Thermal and Electrical Investigation on LTCC Gas Sensor Substrates

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Abstract—Metal Oxide (MOx) semiconductor gas sensors typically operate at temperatures of few hundred Celsius degrees and consume hundreds of milliwatts of power. It is therefore, essential, to investigate the heat flux and power consumption in MOx sensors, especially designed for applications in battery-powered devices. The work presents the thermal and electrical investigations on LTCC substrates (Low Temperature Cofired Ceramic) as a base material for gas sensors. A novel shape of substrates with reduced heat capacity is showed. The sensor temperature was modulated with a pulsed heater voltage, therefore decreasing the average power consumption.

Index Terms—LTCC, gas sensors, thermal investigation, metal oxide.

I. INTRODUCTION

During the last several decades, considerable advances in gas sensor technology have been achieved in various applications such as: safety devices [1], environmental monitoring [2], air quality control [3], and health diagnosis [4]. Metal oxide (MOx) semiconductors have become promising gas sensors due to their low cost, short response time, long life and wide range target selectivity [5]. Therefore, numerous researchers presented the investigation results on the following oxides: CuO [6], In$_2$O$_3$ [7], WO$_3$ [8], TiO$_2$ [9], V$_2$O$_5$ [10], Fe$_2$O$_3$ [11], MoO$_3$ [12], ZnO [13], etc. Different metal oxide based materials have different reaction activation to the target gases. The chemical and physical processes of working sensors are well known [14], but still sensors behaviours are investigated. However, it is well known that the operating temperature is the most important factor that influences the performance of semiconductor gas sensors. The MOx sensors usually work in 350-500°C temperature range [15]. To provide such elevated temperatures a heater is usually embedded into gas sensors substrates. Depending on expected working temperature and substrate built, the consumed power can reach the level of hundreds of milliwatts. The power consumption of gas sensors can be reduced by using a pulsed operation mode, gas sensitive layers working at lower temperatures, changes in substrate design and materials [16]. The search for suitable substrate for semiconductor chemical sensors is still a challenge.

In this paper, thermal and electrical investigations on LTCC gas sensors have been reported. The authors used the LTCC substrates for gas sensors with sensitive layers of SnO$_2$/In$_2$O$_3$ [17], CuO [18], WO$_3$ [19], ZnO [20].

II. DESIGN

A. Gas Sensing Device

Fig. 1 shows the concept of a complete gas sensing device which consists of few elements such as: gas sensitive layer, gas sensor substrate, package, front-end electronics and signal processing. For developed gas sensors the authors used TO-5 package. The electrical connections between sensors pads and case pins were welded with a 0.5 mm thick Pt wire.
as Fourier's law. It can be written as (1):

\[ q_p = -\lambda \cdot \nabla T \]  

(1)

where: \( \lambda \) – thermal conductivity coefficient characteristic for the material, \( \nabla T \) – negative local temperature gradient.

The heat convection is known as Newton's law of cooling and it can be written as (2):

\[ q_k = h_k \cdot A \cdot (T - T_0) \]  

(2)

where: \( h_k \) – the heat transfer coefficient, \( A \) – heat transfer surface area, \( T \) – temperature of the objects surface, \( T_0 \) – temperature of the environment.

The radiative heat transfer between two surfaces is calculated by the Stefan-Boltzmann law (3):

\[ q_r = \sigma \cdot \varepsilon \cdot A \cdot (T^4 - T_0^4) \]  

(3)

where: \( q_r \) is the density of radiative heat flux, \( \varepsilon \) is the emissivity and \( \sigma \) is the Stefan-Boltzmann constant equal \( 5.67 \times 10^{-8} \) W/(m\(^2\)K\(^4\)).

The heat flux in the gas sensor substrate made in LTCC technology with embedded heater is shown in Fig. 3. The total heat transfer can be summarized as (4):

\[ q_{\text{total}} = q_1 + q_2 + q_3 \]  

(4)

where: \( q_1 \), \( q_2 \) – represent convection and radiation, \( q_3 \) – represents conduction.

As can be noticed, when the sensor works at temperature around 250°C, the package heats up to 80-90°C. For operating temperature around 350°C the package is heated up to 120°C. As a consequence, the more power is needed to heat up the substrate and stabilize the temperature. Due to this fact, the sensitive materials working at lower temperature are desirable. Recently, researchers are looking for materials which exhibit gas sensing properties at room temperature [22]-[25].

B. LTCC Fabrication

The authors used LTCC for years as a base material for gas sensors substrates. The first substrates consisted of three Green Tape layers with 6 mm in diameter and 0.2 mm thickness. The heater was deposited by thick film technology on the second layer from the top. The sensor electrodes were placed on the top, while contact pads were placed on the bottom side. Three pairs of gold interdigitated electrodes with 200 µm line and gap width were formed. The main limitation of described substrates was very high heat capacity and therefore higher power consumption [17]. In next version [21] the substrate diameter was reduced to 4 mm, while the thickness remained 0.2 mm. The gold interdigitated electrodes were reduced to 100 µm width of lines and gaps. To provide better temperature distribution, the metal layer was deposited on the bottom side. The authors simulated the shape and thickness of this layer as well as the heater topology to obtain the best temperature distribution in sensitive region [21]. However, the measurements revealed that power consumption is still too high and more uniform temperature distribution does not improve the gas sensing parameters. In the third substrate version the additional metal layer at the bottom was omitted, but the heater geometry was optimized, as well as a new, smaller shape was designed (Fig. 5). Moreover, the electrodes were reduced to 80 µm and 100 µm line and gap width, respectively.

III. RESULTS AND DISCUSSION

A. Power Consumption

The sensors heaters were deposited with two different
pastes, i.e. polymer conductive silver paste 1109-S with resistivity <50mΩ/sq or silver conductive paste 9912 with resistivity <0.15mΩ/sq (ESL Inc, PA, USA). Fig. 6 shows the power consumption as the function of operating temperature for the heaters made of two kind of pastes. As can be seen, there is no significant difference between those two pastes, as the sensor construction is the same. However, paste 1109 has higher resistivity, so it is more suitable for that application. The temperature responses for gas sensors with embedded heater deposited using 1109 paste for different values of voltage supply are shown in Fig. 7.

As can be noticed from Fig. 6, the power consumption is still in the range of few hundred mW for constant power supply. For example, assuming the 300 mW of power consumption (working temperature ca. 250°C), a typical AA battery of capacity ca. 2000 mAh could only suffice for 6 h operation [26]. To overcome this limitation the substrate has to be mounted to package with lower thermal conductivity, e.g. alumina. However, the best results can be obtained by reducing the size of MOx sensors. It is the most effective way to reduce overall power consumption. It has to be underlined, that semiconductor metal oxide gas sensors are highly resistive. Therefore, the interdigitated electrodes are used in order to reduce the resistance to be measured. Nowadays, in LTCC technology, the lines and gaps can be realized with 50 µm and 80 µm width, respectively. Summarizing, three pairs of interdigital electrodes will cover the area of 0.9 mm². Currently used substrates have the area of 4.9 mm². The novel substrates are still under investigation.

The resistance changes in wide range of temperatures for heater deposited using 1109 paste is shown in Fig. 8. The TCR (Temperature Coefficient of Resistance) was approximately 2003 ppm/°C. For comparison, the TCR of platinum heater is 3850 ppm/°C (IEC751 Standard). The temperature changes for gas sensor heated with pulse voltage of various frequencies such as: 20Hz, 50Hz, and 100 Hz is shown in Fig. 9.

The power consumption can be further reduced by operating the heater in pulse mode, while the voltage is applied according to specific function (i.e. 5 V for 20 s and 0 V for 40 s). The average power consumption can therefore be proportionally reduced.

IV. CONCLUSIONS

The proposed LTCC gas sensor substrates are an alternative to a widely discussed membrane substrates based on Si [27], alumina (Al₂O₃) [28] or novel flexible substrates [29]. Thermal, electrical and mechanical stability of LTCC gas sensor substrates meet the challenging conditions for many applications such as: automotive or medicine (i.e. breath analyzers). Moreover, the technology is much easier and cheaper than silicon. However, the main limitation are electrode line and gap dimensions, that cannot be less than tens of microns. It is still too high for nanowires or nanorods based thin films, that exhibit a huge surface to volume ratio which is a highly favorable for gas sensors. It is essential therefore to investigate a new techniques to obtain such low dimensions. Due to this fact, the LTCC substrates have to be improved.
REFERENCES


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