

VISCOSITY SENSOR BASED ON A SYMMETRIC DUAL QUARTZ THICKNESS SHEAR RESONATOR

E. Benes, R. Thalhammer, M. Gröschl, H. Nowotny¹, S. Jary²

Vienna University of Technology, Institute of General Physics, ¹Institute of Theoretical Physics, Vienna, Austria/Europe

²Lenzing AG, Lenzing, Austria/Europe

Abstract - The paper describes a novel quartz crystal sensor for the measurement of the density-viscosity product of Newtonian liquids. The sensor element consists of two circular quartz crystal plates with an air-gap in between and the liquid sample in contact with the outer plate surfaces. Plano-convex AT-cut quartz crystals arranged in mirror symmetric crystallographic orientation and vibrating in an even-symmetric thickness-shear fundamental mode at 2.77 MHz are utilized. The two outer plane sides of the crystals are fully covered by gold electrodes, which are both connected to ground potential. This special mirror symmetric set-up allows the compensation of spurious displacements in the circular clamping zones of the two crystals. The measurement values of the sensor are the fundamental resonance frequency f and the associated resonance Q -value, which are analytically dependent on the density-viscosity product of the liquid in contact with the sensing surfaces. In contrast to an earlier report [1] about a sandwich resonator sensor, which entrapped the liquid sample between two quartz plates, the immersible sensor presented here is not restricted to low viscosity samples. The sensor covers a viscosity range from almost zero (air!) up to 2000 Pa.s, and is not restricted to electrically insulating liquids.

Keywords - Resonant sensors, BAW sensors, viscosity, dough

I. INTRODUCTION

The utilization of thickness-shear mode quartz crystals for the determination of liquid parameters has been investigated by several groups [2-5]. When operated at a properly chosen frequency (series resonance), the quartz crystal performs an in-plane oscillation (thickness-shear mode). This in-plane oscillation radiates a shear wave into the contacting viscous fluid, whereby the decay length of this wave is very small, typically only a few micrometers. The fluid layer contacting the quartz surface causes a decrease both of the resonance frequency and of the resonance Q -value, compared to the unperturbed (dry) crystal. For Newtonian liquids the density-viscosity product can be derived analytically from the measured electrical admittance curve, as will be reviewed in the following Section II.A.

In Section II.D the problem of spurious out-of-plane displacements is treated. It has already been shown in [1] how this problem can be solved by using a dual quartz sensor with the liquid between the two mirror-symmetrically arranged quartz plates. This three layer sandwich resonator was characterized by excitation of an even symmetric shear displacement curve which resulted in an angular momentum compensation. However, the drawbacks of this viscosity sensor with high absolute accuracy were its restriction to rather low viscosity liquids and the difficult cleaning procedures.

In Section III the design of an immersible dual quartz viscosity sensor is described and in Section V first measurement results obtained in rough industrial environment are presented.

II. THEORY

A. One-dimensional model

Fig.1 schematically shows the crystal-liquid interaction. A typical arrangement comprises a thin AT-cut quartz disk with electrodes on both sides. One surface of the quartz sensor is in contact with the liquid, while the opposite surface is air-backed. The quartz crystal is driven by a high-frequency voltage source. This is a one-dimensional, two layer model, with all quantities depending exclusively on the y -direction and on time.

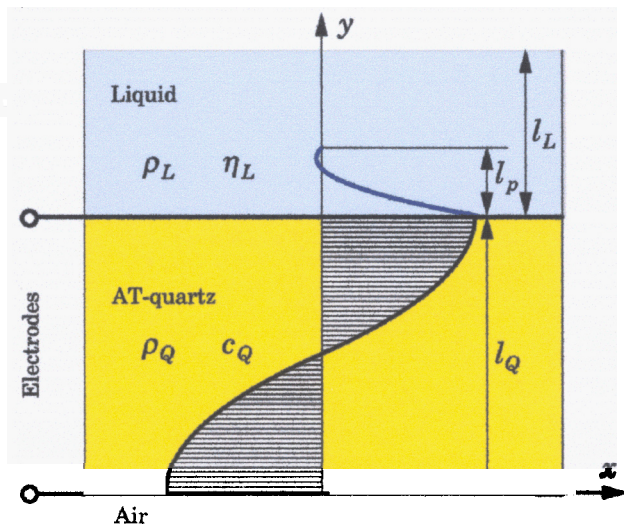


Fig.1 Displacement of the shear wave in the sensor quartz and in the adjacent fluid layer (schematically).

ρ_L, ρ_Q mass density of fluid / quartz

η_L shear viscosity of fluid

z_Q acoustic impedance of quartz

l_Q thickness of quartz layer

l_L thickness of fluid layer ($l_L \gg l_p$)

l_p decay length of shear wave

For the mathematical treatment, the liquid layer can be regarded as semi-infinite in y -direction as long as the penetration depth l_p of the shear wave into the liquid remains much smaller than the liquid layer thickness l_L . The properties of a

general viscoelastic layer can be described by a complex elastic stiffness constant [6].

$$c = c' + jc'' . \quad (1)$$

For the liquid limit the elastic constant becomes purely imaginary

$$c = jc'' . \quad (2)$$

For a Newtonian liquid the imaginary part is equal to the angular frequency ω_n at the regarded overtone number n multiplied by the dynamic viscosity η_L of the liquid, respectively:

$$c'' = \omega_n \eta_L . \quad (3)$$

The primary measurands of the viscosity sensor are the fundamental resonance frequency and the associated resonance Q -value. The product of density and dynamic viscosity of the fluid (liquid) in contact with the quartz crystal can be calculated out of these primary measurands. The calculation is based on the solution of the fundamental piezoelectric equations for a layered composite resonator structure according to Nowotny and Benes [7-9]. This rigorous model delivers the same numerical results as the layered resonator model according to Ballato [10, 11], although it is based on simple matrix multiplications and therefore a C++ program for the most general case was easily developed. For Newtonian liquids the sensor function can be derived as follows (see also [1])

$$\eta_L \rho_L = \frac{n^2 \pi z_Q^2}{4} \frac{1}{Q_{nL}^2 f_{n0}} = n^2 \pi z_Q^2 \frac{\Delta f_n^2}{f_{n0}^3} , \quad (4)$$

whereby n is the mode number, z_Q the acoustic impedance (real part) of the quartz, $\Delta f_n = f_n - f_{n0}$ the frequency shift due to the viscous load of the quartz, f_n , f_{n0} are the resonance frequencies of the loaded and unloaded resonator, respectively. Q_{nL} is the acoustic quality factor of the fluid that can be derived from the quality factors of the loaded and unloaded crystal, respectively:

$$\frac{1}{Q_{nL}} = \frac{1}{Q_n} - \frac{1}{Q_{n0}} . \quad (5)$$

The quality factor Q_{nL} and the series resonance frequency of the unloaded crystal f_{n0} can be determined from the locus of admittance $Y(f)$

$$Q_{nL} = \frac{f_n}{f_{n+} - f_{n-}} , \quad (6)$$

whereby the series resonance frequency f_n can be determined at the maximum of the real part of the locus of admittance $Y(f)$ and the half-value frequencies f_{n-} and f_{n+} are at the maximum and minimum of the imaginary part of the locus of admittance $Y(f)$ curve, respectively (see Fig.2).

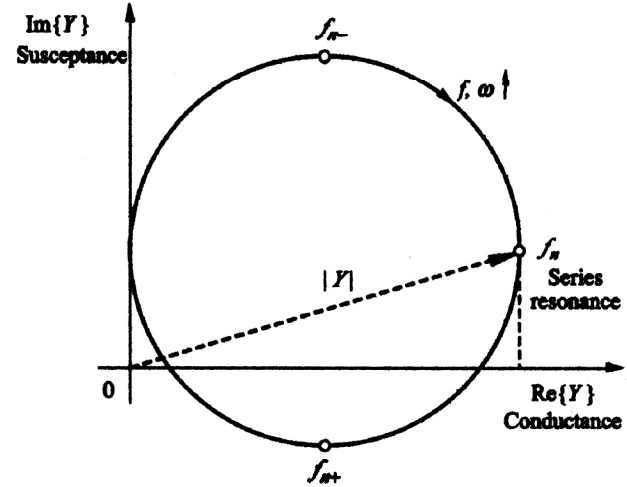


Fig.2 Locus of admittance $Y(f)$ with the used characteristic frequencies.

For lower viscosity values the use of the left part of (4) delivers much more accurate values for the density-viscosity product, while for high viscosities the right part might be used alternatively.

In case of Newtonian liquids Δf_n and Q_{nL} are related by

$$\Delta f_n = \frac{f_{n0}}{2Q_{nL}} . \quad (7)$$

This criterion can be used to discriminate between Newtonian and non-Newtonian liquids.

B. Consideration of real geometry with finite active area

The derived analytical sensor function accurately describes the situation as long as the basic assumption of one-dimensional treatment is justified. To preserve the high quality factor of free quartz, vanishing displacement amplitude at the edge of the quartz plate is desirable. This minimizes the influence of the quartz support on the vibration pattern. A powerful method to ensure more or less vanishing displacement at the edge, called "energy-trapping", can be realized in different ways. One possibility is to restrict the electrodes to the center area of the quartz, another method is to use a quartz plate of plano-convex or biconvex shape, a third method applies both concepts. For a good choice of the center thickness l_Q and the curvature radius R_c of the convex side of the quartz for a given plate diameter D , we optimized the so-called equivalent area A_{eq} , which is a measure for the effective vibrating area of the quartz. Under the as-

sumption of full-size electrodes the equivalent area can be calculated according to Schmid [12] as

$$A_{eq} = \frac{4\pi}{\sqrt{a_n b_n}} = \frac{4}{n} \sqrt{\frac{M_n c_{55}}{\bar{c}_{66}}} R_c \bar{l}_Q^3 = c_n \sqrt{R_c \bar{l}_Q^3}, \quad (8)$$

$$\text{with } c_n = \frac{4}{n} \sqrt{\frac{M_n c_{55}}{\bar{c}_{66}}}. \quad (9)$$

The definitions of the effective elastic constants are according to Tiersten [13]. Numerical values for relevant vibration modes are given in Table I.

TABLE I.

Effective elastic constants of AT-cut quartz depending on the overtone number n [14]; the constant $c_{55} = 68.8$ GPa.

n	M_n (GPa)	\bar{c}_{66} (GPa)	c_n (-)
1	110	29.06	6.92
3	75.8	29.22	2.10
5	90.1	29.23	1.31
7	80.4	29.24	0.911
9	88.7	29.24	0.726

Note that the radius of curvature can be chosen within the range

$$\left(\frac{D^2}{4} + \frac{\bar{l}_Q^2}{2l_Q} \right) < R_c \leq \infty, \quad (10)$$

the lower limit means zero edge thickness.

For the experimental verification of (8) it is useful to relate A_{eq} to the motional capacitance C_1 of the AT-quartz sensor [12] as

$$A_{eq} = \frac{\pi^2 \pi^2 l_Q C_1}{8 \epsilon_{22} k_Q^2}, \quad (11)$$

where ϵ_{22} is the corresponding dielectric constant. Furthermore, the modified Gaussian displacement distribution (33) on the plane surface of the plate ($y=0$) yields the relative displacement at the edge of the plate at $x = \pm D/2$

$$\frac{\xi_n(D/2)}{\xi_{n0}} = \exp \left[-\frac{8\pi}{A_{eq} b_n} \left(\frac{A_Q}{A_{eq}} \right) \right], \quad (12)$$

with the full sensor area $A_Q = D^2 \pi / 4$. For the chosen geometry, the relative amplitude at the edge of the quartz plate is extremely small (about 10^{-6} , see also [1]). This is a good

precondition for clamping of the quartz without mode coupling to the sensor casing.

The link between the infinite model and the real plano-convex geometry is made by introducing an equivalent thickness and the above defined equivalent (effective) excitation area. The equivalent thickness is given by the regarded thickness shear resonance frequency of the plano-convex crystal. The crystal layer thickness in the infinite model is chosen as the thickness of an infinite plate with the same series resonance frequency as the real resonator.

C. Influence of surface smoothness

Hydrodynamic smoothness of the surfaces in contact with the liquid is essential in order to achieve high sensor accuracy and reproducibility of results [14, 15]. Even for Newtonian fluids, viscoelastic effects may occur due to a thin liquid layer entrapped in surface cavities. The size of such cavities is determined by the crystal's surface roughness, which should be less than 100 nm.

D. Importance of even-symmetric displacement

The utilization of thickness-shear mode quartz crystals for the determination of liquid parameters has shown that, in general, these quartz crystals generate not only a damped shear wave, but also compressional waves in the liquid [16-19]. These compressional waves are caused by spurious out-of-plane displacements of the plate surface as a consequence of angular momentum conservation in a shear vibrating finite plate (see Fig.3)

Since the damping of compressional waves in liquids is much smaller than that of shear waves, (high overtone) compressional wave resonances may occur, affecting sensor performance due to mode coupling and additional energy loss. To overcome this fundamental problem, a special geometry of two quartz crystals with the liquid sample in between has been introduced recently [1]. The crystallographic orientation of the crystals and the electric potentials are chosen so that the resulting shear displacement curve of the total arrangement is even symmetric with respect to the center (x - z)-plane. The situation is shown schematically in Fig.3. A pure shear displacement according to Fig.3a is associated with an angular momentum caused by the in-plane movement in x -direction. In case of a free finite plate, the angular momentum conservation principle leads to an additional out-of-plane displacement in y -direction (Fig.3b) [20], thereby generating compressional waves in the contacting liquid.

They have a comparatively low amplitude, but propagate in the liquid with dramatically lower damping than the shear wave, where the sensor function is based upon and which exhibits a very low penetration depth. Typically, the compressional waves easily find a fitting high overtone resonant boundary condition and by coupling to the exciting shear wave cause a strongly frequency-dependent additional damping.

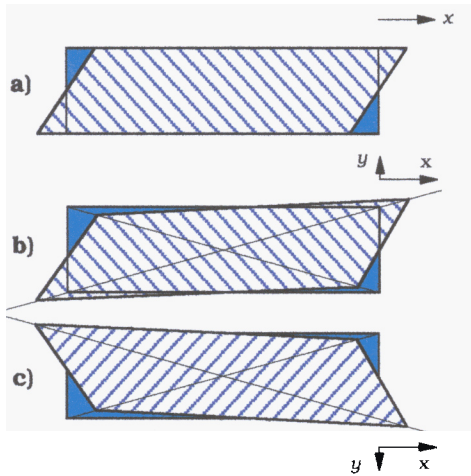


Fig.3. Displacement of a shear vibrating AT-cut crystal plate; a) pure shear displacement, b) shear displacement due to angular momentum conservation, c) mirror-symmetric displacement of second plate in the sandwich arrangement. The arrows indicate the in-plane (x) and the out-of-plane ($y, -y$) displacement directions, respectively.

The often ignored problem of out-of-plane vibration components of the usual odd symmetrically excited thickness shear crystal resonators was addressed in a recent paper of EerNisse [21] in which it was demonstrated that the high overtone bending modes generated by the rotational imbalance of the finite shear vibration mode is - in contrast to the regarded shear wave - not energy trapped. Even the effect of drive level dependence of resonant frequencies can be explained by considering the consequences of rotation imbalances in connection with angular momentum conservation.

With the even-symmetrically excited dual quartz sandwich resonator (Fig.3bc) containing the sample liquid between the two quartzes, a zero total angular momentum along with the compensation of disturbing compressional wave resonances was achieved. However, the drawback of this design was that it is not well suited for higher viscosity liquids ($\eta_L > 20$ Pa.s), which cannot easily be pressed through the small gap between the two crystals. At the same time the above described longitudinal mode coupling plays a much less important role for viscosities in the higher range.

III. DESIGN OF THE DUAL QUARTZ IMMERSIBLE VISCOSITY SENSOR

In contrast to the sandwich resonator viscosity sensor with high absolute accuracy according to [1], here a new version with two even symmetrically excited crystals with an air gap in between and the sample liquid covering the outer crystal surfaces is introduced. Since there is practically no acoustic coupling across the air gap, the only remaining advantage of the even symmetric excitation is that the out-of-plane displacement components of high overtone bending modes, which are not energy trapped, compensate each other at the clamping points. Whereby the clamping points actually form a circular line close to the edge of the circular plate. This

effect is comparable with the displacement compensation of tuning fork resonators in the nodal point.

The utilized sensor crystals are plano-convex 2.77 MHz AT-cut quartz crystals with an equivalent area of 12% (for the fundamental mode). The plane surfaces of both plano-convex crystals have been polished according to DIN 3140, resulting in a surface roughness below 100 nm.

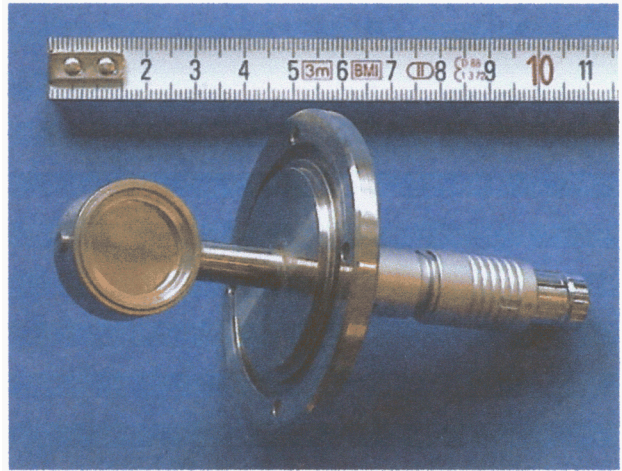


Fig.4 Photograph of the flange mounted immersible dual quartz crystal viscosity sensor (protection grid removed).

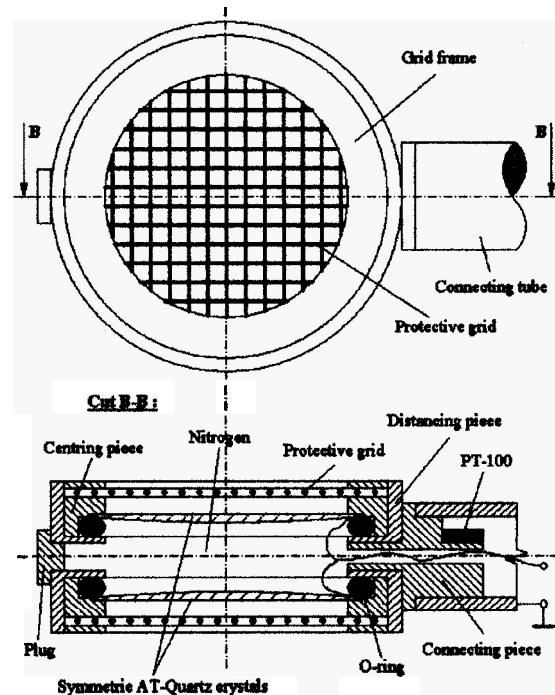


Fig.5 Principal and cross-sectional view of the flange mounted immersible dual quartz crystal viscosity sensor.

The Au-electrodes of both crystals are connected in parallel. The outer electrodes on the plane crystal surfaces are in contact with the liquid and electrically grounded. The space between the two crystals is filled with dry nitrogen. To prevent thermal stresses, the crystal plates are compliantly pressed by a Viton® O-ring to the circular metallic mass contact area on which the crystals are allowed to slip. Frequency alignment < 10 Hz of the two quartz crystals was performed by evaporating a thicker Au-electrode layer on the crystal with the initially higher resonance frequency. The flange mounted immersible dual quartz crystal viscosity sensor is shown in Figs.4 and 5.

IV. RESULTS

The viscosity sensor shown in Figs.4 and 5 was tested in rough industrial environment for on-line monitoring of waffle dough viscosity at the company Manner AG Vienna. Viscosity control is an important issue in the waffle dough production quality control. Fig.5 shows the temporal progress of dough viscosity and temperature. Between measurement numbers 14 and 22 water has been added, to influence the viscosity of the dough and see the sensor response.

In Fig.6 the interesting period between measurement numbers 14 and 22 is shown in an expanded scale.

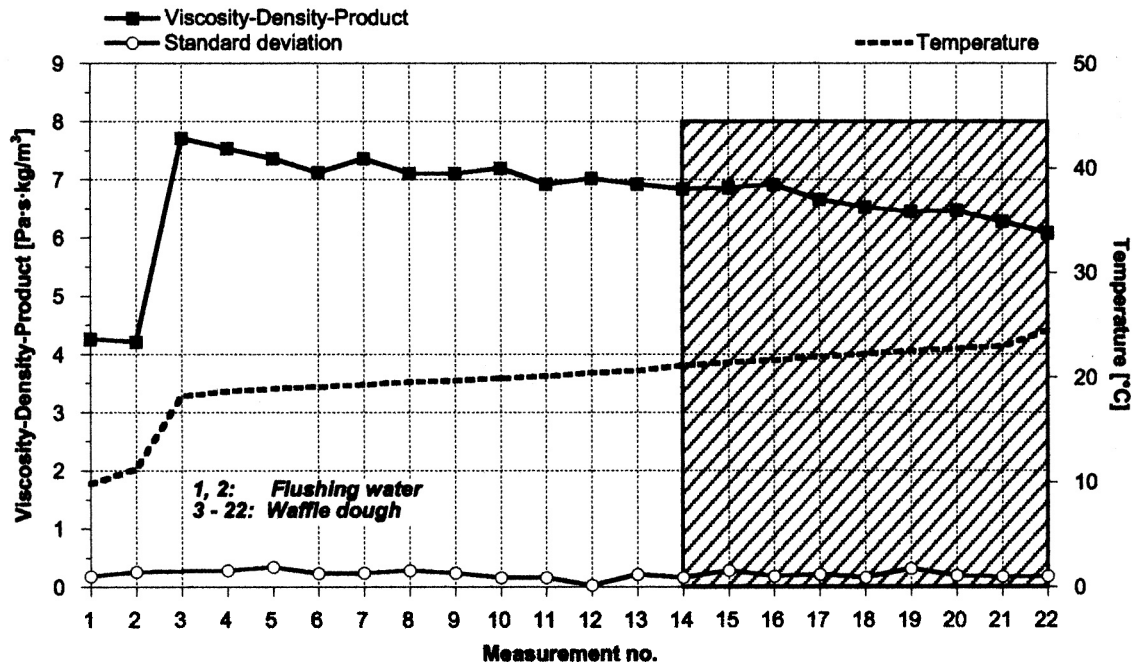


Fig.6 Temporal progress of dough viscosity and temperature. Time interval between two consecutive measurements approximately 10 min. Between measurement points 14 and 22 water was added

V. DISCUSSION

Dough viscosity is an essential parameter for quality control in waffles production. The immersible even symmetric dual quartz viscosity sensor exhibits the following advantages:

- Analytical sensor function free of spurious mode coupling
- No calibration needed, absolute error 2...60 %
- High viscosity range 0...2000 Pa.s
- Insensitive to gas bubbles This is a surprising result of high practical importance. It might be explained by the fact that the gas bubbles never come in direct contact with the crystal surface due to a permanent dough layer on the surface with a thickness higher than the shear wave penetration depth of a few micrometers.
- Measurement of high shear viscosity as it is relevant in many applications, e.g. in jet streaming and high performance engine lubrication.

The present disadvantages of the sensor are its restriction to low allowable pressure variations and its poor absolute accuracy when compared to the sandwich version with the liquid between the crystals. (The absolute error of the three layer sandwich resonator is between 2...5 %.)

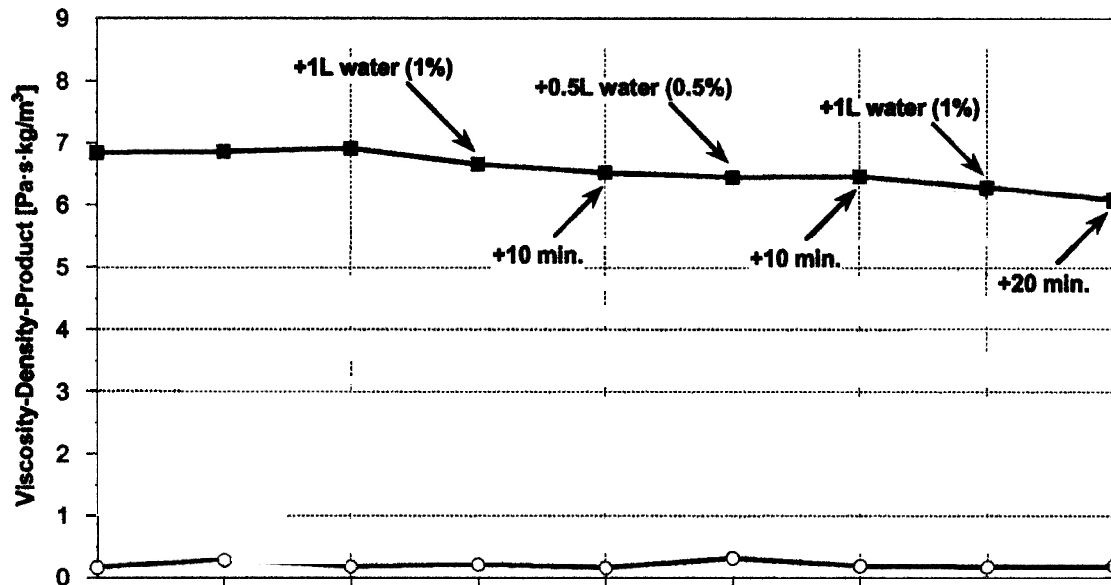


Fig.7 Sensor response when adding small amounts of water (1 vol.% or 0.5 vol.%, respectively). Time interval between two consecutive measurements approximately 10 min.

VI. CONCLUSION

A novel quartz crystal sensor for measurement of the density-viscosity product of Newtonian liquids has been introduced. The sensor element consists of two circular quartz crystal plates with an air-gap in between and the liquid sample in contact with the outer plate surfaces. The quartz crystals, together with a miniature platinum resistance temperature sensor, are mounted in a stainless steel housing.

In contrast to an earlier report [1] about a sandwich resonator sensor, which entrapped the liquid sample between two quartz plates, the immersible sensor presented here is not restricted to low viscosity samples. For Newtonian liquids, the double-quartz sensor features fair absolute accuracy and reproducibility without the need of calibration and without any fitting parameter. The sensor covers a viscosity range from almost zero (air!) up to 2000 Pa.s, and is not restricted to electrically insulating liquids. Non-Newtonian behaviour can be identified and the complex mechanical impedance of non-Newtonian fluids can be determined at high shear rates (10^7 s^{-1}). The flange mounted immersible sensor was employed successfully in rough industrial environment for on-line monitoring the viscosity and temperature of waffles dough flowing through the dough supply pipe of a waffles production plant (Manner®-Schnitten, Wien, Austria). The high viscosity resolution could be demonstrated by adding small amounts of water (1 vol.% or 0.5 vol.%, respectively) for which corresponding decreases in dough viscosity could be observed.

ACKNOWLEDGMENT

This work was supported by MANNER AG, Wien, Austria, LENZING AG, Austria and by the European Commission within the GROWTH shared-cost RTD funding scheme, project "QxSens" No. GRD1-2001-41816.

REFERENCES

For informations on the QxSens Consortium refer to www.qxsens.net

- [1] R. Thalhammer, S. Braun, B. Devcic-Kuhar, M. Gröschl, F. Trampler, E. Benes, H. Nowotny, and M. Kostal, "Viscosity sensor utilizing a piezoelectric thickness shear sandwich resonator," *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, vol. 45, pp. 1331-1340, 1998.
- [2] C. E. Reed, K. K. Kanazawa, and J. H. Kaufman, "Physical description of a viscoelastically loaded AT-cut quartz resonator," *J. Appl. Phys.*, vol. 68, pp. 1993-2001, 1990.
- [3] S. J. Martin, G. C. Frye, and K. O. Wessendorf, "Sensing liquid properties with thickness-shear mode resonators," *Sensors and Actuators A*, vol. 44, pp. 209-218, 1994.

- [4] J. Auge, P. Hauptmann, F. Eichelbaum, and S. Rösler, "Quartz crystal microbalance sensor in liquids," *Sensors and Actuators B*, vol. 18-19, pp. 518-522, 1994.
- [5] J. Auge, P. Hauptmann, J. Hartmann, S. Rösler, and R. Lucklum, "New design for QCM sensors in liquids," *Sensors and Actuators B*, vol. 24-25, pp. 43-48, 1995.
- [6] R. Holland, "Representation of dielectric, elastic, and piezoelectric losses by complex coefficients," *IEEE Trans. Sonics Ultrason.*, vol. 14, pp. 18-20, 1967.
- [7] H. Nowotny and E. Benes, "General one-dimensional treatment of the layered piezoelectric resonator with two electrodes," *J. Acoust. Soc. Am.*, vol. 82, pp. 513-521, 1987.
- [8] H. Nowotny, E. Benes, and M. Schmid, "Vibration modes of contoured piezoelectric resonators with additional film layers," presented at 2nd European Frequency and Time Forum, Neuchâtel, Switzerland, 1988.
- [9] H. Nowotny, E. Benes, and M. Schmid, "Layered piezoelectric resonators with an arbitrary number of electrodes (general one-dimensional treatment)," *J. Acoust. Soc. Am.*, vol. 90, pp. 1238-1245, 1991.
- [10] A. Ballato, "Bulk and surface acoustic wave excitation and network representation," presented at 28th AFCS, 1974.
- [11] A. Ballato, H. L. Bertoni, and T. Tamir, "Systematic design of stacked-crystal filters by microwave network methods," *IEEE Trans. Microwave Theory Tech.*, vol. 22, pp. 14-25, 1974.
- [12] M. Schmid, E. Benes, W. Burger, and V. Kravchenko, "Motional capacitance of layered piezoelectric thickness-mode resonators," *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*, vol. 38, pp. 199-206, 1991.
- [13] H. F. Tiersten and R. C. Smythe, "An analysis of contoured crystal resonators operating in overtones of coupled thickness shear and thickness twist," *J. Acoust. Soc. Am.*, vol. 65, pp. 1455-1460, 1979.
- [14] S. J. Martin, K. O. Wessendorf, C. T. Gebert, G. C. Frye, R. W. Cernosek, L. Casaus, and M. A. Mitchell, "Measuring liquid properties with smooth- and textured-surface resonators," presented at 1993 IEEE Int. Freq. Control Symp., Salt Lake City, UT, USA, 1993.
- [15] S. J. Martin, G. C. Frye, A. J. Ricco, and S. D. Senturia, "Effect of surface roughness on the response of thickness-shear mode resonators in liquids," *Anal. Chem.*, vol. 65, pp. 2910-2922, 1993.
- [16] F. Eggers and T. Funck, "Method for measurement of shear-wave impedance in the MHz region for liquid samples of ≈ 1 ml," *J. Phys. E: Sci. Instrum.*, vol. 20, pp. 523-530, 1987.
- [17] Z. Lin and M. D. Ward, "The role of longitudinal waves in quartz crystal microbalance applications in liquids," *Anal. Chem.*, vol. 67, pp. 685-693, 1995.
- [18] T. W. Schneider and S. J. Martin, "Influence of compressional wave generation on thickness-shear mode resonator response in a fluid," *Anal. Chem.*, vol. 67, pp. 3324-3335, 1995.
- [19] R. Lucklum, S. Schranz, C. Behling, F. Eichelbaum, and P. Hauptmann, "Analysis of compressional-wave influence on thickness-shear-mode resonators in liquids," *Sensors and Actuators A*, vol. 60, pp. 40-48, 1997.
- [20] B. A. Auld, *Acoustic fields and waves in solids*, vol. I. New York: John Wiley & Sons, 1973.
- [21] E. P. EerNisse, E. Benes, and M. Schmid, "The role of localized rotational imbalance in drive level dependence phenomena," presented at 2002 IEEE 56th Annual Symposium on Frequency Control, New Orleans LA, USA, 2002.