A Quartz Crystal Microbalance Based Portable Gas Sensing Platform for On-Demand Human Breath Monitoring

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ABSTRACT The correlation between gas concentrations in human breath and diseases has been increasingly revealed in recent years, triggering the need for cheap and easy-to-use gas sensors that can detect diseases in their early stages. The gas sensors used in these clinical applications need to be portable and sensitive, and preferably provide on-demand measurements for prompt diagnosis. In this paper, we propose a portable and cost-effective sensor platform that can quickly identify gas concentrations in human breath. Specifically, we combined quartz crystal microbalance (QCM) sensors with a single board computer, Raspberry Pi, to enable real-time data processing and display of the sensor data. A web server was operated on the single board computer, and thus, on-demand visualization of the sensor data was possible using a web browser. A gas quantification protocol was also proposed based on Freundlich’s adsorption isotherm. We demonstrated the real-time monitoring of methyl mercaptan (MM) gas over the internet using the developed small sensor device, which can easily be handled in one hand. The limit of detection (LOD) for MM gas was 107 ppb when the sensors were operated for 20 minutes after gas injection. The developed gas sensing platform is unmatched in terms of size and cost compared with analytical devices though less sensitive. These advantages would allow for wide distribution of the developed device to hospitals and individuals in large quantities, leading for behavioral changes for preventive care.

INDEX TERMS Gas sensor, QCM, methyl mercaptan gas, preventive care.

I. INTRODUCTION

Since the concept of the Internet of Things (IoT) was proposed by Ashton [1] in 1999, a considerable amount of small and cheap sensors were developed to enable ubiquitous sensing [2]. In particular, gas sensors have been actively studied due to the high demand of toxic gas detection for environmental monitoring and human breath monitoring for healthcare applications, which are related to recently rising important social problems such as air pollution and aging society [3]–[7]. In recent years, it has become clear that human breath contains more than 1000 types of gases, and many of these are closely related to diseases [8]. As a result, there is a growing need for measuring the concentrations of gases in human breath that are relevant to diseases (Table S.1). Portability and on-demand detection are two important properties that the sensors must have in order to use for such applications. These demands are partly satisfied in the development of other sensors such as tactile sensors [9]–[12]. However, development is still in progress for gas sensors.

To date, various types of gas sensors were developed including semiconductor-type sensors, surface plasmon resonance type (SPR-type) sensors, electrochemical sensors, thermoelectric sensors, and photoionization detectors (PID) [13]–[18]. Among them, semiconductor-type gas sensors are most widely used because of their advantages that the cost is low, the size is small, the power consumption is low and the response is fast [19]–[22]. Most semiconductor-type sensors are portable and are capable of estimating the concentrations of substances in a mixed gas [23]. Leveraging these features, an on-demand gas sensing platform for IoT applications was reported in [7]. However, the semiconductor-type
sensors can detect only substances that cause redox reactions on the surface of semiconductive materials since the gas is detected by the change in conductivity, which severely limits detectable diseases in health monitoring applications. Moreover, it is difficult to distinguish target gas from other interference gases because the sensor responds to various gases that cause redox reactions, though this issue may be partially improved by the use of temperature modulation or multiple partially selective sensors [24], [25].

To overcome this limitation, quartz crystal microbalance (QCM) gas sensors were developed [26]–[28]. The QCM sensors consist of a gas sensing film coated on a resonator. When the target gas adheres to the sensing film on the QCM, the frequency decreases due to the change of the mass, enabling gas quantification using electric circuits. Unlike the semiconductor-type sensors, the QCM gas sensors can selectively detect a large variety of gases by coating a sensing film with high affinity to target gases. Moreover, one can easily coat different adsorption films on the QCM to detect different types of gas, which is in contrast with other types of gas sensors that need to be completely replaced depending on the target gas. Further, the detection limit of the QCM sensors can be very low. For example, Roto et al. [29] fabricated ammonia gas sensors, whose limit of detection (LOD) was 550 ppb, and Tokura et al. [27] developed gas sensors that can detect methyl mercaptan at 20 ppb, which is lower than other gas sensors for sulfur gas [30]–[33]. A multi-gas sensor array was also developed to enable the simultaneous sensing of nitric oxide at 5 ppb and aceton at 5 ppm [26]. However, an on-demand and portable QCM gas sensor platform has yet been developed.

**FIGURE 1.** On-demand and portable gas sensing platform.

In this paper, we propose a portable QCM gas sensing platform capable of monitoring gas concentrations in human breath using web browsers on mobile tablets (Fig. 1). The sensor system is equipped with a single board computer, Raspberry Pi, with a custom Python program for real-time display of the sensor data on the browser of users’ tablets. The dimension of the sensing device is about 20 cm × 10 cm × 10 cm, and thus patients and medical doctors can easily handle the device in one hand and diagnose in real time by accessing the live data using a web browser. In addition, the entire sensor system including the sensors costs less than 500 USD, which is about one tenth of the state-of-the-art analytical equipments [34], [35]. Although the proposed platform is less accurate compared to the analytical equipments such as gas chromatography, these advantages would allow for wide distribution of the developed device to hospitals and individuals in large quantities, leading for behavioral changes for preventive care.

We demonstrated the on-demand nature of the gas sensing platform by displaying the sensor data on the screen of a smartphone and a laptop computer. We also derived a simple formula to quantify the gas concentrations from the sensor data, which are frequency of the resonators based on the Freundlich’s adsorption isotherm. This quantification protocol was illustrated by performing an experiment for methyl mercaptan gas.

**FIGURE 2.** Schematics of the proposed gas sensing platform.

**II. CONCEPT OF GAS SENSING PLATFORM**

**A. OVERVIEW OF THE PROPOSED SYSTEM**

The outline of the developed platform is illustrated in Fig. 2. Specifically, the gas sensing platform consists of three parts: QCM sensors, a frequency counter circuit, and a small single-board computer, Raspberry Pi, for frequency logging and data transmission to mobile devices through the internet (Fig. 2).

Gas molecules in human breath are initially bound to the surface of the QCM sensors, which are thin films attached to quartz crystal resonators driven by the Colpitts oscillator circuits [36]. This causes a small change in molecular mass at the films and results in the deviation of the oscillation frequency. This deviation is then detected by the frequency counter circuit connected to the QCM sensor and is sent to the Raspberry Pi upon the arrival of a read control signal. The Raspberry Pi stores the values of the frequency for later reference and, at the same time, generates a figure showing the measured data. The figure is then displayed on a web page, which is transmitted to users’ tablet devices through the internet. To control and monitor the sensing operation, a small touch-screen display is attached to an HDMI port of the Raspberry Pi circuit board.

The total cost of the sensing system is less than 500 USD. Moreover, the gas sensing films and the resonators, which are disposable parts cost about 15 USD. Because of this advantage, the sensing system can be distributed to hospitals...
and individuals in large quantities and can be used as one of the tools for preventive self check up, which would save on the insurance and social security cost.

**FIGURE 3.** (a) Frequency behavior of QCM sensor. (b) Schematic diagram of QCM coating by cast method.

### B. QUANTIFICATION OF GAS CONCENTRATION USING QCM SENSORS

The QCM sensor is composed of the QCM coated with a thin film that captures gas molecules. Specifically, when the gas molecules adhere to the surface of the film, the resonance frequency of the attached oscillator changes because of the change in mass (Fig. 3(a)). The mass of the attached gas molecules can then be estimated by Sauerbrey’s equation expressed by

$$
\Delta f = \frac{-f_1^2}{S \rho N} \Delta m,
$$

where, $\Delta f$ is the change in resonance frequency, $f$ is the fundamental frequency, $\Delta m$ is the mass of the attached gas molecule, $S$ is the electrode surface area, $\rho$ is the density of crystal, and $N$ is the frequency constant of the QCM [37]. For the quartz crystal used in this study (Crystal Sunlife Co., Tokyo, Japan), $f = 1.0 \times 10^7$ Hz, $S = 2.12 \times 10^{-5}$ m², $\rho = 2.65 \times 10^6$ g·m⁻³ and $N = 1668$ kHz·mm, and thus, $\Delta f = -1.04 \times 10^9 \Delta m$. This means that the frequency changes by about 1 Hz when 1 ng of a substance is attached to the film surface, implying that the sensor has sufficient sensitivity to detect a small amount of gas contained in breath. The gas concentration can be estimated from the frequency of the resonator using a calibration curve that maps gas concentrations to the frequency change as demonstrated in Section IV.

### III. DEVICE FABRICATION AND SOFTWARE DESIGN

#### A. GAS ADSORPTION FILM OF QCM SENSOR

We used the QCM with a methyl mercaptan gas adsorption film developed by Tokura et al. [27], which was made of manganese oxide nanosheets. The materials of the gas adsorption film were prepared based on the method presented in [27] and were ion-exchanged for enhancing sensitivity. The film was then coated by the cast method [28], [38], which is a coating technique that combines drip and natural drying (Fig. 3(b)).

According to the literature [27], this film has the following three properties: (i) 20 ppb of methyl mercaptan gas can be detected in a static system, which is the level required for the detection of periodontitis and other visceral diseases (see Table S.1). (ii) The film has high selectivity for methyl mercaptan gas, and (iii) the sensitivity of the film becomes high at high humidity, which is ideal for the monitoring of breath. Specifically, the response to methyl mercaptan gas was about five-fold higher than that to ethanol, ammonia, acetic acid, toluene and pyridine. In addition, the response was three times and seven times more sensitive at 35% and 50% relative humidity (RH) compared to 16% RH.

#### B. SENSOR HARDWARE SYSTEM

A custom oscillator circuit (Tama Device co. Ltd.) was made based on the Colpitts oscillators circuit to simultaneously drive three QCMs. Then, an AC voltage of 5 V was applied to drive the oscillator at about 10 MHz.

A PIC microcomputer (PIC24FV32KA302) was used to measure the frequency of the oscillators using the built-in frequency counter circuit. The oscillation frequency was then sent to the custom program on Raspberry Pi. The connection with Raspberry Pi was established via serial communication using UART protocol, to enable real-time and on-demand control of the PIC microcomputer using the Raspberry Pi (Fig. 2).

#### C. DATA PROCESSING BY RASPBERRY PI

Figure 4 is a diagram showing the data flow from the sensor to the user. A frequency read signal was sent to the PIC microcomputer every five seconds to measure the frequency of the resonators. Upon the arrival of the frequency data, a custom Python program on Raspberry Pi generated an image file showing the frequency change of the resonators. The data were also recorded in a CSV format for later reference. For the on-demand access to the sensor data, a web server was operated on Raspberry Pi, and an HTML file containing the image file was transmitted upon an access request from web browsers.

#### D. DEVICE ASSEMBLY AND OPERATION

The assembled gas sensing platform is shown in Fig. 5(a). The gas sensing system had three QCM sensors coated with methyl mercaptan gas adsorption films to avoid film-to-film variations due to the chemical manufacturing and the casting process. The sensors were placed approximately equidistant from the gas inlet so that the injected gas arrives at the three films at the same time (Fig. 5(b)). The operation of the device was confirmed by accessing the web page on Raspberry Pi from a laptop computer and a smartphone (Fig. 5(c)).

An example of the time-series data displayed on a laptop computer is shown in Fig. 5(d), where three QCM sensors in methyl mercaptan gas are shown. An embedded script in the web page automatically reloaded the sensor data every five seconds (Fig. 5(d)), enabling real-time monitoring of the output of the portable sensors. These results suggested potential applications of the proposed platform not only to the breath monitoring but also to real-time environmental sensing applications and food analysis [39]–[41].
IV. GAS QUANTIFICATION PROTOCOL

A. QUANTIFICATION BASED ON ADSORPTION ISOTHERM

In this section, we introduce a protocol for quantifying the gas concentrations using a calibration curve based on an adsorption isotherm. We convert the frequency of the QCM sensors to the gas concentrations using an adsorption isotherm identified by calibration experiments. Specifically, we use a modified version of Freundlich’s adsorption isotherm [42] since the methyl mercaptan gas chemically adsorbs on the gas adsorption film (see [27] for details). Freundlich’s adsorption isotherm is given by

\[ v = aP^{\frac{1}{n}} \]  

(2)

where, \( v \) is the adsorption amount, and \( P \) is the equilibrium pressure [42]. This equation implies that the adsorption amount \( v \) is not linear and gradually saturates as \( P \) increases. \( a \) and \( n \) are constants that depend on the adsorbent, the adsorbate and the adsorption temperature. The factor \( 1/n \) represents the saturation of the adsorption when the adsorbent layer becomes thicker.

To reflect the detection limit of the sensor, we add a positive constant \( b \) to Eq.(2) and obtain

\[ v = aP^{\frac{1}{n}} + b \]  

(3)

In Eq.(3), \( v \) is proportional to the frequency of the resonator according to the Sauerbrey’s equation (1). Thus, we identify \( a, b \) and \( n \) by measuring the resonator frequency at different gas concentrations and performing a curve fitting. In the next section, an experiment is performed to illustrate the proposed calibration method.

B. DEMONSTRATION OF METHYL MERCAPTAN GAS QUANTIFICATION

To illustrate the identification process of the calibration parameters in Eq.(3), the QCM sensors were put in a closed container where the gas concentration could be externally controlled (Fig. S.1). More specifically, the flow rate of methyl mercaptan, which is known as biomarkers for periodontal disease and colorectal cancer was controlled by a mass flow controller (MFC; MODEL 5100 SERIES, KOFLOC) attached to a switching valve controlling the timing of injection. Humidity of the closed container was also regulated by bubbling a compressed air through a water bath.

To generate a calibration curve, methyl mercaptan gas was injected at 186, 500, and 1000 ppb, and the frequency of the oscillator was recorded. These experiments were carried out at 61.5% ± 4.5% RH. It should be noted that the relative humidity used in this experiment was lower than that of human breath (90% RH), implying that the sensitivity might have been lower than that with actual human breath. All of the three QCM sensors responded immediately to the methyl mercaptan gas, and the time courses of the frequency change were similar between the sensors, implying that on-demand quantification of the gas concentration was possible with reliable gas sensing films (Fig. 6(a–c)). A measurement was also performed at 0 ppb as control (Fig. S.2). We observed that the frequency decreased over time due to the continuous adhesion of the methyl mercaptan gas molecules to the film.
Thus, the frequency change at 5, 10, 15 and 20 minutes after the injection of the gas was recorded. Using these data, the adsorption isotherm was generated by least square fitting using Eq.(3) (Fig. 6(d)).

As illustrated in Fig. 6(d), the trend of the fitted curve was consistent with the measured data in that the response frequency tends to saturate as the concentration increases. It was observed that the horizontal intercept decreases with time. At 20 minutes, the LOD was 107 ppb [43], though, in practice, more detailed calibration would be necessary around 100 ppb (see Table S.2 for more information). These results suggest that the proposed calibration equation (3) is useful for the estimation of the gas concentration.

V. CONCLUSION

We have proposed a portable and low cost gas sensing platform that can monitor gas concentrations using web browsers. The proposed system consists of QCM gas sensors and a small single-board computer, Raspberry Pi, that enables on-demand display of measured data through the internet. The combination of these technologies allow for cost-effective preventive self check up, which would lead to a detailed secondary diagnosis and brings a behavioral change for disease prevention.

Using the developed device, we have first confirmed using a smartphone and a computer that the measured data could be displayed on the screen in real time. Then, the gas quantification protocol using Freundlich’s adsorption isotherm was proposed. To demonstrate the gas quantification protocol as well as the on-demand nature of the proposed device, we have performed calibration experiments for methyl mercaptan gas.

In the future, combined efforts should be made both in circuit and adsorption film development to improve the accuracy of the sensor system at low concentrations, leading to the detection of diseases at earlier stages. We also envision that the developed multi-channel QCM sensor systems would enable simultaneous multi-gas sensing by coating with different adsorption films.

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