10.1109/ULTSYM.2011.0129

Measurements of the viscosity of liquids in function of pressure and temperature using SH surface acoustic waves

[a]P. KIEŁCZYŃSKI[1], M. SZALEWSKI[1], A. BALCERZAK[1], A. J. ROSTOCKI[2]
[1]Institute of Fundamental Technological Research, Polish Academy of Sciences,
ul. Pawińskiego 5B, 02-106 Warsaw, Poland.
[2]Warsaw University of Technology, ul. Koszykowa 75, 00-662 Warsaw, Poland.

[a]e-mail: pkielczy@ippt.gov.pl

Abstract – **Triolein viscosity was measured 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. Application of 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. The knowledge of changes in viscosity in function of pressure and temperature can help to obtain a deeper insight into thermodynamic properties of liquids. Measurements of phase transition kinetics and viscosity of liquids at high pressures and various temperatures (isotherms) is a novelty.**

Keywords: Viscosity, SH surface acoustic waves, high pressure, phase transitions

I. INTRODUCTION

The knowledge of physical properties of treated substance is essential for understanding, design and control of the process technology. 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. Viscosity is one of the most important parameters of liquids. 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 fuction of hydrostatic pressure and temperature.

Up to date, high-pressure viscosity measurements were performed using conventional mechanical methods [1]. It is very difficult to extend conventional methods to determine the viscosity at high pressure [2]. 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 [3]. 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 Section of Acoustoelectronics of the IFTR PAS in Warsaw for the measurements of the liquid viscosity at the atmospheric pressure [4,5]. Subsequently, Love and B-G waves were successfully applied as a tool to measure the viscosity of liquids at high pressure [6]. At the

beginning, the measurements of the viscosity of triglycerides, vegetable oils and fatty acids were carried out at the ambient temperature [7-9]. In this paper, we present the results of measurements of liquid viscosity in function of hydrostatic pressure for various temperatures. To this end we developed a new measuring setup. The new high-pressure chamber with a thermostatic jacket was also constructed.

II. MEASURING METHOD AND SETUP

Measurements were performed in the experimental setup 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 hydraulical 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.

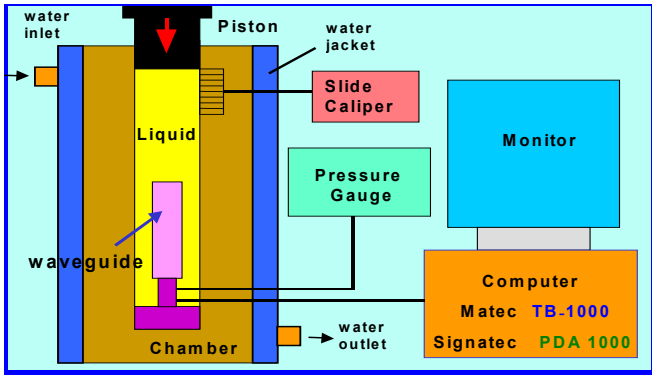

Fig.1. Ultrasonic experimental setup for measuring the viscosity of liquids under high pressure at various temperatures. Temperature of water is stabilized by refrigerated/heating circulator (not presented in the graph).

The viscosity sensor (B-G waveguide) was placed inside the high-pressure chamber. The piezoelectric transducer attached to the B-G wave waveguide, manganine coil, and thermocouple were connected with the external measuring setup by an electrical multichannel 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

III. EXPERIMENTAL RESULTS

A. VISCOSITY

Figure 2 shows the results of high-pressure viscosity measurements of triolein at various temperatures. The pressure was generated by a hand-operated pump in 10 MPa steps, then kept constant for about 2-5 minutes. During that time the pressure and temperature was observed. That allowed identification of the pressure drop due to phase transition and to observe whether the system was reaching thermodynamic equilibrium. The 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. 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

part (red) 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 (black) 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 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, blue, of each curve in Fig. 2). The further increase of viscosity with increasing pressure was observed.

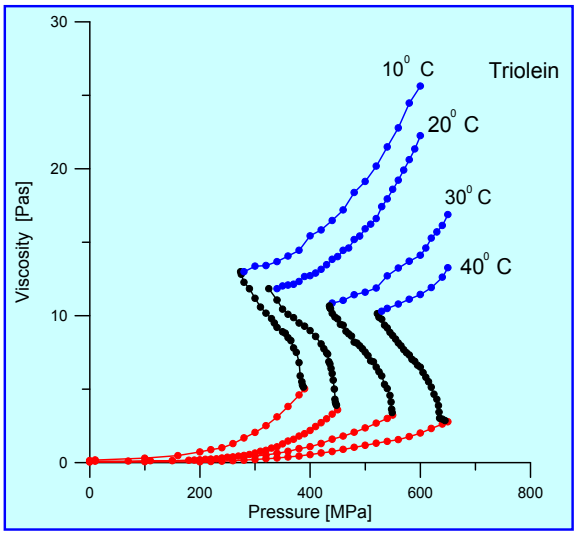

Fig.2. Viscosity of triolein versus pressure along various isotherms (T = 10, 20, 30, and 40° C).

With the increase of temperature, viscosity of triolein high-pressure and low-pressure phases diminishes. We measured also the viscosity during the decompression process. The changes of viscosity during the decompression have shown large hysteresis. Large hysteresis indicates existence of large internal friction forces. We have not presented those parts of curves in Fig. 2 for the sake of readibility of the figure.

B. KINETICS

Figure 3 displays the kinetics of the phase transitions that were observed during viscosity measurements.

Pressure changes, occuring 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.

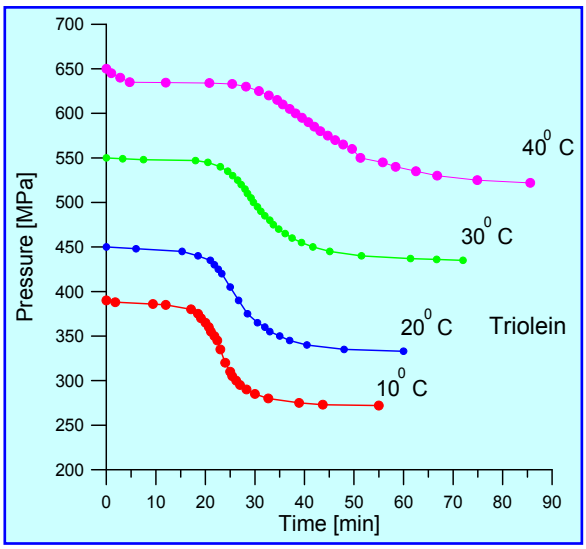

Fig.3. Kinetics of phase transition in triolein at various temperatures (T = 10, 20, 30, and 40° C).

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.

IV. CONCLUSIONS

The benefit of using 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, during the phase transition and during the pressure decompression were not yet reported in the scientific literature.

Investigation of phase transitions is very important in food processing and conservation. 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.

REFERENCES

1. L. Kulisiewicz, A. Delgado, "High-pressure rheological measurements methods, a review", Appl. Rheol., 20 (2010) 13018-1-13018-15.

2. Y.Y. Hou, H.O. Kassim, "Instrument techniques for rheometry", Rev. Sci. Instr., 76 (2005), 101101-1-101101-19.

3. A.F. Collins, E. M. McLaughlin, "Torsional crystal technique for the measurements of viscosities at high pressure", Trans. Faraday Soc. 67 (1971) 340-352.

4. P. Kiełczyński, R. Płowiec, „Determination of the shear impedance of viscoelastic liquids using Love and Bleustein-Gulyaev waves", J. Acoust. Soc. Am., 86 (1989) 818-827.

5. P. Kiełczyński, W. Pajewski, M. Szalewski, A. Balcerzak, "Measurement of the shear storage modulus and viscosity of liquids using the Bleustein-Gulyaev wave", Rev. Sci. Instr., 75 (2004) 2362-2367.

6. P. Kielczynski, M. Szalewski, R.M. Siegoczynski, A.J. Rostocki, „New ultrasonic Bleustein-Gulyaev wave method for measuring the viscosity of liquids at high pressure", Rev. Sci. Instr., 79 (2008) 026109-1-026109-3.

7. R.M. Siegoczynski, A.J. Rostocki, P. Kielczynski, M. Szalewski, „ A viscosity measurement during the high pressure phase transition in triolein", J. Phys.: Conf. Ser., 121 (2008) 142010-1-142010-5.

8. P.Kielczynski, M. Szalewski, A.J. Rostocki, J. Gladysz, „Investigation of high-pressure phase transitions in castor oil using SH surface acoustic waves ", 2008 IEEE International Ultrasonics Symposium Proceedings, Beijing, China, 2154-2157.

9. A.J. Rostocki, R.M. Siegoczynski, P. Kielczynski, M. Szalewski, "An application of Love waves for the viscosity measurements of triglycerides at high pressure", High Press. Res., 30 (2010) 88-92.