MICROMACHINED VISCOSITY SENSOR FOR REAL-TIME POLYMERIZATION MONITORING

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SUMMARY

This paper reports on low-frequency micromachined viscosity sensors intended for real-time polymerization monitoring. The viscosity sensors consist of micromachined membrane resonators featuring electrothermal excitation and piezoresistive detection of transverse membrane vibrations. In contrast to high-frequency (above 1 MHz) sensors, e.g. thickness-shear mode sensors or Lamb wave sensors, the use of low-frequency (1-20 kHz) resonators for viscosity sensing, which more closely mimics conventional resonant viscometers, is investigated in this work. The viscous fluid loads the membrane resonator and changes the quality factor of its fundamental resonance. The change in the Q-factor is most pronounced in the viscosity range from 10^{-3} Pa·s to 1 Pa·s.

Keywords: resonator, viscosity sensor, membrane

INTRODUCTION

Most commercially available viscometers used for on-line process control, e.g., monitoring the degree of reaction in a polymerization, belong to one of the following categories [1]: (a) viscometers based on pressure driven flows (e.g. capillary viscometers), (b) rotational, (c) falling-piston/sphere, and (d) vibrational viscometers. For the case of vibrational viscometers, the damping of the sensor vibration is a function of the product of the density and the viscosity of the fluid sensed. Vibrational viscometers can be relatively small in size, can feature fast response time, and can be installed as immersion units, e.g., in a process vessel [1]. In contrast to "volume loaded" viscosity sensors, such as the capillary viscometers, the vibrational viscometers are "surface loaded" devices, i.e. the deformation of the fluid is limited to a thin layer surrounding the sensor.

Recently, acoustic wave microsensors [2], including thickness-shear mode (TSM) [3, 4] and Lamb wave sensors [5, 6], have been investigated as miniaturized versions of vibrational viscometers. Whereas in the case of the TSM devices the vibrations are parallel to the sensing surface, Lamb wave or flexural plate wave (FPW) sensors exhibit deflections parallel and normal to the surface. Typically, the operating frequencies of the TSM and FPW devices range from 1 to 10 MHz [2]. The thickness of the fluid layer probed is of the order of 0.25 µm in water for a 5 MHz TSM quartz resonator [3]. In addition to acoustic wave devices, passive, micromachined shear-stress sensors [7] have been employed for liquid sensing.

In contrast to the above work, the use of low-frequency (1-20 kHz) resonators for viscosity sensing, which more closely mimic conventional resonant viscometers, is investigated in this work. In particular, the influence of the viscosity of standard polymeric solutions on the characteristics of micromachined membrane resonators, such as quality factor and resonance frequency, is investigated.

MEMBRANE RESONATOR DESIGN

Square membrane resonators with a side length between 1 and 3 mm have been fabricated. The devices are comprised of a monocrystalline silicon layer with one or more dielectric layers (thermal oxide and passivation) on top (see schematic in Fig. 1). P-doped silicon resistors in the membrane center and close to the membrane edge are used for electrothermal excitation and piezoresistive detection of transverse vibrations, respectively.

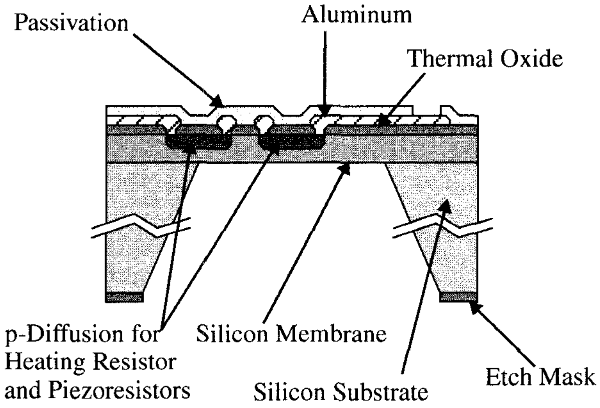

Fig. 1: Schematic of the membrane resonator with heating resistor for excitation and piezoresistors for detection.

The resulting fabrication sequence is compatible with standard CMOS or bipolar IC technology. The membrane resonators are released in a post-processing anisotropic etching step using a 30 % potassium hydroxide solution. Fig. 2 shows a photograph of a finished membrane resonator with heating resistor as well as four piezoresistors in a Wheatstone bridge configuration.

The membrane chips are mounted in standard ceramic Dual-In-Line packages for testing (see Fig. 2). The bonding wires are sealed with a high elastic casting compound on polyurethane-base in order to reduce thermal stresses due to mismatch of thermal expansion coefficients between resonator and packaging.

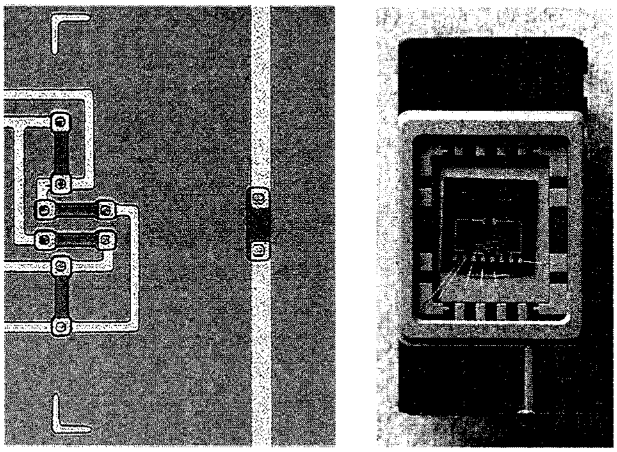

Fig. 2: (Left) Photograph of a fabricated 1.5 mm by 1.5 mm membrane resonator with the driving resistor in the center and the piezoresistors arranged in a Wheatstone bridge close to the edge. (Right) Complete membrane structure after wire bonding but prior to sealing

The membranes investigated in this work are subject to compressive in-plane stresses mainly induced during the growth of the thermal oxide. Due to the compressive stress, the membranes buckle if their thickness becomes smaller than a critical value which itself depends on the membrane dimensions and material properties. Resonators with an axial load close to the buckling load exhibit minimal fundamental resonance frequency and maximum vibration amplitudes [8, 9]. Therefore, membrane resonators with very small resonance frequency can be designed by proper control of the fabrication-induced in-plane stresses.

EXPERIMENTAL RESULTS

To excite transverse membrane vibrations, an ac voltage $U_{ac}\cos\omega t$ superimposed on a dc voltage U_{dc} is applied to the heating resistor (with resistance R). Therefore, one component (P_{dyn1}) of the thermal power P has the same frequency as the applied ac voltage [8, 9]:

$$P = \frac{1}{R}\left[U_{dc}^2 + \frac{1}{2}U_{ac}^2 + 2U_{dc}U_{ac}\cos\omega t + \frac{1}{2}U_{ac}^2\cos 2\omega t\right]$$
$$= P_{stat} + P_{dyn1}\cos\omega t + P_{dyn2}\cos 2\omega t$$

The static heating power P_{stat} causes a static temperature elevation of the membrane resonator and can be used, e.g., to adjust the resonance frequency of the device [8, 9].

Membrane Characteristics in Air

First, the basic characteristics of the membrane resonators, such as resonance frequency, vibration amplitude and sensitivity of piezoresistive detection, were investigated in air. Vibration amplitude and resonance frequency were measured optically using a laser heterodyne interferometer in combination with a network/spectrum analyzer HP 4195A. The ac output signal of the Wheatstone bridge (biased at $U = 5$ V) consisting of four p-doped piezoresistors was amplified with a low-noise differential amplifier (gain $V = 10$) and detected with either a network/spectrum HP 4195A or a gain/phase analyzer HP 4194A.

As an example, Fig. 3 shows the vibration amplitude as well as the piezoresistive output signal after amplification (gain $V = 10$) of a 2.5 mm by 2.5 mm membrane as a function of the driving frequency near the fundamental resonance. With a thermal heating power $P_{stat} = 16$ mW and $P_{dyn1} = 13$ mW, vibration amplitudes of approximately 300 nm are detected at the fundamental resonance frequency $f = 6.88$ kHz. A quality factor $Q = 115$ can be calculated from the measured 3-dB bandwidth at resonance. As expected, the output signal of the Wheatstone bridge with its four piezoresistors is proportional to the vibration amplitude. With respect to the vibration amplitude A, the piezoresistive detection has a sensitivity $S_{piezo} = U_{piezo}/A = 0.27$ µV/nm for a bias voltage of 5 V.

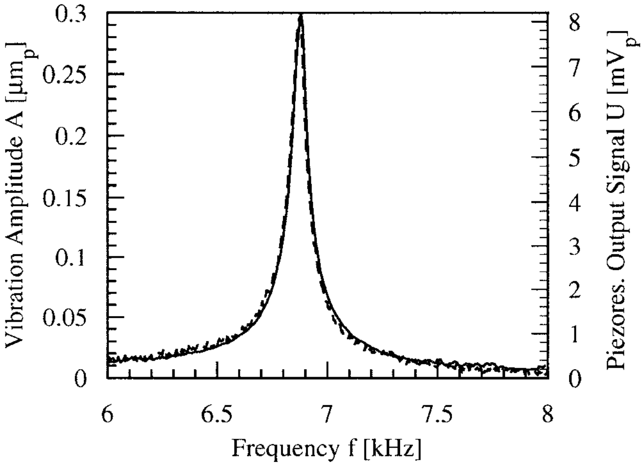

Fig. 3: Vibration Amplitude in the center of a 2.5 mm by 2.5 mm membrane (solid line) and piezoresistive output signal (dashed line; amplified with V = 10) around the fundamental resonance frequency f = 6.88 kHz (heating power: P_{stat} = 16 mW and P_{dyn1} = 13 mW).

The different membrane resonators investigated in this work have fundamental resonance frequencies ranging from 5 kHz to 60 kHz in air. The largest membranes (with 2.5 mm and 3 mm side length) show fundamental resonance frequencies in the range of 5 - 7 kHz if the axial load is close to the buckling load. For these devices, the vibration amplitudes at resonance can be as large as 3 µm even for small dynamic heating power $P_{dyn1} \approx 30$ mW. However, the resonators exhibit nonlinear behavior associated with stress-stiffening and stress-softening at large vibration amplitudes. In order to minimize these nonlinear effects, the membrane resonators are operated at relatively small vibration amplitudes (typically 100 - 300 nm) during characterization. The quality factor of the fundamental resonance in air ranges from 60 - 120 depending on the resonance frequency of the device.

Membrane Characteristics in Viscous Fluids

Polydimethylsiloxanes (PDMS) with viscosities ranging from 1 to $6 \cdot 10^5$ cSt (corresponding to $8 \cdot 10^{-4}$ to 600 Pa·s) were used to investigate the properties of the resonators in viscous polymer solutions. Whereas the viscosity changes over more than five orders of magnitude, the density of the polymer solutions increases only from about 0.82 g/cm^3 to 0.98 g/cm^3 for the solutions with viscosity of 1 cSt and $6 \cdot 10^5$ cSt, respectively. Therefore, the influence of the density of the PDMS solutions on the membrane damping was neglected compared to the viscosity effect.

For testing, the packaged membrane resonators were mounted in a test setup which only allows contact of the test fluid with the front side of the membrane resonator. All measurements were performed at room temperature and the polymer solutions were not stirred. The fundamental resonance frequency f as well as the quality factor Q were investigated as a function of the viscosity of the test fluid. Two different measurement schemes were used to access Q and f: (a) the membrane resonators were excited at their fundamental resonance using a burst signal and the quality factor of the resonance is obtained from the amplitude decay of the piezoresistive output signal or (b) the amplitude transfer characteristic around the fundamental resonance was recorded using a gain/phase analyzer and Q was obtained from the 3-dB bandwidth at resonance. As an example, Fig. 4 displays the response of a 1.4 mm by 1.4 mm sensor to the burst excitation, i.e. the amplified piezoresistive output signal, for two different PDMS solutions having viscosities of 1 and 10 cSt, respectively. The faster amplitude decay and, therefore, the reduced quality factor in the polymer with higher viscosity is clearly visible.

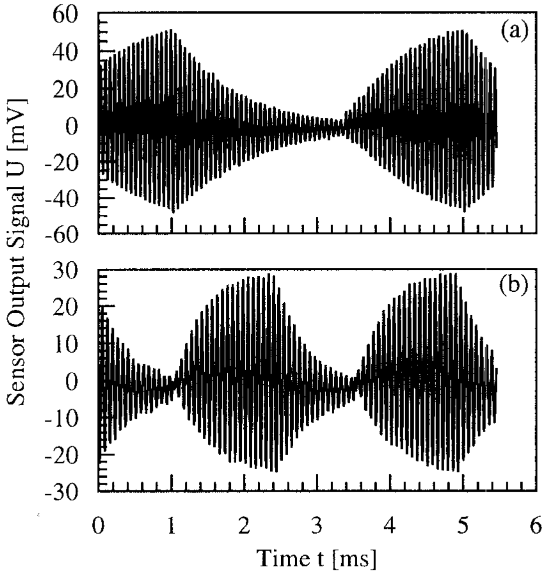

Fig. 4: Output signal of the membrane resonator subject to a burst excitation for (a) a 1 cSt ($8.2 \cdot 10^{-4}$ Pa·s) and (b) a 10 cSt ($9.4 \cdot 10^{-3}$ Pa·s) PDMS solution. Note that trace (a) was measured at a lower burst rate because of the longer decay time in the less viscous solution.

The characteristics of the membrane resonators are compared to the behavior of a commercially available non-micromachined piezoelectric transducer with a diameter of 25 mm in the PDMS solutions. The piezoelectric transducer consists of a circular brass membrane with a piezoceramic glued to it. Applying an ac voltage to the piezoceramic generates transverse vibrations of the brass membrane. The vibrations can be sensed piezoelectrically with a second pair of electrodes. The transducer has a fundamental resonance frequency of about 4.5 kHz in air.

Fig. 5 shows the quality factor Q of a 1.4 mm by 1.4 mm membrane resonator (triangles) and the piezoelectric transducer (circles) as a function of the shear viscosity of the surrounding medium. Measurements were performed in air (open symbols) as well as in different PDMS solutions (solid symbols). The quality factor of the micromachined membrane resonator drops from 78 in air to about 6 in a 10^3 cSt PDMS solution. The behavior found for the larger piezoelectric transducer is very similar, except that the Q-factor changes are most pronounced in the higher viscosity range.

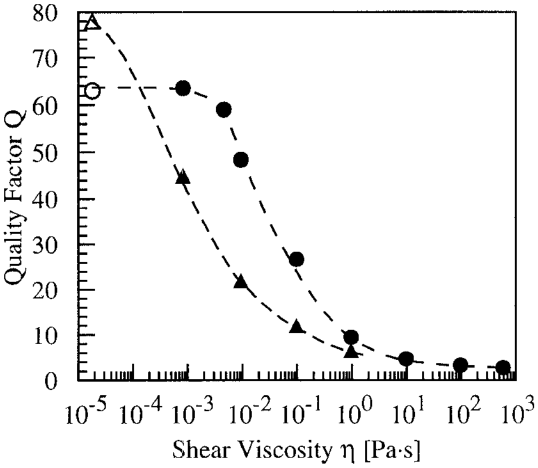

Fig. 5: Quality factor of the piezoelectric transducer (circles) and a 1.4 mm by 1.4 mm membrane resonator (triangles) as a function of the shear viscosity in air (open symbols) and different PDMS solutions (solid symbols).

The resonance frequency strongly decreases if the sensors are operated in a liquid medium instead of air due to the increase in density. As expected, the resonance frequency changes only slightly among the different PDMS solutions since their density stays almost constant. Within the viscous fluids a fundamental resonance frequency of approximately 14 kHz and 3 kHz is found for the 1.4 mm micromachined membrane resonator and the piezoelectric transducer, respectively.

Fig. 6 shows the change in resonance frequency f/f_{air} for the piezoelectric transducer (open circles) as well as for different membrane resonators with sizes varying from 1.4 mm to 3 mm (solid symbols). Whereas the resonance frequency of the piezoelectric transducer is reduced only by about 30 % if immersed in the fluids, the resonance frequency of the membranes typically drop to one fifth of the value in air. The larger effect of the fluid on the resonance frequency of the micromachined devices can be explained by their smaller mass. For the different PDMS solutions, the resonance frequency decreases slightly with increasing molecular weight since the density of the fluids increases by about 20 %.

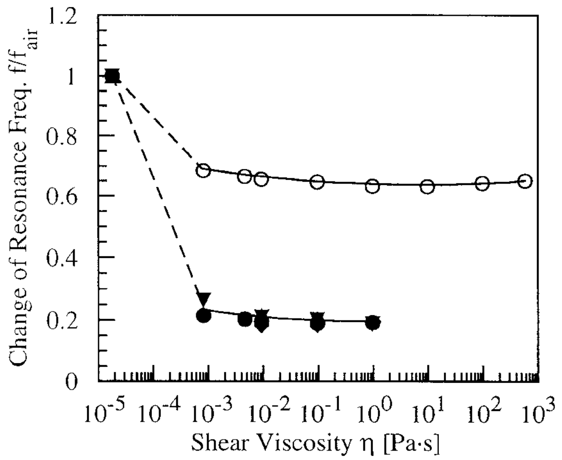

Fig. 6: Change of fundamental resonance frequency f/f_{air} of the piezoelectric transducer (open circles) and the membrane resonators (solid symbols) as a function of the shear viscosity.

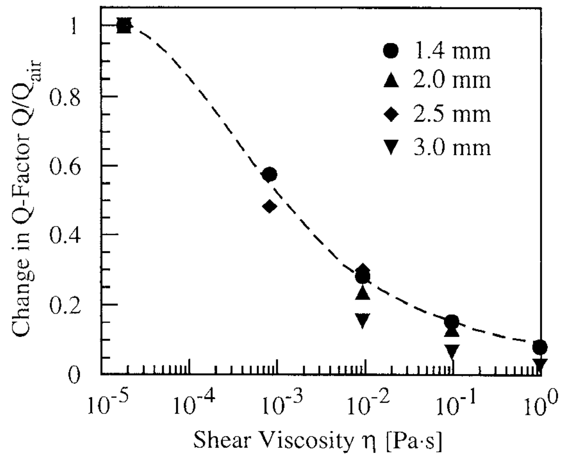

Fig. 7: Change of quality factor Q/Q_{air} of membrane resonators of different side lengths as a function of the shear viscosity.

Normalizing the quality factor Q by the quality factor in air Q_{air} shows that all membrane resonators exhibit a similar behavior (see Fig. 7) within the viscous fluids. The change of the Q-factor is most pronounced in the fluids with viscosities smaller than 1 Pa·s.

CONCLUSIONS

Resonant membrane devices for characterization of polymer solutions have been designed, fabricated, and characterized. Unloaded resonant quality factors between 60 and 120 in air have been achieved. These devices operate at frequencies below 20 kHz in viscous liquids. In viscous liquids, measurement of viscosity over the range of 10^{-3} to 1 Pa·s has been demonstrated.

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