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Miniature plasma generator made of low temperature co-fired ceramics (LTCC) for gas characterization using optical emission spectroscopy (OES)

Abstract. A novel miniature plasma generator made of low temperature co-fired ceramics (LTCC) is presented in this paper. The developed generator is composed of a stack of 9 ceramic tapes, has an optical fibre integrated into the structure and is consisted of an 8.7 x 3.5 mm² plasma chamber placed between two 5 x 5 mm² electrodes made of AgPd. Each electrode is separated from the plasma chamber by a single LTCC tape, forming a 660 µm thick gap. The shape of the plasma chamber and the channel for the optical fibre were cut in green LTCC tapes using an UV laser, and the electrodes were fabricated with the standard screen-print method. During the experiments, the plasma chamber was filled with an ambient air. The plasma was generated between AgPd electrodes connected to an AC power supply. The light of the air plasma was transmitted from the plasma chamber to the miniature spectrometer using the integrated optical fibre. The glow discharge in the air at atmospheric pressure was characterized by optical emission spectroscopy (OES).

Streszczenie. W artykule przedstawiono technologię miniaturowego generatora plazmy. Wspomniany układ został wykonany za pomocą techniki bazującej na niskotemperaturowej współwypalanej ceramice LTCC (Low Temperature Co-fired Ceramics). Urządzenie składało się z 9 warstw ceramiki LTCC. W skład opracowanego generatora wchodziły komora plazmowa o wymiarach 8,7 x 3,5 mm² oraz dołączony do niej światłowód kwarcowy. Komora plazmowa umieszczona była pomiędzy dwiema elektrodami o wymiarach 5 x 5 mm² wykonanymi ze stopu PdAg. Każda z elektrod została odizolowana od komory plazmowej za pomocą pojedynczej warstwy LTCC tworząc szczelinę o grubości 660 µm. Kształt komory plazmowej oraz kanału pod światłowód zostały wycięte w surowych foliach ceramicznych za pomocą lasera UV. Elektrody PdAg zostały naniesione na ceramikę LTCC metodą sitodruku. Podczas eksperymentów komora plazmowe wypełniona była powietrzem z otoczenia o ciśnieniu atmosferycznym. Plazma powietrza generowana była pomiędzy dwiema izolowanymi elektrodami zasilanymi napięciem zmiennym. Promieniowanie optyczne plazmy powietrza było transmitowane z komory plazmowej do miniaturowego spektrometru za pomocą zintegrowanego światłowodu. Obserwowane wykonany techniką LTCC do charakteryzacji gazów za pomocą optycznej spektroskopii emisyjnej (OES).

Keywords: low temperature co-fired ceramics (LTCC), plasma, air, optical emission spectroscopy (OES). Słowa kluczowe: niskotemperaturowa ceramika współwypalana (LTCC), plazma, powietrze, optyczna spektroskopia emisyjna (OES).

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Introduction

The low temperature co-fired ceramics (LTCC) technology was developed in the '80s, during the 20th century. It was one of the technologies used for the production of Ceramic Multichip Modules (MCM-C). The MCM-C is a multilayer structure which is capable of supporting several chips in one package [1]. It can be composed of several layers, mechanically and electrically connected by a network of internal and external conductors and passives. Active components can be mounted on a top and/or bottom surface of the module using the SMT (Surface Mounting Technique) or flip-chip method [2]. Thanks to these advantages, the LTCC technology is mainly used for the production of high volume microwave devices. In additional to three-dimensional electrical connections, channels (e.g. for fluid transportation) and other spatial structures can also be fabricated inside the LTCC module. Thus, the LTCC technology was recently applied to the production of sensors, actuators, microsystems, MEMS (Micro-Electro-Mechanical System) and MOEMS (Micro-Opto-Electro-Mechanical System) The LTCC technology enables the package [3-5]. fabrication of miniature devices consisting of electrical and fluidic components, and is especially suitable for the making of plasma generators. Small plasma generators are able to operate at higher pressure, when compared to those used in classical plasma systems [6]. Miniature devices which can generate plasma at atmospheric pressure make possible the reduction of equipment cost by avoiding the necessity for an expensive vacuum system. Atmospheric pressure plasma can be used for, e.g. surface modification, the cleaning of materials, or gas detection [7-10].

In this paper, a new miniature plasma generator for gas characterization using Optical Emission Spectroscopy (OES) is presented. The device operates at atmospheric pressure. It is fabricated using LTCC technology. The threedimensional model of the miniature plasma generator is presented in Fig. 1.



Fig.1. Three-dimensional model of the plasma generator

The LTCC-based plasma generator consists of a chamber located between two isolated electrodes and incorporates an integrated optical fibre for spectroscopic measurements. The chamber, the channel for the optical fibre, and the electrode terminals were cut in green LTCC tapes by UV laser. The electrodes were made of conductive silverpalladium (AgPd) paste with the screen-print method inside the LTCC module. To minimize the impact of plasma on electrodes sputtering, each electrode was isolated from the plasma chamber with a single LTCC layer with a thickness of ca. 200 µm. The performance of the fabricated LTCCbased plasma generator was examined experimentally. Measurements were performed for an ambient air at an atmospheric pressure. The air plasma was ignited by applying AC voltage to the electrodes. The light emitted by the plasma was transmitted via optical fibre to the miniature spectrometer and the spectrum of the air plasma was recorded and analyzed.

Materials and methods

The DP951 PX LTCC tape was used to fabricate the miniature plasma generator. The thickness of a single tape in a green state was equal to 254 μ m. The device was composed of 9 LTCC layers. All the ceramic layers of the miniature plasma generator are presented in Fig. 2.



Fig.2. The LTCC layers for the miniature plasma generator: a) bottom layers, b) bottom electrode layer, c) layers for plasma chamber and channel for the optical fibre, d) upper electrode layer, e) top layers

The two bottom layers (a) consist of cuts for the bottom electrode's terminal, and define the base of the miniature plasma generator. The bottom electrode is deposited on a single layer (b). The plasma chamber and channel for the optical fibre are cut into three layers (c) and they are closed with another layer (d). On the top surface of the other layer (d), a second electrode is deposited and is sealed with two layers (e). CAD software was used to design the features of each layer. The plasma generator has the designed dimensions of 16 x 19 mm^2 with a working area (where plasma is ignited) of 5 x 5 mm^2 (before firing). The plasma chamber, channel for the optical fibre, and electrode terminals were made using an UV laser operating at λ_{max} = 355 nm (LPKF Protolaser U). The electrodes were made from AgPd thick-film paste (DP 6146) using a standard screen-printing method, through a 325 mesh steel screen. After laser cutting and screen-printing, all LTCC layers were stacked together in the proper order and laminated using an

isostatic press. In order to prevent the deformation of the plasma chamber and the channel for the optical fibre, the lamination process was modified. The multi-step lamination process, combined with the application of the SVM (sacrificial volume material), is presented in Fig 3. In the first step, similar layers were laminated together (Fig. 2a, c, e) with a pressure of 20 MPa, at a temperature of 70°C for 10 min. The laminated layers, defining the plasma chamber and the channel for the optical fibre, were then sealed using the laver with the deposited bottom electrode (Fig. 2b). The second lamination was performed with a relatively low pressure of 2 MPa, at a temperature of 70°C for 10 min. In the next step, the plasma chamber and the channel for the optical fibre were filled with carbon paste. The applied paste worked as a sacrificial volume material. Finally, all LTCC layers were pressed together with a standard pressure of 20 MPa, at a temperature of 70°C for 10 min. The LTCC laminate was co-fired in a box furnace in ambient air using the two-step thermal profile recommended by DuPont®. The peak temperature was set to 875°C. The fired LTCCbased miniature plasma generator structure is presented in Fig. 4.



Fig.4. The LTCC-based miniature plasma generator

After the co-firing process, the silica UV/Vis optical fibre (57-064, Edmund Optics), with a core diameter of 600 μ m and NA = 0.22, was precisely positioned inside the channel and glued to the fired LTCC structure. The optical fibre collected light from the plasma and transmitted it to the computer controlled spectrometer.



Fig.3. Scheme of the modified lamination process

Results and discussion

The characterization of the fabricated miniature plasma generator began from geometrical measurements. The final

LTCC structure was flat, without visible deformations or delaminations. To avoid sagging or contraction of the plasma chamber and the channel for the optical fibre, the SVM was applied. The resulting spatial structures had a rectangular cross-section. The dimensions of the fired LTCC structure were approximately 13.9 x 16.8 x 2 mm³. Therefore, the shrinkage rate was correlatively 13 % in *x*, *y* and *z* directions.

After geometrical characterization, the LTCC-based plasma generator was connected to the miniature spectrometer (BLUE-Wave, StellarNet) via an optical fibre. The AC power supply, operating at 70 kHz and at a power of 5 W, was connected to the electrodes of the miniature plasma generator. The experiment was performed in atmospheric pressure and room temperature. After the connection of the AC power supply, the air plasma glowed intensely inside the chamber (Fig. 5). The air plasma did not expand from the LTCC-based generator, as can be seen in Fig. 5b.





The obtained air plasma was characterized using optical emission spectroscopy. The optical fibre collected light from the chamber and transmitted it to a computer controlled miniature spectrometer. The spectrometer had the spectral resolution of 3.5 nm and operated in the wavelength range of 300 to 1100 nm. Select regions of the recorded emission spectra for air plasma are presented in Fig. 6.

The recorded emission lines for air plasma were characterized by high intensities in the spectral range from 300 to 425 nm. According to the referenced literature [11], this can be interpreted as the vibrational transition from the second positive system of N₂, and the first negative system of N₂⁺. A distinct band with a maximum at ca. 355 nm is characteristic for nitrogen at high pressure. Weaker emission was observed in the 625 to 775 nm wavelength range. Two distinct bands can be noticed. The maximum of the first band appears at ca. 665 nm. It, most likely, can be assigned to a transition in the first negative system of N₂⁺ molecular ion. Moreover, the neutral atomic oxygen line can be noticed at ca. 775 nm [12]. However, it must be

noted that the data within the referenced literature refer to much higher volumes of gas than is available in our experiment. Taking this into account, the interpretation should be treated as a preliminary, particularly in the range of lower emission intensities (600 nm - 800 nm).



Fig.6. Chosen regions of the optical emission spectrum recorded in two wavelength ranges: (a) 325 - 500 nm and (b) 600 - 800 nm (for air at atmospheric pressure).

Conclusions

A novel miniature plasma generator was presented. This device was fabricated using well known LTCC microelectronic technology. The LTCC tapes were cut with a laser system to fabricate spatial structures (plasma chamber and channel for the optical fibre). The electrodes were made of AgPd using the screen-printing method. Deformation and delamination of the structure was prevented by modifying the lamination process. Modification was based on a multi-step lamination process, combined with the application of a sacrificial volume material. The fabricated LTCC-based plasma generator was then used to ignite air plasma.

The optical emission spectrum of the air plasma was recorded by a computer controlled spectrometer connected to the LTCC-based plasma generator via the optical fibre. The recorded emission spectrum was similar to those which were observed in the referenced literature, including high intensity emission lines related to nitrogen and oxygen.

The performed preliminary experiments have demonstrated that the presented miniature plasma generator shows potential in successful application as an integrated part of a more sophisticated system for the characterization of the gases using optical emission spectroscopy. The authors wish to thank National Science Centre (DEC-2013/09/D/ST7/03953) and Wrocław University of Technology (S30066) for financial support.

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