Application of SH surface acoustic waves for measuring the viscosity of liquids in function of pressure and temperature

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ABSTRACT

Viscosity measurements were carried out on triolein at pressures from atmospheric up to 650 MPa and in the temperature range from 10 °C to 40 °C using ultrasonic measuring setup. Bleustein–Gulyaev SH surface acoustic waves waveguides were used as viscosity sensors. Additionally, pressure changes occurring during phase transition have been measured over the same temperature range. Application of ultrasonic SH surface acoustic waves in the liquid viscosity measurements at high pressure has many advantages. It enables viscosity measurement during phase transitions and in the high-pressure range where the classical viscosity measurement methods cannot operate. Measurements of phase transition kinetics and viscosity of liquids at high pressures and various temperatures (isotherms) is a novelty. The knowledge of changes in viscosity in function of pressure and temperature can help to obtain a deeper insight into thermodynamic properties of liquids.

1. Introduction

Monitoring and studying the pressure and temperature effect on liquid physical properties (e.g. viscosity) are becoming increasingly important in the food, chemical, petrochemical, cosmetic and pharmaceutical industry. High pressure research of the physical properties of liquids has been stimulated by the fast development of such technologies as biodiesel production, high-pressure food processing and conservation, modification of biotechnological materials. The knowledge of physical properties of treated substance is essential for understanding, design and control of the process technology. Viscosity is one of the most important parameters of liquids [1–4]. Measurement techniques for "*in situ*" determining of viscosity of liquid under high pressure allow insight into the phenomena governing the microstructural modifications occurring in the treated substance. High-pressure transitions in liquids can be investigated by the measurements of the viscosity in function of hydrostatic pressure and temperature.

Up to date, high-pressure viscosity measurements were performed using conventional mechanical methods [5]. It is very difficult to extend conventional methods to determine the viscosity at high-pressure [6]. One of the problems is to control the trajectory of the falling (rolling) ball and to track its movements. The resetting of the sinker or rolling ball also present difficulties. An eccentric fall of the sinker can cause significant errors in

determining viscosity based on sinker descent time. Falling sinker viscometers and rolling ball viscometers have very long measuring times at high viscosities. Capillary type viscometers pose problems with pressure gradients. The application of rotary viscometers is limited due to the problems with generated heat and leakage during the transmission of the rotation into high-pressure chamber. Due to inherent limitations, the conventional methods cannot operate on-line during technological processes and are only laboratory methods. Recently, the new type of Couette (rotational) viscometer has been developed for pressure range up to 400 MPa [7].

There exist also other methods employing different physical phenomena e.g. magnetic field [8] and light scattering [9] for measuring the viscosity of liquids at high pressure. However, they need very complicated equipment and specially developed high-pressure chambers.

Due to disadvantages of the conventional methods, a need for new measuring methods arose. To this end, ultrasonic methods for the measurements of the viscosity of liquids under high pressure were proposed. For example, a torsionally oscillating piezoelectric quartz rod was applied as an ultrasonic viscosity sensor [10–12]. In this type of ultrasonic sensors bulk type waves were applied. The acoustic energy of bulk waves is distributed in the entire volume of the resonator. The contact with a measured liquid takes place on the surface of the resonator. This results in the moderate sensitivity of this type of viscosity sensors.

To overcome the disadvantage of the bulk wave methods, the authors have proposed to use the SH surface acoustic waves of the Love and Bleustein–Gulyaev (B–G) type. This method was

established in the Institute of Fundamental Technological Research in Warsaw (Poland) for the measurements of the liquid viscosity at the atmospheric pressure [13,14]. Subsequently Love and B–G waves were successfully applied as a tool to measure the viscosity of liquids at high pressure [15]. At the beginning, the measurements of the viscosity of triglycerides, vegetable oils and fatty acids were carried out at the ambient temperature [16–18]. In this paper, we present the results of measurements of liquid viscosity in function of hydrostatic pressure at various temperatures (isotherms). To this end we developed a new measuring setup. The new high-pressure chamber with a thermostatic jacket was also designed and constructed.

In Section 2 we shortly recalled the measuring method. In Section 3 the measuring setup is described. In the Section 4 the results of the measurements of the triolein viscosity in function of the hydrostatic pressure for four values of temperature are presented (isotherms). Triolein ($(C_{17}H_{33}COO)C_3H_5$) has been selected as a very good model-liquid for the whole group of triglycerides. Most of the natural oils consist of triglycerides of various fatty acids.

2. Viscosity measurement using B–G waves

The B–G and Love waves are SH acoustic surface waves having only one component of mechanical displacement, parallel to the propagation surface and perpendicular to the direction of propagation. In the presented measurements we used the B–G wave viscosity sensor.

The penetration depth of the B–G wave is of the order of a wavelength. Consequently, the energy of the B–G waves is mostly concentrated in the vicinity of the surface of a properly polarized piezoelectric ceramic waveguide used as a viscosity sensor [14].

The 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. 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:

$$Z_L = (\rho \cdot G_L)^{1/2} \tag{1}$$

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

By applying the perturbation method [19], one can prove that the change in the complex propagation constant γ of the B–G wave produced by viscoelastic liquid loading is as follows [14]:

$$\Delta\gamma = -j\left(\frac{|u_3|^2_{x_2=0}}{4 \cdot P}\right) \cdot Z_L = -j \cdot K \cdot Z_L \tag{2}$$

where $\gamma = \alpha + j\beta$, α is an attenuation coefficient, $\beta = \omega/v$, v_0 is the phase velocity of the non-perturbed B–G wave on the free surface, ω is the angular frequency of the B–G wave, u_3 is the B–G wave amplitude on the waveguide surface ($u_3 = 0$), 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 [13].

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 [20]:

$$Z_L = R_L + jX_L = \left(\frac{\rho_L \omega \eta}{2}\right)^{1/2}(1+j) \tag{3}$$

where η is the viscosity.

So that we may regard Eq. (4) as holding for the liquids considered in this paper [15]:

$$\eta = \frac{2R_L^2}{\omega\rho_L} = \frac{2X_L^2}{\omega\rho_L} \tag{4}$$

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 [14]. The same relations are valid for the Love viscosity sensors [18].

The model of a Newtonian liquid was used e.g. by Philippoff [10]. 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 experimental setup for measuring the viscosity of liquids under high pressure at various temperatures is presented in Fig. 1. High pressure was generated in a thick-walled cylinder chamber with a simple piston and Bridgman II sealing system. The piston–cylinder assembly was working with a hydraulic press, driven by a hand-operated pump. The piston displacement was controlled by a digital caliper. For pressure measurements a typical 75 Ω manganin transducer was used. Its resistance was measured with a digital resistance bridge calibrated in MPa. The temperature in the chamber was measured using T-type thermocouple (Cu-constantan). A thermostatic bath was circulating in a thermostatic jacket around the chamber. The thermostatic jacket was connected to a precision thermostat (Julabo Labortechnik, Germany) working as a refrigerated/heating circulator. The viscosity sensor (B–G waveguide) was placed inside the high-pressure chamber. The piezoelectric transducer attached to the B–G wave waveguide, manganin coil, and thermocouple were connected with the external measuring setup by an electrical multi-channel lead through.

The sending–receiving piezoelectric transducer, attached to the B–G wave waveguide, was driven by the TB-1000 pulser-receiver computer card (Matec, USA). The TB-1000 pulser generated the rf tone bursts with a frequency $f = 2$ MHz and length equal to 0.5 μs. The repetition period was equal to 0.4 ms. The B–G wave impulse generated by the transducer was reflected in multiple ways between two opposite edges of the B–G wave waveguide. The signals received by the transducer were amplified by the TB-1000 receiver and sent into the PDA-1000 digitizer card (Signatec, USA). This card sampled and digitized the input analog signals. The stored signals were then analyzed by computer software. For each measurement, the ultrasonic signal was averaged 1024 times in order to improve the signal-to-noise ratio. A computer program that controlled the operation of the computer cards and data acquisition was written in C++ language.

4. Experimental results

4.1. Viscosity

The results of high-pressure viscosity measurements of triolein at various temperatures are shown in Fig. 2. The pressure was generated by a hand-operated pump in 10 MPa steps, then kept constant for about 2–5 min. During that time the pressure and temperatures were observed. That allowed identification of the pressure drop due to phase transition and to observe whether the system was reaching thermodynamic equilibrium. The
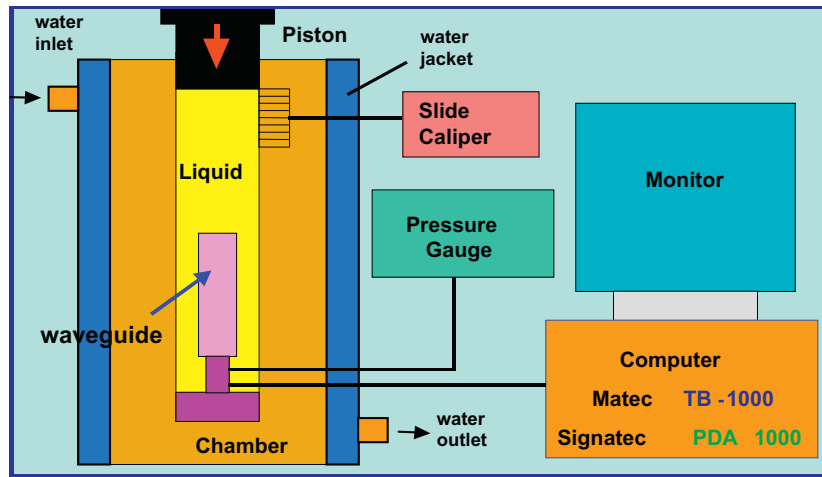
Fig. 1. Ultrasonic experimental setup for measuring the viscosity of liquids under high pressure at various temperatures. Temperature of water is stabilized by refrigerated/heating circulator (not presented in the graph).
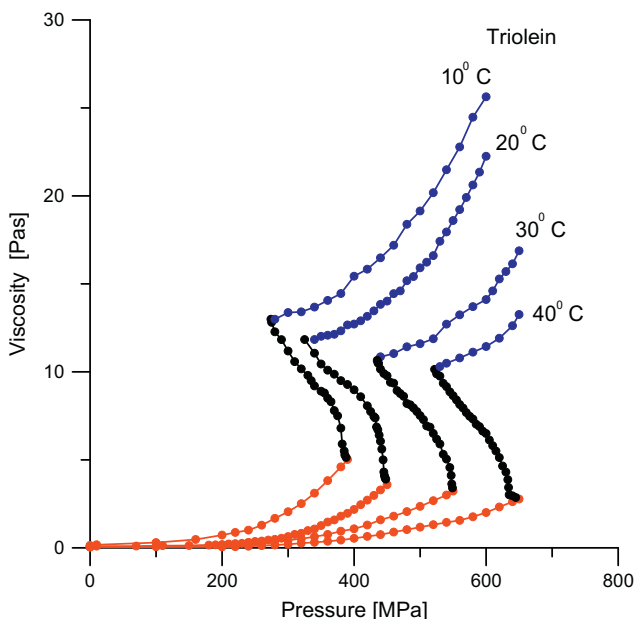

Fig. 2. Viscosity of triolein versus pressure along various isotherms (*T* = 10, 20, 30, and 40 °C).

viscosity of triolein was measured in function of pressure and temperature at 10 °C intervals from 10 °C to 40 °C and from atmospheric pressure to 650 MPa, see Fig. 2. This range of temperatures is the most interesting from the practical and cognitive point of view. The temperature of solidification is 5 °C. On the other hand, in the temperature range above 50 °C, phase transition runs very slowly and starts at a higher value of pressure. The temperatures in the high-pressure chamber was stabilized by the refrigerated/heating circulator and controlled by the T-type thermocouple. In the Fig. 2 three different parts of each curve can be seen. At first we increased the pressure until the first-order phase transition began. The viscosity was increasing almost exponentially according to known empirical Barus formula $\eta(p) = \eta_0 \exp(\alpha p)$, where η_0 is the viscosity at atmospheric pressure and α is the viscosity-pressure coefficient. One can see in Fig. 2 that the coefficient α depends on temperature. When the phase transition started we stopped the compression, and the piston in the high-pressure chamber was

fixed to enable the phase transition to occur undisturbed. During the phase transition a pressure drop of about 120 MPa was observed in the chamber. The viscosity showed the further rise despite the pressure drop. It means that the volume occupied by the resulting high-pressure phase diminished. When the temperature during measurements was higher, the pressure, at which the phase transition began, increased, see Fig. 2. Consequently, the pressure at which transition stopped was also higher. The stabilization of pressure and negligible changes in viscosity indicate that the phase transition is completed. At the termination of the phase transition process we increased the pressure again, in order to measure the viscosity of the new high-pressure phase of triolein (third part of each curve in Fig. 2). The further increase of viscosity with increasing pressure was observed. With the increase of temperature, viscosity of triolein high-pressure and low-pressure phases diminishes, see Fig. 2.

4.2. Kinetics

The kinetics of the phase transition was investigated during viscosity measurements, see Fig. 3. Pressure changes, occurring during phase transition, were registered with the piston locked in a fixed position. At first the pressure remained constant. Subsequently, a rapid decrease of pressure was observed, due to a phase transition in triolein. Finally, the pressure level stabilized. This means that the phase transition was complete. As a result, a new high-pressure phase in triolein, with different microstructure, has emerged. Physical properties of the new high-pressure phase are different than those in the low-pressure phase. One can see in Fig. 3, that with the increase of temperature phase transition starts at higher value of pressure. Moreover, an augmentation in temperature decreases the speed of phase transition (e.g., phase transition at 40 °C runs about 70 min). On the other hand, with the decrease in temperature, phase transition starts at a lower value of pressure and runs with higher speed (e.g., phase transition at 10 °C runs about 40 min).

The phase transitions in triolein at high pressure were investigated using: (1) measurements of changes in permittivity [21], (2) optical methods [22] and (3) measurements of changes in phase velocity of longitudinal ultrasonic waves [23]. All those measurements were carried out at ambient temperature. The results obtained from those three methods conform well with the results obtained using the method presented in this paper.
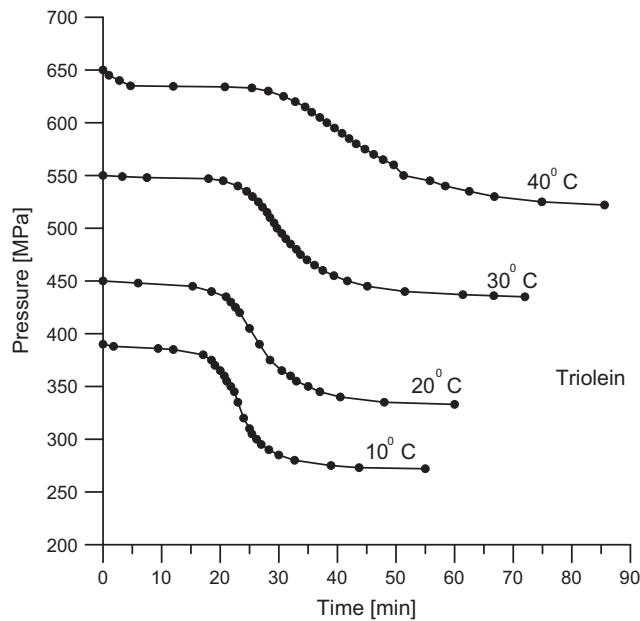
Fig. 3. Kinetics of phase transition in triolein at various temperatures (T = 10, 20, 30, and 40 °C).

5. Conclusions

The usefulness of the modified experimental setup for measuring liquid viscosity at high pressure for various temperatures has been stated. The SH surface acoustic waves (SAW) method enables measuring viscosity during phase transition, after emerging of the high-pressure phase and during phase decomposition. The kinetics of the phase transition has also been measured. To the authors knowledge, the measurements of liquid viscosity under high pressure, for various temperatures (isotherms), during the phase transition were not yet reported in the scientific literature.

Measurement of liquid viscosity at high pressure is important in tribology in rolling bearings, in design and exploitation of ship diesel engines, in the chemical, pharmaceutical and cosmetic industries as well as in bio-fuels production, and food conservation.

Investigation of phase transitions is important in lubricants, since rheological properties of lubricants change during phase transitions [24]. Investigation of phase transitions is also very important in food processing and conservation [25]. Phase transitions can modify irreversibly the molecular structure and quality of food products. The measurements of the rheological properties of liquids during phase transitions are not possible using conventional mechanical methods. Applications of the B–G wave or Love wave method enables both the detection of phase transitions and investigation of their kinetics.

In general, the SH-SAW method has high sensitivity and high reliability. It can be computerized. This enables continuous (on-line) monitoring of the rheological parameters of liquids *in situ* during the course of technological processes. Small dimensions of the viscosity sensor and the absence of moving parts are substantial advantages of this method. The B–G wave or Love wave viscosity sensor is electrically responsive. Owing to this fact, modern methods of the digital signal acquisition and processing can be efficiently used.

The knowledge of changes in viscosity in function of pressure and temperature will help to obtain a deeper insight into thermodynamic properties of liquids.

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