Free Suspended Thin-Walled Nickel Electroplated Tubes for Microfluidic Density and Mass Flow Sensors

Mahdieh Yariesbouei, Remco G. P. Sanders, Taghi Moazzenzade, Remco J. Wiegerink, and Joost C. Lötters

Abstract—In this paper, a novel fabrication method is proposed for microfluidic tubes with a large diameter, circular cross-section, and thin wall. These properties make the tubes especially suitable for density sensors and Coriolis mass flow sensors, because of the resulting low tube mass, low-pressure drop, and low pressure-dependence of the tube shape. A demonstrator sensor was fabricated and the first measurement results of fluid density and mass flow are presented. The low-cost fabrication method is based on electroplating technology and results in tubes with a near-perfect circular cross-section. Diameters ranging from 120 \( \mu m \) to 1 mm and wall thicknesses from 8 \( \mu m \) to 60 \( \mu m \) have been achieved. For the demonstrator sensor presented in this paper a freely suspended tube was realized with a total length of 37 mm, a diameter of 600 \( \mu m \), and a wall thickness of 20 \( \mu m \). Density measurements were performed using various gases, liquids, and liquid mixtures at 21\(^\circ\)C to 23\(^\circ\)C lab temperature. The accuracy of the measured densities of gases such as nitrogen, argon, and helium is 5%. For liquids including DI water, isopropyl alcohol (IPA), and their various mixtures an accuracy of 0.5% was obtained. Preliminary mass flow rate measurements were performed with water and isopropyl alcohol up to 30 g/h with less than 30 mbar pressure drop thanks to the large tube diameter.

Index Terms—Cylindrical tube, density sensor, nickel electroplated tube, mass flow sensor.

I. INTRODUCTION

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VER the past decades, a large variety of applications have been presented for micromachined density sensors, such as the estimation of the state of the charge (SOC) and the state of health (SOH) in batteries [1], controlling the insulation gas in high voltage switchgears [2], and checking the percentage of sucrose and alcohol in the beverage industry [3].

The basic principle of density measurement of a fluid inside a vibrating tube, is that the density of the fluid directly affects the resonance frequency of the tube. This resonance frequency variation is due to the change in the total mass of the structure, the higher the density of the fluid, the lower the resonance frequency. For high mass sensitivity, the tube should have a large diameter to wall thickness ratio, resulting in a large relative change in mass. Furthermore, the tube wall should not deform due to the pressure, as this affects the stiffness of the tube which would also result in a change in resonance frequency. To prevent such deformations, especially with a relatively thin tube wall, a circular cross-section is needed.

The first silicon micromachined vibrating tube-based density and Coriolis mass flow sensors were proposed by Enksson et al. [4], [5] and consisted of a silicon tube with a hexagonal cross-section. The tube diameter was 800 \( \mu m \) and wall thicknesses of 50 \( \mu m \) and 100 \( \mu m \) were realized, giving a diameter to wall thickness ratio of 16.

Surface micromachined tubes as proposed by e.g. Westberg et al. [6] typically result in rectangular cross-sections and a relatively poor height to wall thickness ratio due to the limited thickness of the sacrificial layer. Tubes with rectangular cross-sections can also be made using wafer bonding as proposed by Sparks et al. [3], who presented a micromachined silicon tube with a rectangular cross-section of 500\( \mu m \times 120\mu m \) and a height to wall thickness ratio of about 5. Dijkstra et al. [7] proposed the so-called surface channel technology (SCT) to fabricate semicircular tubes with a thin silicon-rich silicon nitride tube wall. These tubes have a semicircular cross-section with a flat top side. Haneveld et al. [8] fabricated a density and micro-Coriolis mass flow sensor in this technology. The diameter of 40 \( \mu m \) and wall thickness of 1.2 \( \mu m \) result in a large diameter to wall thickness ratio of 33, however, the flat top side does result in some pressure dependence [9].

The use of thermally elongated glass capillaries was proposed by Malvar et al. [10]. Despite the fact that the inner and outer diameter can be varied by adjusting parameters such as laser power orpulling force, the maximum ratio between inner diameter and wall thickness is limited to approximately 4.2[11]. Furthermore, the glass capillaries are straight tubes that are clamped at both ends and differently shaped structures are not possible with this method [12]–[14].

Some of the vibrating tubes can also measure mass flow rate when operated as a Coriolis-type mass flow sensor [15]–[17].
In that case, the fluid flow inside the vibrating tube results in Coriolis forces that excite a secondary vibration mode. This is illustrated in Figure 1 for a U-shaped tube that is actuated in twist mode (Figure 1a) and Coriolis forces due to fluid flow induce a swing motion (Figure 1b), or vice versa.

In this paper, we present a fabrication method based on electroplating of nickel using a sacrificial pre-shaped polymer wire with a circular cross-section as a mold. We also present a demonstrator device and the results of density and mass flow measurements. The basic idea of fabricating microtubes by the electroplating method and some first results have already been presented in our previous study [19], [20].

Surface metallization of polymers has been widely used in different industries for various purposes, such as magnetic shielding, electrical conductivity, abrasion and corrosion resistance, waterproofing, and decoration [21]. In this paper, we use the strength and uniformity of the electroplated coating and the possibility to pre-shape the polymer wire to realize a freestanding tube.

The paper is organized as follows. In the next section, we explain the proposed fabrication process in detail. Next, we present the design and realization of a demonstrator density and mass flow sensor with a tube diameter of 600 μm and wall thickness of 20 μm. Finally, we discuss the results of density and mass flow measurements with various fluids.

II. Fabrication Process

A schematic representation of a cross-sectional view of the fabrication sequence is indicated in Figure 2. The process starts with a wire of acrylonitrile butadiene styrene (ABS), which is a commonly used thermoplastic polymer with a glass transition temperature around 105 °C, figure 2(a). The wire has a circular cross-section and can be pre-shaped as needed for the device that is fabricated so that bending of the fabricated tubes is avoided. In the second step, figure 2(b), the shaped wire is dipped in silver conductive paint (Electrolube SCP26G) and dried at room temperature to create a seed layer for the electroplating process. To avoid a thick layer of the silver paint on the wire during the dip-coating process, the silver paint is first diluted with isopropyl alcohol (IPA). The ratio of the silver conductive paint to IPA was chosen 3 to 1. This ratio resulted sufficient coverage of the ABS surface. A larger ratio resulted in voids in the coating.

The electroplating setup consists of two electrodes immersed in an electrolyte bath and a DC current source. In the electrodeposition process, nickel ions are dissolved in the solution by oxidation of the 99.99% pure nickel anode, and these ions are reduced on the surface of the cathode to form the coating [22]. In the third step, figure 2(c), a layer of nickel is grown on the surface of the conductive ABS wire by electroplating.

We used the electrolyte bath composition and operating conditions according to the bright nickel coating process given in [21], [22]. The duration of the electroplating process and the current define the average wall thickness $d$ of the resulting tube as follows [22]:

$$d = \frac{cIt}{A_e},$$

where $I$ is the current, $t$ is the duration of the electroplating process, $A_e$ is the area that will be electroplated, and $c = 4.426 \text{ m}^3/\text{As}$ is a constant.

In the fourth step, figure 2(d), the nickel-coated wire is immersed in a beaker with acetone in an ultrasound bath for several hours to completely dissolve the ABS wire inside the tube.

With this fabrication process, a wide range of tube diameters from 120 μm to 1 mm, and wall thicknesses from 8 μm to 60 μm have been realized. Figure 3 shows photographs of some of the resulting freestanding tubes. The rough and deformed tube ends are due to cutting it at the desired length.

III. Basic Structure and Operating Principle

The basic structure and operating principle of the density and mass flow sensor are shown in figure 4. The sensor consists of a U-shaped tube that is brought into vibration at its resonance frequency by Lorentz forces. An AC current $i_{act}$ is fed through the tube wall in the presence of a magnetic field $B$. With three permanent magnets, a strong and uniform field...
is generated around the tube. The resulting Lorentz forces are given by:
\[
\vec{F}_{\text{act}} = L \cdot (\vec{v}_{\text{act}} \times \vec{B}),
\]
where \( L \) is the length of the tube segments that are perpendicular to the magnetic field direction, as indicated in figure 4. The direction of the Lorentz force at the left and right sides of the sensor is opposite, resulting in the actuation of the twist mode as indicated by \( \phi \) in figure 1(a).

The sensor can be modeled by a simple lumped-element mass-spring system, giving the following expression for the resonance frequency:
\[
f_o = \frac{1}{2\pi} \sqrt{\frac{K}{m_{\text{tube}} + m_{\text{fluid}}}} = \frac{1}{2\pi} \sqrt{\frac{K}{m_{\text{tube}} + \rho V_{\text{tube}}}}, \tag{3}
\]
where \( K \) is the effective modal spring constant of the structure, \( m_{\text{tube}} \) is the mass of the empty tube, \( m_{\text{fluid}} \) is the mass of the fluid inside the tube, \( \rho \) is the density of the fluid, \( V_{\text{tube}} \) is the volume of the tube. The effective modal spring constant \( K \) depends on the tube dimensions and material properties of the tube wall. It will also depend on the pressure inside the tube, as increasing pressure will deform the tube and increase the stiffness. This can be described by a pressure dependence coefficient \( \alpha \) as follows:
\[
K = K_0 + \alpha P, \tag{4}
\]
with \( K_0 \) the effective modal spring constant at atmospheric pressure and \( P \) the gauge pressure. Inserting (4) into (3) gives the following expression for the resonance frequency:
\[
f_o = \frac{1}{2\pi} \sqrt{\frac{K_0 + \alpha P}{m_{\text{tube}} + \rho V_{\text{tube}}}}. \tag{5}
\]

The same structure can be used as Coriolis mass flow sensor. If there is a mass flow through the tube this will result in Coriolis forces that will induce a swing mode vibration as shown in Figure 1(b) [8]. The amplitude of this vibration is proportional to the mass flow rate and can be detected by monitoring the motion of the midpoint of the tube indicated by point A in Figure 4.

The pressure drop (\( \Delta P \)) along the tube in case of laminar flow of an incompressible fluid inside the tube can be determined by the Hagen-Poiseuille equation:
\[
\Delta P = P_2 - P_1 = \frac{8\mu Q_m L_{\text{tube}}}{\rho \pi r^4}, \tag{6}
\]
where \( P_2 \) is the pressure at the inlet of the tube, \( P_1 \) is the pressure at the outlet of the tube, \( \mu \) is the dynamic viscosity of the fluid, and \( Q_m \) is the mass flow rate.

Because of the compressibility of gases, the density and flow velocity cannot be assumed constant and the model for pressure drop over the tube will be different than for incompressible liquids. The equation for the pressure drop for laminar compressible gas flow through a circular cross-section tube can be derived from the energy balance as follows [23]:
\[
\begin{align*}
\frac{P_2^2 - P_1^2}{2RT} &+ 2 \left( \frac{Q_m}{A_i} \right)^2 \ln \left( \frac{P_1}{P_2} \right) \\
&+ 2f \frac{L_{\text{tube}}}{r} \left( \frac{Q_m}{A_i} \right)^2 + \frac{\kappa Q_m^2}{2A_i^2} = 0, \tag{7}
\end{align*}
\]
where \( R \) is the specific gas constant, \( T \) is the temperature, \( A_i \) is the inner surface area of the tube, \( f \) is the friction factor (\( f \sim 8/Re \)), \( Re \) is the Reynolds number [23], and \( \kappa \) is defined as an additional loss coefficient which depends on the number and the shape of the bends, and changes in the diameter of the tube [24].

The loss coefficient for an increase in diameter of the tube, \( \kappa_{c\ell} \), is defined as [24]
\[
\kappa_{c\ell} = (1 - \frac{R_1^2}{R_2^2})^2, \tag{8}
\]
the loss coefficient for a decrease in the diameter of the tube, \( \kappa_{c\ell} \), is defined as [24]
\[
\kappa_{c\ell} = 0.42(1 - \frac{R_1^2}{R_2^2}), \tag{9}
\]
where \( R_1 \) and \( R_2 \) are the radius of the tube before and after the change in the diameter, respectively. The loss coefficient for a 90-degree turn is approximately 0.33 [24].

IV. EXPERIMENTAL SETUP

As mentioned before, in the first step of the fabrication process the ABS wire should have the desired shape. Figure 5 shows the mold that was used to bend the ABS wire as a U-shaped wire. The mold is a 12 mm×12 mm square that is made of aluminum. During bending, to prevent the breaking of the wire, the mold was placed in a beaker of water on a hotplate at 85 degrees.

Figure 6(a) shows a photograph of the nickel electroplated U-shaped tube which was soldered to a printed circuit board (PCB). The PCB is then mounted between a magnet holder, containing the three permanent magnets, and a PMMA block that provides the fluid inlet and outlet. Figure 6(b) shows a photograph of the complete assembled density and mass flow sensor.

Figure 7 shows a block schematic of the experimental setup. In this setup, a pressure controller (Bronkhorst EL-PRESS) sets the input pressure of a pressurized container.
Fig. 5. Picture of the mold for bending ABS.

Fig. 6. Photograph of (a) the nickel electroplated U shape tube, (b) the density/mass flow sensor.

Filled with liquid. A degasser (Biotech DEGASi CLASSIC) is used to remove air bubbles from the liquid and a filter with a porosity of 2 μm is used to provide a particle free flow. A voltage source is used to apply a periodic chirp or sinusoidal actuation signal with a resulting current amplitude of 20 mA. The motion of the tube is detected by a laser doppler vibrometer (LDV, Polytec MSA-600 microsystem analyzer). A differential pressure sensor (Sensortechnics PTE5005D1A) is used to measure the pressure drop over the tube. A mass flow controller (Bronkhorst mini CORI-FLOW ML120) is used to control the mass flow rate between 0 to 50 g/h. By placing the mass flow controller at the outlet of the setup, the pressure in the system can be controlled by the pressure controller for different flows.

V. RESULTS AND DISCUSSION

A. Density Measurements

The measured vibration spectrum of the air-filled U-shaped vibrating tube when actuated with a periodic chirp signal is shown in figure 8. The resonance frequencies and quality factors are 1375 Hz and 537 for the swing mode, and 3545 Hz and 502 for the twist mode, respectively.

Figure 9 shows the measured relation between the actuation resonance frequency versus actual density of helium, nitrogen, argon, water, IPA, and their mixtures at zero flow rate and ambient pressure. The actual densities of the different mixtures of liquids were measured by a pycnometer. The density of gases is according to Fluidat [25]. By substituting the measured resonance frequency in (5), the density measurement results have an accuracy of 0.5% for the liquids and 5% for the gases listed in figure 9.

Figure 10 shows the measured resonance frequencies for water, IPA, and their various mixtures as a function of pressure. The mass flow is set to zero in order to have the same pressure everywhere in the tube. As the liquid is incompressible, the slight increase in resonance frequency with pressure shows the influence of the pressure on the modal spring constant as expressed by the coefficient α in (4).

Figure 11 shows the measured resonance frequencies for the gases helium (He), argon (Ar), and nitrogen (N2) as a function of pressure. For these measurements the pressurized container and the degasser were removed from the setup in figure 7. As the gases are compressible, the pressure dependence is now due to a combination of two effects: an increase in pressure results in an increase in modal spring constant and an increase in density. We see that for relatively heavy gases like argon the increase in density is the dominant effect and the resonance frequency decreases with increasing input pressure. For a light gas like helium the change in spring constant has a stronger effect and the resonance frequency increases with pressure.

The measurement results from figure 10 and figure 11 were used to fit the parameters in (5) and the resulting values are listed in table 1. These values were subsequently used to include the error margins of ± 0.5% for liquids and ± 5% for gases in the figures.

B. Pressure Drop

Figure 12 shows the measured pressure drop over the tube when the input pressure was set to 4.5 bar for IPA and water, together with the expected pressure drop calculated from (6). The difference in the slope of these curves is equal to 2.4 which corresponds to the viscosity ratio between the two liquids.

Figure 13 indicates the measured and analytically calculated pressure drop over the tube as a function of flow rate for nitrogen, argon, and helium. Because of the compressibility of the gases, the increase in pressure drop is not linear.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$K_0$</td>
<td>$1.56 \times 10^4$ N/m</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>$4.45 \times 10^3$ N/m/Pa</td>
</tr>
<tr>
<td>$m_{\text{tube}}$</td>
<td>$3.11 \times 10^3$ kg</td>
</tr>
<tr>
<td>$V_{\text{tube}}$</td>
<td>$10.61 \times 10^2$ m$^3$</td>
</tr>
</tbody>
</table>
Fig. 8. Measured vibration spectrum of an air-filled tube detected by the Polytec MSA-600 laser doppler vibrometer (using a periodic chirp signal to excite the sensor).

Fig. 9. Actuation resonance frequency \( (f_0) \) vs density. Measured points from left to right: helium, nitrogen, argon, IPA, 90% IPA + 10% water, 75% IPA + 25% water, 50% IPA + 50% water, 25% IPA + 75% water, 10% IPA + 90% water, and water. The slope of the graph is \( 6.8 \times 10^{-13} \text{ m}^3/\text{Hz}^2/\text{kg} \).

Fig. 10. Measured resonance frequency \( (f_0) \) vs pressure. The grey area indicates the theoretical results according to (5) and ± 0.5% of the actual density of the liquids that were measured by a pycnometer.

Furthermore, a higher input pressure causes a lower pressure drop for the same mass flow rate, whereas, for liquids, the input pressure did not affect the shape or slope of the pressure drop. In figure 13 the analytical pressure drop over the tube is calculated according to (7). Besides the pressure drop over the electroplated tube itself, also the pressure drop over the tubing that connected the device to the differential pressure sensor as well as the additional losses caused by the changes in tube diameter were taken into account. The length of additional tubing at each side of the electroplated tube was 20 cm with a diameter of 762 micron. The additional losses due to the change in diameter were calculated according to (8) and (9). Still, the measured pressure drop is somewhat higher than
Fig. 13. The pressure drop over the tube versus flowrate for Nitrogen, Argon, and Helium, at input pressure of 5 and 6 bar. The analytical pressure drop for the tube is calculated with (7).

Fig. 14. Schematic representation of the lock-in amplifier which is connected to the output of the LDV. Current actuation is connected to the mixer as a reference signal.

Fig. 15. The voltage in the center of the tube (point A in figure 4) vs mass flow rate for water and IPA.

C. Mass Flow Measurements

By connecting a lock-in amplifier to improve the output signal of the LDV, the same structure and setup can be used for mass flow measurement. Figure 14 shows the schematic of the lock-in amplifier that was connected to the output of the LDV. Figure 15 shows the measured output voltage, which is proportional to the motion of the midpoint of the tube (point A in figure 4), as a function of mass flow rate of water and IPA, respectively. As the Coriolis forces are proportional to the mass flow, independent of the type of fluid, the measurements for water and IPA should be identical. However, an important requirement is that the actuation amplitude, i.e., the amplitude of the twist mode vibration, is the same for both liquids. Unfortunately, the current setup does not allow accurate control of the actuation amplitude, especially when the resonance frequency of the tube changes due to a change in density of the liquid or a change in pressure. Future work will first focus on an integrated readout that can also monitor the actuation amplitude. This will allow the implementation of a control system to always actuate the sensor at resonance and with constant amplitude. Besides this many other factors may affect the sensitivity and also packaging aspects need to be considered [26].

VI. CONCLUSION

In this paper, electroplating is proposed as a novel fabrication method for thin-walled cylindrical microtubes with a near-perfect circular cross-section. A wide range of diameters, from 120 μm to 1 mm, and wall thicknesses, from 8 μm to 60 μm, have been achieved. A demonstrator density and mass flow sensor was designed and fabricated based on a tube with 600 μm diameter and 20 μm wall thickness. The tube is actuated by Lorentz forces and readout is done by a laser doppler vibrometer. This sensor resulted an accuracy of 0.5% for the density of liquids such as DI water, IPA, and their mixtures, and an accuracy of 5% for the density of gases like nitrogen, argon, and helium. Initial mass flow measurements show that the device also works as Coriolis mass flow sensor, however an improved readout is needed to allow control of the actuation amplitude.

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REFERENCES

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