DETERMINATION OF THE THERMAL CONDUCTIVITY OF GASES UNDER FLOW CONDITIONS

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ABSTRACT

This contribution presents a calorimetric flow sensor that is capable of determining the thermal conductivity (k) regardless of the flow conditions. The sensor can also measure the flow rate (Q) provided that the volumetric heat capacity (ρc_p) or the thermal diffusivity ($\alpha = k/(\rho c_p)$) of the flowing gas is known. No additional sensors or bypass channels are needed. The measuring principle relies on using high frequency heat generation (200 Hz). The thermal conductivity of several common gases is measured.

KEYWORDS

Flow sensor; Thermal flow sensor; Thermal conductivity; Oscillatory excitation

INTRODUCTION

Although the first micro-machined flow sensors were fabricated over 40 years ago [1], active research on this topic still takes place. The lack of moving parts is an advantage for applications, where long-term measurement and reliability are important constrains. Another advantage is the simple electronic instrumentation such sensors require. Possible applications include monitoring of gas, environmental monitoring, gas chromatography, etc. [2].

According to their measuring principle, thermal flow sensors are classified in the following types: anemometric, time-of-flight [3], and calorimetric [4]. Typical anemometric sensors feature at least a single heater that also serves as temperature sensor. They monitor the heat transfer from the heater to the fluid, which increases with the flow speed. An additional temperature sensor allows compensating temperature changes in the fluid. Time-of-flight sensors measure the time it takes for a heat pulse created by a heater to reach a temperature sensor at some location further downstream. Calorimetric flow sensors usually feature a heater and two temperature sensors located up- and downstream from the heater. The temperature profile in the vicinity of the heater changes with flow. The temperature difference between the up- and downstream locations is therefore related to the velocity. This relation holds linear for a bounded range of flow rate. Usually, calorimetric flow sensors are operated in constant power mode; that is, constant heat generation at the heater.

Regardless of the measurement principle, the readout parameters not only depend on the flow rate but also on the thermal properties of the fluid, namely thermal conductivity (k), specific heat capacity (c_p) and density (ρ). Depending on the geometry of the flow sensor and the flow speed, the viscosity (μ) might also exert an influence.

In consequence, thermal sensors still require calibration procedures whenever the sample fluid is changed; or temperature or pressure changes modify the thermophysical properties of the fluid.

To obtain a correct flow measurement, it is necessary to determine the fluid properties while the fluid is moving. Most efforts that address this problem involve using oscillatory heat generation at the heater. The 3-omega method has been widely used to determine k and α of liquids and gases [5]. However, this method has been proven to work well for stagnant fluids or solid materials.

Other researchers have also successfully used oscillatory (AC) thermal excitation to simultaneously determine k and α of stagnant or moving liquids [6, 7]. The main drawback is that the flow speed must be known beforehand. Their method consists in fitting the amplitude and phase of the thermal oscillations to the theoretical frequency response. For this, the sensor geometry has to be geometrically simple to allow easy analytical modeling.

Some of the most recently proposed approaches feature a bypass, through which a small quantity of fluid flows at very low speeds [8, 9]. Inside this bypass, an additional sensor structure determines the thermal properties of the fluid assuming that the flow velocity is almost zero. This extra structure increases the complexity and cost of the finalized sensor.

In this contribution, we present a simple calorimetric flow sensor that is capable of measuring the thermal conductivity of gases under flow conditions without prior knowledge of the flow rate. The measuring principle relies on sinusoidal heat generation at a fixed frequency. The heater-thermistor pair located at the midpoint of the sensor serves as thermal conductivity sensor. Simultaneously, constant heat generation can be used to determine the flow rate provided that the thermal diffusivity of the gas is known.

DESIGN

The sensor is a silicon dice of 1270 × 1270 × 525 μm^3 that holds a SiO_2-SiN_x, 1-mm-diameter membrane. The typical thickness of the membrane is 1.4 µm.

Figure 1 shows a close-up view of the membrane, whereupon a circular chromium heater and five germanium thermistors are situated.

The heater consists of four different arcs that surround the central thermistor (CT). The gaps between the heater arcs allow the interconnection of the central thermistor. The average radius of the arcs is 78.5 µm. They are interconnected in series outside the chip. Chromium was selected as heating material because of its low temperature coefficient of resistance (TCR).

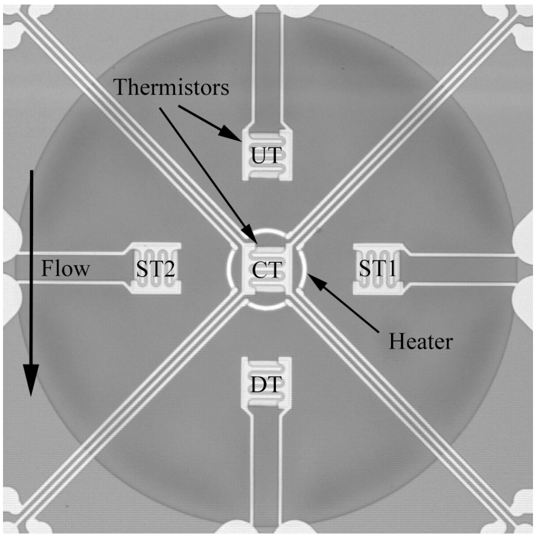

Figure 1: Close view of the sensor's membrane featuring the heater and the temperature sensors. The labels UT, DT, ST1 and ST2 respectively represent the up-, down- and side thermistors.

The central thermistor is intended to provide a closer estimation of the heater's temperature; therefore the average separation between the heater and the central thermistor is just about 13.7 µm. This small separation also allows the determination of the thermal conductivity as shown later on this paper.

The remaining four thermistors rest concentrically around the central thermistor-heater structure. All thermistors are 105 × 105 µm^2 in size. Their midpoint is 231.5 µm away from the center of the membrane. In this work, the gas is always assumed to be flowing along one of the major axes; thermistors are therefore labeled according to the flow direction, as shown in Figure 1. Further details about the fabrication process are described in previous contributions [4, 10].

EXPERIMENTAL

The sensor dice is glued onto a PCB, which serves as electrical interface to the instrumentation circuit. Before gluing, a small window is drilled in the PBC to allow direct access to the backside of the sensor's membrane.

The sensor is then placed at the bottom of a small, PMMA channel with a high-aspect-ratio cross sectional area. Its height (H) and width (W) are 600 µm and 14 mm, respectively. Gases flow into the measurement channel from a larger channel (5 × 14 mm^2), which not only feeds the measurement channel but also a small closed-end channel below the sensor. This small conduit allows the measured gas to statically occupy the volume below the membrane. This ensures that the same gas occupies the volume above and below the membrane. However, below the membrane, the sensor is stagnant.

An analog mass flow controller (Brooks 5850E) regulates the flow rate into the channel from a gas bottle. The flow range spans from 20 to 1000 sccm nitrogen equivalent. The average flow speed in the channel ranges then roughly from 0.04 to 2m/s. This range might change according to the volumetric heat capacity of the gas.

Table 1: Thermal properties of the gases used for the measurements at 25°C and 1 bar [11, 12].

Gas	k (mW m^{-1} K^{-1})	ρc_p (kJ m^{-3} K^{-1})	α (cm^2 s^{-1})
CO_2	16.79	1.511	0.111
Ar	17.84	0.836	0.213
N_2	25.97	1.169	0.222
Air	26.38	1.169	0.226
Ne	49.77	0.833	0.597
He	156.0	0.833	1.873

Table 1 presents the gases used in the experiments and their relevant thermal properties. The outlet of the channel leads to open space. Therefore, the temperature and the pressure are set to room conditions (25°C, 1 bar).

The distance from the sensor membrane to the inlet of the measurement channel is long enough to guarantee a fully developed parabolic flow profile over the sensing surface as follows [13]:

$$u(y) = \frac{6\overline{u}}{H^2} y(y - H), \quad (1)$$

where ū is the average velocity in the channel. As a result, the viscosity of the fluid has no effect on the flow profile under laminar regime, and in turn, the viscosity has not effect on the heat exchange between the heater and the fluid. Nevertheless, the pressure drop along the sensor does depend on the viscosity. A thermal boundary layer develops under flow condition from the heater downstream.

A steady sinusoidal voltage signal is applied to the heater. Since chromium's TCR is small (~10^{-4} K^{-1} [14]), the resulting average heat generation and the amplitude of the heat oscillations are constant. They are set both to be 0.5 mW. The frequency of the heat generation, and the resulting temperature oscillations, is kept constant at 200 Hz.

The small heat generation power ensures small overtemperatures in the vicinity of the heater. In consequence, in this contribution, the thermophysical properties of the measured gas are assumed to be constant across the channel. Moreover, the natural convection processes are also ruled out [15], neglecting any effect on the flow profile.

Before the experiments take place, germanium thermistors in the sensor are calibrated in an oven for a temperature range from 26 to 34 °C.

Each thermistor has a polarization current of just about 1 µA, hence the self-heating effect is negligible. Current-to-voltage converters measure the resistance of the thermistors. A 24-bit acquisition card (NI-9239) digitalizes the resulting voltage signals. The heater excitation and the recording of the output signals are controlled with a LabVIEW program, which converts the voltage signals into temperature values using the calibration curves. This program also implements a lock-in amplifier, which calculates the amplitude, phase shift and mean value of the temperatures measured by the thermistors. The reference frequency is also 200Hz.

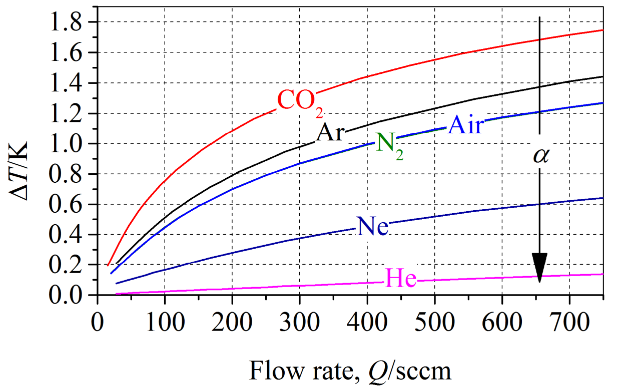

Figure 2: Difference in the DC temperature from the upstream (UT) to the downstream (DT) thermistor vs. the flow rate for various gases.

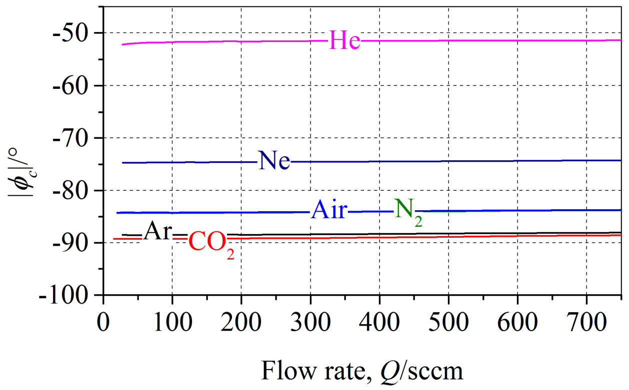

Figure 3: Phase shift measured by the central thermistor vs. the flow rate at 200 Hz for various gases.

RESULTS AND DISCUSSION

The suitability of the presented device as a flow sensor was already proven for air and nitrogen in prior work [4]. For calorimetric flow sensors, the main flow sensing parameter is the average (DC) temperature difference between the up- and the downstream sensors ($\Delta T = T_{DT} - T_{UT}$). The average (DC) power generated by the heater directly affects ΔT; on the contrary, the oscillatory component of the heat generation has no influence on ΔT as long as the thermal properties of the fluid remain homogeneous. This is guaranteed by the small power delivered to the heater.

Figure 2 shows ΔT for different flowing gases in relation to the flow rate (Q). These curves match the typical characteristic of a calorimetric flow sensor. Each one features two distinct regions. First, for low flow rates, ΔT is proportional to the flow rate. Second, for moderate large flow rates, ΔT is proportional to Q^p, where $0 < p < 1$. In both cases, the proportionality constant (sensitivity) decreases as the thermal diffusivity of the gas increases (see Table 1). Moreover, the transition point between the two regions also increases with α [16]. The transition occurs around 70 sccm for CO_2, whereas for He the relation is linear for the whole range shown in Figure 2.

As most of the state-of-the-art works [6-8] suggest, it is necessary to know the flow velocity in order to determine the thermal properties of the fluid. The natural solution is thus to stop the fluid motion or to bring the heat transfer process into a region where the fluid is basically motionless. Inside a flow channel, the flow speed is in theory zero at the channel walls (see Eq. (1)) regardless of the flow speed up to a certain extend. Thus most of the heat transfer has to be constrained to a close vicinity of the membrane to minimize the effect the flow velocity has on fluid properties determination.

Increasing the frequency of the heat generation reduces the penetration depth of the temperature oscillations into the fluid. This effect comes as a result of the heat capacity of the fluid. It can be shown for the one-dimensional case that the penetration depth (δ_p) depends on α and the frequency (f) as follows:

$$\delta_p = \sqrt{\frac{\alpha}{\pi f}} \,. \tag{2}$$

Thus, by placing the heater on the channel wall (membrane) and increasing the frequency, most of the heat transfer is then bounded to the membrane and a thin layer of fluid in the vicinity of the heater. Here, the central thermistor permits estimating not only the temperature of the heater in DC but also, due to its proximity to the heater (~13.7 µm), the thermal properties of the fluid.

Figure 3 presents the phase shift (φ_{CT}) of temperature oscillations at 200 Hz measured by the central thermistor as function of Q. This phase shift is referenced to the heat generation signal. As expected, φ_{CT} remains almost constant up to 750 sccm. Moreover, the maximum increase among all gases is 1.2° (helium), which means a 0.66% change for a 180° full-scale. The amplitude of the temperature oscillations measured by the central thermistor ($|T_{CT}|$) also shows a similar behavior. Nevertheless, $|T_{CT}|$ seems to be more affected by the flow rate as the maximum percentage change is almost as twice as large (1.2% for He).

It is important to note that φ_{CT} in Figure 3 is distinct for each gas, with the exception of air and N_2, which possess similar thermal properties. In consequence, the phase shift at high frequency is a suitable measurement parameter to determine the thermal properties of the flowing gas. Correlating φ_{CT} at low flow rate to each thermal property shows that φ_{CT} seems to be almost entirely dependent on the thermal conductivity.

The relation between k and φ_{CT} is found to be best fitted by

$$k = A\exp(B\varphi_{CT}) + C \,, \tag{3}$$

where A, B and C are real numbers. The resulting fitting curve and parameters are shown in Figure 4. Although R^2 is quite close to 1, measurements with more gases and gas mixtures are necessary to confirm the relation in Eq. (3).

The magnitude of the phase shift increases with the thermal conductivity of the gas. Since $C < 0$, $|\varphi_{CT}|$ has a maximum value. According to Eq. (3), $|\varphi_{CT}|_{max} = 101.5°$, which corresponds to vacuum conditions. $|\varphi_{CT}|_{max}$ is then defined by the thermal conductivity (k_m) and volumetric heat capacity ($\rho_m c_{pm}$) of the membrane.

On the other hand, Equation (3) also suggests that for $k > 1238.8$ mW·m^{-1}K^{-1}, the phase shift $\varphi_{CT} > 0$. Given the nature of thermal transfer, φ_{CT} must be negative. In fact,

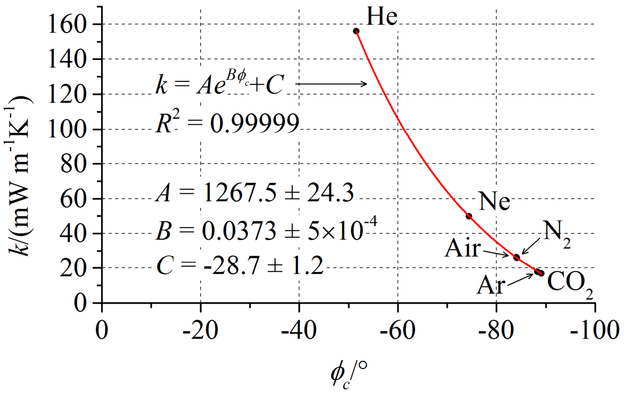

Figure 4: Thermal conductivity (k) vs. central phase shift (φ_{CT}) at 200 Hz. The fitting curve is shown in red.

φ_{CT} must approach asymptotically to zero as k becomes very large. As a consequence, Eq. (3) is not expected to be accurate for fluids with larger thermal conductivity, e.g., liquids.

Since the volumetric heat capacity of the membrane (2.46 MJ·m^{-3}K^{-1}) is over 1000-fold larger than that of any measured gas and δ_p is too small at 200 Hz, the thermal capacity of the membrane becomes too dominant. Therefore, the heat capacity of the gas has a very limited influence on $|T_{CT}|$ and φ_{CT} at high frequencies. The use of lower frequencies might allow the determination of α [7]. The main drawback is that the flow rate would also affect the read out signal.

CONCLUSION

We present a simple calorimetric flow sensor that is able to measure the thermal conductivity of common gases under flow conditions. It operates without bypass or additional sensor structures.

Although the thermal diffusivity cannot yet be measured under flow conditions, this sensor opens the possibility of fluid independent flow measurement for gases with similar molecular structure, i.e., gases with similar volumetric heat capacity. For gases with distinct volumetric heat capacity, determination of their thermal diffusivity is still a required step for autocorrection procedures.

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