Ruhr-University Bochum



ISPC 19

International Symposium on Plasma Chemistry



edited by A. von Keudell, J. Winter M. Böke, V. Schulz-von der Gathen

> Ruhr-University Bochum Germany July 27 – 31, 2009

> www.ispc-conference.org

Generation of Discharges inside the Honeycomb Monolith Assisted by Diffuse Coplanar Surface Barrier Discharge

K. Hensel¹, M. Janda¹, J. Ráhel²

¹Department of Astronomy, Earth Physics and Meteorology, Comenius University (FMFI UK), Bratislava, Slovakia ²Department of Experimental Physics, Comenius University (FMFI UK), Bratislava, Slovakia *e-mail: hensel@fmph.uniba.sk*

Abstract: The discharge generated inside the bundle of quartz capillaries that emulated the honeycomb monolith structure was investigated. The diffuse plasma generated by the diffuse coplanar surface barrier discharge was extended into the capillaries by the application of the additional DC electric field component across the capillaries. The proposed technique resulted in a homogenous plasma formation inside the capillaries. The paper presents essential electrical and optical characteristics of the discharges inside capillaries and describes basic discharge properties.

Keywords: diffuse coplanar surface barrier discharge, corona discharge, honeycomb catalyst, optical emission spectroscopy

1. Introduction

Air pollution generated from mobile sources is a problem of general interest. Burning of hydrocarbon in the car engine ideally leads to the formation of water and carbon dioxide however, due to non-perfect combustion control and the high temperatures reached in the combustion chamber, the exhaust contains significant amounts of pollutants which need to be transformed into harmless compounds. Automotive catalytic converters have been developed and used for abatement of pollutants emitted by various vehicles for more than a quarter of a century. Despite their high efficiency, legislation limits for automobile emissions require still new and new technologies to be applied in order to meet the current emission standards. The present catalytic converts must have high activity (up to 98%) and selectivity even at low temperatures and high thermal stability. The major problems of the catalysts consist of their low activity at low temperatures and performance in non-stiochiometric conditions. Several techniques have been applied to overcome the low temperature problem including hydrocarbon adsorbing trap, electrical or chemically heated catalysts placed upstream of the main catalysts system, closely coupled catalysts, along the effort to develop new catalysts [1].

In the past, we have proposed an idea to solve some of these problems by combination of the catalyst with non-thermal plasma generated by electric discharges. In the proposed system the plasma is generated inside capillary channels of the honeycomb-shaped catalytic monolith. Generation of homogenous and stable plasma directly inside thin and long channels was rather difficult, due to low discharge stability and mechanical breakdown of the washcoat caused by random discharge sparking. Therefore instead of generating the discharge/plasma directly inside the body of the catalysts (in the capillary channels), we first generated auxiliary discharge outside and subsequently extended it into the catalytic monolith. In the past, we have demonstrated this technique by generating such auxiliary AC driven discharge inside a packed pellet bed, which was then extended into the capillary channels by the application of the additional DC electric field component across the monolith [2, 3].

In this paper we present a similar system capable to generate the stable and homogenous plasma inside the channels of the honeycomb monolith. Instead of discharge in the pellet bed, a diffuse coplanar surface barrier discharge (DCSBD) [4] was used as the auxiliary discharge. The DCSBD generates a thin layer of macroscopically uniform plasma over the surface of dielectric barrier with the higher number density of charged particles comparing to the standard volume dielectric barrier discharge. The role of the DCSBD in the presented system was to supply a sufficient amount of seeds (positive ions) to ionize the volume inside the capillary channels.

The paper presents essential electrical and optical characteristics of the discharges generated inside the capillaries, describes basic discharge properties and addresses the effects of applied voltage, discharge power and dimensions of the capillaries.

2. Experimental Setup

Schematic drawing of the discharge reactor is depicted in Fig. 1. The system consisted of alumina (Al₂O₃) made DCSBD discharge panel and a quartz tube ($\emptyset = 26$ mm) packed with a bunch of quartz capillaries ($\emptyset = 1$; 2 mm, L = 2 cm), which was positioned on the top of alumina barrier. The transparent capillaries were used instead of ceramic honeycomb monolith in order to be able to visually observe the discharge.

System of electrodes consisted of those embedded in-



Fig.1. Experimental system including discharge reactor and electrical and optical diagnostics.

side the DCSBD alumina panel, aluminum foil wrapped around the quartz tube and metal mesh placed on the top of the capillaries. The DCSBD electrode was made by 15 pairs of 2 mm wide silver strip electrodes embedded 0.5 mm below the surface of 96% Al_2O_3 ceramics. Mutual distance of silver strip electrodes was 1 mm. The panel was energized by 14 kHz sinusoidal voltage, supplied by HV generator (Lifetech VF700). The power input to the discharge panel was 400 W/200 cm². The mesh electrode was powered by negative DC high voltage, and the foil was grounded.

The electrical parameters of discharges were monitored by the current monitor (Pearson Electronics 2877) and high voltage probes (Tektronix P6015A) and the signals were recorded by the oscilloscope (Tektronix TDS2024).

Optical measurements were performed by a dual fiber-optic compact UV-VIS emission spectrometer (Ocean Optics SD2000). The optical system consisted of lenses, which were set up to be able to record the discharge emission along the axis of the capillaries. The images of the discharge were recorded by a digital camera (Nikon D40x). The experiments were performed in atmospheric pressure ambient air and at room temperature.

3. Results and Discussion

The DCSBD discharge produced diffuse homogenous plasma on the surface of the ceramic panel. The discharge



Fig.2. Images of the discharge [capillary diameter 2 mm, length 2 cm] in ambient air: DCSBD ON $[P_{AC} = 250 \text{ W}]$ (left); DCSBD ON $[P_{AC} = 250 \text{ W}] + \text{DC ON } [U_{DC} = -17 \text{ kV}]$ (right).



Fig.3. Discharge current waveform: DCSBD ON [$P_{AC} = 250$ W] (left); DCSBD ON [$P_{AC} = 250$ W] + DC ON [$U_{DC} = -17$ kV] (right).

was used as an auxiliary source of plasma to ionize the space inside the capillaries. Upon the application of DC voltage across the capillaries placed on the surface of the DCSBD discharge panel a gradual development of a stable streamer discharge inside them was observed. In the end, the plasma generated by the DCSBD was successfully extended into the capillaries. The mechanism of the discharge formation inside the capillaries can be understood as a superposition of the barrier discharge and the DC corona discharge. The first one works a plasma electrode, producing charged particles and ionic space charges. The latter one produces and maintains an ionic wind toward the DC electrode.

Figure 2 shows the photographs of the discharge. The image on the left displays the case when only the DCSBD was turned on. The emission from the capillaries was rather limited and no visible light was coming out from the middle of the capillaries. The situation changed when DC component was applied across the capillaries. As the figure of the right shows, in this case an increase of the light emission intensity inside the capillaries could be clearly recognized. The image also shows the distribution of the discharge and the emission inside the capillaries was relatively homogenous.

Figure 3 shows the typical current waveforms of the discharge inside the capillaries and corresponding to the images in Fig.2. The left figure represents the current waveform measured by the current probe on grounded electrode when only the DCSBD discharge was applied. The other waveform shows the case when also DC component was applied, where additional pulses with much higher amplitudes can be recognized. The pulses occurred in the phase with the maxima of the applied voltage. They were the result of the discharge formation and propagation inside the capillaries.



Fig.4. Amplitude of the current pulses as functions of the DC applied voltage and the DCSBD power $[\emptyset \ 2 \ mm]$.



Fig.5. Emission intensity as functions of the DC applied voltage and the DCSBD power $[\emptyset \ 2 \ mm]$.

Figure 4 shows the amplitude of these current pulses as a function of the DC applied voltage. The amplitude of the pulses was found increasing with the DC applied voltage, but on the other hand found independent of the power of the DCSBD. It follows that the density and homogeneity of the plasma generated by the DCSBD does not play the most important role for the discharge generation inside the capillaries. The result corresponds to the findings obtained in the similar configuration, where the pellet bed was used instead of the DCSBD panel [2].

By increasing the amplitude of DC voltage, stable and homogenous plasma is formed inside the channels. Figure 5 shows the discharge emission intensity as function of DC applied voltage. The emission intensity reflects a concentration of active species and can be considered as a measure of plasma chemical activity. The intensity increased with both the applied DC voltage, as well as, with



Fig.6. Emission intensity as a function of the vertical position $[P_{AC} = 250 \text{ W}].$



Fig.7. Emission intensity as a function of the vertical position $[P_{AC} = 250 \text{ W}].$

the power of the DCSBD, although the effect of the latter was almost negligible. Besides the measuring the emission intensity in the middle of the capillaries, the measurements of the emission along the axis of the capillary were also performed. The axial profile of the emission was recorded to evaluate the level of the plasma lateral homogeneity and the discharge formation inside the capillary channels.

Figures 6 and 7 shows axial profiles of the emission intensity produced by the discharges inside the capillaries. The vertical position in the figures is defined as a distance from the surface of the DCSBD discharge panel. The figures show that the emission intensity decreased with increasing distance from the panel. The result supports the original idea that the discharge inside capillaries is formed from the plasma generated on the surface, which is extended into the capillaries. There it dies out with the distance from the panel just like in afterglow. The decrease of the intensity at close vicinity of the panel (vertical distance $0\sim1$ mm) is caused by the method of the measurement. In this case, the cone of the light focused by the lens is reduced due to the presence of the boundary, the DCSBD panel, which does not contribute to the absolute value of the emission intensity. By comparing the capillaries of different diameter, the emission intensity was found increasing with the diameter. For smaller diameter the volume of the plasma inside the capillaries is smaller. As the emission intensity is proportional to the volume of the plasma, in this respect the obtained result can be well understood.

Regarding the stability of the discharge inside the capillaries, it was found the stable operation is possible within a wide interval of both DCSBD and DC applied voltages. The transition into a spark discharge was observed when too high DC applied voltages was used. The DC sparking voltage decreased with increasing the power of the DCSBD and vice versa. The stability of the discharge can be controlled and the transition to a spark suppressed by using series resistance in the electric circuit (usually several M Ω).

4. Conclusions

Generation of discharge inside spatially confined volume inside the glass capillaries emulating honeycomb monolith structure by using the DCSBD was presented. The discharge inside capillaries was generated by the superposition of DCSBD coupled in series with DC powered honeycomb monolith. The paper introduced the properties of the discharge based on electrical and optical measurements. The homogeneity and the stability of the discharge can be controlled by amplitude and power of the individual power supplies. The presented discharge generates relatively cold plasma with a high level of non-equilibrium.

The research has been supported by the Slovak Research and Development Agency, projects No. APVV-0485-06, APVV-0267-06 and the Slovak Grant Agency VEGA 1/0711/09. The authors thank T. Homola and M. Leštinský for technical assistance during experiments.

References

- J. Kašpar, P. Fornasiero, N. Hickey, Catal. Today 77, 419 (2003).
- [2] K. Hensel, S. Sato, A. Mizuno, IEEE Trans. Plasma Sci. 38, 1282 (2008).
- [3] S. Sato, K. Hensel, H. Hayashi, K. Takashima, A. Mizuno, J. Electrostat. 67 (2-3), 77-83 (2009).
- [4] M. Šimor, J. Ráhel', P. Vojtek, M. Černák, Appl. Phys. Letters 81, 2716 (2002).