EXPLOITATION OF LTCC QUALITIES IN GAS SENSORS APPLICATION

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SUMMARY

In this contribution we deal with technology potential of the low temperature co-fired ceramic (LTCC) in association with design, development and production of organometallic thick film gas sensors. Customer demands to the gas sensors array and to the joint construction of the sensing part with evaluating ASIC led to the exploitation of LTCC potential in the production technology. The presented outcomes are based on the lot of investigations and simulations on the several models of organometallic gas sensors made with one common factor - low temperature co-fired ceramic utilisation

The most important characteristic of LTCC and main reason for its utilisation in technology of sensor production is behind its temperature charactersistics also the possibility to fabricate various shapes in several separate layers which finally create monolithic ceramic module based on its attractive features. In this review, we want to emphasise tailored utilisation of LTCC in thick film based gas sensors, which were successively realised in the following parts: LTCC based multilayer heating systems for achievement of sensor operating temperature with exceedingly flat temperature distribution in the active sensor area, creation of the special LTCC sensor/ASIC carrier for relative high temperature and safety operation and finally LTCC based design for the gas sensor array.

Keywords: *Low Temperature Cofired Ceramics, LTCC, Thick Film Gas Sensors.*

1. INTRODUCTION

The entrenchment of Low Temperature Cofired Ceramics (LTCC) into the electronics technologies gives strong tool to Thick Film Technology (TFT) which enables production of particular three dimensional custom-tailored structures. It has an impact to the several electronics applications including unconventional TFT applications in the field of sensors design, development and production. The field of attention also includes developments in the production technology of substrates for microelectronics, packaging and the multilayer 3D structures for general utilisation.

Raw LTCC material represents “high-tech” microelectronic material which contains 40% Al_2O_3, 45% SiO_2 and 15 % of the organic compound and which is used for its attractive flexibility of shape in conditions enabling creation of various multilayer structures. Utilization of this technology can simplify assembling of the hybrid circuits, other modules or sensors in the frame of thick film technology. Moreover LTCC represents the efficient way of making multilayer structures by replacing the dielectric screen printable paste with dielectric foils. LTCC systems build multilayer using a cost-effective parallel process instead of the sequential process used in conventional thick film hybrids.

LTCC modules are prepared by specific cutting of the raw “Green-tape” foil to a demanded substrate size, punching the interconnecting vias or other designed cavities, filling the vias by thick film paste and patterning of the interconnected lines and/or possibly other layout on the singular raw substrate. These operations are repeated for as many layers as the design requires. Separate raw substrates which create designed module are subsequently collated, stacked, and laminated to the raw multilayer structure. This way created multilayer raw module is co-fired afterwards and the surface layers can be completed by another screen printed layouts like pattern of resistors, conductors and/or assembly of the surface mounted devices.

This article presents some of adjusted results using low temperature cofired ceramics in the development, design and production of the organometallic thick film gas sensors. The principle of gas sensing is based on reaction at effective SnO_2 surface which has to bring more important role than spatial volume changes in semiconductor layer. This is why the specific surface i.e. ratio of layer volume and effective surface could be minimised. The choice of the SnO_2 layer as N-type semiconductor material is based on electric resistance increasing after an oxygen adsorption and consecutive resistance decreasing after further desorption by reduction agent. These resultant changes of sensing layer conductivity are occurred following surface reactions at semiconductive effective surface.

The design philosophy of the organometallic thick film gas sensor goes out from accumulating electrodes system in combination with active and reference organometallic SnO_2 layers and heater system with flat heating shape applied on both sensor surfaces - active and reference. In compliance with theoretical assumptions it was necessary to provide several conditions for the assign of well gas sensing stability, sensitivity and selectivity. Utilisation of the LTCC offers final solution for some of the above demands:

- To determine and to ensure of the optimal operating temperature with really flat behaviour by appropriate design. The higher temperature causes so high reaction speed, the lower temperature could cause slow reaction speed which brings changes in sensing characteristics. Both cases can influence to measurement accuracy and sensor stability. Moreover, the selectivity of gas sensor can be also controlled by regulation of the operating temperature level.

- To secure a reference resistance which could eliminate the perturbing influences of surrounding conditions including the potential operating temperature alteration and brings higher stability to resistance measurement by build in to inner layer of LTCC module.
- To prepare unique and common carrier system for sensor and ASIC, including possibility for sensors array design.

The LTCC Du Pont DP951 Green Tape^{TM} was chosen for thick film gas sensors applied in support of methane sensing and this choice is presented in the article. This material selection was used also as an appropriate alternative for task solution of the temperature field distribution by reason of needs to homogeneous and really flat temperature dispersion at both of organometallic surfaces. LTCC is otherwise convenient material for appliance at relative high operational temperatures (450 – 470 °C for methane sensing) and finally for opportunity to create design of common carrier for controlling ASIC and gas sensors array. The main reasons for using of Green Tape^{TM} techniques for this application are:

- High strength, high flexural strength and simplicity of tape processing in the raw state.
- Thermo-physical properties, e.g. low thermal conductivity and stability.
- Mass production methods can be readily applied.
- Multilayer heating system can be integrated into the electronic modules/carriers.
- Fabrication techniques are relatively simple, inexpensive and environmentally friendly.

2. SENSOR DESIGN

The task to design of organometallic thick film sensor can be split into two subtasks. First problem represents design of the sensorial part with active and reference SnO_2 layers and the second problem poses the sensor heater design for operating temperature acquirement and guarantee with the optimal distribution of temperature field. Used materials are situated at relatively difficult conditions because of the high sensor operating temperatures. The LTCC excellently meets all of the demands.

The optional design is based on the idea that both surfaces of sensor substrate will be effective used (Figure 1). First – face side – for the sensing part construction and second – back side – for the realisation of the heating element. Designed layout enables to attach the sensor-unit to carrier frame by using of standard wire bonding technology and secondary reduces the temperature transmission to a further parts – i.e. ASIC or another sensor units.

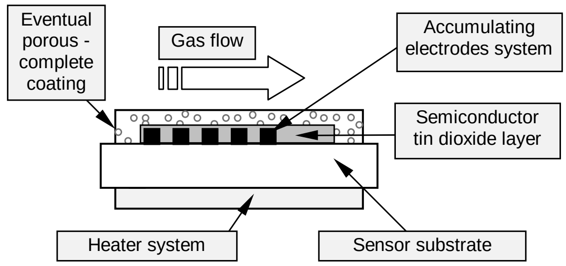


Fig. 1 Cross section of basic design for Thick Film gas sensor unit.

2.1. Sensorial part

The design philosophy is based on accumulating electrodes system in combination with active organometallic SnO_2 layer. The particular design evolution was focused to layout of electrode system in combination with several sensing and covering layers. The choice of the most appropriate grid density is given by Thick Film technology and subsequent dimensions which also influenced by heater design and its capability to secure the optimal operating conditions – i.e. the flat collocation of temperature field and achievement of operating temperature level with respect to limited maximum power consumption.

Numerous of experiments and measurements led to the resultant design of the sensor unit based on LTCC carrier with dimension 5 to 5 mm (Figure 2). In coherence with acquired experience and measured characteristics was chosen the most suitable raster for electrode system represented by values – 0,125 mm width per electrode and 0,125 mm width per gap at layer thickness 0,012 mm. The surface was completed by couple of "dummy" bumps on lower part – with respect to the sensor package and assembling concept. Moreover, these bumps were subsequently appointed for power attachment of heating part after wire bonding in sensor carrier at the package. Conductive interconnections between both sensor unit sides of produced samples were realised by conductive coating across the edge. This technique for joint interconnections realisation was terminated by technological potential at available laboratory.

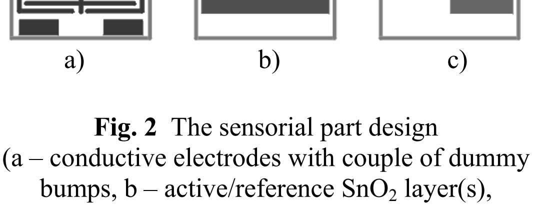


Fig. 2 The sensorial part design
(a – conductive electrodes with couple of dummy bumps, b – active/reference SnO_2 layer(s),
c - covering layer for reference values and evaluation of the gas concentration - LTCC).

2.2. Heating part

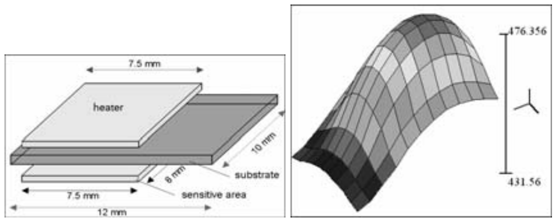
Generally the heater has an essential role in majority of similar gas sensors because the operating temperatures are relatively high and behind of sensor stability operating temperature determines the selection of sensing gases. Particular temperature level for methane sensing is in the range from 420°C to 470°C. The really important factor for sensors characteristics and their stable sensitivity level is the distribution of heating field on the sensitive surface. For that reason the heater unit was situated on the back side of sensor substrate and the increased accent to analysis of temperature conditions on sensor substrate was given.

According to designed assembly and packaging method the ceramics substrate dimensions should be so small as is technologically possible and the power consumption for the operating temperature achievement should has minimum level. Sensors substrate dimensions finished from initial 150 to final approx. 25 square mm and the necessary power consumption varied from initial 6W to final value under 2W during the sensor design development..

The heating elements based on resistive layers are often used in many electronics systems. The examples of their applications could be found among aviation systems, military equipment, systems for temperature control and as well as in manufacturing of different types of sensors (e.g. heaters for gas or humidity detectors). In many cases it is important not only the total value of dissipated power and the highest or average heater temperature but the homogeneity of temperature field distribution on the surface of sensing elements and heaters for gas sensors have a special position in this area. The problem of suitable heating elements construction has an essential role because, besides the relative high operation temperature – typically higher than 200^{o}C, the properly shaped temperature field on the sensor surface is required. For this purpose, it is beneficial application of the thick-film resistor and/or the conductive layers as heaters. One of the most important features of LTCC technology which determine its using in this territory is high level of the flexibility.

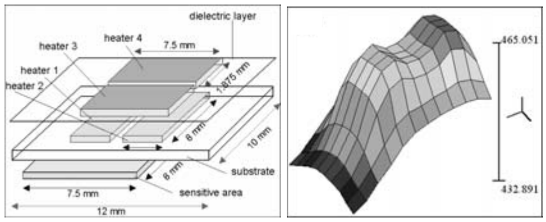
The basic problem in design of such devices is in the selection of proper shape of particular resistors and conductor layout as well as theirs mutual arrangement inside the mentioned structure. The experimental methods in combination with computer simulations are utilised usually in practical solutions of similar tasks progress. This way acquired results represent global information regarding the heat exchange mechanism and temperatures fields' distribution. The main goal of this investigation was presentation of the temperatures field formation.

The basic planar resistive structure should be extended by adding of several further heating elements for improvement the homogeneity of temperature distribution on the sensor surface. The simulations and shaping of the temperature field were realised for several substrates with different geometrical parameters. The effects of the resistors number, their geometry and as well as their arrangement to the temperature field distribution are shown on the Figures 3 and 4.



A) B)

Fig. 3 Simple planar structure of heater element (A) and 3D plot of the temperature field distribution on surface of sensitive area (B).



A) B)

Fig. 4 Multilayer structure with four heating elements (A) and 3D plot of temperature field distribution on the sensitive surface (B).

Considerable substrate dimension reducing based on demands to sensor arrays brought simplified heater layout which consists exclusively from conductive layer. The heating resistor array was substituted by conductive meander.

This heater concept for sensor array brought also relevant simplification to production technology. On the other hand this design could cause decline of temperature field collocation quality and expectation of problem with cracking of ceramic substrates. Acquired results shows that these apprehensions were unsupported because realised sensor samples operated in accordance with theoretical assumptions and there were no problems with eventual sensor cracking. This fact is conditioned by the above mentioned reduction of sensor unit dimensions - approximately to the quarter of the original size. The surface minimisation (under 25 sq. mm), brings beside the power saving also lower requirements on the temperature field collocation and on the decrease of cracking hazard level of substrate carrier. Furthermore the reduction of sensor weight brings reliability improvement of mechanical fixation in the designed carrier. The final dimensions were specified to 4,25 x 4,75 mm and the heater layout design is shown in the Figure 5.

Fig. 5 The heater design based on conductive meander (line to gap thickness rate: 0,175/0,250 mm).

This heater concept brings the possibility to use heating layer for the temperature measurement and power consumption control simultaneously. The synchronized heating and temperature monitoring are based on solving of simple mathematical equation for temperature/resistance dependence in conductive layers which allows calculation of topical temperature based on measured values of heater resistance. These data can be acquired and processing in real time and they could be used for temperature monitoring and secondary for sensor temperature and power consumption control.

3. REALISATION OF SENSOR UNIT, SENSOR CARRIER AND SENSOR ARRAY

Designed sensors were produced in several steps in coherence with suggested design and with progressive acquisition of necessary experience. The realisation of several versions of the sensitive layers (changes of the active surface largeness and thickness) at the small substrate and the latest heating concepts allow collocation of two or more sensors which used different heating temperature into one carrier combined with evaluation ASIC. Moreover the simply change of accumulating electrodes raster enables another controlled influence to the sensorial characteristics including sensing gas selection. It brings possibility for realisation of intelligent gas sensor array based on simply organometallic units by Figure 6.

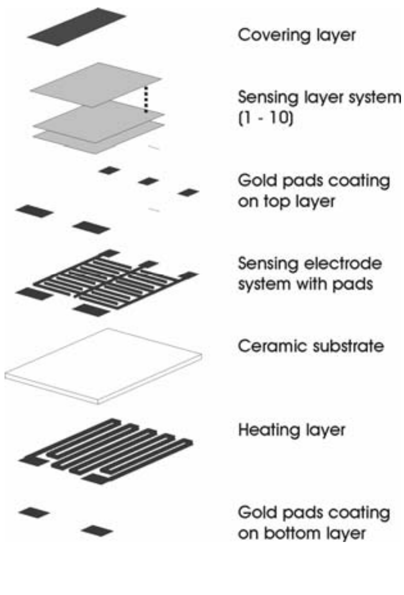


Fig. 6 Schematic design of typical sensor unit.

The ceramic sensor units were mechanically fixed by wire bonding technology in sensor carrier systems prepared by Figure 7. Carriers were made from LTCC which enables to create necessary shapes including the open window for sensor unit assembly. Moreover it enables to use standard thick film technology for realisation of necessary conductive layout for power supply, for the ASIC assembly and/or for realisation of necessary interconnections dedicated for another signal processing. This sensor units assembly solution eliminates the temperature loss and secondary solve the needs to regular electrical connections with additional electronics devices or equipment (ASIC or its computer simulation). Designed ASIC realizes complex of measurements and sensors control which enables using of several sensor substrates in the similar working conditions such as surrounding temperature, humidity, atmospheric pressure and gas concentration. It also enables to monitor and control of the sensor operating temperature influence to the sensing characteristics in the range up to 500°C. This fact brings the advancement with regard to choice of the most appropriate value of operating temperature to correlation between this temperature and sensor sensitivity for scanned gas.

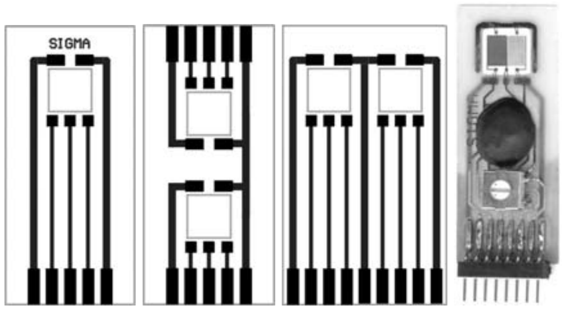


Fig. 7 LTCC sensor carrier systems and demonstration of their utilization .

Another design of sensor array is based on LTCC substrate which enables to prepare multilayer structures with build in heater and both sides of sensor unit can be utilised for gas sensing.

4. CONCLUSION

The LTCC based gas sensors were developed and realized in the frame of research work at Department of Technologies in Electronics FEI TU in Košice. Several sensor versions, including different sensor units based on the resistor and on the conductive meander heater concept, were realized and tested during this work. All of them brought several advantages and on the other hand limitations, too. However the concept with meander heating brought beside the stabilization of operating temperature and sensing characteristics also the possibility of operating temperature evaluation and power consumption saving as long as the temperature field collocation was satisfactory after strong reduction of sensor dimensions, as well.

Particular version for methane sensor design with optimal choice of electrode raster, heater geometry and count of the sensing layers is based on extensive database of measured results. Its sensitivity dependence on operating temperatures and responded sensing characteristic are shown on Figures 8 and 9.

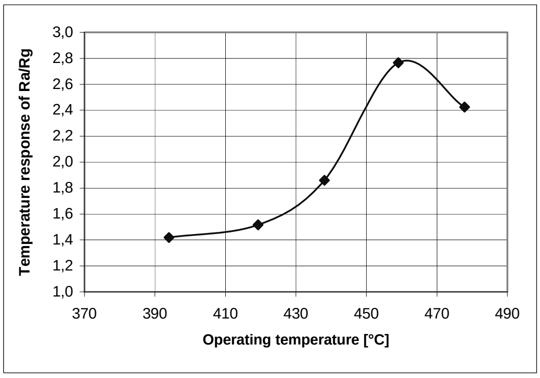


Fig. 8 Dependence of ratio Ra/Rg (sensitivity) on operating temperature at constant gas concentration approx. 2% CH_4.
Ra – reference resistance (covered, protected part),
Rg – resistance at gas surrounding (open part).

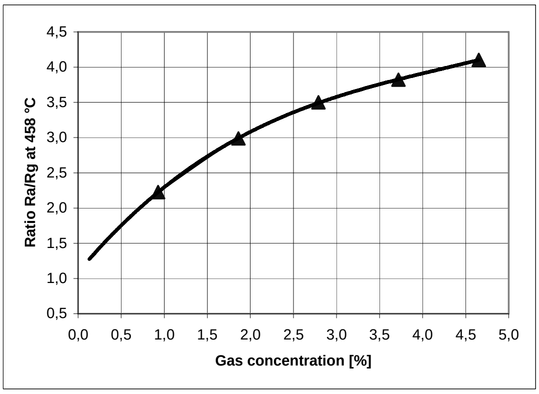


Fig. 9 LTCC sensor carrier systems and demonstration of its utilization .

Exploitation of LTCC qualities and typical features of this technology allow great possibilities for design, development and fabrication of three dimensional structures using multiple layers in numerous applications including sensors production.

In this contribution we tried to show the potential of ceramic tape technology for production of gas sensors with integrated heating systems, as well the possibility to produce gas sensor array. In spite of all their attractive features, the potential of LTCC is far beyond the hybrid technology.

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BIOGRAPHIES

Ján Urbančík, (Ing., PhD.) 1960 – defended his MSc degree at FEI TU Košice in 1984. Afterwards he worked as technologist in industrial praxis. Since 1989 he is working as a researcher at Department of Technologies in Electronics at FEI TU Košice. He defended his PhD. degree in the field of branch 26-35-9 Electrotechnology and Materials in 2004. His present work is oriented to thick film technology with accent to the quality and reliability.

Alena Pietriková (Prof., PhD.) received PhD degree in Material Engineering from the Technical University of Košice in 1986. Since 2007 she has been professor at FEI TU of Košice in the branch of Electrotechnology and Materials. Her research interests include thick film technology and materials for electronics like ceramics, lead free solders and other materials for electronics.

Juraj Banský (Prof., PhD.) graduated at the Slovak Technical University Bratislava, Slovak Republic and received his Ph.D. degree in Physics of Solids. He is author or co-author of the more than 130 scientific papers, research works, books and patents. He spent more than seven years as the scientific worker or Visiting Professor abroad – University of Leeds – England, University of Wuppertal – Germany and Michigan State University – USA. He is the international member of the IMAPS, USA. His professional orientation is concentrated on unconventional applications of the LTCC in sensor's technologies.