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## An application of Love SH waves for the viscosity measurement of triglycerides at high pressures

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A new ultrasonic method of viscosity measurements at a high-pressure conditions has been presented. The method is based on the Love wave amplitude measurement. The same electronic setup as in the Bleustein–Gulyaev (B–G) wave method applied by the authors recently for a high-pressure measurement was adopted. The new sensors were made of metallic materials, which make them more reliable at high-pressure conditions. The method has been successfully applied for the viscosity measurement of some triglycerides at high-pressure conditions up to 1 GPa. The results have been compared with the earlier results obtained using B–G waves. This comparison has shown that Love wave method sensors are more reliable than B–G wave sensors and are also cheaper in fabrication, although the sensitivity of Love wave sensors is lower. During the measurement, the phase transitions in the investigated liquids were observed.

Keywords: Love SH waves; viscosity; phase transitions; sensors

#### 1. Introduction

The high-pressure changes of the properties of liquids have been the subject of intensive research at the Warsaw University of Technology for a many years. Since the beginning, the research focused on the viscosity and density changes of various oils. For this purpose, the classical method of falling body was adopted [1]. Several high-pressure-transmitting media have been examined using this arrangement [2]. The strong, exponential dependence of viscosity on pressure, known in the literature as the Barus formula, suggested that a small difference in the composition of a liquid may cause a big difference in the viscosity at high pressures. Because of this strong dependence, the falling ball viscometer method was abandoned.

The discovery of phase transitions in castor oils [3] and in other liquids has intensified the research on the viscosity of oils. Special attention has been focused on the rheological properties of the new high-pressure phase of castor oil. Due to the problems with the Stokes method, the capillary flow rheometer has been applied [4]. Although the measurements of apparent viscosity of castor oil have shown an abnormal rise in viscosity above the phase transition, much bigger

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than the one resulting from the Barus formula, it became obvious that the capillary method must be replaced by another method because of the overly-complicated high-pressure arrangement.

To overcome these problems, shear-horizontal (SH) surface acoustic wave methods were chosen. Such methods have been established in the Section of Acoustoelectronics of the Institute of Fundamental Technological Research, Polish Academy of Sciences in Warsaw, for viscosity measurements under ambient pressure [5,6]. Two methods based on SH surface acoustic waves were considered for high-pressure applications. The first was the Bleustein–Gulyaev (B–G) wave method and the second was the Love wave method. The B–G wave method has been successfully applied for high-pressure measurements of the viscosity of liquids [7]. Using this method, the changes of viscosity during the phase transition in castor oil [8] and triolein [9] were observed. The results of the tests of the high-pressure SH Love method are presented in this paper.

#### 2. Viscosity measurement using SH Love waves

The SH Love wave is an SH acoustic surface wave having only one component of mechanical displacement, parallel to the propagation surface and perpendicular to the direction of propagation. The SH Love wave method is similar to the earlier presented B–G wave method. Both methods are presented in Figure 1.

Energy of the Love wave is mostly concentrated in the 0.2 mm thick copper layer deposited on the surface of a bulk steel bar. The layer and bar constitute the acoustic waveguide.

The viscoelastic liquid covering the waveguide surface loads it mechanically. The value of this load is proportional to the value of the mechanical impedance Z of the liquid medium.

The mechanical impedance of a layer of liquid loading the surface of the B–G wave waveguide is equal to the characteristic shear impedance of the liquid  $Z_L$  for plane waves:

$$Z_{\rm L} = (\rho_{\rm L} \cdot G_{\rm L})^{1/2},$$
 (1)

where  $G_{\rm L} = G' + jG''$  is the complex shear modulus of the liquid defined as the ratio (T/S) of the shear stress T to the shear strain S,  $\rho_{\rm L}$  is the liquid density and  $j = (-1)^{1/2}$ .

In general, liquid loading of the sensor surface changes the phase velocity v and the attenuation  $\alpha$  of the B–G wave. The complex propagation constant  $\gamma$  of the B–G wave changes:

$$\frac{\Delta\gamma}{\beta} = \frac{\Delta\alpha}{\beta} - j\frac{\Delta v}{v_0},\tag{2}$$



Figure 1. The waveguide assembly for viscosity measurement by the B-G wave method and the Love wave method.

where  $\gamma = \alpha + j\beta$ ,  $\beta = \omega/v$ ,  $v_0$  is the phase velocity of the non-perturbed B–G wave on the free surface and  $\omega$  the angular frequency of the B–G wave.

By applying the perturbation method, one can prove that the change in the complex propagation constant  $\gamma$  of the Love wave produced by viscoelastic liquid loading is as follows [6]:

$$\Delta \gamma = -j \left( \frac{|v_3|_{x_2=0}^2}{4P} \right) Z_{\rm L} = -j K Z_{\rm L},\tag{3}$$

where  $v_3$  is the B–G wave amplitude on the waveguide surface ( $x_2 = 0$ ), *P* is the mean power on the unit width of the B–G wave. The coefficient *K* is the characteristic quantity for each B–G wave waveguide and depends solely on the material parameters of the waveguide and frequency [5]. In the next step, the rheological model describing the investigated liquid should be determined.

In this paper, the liquids investigated under high pressure are treated as the Newtonian liquids. For the case of a Newtonian (viscous) liquid, the shear mechanical impedance (defined as a ratio of the shear stress to the shear vibrational velocity) can be expressed as follows [10]:

$$Z_{\rm L} = R_{\rm L} + j X_{\rm L} = \left(\frac{\rho_L \omega \eta}{2}\right)^{1/2} (1+j), \tag{4}$$

where  $\eta$  is the viscosity.

So that we may regard Equation (5) as holding for the liquids considered in the paper:

$$\eta = \frac{2R_{\rm L}^2}{\omega\rho_{\rm L}} = \frac{2X_{\rm L}^2}{\omega\rho_{\rm L}},\tag{5}$$

where  $R_L$  and  $X_L$  are the real and imaginary parts of the mechanical shear impedance of a liquid.

The shear mechanical impedance of a liquid  $Z_L = R_L + jX_L$  can be determined from the measurement of the change in attenuation and time of flight of wave trains that propagate in the waveguide loaded by a liquid [6]. The model of a Newtonian liquid was used by Philippoff [11]. He stated that the majority of oils in the considered shearing rate (about 1 MHz) and under high pressures are the Newtonian liquids. This can justify the use of a Newtonian liquid model in our paper.

#### 3. Experimental setup

The real advantage of the method presented here is that contrary to the other methods of viscosity measurement, this method does not require any special high-pressure equipment. Any chamber that can contain the ultrasonic Love SH wave sensor with reliable electrical terminals is appropriate for this application. In our case, the acoustic waveguide was made of a rectangular  $8 \times 8 \times 60$  mm bar of steel covered at one side with 0.2 mm layer of copper, with a piezoceramic transducer bonded at its end. The pressure was generated in a thick-walled cylinder of 17 mm internal diameter with a simple piston presented in [12]. This piston–cylinder system was operated by a 20 ton laboratory hydraulic press, which limited the maximum pressure to about 1 GPa. For pressure measurement, a typical 75  $\Omega$  manganin transducer was used. Its resistance was measured with a digital resistance bridge calibrated in MPa. Pressure measurement accuracy was better than 0.1%. The experiments were done at room temperature. Temperature was measured by the copper-constantan thermocouple placed inside the chamber.



Figure 2. Variation in viscosity of triolein, as a function of hydrostatic pressure, measured by the ultrasonic Love wave method. Exponential curve represents the Barus formula. Decompression curve is marked by an arrow.

#### 4. Results of the measurements

The pressure was generated in 10 MPa steps and then kept constant for about 2–5 min. During that time, the pressure was carefully observed. That allowed identification of the pressure drop due to the first-order phase transition and to observe whether the system is reaching thermodynamic equilibrium. After approaching 0.6 GPa, the pressure was kept constant for about 20 h to enable the phase transformation to occur. During the phase transition, a small drop in pressure and increment of viscosity was observed. The results of the viscosity measurement are shown in Figure 2.

As seen in Figure 2, the experimental curve up to about 400 MPa is almost tangential to the exponential curve which represent the Barus formula  $\eta(p) = \eta_0 \exp(\alpha p)$ , where  $\eta_0$  is the viscosity at atmospheric pressure and  $\alpha$  is the viscosity–pressure coefficient. Above 400 MPa, the experimental points rise more slowly than the theoretical prediction. Finally, at 600 MPa, when the pressure rise was stopped for about 20 h, the viscosity has risen to the new value, characteristic for the high-pressure phase of castor oil. The further increment in the viscosity was rather a linear function of pressure.

#### 5. Conclusions

- (1) The Love SH wave sensor was successfully tested at high-pressure conditions.
- (2) The measurements of viscosity performed using the new sensor were very similar to the results obtained by the B–G wave method.
- (3) The new type sensor is more reliable in chemically aggressive media.

The usefulness of the surface Love wave for measuring liquid viscosity at high pressures has been stated. This method enables measuring viscosity during phase transition and high-pressure phase decomposition. To the authors knowledge, the measurements of liquid viscosity under high pressures during the phase transition and during the pressure decompression were not yet reported in the scientific literature.

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