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Investigation of the Acoustic Properties of Viscosity Standards

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Longitudinal and shear ultrasonic wave velocities were measured versus temperature in the viscosity standards of Paragon S8000S, N30000S and Cannon N2700000. The measurements were performed by the through-transmission method at the frequency of 2 MHz. Ultrasonic pulses were sent via polymethyl methacrylate (PMMA) waveguides between the tips of which a small amount of the particular standard liquid was placed. The velocities of longitudinal and shear waves were determined to depend on the viscosity of the liquid and increase with the viscosity.

Keywords: viscosity standard; longitudinal and shear waves; ultrasound velocity.

1. Introduction

Viscosity and density are important parameters that characterize the physical properties of liquids. For calibration of the instruments used for measurement of those parameters, viscosity and/or density standard liquids are used. Most of these standards are manufactured by Paragon and Cannon companies (General purpose viscosity standards, High viscosity standards). The exact composition of these standards is not available. Because they are made of polymers, they must be composed of large molecules with many repeated subunits. In recent years, measurement of those quantities has been performed using acoustic and ultrasonic methods. Different types of ultrasonic waves were used: the torsional mode of waves (Kim, Bau, 1989; Ai, Lange, 2008; Rabani *et al.*, 2011); ultrasonic guided waves (Vogt *et al.*, 2004; Kazys *et al.*, 2013; 2014), Love waves (Kiełczyński *et al.*, 2012; 2014). Therefore, the acoustic properties, such as ultrasound velocities, are acquiring high importance; however, to the best of our knowledge, the velocities of longitudinal and shear waves in such liquids have not previously been measured. Propagation of ultrasonic waves in polymers is rather complicated due to a wide continuous relaxation spectrum. Usually shear waves do not propagate in liquids, however in viscous liquids it is possible to expect that they may propagate short distances. In addition, the attenuation of ultrasonic waves in such liquids may be high or even extremely high. The objective of this work is the determination of the ultrasonic longitudinal and shear wave velocities in the Paragon and Cannon viscosity standards at various temperatures.

2. Measurement method

For measurements of ultrasound velocities in liquid viscosity standards, a pulse through- transmission method was selected. The experimental set-up used for measurements is shown in Fig. 1. For this purpose, a special measurement chamber with the acoustic waveguides was developed (Fig. 1). Taking into account that the expected attenuation in the liquids un-

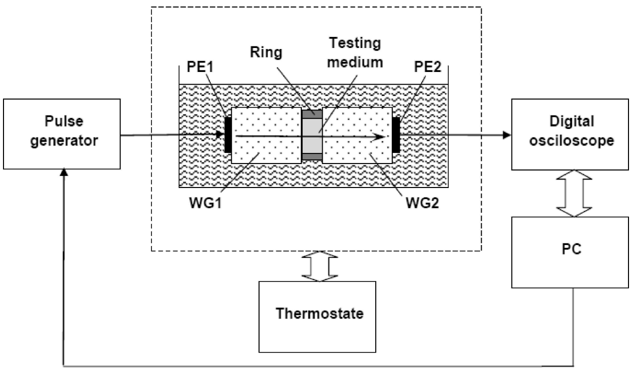

Fig. 1. Experimental set-up for measurement of the ultrasonic longitudinal and shear waves velocities in viscosity standards: PE1, PE2 – PZT 2 MHz 5 mm × 5 mm piezoelectric elements for generation and reception of longitudinal or shear waves, WG1, WG2 – PMMA 12-mm diameter waveguides, with the limiting ring having an internal diameter of 10 mm.

der investigation may be high, the propagation path in the liquids was selected to be quite short: 0.55 mm for longitudinal waves and 0.15 mm for shear waves. The length of the acoustic path in the test medium is defined by the limiting stainless steel ring inserted between the waveguides WG1 and WG2. The ring was machined by lapping and polishing machine M15 from Lapmaster&Wolters Ltd. The thickness of the ring was determined with the uncertainty ±1 µm. A very small amount of the liquid under investigation is placed inside the ring. The length of the waveguides for longitudinal waves and shear waves was $L = 15$ mm and $L = 6$ mm, respectively, and the waveguide diameter was 12 mm. The waveguides were manufactured from polymethyl methacrylate (PMMA) plastic material.

For the generation and reception of the longitudinal and shear waves, piezoelectric rectangular PZT type elements of a corresponding polarization were used. The shear wave piezoelements were cut from a 5-mm-thick PZT piezoelectric element in parallel to the poling direction. The piezoelements were ground to produce the required geometry and operation frequency of 2 MHz. The thickness of the piezoelements for the longitudinal waves and the shear waves was 0.8 mm and 0.4 mm, respectively. Subsequently, the piezoelectric elements were coated by copper electrodes using electroless chemical technology.

The piezoelectric elements were glued to the PMMA waveguides using an epoxy resin. The acoustic impedance of the PMMA material is rather low, so damping of piezoelectric elements is low as well, and the ultrasonic longitudinal and shear wave signals are rather narrowband, which enables avoiding the influence of dispersion.

The piezoelectric transducers were excited by the Agilent Technologies 81150A arbitrary pulse function generator. The excitation signal was a rectangular pulse with the amplitude of 5 V and a duration 200 ns. The duration of the excitation pulse for longitudinal and shear waves was the same. The signals were observed and measured using a Hewlett Packard 54645A oscilloscope and a PC, and the time measurement error was 1 ns. The propagation time of ultrasonic pulses was measured using the zero-crossing method.

Due to the very short propagation paths in the viscosity standards, the measurements of the delay times of the ultrasonic waves were performed via the buffer waveguides WG1 and WG2, which contacted with the medium under investigation. The velocities of the longitudinal as well as shear ultrasonic waves are determined from the following expression:

$$c = d/\Delta t, \tag{1}$$

where d is the distance between the tips of the waveguides (which is defined by the thickness of the ring), $\Delta t = t_1 - t_0$, t_0 is the delay time of the ultrasonic wave in the waveguides, and t_1 is the total delay time in the waveguides and the viscosity standard under investigation.

The pulse propagation time t_0 and, consequently, the ultrasound velocity in waveguides depend on temperature. To achieve the objective of this investigation of the measurement of the velocities of longitudinal and shear waves in a temperature range, the dependence of the propagation time t_0 versus temperature must be determined.

The pulse propagation time t_0 at different temperatures is measured when the waveguides are pressed together, i.e. the limiting ring is taken out. The acoustic chamber with the waveguides is immersed into a water bath, the temperature T of which is controlled by a thermostat with accuracy 0.1°C.

The measured reference temperature dependence $t_0(T)$ of the longitudinal 2 MHz pulse delay time t_0 in the waveguides, coupled together via zero layer of the viscosity standard, can be approximated as follows:

$$t_0(T) = 9.885 \times 10^{-3} T + 12.523, \ \mu\text{s}, \tag{2}$$

where T is the temperature in degrees of Celsius.

The measured reference temperature dependence $t_0(T)$ of the shear 2 MHz pulse delay time t_0 in the waveguides may be approximated as

$$t_0(T) = 2.8 \times 10^{-5} T^2 + 2.89 \times 10^{-3} T + 9.757, \ \mu\text{s}. \tag{3}$$

The propagation time of the ultrasonic waves in the test medium inside the plastic ring at the temperature T is found from the following expression:

$$\Delta t(T) = t_1(T) - t_0(T), \tag{4}$$

where $t_1(T)$ is the total delay time in the waveguides and the viscosity standard under investigation, and $t_0(T)$ is the delay time in the waveguides at the given temperature T. Note that the delay times $t_1(T_1)$ and $t_0(T_2)$ are usually measured at slightly different temperatures T_1 and T_2 because it is impossible using the thermostat obtain the same temperature of the waveguides during the calibration procedure and the measurements. Therefore, the exact value of t_0 at the temperature T_1 during measurements is found from Eq. (2) or Eq. (3).

Finally, the corresponding longitudinal or shear wave velocities in the viscosity standard are found from the expression:

$$c(T) = d/\Delta t(T). \tag{5}$$

3. Measurement results

Measurements of ultrasonic velocities were performed in three different viscosity standards: Paragon S8000S, N30000S and Cannon N2700000 (General purpose viscosity standards, High viscosity standards).

The properties of those standards at 20°C are presented in Table 1. The small quantity of the viscosity standard is placed inside the limiting ring while being careful (Fig. 1) not to overfill the volume; otherwise, the actual thickness of the viscosity standard may be incorrect.

Table 1. Properties of investigated viscosity standards at 20°C.

Viscosity standard	Viscosity η [Pa·s]	Density ρ [g/cm^3]
Paragon S8000S	31.43	0.891
Paragon N30000S	115.16	0.897
Cannon N2700000	7900	0.911

The waveforms of the longitudinal and shear wave pulses transmitted via the waveguides and the viscosity standard are shown in Figs. 2a and 2b, respectively. The presented waveforms indicate that even in the case of the shear waves, the amplitude of the pulse transmitted via the viscosity standard is suitable for accurate delay time measurements. The amplitude of the shear wave pulse transmitted through the viscosity standard Paragon S8000S reduces only 1.35 times in comparison with the shear wave pulse transmitted directly via the waveguides.

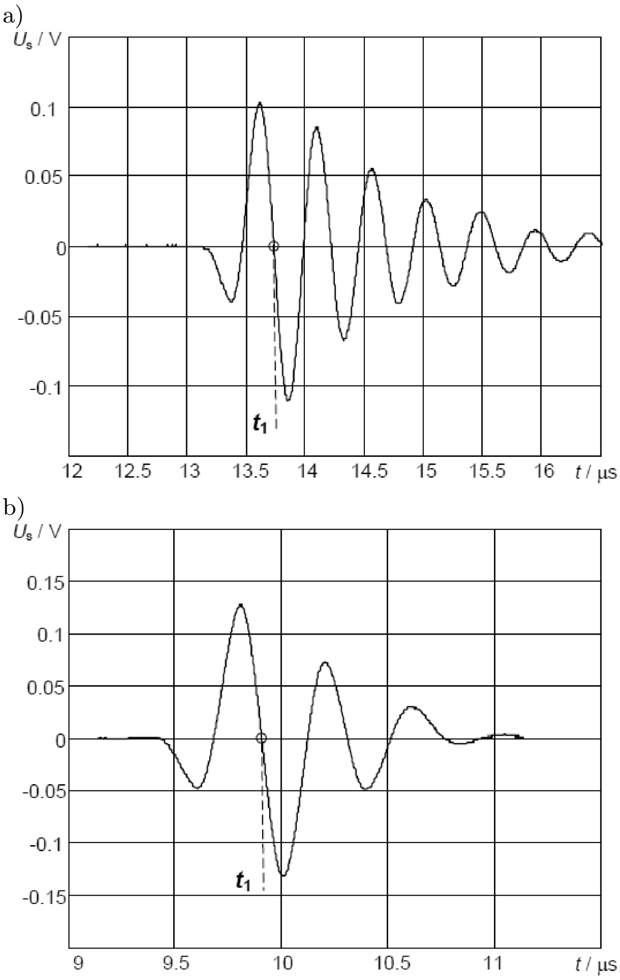

Fig. 2. Waveform of the 2 MHz wave pulse transmitted via the waveguides and the layer of the viscosity standard Paragon S8000S: a) longitudinal wave; thickness 0.55 mm; b) shear wave; thickness 0.15 mm.

The measured ultrasonic longitudinal wave velocities versus temperature in all three viscosity standards are shown in Fig. 3a. The viscous liquids at the beginning were heated up to the 40°C and after that slowly cooled down. The measurements were performed at manually selected time instants. The estimated measurement uncertainty in all cases was less than 1%. The measurement error due to diffraction may be neglected as the measurements are in principle differential.

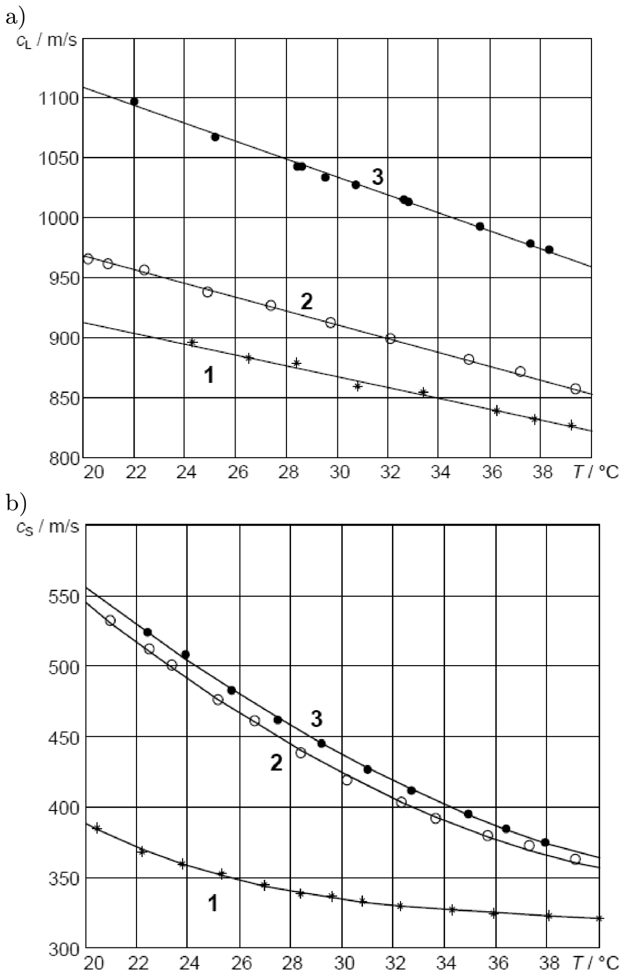

Fig. 3. Temperature dependencies of ultrasonic 2 MHz wave velocities in the viscosity standards: 1 – Paragon S8000S, 2 – Paragon N30000S, 3 – Cannon N2700000. a) Longitudinal wave velocity c_L; b) shear wave velocity c_S.

From the measurement results, the velocity of the longitudinal wave c_L in the investigated temperature range is found to increase with the viscosity of the liquid. Considering that the viscosity reduces with the increased temperature, the ultrasound velocity reduces as well.

The velocities of ultrasonic longitudinal waves versus temperature in the Paragon S8000S, Paragon

N30000S and Cannon N2700000 viscosity standards may be approximated by the following linear dependences of Eqs. (6), (7), and (8), respectively:

$$c_L(T) = -4.540 \times 10^{-3}T + 1003.4, \tag{6}$$

$$c_L(T) = -5.689 \times 10^{-3}T + 1081.5, \tag{7}$$

$$c_L(T) = -7.358 \times 10^{-3}T + 1254.2. \tag{8}$$

The ultrasonic longitudinal wave velocities at 20°C and the corresponding temperature coefficients are presented in Table 2.

Table 2. Longitudinal wave velocity data in the standards of viscosity.

Viscosity standard	Velocity c_L at 20°C [m/s]	Temperature coefficient $\Delta c/\Delta T$ [m/s/°C]
Paragon S8000S	920	−4.54
Paragon N30000S	970	−5.69
Cannon N2700000	1145	−7.36

The measurement results of the shear waves are shown in Fig. 3b. The velocity of the ultrasonic shear wave c_S in the investigated standards is also higher in more viscous liquids and decreases with an increase in temperature. However, unlike the case of the linear dependence of the longitudinal waves, the temperature dependence of the ultrasound velocity is not linear.

The ratio of shear and longitudinal wave velocities at a few different temperatures is presented in Table 3.

Table 3. Ratio of ultrasonic shear to longitudinal wave velocities.

Viscosity standard	Ratio c_S/c_L		
	20°C	30°C	40°C
Paragon S8000S	0.42	0.39	0.39
Paragon N30000S	0.56	0.46	0.42
Cannon N2700000	0.49	0.42	0.37

4. Conclusions

For the first time, the ultrasound velocities of longitudinal and shear waves in the viscosity standards of Paragon S8000S, N30000S and Cannon N2700000 were measured as a function of temperature using the through-transmission method. Taking into account that expected attenuation of ultrasonic waves may be high, or in the case of shear waves extremely high, measurements were performed at very short distances between the tips of the transmitting and receiving waveguides. This distance in the case of longitudinal waves was 0.55 mm, what at the operation frequency $f = 2$ MHz and room temperature is in the range (0.9–1.1) λ_{L}, where λ_{L} is the wavelength of the longitudinal wave in the viscosity standard. Respectively, the measurement distance for shear waves was selected 0.15 mm, what for above mentioned conditions corresponds to (0.6–075) λ_{SH}, where λ_{SH} is the wavelength of the shear wave. The experiments have proved that a high viscosity of the liquids and the short measurement distance enabled transmission and delay time measurements of the shear wave pulses.

It was found that both velocities reduce with the increasing temperature. The temperature dependence for longitudinal waves is linear, but for shear waves is essentially non-linear. On the other hand from the measurement results follows that the velocities of both types of waves are directly proportional to the viscosity of the liquid. It means, for a given viscous liquid ultrasound velocity measurements may be used for monitoring of the liquid viscosity. Taking into account that ultrasound velocity measurements may be performed with a high accuracy the viscosity may be determined with a high accuracy as well.

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