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Indoor air quality monitor based on Solidly Mounted Resonators for the detection of VOCs

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***Abstract*—This work describes the development of an indoor air quality monitor based on high frequency solidly mounted resonators (SMR) for the detection of volatile organic compounds (VOC). The sensor system operates in a differential mode with two SMR devices driven by Pierce oscillators. The active resonator was spray-coated with a selective polymer compound, while the second one was used as a reference. The absorption of vapor molecules into the polymer layer causes a frequency shift in the resonator. A study of suitable polymer coatings and finite element simulations on the sensor response were performed. Experimental results demonstrated that the VOC monitor is capable of detecting toluene concentrations down to 5 ppm with a high sensitivity of 60 Hz/ppm for a 180 nm PDMS polymer coated SMR resonating at 900 MHz. We believe that the system could be used as a robust, low-cost and low power microsensor for the real-time and ubiquitous monitoring of VOCs, particularly in a personal air quality monitor.**

Keywords— Acoustic wave sensor, indoor air quality monitoring, Pierce oscillator, solidly mounted resonator, toluene detection, VOC sensor.

I. INTRODUCTION

VOCs emitted from both natural and anthropogenic sources pose significant adverse effects on human health. Acute effects such as dizziness, headaches and skin irritation are associated with the inhalation and direct contact to VOCs in the ambient air [1]. Other respiratory problems, such as asthma and lung cancer, have also been reported [2]. The aromatic VOCs of benzene and toluene have been found to pose the most significant health risks, particularly benzene which has been associated with cancer and leukemia [3].

Instruments such as gas chromatographs and mass spectrometers are widely used for the detection of VOCs. However, these are bulky devices unsuitable for real-time, in-situ applications. Additionally, more stringent exposure limits have been introduced by legislators, resulting in an increased interest in portable and real-time technologies capable of detecting indoor VOCs.

Portable technologies such as electrochemical sensors, metal oxide semiconductor (MOX) sensors and photoionization detectors (PID) are used to this purpose. Electrochemical sensors offer an affordable solution to detect a wide range of VOCs but their use as air quality monitors is limited due to their susceptibility to poisoning, short life span and bulky design [4]. MOX sensors offer fast response and long life span but have high power consumption. PIDs are the gold standard for VOC detection with high sensitivity and fast response time. However, they are bulky, have limited selectivity and require periodic calibration.

Acoustic wave based sensors such as Quartz crystal microbalances (QCMs) and surface acoustic wave devices (SAWs) have been widely investigated for the detection of VOCs. These devices are low-cost and have fast response times. However, they exhibit limited sensitivity, selectivity and are susceptible to noise. Due to their suitability to operate at higher frequencies, SMR devices exhibit improved sensitivity. Furthermore, their small footprint and compatibility with CMOS technology make them a suitable candidate for the development of a miniature, low-cost and personal indoor air monitor for VOC detection.

Toluene is the most commonly found VOC in indoor environments [5]. For this reason, it is important to monitor the indoor concentration levels of this vapour to ensure they are kept within the safe exposure limits set at 50 ppm and 100 ppm for the long-term (8-hr reference period) and short-term work exposure limit (15-minute reference period), respectively [6].

In this work, we present the development of a low-cost, highly sensitive indoor air quality monitor based on Solidly Mounted Resonators for the detection of VOCs, such as toluene.

II. VOC SENSOR SYSTEM

The air quality monitor operates in a dual oscillator configuration to ameliorate environmental and other common mode interference. Two SMR devices are driven by Pierce oscillators in a differential mode implemented using a mixing circuit. One oscillator acts as the reference while the second is the sensing element. The overall structure of the system is depicted in Fig. 1.

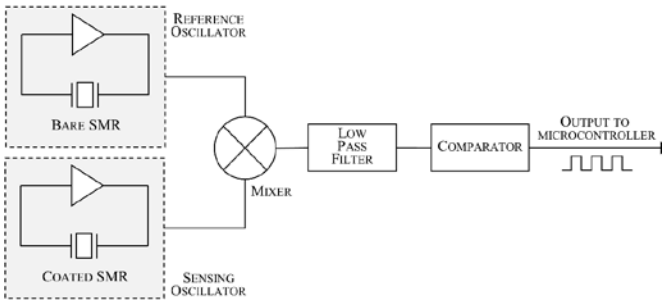

Fig. 1. Block diagram of the SMR based VOC sensor and the associated drive circuitry.

The high frequency signals from the reference and sensing oscillators are mixed in a heterodyne process using an RF mixer. The low pass filter provides a differential output (suppressing the noise due to harmonic signals). The sinusoidal output from the low pass filter is converted to a square wave having the same frequency, for digital processing using a microcontroller.

A. *Solidly Mounted Resonator*

The sensing element of the system is a ZnO based solidly mounted resonator working at a resonant frequency of 900 MHz. An electric field applied to the piezoelectric layer generates a mechanical wave that propagates through the bulk of the material.

For the detection of VOCs, a thin polymer film is coated onto the sensing area of the SMR as depicted in Fig. 2. The absorption of the vapour molecules into the polymer causes a change in mass, which in turn results in a frequency shift of the resonator. The sorption of the vapour molecules is a reversible process, where the vapour can desorb and recover its baseline; thus the sensor is capable of continuous, real-time monitoring.

The SMR devices were fabricated in a 3-mask photolithographic process. They consist of a ZnO thin layer (2.9 µm) sandwiched between Al electrodes (200 nm) with an acoustic mirror formed of three layer pairs of Mo/SiO_2 (1.85 µm/1.5 µm). Details on the design and fabrication process of these devices are reported elsewhere [7].

B. *Driving Circuitry*

A Pierce oscillator was designed to drive the SMR devices. For acoustic resonators operating at high frequencies, this type of oscillator is preferred as less parasitic effects are introduced, providing good frequency stability and large output at low power level with only a few components [8]. To simulate the designed circuit, the electrical equivalent circuit of the resonator was extracted. The oscillator was implemented in a four-layer printed circuit board with separate power and ground layers to reduce cross talk between high frequency signals, resulting in stable oscillations. A separate PCB was designed to implement the mixing circuit comprising the RF mixer, low pass filter and comparator. A typical output signal of the complete system is shown in Fig. 3. This is the differential signal output of the comparator which is acquired by the microcontroller.

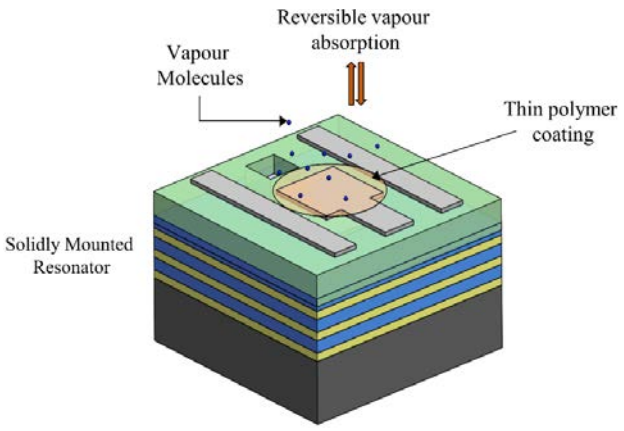

Fig. 2. Schematic of polymer-coataed SMR for VOC detection.

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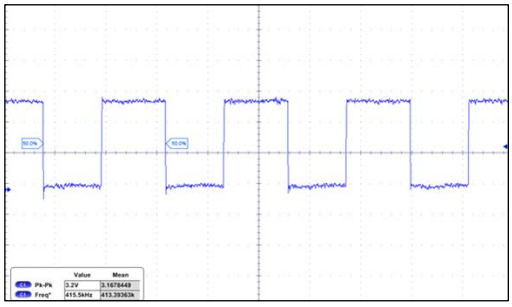

Fig. 3. Typical output of the complete system. Frequency count is performed by the microcontroller.

III. POLYMER COATING

A. *Polymer selection and spray-coating*

Appropriate selection of the polymer coating is essential to ensure good sensitivity and selectivity to the target vapour. The strength of the interaction between the vapour and polymer molecules is described by the vapour-polymer partition coefficient K_p, which relates the concentration of vapour molecules absorbed in the polymer film, C_p, to the concentration of the sampled vapour in the gas phase, C_v. The larger the partition coefficient, the stronger the vapour sorption.

Partition coefficients were calculated for different polymer-vapour pairs using linear solvation energy relationships and polymer solvation parameters available from published data. A PDMS coating was chosen for the detection of toluene, with a partition coefficient value of 1165.

Polymer coating of the device was performed using a spray-coating method. The spray system consisted of a single action airbrush with compressed air as carrier gas. A 0.5% w/v solution of PDMS was continuously sprayed onto a rotating disk driven by a DC motor. An aperture in the disk allows the solution to reach the surface of the device located underneath. A LabVIEW™ virtual instrument was used to control the spraying time and rotation speed of the disk. In this way, small amounts of polymer solution can be deposited onto the sensor surface resulting in thin and uniform polymer films. Once coated, the SMR devices were baked overnight at 100°C and left in ambient air for a period of 20 hours for stabilization.

B. *Finite Element Modelling*

Frequency response of the PDMS coated SMR due to toluene absorption was simulated in COMSOL Multiphysics v4.2 using a 2D model of the device. The absorption of toluene in the polymer film was represented as an increase in the density of the polymer film given by (1)

$$\rho_{VOC/polymer} = K_p \cdot M \cdot c \qquad (1)$$

with M the vapour molar mass and c the VOC concentration in air given as

$$c = (c_0 P)/(RT) \qquad (2)$$

A linear relationship between frequency shift and toluene concentration was observed while the sensitivity of the device exponentially increased with the polymer thickness as shown in Fig. 4.

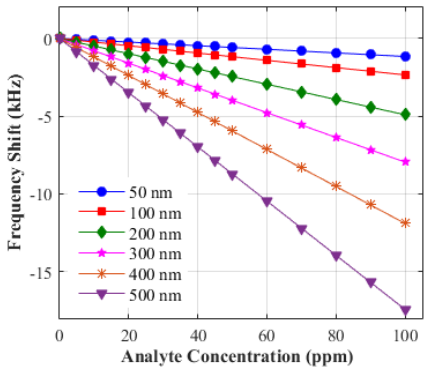

Fig. 4. Simulated frequency shift of PDMS coated SMR to different concentration of toluene and different polymer thicknesses.

IV. Experimental

Testing of the VOC detection system was conducted within a fully automated test station depicted in Fig. 5 where sensors can be exposed to different concentrations of target vapours (down to ppb levels) over long periods of time with repeatability and minimum user interaction.

Sensors were placed in a gas exposure chamber (3.5 mL dead space volume) kept at a constant temperature of 35ºC. Fig. 6 shows the measured frequency shifts for three different PDMS thicknesses when exposed to concentrations of toluene ranging from 5 ppm to 100 ppm. As expected from the simulations, the sensitivity of the device increased with thicker coatings. For the 180 nm PDMS coated sensor, a sensitivity of ~60 Hz/ppm was observed.

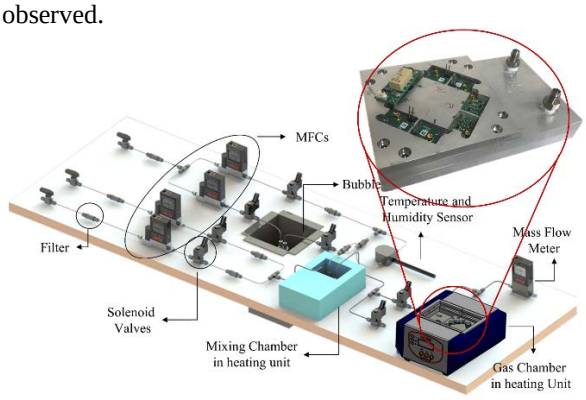

Fig. 5. VOC automated test station and gas exposure chamber.

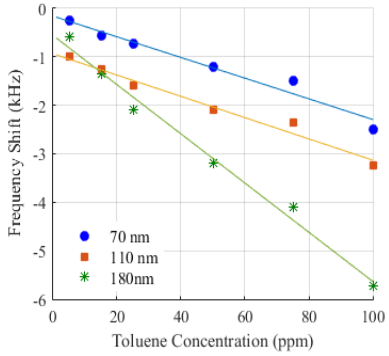

Fig. 6. Measured frequency shifts of PDMS coated SMR to different concentration of toluene for three different polymer thicknesses.

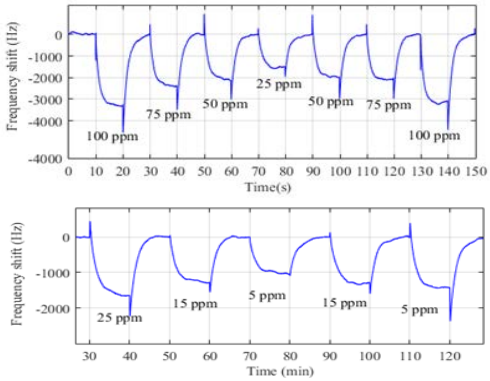

Fig. 7. SMR sensing system response to different concentrations of toluene (5 ppm to 100 ppm). PDMS thin film is 110 nm thick.

Repeatability of the results was tested with the experiments performed at least three times. Vapour desorption was observed with the sensors returning to a baseline of synthetic air at a constant flow rate of 200 mL/min. Fig. 7 shows the time response plot of the sensor system with a 110 nm polymer film to different concentrations of toluene. A linear fit was applied to the data to remove the drift observed in the sensing system. The system was capable of detecting toluene concentrations down to 5 ppm, well below the maximum safe exposure limit.

V. Conclusions

A VOC sensor based on polymer-coated high frequency solidly mounted resonators operating in a differential configuration was developed. Simulations on the frequency response were performed and experimental results were obtained for the detection of toluene. A sensitivity of 60 Hz/ppm was measured for a 180 nm PDMS coated device with a resolution of 5 ppm, demonstrating a good agreement with the simulations results. The sensor was capable of real-time detection of concentrations well below the safe exposure limit, making it suitable for low-cost indoor air quality monitoring. Further work is underway towards the development of a low power, portable, and smart microsensor system by integrating the SMRs and the associated circuitry into a single monolithic system, capable of being used as a personal air quality monitor.

References

[1] Rumchev, K., Brown, H. and Spickett, J., "Volatile Organic Compounds: Do they present a risk to our health?", Reviews on Environmental Health, vol. 22, no. 1, pp. 39-55, 2007.

[2] Cakmak, S., *et al.*, "Residential exposure to volatile organic compounds and lung function: Results from a population-based cross-sectional survey", Environmental Pollution, vol. 194, pp. 145-151, 2014.

[3] IARC, "Chemical agents and related occupations Volume 100F: A review of human carcinogens", 2012.

[4] Piedrahita, R., *et al.*, "The next generation of low-cost personal air quality sensors for quantitative exposure monitoring", Atmos. Meas. Tech., vol. 7, no. 10, pp. 3325-3336, 2014.

[5] AGÖF, "AGÖF Guidance Values for VOCs in Indoor Air", 2013.

[6] Health and Safety Executive (HSE), UK "EH40/2005 Workplace exposure limits", 2013.

[7] F. H. Villa-López *et al.*, "Design and modelling of solidly mounted resonators for low-cost particle sensing," Meas. Sci. Technol, vol. 27, no. 2, p. 025101, 2015.

[8] Matthys, R. J.. "Crystal oscillator circuits". Florida: Krieger Co., 1992.