Novel tube-type LTCC transducers with buried heaters and inner electrodes for high-temperatures gas sensors

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Abstract

In this work, a fully novel setup for high temperature gas sensor applications is presented. It consists of small tubular devices manufactured in Low Temperature Co-fired Ceramics (LTCC) Technology. It provides buried heaters and temperature sensors - all integrated in the tube - and interdigitated electrodes on the inner side of the tube. The sensitive film is also located inside of the tube, featuring an excellent homogenous temperature distribution on the sensitive film. As an example for a suitable application, a tubular dosimeter-type NO sensor for low level amount detection in varying gas velocities was developed.

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1. Introduction

Gas sensors with electrodes and sensitive coatings on the outer side of a small ceramic tube and a platinum wire heater inside the tube have been serialized already in the 1970’s [1]. Today’s gas sensors are typically realized as planar structures, usually due to the ease of depositing electrodes, heaters, and gas sensitive layers by film techniques. A tubular substrate offers advantages for gas sensing [2]. The symmetrical geometry of the sensor ensures a uniform temperature profile along the gas sensitive layer, the distortion of gas flow in the pipe is reduced, and the analyte-containing gas passes the whole area of the sensitive layer uniformly. For creating of modern tubular substrates the Low Temperature Co-fired Ceramics Technology (LTCC) seems to be most suitable, because of easiness of forming and patterning of
unfired tapes, integration of passive elements inside the structure and possible incorporation of additional elements like channels or cavities. The suitability of LTCC technology for construction of small structures for high temperature applications with integrated buried heater, temperature sensors and electrodes was confirmed in the last years [3-7]. First tubular substrates in LTCC technology were presented in [8-10]. They were equipped with buried heaters, temperature sensors, and with interdigitated electrode structures (IDEs) on the inner tube side. Whereas mentioned area of applications covered rather low-temperature range (drift tube for ion mobility spectrometer, so-called “smart-channels” with polymer chemiresistor), tubular substrates could be successfully used in high-temperature gas sensor technology as well.

2. Tube preparation

In our previous initial study [11], we reported on different methods to produce a tube. The most promising is to wrap the tape around a form, to laminate the wrapped tape to achieve a cylindrical shape, to remove the rod after lamination and to finally fire the structure. This process is depicted in Fig. 1.

![Fig. 1. Technological steps necessary to manufacture a tubular sensor.](image)

The here-presented tubes were made of CT 702 tapes (Heraeus). On the back right side, a platinum heater was printed (LPA 88-11S, Heraeus), while on the front left side, IDEs with 125 μm line/space (TC 7102, Heraeus) were deposited (Fig. 2a). The tape stripes were wrapped on an 8.4 mm alumina rod, covered with metallized polymer foil. After lamination at 14 MPa, the alumina rod and the metallized foil were removed before the structure was fired according to the manufacturer’s recommendation. After wrapping and firing, the heater is buried between the LTCC layers and the IDEs are located inside the tube (Fig. 2b,c). These inner IDEs are abbreviated as IIDE in the following.

![Fig. 2. Tube preparation: a) unfired tape with screen-printed structures: gold IIDE electrodes (top) and platinum heater (bottom); b) fired LTCC tube with buried heater and IIDE electrodes inside (c).](image)

3. Characterization of the buried heater

In the case of high temperature gas sensors applications, a homogenous temperature distribution over the gas sensitive layer is essential for the sensor’s functionality. Therefore, the heater of the tube sensors was FEM-modeled and optimized using Comsol-Multiphysics to ensure a homogenous temperature of the sensitive layer area (Fig. 3). The results were confirmed via an infrared camera (Fig. 3a). The temperature difference over the gas sensitive layer is about ±8°C at 350 °C (Fig. 3b).
Fig. 3. Comparison between a) the simulated (left) and measured (right) temperature distribution over the sensitive layer, b) the temperature profile along the z-axis of the tube.

4. NO gas sensing with the tubular setup

The suitability of the LTCC tubes for high-temperature gas sensing applications was tested using the novel integrating-type sensing principle (also accumulating principle or dosimeter-type sensor) [12,15].

By accumulating NO molecules in a sensitive NO trapping layer, even small NO amounts contribute to the sensor signal, enabling a reliable low level amount detection which is characterized by a fast sensor response and a missing baseline drift [12]. Since the real analyte amount depends on the gas velocity (see [12]), channel-type sensors ensuring that all incoming NO is stored are required in applications with varying flow rates (see [13]). Therefore, tubular structures with small diameters and large-scale sensitive trapping layers are expected to show correct amount accumulating properties. The relative resistance change, \( \Delta R/R_0 \), as the sensor signal of a 46 mm tube sensor with a 120 \( \mu \)m Au-IDE (4.4 x 14.4 mm\(^2\)) coated with a NO\(_x\) trap layer by dip coating (from [16]) upon NO exposure is shown in Fig. 4a for varying gas flows, \( \dot{V} \). The characteristic line (Fig. 4b) correlates \( \Delta R/R_0 \) with the total amount of NO, \( A_{\text{NO}} = \int_{\text{NO}} c_{\text{NO}}(t) \dot{V}(t) dt \), demonstrating full dosimeter-type properties independently on the actual gas velocity with the presented tubular sensor setup.
5. Conclusions

Novel tube-type LTCC transducers with buried heaters and inner interdigital electrodes were developed for high-temperature gas sensors and their suitability in the application of integrating-type sensors was demonstrated. The structures of buried heaters were optimized by modeling and the homogenous temperature distribution was verified by infrared camera. The rotational symmetric gas stream, the homogeneous temperature profile of the sensitive layer, and its close contact to the gas stream is advantageous for the amount detection properties of integrating-type sensors at varying flow rates. The next steps will be on advanced tube-type sensors with multiple sensitive devices at different temperatures.

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