An ultrasonic shear wave viscometer for low viscosity Newtonian liquids

OPEN ACCESS
IOP Publishing
Measurement Science and Technology
Meas. Sci. Technol. **32** (2021) 125305 (11pp)
https://doi.org/10.1088/1361-6501/ac200f

An ultrasonic shear wave viscometer for low viscosity Newtonian liquids

S Mastromarino[1], R Rook[1], D De Haas[1], E D J Verschuur[2], M Rohde[1,*] and J L Kloosterman[1]

[1] Department of Radiation Science and Technology, Delft University of Technology, Mekelweg 15, Delft, JB 2629, The Netherlands
[2] Departments of Imaging Physics, Delft University of Technology, Lorentzweg 1, Delft 2628 CJ, The Netherlands

E-mail: M.Rohde@tudelft.nl

Received 2 March 2021, revised 19 July 2021
Accepted for publication 23 August 2021
Published 17 September 2021

Abstract

A method based on ultrasonic wave propagation is applied for the determination of the viscosity of low viscous liquids. A waveguide is used to remotely transmit the ultrasonic waves from a shear piezoelectric transducer into the liquid. At the solid–liquid interface, a guided wave mode, the shear mode, is used to extract the liquid viscosity. The energy of the reflected ultrasonic wave depends upon its operating frequency, the physical properties of the liquid (viscosity and density), and the waveguide (density and shear modulus). The results show that the attenuation of the waves, and thus the viscosity of the liquid, can be retrieved using this method. Measurements on water, ethanol, and mixtures of water/glycerol illustrate that the method can monitor changes in attenuation due to the viscosity of the liquid. The range of viscosities measured was between 0.8 and 60 mPa s. Compared to literature values, the relative error for these measurements was lower than 12% while the uncertainty in the measurements was lower than 5%. Besides its ability to measure low viscosities, this method offers advantages such as the capability to perform *in-situ* measurements of liquids in harsh environments, the omission of mechanical parts, and the possibility to handle small volumes of liquid. These features make this method suitable for low viscous liquids that are radioactive, corrosive and at high temperature.

Keywords: ultrasonic viscometer, shear waves, guided waves, acoustic bulk attenuation, viscosity

(Some figures may appear in colour only in the online journal)

1. Introduction

In many industrial applications one can find low viscosity liquids in harsh environments, such as high temperatures, high corrosion or high radioactivity. One example is the nuclear fuel used in molten salt nuclear reactors. Measuring the viscosity of liquid nuclear fuels is difficult because of the high temperatures and levels of corrosion and radioactivity. These conditions impose severe requirements on the measuring equipment: a remote measurement where the salt is not in close contact with the electronic instrumentation is preferred.

A technique that may overcome the aforementioned difficulties consists of the use of ultrasounds in a waveguide. The usage of ultrasound for measuring material properties started in 1949 with Mason *et al* who placed the transducer in direct contact with the liquid [1]. This is, however, not possible for measuring corrosive liquids that can destroy the piezoelectric element in the transducer. The high temperature is also a limitation for the transducer. Above the Curie temperature of the transducer, the piezoelectric activity is lost and high-temperature transducers are not yet available [2]. Therefore,

* Author to whom any correspondence should be addressed.

a waveguide is used to separate the transducer from the liquid.

In 1971, Hunston *et al* [3] proposed the measurement of the shear properties of liquids using a strip waveguide immersed in a liquid. In their experiment, an aluminum strip is connected to a transducer and placed in a empty cell. The cell is then filled with the liquid sample to be measured until the sample touches the transducer. The change in attenuation of the waves is measured when the strip is in air and then when it is immersed in the liquid sample. The attenuation is expressed in terms of the viscosity. With this technique, Hunston measured a viscosity as low as 3.6 mPa s with an error respect to the literature value between 30% and 40%. Knauss *et al* [4] repeated the measurement proposed by Hunston to obtain a better accuracy of the results. Through the use of a longer waveguide and a different frequency, a higher attenuation was achieved and the measurements of the viscosity had a maximum relative error of 11%. The method proposed by the group of Hunston and Knauss is based on the attenuation of the wave in a waveguide completely immersed in the fluid. In their system the transducer is in contact with the fluid. This makes this system not suitable for corrosive fluids or at high temperature.

The potential applications of ultrasounds to measure viscosity at high temperature was first reported in 1996 by Shah [5]. Shah measured liquids with viscosities between 2 and 10 Pa s up to 60 °C. Prasad *et al* [6] have reported ultrasonic studies based on longitudinal waves on glass melts using a measuring system comprising ultrasonic waveguides of alumina. They have developed an experimental apparatus to measure, *in situ*, the viscosity of molten glass at elevated temperatures (up to 1250 °C). In their method, guided waves are generated through a cylindrical buffer rod and the reflection coefficient is measured. From the relation between the reflection coefficient of a wave at a solid-liquid interface and the acoustic impedance of a liquid, the product of density and acoustic velocity, the viscosity can be obtained knowing the density of the liquid [7]. Since the impedance determines the ability of an ultrasound wave to pass from one medium to another, in the reflection coefficient method, attention should be given to the choice of the material of the buffer rod. The effectiveness of this measurements method is reduced when there is a mismatch of the impedance between the solid and the liquid, which reduces the possible materials for the buffer rod suitable for low viscous liquids at high temperature.

More recently, Vogt *et al* [8] measured the viscosity of glycerol (1.15 Pa s) and the commercial Cannon VP450 viscosity standard (1.42 Pa s) measuring the change in the attenuation coefficient, as proposed by Hunston, but using a cylindrical wire as the waveguide. A rectangular waveguide was used instead by Cegla [9], who obtained the density and viscosity of a liquid simultaneously by measuring the attenuation and the velocity of the guided waves [10]. Both Cegla and Vogt have measured the viscosity of different liquids, from 1 to 13 Pa s with errors relative to the literature values of maximum 4%.

Gitis *et al* [11] also used the measurement of the attenuation coefficient, but proposed an alternative approach to using a rectangular waveguide. Gitis *et al* identified the limit of the method in placing a transducer on the strip end surface of the waveguide. The attenuation, and the accuracy of the measurement, can be increased if the surface of contact between the piezoelectric element and the waveguide is higher. Gitis *et al* designed a set up in which the waveguide is shaped as a ribbon and it is hermetically sealed in a vessel filled with the liquid sample. The intensity of the waves and their attenuation is optimized by using an angle ultrasonic transducer glued to the waveguide, obtaining a higher surface of contact with the waveguide. The difference in amplitude between the signal when the vessel is empty and then filled with the sample is used for the measurement. With this method Gitis *et al* measured the viscosity of liquids as low as 1 mPa·s with a relative error of 1%. A substantial increase in the accuracy was achieved for the attenuation method for measuring the viscosity. The disadvantage of this method for measuring aggressive liquids is that the waveguide has to be completely immersed in the liquid, therefore a large volume (more than 200 ml approximately) is needed. When measuring molten salt fuels, for example, a large volume has to be avoided because of their high radioactivity.

Table 1 summarizes the previous measurements that have been done using a waveguide for measuring the viscosity of fluids. These works have been based on the measurement of either the reflection coefficient at the solid-liquid interface or the attenuation of the longitudinal, torsional or shear modes in cylindrical or rectangular waveguides when immersed in a liquid sample. Both the techniques have been successful in different ranges of viscosity reporting results with relative error as low as 1% [11]. While the reflection coefficient technique has the difficulty of finding a waveguide that is compatible to the liquid, the attenuation measurements have met the difficulty to reach the necessary accuracy of the measurement. For many of these techniques a large amount of liquid is needed, an incompatible characteristic for harsh liquids.

In this work we have chosen to measure the viscosity from the attenuated shear waves in a plate waveguide for several reasons. First, the shear waves do not show dispersion, the wave packets do not change phase velocity while travelling along the waveguide and maintain their shape [8]. Second, these waves do not convert into other modes when the plate is immersed in a liquid, making the interpretation of the measured signal easier [9]. Third, the shear waves are highly attenuated by the viscosity of the liquid resulting in a high sensitivity to low viscous liquids [13]. The advantage of the set up proposed in this work is that a very thin plate and a small amount of liquid are used for the measurement, differently than the set up proposed by [11]. The set up proposed is very similar to the one used by [9], but we used this method for measuring low viscous liquids.

This non-destructive method is based on wave pulses of short ultrasonic waves at high frequencies (MHz range) travelling through a plate that is partially immersed in the liquid. The piezo-electric transducer is placed on top of the waveguide, and is, thus, physically separated from the liquid, eliminating problems that might arise from a liquid under harsh conditions. The distance of the transducer from the liquid and the small interface between transducer and plate waveguide will allow the transducer to remain at low temperature. By doing

Table 1. Previous measurement of the viscosity using a waveguide.

	Hunston (1972) [12]	Knauss (1973) [4]	Shah (1996) [5]	Vogt (2004) [8]	Cegla (2005) [9]	Prasad (2008) [6]	Gitis (2012) [11]
Viscosity (Pa s)	0.00364–100	0.001–0.02	1.994–10.45	1	1–13	18–6300	0.001–1
Error (%)[a]	30–40	11	3	2	4	3	1
Method	Attenuation of the SH0 mode	Attenuation of the SH0 mode	Reflection coefficient technique	Attenuation of T(0,1) and L(0,1)	Attenuation of the Quasi-Schoelte mode	Reflection coefficient technique	Attenuation of T(0,1) and L(0,1)
Frequency (MHz)	1.6	2.1	1–2.25	0.25–0.4	0.5	0.25	2–5
Fluid	Polydimethyl-siloxilane, toluene solution of polystyrene	Toluene, water, propanol, dibutyl phthalate, diethyl phtalate	Calibration fluids	Glycerol (1% water) and Cannon VP450	Glycerol, honey, water with suspensions	Glass	40%, 50% and 60% solutions of saccharose in water
Waveguide	Aluminum plate (1.1 mm thick)	Aluminum plate (1.1 mm thick)	Plexiglas, graphite, aluminum plates	steel rod (0.25 to 2.5 mm radius)	Steel plate (1 mm thick)	Aluminum rod (4.75 mm radius)	Aluminum plates (0.3 × 140; 0.2 × 200 mm)
Temperature (° C)	30	25	30–60	Room temperature	Room temperature	Up to 1250	20–40

[a] The error is relative to the literature value.

measurements at different immersion depths the determined attenuation of the wave pulse can be used to derive the viscosity. The attenuation is measured while the waveguide is immersed in the liquid, therefore it is easier to perform an experiment varying the temperature.

In the section 2 of this paper, the theory on which the ultrasonic technique is based is described. A more descriptive section is dedicated to the shear horizontal wave attenuation method since this is the chosen mode for the measurements presented here. Then, in section 3, the experimental set-up and procedure are described. Section 3.2 is dedicated to the data processing and section 3.3 to the determination of the accuracy of the method. Results are shown in section 4, followed by conclusions.

2. Background

The principle used for evaluating the viscosity of liquids is based on a high frequency wave that is transmitted in a solid guide and attenuated by the shear motion within the liquid in which the guide is immersed. A transducer generates a wave and sends it through the waveguide. The wave's mode can be generally of two different types (longitudinal and shear), depending on the way in which the piezoelectric material inside the transducer is cut. While the longitudinal waves transmit in liquid, gas and solids, shear waves can be sustained by solids only [14, 15]. When using a waveguide embedded in another material, the shear waves can be transmitted into the waveguide and their energy can partially leak towards this material via their mutual interface and be attenuated.

When the embedding material is a liquid, the waves are transmitted in the liquid as bulk waves [16]. In this case there is a distinction between the transmission of waves that can occur if the liquid is viscid or inviscid. In inviscid liquids, shear stresses do not exist and only longitudinal waves can be transmitted [17]. When the plate is immersed in a viscous liquid, shear stresses may appear as well in the surrounding liquid and the guided waves, both longitudinal and shear, propagating in the plate can be attenuated due to viscous dissipation [10].

The shear wave that is excited in this work is the plate mode commonly referred to as SH0 mode. This mode does not introduce longitudinal waves in the surrounding liquid. Therefore, shear is the only mechanism that causes energy loss in the plate. The rate of the attenuation of the shear wave depends on the material properties of the waveguide and of the surrounding liquid, together with the frequency of the propagated wave. By measuring the attenuation of the reflected shear wave in the plate, the viscosity of a Newtonian fluid can be calculated by the approximation derived by Cegla *et al* [18],

$$\alpha = -\frac{1}{2\,h}\left(\frac{2\,\rho_f \omega \eta}{G \rho_s}\right)^{1/2}, \tag{1}$$

where α is the attenuation of the wave, h is the thickness of the plate, ρ_f is the liquid density, η is the liquid dynamic viscosity, G is the shear modulus of the plate, ρ_s is the plate density and ω is the angular frequency of the shear wave. This formula works well if the frequency thickness, being the product $h \cdot \omega/2\pi$, is up to about 2 MHz mm. Above this value, the SH0 mode shape becomes frequency dependent. Cegla *et al* [18] showed that the deviation between equation (1) and its exact solution is 0.04% when the frequency-thickness is 1 MHz mm. For this reason, this value for the frequency-thickness will be used in this work.

The dissipation of the shear waves in the liquid depends on the viscosity, and thus on the shear stress. It can be shown that the attenuation shows an exponential decay along the direction of propagation, i.e. $\sim e^{-\alpha x_1}$. From the derivation of the velocity profile in the liquid one can recognise a length scale over which the shear waves propagate in the liquid perpendicularly to the plate surface. This length scale is the viscous skin depth of the wave that defines the distance a wave will travel before the amplitude of the wave decays with a factor of $1/e$. The viscous skin depth is defined as:

$$\delta = \left(\frac{2\eta}{\rho_f \omega}\right)^{1/2}. \tag{2}$$

For example, the measurements in water with a viscosity of ca. 1 mPa s and a density of 1000 kg m^{-13} using a transducer with a frequency in the MHz range, result in a the viscous skin depth in the µm range. The same is valid for the highest viscosity we measured (∼50 mPa s). This means that the container holding the liquid can be very small and will allow measurements with merely a few ml of liquid.

3. Experimental setup

The setup in figure 1(a) was designed to excite and transmit shear waves through a waveguide, being a metallic plate, immersed in a liquid of which the viscosity has to be measured. In this setup an ultrasonic wave is generated by a transducer placed at one end of the waveguide. The transducer converts the electrical signal into a mechanical signal, a displacement wave, which is transferred to the waveguide. A shear transducer V-155RM by Olympus (Normal incidence shear wave) with a peak frequency of around 5 MHz and a diameter of 13 mm is used for the production of shear waves. The transducer is clamped at the top of the plate and is used to excite SH (shear horizontal) waves, with displacements in the x_1x_2 plane. Note that by rotating the transducer around the x_1 axis, by $\pi/2$ rad, the polarisation of the waves can be chosen to flexural waves if desired (not in this work). The transducer is such that the direction of the polarization of the shear wave is in line with the right angle connector. After transmission to the plate, the wave moves along the x_1 direction. Its path should be free of obstructions that may cause distortion of the wave. As such, the clamps (indicated by Clamping in figure 1(a)) are put at some distance from the middle of the plate to prevent the distortion. By putting pressure at the edges and at the centre of the plate, it was tested if the clamps would be of influence on the wave attenuation.

No change in the amplitude of the reflected wave was observed when pressure was applied on the edges of the plate, while the wave was strongly attenuated when pressure was applied in the centre, confirming that the clamping can be

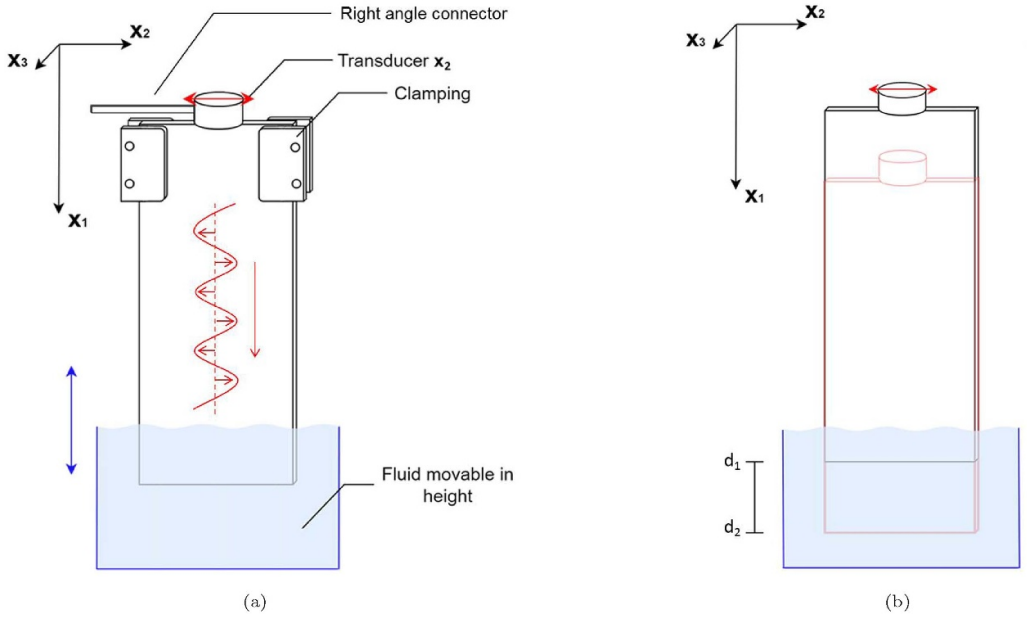

Figure 1. Schematic of the waveguide immersed in liquid. The clamping is shown (a) as well as the mechanism of immersion depth (b).

safely attached to the sides of the plate. To install the transducer, pressure is applied downwards on top of the transducer, after which it is clamped on the sides and the pressure on the top is released. To enhance traction between the transducer and the very thin plate (0.1 mm thickness), a shear couplant SWC-2 by Olympus (couplant and adaptors) is used. The connection between the transducer and the plate is essential in the stability of the transmitted signal. This connection depends on the alignment of the transducer on the place. To obtain an intense signal, the middle of the transducer has to be exactly on the place as this maximizes the contact area between the transducer and the plate.

Figure 2 shows the experimental setup. Connected to the transducer, an arbitrary waveform generator DG1022A by Rigol (Arbitrary Waveform Generators, AWG) is used to generate a sinusoidal wave packet of 40 cycles with a frequency in the ultrasonic range. Each wave packet consists of 40 cycles to assure that a sinusoidal wave is obtained at a constant chosen frequency. A wave packet consisting of few cycles shows an exponential decay that interferes with the measurement of the attenuation. The wave packet is repeated with a delay of 30 ms. This delay is needed as the waves get reflected many times in the waveguide before they are fully attenuated. The amplitude of the generated signal is chosen as high as possible, to get the highest signal to noise ratio. The amplitude of the generated signal can be limited by a couple of things: the generator, the amplifier, the oscilloscope, and the transducer. The signal generator has a maximum output voltage of 10 Vp-p (Peak-to-Peak Voltage). The transducer is preferably used with no voltages higher than 100 V. If the transducer is used a long time at continuous excitation voltages of 300 V or higher, it could be re-poled to a longitudinal wave transducer. This is an irreversible effect, so good care has to be taken in not exposing the transducer to these voltages continuously. The oscilloscope used is the Keysight InfiniiVision DSOX2024A, which has a maximum input voltage of 200 Vp-p. The maximum voltages for the transducer and the oscilloscope are higher than the maximum output voltage of the signal generator. Therefore, a radio frequency amplifier 2100 L by E & I (E. I. Innovation, 100 W, 10 kHz–12 MHz RF amplifier) is used to amplify the signal outputted by the generator. The amplifier used in this work can only withstand an input signal of $+13$ dBm without damage, corresponding to 1 V. This limits the output signal of the generator to $2\sqrt{2}$ Vp-p. To be below the limit, the amplitude of the wave is set to 1 Vp-p. The amplifier has a gain of 50 dB, which amplifies the 1 Vp-p signal to around 300 Vp-p. This is higher than recommended for the transducer, but as the waves are sent in bursts, it is not excited continuously with this voltage. The oscilloscope is not made to withstand these high voltages, and therefore a delimiter was fabricated to limit the voltage that is sent to the oscilloscope. The delimiters is placed between the amplifier, the transducer and the oscilloscope. The delimiter splits the input signal from the transducer and the received signal and decreases the voltage sent to the oscilloscope while sending the amplified signal to the transducer.

The SH0 waves propagate along the plate until the location where the plate is in contact with the liquid. By inserting the plate into the liquid, as shown in figure 1(b), the shear waves are attenuated due to the viscous shear the liquid exerts on the plate. The signal is then entirely reflected at the end of the plate, travels back to the transducer and is recorded.

The measurement of the attenuation α is based on the intensity of the waves at two immersion depths of the plate in the liquid as follows:

$$\alpha = -\frac{1}{2(d_2 - d_1)} \ln \frac{S_2(\omega)}{S_1(\omega)}, \tag{3}$$

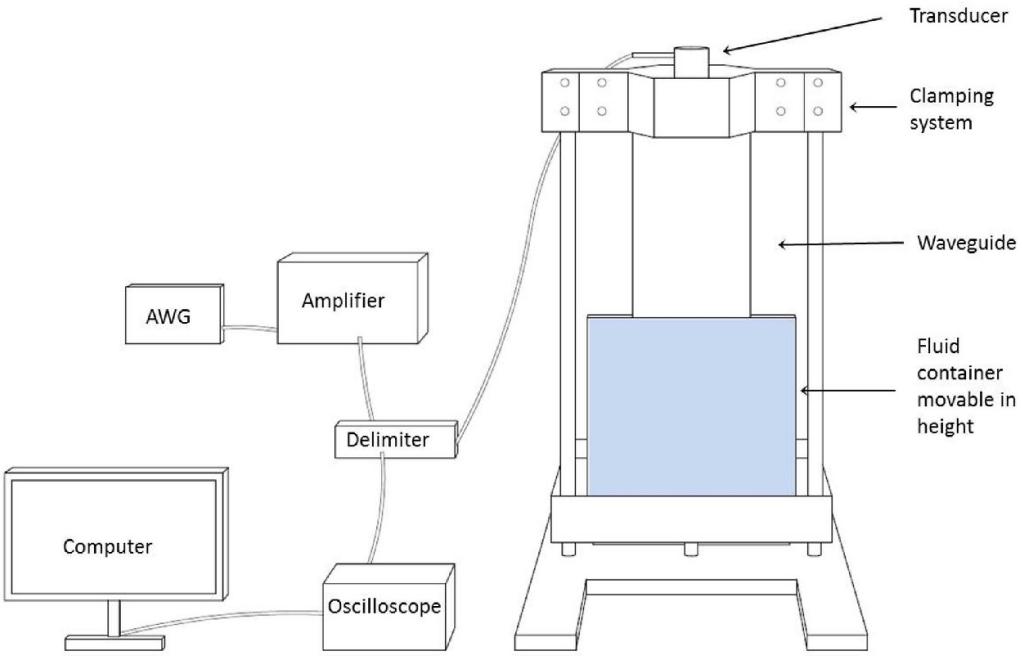

Figure 2. Schematic of the setup including the electronics to record the waves.

where $d_1 < d_2$ are two different immersion depths, and $S_2(\omega)$ and $S_1(\omega)$ are the received signal amplitudes at the respective immersion depths and at frequency ω.

The immersion depth is altered by moving the liquid container, placed on an elevator that can be steered automatically with an electromotor. A ruler can be used to gauge the insertion depth, although it can also be set at a regular insertion depth by the electromotor. In principle it is sufficient to perform a measurement at only two different depths to evaluate the attenuation and consequently from equation (1), calculate the viscosity. However, to increase the accuracy of the measurements, the plate was immersed in the liquid at several immersion depths. Each measurement was performed by increasing the immersion depth of the stainless steel plate by steps of 0.40 cm. The height of the container (82 mm) allowed to obtain the signal from a minimum of ten immersion depths. The maximum volume of the container is 45 ml, this is a small volume for a density measurement and it is ideal for measuring liquids in a harsh environment.

It must be noted that hysteresis in the level of the liquid was observed, where the liquid could differ in height by approximately 0.5 mm between the cases where the plate was inserted into the liquid and the cases where the plate was extracted from the liquid. This phenomenon, caused by the creation of a thin film of liquid during extraction of the plate, was mainly observed for viscous liquids, in the order of 50 mPa s, while it was not observed for water. Therefore, it was chosen to perform measurements by inserting the plate in the liquid only.

The liquid temperature was recorded during the experiments using a thermocouple that was immersed in the liquid.

3.1. Materials

A thin metallic plate of stainless steel was used as the waveguide. The properties of the plate, such as shear modulus and density, have to be known in order to accurately evaluate the liquid viscosity in equation (1). The density of the plate was calculated by the ratio of mass and volume, thereby measuring the weight with a common balance. Three measuring instruments were used to measure the dimensions of the plate. The thickness of the plate was measured with the 3732XFL-1 electronic micrometer by Starret. The micrometer was NIST calibrated and had accuracy of 0.002 mm. The thickness of the plate was measured 0.202 ± 0.002 mm. The width of the plate was measured with the 799A-6/150 electronic caliper by Starret, with accuracy of 0.02 mm. The caliper was also NIST calibrated. The width of the plate was measured to be 80 ± 0.02 mm. The length of the plate was measured with a measuring tape and it was 300 mm with an uncertainty of 0.1 mm, as reported by the manufacturer of the measuring tape. The density of the plate was then found to be 7874.3 ± 78.1 kg m^{-13}. The shear modulus, together with the density of the waveguide, defines the shear wave speed in the plate, i.e. $G = v_s \rho_s$. Therefore, the shear modulus can be obtained by knowing the density and the velocity of the shear wave in the metallic plate. The shear modulus, 74.9 ± 0.7 GPa, is in agreement with the range of values (dependent by the composition of the stainless steel) in literature [19]. The velocity of the shear wave in the metallic plate, v_s, was measured from the time of arrival of the first shear signal. Between each reflection, the wave travelled from the top of the plate to the bottom and then back to the transducer, which corresponds to two times the length of the

plate. This means that the shear wave velocity can be calculated using $v_s = \frac{2l}{\Delta t}$. The velocity was found to be 3083.33 ± 2.78 m s^{-1}, in agreement with the range of literature values for the different grades of stainless steel [20].

Five different Newtonian liquids were analysed. The liquids have a dynamic viscosity in the range between 0.8 and 60 mPa s at room temperature. Water, ethanol, both with a low viscosity, and three mixtures of water and glycerol with increasing volume fraction of glycerol, were measured. For the mixtures, the two liquids were stirred until they were homogeneously mixed to last for the duration of an experiment (ca. half an hour). The mixtures were then stirred again and the experiment was repeated in order to check for the repeatability. The ratio between water and glycerol was chosen in order to have viscosity between 10 and 60 mPa s.

The density of the liquids was calculated from the mass and volume ratio using an Erlenmeyer flask of 25 ml to measure the volume.

3.2. Data processing

The data were recorded using LabVIEW. The program recorded a predefined number of signal windows, 128 in this work, displayed on the oscilloscope. Each signal had a number of wave periods visible on the oscilloscope that depends on the used frequency. For the used frequencies in this research, each recorded signal contained 40 wave periods.

To obtain the signal strengths at each immersion depth, the 128 windows showing the reflected waves were averaged before calculating the intensity of the averaged wave. The background noise was considered to be a random signal with zero mean and a fixed variance, so when the windows were averaged, the background noise cancelled out. On top of that, the amplitude was constant over the whole signal, which means that the rms of this signal was the amplitude divided by $\sqrt{2}$.

For Newtonian liquids, the intensity of the reflected signal exponentially decays as a function of the immersion depth of the waveguide in the surrounding liquid, following:

$$S(x) = S_0 e^{-2\alpha(x-x_0)}, \tag{4}$$

which is a rearrangement of equation (3). Therefore, an exponential curve was fitted through the measured rms of the intensity reflections at each immersion depth:

$$S_{fit}(x) = C_1 e^{C_2(x-x_0)}, \tag{5}$$

where x is the immersion depth of the plate and the two coefficients C_1 and C_2 are determined in such a way that the exponent fits the data best. Equations (4) and (5) were combined to obtain the attenuation as:

$$\alpha = -C_2/2. \tag{6}$$

Table 2. Parameters used for the analysis.

Property	Value and uncertainty
Waveguide density (kg m^{-3})	7874 ± 78
Waveguide shear modulus G (Gpa)	74.9 ± 0.7
Waveguide thickness (mm)	0.202 ± 0.002
Waveguide length (mm)	103.4 ± 0.1
Waveguide width (mm)	80.20 ± 0.02
Transducer frequency (MHz)	2.6–3.6± 3 × 10^{-6} MHz
Density of the liquids[a]	0.2%

[a] The density uncertainty was calculated from the propagation of the errors in the volume uncertainty and mass uncertainty of the fluid.

3.3. Accuracy of the viscosity evaluation and of the attenuation measurements

The uncertainty of the viscosity derivation has been determined by applying:

$$\epsilon(\mu) = |\mu_0| \left(2\left(\frac{\epsilon(h)}{h}\right)^2 + 2\left(\frac{\epsilon(\alpha)}{\alpha}\right)^2 + \left(\frac{\epsilon(\rho_s)}{\rho_s}\right)^2 + \left(\frac{\epsilon(G)}{G}\right)^2 + \left(\frac{\epsilon(\rho_f)}{\rho_f}\right)^2 + \left(\frac{\epsilon(\omega)}{\omega}\right)^2 \right)^{1/2}. \tag{7}$$

In equation (7), $\epsilon(h)$ is the uncertainty in the thickness of the plate, $\epsilon(\rho_s)$ and $\epsilon(G)$ are the uncertainties in the density and shear modulus of the plate, $\epsilon(\rho_f)$ the uncertainty in the density of the liquid and $\epsilon(\omega)$ the uncertainty in the frequency. The $\epsilon(h)$ is based on the uncertainty of the caliper used for measuring the thickness of the plate, that is 0.002 mm. The uncertainty of the balance (i.e. 0.01 g) and of the volume measurement (i.e. 0.04 ml) is used to calculate $\epsilon(\rho_f)$ resulting in a relative uncertainty between 1.1% and 2.0%. The relative uncertainty of 1% for both the density and shear modulus of the plate is retrieved experimentally according to the method given in section 3.1. The uncertainty in the angular frequency, $\epsilon(\omega)$, is the uncertainty of the frequency of the waves sent by the generator multiplied by 2π. The frequency of the generator has an uncertainty of 1 ppm, which corresponds to 3 $\times 10^{-6}$ MHz for a frequency of 3 MHz. The properties of the plate and the source of errors for these measurements are reported in table 2. The uncertainty in the attenuation $\epsilon(\alpha)$ was calculated through a total least-squares approximation of the data. The confidence bounds for the exponential fit from equation (5) were calculated considering parameters that are a measure of the fit: the inverse R factor from QR decomposition of the Jacobian, the °s of freedom for the error, and the root-mean-squared error [21]. By combining these parameters, 95% confidence intervals for the fit parameters and thus the attenuation was obtained. The difference between the fitted coefficient C_2 and one of the 95% confidence bounds was used for calculating the uncertainty of the attenuation. The relative uncertainty measured in α was up to 1%. Substituting the standard deviations of the experimental data to the total error on μ, one obtains the relative uncertainty of 5% in the viscosity.

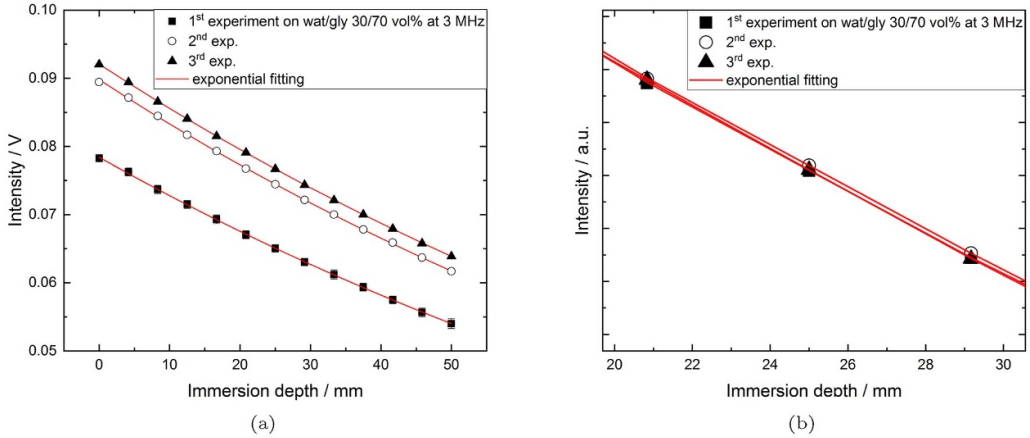

Figure 3. Amplitude of the SH0 mode for a stainless steel plate immersed in a mixture of water and glycerol 30/70 vol% at 3 MHz. The intensity was measured each 0.40 cm of immersion of the plate in the liquid. An exponential curve was fitted through the measured rms of the intensity reflections to retrieve the attenuation of the waves ($\alpha = 3.82 \pm 0.03$, ■; $\alpha = 3.79 \pm 0.03$, ◯ ;$\alpha = 3.72 \pm 0.02$, ▲). The experiment was repeated three times in order to increase the accuracy by averaging. The amplitude was normalized (b) to show the repeatability of the three measurements in retrieving the attenuation.

4. Results

As described before in section 3, the attenuation of the shear waves in liquids was retrieved from the amplitude of the measured shear waves at a range of immersion depths.

Figure 3(a) shows the measured decrease in intensity of the SH0 mode as a function of the immersion depth of the stainless steel plate in a mixture of water and glycerol 30/70 vol%. To show the repeatability of the method, the three experiments shown in figure 3(a) were performed at three different times. The intensity differs between each set because the energy transferred by the transducer to the plate may vary (e.g. by disconnecting it in between two measurement series). According to equation (3), however, only the ratio of two intensities for two or more immersion depths is of importance. The intensity was fit with an exponential curve and after normalization, figure 3(b) shows that the three curves are in agreement and the derived attenuation has a maximum of 2.6% difference between two measurements. The experiments were therefore considered to be sufficiently repeatable.

This procedure was followed for each liquid analysed. The viscosity was then obtained using equation (1). At room temperature, liquids with a viscosity ranging from 0.8 to 60 mPa s have been measured. The measured attenuation and the viscosity calculated from the averaged attenuation are reported in table 3. The results are compared to the literature values and to our own experimental results obtained with a commercial falling ball viscometer [22]. The last one was used to validate the results obtained with the waveguide viscometer. For the used liquids, there are experimental data in the literature that can be used as a reference. However, for some liquids, like the water/glycerol mixtures, few experimental data are available in the literature, and the fractions in the mixture may change over time. The advantage of using this falling ball viscometer is that a reference viscosity for the liquids can be obtained at the same time, under the same conditions, and with exactly the same composition as the viscosity measurements with the ultrasonic waveguide.

For the low viscosity of water, with measured viscosity of 0.88 ± 0.04 mPa s, the relative error to the literature value is 2%, while it is 4% to the measurement performed with a falling ball viscometer. For ethanol, with a measured viscosity of 1.19 ± 0.07, the relative error is higher: the relative error to the literature value is 12% and it is 4% to the falling ball viscometer. The ethanol 95% was purchased by Sigma Aldrich, but impurities might be present in the liquid. A slightly different composition of the purchased ethanol might change its viscosity. In fact, the measurement of the viscosity of ethanol performed with the falling ball viscometer has a lower relative error (in agreement with the error found for all the other liquids) with the measurement done with the acoustic method. This supports the hypothesis that the purchased ethanol has a different composition (due to impurities, moisture or different percentage of methanol and 2-propanol) that the one found in literature.

The viscosity of three water/glycerol mixtures, varying from 14.5 to 57.4 mPa s, has been measured and the results as a function of glycerol content are plotted in figure 4. The results show that this technique is sensible to change in viscosity of water/glycerol mixtures when the amount of water content differs. With a very small addiction of water in the mixture, the viscosity drops significantly: the viscosity of a water/glycerol mixture with only 1% water is almost 20% lower than the viscosity of pure glycerol [25, 27]. These are results for a liquid temperature of 22 ± 0.5 °C. The experimental values are compared in figure 4 with the results obtained by Cheng [25] and revised by Volk and Kähler [26], shown by the dotted line.

The viscosity measured in this work agrees within 3% to the results of Cheng. Besides the strong dependence on the water content, the viscosity of the mixtures is also very sensitive to the temperature fluctuations of the liquid as it

Table 3. Viscosity μ calculated from the attenuation measured with SH0 waves. These results are compared with the viscosity obtained from literature and measured in this work with a commercial falling ball viscometer.

Liquid	T (°C)	f (MHz)	α (np m^{-1})	μ (mPa s) This work	lit. μ (mPa s) Literature	Reference	E_R to lit. (%)	μ^a (mPa s) Falling ball	$E_R{}^a$ (%)
Water	26 ± 0.1	3.0	0.59 ± 0.01	0.88 ± 0.04	0.86	[23]	2	0.84	4
Ethanol 95%	26 ± 0.1	3.0	0.64 ± 0.02	1.19 ± 0.07	1.05	[24]	12	1.2	4
Wat/Gly 40/60 vol%	22 ± 0.1	3.0	2.58 ± 0.02	14.5 ± 1.1	14.4	[25, 26]	1.4	13.9	4
Wat/Gly 30/70 vol%	22 ± 0.1	3.0	3.78 ± 0.03	31.0 ± 0.6	31.9	[25, 26]	3	29.8	4
Wat/Gly 25/75 vol%	19 ± 0.1	3.0	5.62 ± 0.01	57.4 ± 0.2	57.9	[25, 26]	0.8	55.8	3
Measurements varying the frequency									
Wat/Gly 40/60 vol%	22 ± 0.1	[2.6; 3.6]	—	15.0 ± 1.0	14.3	[25, 26]	1.4	13.9	4
Wat/Gly 30/70 vol%	22 ± 0.2	[2.6; 3.6]	—	30.3 ± 0.6	32.0	[25, 26]	3	29.8	4
Wat/Gly 25/75 vol%	22 ± 0.1	[2.6; 3.6]	—	48.5 ± 0.3	47.8	[25, 26]	1.5	46.5	4

[a] These values were obtained with the falling ball viscometer.

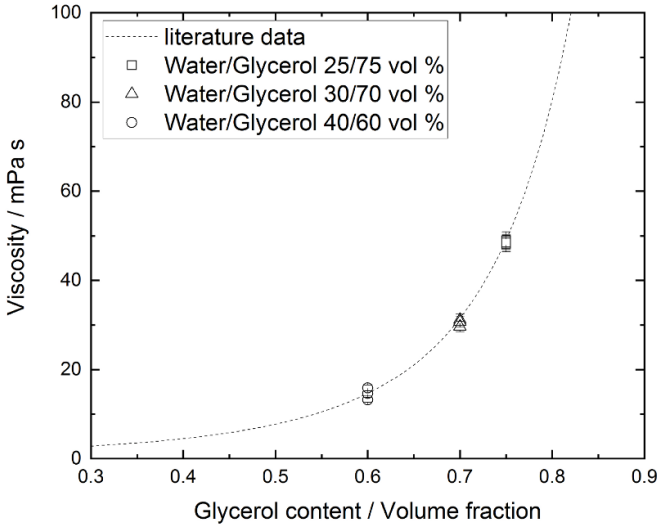

Figure 4. The viscosity of the water/glycerol mixtures, (◯ 40/60 vol%, △ 30/70 vol%, □ 25/75 vol%) as a function of volume % of glycerol at 22 ± 0.5 °C measured with the waveguide and calculated with an empirical formula (dashed line) by Cheng [25, 26].

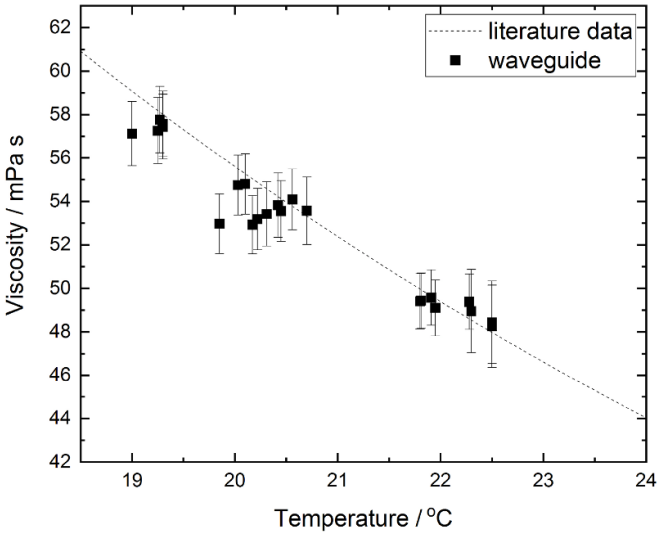

Figure 5. The viscosity of the 25/75 volume % water/glycerol mixture measured with the ultrasonic viscometer ■ as a function of the liquid temperature, compared to the literature data by Cheng [25] (dashed line).

is shown in figure 5, where the results between 19 °C and 22 °C are shown. In this temperature range between 19 °C and 22 °C the viscosity drops by almost 7%. Therefore, it is important to measure the temperature of the liquid during measurements. The results are compared to the literature data by Cheng [25] in figure 5 showing agreement within 4%.

4.1. Influence of the frequency on the viscosity

For the three mixtures of water and glycerol, the attenuation was measured at several frequencies between 2.4 and 3.6 MHz. For Newtonian fluids, the measured viscosity should be independent of the applied transducer frequency [28]. However, these measurements were performed to evaluate if the accuracy of the result could be increased by averaging the viscosity at different frequencies.

The attenuation measured experimentally for the three mixtures at different frequencies is shown in figure 6(a), where the predicted curve (the solid line in the figure) for each liquid is calculated by using the viscosity published by Cheng [25]. The attenuation is found by taking the mean attenuation from three measurements at each frequency. According to equation (1), the attenuation follows a square root dependency on the frequency of the waves. From the viscosity in figure 6(b), it is shown that the viscosity measured at different frequencies deviated from the mean viscosity of maximum 5%, within the uncertainty of the measurements.

Table 3 summarizes all the measured attenuation and the viscosity compared to the values found in literature and with the falling ball viscometer. In figure 7(a), the comparison between the viscosity measured with the ultrasonic waveguide viscometer and the literature values shows good agreement. The relative error between the literature values and

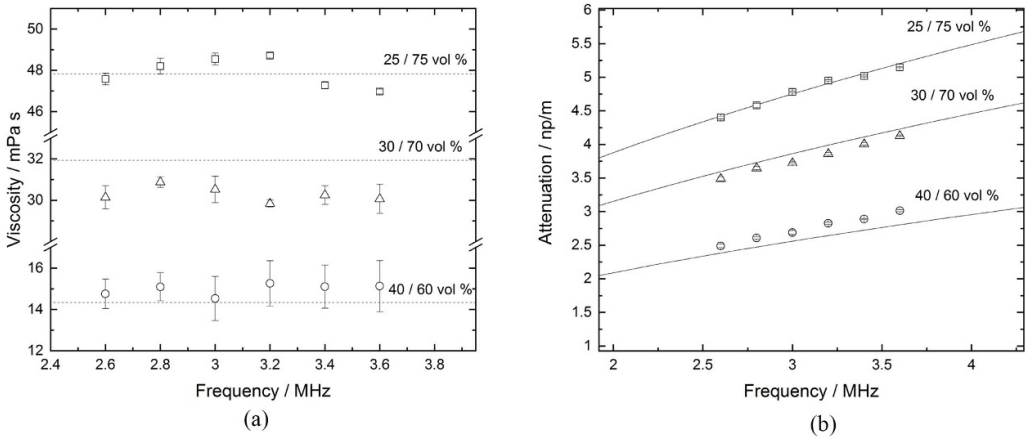

Figure 6. The measured attenuation of the mixtures water/glycerol as a function of frequency (a) is used in order to retrieve the viscosity of the liquid. The liquid viscosity for the water/glycerine mixtures from the ultrasonic measurement are compared to the reference values.

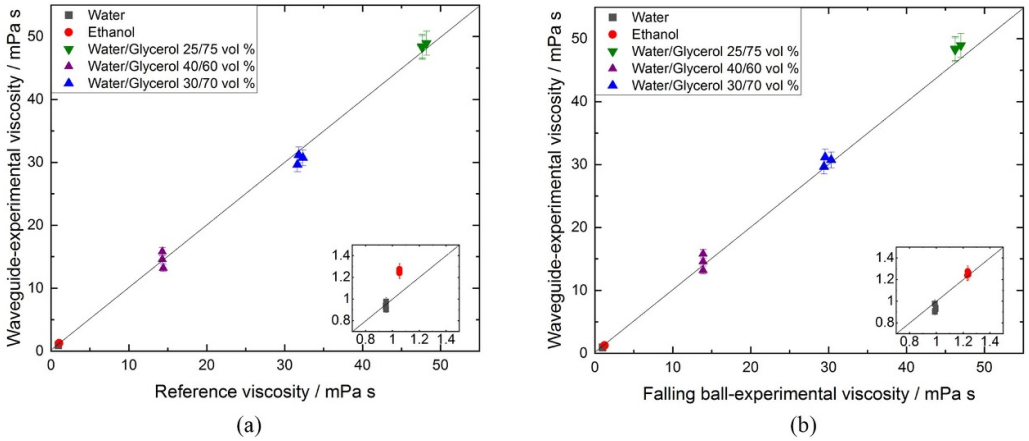

Figure 7. The viscosity from the shear guided waves attenuation for all the liquids measured is compared to the theoretical viscosity (a) and to the results from the falling ball viscometer (b) for the liquids at the same temperature. The error bars indicate the uncertainty of the method calculated as in section 3.3.

the experimental ones is less than 4% for all the liquids except ethanol. A comparison between the viscosity for all the liquids measured by the SH0-wave technique versus the viscosity obtained with the falling ball viscometer is presented in figure 7(b). The results obtained with the waveguide viscometer are slightly higher than the viscosity obtained with the falling ball viscometer and generally the viscosity measured with the ultrasonic setup corresponds better to the literature data than to the falling ball viscometer.

5. Conclusions

The technique based on ultrasonic wave propagation for deriving the viscosity of liquids is a promising technique for measuring a wide range of low viscosity liquids between 0.8 and 60 mPa s. The shear horizontal mode (SH0) was used to measure the dynamic viscosity of Newtonian fluids at room temperature.

The experimental results compare quite well to literature values with a relative error within 3% except for ethanol that showed results off by 12% compared to literature data. The results by the ultrasonic viscometer were also compared to the results by a commercial falling ball viscometer. In this last case the relative error was slightly more significant, being less than 4% for all the liquids. The ultrasonic viscometer showed the capability of measuring small variations of viscosity for liquids with low viscosity, as for the measurements of the water/glycerol varying the glycerol content and the temperature. The viscosity of the water/glycerol mixtures was measured at different frequencies ranging between 2.6 and 3.6 MHz. The results showed, as expected, that the viscosity of the mixtures is independent of the wave frequency.

Finally, this work shows that this type of viscosity measurement is suitable for liquids with low viscosity in a harsh environment, where the electronic parts should be kept apart from the liquid. This work show also that a small volume of liquid (less than 40 ml) is enough to obtain viscosity results with an error below 4%. A more accurate measurement can be performed by using another type of transducer to maximize the contact between the transducer and the plate and by increasing the intensity of the signals and the attenuation of the waves due to the viscous liquid.

Future applications of this ultrasonic viscometer will address the possibility of performing measurements at high temperatures. The possibility to measure non-Newtonian fluids will also be considered. The use of a smaller waveguide will also be studied in order to make the amount of liquid necessary for the measurements as minimal as possible.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Acknowledgments

The authors acknowledge Martin Verweij and the acoustic group of TUDelft for the private communications and for the borrowing of their instruments and laboratory. This project has received funding from the Euratom research and training programme 2014–2018 under Grant Agreement No. 661891.

ORCID iD

S Mastromarino https://orcid.org/0000-0002-6046-1605

References

[1] Mason W P, Baker W O, Mcskimin H J and Heiss J H 1949 Measurement of shear elasticity and viscosity of liquids at ultrasonic frequencies *Phys. Rev.* **75** 936–46
[2] Stevenson T, Martin D G, Cowin P I, Blumfield A, Bell A J, Comyn T P and Weaver P M 2015 Piezoelectric materials for high temperature transducers and actuators *J. Mater. Sci.: Mater. Elect.* **26** 9256–67
[3] Hunston D L 1971 Determination of the high frequency viscoelastic properties for polymers using shear mode strip delay lines *J. Poly. Sci.: Part C* **35** 201–10
[4] Knauss C J, Leppo D and Myers R R 1973 Viscoelastic measurement of polybutenes and low viscosity liquids using ultrasonic strip delay lines *J. Polym. Sci.: Polym. Symp.* **43** 179–86
[5] Shah V, Balasubramaniam K and Mayer P W G 1996 Effect of viscosity on ultrasound wave reflection from a solid/liquid interface *Ultrasonics* **34** 817–24
[6] Prasad V, Balasubramaniam K, Kannan E and Geisinger K L 2008 Viscosity measurements of melts at high temperatures using ultrasonic guided waves *J. Mater. Process. Technol.* **207** 315–20
[7] Shah V V and Balasubramaniam K 2000 Measuring Newtonian viscosity from the phase of reflected ultrasonic shear wave *Ultrasonics* **38** 921–7
[8] Vogt T K, Lowe M J S and Cawley P 2004 Measurement of the material properties of viscous liquids using ultrasonic guided waves *IEEE Trans. Ultra. Ferroelect. Frequency Control* **51** 737–47
[9] Cegla F B, Cawley P and Lowe M J S 2005 Material property measurement using the quasi-Scholte mode—a waveguide sensor *J. Acoust. Soc. Am.* **117** 1098–107
[10] Nagy P B and Kent R M 1995 Ultrasonic assessment of Poisson's ratio in thin rods *J. Acoust. Soc. Am.* **98** 2694–701
[11] Gitis M B and Chuprin V A 2012 Application of surface and normal ultrasonic waves for measuring the parameters of technical fluids: I. Shear viscosity measurements *Tech. Phys.* **57** 671–6
[12] Hunston D L, Knauss C J, Palmer M B and Myers R R 1972 Viscoelastic measurements of poly(dimethylsiloxanes) and a polystyrene solution using the layered waveguide technique *Trans. Soc. Rheol.* **16** 45–57
[13] Greenwood M S and Bamberger J A 2002 Measurement of viscosity and shear wave velocity of a liquid or slurry for on-line process control *Ultrasonics* **39** 623–30
[14] Wang L 2007 Chapter 6—one-dimensional visco-elastic waves and elastic-visco-plastic waves *Foundations of Stress Waves* ed L Wang (Oxford: Elsevier) pp 219–64
[15] Haldar S K 2018 Chapter 6—exploration geophysics *Mineral Exploration* 2nd edn, ed S K Haldar (Amsterdam: Elsevier) pp 103–22
[16] Vogt T, Lowe M J S and Cawley P 2002 Ultrasonic waveguide techniques for the measurement of material properties *AIP Conf. Proc.* **615** 1742–9
[17] Kostek S, Sinha B K and Norris A N 1993 Third-order elastic constants for an inviscid fluid *J. Acoust. Soc. Am.* **94** 3014–17
[18] Cegla F B, Cawley P and Lowe M J S 2006 Fluid bulk velocity and attenuation measurements in non-Newtonian liquids using a dipstick sensor *Meas. Sci. Technol.* **17** 264–74
[19] ASM International. Handbook Committee 1990 Properties and selection: irons, steels, and high-performance alloys (ASM International)
[20] Weast R C, Astle M J, Beyer W H and Van Allen J A 1986 *Handbook of Chemistry and Physics* 65th edn (CRC Press)
[21] Draper N R and Smith H 1998 *Applied Regression Analysis* 3rd edn (Hoboken, NJ: Wiley Interscience)
[22] Fungilab Leading Viscosity Technology, kinematic-viscometers: viscoball, Fungilab
[23] ToolBox E 2004 Engineering ToolBox water—dynamic and kinematic viscosity
[24] ToolBox E 2018 Engineering ToolBox ethanol—dynamic and kinematic viscosity
[25] Cheng N-S 2008 Formula for the viscosity of a glycerol–water mixture *Ind. Eng. Chem. Res.* **47** 3285–8
[26] Volk A and Kähler C J 2018 Density model for aqueous glycerol solutions *Exp. Fluids* **59** 75
[27] Trejo González J A, Longinotti M P and Corti H R 2011 The viscosity of glycerol–water mixtures including the supercooled region *J. Chem. Eng. Data* **56** 1397–406
[28] Schirru M, Li X, Cadeddu M and Dwyer-Joyce R 2018 Development of a shear ultrasonic spectroscopy technique for the evaluation of viscoelastic fluid properties: theory and experimental validation *Ultrasonics* **94** 364–75