A PHOTONIC MICROSYSTEM FOR HYDROCARBON GAS ANALYSIS BY MID-INFRARED ABSORPTION SPECTROSCOPY

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ABSTRACT

This paper demonstrates the functional integration of a linear variable optical filter (LVOF) and a gas cell at the wafer level, i.e. a gas-filled LVOF, where the resonator cavity of the filter is also used for storing the gas sample. A mm-level effective optical absorption path length is achieved from a µm-level physical path length, by exploiting multiple reflections from highly reflective Bragg mirrors and the high-order operation of the filter. The wideband spectral response of the device is ensured by using a tapered cavity, where the cavity length changes linearly along the length of the filter. Therefore, combined with a detector array and a light source, the gas-filled LVOF enables wideband operation and long absorption path in a single MEMS device, thus ensuring a highly sensitive on-chip gas absorption microspectrometer.

INTRODUCTION

Natural gas is the fastest growing fossil fuel in use due to abundant resources and robust production [1]. The expanding energy trade leads to mixing imported natural gas of diverse origin to meet the consumer demands. This gives rise to an unpredictable composition of natural gas for both industrial and residential applications, thereby degrading the combustion efficiency. Therefore, the constituents of natural gas must be regularly monitored to optimize the burning process.

Measuring the composition of natural gas in such a large scale application requires an autonomous sensor network that can communicate with the outside world. The high volume imposed by the application calls for bulk-micromachined miniaturized devices that do not require sample handling. Silicon sensors are highly suitable for miniaturization and integration with the circuits for signal processing and communication [2].

The main components of natural gas, i.e. hydrocarbons have very similar molecular structures. As a result, a highly selective technique is required to distinguish these gas molecules. Gas chromatography is a highly selective method that separates the sample gas into its constituents based on the chemical affinity of each component [3]. However, the need for carrier gas supply and sample handling renders this technique ineligible for this application. In contrast, calorimetric sensors offer miniaturized low-cost solution to gas sensing [4]. However, the selectivity of these sensors is too limited for measuring the complex composition of natural gas.

Optical absorption spectroscopy is a commonly used self-referencing and nondestructive analysis method that provides comparable selectivity and high-level of miniaturization in gas sensing. In this method, the spectral distribution of the light that is absorbed by the sample is analyzed and compared to a material database for identification [5]. Hydrocarbons have distinctive absorption features in the mid-IR as shown in Figure 1. Therefore, a bulk-micromachined optical absorption microspectrometer in the mid-IR with integrated electronics would be suitable to measure the composition of natural gas in a large sensor network.

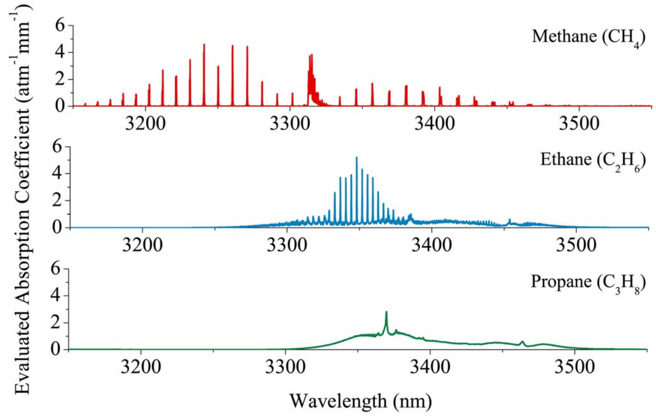

Figure 1: The evaluated absorption coefficient spectra of methane, ethane and propane per unit pressure and optical absorption path extracted from Pacific Northwest National Laboratory database at 0.112 cm^{-1} resolution [6].

The sensitivity of an absorption spectrometer is strongly dependent on the interaction between the light beam and the gas sample. Increasing the optical absorption path length allows the gas sample to absorb more light and thereby, improves the sensitivity of the spectrometer. The key challenge in the miniaturization of spectrometers is maintaining a sufficiently long absorption path in an on-chip implementation. In this paper, we present an interference filter based microspectrometer with a functionally integrated gas cell. The high-finesse resonator cavity of the optical filter is used also as a gas cell and the effective optical absorption path length is elongated beyond the physical dimensions of the device.

GAS-FILLED LVOF

A linear variable optical filter (LVOF) is a wideband interference filter that is composed of a flat and a tilted mirror with a tapered resonator in-between. Multiple reflections between these mirrors create a phase difference between the waves based on the resonator thickness. When this phase difference meets the constructive interference criterion for a specific wavelength, the filter transmits this particular spectral component of the input light. Therefore, if the LVOF is combined with a detector array, every pixel in the detector would measure a different narrow band in the spectrum depending on the resonator thickness at that particular position.

The gas-filled LVOF is a specialized implementation, where the wideband optical filter and the gas cell are functionally integrated in a single device as shown in Figure 2. In the gas-filled LVOF, a tapered resonator cavity is formed instead of a dielectric resonator layer by combining a flat and a tapered mirror. The sample gas to be measured flows through the cavity and the interaction of the light and the sample gas is enhanced through multiple reflections. Therefore, wavelength selection and optical absorption are achieved simultaneously with a sensitivity that exceeds the dimensions of the device.

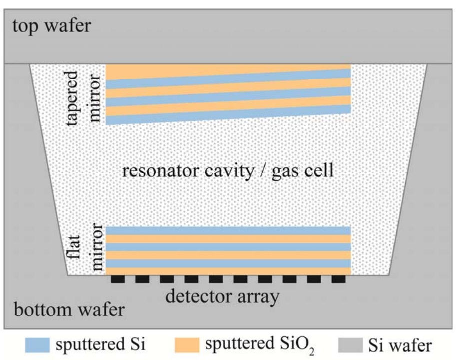

Figure 2: Structure of the gas-filled LVOF. The tapered resonator cavity acts also as a gas cell, where wavelength selection and sample absorption occur simultaneously.

Optical Design

Achieving an effective optical path length that is comparable to that of an external gas cell requires the use of a relatively long resonator cavity in addition to highly reflective mirrors. This forces the filter to operate at a high order and limits the free spectral range (FSR) of the device. Therefore, the gas-filled LVOF is designed at the 15th order in the 3.2 μm to 3.4 μm wavelength range with a 200 nm FSR, corresponding to a 14 mm long filter with a cavity length ranging from 24 μm to 25.5 μm.

The spectral response of an LVOF is conventionally approximated to an array of Fabry-Perot (FP) filters with flat resonators. The wideband transmission behavior is extracted by varying the resonator thickness for a specific pixel width based on the taper angle. However, the high-order operation and the use of highly reflective mirrors render this approximation inadequate for the gas-filled LVOF. Instead, the Fizeau interferometer model is employed, where the varying trajectory of the light beam due to the taper angle of the resonator is taken into account [7, 8].

Gold and aluminum are commonly used as reflective coatings in the mid-IR. However, since these are also highly absorbing materials, they cannot be used in transmission filters. On the other hand, Bragg mirrors that are composed of alternating layers of thin-films with quarter-wavelength optical thickness can be designed as both highly reflective and nonabsorptive. High reflectivity is assured by selecting thin-films with a high refractive index contrast and increasing the number of layers, while absorption is minimized through the use of materials with negligible extinction coefficient in the desired wavelength range. For this application Si (n_{Si} =3.61) and SiO_2 (n_{SiO_2} =1.45) are selected as nonabsorptive thin-film materials.

The transmission response is calculated by scanning the filter along its length. At every position, the interference of multiply reflected beams is calculated using the combined reflectivity of the mirrors and the cavity length at that particular position. Since the tapered mirror is formed by tapering the first SiO_2 layer, the combined reflectivity of the system becomes position dependent as shown in Figure 3.

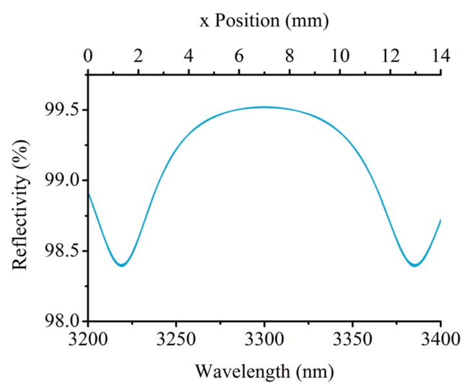

Figure 3: The combined reflectivity of the tapered and the flat Bragg mirrors calculated taking into account the incoherence of the thick Si wafer at normal incidence.

The transmission curves for eleven different wavelengths are calculated as shown in Figure 4. The effect of methane is included in the numerical analysis by inserting an exponential attenuation factor at every reflection. The accumulated effect of absorption is represented by a decrease in the transmittance. The varying peak transmittance of the curves without methane is mainly due to the varying mirror reflectivity, while the peak transmittance of the curves with methane is an indication of the methane absorption at that particular wavelength.

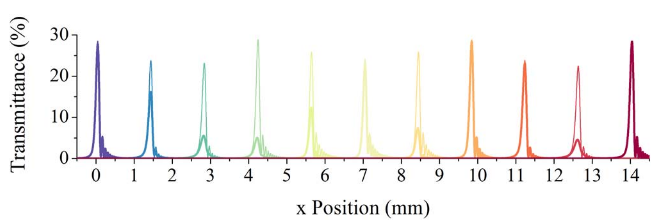

Figure 4: The transmission response in the 3.2 μm to 3.4 μm wavelength range of the LVOF with methane (thick

lines) and without methane (thin lines) at normal incidence. The x-position represents the position along the length of the filter.

Fabrication

The gas-filled LVOF is fabricated by integrating the flat and the tapered mirror through direct wafer bonding [9]. The flat mirror layers are sputtered in a KOH etched cavity. The tapered mirror on the other hand is formed on the surface of a wafer. To fabricate the tapered SiO_2 layer, first a flat layer of SiO_2 is deposited, which is then covered with a thick photoresist. Then, the photoresist is patterned such that there are linearly variable distanced trenches along the length of the filter [10]. Later, the patterned photoresist is converted into a tapered structure by a thermal-chemical treatment with a solvent. Finally, the tapered photoresist profile is transferred onto the flat SiO_2 layer underneath via one-to-one plasma etching. Using the tapered SiO_2 layer as the basis, the rest of the thin-film layers are sputtered to finalize the fabrication of the tapered mirror. The surface profile of a fabricated tapered mirror is shown in Figure 5. The cavity length is that is defined by the level difference between the ends of the mirror is extended to approximately 2 µm to account for the fabrication errors. Consequently, gas inlet and outlet are formed on both wafers by DRIE to allow for the controlled gas flow and the wafers are bonded.

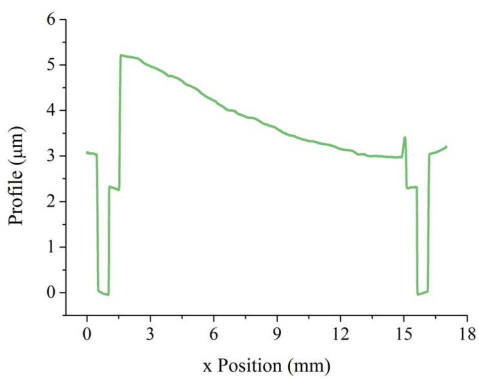

Figure 5: The raw profile of the tapered Bragg mirror measured before wafer bonding.

To analyze the effect of the path length elongation in the gas-filled LVOF, the sample gas must remain only in the cavity during the measurements. Therefore, dispensing tips are connected to the inlet and outlet of the device as shown in Figure 6 .

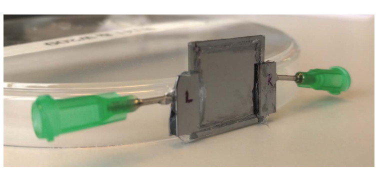

Figure 6: Packaged device.

Characterization

The gas-filled LVOF is to be integrated to a micromachined detector array and wideband light source in the final device. However, a light source with higher resolution is required to extract the optical performance of the filter. Moreover, a single detector combined with a variable slit instead of a detector array with fixed pixel width gives more freedom in choosing the pixel size during experiments.

An optical setup with a mid-IR laser and a large-area PbSe detector is built as shown in Figure 7. The light beam is spatially filtered right before reaching the detector using a 15 µm wide slit to replicate the effect of a pixel in a detector array. The gas-filled LVOF, which is mounted on a motorized translational stage, is scanned along its length to construct the transmission response at the wavelength of the laser. The optical chopper placed at the output of the laser improves the signal-to-noise ratio of the measurement. The incidence angle of the light beam impinging on the device is adjusted by the beam-steering mirror to improve the spectral performance [11].

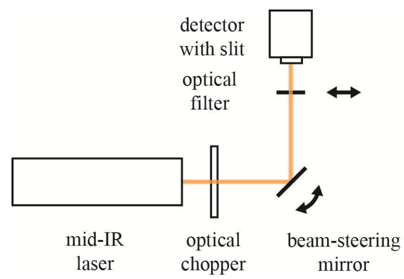

Figure 7: Schematic illustration of the characterization setup. The collimated output of the laser propagates through the optical chopper, reflects off the beam steering mirror, passes through the filter and reaches the detector with a slit.

To extract the response of the filter to the sample gas, first a reference measurement is performed using the IR-inactive gas, nitrogen, flowing through the resonator cavity. Then, the sample gas is fed through the cavity via the gas inlet and another spectral measurement is performed. The ratio of the measurement with the sample gas to the measurement with nitrogen gives the transmission response of the gas-filled LVOF to the sample gas.

The selectivity of the gas-filled LVOF is ensured by the wavelength-selection capability in a wide wavelength range. Methane has highly oscillating absorption peaks in the 3.2 µm to 3.4 µm wavelength range as shown in Figure 1. The absorption features of ethane start at 3.25 µm, while propane begins to absorb light at approximately 3.3 µm wavelength. Therefore, the operation band of the gas-filled LVOF is highly suitable to distinguish the main constituents of natural gas. The sensitivity to different concentrations on the other hand is related to the transmittance values. The concentration is extracted from the measured transmittance, the absorption coefficient, and the optical path length as described by the Beer-Lambert Law [12].

Measurements at 3270.75 nm and 3392 nm demonstrate the wavelength selectivity of the sensor as shown in Figure

8. The incidence angle of the light beam is selected as -6° at a filter-detector separation of 7 mm for the measurement at 3270.75 nm wavelength. The peak transmittance of the gas-filled LVOF with nitrogen was measured as 16.36%, while inserting methane reduced this value to 12.02%. The measurement at 3392 nm wavelength is performed at the optimum incidence angle of -8.8° at a longer filter-detector distance. The peak transmittance measured as 39.9% with nitrogen decreased down to 7.8% with methane, corresponding to more than 80% absorption. When correlated to the absorption coefficient of methane, the effective optical path length is calculated as 1.58 mm. This translates into a 62.2-fold elongation of the 25.44 µm long resonator cavity, experimentally.

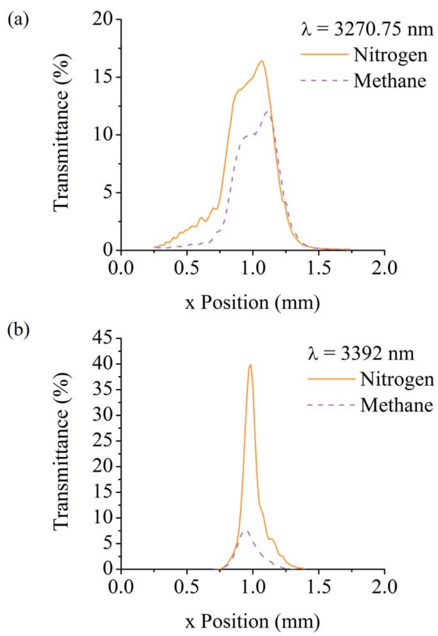

Figure 8: The measured spectral response of the gas-filled LVOF at (a) 3270.75 nm and (b) 3392 nm wavelength with nitrogen as the reference and methane as the sample gas.

CONCLUSIONS

The design, fabrication and characterization of the gas-filled LVOF as an on-chip photonic microsystem is presented. The wavelength selection in the 3.2 µm to 3.4 µm range allows for distinguishing the main constituents of the natural gas, namely, methane, ethane and propane. The selectivity is improved beyond the dimensions of the device by using the resonator cavity of a high-order LVOF with highly reflective mirrors also as a gas cell. The optical design of the LVOF with Bragg mirrors is reported using the Fizeau interferometer approach. The operation of the device is demonstrated at 3270.75 nm and 3392 nm wavelengths in the mid-IR with actual gas measurements using methane, proving the selectivity of the gas-filled LVOF. The improvement in sensitivity is validated by elongating the 25.44 µm long resonator cavity into the 1.58 mm effective absorption path, experimentally.

ACKNOWLEDGEMENTS

This work has been supported by the Dutch technology foundation STW under grant DEL.11476. The devices have been fabricated at DIMES (Else Kooi Laboratory) of TU Delft and the Nanofabrication Laboratory of Chalmers University of Technology. The authors are indebted to René H. Poelma for assistance in device packaging.

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