Miniaturized Sensors and Sensing Systems for Liquid Media

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One of the major research targets in our recent research has been the sensing of physical liquid parameters by means of miniaturized sensors. Such sensors and sensor systems can be utilized in applications, where liquids in industrial processes are monitored in order to maintain the quality of a process or the associated product. Due to the adverse properties commonly associated with chemical interfaces (lacking reversibility, drift, etc.), we concentrate our research on physical parameters as indicators for the state of the liquid, in particularly density, viscosity (or more general rheological properties), and infrared absorption. The miniaturization of suitable sensor principles facilitates the implementation of these devices online. At the same time, scaling effects have to be taken into account, which, e.g., in case of viscosity sensors, lead to issues when it comes to applications in complex liquids such as suspensions.

In our paper we provide an overview on our recent work discussing the device design, the associated modeling, and the application of the devices.

Keywords: Viscosity sensing, resonating sensors

Introduction

In this contribution we report on recent research on miniaturized viscosity sensors, which can be used in industrial process control and related applications wherever process liquids and changes in the liquid's condition are of interest (e.g., in food industry and engine oil quality monitoring). The viscosity (more precisely the "shear viscosity") can be defined in terms of a simple experimental arrangement, where the liquid under test is sheared between two laterally moving plates. The viscosity η is then defined as the ratio between the applied shear stress τ (maintaining the movement) and the resulting gradient of the flow velocity v (i.e. the shear rate $\dot{\gamma}$) of the liquid sheared between the plates:

$$\eta = \frac{\tau}{\partial v_x / \partial y} = \frac{\tau}{\dot{\gamma}} \tag{1}$$

The such defined viscosity (as already proposed by Newton) can be measured in a straightforward manner by laboratory instruments, which in some way impress a shear deformation on the liquid and measure the required torque (or vice versa, i.e. impress torque and measure resulting shear deformation). Such devices typically either utilize

continuous rotational movements (avoiding extended translational movements required by the simple plate setup) or oscillatory rotational movements. Most often, the viscosity as defined by (1) crucially depends on the used process parameters, e.g., the impressed shear rate. For instance consider a simple schematic arrangement as shown in Fig. 1 for the measurement of viscosity: a pivoted cylinder is immersed in a viscous liquid. By means of some motor, the cylinder can be driven to perform a continuous rotational movement or rotational oscillation.
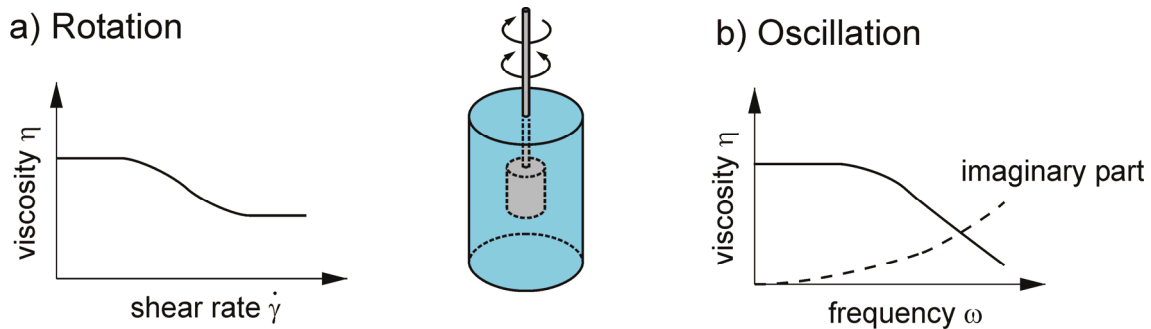
a) Rotation

b) Oscillation

Fig. 1: Basic measurement approaches for viscosity: rotational and vibrational (oscillatory) method.

For the continuous rotation, the ratio between the applied torque and the rotational speed will be related to the viscosity of the liquid. Now for a so-called "Newtonian" liquid, this ratio yielding the viscosity would not depend on the actually applied rotational speed or, in terms of the definition (1), the viscosity would not depend on the shear rate. For real liquids, such a dependence can, however, occur if, e.g., (macro)-molecules in the liquid become disentangled by the shear movement, which would be an example for so-called "non-Newtonian" behavior. This behavior is known as shear-thinning behavior (the viscosity drops with increasing shear rate) and can be reversible or (also partly) non-reversible. This behavior represents a non-linear response, which can be accounted for by describing the viscosity a function of the shear rate. For oscillatory (vibrational) measurements, the cylinder would perform, e.g., sinusoidal oscillations, where, similarly as before, the ratio of the applied torque-amplitude and the amplitude in angular speed could be used as measures for the viscosity. The above-mentioned non-linear behavior here would lead to a dependence of the obtained viscosity on the amplitudes for angular speed and torque. Moreover, another kind of non-Newtonian behavior can appear: the measured viscosity can depend on the applied frequency. More specifically, a phase shift between the torque and the angular speed signals can appear, which represents the onset of elastic behavior. This phenomenon is often referred to as viscoelastic behavior and can be modeled by adopting the common complex notation, where an imaginary part in the viscosity accounts for the elastic part. (This is in analogy to an ohmic resistor supplemented by an additional capacitive part.) In terms of system theory, this kind of non-Newtonian behavior corresponds to a linear distortion in the system response. We note that also the reverse effect can appear at higher frequencies, i.e., dominantly elastic behavior such as the bulk compressibility of a liquid, can show viscous contributions such that the compressibility coefficient shows an imaginary part, which corresponds to the so-called bulk viscosity of a liquid. In contrast to the shear viscosity, the bulk viscosity is difficult to measure and plays a minor role in the analysis of liquid behavior such that it is often neglected.

In general both, linear and non-linear effects can occur, leading to sometimes complicated non-Newtonian behavior of liquids. In rheology, this behavior is a vibrant issue in research (see, e.g., [2] for a thorough account on this).

Considered Technologies for Miniaturized Viscosity-Sensors

As discussed above, the measurement of viscosity involves some kind of interaction of moving or vibrating parts with a liquid. In the following we discuss some fundamental principles underlying our work on miniaturized viscosity sensors.

Thickness Shear Mode Resonators and Related Devices

A very elegant and well defined way to achieve interaction of vibrating bodies with a viscous sample is that of a shear-vibrating plane being in contact with a viscous liquid. Consider an infinitely extended plane orthogonal to the *z*-axis performing a sinusoidal shear vibration (oriented in *x*-direction) with a radian frequency ω and being loaded with a viscous liquid occupying the half-space *z* > 0 above the plate. For this elementary problem, a simple solution for the flow field in the viscous liquid can be obtained in terms of an attenuated shear wave (using time-harmonic complex notation) [3]

$$v_x(z) = v_x(0)\, e^{j\omega t}\, e^{-jz/\delta}\, e^{-z/\delta}\, . \tag{2}$$

Here v_x denotes the velocity component in x direction and δ is the so-called penetration depth δ given by

$$\delta = \sqrt{\frac{2\eta}{\rho\omega}}\, . \tag{3}$$

This interaction can be utilized for viscosity sensing. Prominent examples are so-called thickness shear mode resonators, which are most often embodied by piezoelectric disks. The disks (most often quartz) feature electrodes on both faces. By applying an AC voltage, mechanical vibrations can be excited by means of the piezoelectric effect. Choosing an appropriate crystal cut, shear vibrations can be excited. If the disk is immersed in a viscous liquid, the interaction described above is enabled.

The impedance appearing between the electrodes features a piezoelectrically induced part, which represents the mechanical vibration. Close to the mechanical resonance frequency, this so-called motional arm can be represented as LC-resonance circuit in an equivalent circuit (see Fig. 2, see also [4]).
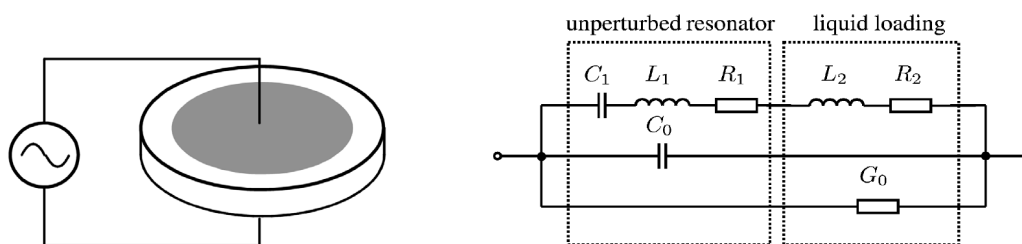

Fig. 2: TSM resonator: basic embodiment and equivalent circuit near mechanical resonance.

A straightforward analysis [4] yields that loading the resonator with a liquid leading to coupling of the shear-vibrating surfaces with the liquid as described above results in additional components L_2 and R_2 in the motional arm of the equivalent circuit. These components can be interpreted as the consequence of the additional mass loading by the entrained liquid layer resulting change in the resonance frequency and the losses

associated with the viscous dissipation in the liquid. Both parameters are approximately proportional to the viscosity-density product of the liquid

$$L_2, R_2 \propto \sqrt{\rho \eta} \tag{4}$$

There are other piezoelectric devices based on shear polarized surface bound modes such as, e.g., Love waves [5] and surface transverse waves [6], which yield similar interaction and sensitivities with respect to the viscosity-density product in terms of the wave's damping and wavenumber.

Piezoelectric excitation can be replaced by other excitation mechanisms such as excitation by Lorentz-forces. In [7] the excitation of modes in conducting (e.g., metallic) disks and membranes in terms of induced eddy currents in an external magnetic bias field is described. Here the excitation of dominantly shear-polarized modes is crucially influenced by the geometry of the excitation coil inducing the eddy currents.

Known issues in the design of shear-mode devices include spuriously excited compressional waves [8] – [10], which may lead to disturbing interferences upon reflections from nearby obstacles as, in contrast to shear waves, compressional waves are scarcely damped even in viscous liquids.

In terms of rheology, shear vibrating devices at higher frequencies show a comparatively small penetration depth δ as given by (3), e.g., for water and a frequency of 6 MHz, δ is in the order of 0.2 microns. Thus structural effects influencing the viscosity, which are related to microstructures with typical dimensions in the order of δ or above, will not be captured by the sensor. For instance, in emulsions featuring droplets in the dimensions 10 microns or above, the viscosity of the continuous phase rather than that of the entire mixture is sensed. This effect does not occur for microemulsions with droplet sizes below one micron [11]. Similar experiments have been reported with suspensions, see also the discussion for vibrating beams below [12].

Apart from the fact that kind of a "thin film viscosity" is determined, compared to oscillatory lab viscometers, the devices typically feature significantly higher vibration frequencies (some 10 kHz to some 100 MHz. At the same time the vibration amplitudes are very small (can be in the nm range). This yield shear rates in the range of $10^7 s^{-1}$ [13], where due to the sinusoidal vibration, we strictly have to speak about an averaged shear rate [14].

Non-Newtonian behavior can be detected in terms of deviations from the relation (4), which holds for Newtonian liquids only. Specifically, based on (4) a linear relation between L_2 and R_2 can be established. If a measured parameter pair yields a deviation from this linear relation this indicates non-Newtonian behavior as will be shown in an example (monitoring of zeolite synthesis) below.

Summarizing, compared to common lab instruments, shear vibrating miniaturized viscosity sensors excite the liquid under test in a significantly different rheological regime, which can be an issue for complex liquids, if the conventional "macroscopic" viscosity values have to be obtained. Still, these sensors can have benefits also for the monitoring of complex non-Newtonian liquids if a proper calibration routine or, more generally, a suitable interpretation scheme for the obtained data is worked out. The latter can be sufficient as in process control, it is often more important to detect changes in the process rather than measure absolute viscosity values. For broad application also in the laboratory, it will be essential, to standardize the method, similarly as it has been done with other novel viscosity measurement methods (e.g., the high-temperature-high-shear or HTHS method in lubrication technology (see, e.g., the standard ASTM D5481 - 04).

Vibrating Beam Devices and Membranes

Another simple vibrating structure that can be utilized to determine viscosity and density of liquids is that of a vibrating beam. Even though the structure is simple, its interaction with the liquid is more involved than that of a plane surface performing shear vibrations. However, in comparison to the thickness shear resonators discussed above, the vibrating beams described in the following feature the lower vibration frequencies and larger amplitudes such that the probed rheological regime can be expected to be more comparable to that of conventional lab viscometers, which indeed holds true in the experiment.

In our research, we considered beams implemented in silicon micromachining technology as well as beams implemented by means of thin vibrating polymer foils. The beams are excited by Lorentz forces stemming from an external magnetic field (provided by a permanent magnet) and AC-currents in conductive paths along the beam. For the readout, the induced voltage in a pickup coil (whose windings are partly on the beam and which is thus vibrating in the magnetic field) or an optical readout method can be utilized. Fig. 3 shows basic beam designs.
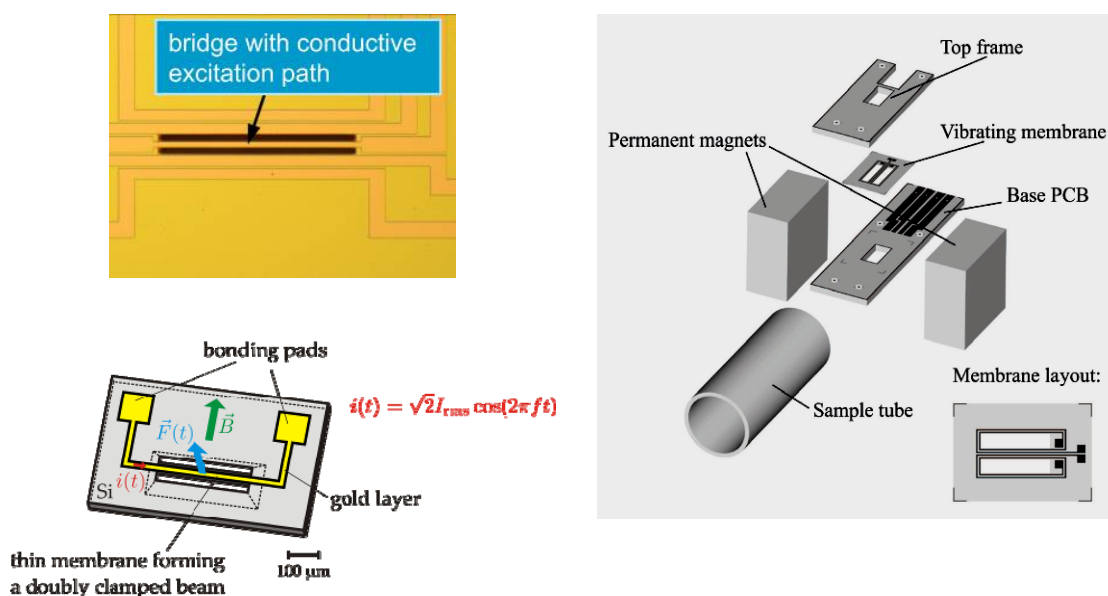

Fig. 3: Clamped-clamped beams in Si-micromachining technology (left, [15]) and polymer technology (right, [16]).

Modeling the interaction of the beam with the liquid is less straightforward. A suitable method considered is the implementation of Euler Bernoulli beam theory by introducing a distributed load along the beam, which represents the interaction with the liquid and thus depends on the transversal motion of the beam given by its lateral displacement $w(x,t)$ [17]. It can be shown [10] that for the calculation of the interaction between beam and liquid, the Navier-Stokes equations can be linearized if the displacement amplitudes in the liquid are sufficiently small. Using complex time-harmonic notation, the distributed load due to interaction with the liquid at some position x along the beam can be linearly related to the displacement amplitude at x by means of a complex-valued coefficient ("mechanical impedance"). In order to determine this coefficient, a 2D approximation can be made if it is assumed that the liquid movements in the direction of the beam axis are negligible. Thus, considering a vibrating cross section of the beam in a 2D model, the ratio between complex resistance force and associated displacement amplitudes can be determined to obtain said coefficient. In [17] the model involving a

spectral domain approach is described. Fig. 4 shows the problem and the plane of computation for the 2D problem as well as an example for the flow field around the vibrating beam cross section computed with the spectral method in 2D.
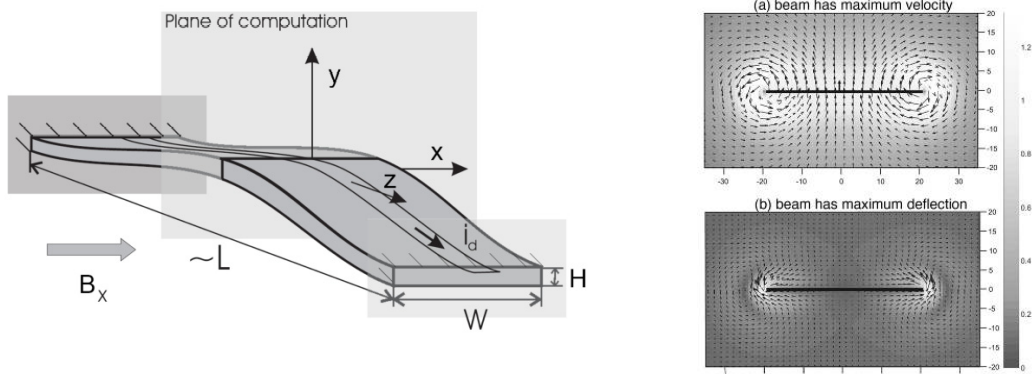

Fig. 4: Beam geometry and associated 2D plane of computation for the fluid resistance force (left) and example for the resulting flow field in the 2D problem (right).

The resulting effect on the beam's frequency response is involved but it can be roughly stated that the density mainly affects the resonance frequency while the viscosity dominantly determines the Q-factor (or damping coefficient D) of the resonances.

To compare the rheological behavior of beams and TSM resonators, suspensions and Newtonian liquid samples have been characterized with both devices [12]. A glimpse on the results is shown in Fig. 5. These results indicate that a TSM resonator is not suitable for the measurement of viscosity or concentration of the considered SiO_2–in–H_2O suspensions (Samples 1 to 8), whereas the results for Newtonian liquids (alcohols) follow the expected relation (4). For the vibrating bridge, all these liquids yield a damping coefficient nicely correlating with the liquid's viscosity as determined by a lab instrument [12].
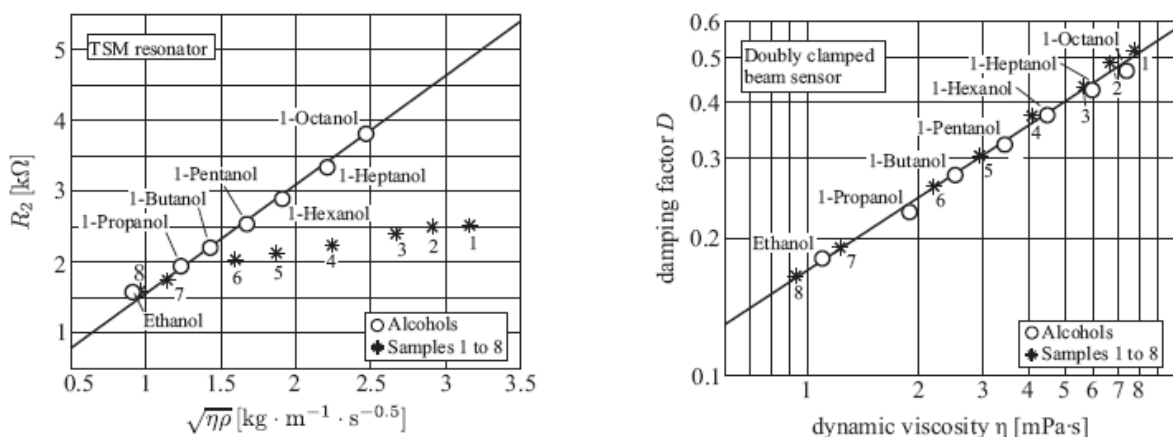

Fig. 5: Viscous loss resistance R2 of a 6 MHz quartz TSM resonator vs. square root of the viscosity-density product (left) showing deviations for non-Newtonian liquid samples. In contrast, the damping factor of a vibrating bridge shows a nice correlation with the viscosity also for complex liquids (right).

We finally note, that the concept of the beam devices is related to the concept of vibrating membranes. In [18] a device is described where the liquid under test is contained in a liquid cell featuring flexible bottom and top walls. Exciting vibrations in these walls, similarly as in the case of the vibrating bridges discussed above, the fluid properties of the liquid will influence the associated resonance properties. This device is suitable for integration in miniaturized fluidic systems. Figure 6 shows an associated device and typical results illustrating that the relation between the liquid parameters viscosity and density to the device parameters frequency and damping is more complex than for the devices described above (both liquid parameters influence both device parameters) [18].
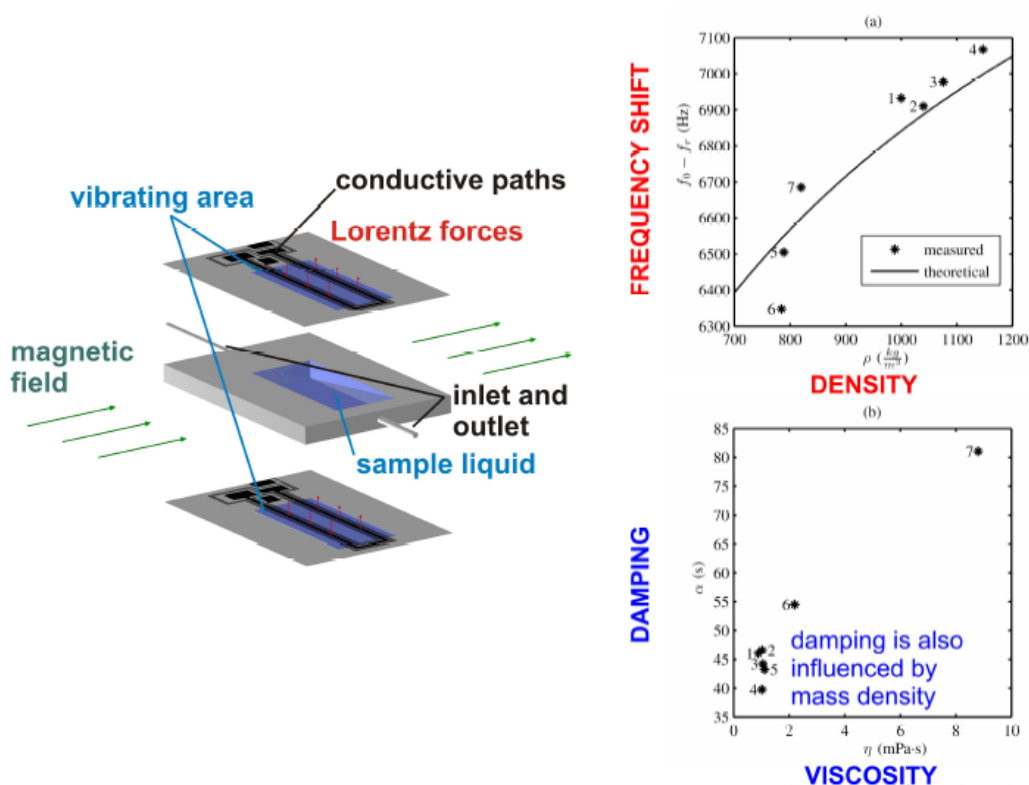

Fig. 6:   Double membrane device where the liquid is contained in a cell with vibrating top and bottom walls. Measurements for liquids with various viscosity and density values show that both values influence damping as well as viscosity (right)

Other Technologies for Liquid Sensor Systems

A lot of monitoring applications involve liquids containing particles. Separating particles with respect to size and/or density is thus an interesting approach to gain knowledge about a liquid sample (suspension). A suitable technology for particle sorting is the utilization of the dielectrophoretic effect. At our lab we are particularly working on 3D AC electro-osmotic (3D-ACEO) pumps, utilizing periodic arrays of interdigitated, stepped electrodes at the channel boundary. These pumps have on the one hand the ability to achieve flow rates up to mm s^{-1} by application of an AC voltage in the range of only a few volts and can on the other hand be utilized for simultaneous particle separation in the microchannel with respect to size and density. This novel separation mechanism is based on the competitive interplay of the forces acting upon the particles in the microchannel during operation of the pump, namely dielectrophoretic, viscous drag as

well as gravitational and buoyancy forces. By varying amplitude and frequency of the driving voltage, the ratio of these forces is tunable, which provides an effective means for size and density dependent particle manipulation, which we have studied both experimentally [19] and theoretically [20]. Based on these findings we have developed pump designs, capable of pumping and separating particle mixtures in suspensions simultaneously.

Infrared absorption measurements are another powerful way to characterize liquid samples. In a joint strategic research activity with Integrated Microsystems Austria, we work on the design of a fully integrated infrared absorption sensor, which is capable to determine chemical properties by investigating infrared absorption at selected distinct wavelengths in the mid infrared region. For example, in order to reduce the frequency of scheduled maintenance of lubrication oil, e.g., in combustion engines, the quality of the deteriorated oil has to be determined, which has been selected as pilot application for our development (this has been investigated in a previous project with AC2T research). Based on results of transmission spectroscopy, it was shown that by means of a properly designed optical waveguide structure, a sensitivity similar to that obtained by transmission spectroscopy, as used in the laboratory, can be achieved by an integrated system. To achieve a highly sensitive absorption element, a planar monomode waveguide is utilized which requires an appropriate coupler to couple the infrared light into the waveguide. The employed grating couplers have been modeled in order to derive a design yielding the desired sensor performance. As the targeted wavelength is located in the mid-infrared region, a suitable infrared source or detector, respectively, can be fabricated by thermal components [21].

Conclusions

Microsensor technology facilitates the implementation of sensors for online applications in industrial processes and plants. Devices involving vibrating structures immersed in liquids can be used to determine mechanical and in particular, rheological properties. Due to the associated small vibration amplitudes and high frequencies, the rheological domain can be quite different to that probed by common lab equipment. In particular, this may be an issue for non-Newtonian liquids such as emulsions and suspensions. For the monitoring of changes in the liquid such as phase transitions, the specifically considered rheological domain plays a minor role and the sensor output signals can be subjected to a calibration procedure of some sort.

Acknowledgements

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