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Novel Miniaturized Thermoelectric Hydrogen Pressure Sensor

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Summary:

In conventional hydrogen pressure sensors, the stress-sensitive diaphragm is the weakest point in terms of construction. Leading to signal drift of the sensor as hydrogen causes material embrittlement and permeation. We address this issue by using a miniaturized gas pressure sensor without a mechanical transducer. Our sensor structure consists of a heating element and thermopiles arranged in a thin perforated silicon nitride membrane. In the thermal domain, the structure exhibits a characteristic low-pass behavior whose phase shift is dependent on the hydrogen pressure. A dynamically excited heating element and the evaluation of the thermopile responses are used to measure pressures ranging from 100 and 2850 kPa. So far, this is the highest measurement range detected with a thermal pressure sensor that has ever been published in literature. Compared to diaphragm-based MEMS pressure sensors, this chip design offers an intrinsic overload protection as well as low-cost fabrication and simple packaging requirements.

Keywords: thermal pressure sensor, pressure-dependent gas properties, heat transfer, MEMS, thermoelectric transducer

Introduction and Motivation

Sustainably and economically produced hydrogen is a key factor in reducing greenhouse gas emissions in the energy, transportation and industrial sectors [1]. As it has an enormous potential as an energy carrier. The easiest way to store and transport large quantities of hydrogen is to supply it in the form of a high-pressure gas [2] which requires precise and long-term stable sensors for monitoring and controlling static and dynamic hydrogen pressures.. Conventional absolute MEMS pressure sensors consist of a vacuum reference cavity and a mechanical diaphragm whose deformation is sensed capacitively or piezoresistively. This design demands complex chip fabrication and costly packaging technologies to avoid signal hysteresis and parasitic stress generation during operation [3], [4]. Additionally, if these components are exposed to hydrogen, it can be observed that hydrogen diffuses into the cavity, which is progressively manifested in a signal drift of the device [5]. We aim to solve these well-known challenges by eliminating the pressure-sensitive diaphragm and applying a thermal measurement principle that is capable of detecting pressure-dependent gas properties. A traditional micro- Pirani sensor measures the thermal conductivity of a gas and converts a pressure variation into a change in the resistance of a heated structure [6], [7]. Unfortunately, this method can only be employed in specific pressure ranges, usually significantly below atmospheric pressures [6], [8]. However decreasing the gap between the heating element and a heat sink in the sub-micrometer range allows the operating range to be shifted to higher pressures. In the past, pressures above 100 kPa have already been detected with nano-gap Pirani gauges [9]. Nevertheless, operation ranges well above 1000 kPa cannot be addressed with that static thermal principle due to signal saturation [6].

In this work, the measuring range is extended using a dynamic measuring method. This allows the detection of pressure-dependent properties (e.g. thermal diffusivity or density) that do not exhibit a saturation behavior. For the first time in literature, we present gas pressure measurements with a thermal principle that far exceed 1000 kPa. In the paper, a thermoelectric pressure sensor is analyzed with gaseous hydrogen in order to gain a better understanding of the pressure-dependent heat transport phenomena and to provide the basis for future structural optimization of the sensor.

Sensor Theory

The measuring principle of the thermoelectric pressure sensor is based on the measurement of dynamic heat propagation. This method has already been used in the past for the thermal characterization of thin films [10] and for the realization of wide-range temperature sensors [11]. In order to qualitatively understand the behavior of the sensor, an analytical model for describing the temperature propagation is derived. In our device, the heating power is created locally by an electrical excitation of the heating element. Thermopiles at a distance *d* then detect the temperature fluctuations. In our model, we approximate the heater as a point source in an infinite homogeneous heat-conducting medium leading to a one-dimensional differential equation in *x*-direction:

$$\rho(p) \cdot c_p \frac{\partial T}{\partial t} = k \frac{\partial^2 T}{\partial x^2} \tag{1}$$

The propagation of the temperature is determined by density ρ, thermal conductivity k and specific heat capacity c_p of the surrounding medium. The solution to Equation 1 for a sinusoidal heat generation is an oscillating thermal wave with a certain temperature amplitude that reaches the temperature detectors with a specific time difference.

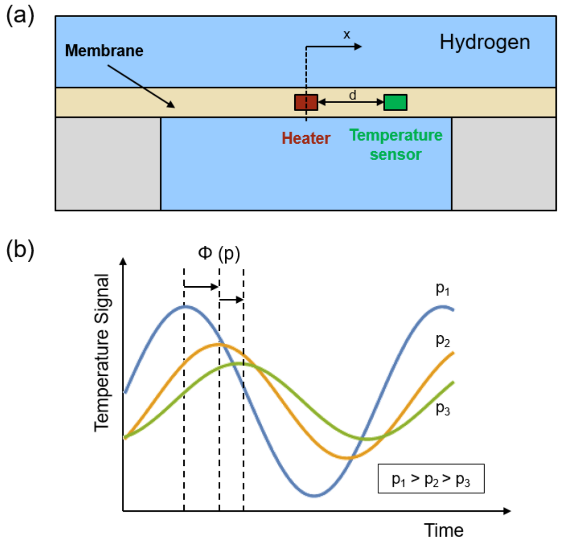

Fig. 1. (a): Schematic cross section of the sensor with a spatially separated heating element and sensor embedded in thin membrane without perforations. (b): Temperature signal for different gas pressures. As the pressure increases, the attenuation during the propagation of the temperature wave intensifies and influences the phase lag.

In this work, the phase shift Θ between electrical excitation and arrival of the temperature signal at the temperature sensors is evaluated in order to monitor hydrogen pressure. This phase lag depends on the thermo-physical properties, the excitation frequency ω and the distance d between the heating element and the temperature sensor. All the thermal properties of our device can be summarized in a single parameter called thermal diffusivity α.

$$\theta = -\sqrt{\frac{\omega \cdot \rho(p) \cdot c_p}{2k}} \cdot d = -\sqrt{\frac{\omega}{2\alpha}} \cdot d \sim -\sqrt{\omega \cdot p} \tag{2}$$

As the active area of our device is located on a thin membrane, not only the thermo-physical properties of the gas but also the parasitic heat transport through the membrane are measured. However, as a first approximation, it can be assumed that the pressure p only affects the gas density, while the other thermal properties are pressure-independent.

Device Fabrication and Measurement Setup

Micromachining allows the production of small and thermally isolated areas that form the active area of our MEMS sensor with a heating element and the thermopiles (TP). The process flow of the novel pressure sensor is based on the production of thermal flow sensors.

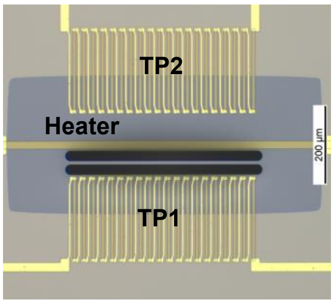

Fig. 2. Top view of the sensor with a DRIE-etched membrane. Both TPs are arranged at a distance of 70 µm from the heater. Between TP1 and the heater, slits in the membrane are introduced in order to improve the thermal interaction with hydrogen gas.

The process starts with the growth of a thermal oxide and the deposition of a stoichiometric LPCVD silicon nitride. After that, n-doped polysilicon is deposited and structured to realize heater and thermopiles. Then an aluminum layer is deposited to implement connection lines, bond pads and thermopiles. Afterwards, the structures are passivated with a PECVD silicon nitride, slits are etched from the front side and the membrane is exposed from the back using a dry etching process (Fig. 2). For the characterization, the silicon chip is mounted on a PCB, contacted with wire bonds and then clamped in a test fixture. In addition to the membrane perforation, the chip is ventilated via the PCB. This guarantees that no pressure gradients occur between the front and back cavity, so that the membrane remains mechanically in a neutral position. The heating element of the MEMS chip is excited with a lock-in amplifier from Zurich Instruments with a sinusoidal voltage. The thermopiles are evaluated at twice the excitation frequency. Hydrogen pressure variations in the measuring chamber are generated by volume compression with a syringe pump. During the measurement, a reference sensor (Keller PAA-33X) monitors the pressure inside the measuring chamber.

Results

In this section, the thermal pressure sensing behavior is evaluated based on the phase signal and qualitatively compared with the analytical dependence derived in equation 2.

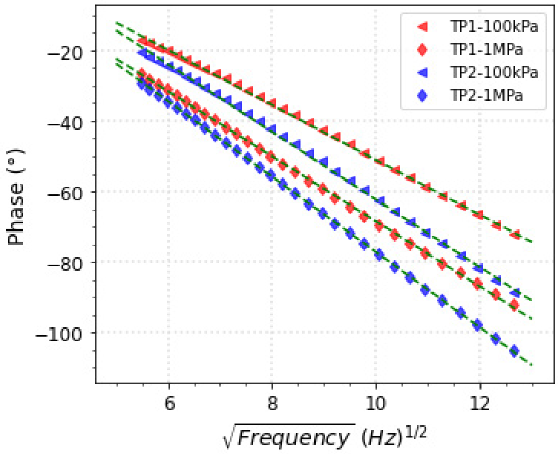

Fig. 3. Measured phase shift for both TP and two hydrogen pressures in relation to the excitation frequency. The dashed lines show the linear fit based on equation 2.

First, we examine the phase signal as a function of the excitation frequency. We consider the frequency range from 30 to 160 Hz. At lower frequencies, the thermal penetration depth is greater than the distance to the heat sink. However, the model is only valid as long as there is an infinitely large distance to the heat sink, hence larger excitation frequencies are required. Measurement have been performed at hydrogen pressures of 100 kPa and 1 MPa (Fig. 3). The phase shift increases with increasing excitation frequency, indicating a characteristic low-pass behavior, which is caused by the thermal inertia of the membrane and the surrounding gas. As the frequency increases, the detected phase lag at the TPs grows. For both TPs, an increase in pressure leads to an increase in the gradient of the characteristic curve. For a constant pressure, the phase shift over the entire frequency range is smaller for TP1 than for TP2. A comparison of the slopes of the measurement curves also reveals that the phase sensitivity of TP2 is greater for the evaluated pressures. These phenomena are caused by different thermal diffusivities in the heat transfer path. In the region of the slits, the membrane is replaced by a medium with deviating thermal properties. As hydrogen at room temperature has a higher thermal diffusivity than the membrane material at 100 kPa and 1000 kPa, the slope as well as the absolute phase value of TP1 is slightly lower.

An excitation frequency of 80 Hz is selected to investigate the pressure dependence. At this frequency, the signal-to-noise ratio of our measurement setup is at its highest. With increasing pressure, the phase shift between excitation and arrival of the temperature signal at the thermopiles increases. While the thermal diffusivity of the membrane is independent of the gas pressure, the thermal diffusivity of hydrogen decreases with increasing pressure. In the pressure range above 400 kPa, the phase signal of both TPs follows the analytical model from equation 2. In this range, the gas is so dense that the parasitic thermal effects caused by the membrane can be mostly neglected and the heat transport through the surrounding gas is dominant. Nevertheless, the sensitivity of TP1 to pressure changes is somewhat higher, as the gas influence on the heat transfer is stronger due to the membrane perforation.

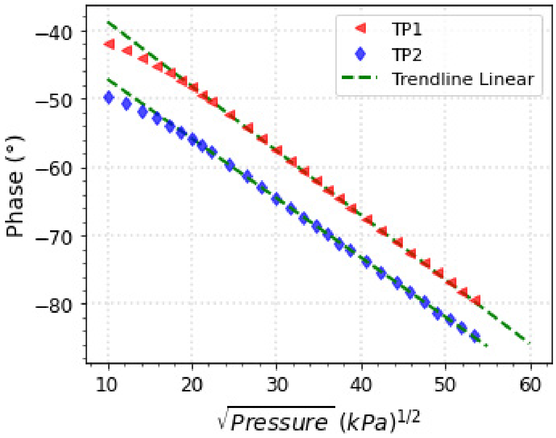

Fig. 4. Measured phase shift for both TPs when the heating element is excited at 80 Hz as a function of the hydrogen pressure. The dashed lines show the linear fit based on equation 2.

Conclusion and Outlook

In this paper, a miniaturized thermoelectric sensor for measuring hydrogen pressure was presented, which is highly sensitive in a wide pressure range due to the dynamic measuring principle. A model was introduced and qualitatively validated with experimental data. We were able to prove that the measurement signal does not saturate even for pressures around 3000 kPa, which is currently the limit of our experimental set-up. Compared to conventional stress-sensitive pressure sensors, we presume that the packaging has less influence on the long-term stability, as a pressure-dependent gas property can be measured directly. Our thermal operation principle can also be easily transferred to other gases like CO_2 or N_2 as well as defined gas mixtures. The knowledge of the pressure-dependent heat propagation within the investigated MEMS device forms the basis for further geometric optimizations. The aim is to improve the sensitivity and measuring range of our thermoelectric pressure sensor. In the future, novel sensor geometries will be investigated and measurements will be carried out at even wider pressure ranges. Additionally temperature compensation algorithms are to be developed to ensure accurate pressure sensing since the evaluated gas properties are temperature dependent.

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