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DISCHARGE FORMATION INSIDE THE HONEYCOMB STRUCTURES ASSISTED BY SURFACE BARRIER DISCHARGE

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In this paper, we experimentally investigated discharge formation inside the honeycomb catalysts. Honeycomb structure of the catalyst was simulated by a bundle of glass capillary tubes. The discharge inside the capillary tubes was formed with the assistance of surface barrier discharge generated by a perforated ceramic substrate and by applying DC high voltage across the capillaries. We evaluated basic electrical and optical characteristics of the discharge using ambient or synthetic air as carrier gases with various flow rates. The effect of polarity of the applied DC high voltage was also studied. The results showed the positive effects of air humidity and air flow rate on stability and quality of the discharge inside capillary tubes. With negative DC high voltage, the highest emission intensity of the capillary discharge was observed for air flow rate of 0.5 L/min, while with positive DC high voltage, it was for air flow rates of 1.0 and 2.4 L/min. These results and the other details related to properties of the surface barrier discharge will be presented during the conference.

1. Introduction

Plasma catalysis is gaining an increasing interest in many environmental applications, e.g. air and water pollution control. It is characterised by high catalyst selectivity as well as high plasma reactivity which can lead to higher pollutant removal efficiencies with lower energy consumption when compared to individual catalytic or plasma processes [1, 2]. In the catalytic processes, besides the catalyst composition also its shape and structure play a crucial role [3]. In plasma catalytic systems, the catalyst can be present as a coating or as a layer on the plasma reactor walls or electrodes, or it can take the form of powder, beads or pellets packed in the reactor volume. In addition to these packed bed reactors, also honeycomb catalysts are sometimes used as they have an advantage of low pressure drop and high surface-to-volume ratio. However, generation of stable and homogeneous non-thermal streamer discharge plasma inside thin and long channels of the honeycomb is relatively difficult. Formation of plasma requires high onset voltages due to losses of charged particles by interaction with the channel walls [4] and it is often unstable and associated with frequent sparking that is undesirable due to possible mechanical damage of ceramics and even with a respect to some applications.

In order to understand plasma discharge generation and propagation inside the honeycomb structures, the processes have been studied by numerical modelling by various research groups [5–7]. *Zhang et al.* found, that in the case when streamer is initiated along the dielectric surface (i.e. wall of honeycomb channel), the discharge will be further enhanced due to surface charging along the dielectric surface [5]. *Jánsky et al.* found, that discharge propagation velocity inside the thin tube increases with the increase of the applied voltage and with the decrease in both the tube radius and relative permittivity [6, 7]. In addition to modelling, experimental investigations of plasma discharge formation in honeycomb catalyst have been also studied [8–12]. In these works, the honeycomb structure was simulated by a bundle of glass capillary tubes. The discharge inside capillaries (also called honeycomb or capillary discharge) was generated by a three-electrode geometry. Firstly, discharge plasma was formed by auxiliary pellet bed discharge [8–10] or surface barrier discharge [11, 12]. Secondly, the plasma of auxiliary discharge was extended into the honeycomb structure upon the application of DC high voltage (HV) across the capillaries. *Hensel et al.* and *Sato et al.* found, that onset DC HV amplitude increases with the length and decreases with the diameter of capillaries. Further, with extending the length of capillaries at a given DC voltage, the discharge current and power decreases. In contrast, the diameter of the capillaries (1-2 mm) showed marginal effect on the discharge power [8, 9]. The average electric field strength for the onset of the capillary discharge was about 10 kV/cm for capillaries with 2.0 mm in inner diameter and 20 mm in length [10]. In addition to

investigation of basic discharge properties, experiments focused on regeneration of diesel particulate filter [13] or removal of NO_x from simulated diesel exhaust gas [14, 15] have been also performed.

The objective of this work was to study electrical and optical characteristics of the discharge formed inside honeycomb structure which was simulated by a bundle of glass capillaries. In the past, we investigated the discharge generation assisted by a packed bed discharge [8, 9] or a diffuse coplanar surface barrier discharge (DCSBD) [12]. Here, the capillary discharge was formed with the assistance of surface barrier discharge (SBD) generated by a perforated ceramic substrate (also called as a plasma actuator). The effects of air humidity, air flow rate and polarity of DC HV applied across the capillaries were investigated on quality (stability) and homogeneity of the capillary discharge.

2. Experimental setup and methods

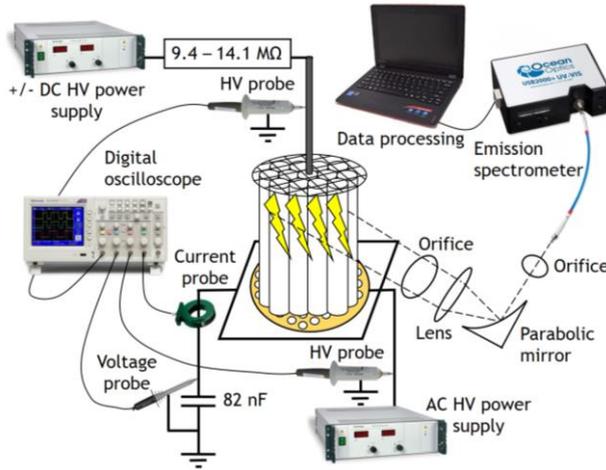


Fig. 1. Experimental setup.

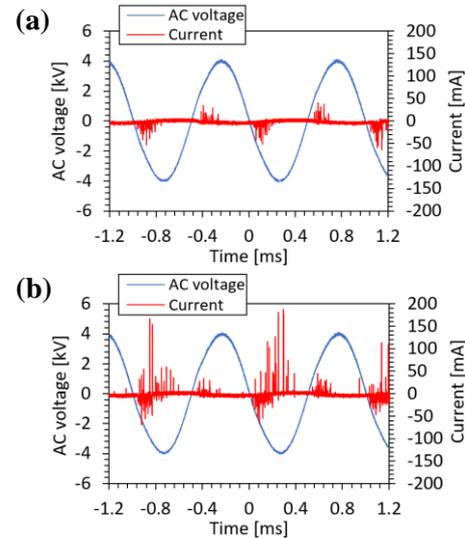


Fig. 2. Voltage and current waveforms of (a) surface barrier discharge (AC HV 4 kV @ 1 kHz) and (b) capillary discharge (AC HV 4 kV @ 1 kHz; DC HV +16 kV; R 9.4 MΩ).

The experimental setup is depicted in the Fig. 1. The ceramic substrate (*Kyocera*) with the dimensions of 50 x 50 x 1 mm and perforated by 170 holes with an inner diameter of 1.5 mm, consisted of one Ni/Au electrode embedded within the ceramic and the other one printed on the ceramic surface. The substrate was powered by AC HV power supply consisting of function generator (*Gwinstek SFG-1013*), signal amplifier (*Omnitronic PAP-350*) and high voltage transformer. The SBD was formed by applying AC HV to the air-exposed electrode (3–7 kV at 1 kHz) while the embedded electrode was grounded. The bundle of glass capillaries was placed perpendicularly on the ceramic substrate, while a metal mesh serving as a third electrode was placed on the top of them and was powered by DC HV power supply (*Technix SR20-R-1200*). The waveforms of the applied AC and DC voltages were measured by HV probes (*Tektronix P6015A*) and the discharge current pulses were measured by a current probe (*Pearson Electronics 2877*) connected to a digital oscilloscope (*Tektronix TDS2024C*). The power consumption of the SBD was evaluated using the Lissajous figure method [16] with an 82 nF capacitor and a voltage probe (*Tektronix P2220*). An optical emission spectroscopy system consisted of dual-fibre optic spectrometer (*Ocean Optics SD2000*), optic fibre, two orifices, parabolic mirror and lens. Photographs of the discharge were taken with a digital camera (*Sony Alpha DSLR-A230*) with manually adjustable aperture and exposure. Ambient and synthetic air were used as the carrier gases and their flow rate (0.5, 1.0 and 2.4 L/min) was controlled by flow meters. A relative humidity of the ambient air was approx. 60%.

3. Results and discussion

The Fig. 2. (a) shows typical voltage and current waveform of the SBD while the Fig. 2. (b) shows the waveform recorded during the operation of both SBD and the capillary discharge. In contrast to the

waveform of SBD, formation of capillary discharge can be recognized by the presence of current pulses of much bigger amplitude during the maximum amplitude of the AC applied voltage (Fig. 2. (b)) that occurred only when both AC and DC HV were applied. When positive DC HV was used, capillary discharge formed only during the negative half-period of the applied AC voltage and vice-versa, because of the biggest potential difference (approx. 20 kV) across the capillaries.

The Fig. 3. shows the plasma ceramic substrate with glass capillaries in different conditions: (a) without applied HV; (b) with applied only AC HV and (c) with applied both AC and DC HV. The photographs in the Figs. 3. (b) and (c) correspond to voltage and current waveforms in the Figs. 2. (a) and (b), respectively. In the Fig. 3. (b), light was emitted by the SBD only, while upon application of DC HV across the capillaries, the streamer propagation formed the homogeneous capillary discharge inside them (Fig. 3. (c)). On the other hand, upon application of only DC HV without the assistance of SBD, the discharge in capillaries had significantly lower emission intensity and was localised only inside one or two capillaries. The mechanism of capillary discharge formation can be explained by a superposition of the AC powered surface barrier discharge and the DC powered streamer corona discharge. The first one serves as an ionizer producing charged particles while the latter one produces and maintains ionic wind toward the DC electrode [9].

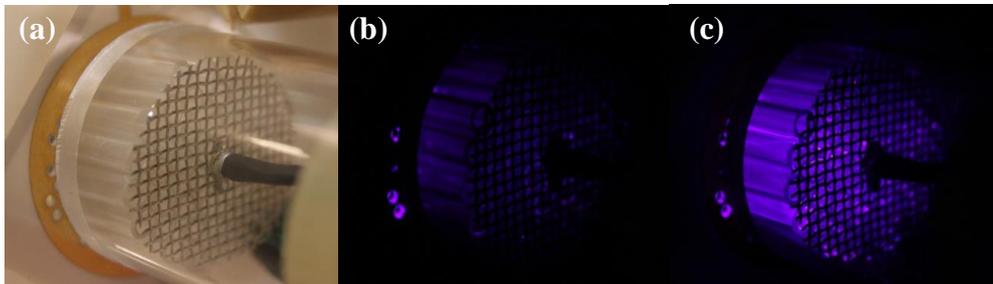


Fig. 3. The ceramic substrate with glass capillaries: (a) without discharge; (b) with applied AC HV (4 kV @ 1 kHz); (c) with applied both AC and DC HV (4 kV @ 1 kHz; +16 kV, respectively) (ambient air, 2.4 L/min) (Exposure time 4 s, f/5.6, ISO 400).

We evaluated the quality (stability, activity) of the capillary discharge by measuring its light emission intensity. Higher emission intensity reflects higher number of stable discharges maintained in the streamer discharge regime. The emission intensity is also a measure of discharge activity, i.e. concentration of active species generated by the discharge. Possible instability of the discharge is associated with its transition from the streamer to the spark regime, what is usually undesirable in practical applications. The discharge emission intensity was evaluated based on the 0–1 spectral band (357 nm) of 2nd positive system of N₂ instead of 0–0 spectral band because of strong attenuation of the intensity of this spectral band occurred when light passed through the glass capillaries.

The Fig. 4. shows the comparison of emission intensity of the capillary discharge in ambient and synthetic air with various air flow rates in the case of negative (Fig. 4. (a)) and positive DC HV (Fig. 4. (b)). The positive effect on the discharge quality was observed with the increasing of the air humidity. When dry synthetic air was used as a carrier gas, the capillary discharge did not form, regardless of air flow rate and the polarity of the DC HV. On the other hand, when humid ambient air was used, a stable discharge dominant in terms of emission intensity was observed – for negative DC HV with flow rate of 0.5 L/min (Fig. 4 (a)) and for positive DC HV with flow rates of 1.0 and 2.4 L/min (Fig. 4 (b)). The positive effect of the air humidity on the capillary discharge quality and stability was also reported in [9, 15].

We also studied the effect of air flow direction on quality (stability) of the capillary discharge. The best result was obtained in case when the air passed firstly through the ceramic substrate and then through the bundle of capillaries. In such case, the air was preionised by the SBD and discharge inside the capillaries was ignited with lower DC HV amplitudes. When air flow direction was reversed, the discharge remained localised especially near to the mesh electrode, its emission intensity was lower and was quite unstable with occasional sparking.

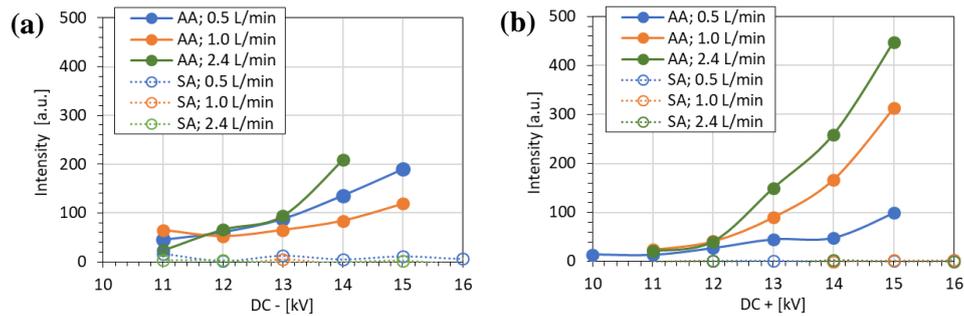


Fig. 4. Emission intensity of the capillary discharge as a function of amplitude of the applied (a) negative or (b) positive DC HV in ambient air (AA) and synthetic air (SA) with various flow rates (0.5; 1.0 and 2.4 L/min).

4. Conclusion

In this paper, we experimentally studied formation of plasma discharge inside the honeycomb structure simulated by a bundle of glass capillary tubes. The discharge inside the capillaries was formed by a superposition of AC powered surface barrier discharge coupled in series with DC high voltage applied across the capillaries. The capillary discharge was maintained in the streamer discharge regime and positively supported and stabilised by increasing the air humidity. The effect of polarity of the DC high voltage, air flow rate and its direction were also studied. With negative DC high voltage, the highest emission intensity of the capillary discharge was observed for air flow rate of 0.5 L/min, while with positive DC HV, it was for air flow rate of 1.0 and 2.4 L/min. In general, we found that air humidity and air flow rate have positive effect on homogeneity, quality and stability of the capillary discharge. Further investigation focused on long-term stability and chemical activity of the capillary discharge is needed in order to determine the optimal conditions of the capillary discharge operation with the respect to eventual applications.

5. Acknowledgements

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