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# ELECTRICAL AND OPTICAL PROPERTIES OF AC MICRODISCHARGES IN POROUS CERAMICS

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**ABSTRACT.** Generation of microdischarges inside porous ceramics by AC high voltage has been investigated. Electrical and optical measurements were performed to explore the physical properties of the microdischarges. The effect of pore size, discharge power and gas mixture on the discharge properties and development is described.

# **1. INTRODUCTION**

Research of various types of non-thermal plasmas (i.e. plasmas with electron temperature elevated with respect to the gas temperature) generated by electrical discharges at atmospheric pressure has received a fast development in the last two decades. The environmental applications of such non-thermal plasmas are based on their plasma-chemical effects, where generated electrons, ions, excited species and radicals initiate processes that occur neither in classical chemistry, nor in thermal plasmas. Streamer and pulsed coronas, and various types of dielectric or ferroelectric barrier discharges are mostly used. They are typical with nonequilibrium character and a large amount of thin filamentary channels, called microdischarges. The microdischarges produce a large density of energetic electrons and free radicals at relatively low energy consumption; therefore they represent a potential method for car exhaust cleaning [1-3]. The exhaust cleaning effect can be even enhanced when the discharge plasma is combined with catalysts. The catalyst, typically of ferroelectric bed type or pellets or of honeycomb structure, can be placed either behind the plasma zone or directly in it. In comparison with plasma-only flue gas treatment systems, the plasma-assisted catalysis performs 0-order kinetics (thanks to the heterogeneous catalytic surface reactions), excellent carbon balance, and minimal aerosol production [4-7]. The pressure drop on the catalyst appearing at high gas flow rates and a small volume of the generated plasma associated with the pellet bed type plasma reactors leads to a more prospective use of porous materials, such as honeycomb catalysts or porous ceramics.

In the recent years, there appeared a few pioneering works dealing with the generation of microdischarges in narrow cavities and capillaries of porous dielectric materials with the aim to investigate their physical properties and use them for the removal of volatile organic compounds and nitrogen oxides [8-10]. Unlike most of pulsed discharges that need a special high voltage power supplies generating pulses with a rise time of some tens of nanoseconds in

order to efficiently produce radicals and oxidants, microdischarges can be generated even with an AC or a DC high voltage power supply [11-13]. This fact substantially enlarges the possibilities of the microdischarge use in porous materials because high investment costs for the construction of a pulsed power supply are reduced.

The objective of the paper was to investigate the physical properties of the microdischarges generated inside porous ceramics by AC high voltage power supply. In the previous works we described the properties of the microdischarges generated using DC power [11, 13]. The methods of the investigations in the presented work consisted of electric and optical measurements focused on determining the optimal conditions of the discharge generation with main respect to the pore size.

#### 2. EXPERIMENTAL SET-UP

The experimental set-up consisting of a discharge reactor and electric and optical circuits is depicted in *Figure 1*. A porous ceramics was placed between two stainless steel mesh electrodes. The ceramics were sintered from alumina or fused silica and their diameter and thickness were 31 and 7 mm, respectively. The pore size of the ceramics was 2, 10, 30, 80 (alumina) and 50, 120, 200  $\mu$ m (silica). The ceramics was set inside the quartz cylinder sealed by silicon stoppers at the both ends. The discharge reactor was placed in a Faraday cage to reduce induced noise signals and correctly measure voltage and current waveforms.

AC regulated high voltage power supply connected via a 5 M $\Omega$  series resistor limiting the total discharge current was used to excite the discharge reactor. The total power including the power losses in the electrical circuit was measured by digital multimeter Metex 3860M. The voltage at the reactor was measured by a high voltage probe Tektronix P6015A and the discharge current was measured using a current probe Pearson Electronics 2877 (1V/A) linked to the digitizing oscilloscope Tektronix TDS1012 (100 MHz, 1 GS/s).



FIGURE 1. Experimental set-up.

Emission spectroscopy optical system consisted of a dual fiber-optic compact spectrometer Ocean Optics SD2000 with CCD detector used for fast scanning in the UV and VIS-NIR region (200-500 and 400-1050 nm). The photographs of discharge were taken by the digital camera Nikon E4300 with manually adjustable aperture and exposure time.

All experiments were carried out in atmospheric pressure nitrogen, oxygen or air at room temperature. The pressure drop across the discharge reactor was measured by the digital manometer PCE P-30.

#### 3. RESULTS AND DISCUSSION

The effect of the pore size, discharge power, and gas mixture on the properties and development of the discharge was investigated means of electrical and optical by measurements. The pore size of the used ceramics was a critical parameter for selecting a total gas flow rate. The smaller the pore size, the bigger was the pressure drop across the reactor. Figure 2 shows the dependence of the pressure drop on the gas flow rate for various pore sizes. For the tests, we have used the gas flow rate of 0.4 l/min, as the pressure drop at this flow rate was almost negligible (< 200 Pa) for all ceramics, except for the one with 2  $\mu$ m pores.



**FIGURE 2.** Pressure drop across the reactor as a function of gas flow for various pore sizes.

Generation of microdischarges inside the ceramics was possible only at the specific discharge power depending on the pore size of the ceramics. In the previous studies, we reported that at a small applied voltage, the surface discharge development over the surface of the ceramics could be observed. It mainly occurred with the ceramics of the smallest pore size. By the increase of the applied voltage, however, a gradual change of the discharge mode was observed. At a certain voltage, the surface discharge "leaked into" the ceramics and the microdischarges inside the material were formed. Figure 3 represents the instant values of the applied voltage and the total power in the moment of the surface discharge transition to microdischarges related to the pore size. As the figure shows, the voltage and the power at the onset of the microdischarges decreases with the pore size. Figure 4 displays the total powerto-voltage (P-U) characteristics of the microdischarges. For better readability of the figure, the results of three ceramics are presented only. The P-U characteristics of the surface discharge for all ceramics were almost identical. After the transition to microdischarges, the slope of the P-U characteristics increased. The slope of the characteristics increased with the pore size. The results presented in the Figure 3 and 4 are in the agreement with those obtained for microdischarges generated by DC power [11-13].



**FIGURE 3.** Total power and applied voltage in the moment of transition to microdischarges.



**FIGURE 4.** Total power as a function of discharge voltage for the selected pore size (in air).

We also recorded waveforms of both the applied voltage and the current pulses. *Figure 5* presents the waveforms obtained for 30  $\mu$ m ceramics at the various timescales. The discharges occurred in both negative and positive polarities of the applied high voltage. The amplitude of the current pulses increases with the applied voltage and the power and reached several tenths of amps (*Fig. 6*). For a given power, the maximal amplitude of the current pulses was observed with 50 and 80  $\mu$ m pore size ceramics (*Fig. 7*).



**FIGURE 5.** Voltage and current waveforms of the discharge in porous ceramics at the different timescales (pore size 30  $\mu$ m, U = 16.2 kV, P = 23 W, air).



**FIGURE 6.** Current pulse amplitude as a function of applied voltage (in air).

**FIGURE 7.** Current pulse amplitude as a function of pore size for a different power (in air).

The optical characteristics of the discharge were recorded simultaneously with their electric characteristics. *Figures 8-10* presents three sequences of photographs visualizing the effect of the power, the gas and the pore size on the discharge character and spatial development. All photographs were taken with the same exposition time. The light emission of a single microdischarge channel was not constant in time, as well as, the spatial distribution of the channels randomly changed, too. The figures show that the light intensity increased with the discharge voltage and the power. It is also evident, that the increase of the oxygen contents in the gas mixture resulted into redistribution of the microdischarge channels inside the ceramics. In oxygen rich mixtures, the channels concentrated mainly around the outer circumference of the mesh electrodes and almost no emission from the center the ceramics was observed. On the other hand, in nitrogen the light emission was relatively homogenously distributed over the whole surface of the ceramics. The oxygen contents cause the number of discharge channels decreased resulting in a decrease of the discharge power for a given voltage.



**FIGURE 8.** The effect of power (left to right): 17, 19, 21, 23, 26 W (pore size 80  $\mu$ m, exposure time 1s, f-number f/3.5, air).



**FIGURE 9.** The effect of gas (left to right):  $O_2$ , air,  $N_2$  (pore size 80  $\mu$ m, exposure time 1s, f-number f/3.5, P = 22 W).



**FIGURE 10.** The effect of pore size (left to right): 10, 30, 80, 120  $\mu$ m (exposure time 1s, f-number f/3.5, P = 20 W, air).

The emission spectroscopy was employed as a powerful technique of plasma diagnostics to give us valuable information on excited atomic and molecular states and the insight in

ongoing plasma chemistry. Figure presents 11 the emission spectra of microdischarges in VIS-NIR region taken at the constant power various in gas mixtures. In nitrogen and air, two principal systems of bands have been observed. The  $2^{nd}$  positive system of  $N_2$ violet in the region corresponding to the transition  $C^{3}\Pi_{u} - B^{3}\Pi_{g}$  of N<sub>2</sub> excited states, and the 1<sup>st</sup> positive system of N<sub>2</sub> in the visible orange-red region  $\begin{array}{ll} corresponding & to \\ transition \ B^{3}\Pi_{g} - A^{3}\Sigma_{u}^{\ +}. \end{array}$ the



**FIGURE 11.** Emission spectra of microdischarges in VIS in various gas mixtures (pore size  $80 \ \mu m$ , P =  $30 \ W$ ).

Significant changes in emission spectra depending on the  $O_2$  content and many other unidentified bands have been observed (502, 518, 568, 695, 718, 795 nm, etc.). They may belong to the bands of Fe (electrodes) and/or to various atomic spectra of N, O, NO,  $O_3$ , etc. Their interpretation, however, needs further investigation.

### 4. CONCLUSIONS

Electrical and optical properties of microdischarges generated inside the porous ceramics by AC high voltage power have been investigated. The effect of the pore size, discharge power and gas mixture on the discharge properties and development was described. It was found the onset voltage of the microdischarges decreases with the pore size, while that the slope of P-U characteristics slope increases with the pore size. Amplitude of the current pulses increased with the applied voltage. The maximal amplitude was observed for 50 and 80  $\mu$ m pore size ceramics. In nitrogen, the light emission was relatively homogenously distributed over the whole surface of the ceramics and a decrease of the discharge power was observed. Emission spectroscopy also showed significant changes in emission spectra depending on the O<sub>2</sub> content. In total, the optimal generation of the microdischarges and their distribution in the ceramics was observed namely for 30-80  $\mu$ m pore sizes. The discharge formation inside porous ceramics presents a novel way to generate large volume stable atmospheric pressure plasmas in hybrid plasma–catalyst reactors and can be effectively used for flue gas treatment.

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# REFERENCES

- [1] Oda T, Kato T, Takahashi T, Shimizu K (1998) IEEE Trans. Industry Appl. 34, 268-272.
- [2] Yamamoto T, Okubo M, Nagaoka T, Hayakawa K (2002) IEEE Trans. Ind. Appl. 38, 1168-1173.
- [3] Kim H-H, Prieto G, Takashima K, Katsura S, Mizuno A (2002) J. Electrostat. 55, 25-41.
- [4] Hammer T. (2002) Plasma Sources Sci. Technol. 11, 1.
- [5] Holzer F, Roland U, Kopinke F-D (2002) Appl. Catal. B: Environ. 38, 163-181.
- [6] Kim H-H (2004) Plasma Process. Polym. 1, 91-110.
- [7] Penetrante BM, Hsiao MC, Bardsley JN et al. (1997) *Plasma Sources Sci. Technol.* 6, 251-259.
- [8] Gherardi N, Gouda G, Gat E, Ricard A, Massines F (2000) *Plasma Sources Sci. Technol.* 9, 340-346.
- [9] Blin-Simiand N, Tardiveau P, Risacher A, Jorand F, Pasquiers S (2005) *Plasma Proc. Polym.* 2, 256-262.
- [10] Jidenko N, Petit M, Borra J-P (2006) J. Phys. D: Appl. Phys. 39, 281-293.
- [11] Hensel K, Katsura S, Mizuno A (2005) IEEE Trans. Plasma Sci., 33, 574-575.
- [12] Sawada J, Matsui Y, Hensel K, Koyamoto I, Takashima K, Katsura S, Mizuno A(2004) "Microdischarge in Porous Ceramics for Exhaust Gas Cleaning", 5<sup>th</sup> International Conference on Applied Electrostatics ICAES 2004, Shanghai, November 2-5, 128-131.
- [13] Hensel K, Matsui Y, Katsura S, Mizuno A (2004) Czech. J. Phys., 54, C683-C689.