PHYSICAL SENSORS FOR FLUIDS

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ABSTRACT

Physical sensors are suited for many applications where the state of a fluid has to be monitored or evaluated. In contrast to chemical sensors they do not employ chemical reactions with the fluid and thus avoid implications associated with these reactions. In this paper, we focus on our recent work on sensors for mechanical fluid properties such as viscosity.

KEYWORDS

Physical Sensors, Fluidic Sensors, Resonant Sensors.

INTRODUCTION

Modern process control and condition monitoring applications frequently require information on the state or composition of (process) fluids. Often such sensors are chemical sensors achieving the required specificity by means of chemical reactions taking place in a chemical interface which is placed on a physical sensor. Here, chemical reactions with substances in the fluid change some physical property of the interface (e.g., density or conductivity), which is the final parameter being sensed. However, chemical interfaces often do not conform to the requirements in terms of robustness, e.g., in industrial applications. As an alternative approach, physical parameters of the fluid can be sensed such that no chemical interface is required, which is sometimes referred to as "physical chemosensing" [1]. This approach works is particularly feasible, if the monitored process is well understood and thus the physical parameters are clearly correlated to the chemical information that is actually required. The quality of this relation can often be further improved by utilizing physical sensor arrays yielding additional correlations.

Examples for physical parameters are thermal and electrical conductivity, permittivity, viscosity, speed of sound, and density. In this contribution, we focus on mechanical fluid properties. For a review on other physical parameters we, e.g., refer to our recent review papers [2] and [3]. We discuss issues arising with complex fluids, suitable sensor designs in different technologies, and illustrate these aspects by means of examples.

MECHANICAL FLUID PARAMETERS

Density and speed of sound

The mass density ρ of a fluid is a parameter that, compared to viscosity, varies only slightly with temperature. Increasing the temperature generally yields decreasing densities, where water represents a well-known exception featuring an increase in density when increasing the temperature in the range from 0°C to 4 °C. Pressure also affects the density; higher pressures yield higher densities as the molecules become more densely packed.

Besides these moderate dependencies, also most liquids feature densities in a similar order of magnitude. The density of water is around 1 kg/m^3, that of ethanol and other alcohols are in the order of 0.8 kg/m^3, and the densities engine oil as well as diesel fuel are around 0.9 kg/m^3 (all at room temperature).

This small numerical range means that densities have to be measured with a comparatively high accuracy if they are to be used as key parameters in, e.g., condition monitoring. On the macroscopic scale, densities are often determined by weighing a defined volume of a sample. In case of microsensors, due to the small dimensions, gravitational effects often become negligible such that the effect of inertial forces on the sample, preferably during high frequency vibrations, is used to determine mass densities.

The speed of sound c in a fluid is closely related to the density as it can be expressed in terms of the fluid's adiabatic compressibility β_s (the inverse of the bulk modulus K) and the density ρ: $c = \sqrt{1/(\rho\,\beta_s)} = \sqrt{K/\rho}$

It can be measured by using ultrasonic pressure waves. The compressibility or bulk modulus is also related to the second coefficient of viscosity discussed below. Using complex notation, the latter can be formally represented as an imaginary part in the bulk modulus or vice versa.

Viscosity

The physical quantity viscosity represents a constitutive parameter associated with flow behavior, which, for many fluids, can exhibit surprisingly complex. To put this into context, we review the fundamental governing equations of viscous fluid flow.

The equation of motion in continuum mechanics can generally be formulated as (Cauchy's first law of motion)

$$\rho\left(\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\nabla)\mathbf{v}\right) = \nabla\cdot\mathbf{T}\cdot \qquad (1)$$

Here, the large brackets on the left hand side contain the so called material derivative of the velocity $\mathbf{v}$, which contains the partial derivative $\partial\mathbf{v}/\partial t$ as well as the so-called convective term $(\mathbf{v}\cdot\nabla)\mathbf{v}$ containing the del operator ∇. The term $\partial\mathbf{v}/\partial t$ accounts for the fact that the velocity field may be non-stationary (i.e. at a fixed point in space the velocity field varies with time). Both terms together yield the total acceleration that an individual particle experiences. On the right hand side, the divergence of the Cauchy stress tensor $\mathbf{T}$ yields the force per volume on a volume element located at a certain position in space. The equation in this notation corresponds to the so-called Eulerian description assuming fixed spatial coordinates as it is common in fluid mechanics. In this form, the equation is fairly general. In a number of cases (see also below), the convective term can be neglected yielding an equation that is in a similar way also used in the classical linearized theory of elasticity.

To relate the stress tensors to the strain tensor characterizing the current configuration in space, constitutive equations are established. These are based on models for material behavior, which most often employ significant approximations.

Regarding viscosity, the most fundamental model characterizing the behavior of fluids is that of a so-called Newtonian liquid. The associated linear model is in analogy to a Hookean solid and can be characterized by Stokes' postulates [6]

a. shear stresses are proportional to the rate of shear strains
b. the stress to rate-of-strain relation is isotropic
c. for zero strain rates the stress tensor is solely determined by the hydrostatic (or thermodynamic) pressure p, i.e. $T_{ij}=-p\,\delta_{ij}$, where δ_{ij} denotes the Kronecker delta

By implementing these requirements, one ends up with the following constitutive equations containing two material parameters μ and λ [6]

$$T_{ij}=-p\delta_{ij}+\lambda\nabla\cdot\mathbf{v}\delta_{ij}+\mu\left(\frac{\partial v_i}{\partial x_j}+\frac{\partial v_j}{\partial x_i}\right). \qquad (2)$$

Here μ is the so-called dynamic shear viscosity, which is complemented by a second coefficient of viscosity λ, which basically concerns viscous effects associated with compressive stresses. The thermodynamic pressure p is the pressure that would be present without any viscously induced pressure components. Interestingly enough, the above constitutive equations yield that the normal stresses are not equal to p as one might expect. Defining the so-called mechanical pressure as the average of the three normal stresses (which is an invariant of this tensor), i.e. $p_m=(T_{xx}+T_{yy}+T_{zz})/3$, one finds that p_m is different from p. For some time, this caused considerable discussions leading Stokes to the postulate that $\lambda=-2\mu/3$ (the so-called Stokes assumption), which enforces $p=p_m$. Nowadays it is well-known that this assumption is generally invalid. The second coefficient of viscosity is difficult to measure if the liquid is nearly incompressible such that it is not accurately known for many liquids. For ideal, incompressible liquids with $\nabla\cdot\mathbf{v}=0$ its impact vanishes at all.

In this notation, the symbols chosen for the viscosity parameters resemble those of the Lamé parameters, which, however, refer to displacements rather than velocities. Other notations are used as well, the shear viscosity μ is often denoted by the letter η and a lot of authors introduce a bulk or volume viscosity defined by $\eta_B=(2/3)\mu+\lambda$, see, e.g., [7]. In terms of η_B the Stokes assumption reads $\eta_B=0$.

Introducing the constitutive relations in (1) one obtains

$$\rho\left(\frac{\partial\mathbf{v}}{\partial t}+(\mathbf{v}\cdot\nabla)\mathbf{v}\right)=-\nabla p+(\lambda+\mu)\nabla(\nabla\cdot\mathbf{v})+\mu\nabla^2\mathbf{v}\cdot \qquad (3)$$

This equation is non-linear due to (i) the convective term and (ii) due to the slight variations $\delta\rho$ of the density $\rho=\rho_0+\delta\rho$ upon compression of the fluid. The latter is, in general, small compared to the static density ρ_0 and can be neglected, i.e. $\delta\rho<<\rho_0$. The convective term can also be neglected under certain circumstances. The sensors discussed further below utilize sinusoidal excitation, which leads to wave phenomena in the fluid. Let us assume that such a wave is present featuring displacement amplitudes A, a radian frequency ω, and a wavenumber $k=2\pi/\lambda$. First we note that for a plane shear wave, the convective term vanishes as $\mathbf{v}$ is orthogonal to the wave propagation direction, which leaves us with the case of a wave containing longitudinal components. The velocity amplitude will then be in the order of ωA. We thus find the magnitude of the convective term $(\mathbf{v}\cdot\nabla)\mathbf{v}$ to be in the order ω^2A^2k. We compare this to the order of magnitude of the partial derivative $\partial\mathbf{v}/\partial t$, which will be in the order of ω^2A. Thus we can neglect the convective term also for a longitudinal wave if we have $\omega^2A^2k<<\omega^2A$ yielding the condition $Ak<<1$, which means that the convectional term can be neglected if the longitudinal components displacement amplitude of a wave-like motion are much smaller than the wavelength of the wave. This is the case for many problems. Under this assumption, the linearized Navier Stokes equation for Newtonian liquids can be established as

$$\rho_0\frac{\partial\mathbf{v}}{\partial t}=-\nabla p+(\lambda+\mu)\nabla(\nabla\cdot\mathbf{v})+\mu\nabla^2\mathbf{v}\cdot \qquad (4)$$

The pressure p can be expressed as a static term p_0 plus a variable term δp representing the pressure variations due to compression, i.e. $p=p_0+\delta p$. Assuming small displacements, δp can be expressed in terms of the compressibility β_s yielding $p=p_0-(1/\beta_s)\nabla\cdot\mathbf{u}$, where $\mathbf{u}$ denotes the displacement and the divergence $\nabla\cdot\mathbf{u}$ represents the relative volumetric change due to compression [8]. As the velocity and the displacement are related by $\mathbf{v}=\partial\mathbf{u}/\partial t$, (4) can be rewritten as a differential equation for the displacement. In a homogeneous medium (i.e. with spatially independent material parameters) we thus obtain for time harmonic vibrations using complex notation and suppressing the common factor $\exp(j\omega t)$

$$-\omega^2\rho_0\mathbf{u}=\left(\frac{1}{\beta_s}+j\omega(\lambda+\mu)\right)\nabla(\nabla\cdot\mathbf{u})+j\omega\mu\nabla^2\mathbf{u}\cdot \qquad (5)$$

This equation resembles the equation of motion for isotropic elastic media, where the bulk modulus is represented in terms of the compressibility and an imaginary part due to the viscosity (i.e. the expression in the first pair of brackets), and the shear modulus is purely imaginary and determined by the shear viscosity.

In the following we discuss some elementary cases for flow fields, which are the basis for viscosity measurements. In the case of **simple shear** a layer of fluid is subjected to shearing stress by enclosing it by two actuated plates, see Fig. 1a. As a consequence, the flow field has only an x-component, which varies only in y-direction. Hence the convective term as well as, in the stationary case, $\partial\mathbf{v}/\partial t$ vanish such that we are facing equilibrium where the divergence of the stress tensor vanishes: $\nabla\cdot\mathbf{T}=0$. The diagonal elements of **T** are given by the negative hydrostatic pressure, and the only non-vanishing off-diagonal components are

$$T_{xy} = T_{yx} = \mu \frac{\partial v_x}{\partial y}. \quad (6)$$

Due to $\nabla \cdot \mathbf{T}=0$, these shear strass components do not vary with respect to y and are equal to the externally applied shear stress τ. Hence the ratio of τ to the resulting velocity gradient is equal to the shear viscosity μ. This simple arrangement is often used to define the shear viscosity and many instruments such as rotational viscometers are based on it.

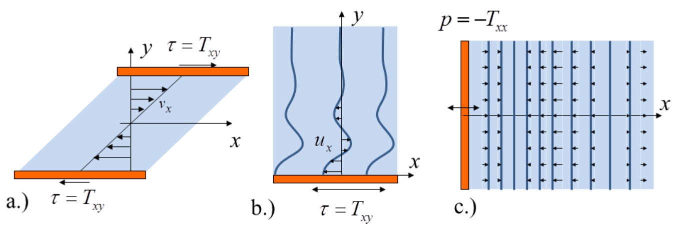


Figure 1: Fundamental excitation patterns in a liquid.

Many miniaturized sensors utilize an in-plane oscillating plate generating **shear waves** in an adjacent liquid. This is depicted in Fig. 1b. The governing equation for this situation can be obtained from (5) and is given by

$$-\omega^2 \rho_0 u_x = j\omega\mu \frac{\partial^2 u_x}{\partial y^2}. \quad (7)$$

The solution of this equation are attenuated shear waves propagating in y-direction where the characteristic penetration depth is given by $\delta = \sqrt{2\mu / \omega\rho}$. The resulting loading for the vibrating plate can be given in terms of a specific acoustic impedance

$$z_{ac,shear} = \frac{-T_{xy}}{v_x} = \frac{-T_{xy}}{j\omega u_x} = (1+j)\sqrt{\frac{\omega\eta\rho_l}{2}}. \quad (8)$$

Finally we discuss the case where a **planar pressure wave** is excited in the fluid by means of a plane featuring harmonic out-of-plane vibrations as shown in Fig. 1c. In this case the governing equation is

$$-\omega^2 \rho_0 u_x = \left(\frac{1}{\beta_s} + j\omega(\lambda + 2\mu)\right)\frac{\partial^2 u_x}{\partial x^2}. \quad (9)$$

This yields attenuated pressure waves, where the attenuation is much weaker than in case of the shear waves above. The attenuation is determined by the viscosity term $(\lambda + 2\mu)$, which is sometimes also called "acoustic viscosity". Again the loading of the plate can be specified in terms of a specific acoustic impedance which is given by

$$z_{ac,pressure} = \frac{-T_{xx}}{v_x} = \frac{-T_{xx}}{j\omega u_x} = \sqrt{\rho_0\left(\frac{1}{\beta_s} + j\omega(\lambda + 2\mu)\right)}. \quad (10)$$

For vanishing viscosities, this assumes the well-known value $\sqrt{\rho_0 / \beta_s}$.

Up to now, we only treated the linear constitutive model for viscosity. Viscosity is a material parameter which can exhibit very complicated behavior. The above model can be simply expanded towards linear viscoelasticity by introducing a (frequency dependent) imaginary part in the shear viscosity and taking a frequency dependence of all material parameters into account. As the miniaturized sensors discussed below use small vibration amplitudes, they commonly do not induce nonlinear effects but laboratory instruments frequently do. Among such effects are dependence of the constitutive parameters on the amplitude of the shear strain or stress, memory effects (such as hysteresis), and complex rheological behavior such as thixotropic (shear thinning) and rheopectic (shear thickening) behavior, where the apparent viscosity drops or increases with the duration of the shear stress, respectively. Also non-linear viscoelastic effects leading, e.g., to different normal stresses in case of shearing, can come into play. A well-known effect related to normal stresses is the Weissenberg effect where a liquid polymer solution is drawn into the direction of an immersed spinning rod rather than being thrown outward. For more details on such non-Newtonian behavior, we refer to the vast literature covering rheology, see, e.g. [9].

RESONATORS FOR FLUIDIC SENSING

Utilizing resonators as sensors offers the potential to achieve high resolutions (as small changes in frequency can be accurately measured if frequency is used as a readout parameter) and sensitivity (as resonant phenomena tend to be highly sensitive to changes in their environment). At the same time, the overall sensor performance is crucially determined by other influences such as noise and spurious cross-sensitivities, which have to be properly considered when designing resonant sensors.

In a simplified lumped element model, a mechanical resonator can be represented by a spring mass system, see also Fig. 2 [10].

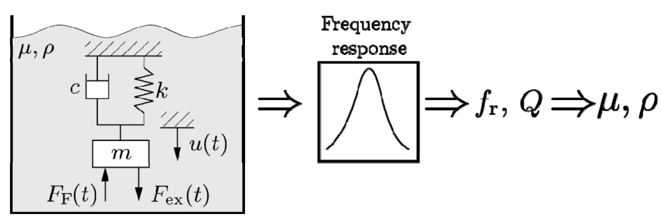


Figure 2: Lumped element representation of a resonator [10].

The lumped mass m represents the moving mass of the system, the spring constant k accounts for the elastic restoring forces, and the damping element featuring a constant c represents the dissipation in the system. The system is driven by an external force F_{ex} while the force F_F accounts for forces due to interaction with the liquid environment. The differential equation for the lumped system is given by

$$m\frac{d^2u}{dt^2} + c\frac{du}{dt} + ku + F_F = F_{ex}, \quad (11)$$

where u represents the resonator's displacement such that $v = dx/dt$ is the characteristic velocity. Assuming $F_F=0$, we obtain the following transfer function in frequency domain

$$\frac{v(\omega)}{F_{ex}(\omega)} = \frac{j\omega\, u(\omega)}{F_{ex}(\omega)} = \frac{1}{c + j\omega m + \dfrac{k}{j\omega}} = \frac{1/(\omega_0 m)}{\dfrac{1}{Q} + j\left(\dfrac{\omega}{\omega_0} - \dfrac{\omega_0}{\omega}\right)}, \quad (12)$$

where we introduced the resonance frequency ω_0 and the Q-factor Q (without liquid loading) as

$$\omega_0 = \sqrt{k/m}, \quad Q = \sqrt{km}/c \quad (13)$$

If a mechanical resonator, e.g. a tuning fork, is immersed in a liquid, an additional effective mass δm (proportional to the liquid's ρ) is moved with the vibration which increases the effectively vibrating mass m of the system by δm such that the resonance frequency becomes $\omega_0 = \sqrt{k/(m+\delta m)}$ yielding an approximate relative change

$$\frac{\delta\omega_0}{\omega_0} = \frac{1}{\omega_0}\frac{\partial\omega_0}{\partial m}\delta m = -\frac{1}{2}\frac{\delta m}{m}. \quad (14)$$

Thus, in order to maximize the sensitivity, it is beneficial to minimize the resonator mass m.

Besides added mass phenomena, immersion in a liquid leads to additional forces (F_F in the model). Neglecting non-linear effects as discussed in the previous section, these forces can be expressed in frequency domain by means of an acoustic impedance Z_{fluid} giving the relation of said forces to the velocity, i.e. $F_F = Z_{fluid}\, v$. Decomposing the impedance in its real and imaginary part $Z_{fluid} = Z_R + jZ_I$, it is found that (13) can be reformulated to include liquid loading by modifying the mass m and the damping factor c as

$$m \to m + \frac{Z_I}{\omega}, \quad c \to c + Z_R. \quad (15)$$

The resonance frequency and the Q-factor given in (13) change accordingly. Strictly, the additional terms Z_I/ω and Z_R are frequency dependent such that the frequency response does not correspond to a pure second order system any more. However, the variation in the vicinity of the resonance frequency is moderate such that as an approximation, their value at ω_0 can be used.

Depending on the geometry and the vibration mode shape of the used resonator, the interaction with the fluid may be close to the fundamental cases of a pure in-plane or out-of-plane vibration. In these cases, the impedance Z_{fluid} can be approximately proportional to the specific impedances presented above. In the general case, there will be a mixed dependence on the viscosity, density and compressibility (and thus also the speed of sound) of the liquid. In [10] a generic model for this interaction is proposed and evaluated for some device designs.

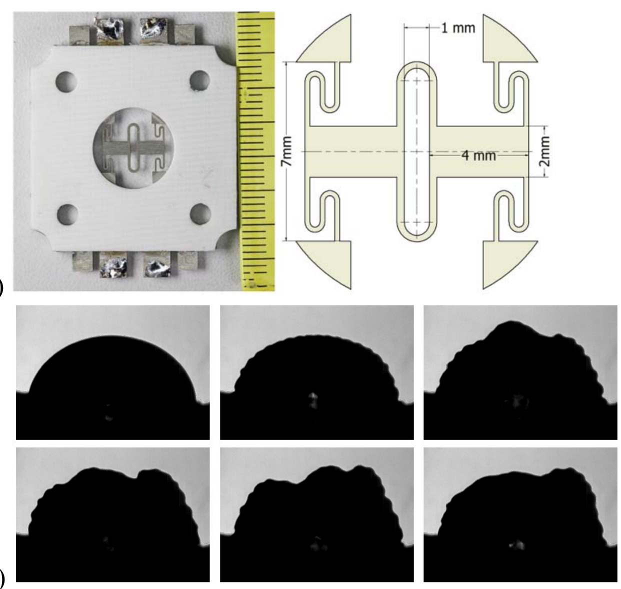
With a suitable model, the parameters sought can be determined, if the characteristic parameters ω_0 and Q are extracted from the frequency response. Doing so, it is essential to properly take spurious effects into account. In [11] and [12] an algorithm for the proper consideration of such spurious effects and the extraction of the desired parameters is presented.

EXAMPLES

In the following we briefly review some concepts for viscosity (and in some cases also mass) sensing. In view of the broad area of applications and technologies, we refrain from attempting to provide a complete picture in terms of a literature survey but concentrate on our own results. The list of references is thus by no means complete.

Many of these devices operate at lower frequencies than the earlier investigated piezoelectric devices, e.g., Quartz crystal resonators or Love wave devices, see the overview in [13] . The reason for that lies in the limited penetration depth δ of the employed shear waves, see also Fig. 1b and the associated discussion above. In many applications, the liquid shows a microstructure such as in case of emulsions or suspensions. If the characteristic dimension of said microstructure (e.g., droplet diameter in case of an emulsion) is in the order of or even larger than δ, the sensor will not capture the macroscopically apparent viscosity [4], [14]. The penetration depth can be increased by using lower frequencies or other modes of vibrations than shear waves, see the discussion in [13].

A straightforward alternative to piezoelectric shear wave devices in the MHz range are **in-plane vibrating plates**. We investigated micromachined designs in silicon technology [15] as well as similar devices based on metal plates [16],[17]. Most recently a balanced setup utilizing two plates has been presented (Fig. 3a, [18]), which reduces the impact of the plate's suspension on the resonance characteristics. These plate waves are driven by Lorentz-forces acting on AC-currents flowing in the plate and are read out using the motion induced voltage. The effect of this voltage is either seen in the impedance of the excitation path or as induced voltage in a separate readout port.



a)

b)

Figure 3: Double plate resonator (a) and vibrating droplet (b).

The in-plane vibrating plate can also be used to accelerate an entire **droplet** which allows characterizing its constitution (e.g., sedimentation of particles) or inducing mixing processes within the droplet. The flow field within the droplet is fairly complex and gives rise to interesting capillary waves allowing to study their interaction with the internal acoustically induced streaming (Fig. 3b, [19]).

Besides shear mode devices, devices with **vibrating beams or wires** have been investigated. Some of them are driven piezoelectrically (like quartz tuning forks) but again Lorentz force actuation can be used as well. A classical approach is the vibrating wire where a vibrating string (or wire) is oscillating in a liquid and the motion induced voltage is used as readout signal. The principle can be modified yielding a tunable resonance frequency by changing the string tension, which also works for suspended plate resonators [20].

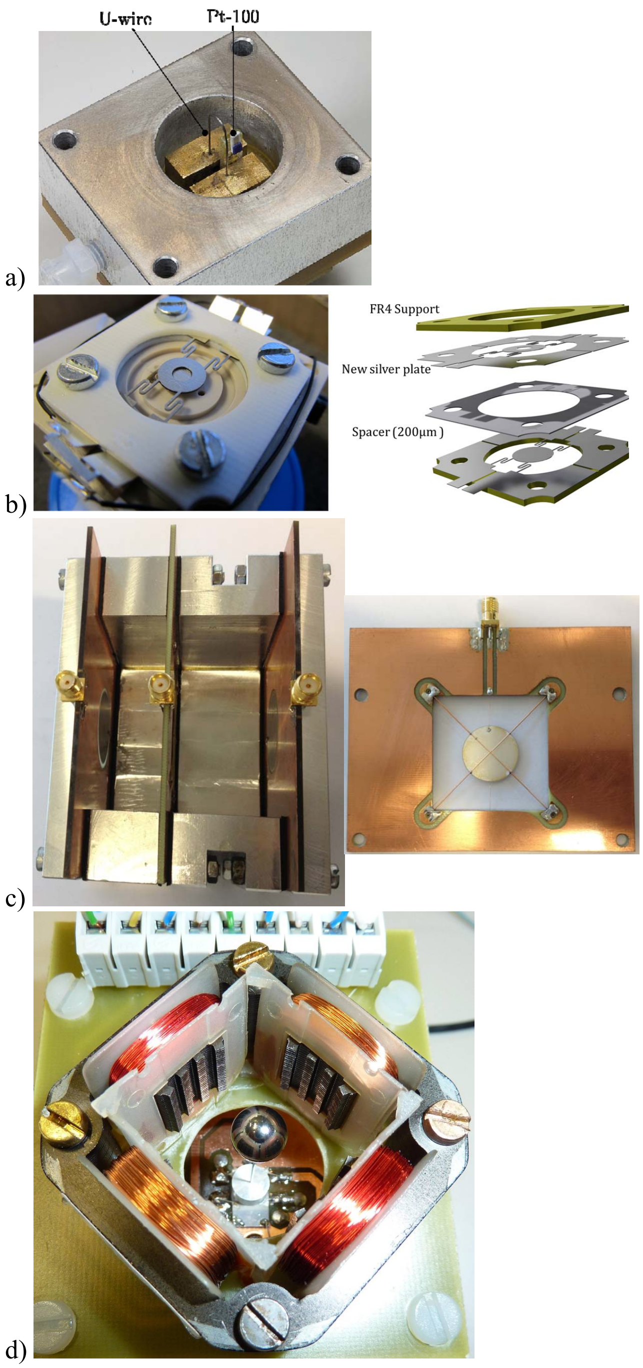


Figure 4: Selection of devices used for viscosity sensing (see text for details and references).

Similarly, beams and wires without mechanical pre-stress can be utilized. In [21] and [22] a U-shaped cantilever in silicon-micromachining technology is used for viscosity sensing. The principle can also be applied to U-shaped metal wires, which, if they are filled with a sample liquid, can also be used for density sensing as the vibrating mass is increased (Fig. 4a, [23]). Also tuning forks fall into this category, these have also been successfully used for gases [24],[25],[26].. The general interaction of a moving beam with a fluid is investigated in [27].

A principle that is particularly suited for online mass sensing in microfluidic circuits, is the so-called **vibrating membrane sensor**. Here the liquid under investigation is led through a chamber with flexible bottom and top membranes. These are actuated in certain mode shapes. Depending on the chosen vibration mode the density or viscosity of the fluid can be sensed [28],[29] by means of their impact on the resonant properties.

For **high viscosities**, shear-wave based resonators become generally highly damped. As an alternative, pressure waves aiming at the determination of the second coefficient of viscosity can be used. Recently an alternative concept, which still utilizes shear waves, has been devised. It is based on the vibrating plate setup but uses two plates which are arranged in parallel in some defined distance of each other. The liquid is placed between the plates rather than immersing one or both plates entirely in liquid. Upon actuation of one plate, the second plate becomes viscously entrained by the shear wave generated in the fluid. This entrainment becomes even more effective for highly viscous liquids. Based on a model for this signal transmission, the viscosity can be extracted from the frequency response of the transfer function (Fig. 4b, [30]).

In order to utilize pressure waves for viscosity sensing, transducers featuring out of plane vibrations have to be applied. The acoustic viscosity $2\mu+\lambda$ but also the density and compressibility are major liquid parameters affecting the properties of the excited wave. One concept is based on establishing an acoustic resonator and determining the frequency response of the excitation transducer, which is influenced by the fluid-filled acoustic resonator it is attached to [31]. Doing so, the Q-factor of the resonance characteristics will be influenced by the viscosity whereas the resonator's resonance frequency will depend on the speed of sound in the liquid (and the geometry of the resonator). Alternatively, a transmission setup can be used, where the damping of the wave is utilized to determine the acoustic viscosity [32]. However, the measured response also depends on the transducer properties, which thus have to be precisely known. When using, e.g., piezoelectric PZT transducers, one is facing significant tolerances in their parameters, which affects the accuracy of this approach. As an alternative, a three transducer setup featuring a transmitting transducer (immersed in the liquid), generating pressure waves of equal amplitude in two opposite directions and two equal receiving transducers, which are positioned in two different distances d_1 and d_2 from the transmitter, can be used. In that manner the ratio of the two receiving signals only depends on the acoustic attenuation of the waves and the difference in the acoustic propagation paths $d_1–d_2$. To avoid interference effects bursts can be used and settling effects can be eliminated by signal processing (Fig. 3c, [33]). Also, spurious effects like diffraction need to be accounted for. This can be done by numerical correction of the measured attenuation values but also by using closed cylindrical geometries, where no diffraction losses can occur [34],[35]. The general principle applied here is similar to so called ultrasound attenuation spectroscopy where attenuation of acoustic pressure waves is measured of comparatively large distance. We demonstrated the potential for miniaturization particularly for highly viscous fluids.

Finally we mention an attempt to reproduce the characteristics of a classical measurement method: the falling ball viscometer, where a metal ball is driven by gravitational forces through a liquid-filled tube. The stationary velocity depends on the viscosity and is determined from the measured time of flight for a certain distance. Particularly for complex fluids, such classical methods yield different viscosity values than the micro-resonators discussed above as the actuation of the liquid is fundamentally different (high frequencies but low amplitudes, etc.). Many users adhere to these old-fashioned methods, as they can interpret the rheometric results achieved at the measurement parameters used. To mimic the behavior of a falling ball viscometer, we devised a setup, where a ball is orbiting through the fluid and the force for a certain speed is determined to evaluate the viscosity. The ball is mounted on a spring wire and can alternatively be operated in a resonant vibrating mode (Fig. 3d, [36]).

CONCLUSION

Viscosity sensing using microsensors primarily enables sensing in the linear viscoelastic domain. The choice of method or device crucially depends on potentially apparent non-Newtonian behavior, viscosity range, and microstructure of the fluid.

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