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Low Temperature Co-fired Ceramics Plasma Generator for Atmospheric Pressure Gas Spectroscopy

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Abstract

Accurate and reliable measurements of gas composition are very important in science and industry applications. One way to accomplish this task is to measure spectrum of plasma gas discharge. In our work we propose integrated microcell for optical measurements of gas discharge at atmospheric pressure. This device is fabricated using Low Temperature Co-fired Ceramics (LTCC) technology. Investigated gas is excited in buried air-cavity, placed between two electrodes. Discharge is created using high alternating voltage (30 kV). Optical signal is then transmitted through an integrated optical fiber to the miniature spectrometer. Because different gases have different emission lines, plot of light intensity vs. wavelength provides information about molecular composition of the investigated gas sample. In this work, design, fabrication and performance of the LTCC-based gas discharge chamber is presented. Spectral measurements are performed for air, argon and nitrogen. Moreover, influence of gas flow rate on obtained emission spectrum is discussed.

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Keywords: LTCC; gas sensor; atmospheric pressure plasma; gas spectroscopy

1. Introduction

Plasma is an ionized gas. It has some special properties, which are used in numerous scientific and industrial applications. Some examples can be enumerated, including surface modification, deposition of thin films, etching, ion implantation, activation and functionalization of polymers, emitting light, generation of ozone, improvement of

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material biocompatibility or sample sterilization [1]. Plasmas subdivision can be made according to its ionization degree, pressure of gas and state of thermal equilibrium. If temperature of all plasma species (electrons, ions and neutral species) is the same, the term local thermal equilibrium (LTE) is used. In other case, when the temperature of electrons is higher than the rest of species, plasma is referred as non-LTE. Non-LTE plasmas are used in applications, where heat is not desirable [2].

Condition for plasma generation is included in Paschen rule. It states, that breakdown voltage of gas depends on product of pressure and distance between electrodes [3]. The breakdown voltage decreases with decreasing gas pressure and interelectrode distance reaching a minimum. Therefore, if the distance between electrodes is relatively small the plasma can be created at high pressure. It is one of motivation for miniaturization of such devices. Some constructions of miniature plasma generators (e.g. [4-5]) can be found in the literature.

One of that was presented in [6] and was made in LTCC technology. It consisted of nine 250 μm thick (in green state) tapes. Distance between electrodes was approximately 200 μm (after firing). Its final dimension was 17.4 x 26.1 mm^2. Plasma was created between electrodes excited by the RF power supply system (13.56 MHz and maximum 200 W). The pressure of working gas was in the range of 1 to 4 Torr (ca 133 and 533 Pa, respectively). The authors have measured spectra of a few gases. However, the relatively low pressure on the generator operation significantly limits its possible application for gas characterization.

Increasing attention has recently been put on gas chromatography based on optical emission spectrometry [7]. In our work, we present a miniature LTCC-based plasma generator for gas spectroscopy, which utilizes discharge in gas at atmospheric pressure. We describe construction, fabrication process of this device with results of measurements for selected gases and different flow rates.

2. Construction and fabrication

2.1. Plasma generator design

The structure of the plasma generator consisted of ten LTCC layers. Their layout is presented in Fig. 1. On each layer four structures were placed. In layers 1 and 2 inlets for a gas were cut. Bottom electrodes for plasma generation and tees were made on layer 3. Via holes for gas transportation are cut in layer 4. They connect with plasma chamber made in layers 5 – 8.

Empty volumes in layers 5 – 8 fulfill few functions. A groove at the left middle side of the layer was formed for the alignment of an optical fiber, which guides optical signal from the plasma chamber to the spectrometer. Channels above and below it connects the inlet with plasma chamber. Gas outlet is formed on the right side of the layers 5 – 8.

Second electrode is placed on layer 9. It is important to notice, that both electrodes are separated from the plasma chamber by a single layer of ceramic. The structure was sealed with layer 10, which mechanically supported structure and separates top electrode from the environment. After firing dimensions of the structure were 17.3 x 17.3 x 2.3 mm^3. Height of the plasma chamber was 0.85 mm, and the distance between the electrodes was 1.28 mm.

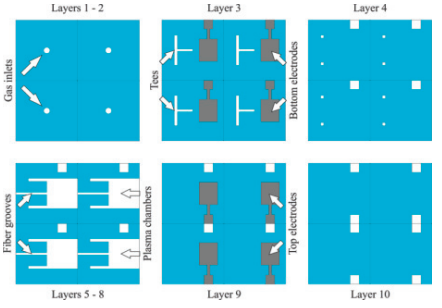

Fig. 1. Schematic view of structure layers.

2.2. Fabrication

For the device manufacturing we chose DuPont 951PX tape system [8]. Firstly, the LTCC sheets were cut into pieces with size appropriate for making orifices for stacking and lamination using UV laser system (LPKF ProtoLaser U). Then, the silver/palladium electrodes were deposited using screen-printing method.

In the next step, layers 1 – 2 and 5 – 8, were separately laminated. We conducted an isostatic lamination using recommended by the tape manufacturer process parameters. After that in all layers appropriate shapes were cut using UV laser system.

Raw LTCC material with air cavities is very susceptible for deflections during the lamination process. To avoid sagging of structures the sacrificial volume material (SVM) was used. After SVM deposition (ESL 49000 carbon tape), all LTCC layers were laminated using an isostatic press.

Finally, the ceramic laminate was cut into single structures and fired. In order to ensure complete burnout of the SVM the firing profile was modified. For rapid temperature changes intensive oxidizing of carbon could deflect or even destroy ceramic structures. Gas connectors (Upchurch N-333) and silica optical fibers (57-064, Edmund Optics) were glued to the final structures. Exemplary LTCC-based plasma generator is presented in Fig. 2.a.

3. Results and discussion

Performance of the fabricated LTCC-based plasma generator was examined experimentally. It was connected to the AC power supply (30 kHz frequency and approximately 30 kV amplitude) and the gaseous discharge plasma at atmospheric pressure was created (see Fig. 2.a). The optical signal from excited gas molecules and ions was transmitted from the plasma chamber to the compact BlueWave-spectrometer (StellarNet) via optical fiber. Measurements were provided in the range from 340 nm to 1160 nm. Emission spectra recorded for air, argon and nitrogen at atmospheric pressure are presented in Fig 2.b. The obtained emission spectra for LTCC-based plasma generator were comparable with the ones presented in the literature [6, 9, 10, 11].

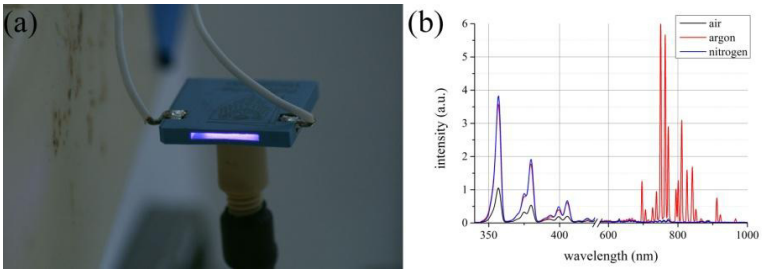

Fig. 2. (a) plasma generation in atmospheric pressure; (b) spectra of different gases.

In the next experiment, the emission spectra were taken for different gases at various flow rates (2, 4 and 8 dm^3/min). Generally, for certain gas light intensity decreases for higher flow rate. It is probably caused by more intense removing of the ionized molecules from plasma chamber by the gas pressure. As a result less gas molecules can take part in the avalanche process. Rate of this descent depends on the gas type. In the Fig. 3. spectra recorded for air are presented. The recorded peaks for air plasma in the range from 350 to 450 nm are characteristic for nitrogen at high pressure. The oxygen emission can be observed for a higher wavelengths (500-800 nm). However, intensity of oxygen emission is relatively low in comparison with nitrogen or argon. Therefore, presence of the oxygen in the gas mixture results with decreasing of the emission spectrum intensity. This decrease is proportional to the oxygen content in the gas mixture. In Fig. 4 are presented results for maximum emission intensity for argon/oxygen mixture measured at 670 nm. One may observe, that emission intensity for fixed flow rate decreases exponentially with increasing content of oxygen in a gas mixture ($R^2 = 0.999$). For higher flow rate R^2 coefficient

was worse than 0.66. It means, that measuring emission intensity at a single wavelength, for relatively low flow rate, we can obtain information about content of oxygen in the mixture.

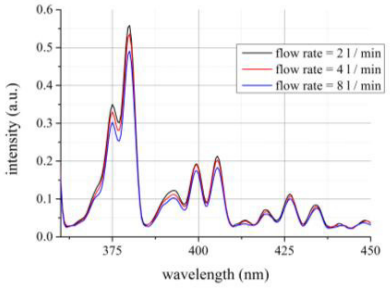

Fig. 3. Spectra of air discharge for different flow rates.

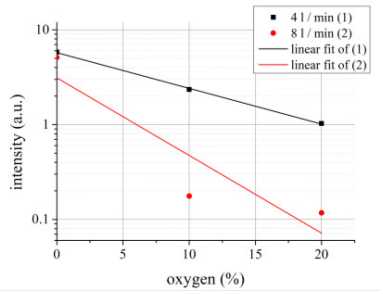

Fig. 4. Intensity at 670 nm for different air flow and oxygen content

4. Conclusions

Fabrication and properties of a novel miniature LTCC plasma generator for atmospheric gas spectroscopy was described. The applied technology allowed fabricating multilayer ceramic structures with plasma chamber placed between two embedded electrodes and microgroove for an optical fiber.

Fabricated device was examined experimentally. Performed experiments have shown that generation of gas discharge plasma at atmospheric pressure is possible. Plasma ignition inside the microchamber was achieved for different gases (argon, nitrogen, air) and flow rates (2, 4 and 8 dm^3/min). Moreover, the emission spectra of the gases were measured and results were similar to those known from literature. It was presented, that content of single, known gas can be measured as well as mixtures with different species.

There are still some problems to solve. Longer usage of the device causes overheating of the LTCC structure. The optical connection is very sensible to mechanical stresses. Future work will focus on optimization of the device construction and AC power supply as well as more accurate calibration.

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