APPLICATION OF ULTRASONIC METHODS TO INVESTIGATE THE PHYSICAL PROPERTIES OF LIQUIDS AT HIGH PRESSURE

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June 17, 2011, Warsaw, Poland.

OUTLINE

1. Motivation
2. Mechanical and ultrasonic methods for measuring viscosity of a liquid under high pressure
3. Surface waves of SH type (shear horizontal)
   - a) Love
   - b) Bleustein-Gulyaev
4. The measuring system for determining the viscosity of a liquid under high pressure
5. The results of measurements
6. Ultrasonic wave velocity measurements in liquids under high pressure
7. The measurement of viscosity as a function of pressure and temperature
8. Phase transitions caused by high pressure
9. Conclusion

Monitoring and studying the pressure effect on liquid physical properties is very important in:

- Chemical, pharmaceutical and cosmetic industry
- Food processing and conservation
- Biodiesel production
- Lubrication processes
- Oil-based drilling fluids exploitation
- Oceanography and Geology (petroleum cuts)
- Astrophysics
- Glass processing

Conventional mechanical methods for the measurement of liquid viscosity adapted for high-pressure conditions:

- Rolling ball
- Falling ball (Stokes)
- Falling needle
- Falling cylinder
- Rotational viscometer (Couette - 1890)
- Capillary tube viscometer (Poiseuille - 1846)

Disadvantages of mechanical methods:

- Presence of moving parts
- Require special sophisticated equipment
- Measurements are tedious and time consuming
- Large dimensions
- Difficult to computerize
- Cannot operate in real-time
- Only laboratory methods

Ultrasonic methods
Bulk acoustic waves

- Standing waves (resonators)
- e.g. torsionally oscillating piezoelectric quartz rod (1950)
- Travelling waves (waveguides)

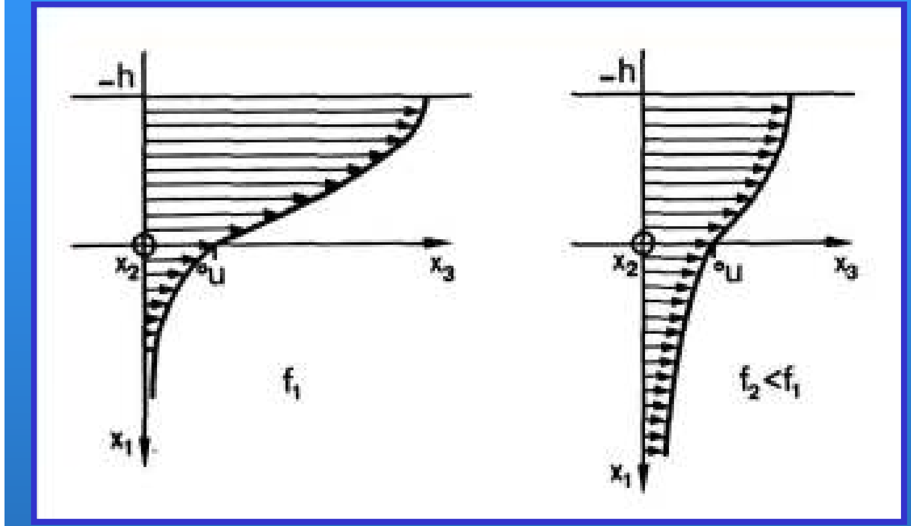
The acoustic energy is distributed in the entire volume of resonator or waveguide. The contact with an investigated liquid takes place on its surface.

Shear horizontal surface acoustic waves (SH-SAW)

- Love waves
- Bleustein-Gulyaev (B-G) waves (1989)

The energy of SH-SAW is concentrated in the vicinity of the waveguide surface. Thus the SH-SAW velocity and attenuation strongly depend on the boundary conditions on the waveguide surface. In consequence, the sensitivity of the viscosity sensors using SH-SAW can be several orders larger than the sensitivity of the sensors employing bulk shear acoustic waves.

Love Waves



- Fig.1. Love wave amplitude in function of depth

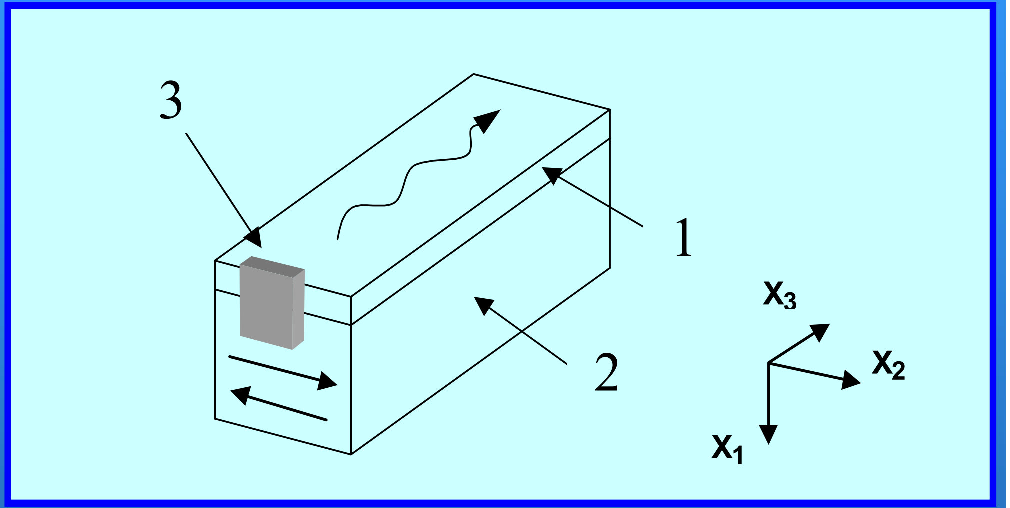
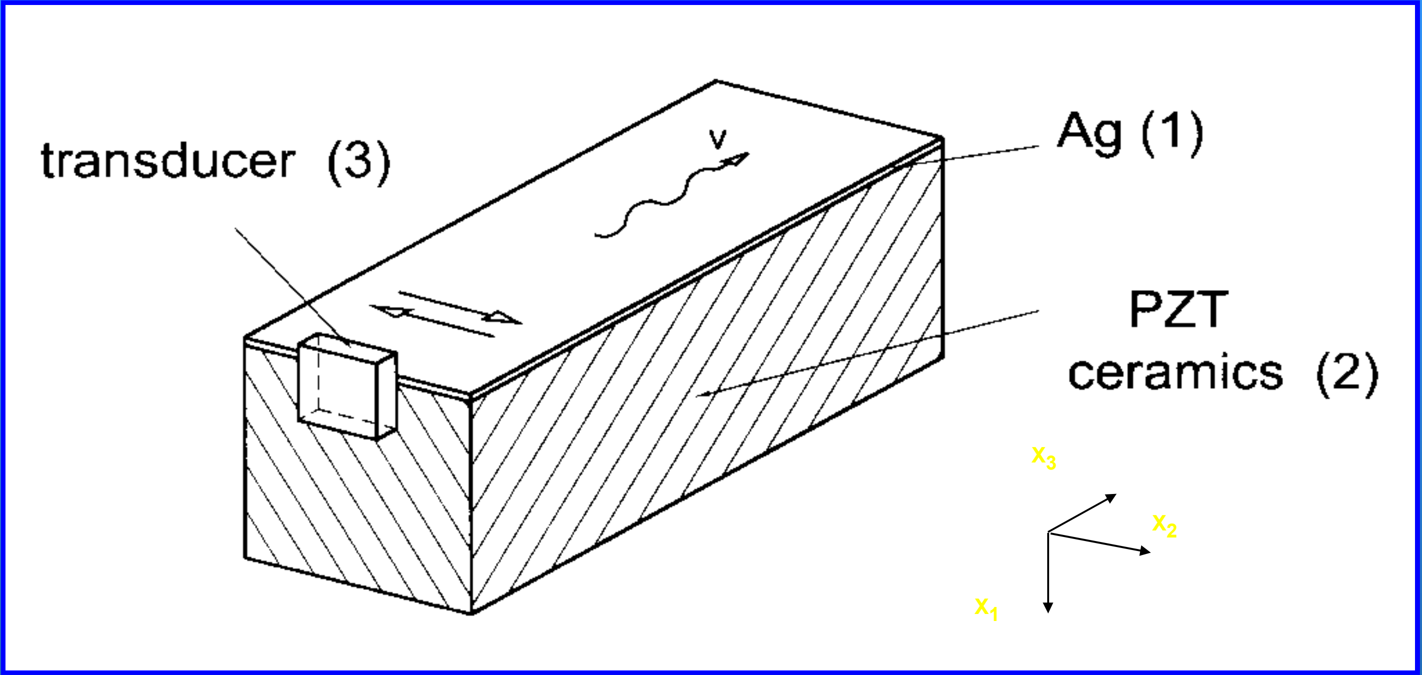


Fig.2. Excitation of the Love wave, (3) PZT plate transducer, (1) Cu surface layer, (2) steel substrate

Bleustein-Gulyaev (B-G) Waves



- Fig.3. Excitation of the B-G wave in a piezoceramic PZT waveguide (2) covered on the surface by a very thin metallic (Ag) layer (1) by means of the PZT plate transducer (3). PZT ceramics (both in the transducer and waveguide) is polarized along the axis.

x_2

Application of SH-SAW for determining the rheological parameters of liquids at atmospheric pressure

$$Z_L = \left(\rho_L \cdot G_L\right)^{1/2}$$

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

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

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

$$\Delta\gamma = -j\left(\frac{\left|v_2\right|^2_{x_1=0}}{4P}\right)Z_L = -jKZ_L$$

where: v_2 is the SH surface wave amplitude on the waveguide surface ($x_1 = 0$), P is the mean power on the unit width of the SH surface wave. K is constant depending solely on the material parameters of the waveguide and frequency

Application of SH-SAW for measuring the viscosity of liquids at high pressure

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

where: η is the viscosity, ρ_L is the density of a newtonian liquid

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

where: R_L and X_L is a real and imaginary part of the mechanical shear impedance of a liquid.

$$R_L = \frac{\ln\left(A_1^0 / A_1^1\right)}{2KL}$$

where: A_1^0 and A_1^1 represent amplitudes of the first echo of the SH surface wave for an unloaded loaded waveguide respectively, L is the length of the waveguide covered with an investigated liquid.

Advantages of the SH-SAW method for measuring the viscosity of liquids at high pressure:

- Absence of moving parts
- Operation in real time
- Short measuring time
- High sensitivity
- Low power consumption
- Small dimensions, simple and robust construction of the sensor
- Possibility of computerization
- Output signal is electrical
- No leakage problems
- No heating caused by shear

Ultrasonic set up for measuring the viscosity of liquids under high pressure

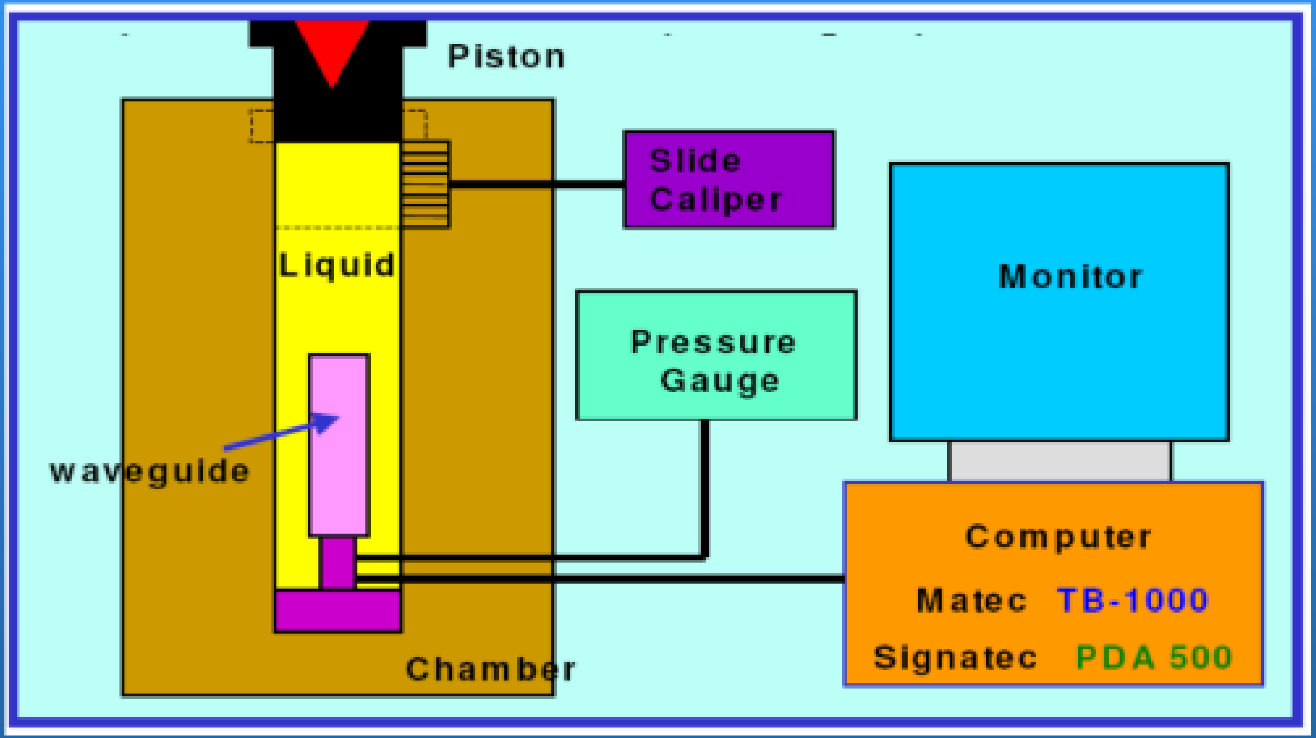
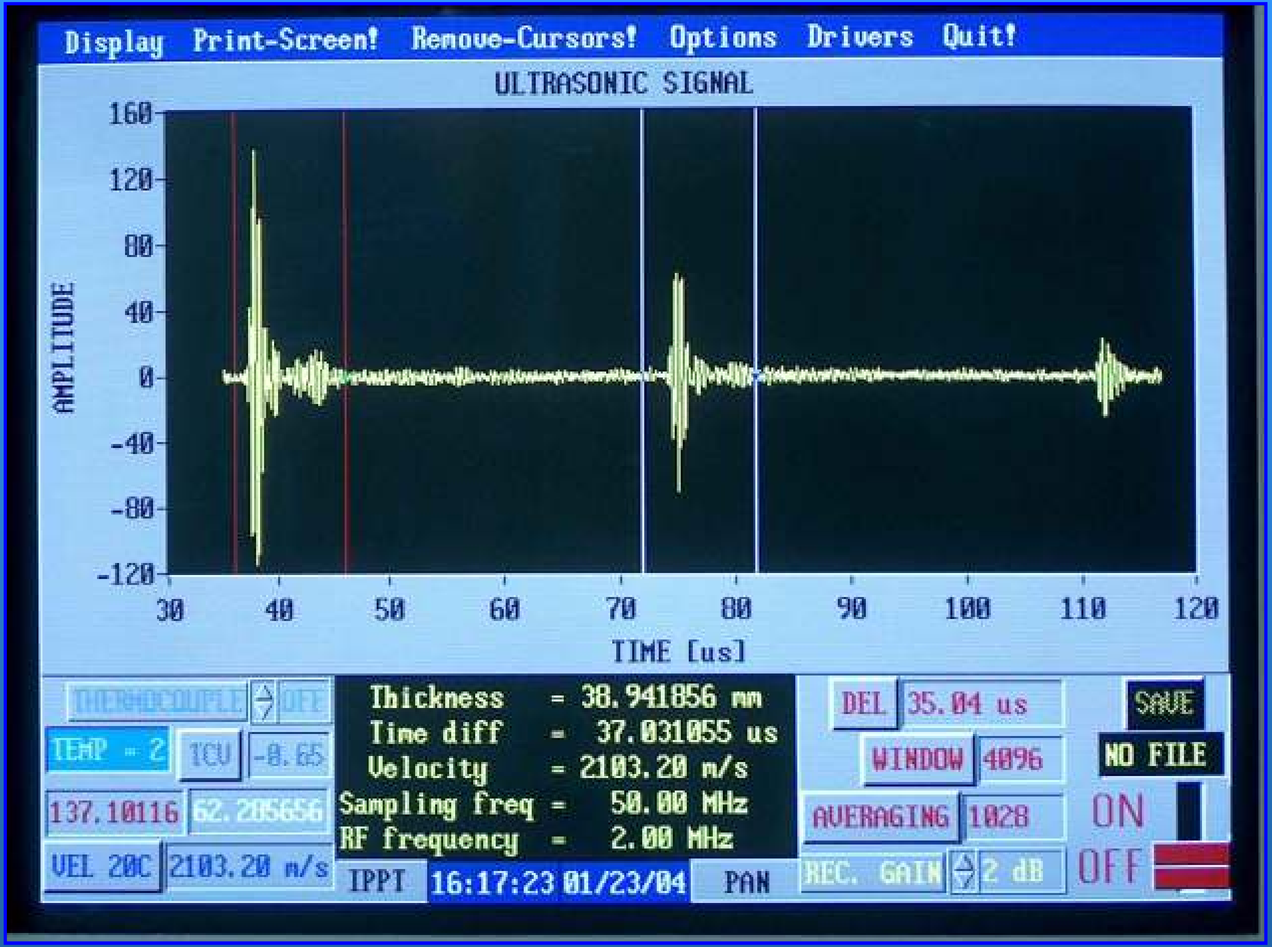


Fig.4

Ultrasonic set up for measuring the viscosity of liquids under high pressure

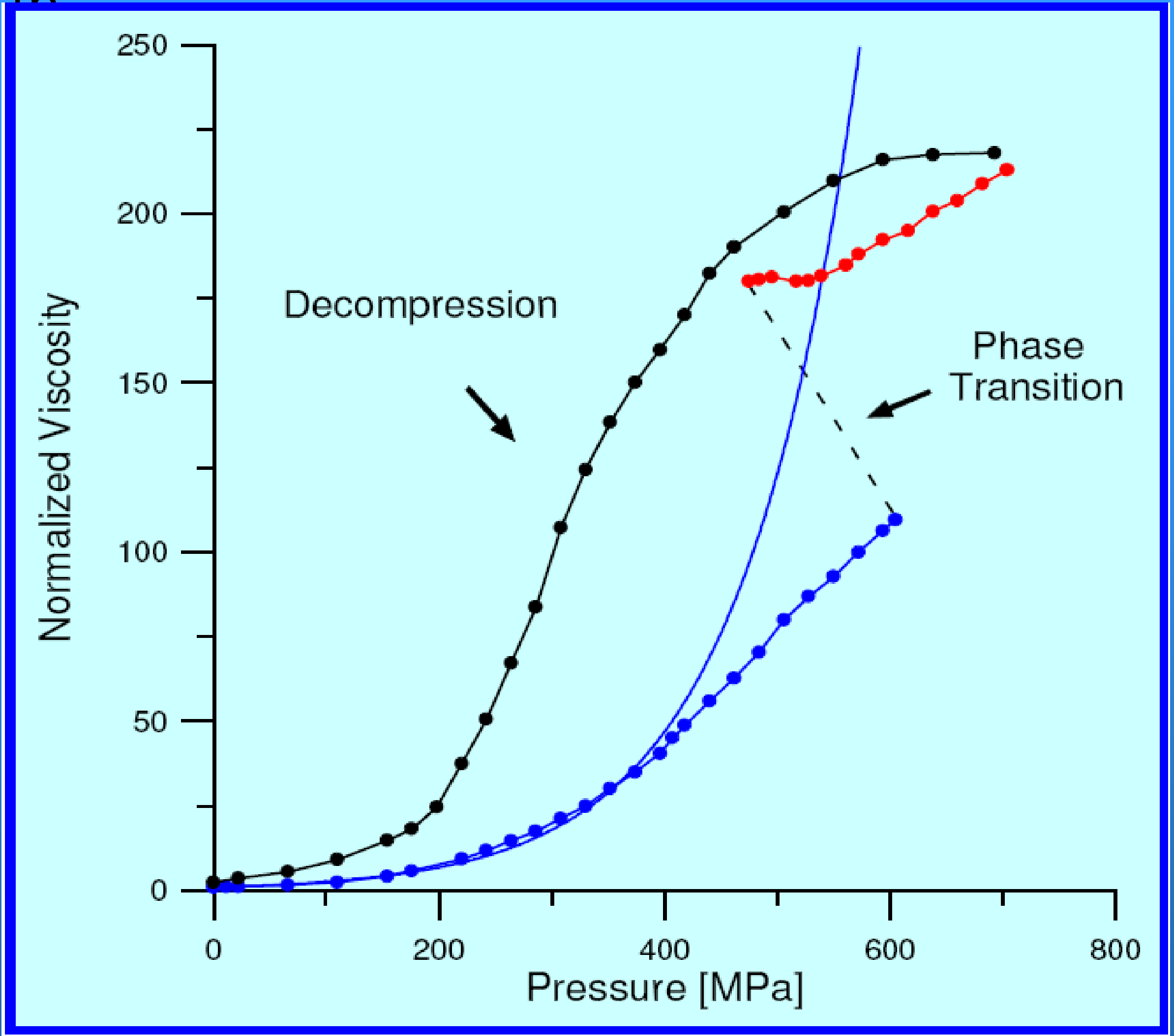
Fig.5. Love wave waveguide, Cu surface layer on a steel substrate (on the right), connected to the high-pressure lead through (on the left).



```
Thickness      = 38.941856 mm
 Time diff     =  37.031055 us
 Velocity      = 2103.20 m/s
Sampling freq  =   50.00 MHz
RF frequency   =   2.00 MHz
```

TEMP = 2
-0.65
137.10116
62.205656
VEL 20C 2103.20 m/s
16:17:23 01/23/04
DEL 35.04 us
WINDOW 4096
AVERAGING 1028
REC. GAIN 2 dB
NO FILE

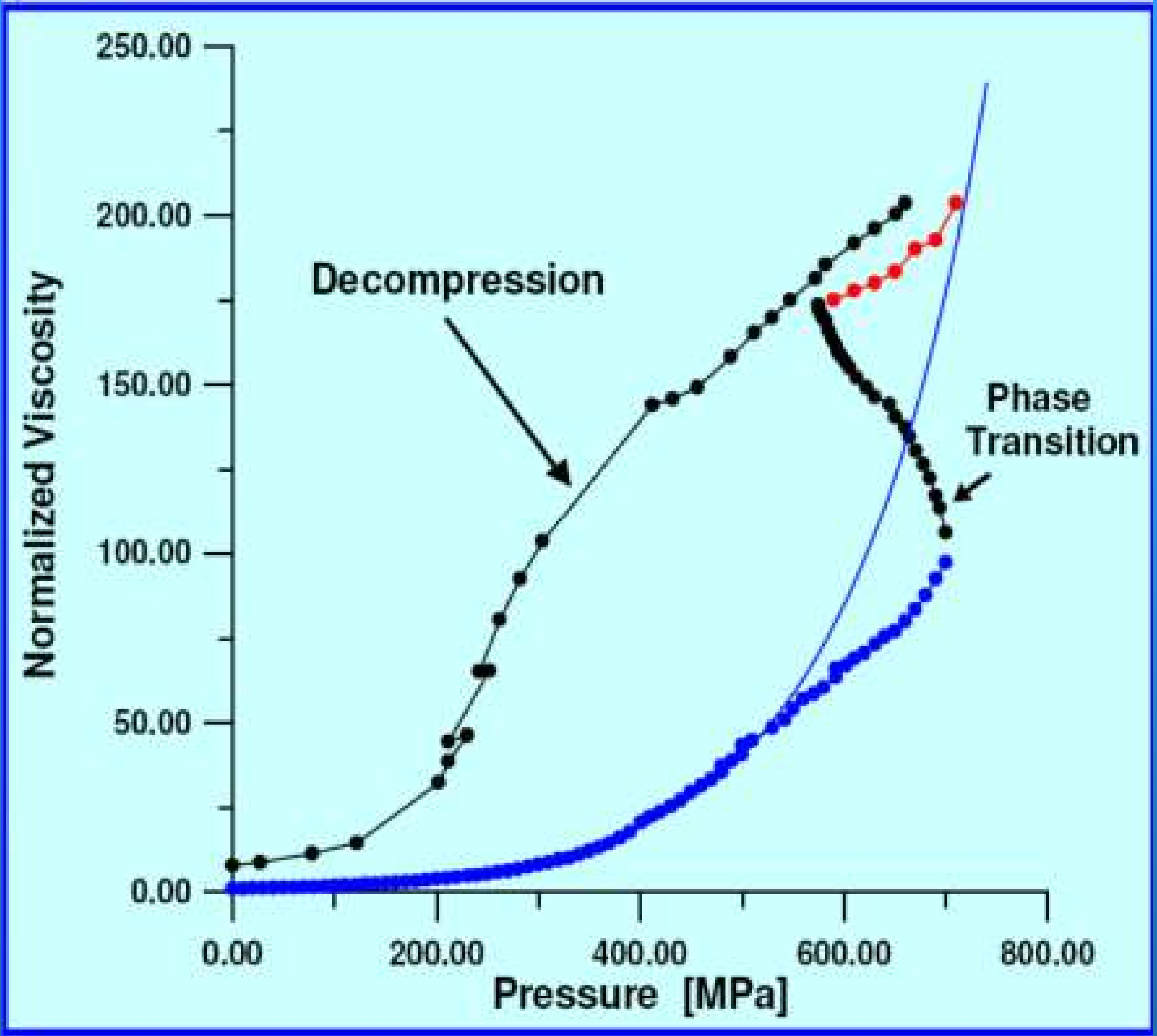
Fig.6. Oscillogram of the SH surface wave impulses reverberating in the waveguide



continuous exponential curve represents the Barus formula

$$\eta(p) = \eta_0 \exp(\alpha p)$$

Fig.7. Variations in viscosity of castor oil, as a function of hydrostatic pressure, measured by the Love wave method



continuous exponential curve represents the Barus formula

$$\eta(p) = \eta_0 \exp(\alpha p)$$

Triolein = ester =
glicerol + 3 oleic acids

The main constituent of:
vegetable oils,
animal fats

Fig.8. Variations in viscosity of triolein, as a function of hydrostatic pressure, measured by the Bleustein-Gulyaev (B-G) wave method

Measurement of sound speed in liquids at high pressure

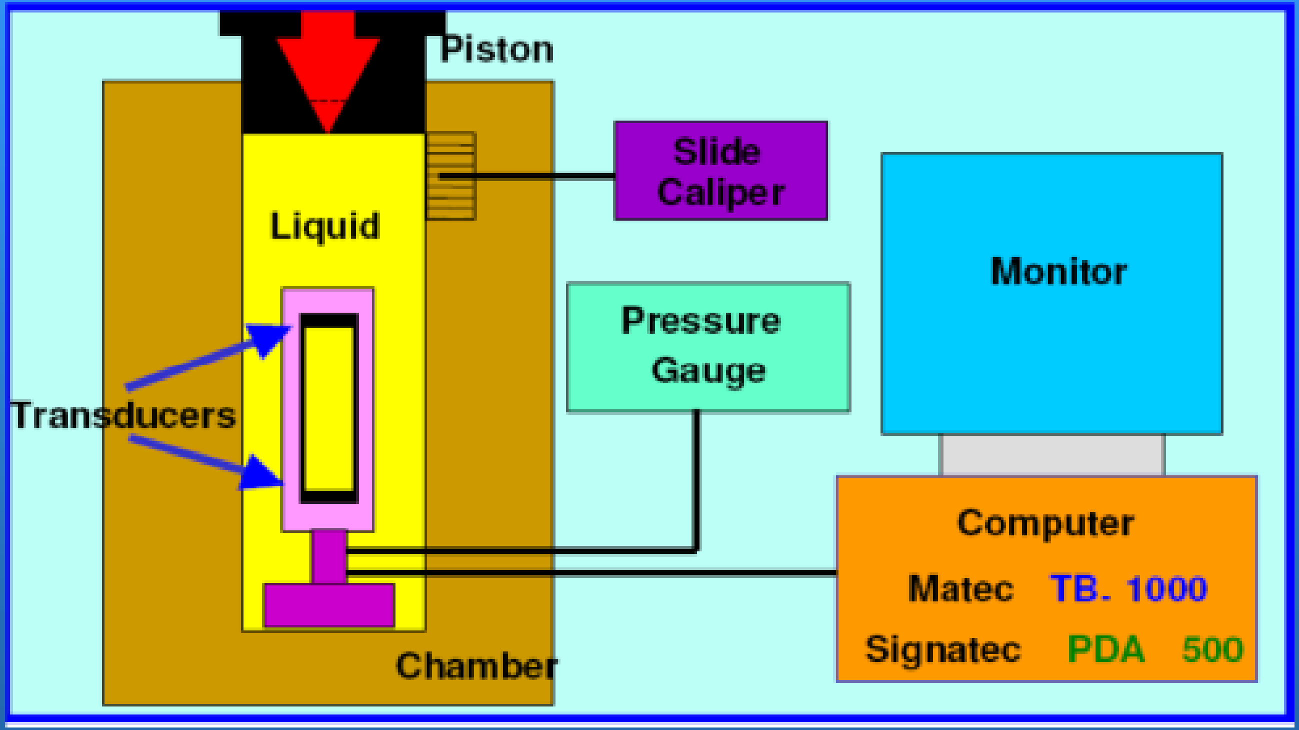


Fig.9 $LiNbO_3$ (Y_{36} cut) transducers, f = 5 MHz

Phase velocity of longitudinal acoustic waves in triolein in function of hydrostatic pressure

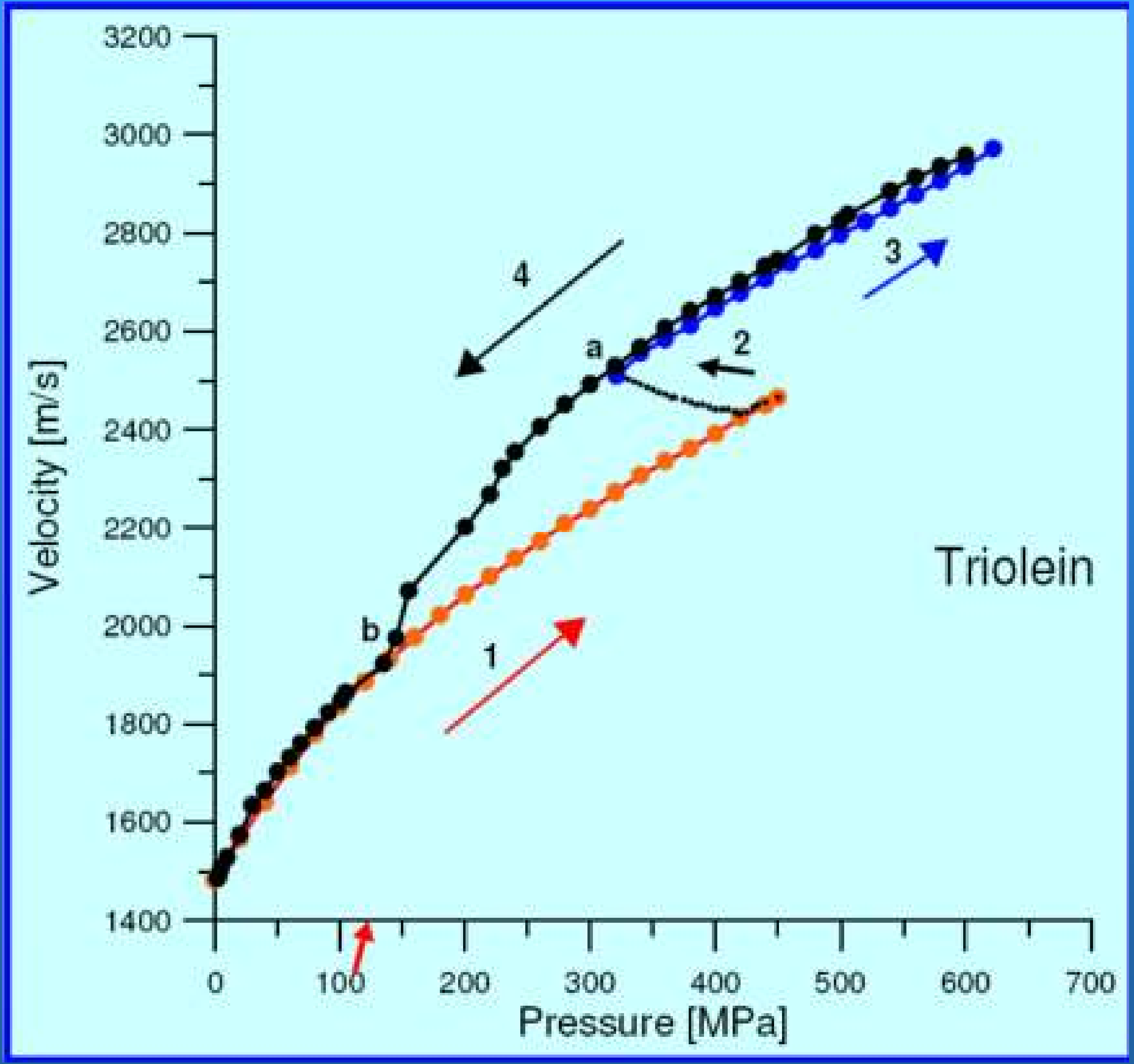


Fig.10

1 – low-pressure phase
2 – phase transition
3 – high-pressure phase
4 – decompression

Between points marked by **a** and **b** two phases coexisted in triolein.

$$c_0^2 = \frac{1}{\rho \beta_S}$$

$$\beta_S = -\frac{1}{V}\left(\frac{\partial V}{\partial p}\right)_S$$

Isothermal compressibility of triolein as a function of hydrostatic pressure

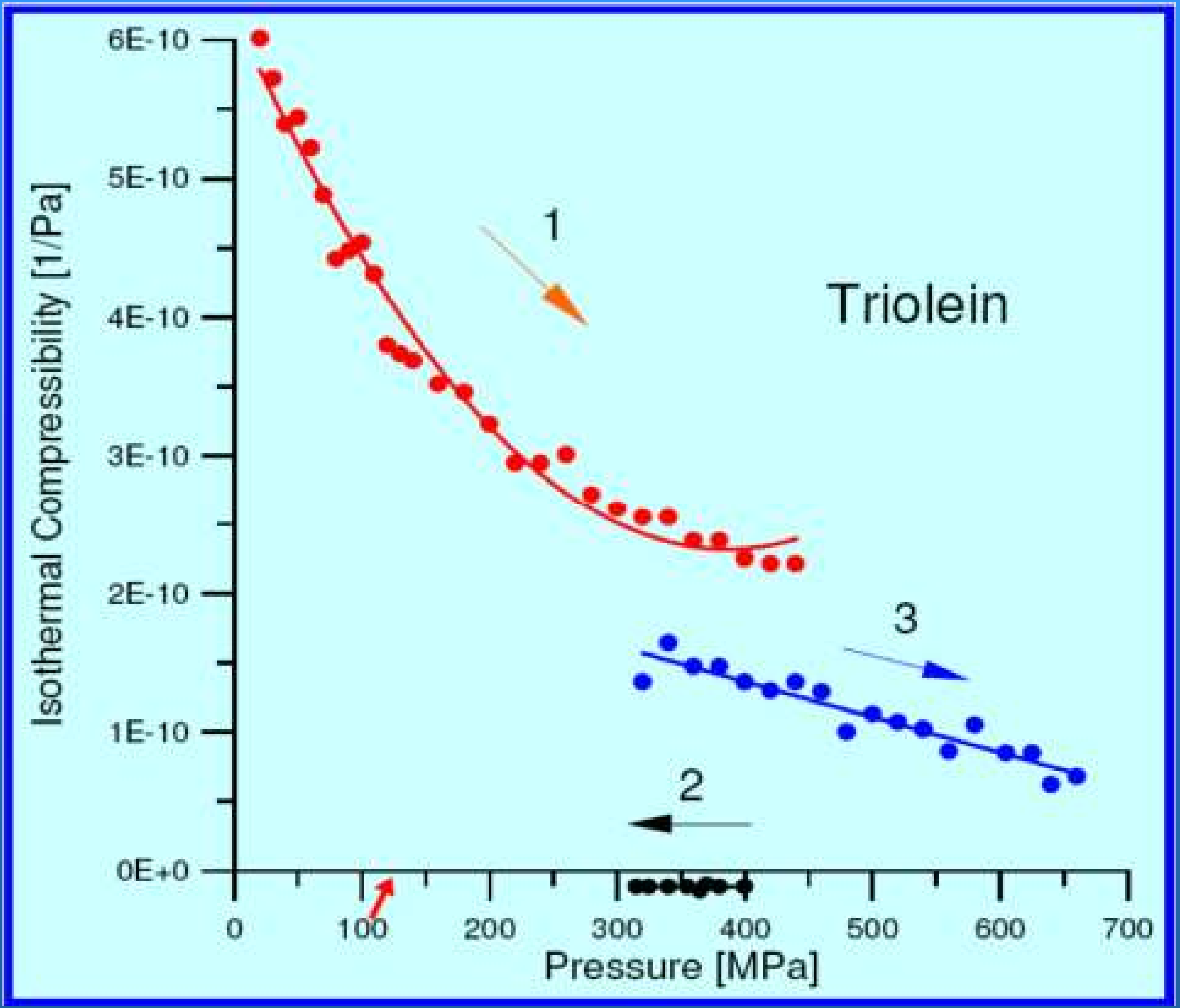


Fig.11

1 – low-pressure phase

2 – phase transition

3 – high-pressure phase

$$\beta_T = -\frac{1}{V}\frac{\partial V}{\partial p}$$

Possibility of measurement of various physical (thermodynamic) parameters

- Isothermal compressibility
- Isentropic compressibility
- Isobaric heat capacity
- Isobaric thermal expansion coefficient
- Internal pressure
- Free volume
- Non-linearity parameter B/A

$$\boxed{\frac{B}{A} = \rho_0 c_0 \left(\frac{\partial c}{\partial p}\right)_T + \frac{\beta T \rho_0}{c_P}\left(\frac{\partial c}{\partial T}\right)_P}$$

Ultrasonic set up for measuring the viscosity of liquids under high pressure at various temperatures

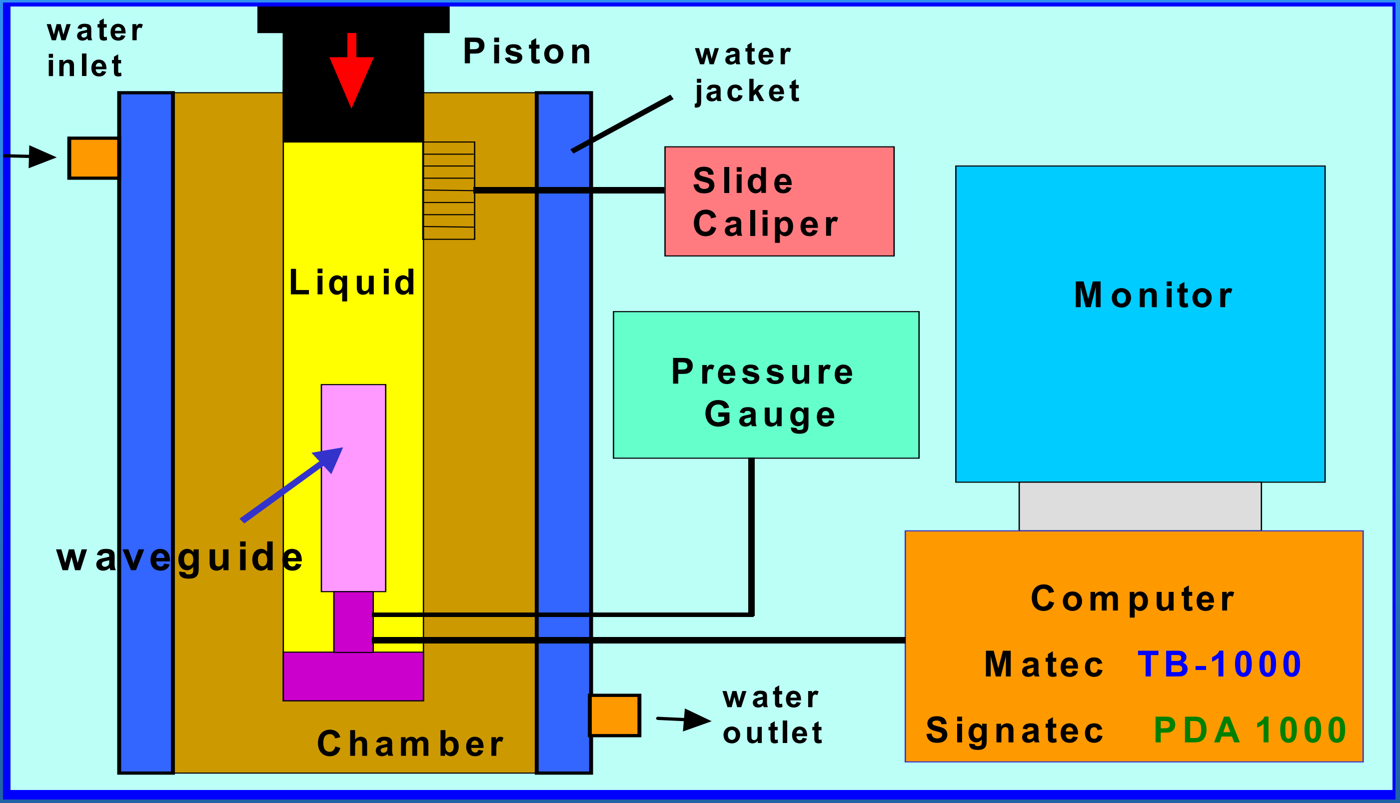


Fig.12

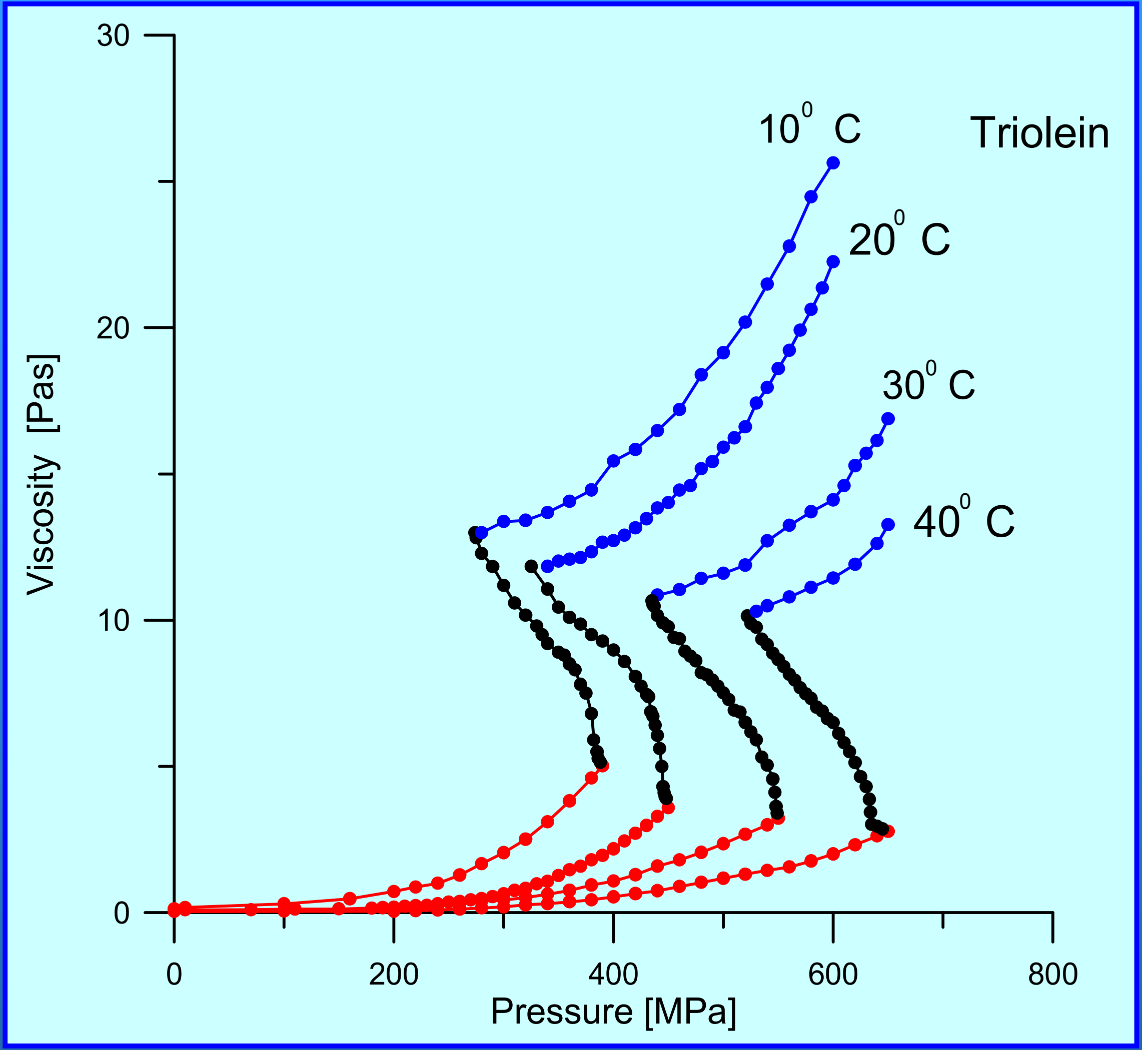


Fig.13. Viscosity of triolein versus hydrostatic pressure along various isotherms

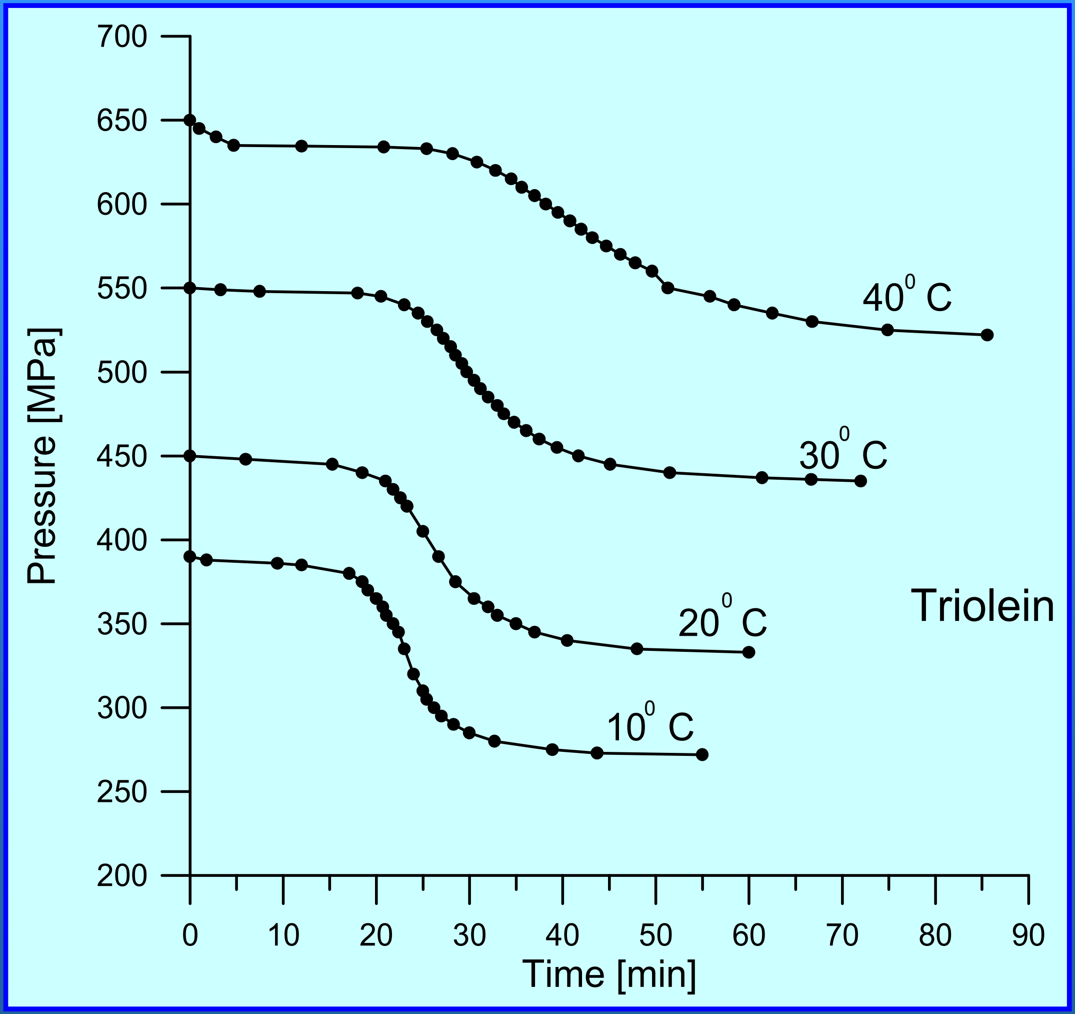
Measurement of the physical properties of liquids during phase transition

Investigation of phase transition is very important in lubricants exploitation and in food processing and conservation.

During phase transition a step change in liquid viscosity, phase velocity and compressibility occurs.

Investigation of phase transitions was impossible with conventional mechanical methods.By contrast, the presented novel SH-SAW methods enable the measurement of the rheological parameters of liquids during phase transitions.

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



The piston was locked in a fixed position

Fig.14

Conclusions

- New methods for measuring the viscosity of liquids at high pressure have been established.
- The SH-SAW viscosity sensor is electrically responsive. Owing to this fact, modern methods of the digital signal acquisition and processing can be efficiently used.
- The measuring set up operates in real time and can be employed for measuring liquid viscosity under high pressure in the course of the technological processes.
- We measured the viscosity of liquids and speed of sound not only in the exponential range but also during the phase transition, at high pressure phase and during the decompression. This is a novelty.

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