Resonant pressure wave setup for simultaneous sensing of longitudinal viscosity and sound velocity of liquids

Roman Beigelbeck, Hannes Antlinger, Samir Cerimovic, Stefan Clara, Franz Keplinger and Bernhard Jakoby

1 Institute of Sensor and Actuator Systems, Vienna University of Technology, Austria
2 Institute for Microelectronics and Microsensors, Johannes Kepler University Linz, Austria

E-mail: roman.beigelbeck@tuwien.ac.at

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Abstract
Increasing demands for online monitoring of liquids have not only resulted in many new devices relying on well-established sensing parameters like shear viscosity but also initiated research on alternative parameters. Recently, the longitudinal viscosity has been evaluated as a promising candidate because the devices arising enable the bulk of the liquid to be probed rather than a thin surface layer. We report on a multi-purpose sensor which allows simultaneous measurement of the sound velocity and longitudinal viscosity of liquids. The device embodiment features a cube-shaped chamber containing the sample liquid, where one boundary surface carries a flush-mounted PZT transducer. In operation, the transducer induces standing, resonant pressure waves in the liquid under test. We studied the influences of sound velocity and longitudinal viscosity on the generated pressure waves by means of the Navier–Stokes equation for adiabatic compressible liquids and exploited both parameters as the basic sensing mechanism. Furthermore, a three-port network model describing the interaction of the transducer and sample liquid was developed in order to be applied for extracting the parameters of interest from the raw measurement data. Finally, we demonstrate the device and method by carrying out and discussing test measurements on glycerol–water solutions.

Keywords: liquid property sensing, online monitoring, pressure waves, sound velocity, longitudinal viscosity, resonant setup

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

1. Introduction
Sensing fluid properties is nowadays of utmost importance in many technical processes. In the past two decades, the devices and methods utilized as well as their demands have evolved continuously [1]. In particular, sensors for online condition monitoring of fluids have recently received great interest. In this context, several efforts have been made to evaluate thermal conductivity, permittivity, viscosity and mass density regarding their suitability as monitoring parameters [2–4]. While thermal conductivity measurements are traditionally applied for gas sensing (e.g., in gas chromatographs [5]) and permittivity monitoring is frequently used together with capacitive readout mechanisms (e.g., in fuel level indicators [6, 7]), the focus of attention has lately shifted towards viscosity detection [8]. The latter is often utilized in combination with the determination of additional parameters like mass density or sound velocity. In rheology, many different definitions of viscosity are in use [9]. The most important one is the shear viscosity which is often referred to simply as viscosity.
Modern online monitoring systems for liquids based on viscosity sensing aim at miniaturized, highly integratable, robust and reasonably priced devices. However, conventional laboratory appliances for viscosity measurements often involve motors combined with bulky, rotating cylinders hampering efficient miniaturization [9]. Moreover, most of these devices require manual liquid withdrawal which is time consuming, error prone and barely compatible with autonomous monitoring processes. Promising alternatives to such classical apparatuses are miniaturized vibrating structures immersed in the liquid under investigation. A prominent technique to determine the liquid’s shear viscosity by means of such sensors relies on thickness shear mode (TSM) resonators operating in the MHz regime. They basically embody an AT-cut quartz disc in contact with electrodes on both faces [10]. Applying an ac voltage to the electrodes excites shear vibrations of the disc through the inverse piezoelectric effect. Operation in the sample liquid imposes a trapped shear mode in the surrounding media. This interaction depends on the shear viscosity of the liquid and can be detected electrically by measuring the frequency response of the impedance between the quartz electrodes [11]. TSM resonators are characterized by high mechanical stability, but suffer from two serious drawbacks when used as viscosity sensors. First, the induced shear waves are strongly attenuated, so that only a small liquid layer of the order of microns or below is probed [12]. Consequently, such devices are prone to surface contamination and do not acquire bulk properties of liquids. The latter has a severe impact on the obtained results when complex liquids like emulsions are investigated [13]. Second, the trapped mode also generates compressional waves that are by nature scarcely damped and may cause spurious interferences when reflected by nearby obstacles [14, 15]. Other commonly used miniaturized devices for viscosity monitoring rely on vibrating beams, bridges or membranes [16, 17]. They typically operate in the frequency range between 1 kHz and a few 100 kHz, yielding a larger penetration depth of the generated shear waves. Such devices are more suitable for characterizing complex and non-Newtonian liquids than TSM resonators [13]. Notably, as the viscosity is in general a frequency-dependent quantity, results achieved by the described miniaturized sensors are not always directly comparable to those received from conventional viscometers which usually are based on the utilization of stationary or low-frequency oscillatory motion.

All previously mentioned methods depend on excitation and detection of shear waves in order to deduce the shear viscosity of the liquid. Recently, several efforts have been made for studying the longitudinal viscosity (i.e., a superposition of shear and dilatational viscosity) as an alternative monitoring parameter. Although the existence of this second coefficient of viscosity has been known for a very long time [18], it is still insufficiently investigated so that reliable material data are scarce. However, it may pave the way for a new kind of online monitoring devices which are able to sense the bulk of the liquid rather than a surface layer. First prototypes operate on the basis of acoustic spectroscopy [19, 20]. They utilize the damping behaviour of pressure waves in fluids which is well known from the theory of acoustic wave propagation in gases [21, 22]. In contrast to the large-scale measurement setups published in [19, 20], we presented in [23] a compact prototype founded on a similar principle. This device monitors the viscous attenuation of a pressure wave, generated by a microacoustic actuator in the sample liquid, with a spatially separated pressure sensor.

In this contribution, we report on a multi-purpose sensor which allows simultaneous measurement of the sound velocity and longitudinal viscosity of liquids. After introducing the device setup, the sensing mechanism based on generation and detection of standing, resonant pressure waves in the liquid under test is studied by means of the Navier–Stokes (N–S) equation. Then, a three-port network model describing the interaction of the transducer and sample liquid is introduced, which will further be applied for extracting the parameters of interest from the raw measurement data. Next, measurement results on glycerol–water solutions are presented and discussed in detail in order to finally give an outlook for future improvements.

2. Sensing device and principle

Figure 1 depicts the basic sensor configuration. A sample chamber for the liquid under test is formed by two rigid boundaries separated by a distance h. The lower boundary covers a PZT (lead zirconate titanate, chemical formula Pb(Zr\textsubscript{x}Ti\textsubscript{1-x})O\textsubscript{3}) transducer which is liquid loaded on top and backed by air at the bottom.

In operation, the vibrating transducer serves as actuator and sensor simultaneously. The sensing effect is composed of two different mechanisms. On the one hand, the liquid loading on the surface affects the mechanical resonance of the PZT disc. On the other hand, the transducer imposes a viscously attenuated pressure wave in the liquid. This wave is reflected by the opposite boundary back to the transducer and influences its vibration characteristics. Both effects depend on the liquid properties density, sound velocity and viscosity and can thus be exploited as a sensing mechanism. Through the piezoelectric effect, they are transformed into a change of the electrical transducer impedance.

3. Sensor modelling

Two different models are considered to study the influence of transducer and liquid properties on the envisaged sensing
The unit vectors in the liquid reads

\[ \text{Figure 2. Simplified analytical model to analyse the wave propagation in the liquid. Two rigid walls separated by a distance } \, d \, \text{enclose the liquid under test. The transducer featuring a lateral extension } \, d/2 \text{ and a thickness } \, l \text{ is flush mounted with the lower wall. The unit vectors in the } \, x, \, y, \text{ and } \, z \text{-directions of the indicated Cartesian coordinate system are denoted by } \, \hat{e}_x, \, \hat{e}_y, \text{ and } \, \hat{e}_z, \text{ respectively.} \]

mechanism. One is based on investigations of the linearized N–S equation for adiabatic compressible liquids demonstrating the basic sensing mechanism in the liquid. This approach covers the propagation of pressure and shear waves as well as their interaction, but neglects the feedback to the transducer. The second model relies on a three-port network configuration which is less general than the N–S model, but capable of incorporating the complete electromechanical transduction chain composed of transducer, liquid and reflecting boundary interaction. The latter model is essential for extracting the parameters of interest from the raw measurement data.

3.1. Derivation of the sensor function

This section focuses on analytical analyses of the sensor model shown in figure 2 where two parallel rigid walls, infinitely extended in the } x \text{-direction, are separated by a distance } \, d \text{ and the transducer is flush mounted with the wall at the bottom. The sample liquid is assumed to behave in a Newtonian and linear viscous manner. In this case, the stress tensor in the liquid reads

\[ \mathbf{T} = (\nabla \rho) \mathbf{I} + \mu [\nabla \otimes \mathbf{v} + (\nabla \otimes \mathbf{v})^T] + \lambda (\nabla \cdot \mathbf{v}) \mathbf{I}, \]

where \( \mathbf{v} \) is the velocity vector, \( \rho \) the pressure, \( \mathbf{I} \) the identity tensor, \( \nabla \) the del operator and \( \otimes \) the noncommutative tensor product [24]. The superscript ‘tr’ denotes the transpose operator. First and second coefficients of viscosity are indicated by \( \mu \) and \( \lambda \), respectively. \( \mu \) is related to shear stress components and therefore called shear viscosity while \( \lambda \) is associated with compressional stress components and accordingly termed dilatational viscosity. However, the definition of both coefficients is not always consistent within the literature and must always be checked to avoid misinterpretations. The notation used in this paper follows White [24].

Since the expected displacement amplitudes in the liquid are small compared to both the dimensions of the transducer and the wavelength of the excited acoustic waves [12], the nonlinear convective term in the equations of motions can be neglected [15] and Cauchy’s second law of continuum mechanics simplifies to

\[ \dot{\varrho}_0 \mathbf{v} = \nabla \cdot \mathbf{T}, \]

where \( \varrho_0 \) is the static density and the dot above a quantity denotes partial differentiation with respect to time \( t \) [24]. Introducing (1) into (2) yields the linearized N–S equation

\[ \varrho_0 \mathbf{v} = -\nabla p + \mu \nabla^2 \mathbf{v} + (\mu + \lambda) \nabla (\nabla \cdot \mathbf{v}) \]

describing the velocity field in the liquid. Here, \( \nabla^2 \) defines the vector Laplacian [25]. We assume an adiabatic compressible liquid and prescribe the relation between pressure and density by the adiabatic compressibility coefficient

\[ \varsigma = \frac{1}{\varrho_0} \left( \frac{\partial \varrho}{\partial p} \right)_s = \frac{1}{\varrho_0} \frac{\partial^2 \varrho}{\partial p^2}, \]

where the index ‘s’ refers to a process of constant entropy [26].

In case of plane shear waves \( \mathbf{u}(x, z) \), (12) and (13) reduces to

\[ \frac{\partial^2 u_z}{\partial z^2} + (1 + \alpha_i) \frac{\partial^2 u_z}{\partial x^2} + \alpha_i \frac{\partial^2 u_z}{\partial x \partial z} + \alpha_2 u_z = 0, \]

where \( k_{s,\text{visc}} \) is the complex-valued wavenumber for shear (denoted by the index ‘s’) displacements \( u_z \) determined by

\[ k_{s,\text{visc}}^2 = -\frac{j \varrho_0}{\mu} \]

which is independent of \( \lambda \).
According to (12) and (13), plane pressure waves of the form \( u(x, z) = u_z(z) \hat{i} \), are governed by

\[
\frac{\partial^2 u_z}{\partial z^2} + k_{p,\text{visc}}^2 u_z = 0. 
\tag{16}
\]

Here, \( k_{p,\text{visc}} \) is the complex-valued wavenumber for longitudinal (the index ‘p’ stands for pressure) displacements of the relevant liquid, the approximation \( \omega_c \approx \frac{\varrho c^2}{2\mu + \lambda} \)

\[
\omega_c = \frac{\varrho c^2}{2\mu + \lambda} 
\tag{18}
\]

The frequency \( f_c = \omega_c / (2\pi) \) characterizes the boundary between the regions where either 1 or \( j \) is dominant. As long as \( \omega / \omega_c \ll 1 \), which is valid in the case of operating frequencies in the MHz regime for almost any technically relevant liquid, the approximation

\[
k_{p,\text{visc}} = \frac{\omega / \varrho c}{\sqrt{1 + j\omega / \omega_c}} \approx \frac{\omega}{\varrho c} \left( 1 - \frac{j\omega}{2\omega_c} \right) 
\tag{19}
\]

holds. For example, glycerol and water feature \( f_c \)-values in the GHz and the THz regimes, respectively. From (16)–(18), it can be concluded that the displacement propagation in plane pressure waves always depends on the longitudinal viscosity \( \nu = 2\mu + \lambda \) (sometimes also called acoustic viscosity).

In the general case, shear and pressure components appear simultaneously. They are related through the mixed partial derivatives \( \partial^2 u_z / (\partial x \partial z) \) and \( \partial^2 u_z / (\partial z \partial x) \) implied in (12) and (13), respectively. As a consequence, shear waves in the \( z \)-direction with a non-uniform distribution in the \( x \)-direction (i.e., \( u_z(x, z) \)) generate associated pressure waves [15]. Unfortunately, the inverse effect, i.e., the formation of shear waves as a consequence of imposed pressure waves with non-uniform distribution in the \( x \)-direction, cannot be used as a mechanism to solely sense \( \mu \) because the influence of the shear components is always overshadowed by other effects. Not even an array of transducers entraining highly non-uniform pressure waves is able to generate a measurable effect [23]. This sensor setup relying on viscously attenuated, resonating pressure waves can therefore not be utilized to determine \( \mu \) and \( \lambda \) separately, but it essentially allows the measurement of the longitudinal viscosity \( \nu = 2\mu + \lambda \).

Supplementing (16) with the boundary conditions at the transducer (i.e., \( u_z(0) = u_{0z} \)) and the rigid wall (i.e., \( u_z(h) = 0 \)) results in a well-defined boundary value problem for plane pressure waves. Using (19), an approximate solution for the normalized displacements can be given by

\[
\frac{u_z(z)}{u_{0z}} \approx \cos \left[ \kappa_p (1 - j \kappa_p \theta) \frac{z}{h} \right] 
- \cot[\kappa_p (1 - j \kappa_p \theta)] \sin \left[ \kappa_p (1 - j \kappa_p \theta) \frac{z}{h} \right], 
\tag{20}
\]

where

\[
\kappa_p = \frac{\omega h}{\varrho c} 
\tag{21}
\]

and

\[
\theta = \frac{c_i}{2\omega h} 
\tag{22}
\]

are the normalized wavenumber of the undamped pressure wave and the normalized damping coefficient, respectively. The associated displacement magnitude follows subsequently from a straightforward auxiliary calculation to

\[
\frac{u_z(z)}{u_{0z}} \approx \sqrt{\frac{\cosh[2\kappa_p^2 \theta(1 - z/h)] - \cos[2\kappa_p(1 - z/h)]}{\cosh[2\kappa_p^2 \theta] - \cos[2\kappa_p^2 \theta]}} 
\tag{23}
\]

which describes a standing pressure wave in the liquid as illustrated exemplarily in figure 3 for \( \kappa_p = 5\pi \) and three different damping values \( \theta \). Equation (23) allows us the following conclusions. First, the resonant behaviour at a coordinate \( z \) in the liquid is approximately determined by the term \( \cosh[2\kappa_p^2 \theta] - \cos[2\kappa_p \theta] \) and therefore, because of \( \cosh[2\kappa_p^2 \theta] \approx 1 \), which is applicable for any technically relevant liquid in the MHz range, given by \( \kappa_p = n\pi \) with \( n \in \mathbb{N} \). The resonance step-ups of the pressure waves are rather sensitive to changes of the longitudinal viscosity \( \nu \).

\[\text{Figure 3. Illustration of the developed standing, resonant pressure wave in the liquid between the two walls. On the left boundary at } z = 0, \text{ a transducer imposes a pressure wave which is reflected by a rigid wall at } z = h \text{ resulting in standing, viscously damped pressure waves. Shown is the normalized displacement amplitude } |u_z(t)/u_{0z}| \text{ for } \kappa_p = 5\pi \text{ and three different damping values } \theta = c_i/(2\omega h). \text{ The box operates at resonance in the case of } \kappa_p = \omega h/c_i = n\pi \text{ with } n \in \mathbb{N}. \text{ The resonance step-ups are rather sensitive to changes of the longitudinal viscosity } \nu. \]\n
3.2. Three-port network model of the sensor device

3.2.1. Basic properties. The three-port network model shown in figure 4 featuring one electrical port and two acoustic ports is commonly used to analyse liquid-loaded PZT discs [27]. In figure 4, \( A \) denotes the area of the PZT disc, \( f \) the thickness of the disc, \( T \) the normal stress component in the \( z \)-direction, \( v_1 \) and \( v_2 \) are the velocity components at the surface (positive inward to the transducer) and \( Z_1 \) and \( Z_2 \) are the acoustic load impedances. \( U_1 \) and \( I_1 \) are voltage and current at the electrical port, respectively.
3.2. Transducer equations. After Kino [27], the governing equations for time-harmonic excitation of the transducer can be written in matrix form as

\[
\begin{bmatrix}
F_1 \\
F_2 \\
U_3
\end{bmatrix} = -j \begin{bmatrix}
Z_C A \cot(\beta_{\text{PZT}} l) & Z_C A \csc(\beta_{\text{PZT}} l) & e \\
Z_C A \csc(\beta_{\text{PZT}} l) & Z_C A \cot(\beta_{\text{PZT}} l) & e \\
e & e & 0
\end{bmatrix} \begin{bmatrix}
v_1 \\
v_2 \\
I_3
\end{bmatrix},
\]  

where \( A \) is the electrode area on one side of the disc and \( \omega \) is the excitation angular frequency. The piezoceramic material is characterized by the piezoelectric coefficient \( e \), the density \( \rho_{\text{PZT}} \), the strain-free dielectric constant \( \varepsilon_S \) and the stiffened elastic constant \( c_D \). Furthermore, clamped (zero strain) capacitance \( C_0 \), acoustic impedance \( Z_C \) and effective wavenumber of longitudinal waves \( \beta_{\text{PZT}} \) are given by

\[
C_0 = \frac{\varepsilon_S A}{l}, \quad Z_C = \sqrt{\rho_{\text{PZT}} c_D}, \quad \beta_{\text{PZT}} = \omega \sqrt{\frac{\varepsilon_S \rho_{\text{PZT}}}{c_D}}.
\]  

(24)

Importantly, all material parameters refer to the longitudinal (‘33’) operation of the piezoceramic disc. Without the loss of generality, we postulate the acoustic loads positive in the outward direction to the transducer surfaces (indicated by \( A \) in figure 4(b)) and define the associated acoustic load impedances by

\[
Z_1 = -\frac{F_1}{Av_1} = \frac{T(-l/2)}{v(-l/2)} , \quad Z_2 = -\frac{F_2}{Av_2} = \frac{T(l/2)}{v(l/2)}.
\]  

(26)

Then, the electrical input impedance of the transducer follows from (24) to

\[
Z_3 = \frac{U_3}{I_3} = \frac{1}{j\omega C_0} \times \left[ 1 + k_T^2 \right] \left[ j(\zeta_1 + \zeta_2) \sin(\beta_{\text{PZT}} l) - 2[1 - \cos(\beta_{\text{PZT}} l)] \right],
\]  

where \( k_T \) is the electromechanical coupling factor of the piezoceramic material. The normalized acoustic load impedances \( \zeta_1 = Z_1/Z_C \) and \( \zeta_2 = Z_2/Z_C \) were introduced in order to identify numerically dominant terms in (27) more easily.

3.2.3. Acoustic load impedance of the air backing. Port 1 in figure 4 is terminated by the acoustic impedance

\[
Z_1 = \rho_{\text{air}} c_{\text{air}}
\]  

(28)

which accounts for the air backing of the PZT transducer (see figure 1). From [22], we obtain at 20 °C for mass density and sound velocity in air values of \( \rho_{\text{air}} \approx 1.204 \text{ kg m}^{-3} \) and \( c_{\text{air}} \approx 343.46 \text{ m s}^{-1} \), respectively, so that (28) yields \( Z_1 \approx 413.6 \text{ Pa m}^{-1} \).

3.2.4. Acoustic load impedance of the liquid and the reflecting wall. This section deals with one-dimensional modelling of the acoustic wave propagation in the liquid and the reflection at the opposite wall by means of an equivalent electric transmission line. In general, viscous losses and diffraction effects in acoustic wave propagation interrelate in a nontrivial way [28, 29]. However, experience has shown that they can be approximately considered independently as presented in the following [30]. Referring to the coordinate system in figure 2, we consider two plane pressure waves travelling in opposite directions along the z-axis. Inferred from (1) and (16)–(19), they impose a stress distribution of the form

\[
T_{zz}(z) = l_z \cdot T \cdot l_z = T_{10} \exp[-jk_{\text{PZT}}]D(z)
\]  

(29)

in the liquid. The damping coefficients \( D(z) \) and \( D(-z + h) \) may describe the diffraction losses for the transmitted and reflected waves, respectively and will be discussed in detail below. Both integration constants \( T_{10} \) can be determined from two linearly independent boundary conditions, e.g., at \( z = 0 \) and \( z = h \). Inserting (29) into (2) gives

\[
v_z(z) = \frac{1}{j\omega \rho_{\text{li}}} \frac{\partial T_{zz}}{\partial z} = \frac{1}{j\omega \rho_{\text{li}}} [T_{10} \exp[-jk_{\text{PZT}}]D(z) + D'(z)] + T_{20} \exp[jk_{\text{PZT}}(z - h)]D(-z + h) + D'(-z + h)],
\]  

(30)

where the prime denotes an ordinary derivative with respect to \( z \). The boundary condition at the reflecting wall \( v_z(h) = 0 \) demands

\[
T_{20} = T_{10} \exp[-jk_{\text{PZT}}]D(h)/D(0),
\]  

(31)

\[
\frac{D(h)}{D(0)} = 1 + jD'(0)/[k_{\text{PZT}}D(0)].
\]  

(32)
The acoustic interaction at the reflecting wall is taken into account by the symmetries of the acoustic impedance at port 2 by taking into account the input impedance of the transmission line acting as load at port 2. Arrangement to study the diffraction effect of the transmitted pressure beam. A baffled circular piston radiates time harmonically into a half space. Beam components enclosed in the cylinder labelled 'effective range' can be detected by a virtual, reaction-free receiver (diameter d, coaxially placed at z) which averages the pressure over its surface.

Evaluating the expression

\[ Z_2 = -\frac{T_z(0)}{v_z(0)} \]  

for the acoustic impedance at port 2 by taking into account the symmetries of \( D(z) \) and \( D(-z+h) \) at \( z = h/2 \) and equating the result to the input impedance of the transmission line model shown in figure 5(a)

\[ Z_2 = -jZ_0 \cot(k_ph) \]  

yields after an extensive auxiliary calculation the equivalent transmission line parameters of our pressure wave resonator

\[ Z_h = \frac{Z_0}{\sqrt{1 + j\omega / \omega_c}} \left[ 1 + j \frac{D'(0)}{k_p \text{visc}D(0)} \right]^{-1}, \]

\[ k_p = k_p \text{visc} + k_p \text{diff} + k_p \text{offset} \]

with

\[ k_p \text{visc} = \frac{\omega}{c_l \sqrt{1 + j\omega / \omega_c}}, \]

\[ k_p \text{diff} = \frac{j}{2h} \ln \left[ \frac{D(h) + jD'(h)/[k_p \text{visc}D(h)]}{D(0) + jD'(0)/[k_p \text{visc}D(0)]} \right]. \]

Here, \( Z_0 \) corresponds to the characteristic impedance while \( k_p \), \( k_p \text{visc} \) and \( k_p \text{diff} \) denote the wavenumbers associated with total, viscous and diffraction losses, respectively. Further loss mechanisms besides \( k_p \text{visc} \) and \( k_p \text{diff} \) may also contribute to \( k_p \) substantially. Prominent examples are mechanical dampings induced by the transducer mounting, non-ideal reflections of the pressure waves at boundaries, energy losses through mode conversion at edges (e.g., by the transducer mounting) and absorption anomalies that occur in certain liquids [31]. Such spurious effects generally elude precise modelling, but can be empirically summarized by the parameter \( k_p \text{offset} \) introduced in (35). This additive wavenumber allows compensation of a damping offset between theory and measurement. Its value must be determined by means of calibration measurements using well-defined reference liquids.

\[ D(z) = 2a \exp \left( \frac{j2za}{d} \right) \int_{0}^{\infty} \frac{J_1^2(u)}{u^2} \exp \left( -j2z\sqrt{u^2 - a^2} \right) du \]

with the abbreviation

\[ a = \frac{\pi fd}{c_l} \]

and

\[ J_n(x) = \sum_{m=0}^{\infty} \frac{(-1)^m}{m!(m + n + 1)!} \left( \frac{x}{2} \right)^{2m+n} \]

as Bessel functions of the first kind and \( n \)th order [40]. The first and second integrals in (38) are related to the far- and near-field components, respectively. To the best of our knowledge, no closed-form solution of (38) is known for arbitrary values of \( z \). Aggravatingly, the kernel of the first integral is highly oscillating and both integrands feature singularities complicating an efficient numerical evaluation. Therefore, their numerical integration must be handled wary.
the special cases reflecting wall can be modelled by a rigid boundary characterized by measurement results. Consequently, the acoustic behaviour of the comb-like resonances. Such an effect has not been observed in our rigid. Second, reflection on a non-rigid wall would shift the of all boundaries as well as the PZT disc. The thickness vibrations reflecting wall for two reasons. First, we utilized a microsystem acoustic impedance of air) to account for the finite thickness of the an additional transmission line (terminated on the right side by the Figure 6. Meas. Sci. Technol. 24 (2013) 125101 R Beigelbeck et al

4. Device prototype and design

We fabricated the prototype shown in figure 7 to facilitate first experiments. The chamber features a maximum carrying volume for the sample liquid of about 53 ml. Its bounding box is made of copper-coated FR-4 material which is a composite material composed of woven fiberglass cloth with an epoxy resin binder. The copper coating and the FR-4 material exhibit commercially available standard thicknesses of 35 μm and 1.5 mm, respectively. One boundary carries a piezoceramic disc (type PIC255, diameter \(d \approx 9 \text{ mm}\) and thickness \(l \approx 1 \text{ mm}\)) connected to an SMA plug. This disc is flush mounted with the wall such that it faces air on one side and the sample liquid on the other side. Transmitting and reflecting wall are separated by a distance \(h \approx 29.2 \text{ mm}\).

The box features theoretically an infinite number of eigenmodes which are determined by the box geometry. Notably, owing to the one-sided free surface condition at the interface between liquid and air in the y-direction, in contrast to the double rigid wall conditions in the other two directions, the possible eigenmodes in the y-direction (edge waves) differ from those in the x- and z-directions. As a design rule, the distance between the side walls must be larger than \(h\) in order to reproduce the parallel plate structure sketched in figure 1. The transducer mainly excites pressure waves in the z-direction, i.e., other resonance modes are not directly excited, but could be stimulated by multipath-propagation of viscously-attenuated pressure waves in the box. Such modes must be in the minority, but the PZT disc preferably detects box resonances in the z-direction. This argument is supported by the fact that we observed no noticeable evidence of spurious interactions with other standing wave patterns like edge waves, waves between the container side walls or waves caused by mode conversion in our measurement results (see later in figure 10, left subfigure).

5. Measurement setups, parameter extraction, experimental results and discussion

Our device was applied to study the sound velocity and longitudinal viscosity of aqueous glycerol solutions with special attention to high-viscosity solutions. We have chosen

resonances of the PZT transducer which are superimposed by comb-like resonances of the mismatched transmission line \(Z_L\). The latter repeats whenever the length \(l\) of the line is increased by a half acoustic wavelength of the distance \(h\). Consequently, the spacing of the peaks can be used to ascertain the sound velocity in the liquid. Moreover, as the quality factors of these resonances are affected by the transmission line, i.e., they depend on the attenuation of the acoustic waves in the liquid and thus on \(v = 2\mu + \lambda\), the longitudinal viscosity can be determined from the magnitude of the resonance peaks.

3.2.5. Network model of the entire sensor device. Figure 6 shows the combined network model to describe the entire transmission chain consisting of PZT disc, air backing, liquid and reflecting wall. Here, the electrical impedance \(Z_L\) follows by inserting (28) and (33) into (27). \(Z_L\) comprises the intrinsic

\begin{align*}
D(0) &= 2a \int_{0}^{\infty} \frac{J_1(u)}{u} \exp \left( \frac{juh}{uh} \right) \frac{1}{u\sqrt{a^2 - u^2}} \, du \\
&= 1 - \frac{1}{a} J_1(2a) + jH_1(2a), \quad (41)
\end{align*}

\begin{align*}
D'(0) &= -\frac{4a}{d} \int_{0}^{\infty} \frac{J_1(u)}{u} \exp \left( \frac{juh}{uh} \right) \frac{1}{u\sqrt{a^2 - u^2}} \, du \\
&= \frac{2[H_1(2a) - J_1(2a)]}{d}, \quad (42)
\end{align*}

where

\begin{equation}
H_1(x) = \frac{2}{\pi} \left[ 1 - J_0(x) \right] + \frac{4}{\pi} \sum_{m=1}^{\infty} \frac{J_{2m}(x)}{4m^2 - 1} \quad (43)
\end{equation}

is the Struve function of the first order [40]. A convenient approximation of (38) is known when the source is much larger than the wavelength \(\lambda\), i.e., \(d \gg \lambda\). In this case, the correction factor (38) for large distances \(h \gtrsim 4d\) approaches the limit [30, 33]

\begin{align*}
D(h) &\approx 2 \int_{0}^{\infty} \frac{J_1(u)}{u} \exp \left( \frac{juh}{uh} \right) \frac{1}{u\sqrt{a^2 - u^2}} \, du \\
&= 1 - \exp \left( -\frac{ad}{2h} \right) \left[ J_0 \left( \frac{ad}{2h} \right) + jJ_1 \left( \frac{ad}{2h} \right) \right] \quad (44)
\end{align*}

from which we attain another useful analytical approximation

\begin{equation}
D'(h) = \frac{1}{h} \exp \left( -\frac{ad}{2h} \right) J_1 \left( \frac{ad}{2h} \right). \quad (45)
\end{equation}

\[\text{Figure 6. Network model to analyse the interaction of PZT disc, liquid and reflecting wall. In our setup, it is not necessary to insert an additional transmission line (terminated on the right side by the acoustic impedance of air) to account for the finite thickness of the reflecting wall for two reasons. First, we utilized a microsystem analyser manufactured by Polytec to visualize potential vibrations of all boundaries as well as the PZT disc. The thickness vibrations of the PZT disc were very clearly seen while all four walls remained rigid. Second, reflection on a non-rigid wall would shift the comb-like resonances. Such an effect has not been observed in our measurement results. Consequently, the acoustic behaviour of the reflecting wall can be modelled by a rigid boundary characterized by } Z_L \xrightarrow{\text{rigid}} \infty.\]
glycerol–water solutions as the sample liquid for two reasons. First, owing to their well-known dependences $\rho_i(\chi)$, $c_i(\chi)$ and $\mu(\chi)$, where $\chi$ represents the amount (in weight percent) of glycerol in distilled water ranging from zero for pure water to one for pure glycerol [41, 42]. Second, glycerol–water solutions span a shear viscosity range of more than three order of magnitude which predestines them as a reference liquid for viscosity investigations [43].

The actual temperature of the liquid affects the sound velocity and both coefficients of viscosity significantly and thus must be monitored. For example, an increase from 20 to 25 °C causes in water relative changes of the shear viscosity, longitudinal viscosity and sound velocity of $-11\%$, $-13\%$ and $-1\%$, respectively [20, 44, 45]. In order to minimize temperature-related stability problems, all our measurements were carried out under very well controllable and sufficiently stable temperature conditions in a climatic chamber manufactured by CTR (Clima Temperatur Systeme). In our measurement arrangements, the device and an additional beaker both filled with the sample liquid were placed in the chamber. In the latter, we immersed the temperature sensor of the climatic chamber to monitor and control the temperature of the sample liquid. Both liquid and chamber temperature for all results presented in this paper were constant at $(20 \pm 0.1)$ °C for which we accurately determined $\nu$.

Before a new measurement run can be started, liquid residuals must be removed by cleaning the chamber, e.g., with a rag soaked with isopropyl alcohol followed by a drying process. During the frequency domain measurements, the SMA plug of the sensor device was electrically connected to an impedance analyser (Agilent 4294) as shown in figure 8 and the frequency response of $Z_3$ was recorded. The evaluation of $c_i(\chi)$ and $\nu(\chi)$ should be carried out in the vicinity of either the series or the parallel resonance frequency of the PZT. Both should principally yield equal results. We favoured the former because the resonance peak stability around the parallel resonance exhibited occasionally small fluctuations (during the frequency sweep redraw process of $Z_3$) while the peaks near the series resonance were always rock solid. The applied overall measurement procedure comprises device calibration and liquid data acquisition with two subsequent tasks for extracting both parameters of interest from the raw measurement data. All four steps will now be explained in detail.
caused by the mounting is not covered by (46). However, both effects can be empirically incorporated (e.g., see [46, 47]) by substituting $\varepsilon_s \rightarrow \varepsilon_s/[1 + j\tan(\delta)]$ and $c_p \rightarrow c_p[1 + jQ_m]$ with appropriately adjusted values of the electrical loss factor $\tan(\delta)$ and the mechanical quality factor $Q_m$. For example, the rows ‘original’ and ‘fit’ in table 1 contain the starting values and the optimum fit obtained for our device prototype, respectively. Figure 9 contrasts measured and fitted $|Z_{30}(f)|$ characteristics.

### 5.2. Liquid measurements

In this step, the chamber is filled with the sample liquid and the frequency response $Z_3(f; \chi)$ is measured. Selected measurement results of $Z_3(f; \chi)$ near the series resonance of the PZT disc are collected in figure 10. The comb-like behaviour induced by the standing pressure wave in the liquid can be clearly seen. However, due to the superimposed intrinsic resonance of the PZT, the frequency response of the transmission line describing the liquid does not transfer directly into associated peaks in the electric impedance. Still, it can be qualitatively concluded that the glycerol content (and thus the viscosity) influences the peak-to-peak magnitudes of $|Z_3(f; \chi)|$. Now, (27) is rearranged to obtain the acoustic impedance

$$Z_3(f; \chi) = \frac{2k_2^2 - jB_{\text{PZT}}l\xi_1[1 - j2\pi fC_0Z_3(f; \chi)]}{N \cos(B_{\text{PZT}}l) + B_{\text{PZT}}l[1 - j2\pi fC_0Z_3(f; \chi)] + jk_1^2\xi_1}$$

$$= \frac{2k_2^2 - jB_{\text{PZT}}l\xi_1}{2N} \frac{j\sin(B_{\text{PZT}}l)}{N}$$

(53)

where the abbreviation

$$N = jB_{\text{PZT}}l[1 - j2\pi fC_0Z_3(f; \chi)] \cos(B_{\text{PZT}}l) - \beta_{\text{PZT}}l\xi_1[1 - j2\pi fC_0Z_3(f; \chi)] \sin(B_{\text{PZT}}l) - jk_1^2 \sin(B_{\text{PZT}}l)$$

(52)

has been used, and the measured characteristics for $Z_3(f; \chi)$ is inserted. In this way, the influence of the superimposed PZT series resonance is computationally eliminated, yielding a characteristic $|Z_3(f; \chi)|$ featuring (nearly) equal maximum magnitudes in the vicinity of $f_{\text{PZT}}$. Examples of $|Z_3(f; \chi)|$ and $|Z_3(f; \chi)|$, both obtained for water are shown in figure 11.

### 5.3. Sound velocity determination

Being completely accurate, as in viscous liquids the wavenumber $k_{\text{p,visc}}$ is generally complex valued, there always exists an interdependence between the spacing of the resonance peaks of $|Z_3(f; \chi)|$, the sound velocity and the viscosity of the liquid. This may lead to erroneous values of the phase velocity $c_0$, in very high-viscosity liquids when a spacing-based measurement method like the one described below is utilized. In such cases, an intricate dual-parameter fit must be carried out to determine $c_0$ and $\nu$ simultaneously. However, as long as $\omega \ll \omega_c$ holds, which is practically fulfilled for any moderate viscous liquid (and even for the highly viscous glycerol) in the lower MHz range, both parameters can be determined independently from each other.

From (33) it follows that, in principle, the sound velocity in the liquid can be calculated using

$$c_0 = 2\pi f\Delta f,$$

(54)

where $\Delta f$ is the difference frequency of either two adjacent parallel or alternatively two series resonance peaks of the comb-like resonances in $|Z_3(f; \chi)|$. However, this method is heavily prone to measurement uncertainties. Moreover, it may also suffer from uncertainties induced by the superimposed PZT resonance on the one hand and flat extremes (present in high-viscosity liquids) that are hard to accurately measure on the other hand. A more appropriate approach is to align the calculated and (indirectly) measured resonance characteristics of $|Z_3(f; \chi)|$ progressively in order to obtain an averaged

| $d$ | $l$ | $h$ | $\rho_{\text{PZT}}$ | $\varepsilon_3/\varepsilon_0$ | $c_p$ | $k_l$ | $Q_m$ | $\tan(\delta)$ |
|-----|-----|-----|------------------|------------------|-----|-----|-----|-----|
| Original | 9 | 1 | 29.2 | 7800 | 857 | 1.22 × 10^11 | 0.471 | 80 | 20 × 10^{-3} |
| Fit | 8.7 | 1 | 29.2 | 7800 | 857 | 1.553 × 10^11 | 0.456 | 44.7 | 20 × 10^{-3} |

Table 1. Geometry and PZT material data (obtained from [48]) are listed in the row ‘original’. The slightly adjusted values of the row ‘fit’ enable a good agreement between the calculated and measured frequency response of the transducer vibrating in air. $\varepsilon_0$, $Q_m$ and $\tan(\delta)$ denote permittivity of free space, mechanical quality factor and electrical loss factor, respectively.

Figure 9. Double-logarithmic plots of the measured and calculated (with and without damping) characteristic of $|Z_{30}(f)|$. The extremes at resonance are indicated by small circles. Measurement and theory (including damping, fit values chosen according to table 1) match perfectly in the relevant range around the (first) series resonance at $f_{\text{PZT}} \approx 2.023$ MHz. However, the theoretically predicted magnitude at the (first) parallel resonance $f_{\text{PZT}} \approx 2.234$ MHz is higher than the measured one because the utilized model does not account for the spurious mode around 2.13 MHz. This spurious mode is a radial mode and is not caused by the transducer mounting since it has also been observed in an alternative resonator design where the PZT disc was placed almost free floating (only fixed by a couple of wires).
corresponding optimum fit calculated by means of ascertainment by the theory and exhibit nearly the same height. Solutions measured in the vicinity of the PZT’s series resonance at increasing. (f small measurement uncertainties of numbering of all measured resonance peaks and singling out adjusted to fulfill (56) again. Reapplication allows consecutive natural number. Next, the frequency values must be slightly low-frequency peak is calculated and rounded to the nearest adjacent resonances are inserted into (56) and the present case. First, the measured frequency values of two PZTs are well.

value of $c_h$. According to (33), the $n$th parallel $f_{pn}$ and series $f_{sn}$ resonance frequencies are approximately governed by

$$\frac{2f_{pn} h}{c_h} = n, \quad \frac{2f_{sn} h}{c_h} = \frac{2n + 1}{2},$$

(55)

from which we infer the conditions

$$f_{p(n+1)} = \frac{n + 1}{n}, \quad f_{s(n+1)} = \frac{2n + 3}{2n + 1},$$

(56)

valid for all $n \in \mathbb{N}$. These equations specify a so-called mixed algebraic-Diophantine problem for the unknowns $n$ and $c_h$, i.e., a system of equations where at least one unknown is restricted to the class of natural numbers [49], which is easily solvable in the present case. First, the measured frequency values of two adjacent resonances are inserted into (56) and $n$ referring to the low-frequency peak is calculated and rounded to the nearest natural number. Next, the frequency values must be slightly adjusted to fulfill (56) again. Replication allows consecutive numbering of all measured resonance peaks and singling out small measurement uncertainties of $f_{pn}$ and $f_{sn}$, enabling a very close fit between model and measurement. Finally, $c_h$ is ascertained by rearranging and evaluating (55) using the revised frequency values. Comparison of measurement and fit obtained for water as sample liquid are shown in figure 11.

In the following, we briefly discuss the temperature stability of our device in terms of sound velocity measurements. This characteristic is mainly influenced by the PZT disc and the FR-4 container material, whereas the latter exhibits a significantly higher impact because the distance $h$ directly affects the measurement data evaluation (54). We obtain from (54) the relative change

$$\frac{1}{c_h} \frac{\partial c_h}{\partial T} = \frac{1}{h} \frac{\partial h}{\partial T} + \frac{1}{\Delta f} \frac{\partial \Delta f}{\partial T},$$

(57)

where the first term describes the relative length change owing to thermal expansion of the container material. Our utilized container material behaves thermally anisotropically with coefficients of linear thermal expansion (CTEs) in out-of-plane and in-plane directions of $\text{CTE}_{\text{op}} \approx 250 \text{ ppm K}^{-1}$ and $\text{CTE}_{\text{ip}} \approx 17 \text{ ppm K}^{-1}$, respectively [50]. Thermal expansion of the side walls by $\text{CTE}_{\text{ip}}$ and the container material thickness $w$ by $\text{CTE}_{\text{op}}$ affects the distance $h$. The second term in (57) can be associated with the peak distance shift imposed by the temperature-related change of the sound velocity. Aqueous glycerol features a much lower thermal diffusivity than FR-4 material and is therefore more resistant to thermal fluctuations [50, 51]. Let us assume small temperature variations such as $\Delta T = 1$°C.

Figure 10. Magnitude $|Z_r(f; \chi)|$ and phase $\arg[Z_r(f; \chi)]$ response of the transducer impedance $Z_r(f; \chi)$ for different glycerol–water solutions measured in the vicinity of the PZT’s series resonance at $f_{PZT} \approx 2.023$ MHz. The characteristic in air is also plotted as reference.

Figure 11. Double-logarithmic plots for water as sample liquid around the PZT’s series resonance. Circles mark measured values while squares and triangles label extremes according to theory (neglecting diffraction) and fit (with diffraction), respectively. Theory and fit were obtained using data from [20, 41, 42] and table 1. Both characteristics will be discussed later in section 5.5. (a) $|Z_r(f; 0)|$ with the corresponding optimum fit calculated by means of $Z_r(f; 0)$. The $n$th parallel $f_{pn}$ and series resonances $f_{sn}$ in the liquid are numbered increasingly. (b) $|Z_r(f; 0)|$ deduced from $Z_r(f; 0)$ using (52). It can be seen that the measured maxima in the vicinity of $f_{PZT}$, are well ascertained by the theory and exhibit nearly the same height.
\[ \frac{\partial \Delta f}{\partial T} = 0 \] while the container material is already influenced. This special case facilitates a very rough estimate for the ultimate temperature sensitivity

\[ \left. \frac{1}{c_{li}} \right|_h \frac{\partial c_{li}}{\partial T} \approx \frac{3}{h} \frac{\partial h}{\partial T} \approx \text{CTE}_{\text{op}} - \left( \text{CTE}_{\text{op}} - \text{CTE}_{\text{op}} \right) \frac{w}{h}. \] (58)

This value is not very realistic for practical applications as other non-thermally related effects may significantly overshadow it. Nevertheless, it determines the lower bound under ideal circumstances. Similar considerations yield the more practically relevant relationship \( \Delta c_{li}/c_{li} = \Delta h/h \) describing how a relative uncertainty in \( h \) impacts the determined sound velocity value. For instance, the sound velocity of water amounts \( 1482.358 \, \text{m s}^{-1} \) at \( 20 \, ^\circ\text{C} \) [45]. In this case, a measurement uncertainty of \( h = (29.2 \pm 0.1) \, \text{mm} \) may cause an absolute measurement error of \( \Delta c_{li} \approx \pm 5 \, \text{m s}^{-1} \).

### 5.4. Longitudinal viscosity determination

In order to achieve an accurate fit of the calculated and measured resonance magnitudes of \( |Z(f; \chi)| \), a proper alignment of both characteristics by the method described in the preceding section is mandatory. Afterwards, the magnitude of \( |Z(f; \chi)| \) can be straightforwardly fitted by varying the damping through the parameter \( \nu \).

### 5.5. Results and discussion

As a reference, we also determined the sound velocity by means of well-established time-of-flight (TOF) measurements (see figure 12) and compared the results to those of the impedance measurements. Here, the transducer was driven with a sinusoidal burst signal supplied by a function generator, where the fundamental frequency of the sine wave was set close to the PZT’s parallel resonance to achieve a reasonable signal-to-noise ratio in the received signal. Then, the sound velocity was calculated from \( c_{li} = 2h/\Delta t \) where the time delay \( \Delta t \) between input and reflected burst signal was measured using a digital storage oscilloscope. Evidently, the input burst signal has to be shorter than \( \Delta t \). Furthermore, sufficiently short cables must be used to minimize spurious time delays induced by the cabling.

Figure 13 comprises characteristics for the sound velocity versus the glycerol content obtained by our impedance and TOF measurements as well as experimental data published by Prugne et al and Elvira-Segura [41, 52]. The latter two were carried out in the low-frequency range below 50 kHz at liquid temperatures of \( 25 \, ^\circ\text{C} \) and \( 23.5 \, ^\circ\text{C} \), respectively, while our results were ascertained in either the ultrasonic range or the time domain, both for \( 20 \, ^\circ\text{C} \). These different measuring conditions are the main reason why the four characteristics deviate within a few per cent.

In the presented sensor design, viscous and diffraction losses in the sample liquid compete as useful and spurious signals, respectively. For example, figure 11(a) illustrates the significance of diffraction effects in water measurements. Neglecting them causes measured (signified by circles) and theoretically predicted values (solid line labelled by squares) to differ considerably. In this case, a parameter fit would yield far too large longitudinal viscosities, whereas with diffraction, a nearly perfect fit between theory (solid line marked with triangles) and measurement can be achieved based on physically reasonable data. In order to compare viscous and diffraction losses in aqueous glycerol solutions to each other, damping coefficients at \( f_{\text{res}} \), induced by viscous

\[
\alpha_{\text{p, visc}} = -\text{Im}\{k_{p, \text{visc}}\} \approx \frac{2\pi^2 f_{\text{PZT}}^2}{\rho h c_{li}^3} \] (59)

and diffraction

\[
\alpha_{\text{p, diff}} = -\text{Im}\{k_{p, \text{diff}}\} = -\text{Im}\left\{ \frac{1}{2h} \ln \left[ \frac{D(h) 1 + jD'(h)/|k_{p, \text{visc}}|D(h)}{D(0) 1 + jD'(0)/|k_{p, \text{visc}}|D(0)} \right] \right\} \] (60)

losses were defined from (36) and (37), respectively. Unfortunately, (59) cannot be used to appraise \( \alpha_{\text{p, visc}} \) for a comprehensive range of different glycerol concentrations \( \chi \) because reliable reference data for \( \nu(\chi) \) or \( \lambda(\chi) \) are scarce. However, a rough estimate of the lower bound of \( \nu(\chi) = 2\mu(\chi) + \lambda(\chi) \) can be made by setting for now \( \lambda(\chi) \equiv 0 \)
and utilizing $\eta_0(\chi), c_0(\chi)$ and $\mu(\chi)$ taken from [41, 42]. This enables a qualitative comparison of both damping coefficients. A representative example evaluated for our device is shown in Figure 14. Here, the threshold concentration $\chi_{th}$ at 0.70, where both damping effects differ approximately by a factor 5, separates the sample liquids in two categories. Below, i.e., from low to moderate viscous liquids, the overall damping coefficient $\alpha_p(\chi) = \alpha_{p, visc}(\chi) + \alpha_{p, diff}(\chi)$ is dominated by (60) and the sensor operation based on viscous damping is substantially overshadowed by spurious diffraction effects. This characteristic enables an empirical quantification of the diffraction losses for glycerol–water solutions by means of calibration measurements and will be briefly discussed later. We note that for water, a better quantification than setting $\lambda(0) \equiv 0$ exists as $v(0)$ narrows down to an accurately known range. For example, Pinkerton as well as Litovitz and Davis reported for the volume viscosity $\mu_v(0)$, also called bulk viscosity, of distilled water at 15 °C within the frequency range 7.5–67.5 MHz an average value of 3.09 mPa s [54, 55]. In contrast, Holmes et al published for Millipore water at 20 °C and 15 MHz an amount of (2.93 ± 0.011) mPa s [20]. Although both volume viscosities are approximately three times larger than $\mu(0)$, they yield longitudinal viscosities determined by [24, 55]

$$v(\chi) = \frac{4\mu(\chi)}{3} + \mu_v(\chi)$$

(61)

which, when evaluated for $\chi = 0$, are still too low to compete with diffraction-induced damping. In summary, the current design lacks sensitivity for concentrations $0 \leq \chi \lesssim \chi_{th}$ and is therefore moderately applicable for longitudinal viscosity measurements in this range. Above $\chi_{th}$, the viscous part becomes more and more apparent so that the device seems well suited for such high-viscosity liquids. Figure 15 depicts two nearly congruent examples of measured and fitted characteristics in this range where viscous damping is the dominant loss mechanism.

Figure 16 displays calculated shear and measured longitudinal viscosities versus the glycerol content while Table 2 contrasts absolute values obtained by theory and measurement. Measured values below the concentration $\chi_{th}$ may exhibit a wide uncertainty range as previously explained. Unfortunately, a well-founded validation of the measured values over $0 \leq \chi \leq 1$ must be stuck for an answer owing to lack of published data so that our results currently represent the state-of-the-art for aqueous glycerol solutions and can be therefore utilized as a basis in future investigations. However, it is well-known that in high-viscosity liquids, where viscosity is the predominating loss factor, classical (Stokes–Kirchhoff) absorption can be observed [55, 56]. This applies also to monatomic liquids like mercury [56, 57]. In all these cases $\mu_v/\mu$ is around 1. Therefore, the viscous properties of such liquids, which are sufficiently documented [58, 59], should also apply to high-concentration glycerol–water solutions, i.e., $\mu_v/\mu$ (glycerol) $\approx 0.8–1.2$. Comparison with our results reveals good agreement, yielding the conclusion that the presented device is well suited for such liquids.

Finally some remarks about a possible empirical estimation of diffraction losses for our specific device design. The basic idea follows from two properties reflected by Figure 14. First, at very low glycerol concentrations $\chi \ll \chi_{th}$, which is equivalent to low viscosities, diffraction is the dominant loss mechanism. Second, over the whole concentration range $0 \leq \chi \leq 1$, diffraction losses are approximately constant compared to the large-scale variation of the viscous losses. Consequently, the diffraction losses can be estimated from damping measurements on a low-viscous sample liquid (e.g., water) and approximated by a constant $k_{p, diff} \approx -\alpha_{p, diff}$. This approximation does not require evaluation of (38)–(45) and is therefore a purely empirical approach with the drawback that it is restricted to the given geometry. Importantly, the fulfillment of the dominant and nearly constant diffraction losses must be checked again if the design geometry is altered.
Figure 16. Measured longitudinal viscosity of aqueous glycerol solutions depending on the glycerol content. Depicted are the absolute $\nu(\chi)$ and the normalized $\nu(\chi)/\mu(\chi)$ characteristic received at a climatic chamber-controlled ambient temperature of 20 °C. Reference values $\mu(\chi)$ were calculated using [41].

Table 2. Comparison of theory and measurement. Two vertical lines separate (from left to right) chosen concentrations in weight percent of glycerol, values taken from the literature [41, 42] and measured mean values, respectively. Importantly, the latter are valid for 20 °C while [41] refers to 25 °C. Values marked by the superscripts '#' and '*' were obtained from [20] at 20 °C and [55] at −14 °C, respectively.

| $\chi$ | $\rho_0$ [41] (kg m$^{-3}$) | $c_0$ [41] (m s$^{-1}$) | $\mu$ [42] (mPa s) | $\nu$ (mPa s) | $\nu/\mu$ | $c_0$ [42] (m s$^{-1}$) | $\nu$ | $\nu/\mu$ |
|-------|-----------------|------------------|------------------|------------|----------|------------------|------|----------|
| 0.00  | 998             | 1500             | 1.005            | 4.284      | 4.26$^a$ | 1480             | 4.351 | 4.33     |
| 0.50  | 1131            | 1775             | 6.002            | –          | –        | 1662             | 22.989 | 3.83     |
| 0.70  | 1184            | 1847             | 23.094           | –          | –        | 1714             | 78.056 | 3.38     |
| 0.80  | 1211            | 1868             | 60.859           | –          | –        | 1802             | 182.577 | 3.00     |
| 0.85  | 1224            | 1874             | 110.829          | –          | –        | 1797             | 315.863 | 2.85     |
| 0.90  | 1237            | 1879             | 223.746          | –          | –        | 1810             | 579.502 | 2.59     |
| 0.95  | 1251            | 1880             | 515.624          | –          | –        | 1848             | 1185.930 | 2.30     |
| 1.00  | 1264            | 1881             | 1413.830         | 3336.640   | 2.36$^c$ | 1854             | 2983.180 | 2.11     |

published another method to incorporate diffraction effects in ultrasonic attenuation measurements by PSpice modelling [60] where they used the distributed resistance $R'$ and conductance $G'$ in a lumped transmission line model to account for viscous and diffraction losses, respectively. Their method could be applied in a modified form to our device, but still requires a mathematical model like (38)–(45) to calculate $G'$ and thus offers no computational savings compared to our approach. However, it could be combined with the previously described empirical approach.

6. Conclusions and outlook

We reported on the entire development chain ranging from the basic sensor idea to first test measurements of an ultrasonic pressure wave resonator applicable for simultaneous determination of sound velocity and longitudinal viscosity of liquids. Mathematical modelling comprised a Navier–Stokes and a three-port network model to study the influence of transducer and liquid properties on the envisaged sensing mechanism in the spatial and the impedance domains, respectively. Although the three-port network model is able to reflect almost all major effects, the Navier–Stokes model is didactically helpful because it reveals basic mechanisms like the spatial formation of the resonance step-ups in the device and their relation to the sound velocity and the viscosity more clearly than the three-port network model. Furthermore, many interim results deduced by the Navier–Stokes model are required for the derivation of the network model parameters. Additionally, it provides the starting point for the mathematical estimation of the diffraction losses in the liquid. Measurement results obtained on aqueous glycerol–water showed that the device combined with the applied raw data evaluation in its present form is well suited for the determination of the sound velocity, but lacks accuracy when investigating the longitudinal viscosity of low-viscosity liquids. This constraint can be attributed to spurious wave propagation phenomena such as diffraction losses overshadowing the viscous damping effect to be determined. However, the results achieved for high-viscosity liquids are promising as they agree very well with the sparsely published data on high-concentration glycerol mixtures. Unfortunately, the lack of comprehensive longitudinal viscosity data as a function of the glycerol content hampers an in-depth characterization and benchmarking of the device over a wide viscosity range. On the other hand, our measured longitudinal viscosities partially bridge the gap between the known values for water and glycerol and may serve as reference for future measurements on aqueous glycerol–water solutions. Moreover, online monitoring tasks often focus on detecting relative changes rather than accurate absolute values. In such cases, this device seems very attractive due to its comparatively low manufacturing costs.

Further mathematical investigations revealed that an analytical closed-form solution of the Navier–Stokes equation (9) might be possible describing the pressure wave excitation by the transducer, the wave propagation in the liquid
as well as their interaction. This solution could be used to
tain deeper insight in the sensor functionality, enabling an
improved device design, better estimations of spurious effects
like diffusion phenomena and their computational correction,
and optimized measurement data processing.

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