicular axis to the waveguide surface, \( \beta \) and \( \gamma \) are the propagation parameters. They present complex numbers connected by equation [7]:

\[
 \beta^2 + \gamma^2 = \omega^2 / c_s^2.
\]

The expression of the tangential component of the stress tensor for the horizontal-polarized wave in the solid is given with the equation

\[
 \sigma_{yz}^w(z = \pm b) = \mu \frac{\partial u_y^w}{\partial z}(z = \pm b).
\]

Substituting the expression (1) in (3), we find

\[
 \sigma_{yz}^w(z = b) = \mu \frac{\partial u_y^w}{\partial z}(z = b) = -A_y^w \mu \beta \sin(\beta b) \exp(i(\gamma x - \omega t)).
\]
The expressions of the tangential components of the stress tensor for the horizontal-polarized wave in liquid are given by
\[ \sigma_{x}^L = \frac{\partial u_{x}^L}{\partial z}, \sigma_{y}^L = \frac{\partial u_{y}^L}{\partial x}. \] (5)

In order to assure the fulfillment of the boundary condition for every point of the surface on the wave propagation way, the horizontal-polarized wave in the waveguide has to be accompany by the wave in the liquid. This wave is written down in form [8]:
\[ u_{y}^L = A_{y}^L \exp(-z/\delta) \exp[i(\gamma x - \omega t)], \delta = \sqrt{2\eta_s / \omega \rho^L}. \] (6)

Numerical estimates show that the condition \(1/\delta >\gamma\) is good fulfilled for liquids whose shear viscosity does not exceed \(10^{-100}\text{P}\). Consequently, \(\sigma_{x}^L >> \sigma_{y}^L\) and the influence of the waveguide-liquid interface has been expressed by the force in the contact plane. In the first approximation \(\sigma_{x}^L\) coincides with the frictional force calculated in [9] for the case, when the surface of the half-space has the contact with the viscous liquid and it oscillates in its plane. If it is proposed that a contacting liquid is Newtonian then the expression of the frictional force has followed
\[ \sigma_{fric} = \left[ \frac{\omega \eta_s \rho^L}{2}(i-1)u_{x}^L; u_{y}^L = A_{y}^L \exp\left[\frac{z-b}{\delta}\right] + i(\gamma x - \frac{z-b}{\delta} - \omega t)\right]. \] (7)

If the tangential components of the particle velocities should be equal on the two sides of the interface, one can find the dispersion relation
\[ -\beta \tanh(\beta b) = \frac{\omega}{\mu} \sqrt{\frac{\omega \eta_s \rho^L}{2}} (1+i). \] (8)

The condition \(\frac{\omega}{c_w} Z_i^L / Z_i^w << 1\) is good fulfilled up to \(\eta_s \approx 10^{-100}\text{P}\) in the frequency range 1–5MHz. Therefore, limiting only the linear members, the left part of the dispersion relation can be written down:
\[ \gamma^2 = \frac{\omega^2}{(c_w^L)^2} (1 + \frac{1+i}{\rho \omega b} \sqrt{\frac{\omega \eta_s \rho^L}{2}}), \] (9)
or
\[ \text{Re} \gamma \approx \frac{\omega}{c_w^L} \left(1 + \frac{1}{2 \rho \omega b} \sqrt{\frac{\omega \rho^L \eta_s}{2}}\right); \quad \text{Im} \gamma = \frac{\sqrt{\omega \eta_s \rho^L}}{2 \sqrt{2} \rho^w c_w^w b}. \] (10)

The comparison of values of the mixture magnitude \((\sqrt{\rho^L \eta_s})_{exp}\) calculated by the relation (10), using the measured values of the attenuation coefficient, with the table values [10, 11] of this magnitude \((\sqrt{\rho^L \eta_s})_{tab}\) is presented in the figure 2. The shear sound velocity and the density of the waveguide have been measured in addition.
This comparison has confirmed the results of Shah et al [12] and Sheen et al [4] that the Newtonian liquid assumption is in force if liquid shear viscosity does not exceed a few tens of centipoises. This result has to be taken into consideration when the magnitude $\sqrt{\rho'\eta_S}$ is computed with the use of the relation (10) from ultrasonic measurements, having the industrial control a technological process as the measurement aim.

It is clear that the magnitude $\sqrt{\rho'\eta_S}$ is no liquid parameter. Therefore, its numerical values determined by norms are absent. To compute the numerical values of regulated liquid parameters, the measurements on the horizontal-polarized waves have to be added with ultrasonic (or other compatible) measurements of the density. The reflection of the longitudinal wave on a liquid-solid boundary [12] or the Rayleigh surface waves and Lamb waves [13] can be used as the ultrasonic source of such information. These waves radiate the part of their energy in the liquid that is the physical cause of the interaction with a liquid-solid interface. Each wave type has its own advantages and disadvantages.

In this work the use of the lowest symmetrical Lamb waves has been considered. Such approach has possessed determined advantage. Firstly, main advantage consists in the large possible signal amplitude changes due to liquid-waveguide interaction. Secondly, the integration of these measurements together with the horizontal-polarized waves is simply to perform. Besides, these waves have the maximal propagation velocity and therefore it is free from hindrances. However, Lamb waves have possessed two components of the particle velocity. The normal component has caused the energy radiation in a liquid, while the tangential component has responded to the above-considered frictional losses. This circumstance has made the interpretation of attenuation coefficient $\alpha_{Lamb}$ measurements more difficult.

The theory of the propagation of Lamb waves in a plate bordered with a viscous liquid has been considered by Zhu and Wu [14]. The left part of the dispersion equation calculated by them has coincided with one for the free plate calculated by Viktorov [15]. The right part of

\[ (\sqrt{\rho'\eta_S})_{exp} \]

\[ (\sqrt{\rho'\eta_S})_{lab} \]

--- ideal dependence,

\(\Delta\) - experimental values.

Figure 2. The relationship between the tabulated and calculated values $\sqrt{\rho'\eta_S}$
the equality has presented complicate function of liquid-waveguide interface parameters (densities and sound velocities of a liquid and waveguides, a shear viscosity of a liquid, the waveguide thinness).

In the general case the dispersion equation is very complicate. The possibility to calculate the dependency $\alpha^W_{\text{Lamb}}(\rho^L/\rho^W)$ without a priori known shear viscosity values with the accuracy acceptable for practical application has to be considerate in addition. Therefore, it is worthwhile, basing on qualitative analyze of the theoretical dispersion equation to build the relation connecting the measured attenuation coefficient of Lamb waves and the density of contacted liquids. This relation has to contain free parameters whose numerical values have to be selected in such a way as to ensure the optimally coincidence of calculated and experimental values of attenuation coefficients.

One can wait that energy losses because of the Lamb wave longitudinal component have lead to the sound attenuation coefficient which is identical to above-considered case of the horizontal-polarized waves. In order to choose the expression for the attenuation coefficient of another component of Lamb wave, it is worthwhile to note the following. The numerical estimates have show that the items taking into account the viscosity influence on the boundary condition for the normal component of the stress tensor has given the correction not exceeded 0.1–0.3% if the viscosity of a liquid is less than 1 Poise. Moreover, one can suppose that the expression of attenuation coefficient due to the radiation in the viscous liquid has weakly differed from one found for the liquid without the viscosity. The last one had been calculated by Victorov [15].

It is proportional to \((Z^L_i/Z^W_{\text{Lamb}})\sqrt{1-(c^L_i)^2/(c^W_{\text{Lamb}})^2}\); \(Z^L_i = \rho^L c^L_i\) is the waveguide longitudinal impedance, \(Z^W_{\text{Lamb}} = \rho^W c^W_{\text{Lamb}}\), \(c^L_i\) is the longitudinal sound velocity in a liquid, \(c^W_{\text{Lamb}}\) is the propagation velocity of null-order symmetrical Lamb wave.

One can write down the expression of the whole attenuation coefficient for the null-order Lamb wave in the form of the sum of losses due to the both components

$$\alpha^W_{\text{Lamb}} = b \cdot (Z^L_i/Z^W_{\text{Lamb}}) \sqrt{1-(c^L_i)^2/(c^W_{\text{Lamb}})^2} + a \cdot \alpha^W_{\text{SH}}. \quad (11)$$

Demanding, for example, the coincidence of numerical values of the measured and calculated attenuation coefficients for equation pairs, the numerical values of the fitted parameters \(a\) and \(b\) can be calculated. In figure 3 is presented the comparison of the dependence $\alpha^W_{\text{Lamb}} = f(Z^L_i/Z^W_{\text{Lamb}})$ calculated from the relation (11), using the values \(a = 0.8\) and \(b = 8.04 cm^{-1}\). The numerical values of sound velocity in the mixture are borrowed from [15]. These parameter values have been got to fit of the calculated and experimental values in pure water and glycerol. This comparison has shown the quantitative agreement the calculated curvature with experimental results although some systematic differences (up to 10 – 15%) have taken place. The latter circumstance demands an additional investigation since such measurement errors impair the possibility of practical use.
values calculated in accordance with the relation (11),
\(\Delta\) - experimental values.

Figure 3. Dependence of the Lamb wave attenuation coefficient (symmetrical mode) on the ratio \(Z_L^L / Z^W_{Lamb}\)

5. Conclusion

Experimental investigations have shown that the presented sensor has ensured a high sensitivity to the magnitude of liquid acoustical impedances and their changes. Its use is the most effective if the direct connection of the acoustical impedance with density and/or shear viscosity of a liquid can be find out. Such cases have been taken place on measurements of the density-shear viscosity product in liquids with a low/middle viscosity using described horizontal-polarized waves. If propagation parameters of the waveguide have been known/measured and a liquid viscosity has not been more than 0.5–0.6 P, the measurement accuracy of the density-shear viscosity product has not exceeded 1%. For more viscosity liquid such simple connection has got broken because of the influence some additional factors whose physical nature is not clear.

The attenuation of Lamb waves propagating along the waveguide-liquid interface has depended on the both liquid impedances. Therefore, the sensor calibration is necessary in order to choose and to fit an approximation describing the relation between these magnitudes and measured ultrasonic parameters with the suitable for practical use accuracy.

References