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Investigation of High-Pressure Phase Transitions in Castor Oil Using SH Surface Acoustic Waves

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Abstract - **Measurement techniques for in-situ rheological investigations under high pressure allow insight into the phenomena governing the microstructural modifications. The conventional mechanical methods can not be operated to this aim due to their inherent limitations. This is why ultrasonic methods for the measurement of the viscosity of liquids under high pressure were introduced. To this end, the authors have applied new ultrasonic methods, i.e., the Love wave method and the Bleustein-Gulyaev (B-G) wave method. The measurements of the viscosity of liquid (castor oil) were carried out in function of hydrostatic pressure up to 800 MPa. During the measurement we stated the phase transformation of castor oil and the presence of the hysteresis of the dependence of viscosity on pressure. To the authors' knowledge, the measurement of the viscosity of liquids under high pressure during the phase transition and during the decompression process is the novelty.**

Keywords: Phase transitions, high pressure, SH surface acoustic waves, viscosity sensors

I. INTRODUCTION

Monitoring and studying the pressure effect on liquid viscosity are becoming increasingly important in the following industries:

1) food (e.g., conservation and processing)
2) in bioengineering (inactivation of enzymes and microorganisms)
3) chemical
4) cosmetic and pharmaceutical
5) in lubricant technology (rolling bearings)
6) investigations of bio-diesel fuels.

High-pressure technologies (pressures up to 1 GPa) have proved a great potential in modern bioengineering as a method of modification of biotechnological materials. The knowledge of rheological properties of treated substance is essential for understanding, design and control of the process technology. Measurement techniques for in-situ rheological investigations under high pressure allow insight into the phenomena governing the microstructural modifications occuring in the treated substance (e.g., vegetable oil). Many vegetable oils undergo some form of phase trasitions during high-pressure manufacture.

In this study we investigate the rheological behavior of castor oil under high pressure [1-7]. Castor oil is a vegetable oil, that is a triglyceride in which approximately ninety percent of fatty chains are ricinoleic acid. Oleic and linoleic acids are the other significant components. Castor oil and its derivatives have applications in the manufacturing of soaps, lubricants, hydraulic and brake fluids, paints, dyes, coatings, inks, cold resistant plastics, waxes and polishes, nylon, pharmaceuticals and perfumes.

II. MEASURING METHODS

Usually the conventional mechanical methods are applied for the viscosity measurements at high pressure. The methods using falling ball, falling cylinder, capilary flow and rotating cylinders (Couette type) are the most popular. However, measurements using this method are time – consuming, tedious, and require special sophisticated equipment. The re–setting of the ball also presents difficulties. 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 the inherent limitations, the conventional methods cannot operate in real-time, and are only laboratory methods.

This is why, 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. In this type of viscosity sensors bulk ultrasonic torsional waves were applied. In this type of viscosity sensors the acoustic energy is distributed in the entire volume of the rod. The contact with a measured liquid takes place on the surface of the rod. This results in the moderate sensitivity of this type of viscosity sensors.

To overcome this discrepancy, shear horizontal (SH) surface acoustic waves (i.e., Love waves and Bleustein-Gulyaev waves) were introduced recently into the measurement of a liquid viscosity [8,9].

III. SH SURFACE ACOUSTIC WAVE SENSORS

To measure the viscosity of liquids under high pressure, the authors applied the Love and Bleustein-Gulyaev (B-G) SH surface acoustic waves. The measuring method proposed by the authors has many advantages [10]:

1) High sensitivity
2) Simple and robust construction
3) Operation in real-time
4) Small dimensions
5) No leakage problems
6) No heating caused by shear

Two types of sensors have been used: 1) Love wave waveguide made of steel substrate coated by Cu layer and 2) B-G wave waveguide made of Pz 46 piezoceramics (Ferroperm) coated by Au layer.

IV. MEASURING SETUP

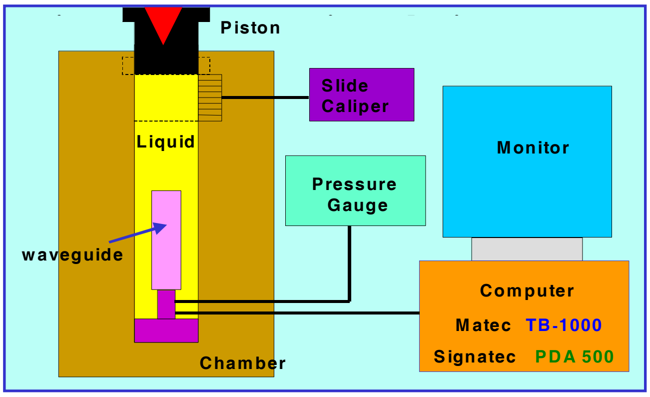

Fig.1. Ultrasonic set up for measuring the viscosity and pressure of liquids under high pressure.

In the setup (see Fig.1) for measuring viscosity using SH surface acoustic waves, the sending–receiving piezoelectric (PZT) transducer is driven by the TB-1000 pulser–receiver computer card (Matec, USA). The TB-1000 pulser generates the rf tone burst with a frequency f = 2 MHz and length equal to 0.5 μs. The SH surface acoustic wave impulse generated by the transducer is reflected in multiple ways between two opposite edges of the piezoceramic waveguide. The signals received by the transducer are amplified by the TB-1000 receiver and sent into the PDA-500 digitizer card (Signatec, USA). This card samples and digitizes the input analog signals. The stored signals are then analyzed by computer software.

V. ANALYTICAL FORMULAS

The presence of a liquid on the waveguide surface changes the velocity and attenuation of the ultrasonic SH surface wave. We measure:

amplitude of the wave pulses:
$\Rightarrow$ viscosity

The liquids investigated under high pressure are treated as the Newtonian liquids. Hence:

viscosity: $$\eta = \frac{2R_L^2}{\omega \rho_L} \qquad (1)$$

where: R_L is the real part of the shear mechanical impedance of a liquid:

$$R_L = \frac{\ln(A_2/A_0)}{2KL} \qquad (2)$$

where: A_2 and A_0 represent the amplitude of the SH surface wave,
L is the length of the waveguide,
K depends on the material parameters of the waveguide and frequency,
ω is an angular frequency, ρ_L is the density of a liquid.

VI. EXPERIMENTAL RESULTS

Variations in viscosity of castor oil as a function of hydrostatic pressure have been measured using the ultrasonic Love wave (Fig.2) and B-G wave (Fig.3) method. For pressure measurements a manganin transducer was used. The pressure was generated in 10 MPa steps. After approaching 600 MPa the compression was stopped and the piston in the high-pressure chamber was fixed. This would enable the phase transformation to occur undisturbed. During the phase transition the viscosity showed the further rise. The changes of viscosity during the decomposition process (upper curve) have shown large hysteresis.

A. *Viscosity Measurements*

1) Love Waves

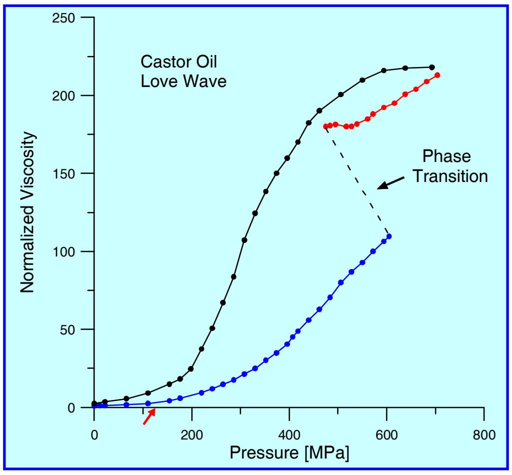

Fig.2. Variations in viscosity of castor oil, as a function of hydrostatic pressure, measured by the Love wave method, f = 2 MHz.

2) Bleustein-Gulyaev Waves

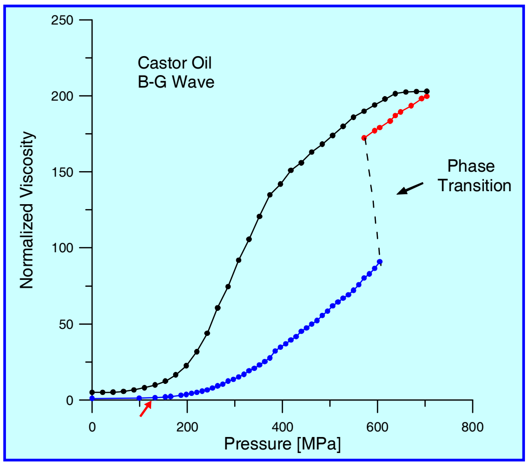

Fig.3. Variations in viscosity of castor oil, as a function of hydrostatic pressure, measured by the B-G wave method, f = 2 MHz.

VII. DISCUSSION OF RESULTS

The measurements were performed in castor oil in the pressure range from 0 to 800 MPa. During the measurements we stated:

a) Phase transformations in liquids
b) Presence of high-pressure phase
c) Presence of the hysteresis in the viscosity-pressure relationship

During the phase transition the drop of pressure and large increase of viscosity were observed. After the phase transition the viscosity has risen to the new value characteristic for the high-pressure phase of castor oil. The decomposition of the high-pressure phase during the decompression precess has shown large hysteresis of the dependence of viscosity on pressure.
Large hysteresis indicates existence of large internal friction forces.

VIII. CONCLUSIONS

The results of measurements presented show:

1. Usefulness of the SH surface B-G and Love wave methods for measuring liquid viscosity at high pressure

2. Advantages of the new SH surface wave method:

a) can be computerized and operates in real-time. This enables continuous (on-line) monitoring of the rheological parameters of a liquid during the course of technological processes. Moreover, this makes it also possible to detect and analyze phase transitions
b) simple sensor fabrication
c) no high pressure leakage problems
d) only electrical lead-through. Therefore it is easy to assemble the sensor into the high-pressure chamber.
e) sensor is electrically responsive. Owing to this fact, modern methods of the digital signal acquisition and processing can be efficiently used.
f) absence of moving parts

Measurement of the viscosity of liquids under high pressure during the phase transition and the decompression is an original author's contribution. To the authors knowledge such measurements were not reported in the scientific literature.

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