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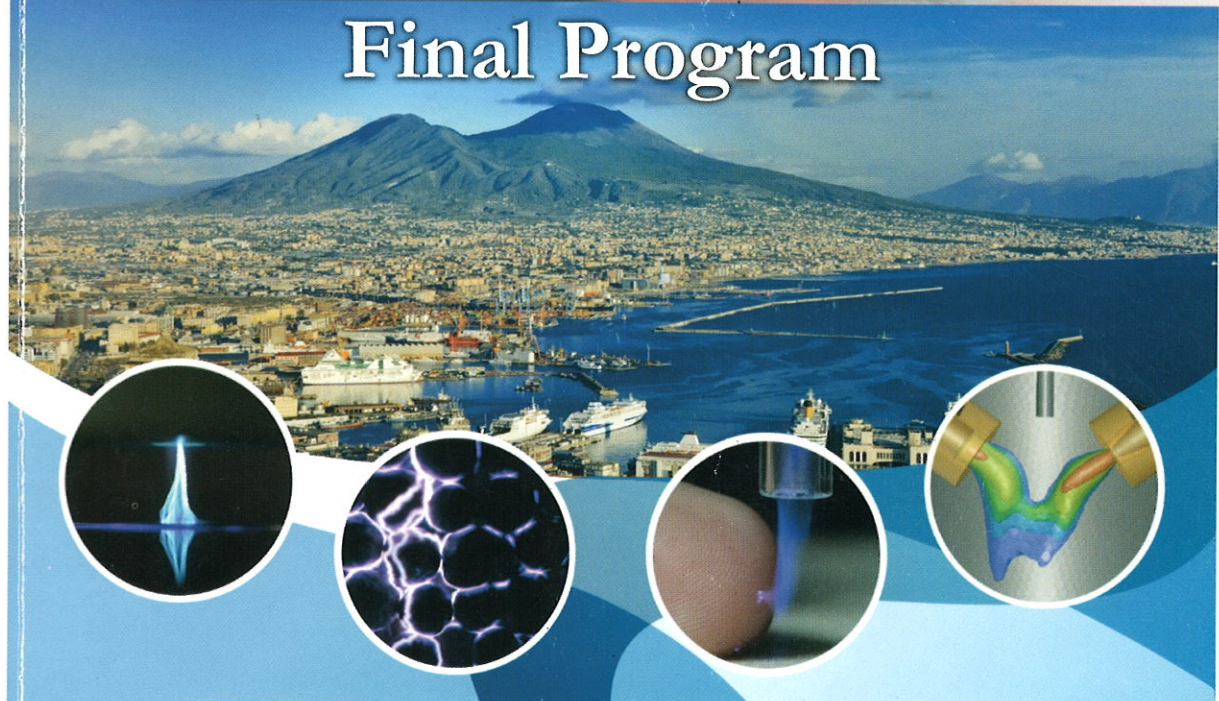
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Discharge formation inside the honeycomb structures assisted by surface barrier discharge

R. Cimerman, E. Maťaš and K. Hensel

Faculty of Mathematics, Physics and Informatics, Comenius University,
Mlynská Dolina F2, 842 48, Bratislava, Slovakia

Abstract: The objective of this study was to experimentally investigate discharge formation inside the honeycomb catalysts. Honeycomb-shaped 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 surface barrier discharge and capillary discharge as well.

Keywords: surface barrier discharge, capillary discharge, honeycomb catalyst

1. Introduction

In systems combining non-thermal plasma with catalysis, 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 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 [1] 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 of plasma discharge formation in honeycomb catalyst have been studied by numerical modelling [2–4] and also experimentally [5–9]. 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 in a three-electrode geometry. Firstly, discharge plasma was formed by auxiliary pellet bed discharge [5–7] or surface barrier discharge [8, 9]. Secondly, this plasma was extended into the honeycomb structure upon the application of DC high voltage (HV) across the capillaries. In addition to investigation of capillary discharge basic properties, experiments focused on regeneration of diesel particulate filter [10] or removal of NOx from simulated diesel exhaust gas [11, 12] have been also performed.

The objective of this work was to study electrical and optical characteristics of the discharge formed inside honeycomb structure simulated by a bundle of glass capillaries and its plasma chemical effects. In the past, we investigated the discharge generation assisted by a packed bed discharge [5, 6] or a diffuse coplanar surface barrier discharge (DCSBD) [9]. Here, the capillary discharge was formed with the assistance of surface barrier discharge (SBD) generated by two types of perforated ceramic substrate (also called plasma actuators [13–15]). The effects of the applied voltage, air humidity and air flow rate on characteristics of SBD as well as capillary discharge were studied. Moreover, the effect of the polarity of DC HV applied across the capillaries were investigated on quality (stability) and homogeneity of the capillary discharge. Plasma chemical effects of the capillary discharge were also briefly evaluated.

2. Experimental setup and methods

The experimental setup is depicted in the Fig. 1. The ceramic substrates (*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 two electrodes. The first substrate (S1) consisted of one Ni/Au electrode embedded within the ceramic and the other electrode printed on the ceramic surface (air-exposed electrode), while the second substrate (S2) had both electrodes embedded inside the ceramic. The substrates were 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 one electrode (3 – 7 kV at 1 kHz) while the other one was grounded. In the case of the S1, HV was applied to the air-exposed electrode. The bundle of glass capillaries was placed inside the quartz glass tube perpendicularly to the ceramic substrates, while a metal mesh serving as a third electrode was placed on the top of capillaries and powered by DC HV power supply (*Technix SR20-R-1200*). The bundle consisted of 48 capillary tubes of 20 mm in length and 2.8 mm in diameter. 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 cylindrical lens. Photographs of the discharge were taken with a digital camera (*Sony Alpha DSLR-A230)* with manually adjustable aperture and exposure. Synthetic air was used as the carrier gas and its flow rate (0.5, 1 and 2.4 L/min) was controlled by flow meter. The air passed through a water cell and its relative humidity (RH) was controlled (0 – 80%) and monitored by electrochemical probe (*Arduino*). Chemical effects of the discharges in the gas phase were evaluated by means of FTIR spectroscopy (*Shimadzu IR-Affinity 1S*). Ozone production was also evaluated by UV absorption at 254 nm.
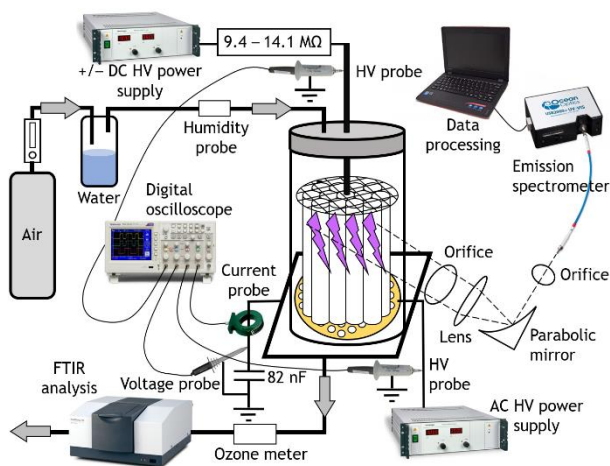

Fig. 1. Experimental setup.

3. Results and discussion
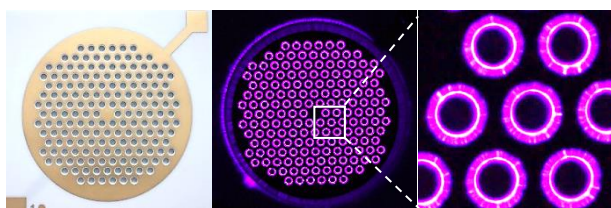### 3.1 Surface barrier discharge

Fig. 2. The surface barrier discharge generated by ceramic substrate with the air-exposed electrode (S1) (AC 4 kV @ 1 kHz) [Exposure time 3 s, f/5.6, ISO 400].

The Fig. 2. shows the SBD generated by the S1, while the Fig. 3. (a, b) shows the electrical characteristics of the SBD (discharge power and amplitudes of both positive and negative current pulses) as a function of applied AC HV amplitude for dry (Fig. 3. (a)) and humid air (RH 60%) (Fig. 3. (b)). The trend in amplitudes of the current pulses with the increasing AC HV was not uniform. In dry air (Fig. 3. (a)), the amplitudes initially increased (at 2 – 4 kV), then reached a maximum (at 4 – 5 kV) and finally decreased (at 6 – 7 kV). The effect of air humidity on the electrical characteristics of the SBD was found significant. When humid air was used as a carrier gas, the discharge power and amplitudes of the current pulses were much smaller in contrast to dry air. On the other hand, the effect of air flow rate on the electrical characteristics of the SBD was found negligible.

Further, from the recorded SBD voltage and current waveforms, the discharge behaved differently during the positive and negative half-periods of the applied voltage. Amplitudes of the positive current pulses (up to 260 mA) were usually found higher than amplitudes of negative current pulses (up to 150 mA). Similarly, the number of positive current pulses was significantly higher than that of negative current pulses, up to 450 and 100 pulses, respectively. Therefore, the positive half-period of the applied voltage corresponds to a corona-like discharge, while during the negative half-period of the applied voltage, the current trace evokes a glow-like regime [13, 14].

The electrical and optical characteristics of the SBD generated by the S2 were different to that of S1. The emission intensity of the SBD generated by the S2 was found substantially lower accompanied with low amplitudes and number of the current pulses when compared to that of S1 at the same applied voltage.
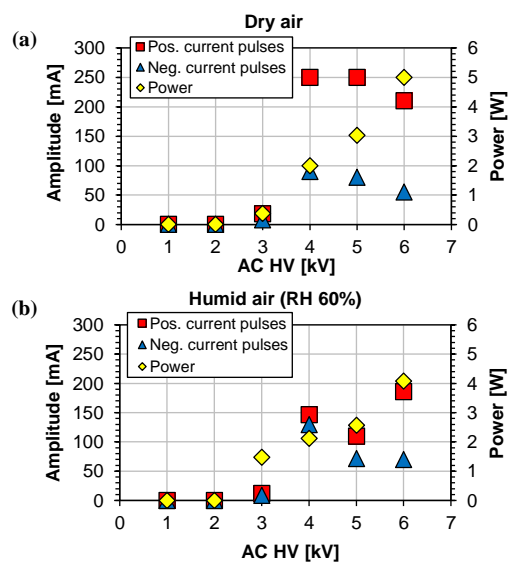

Fig. 3. Amplitude of the current pulses and discharge power of the SBD (S1) vs. amplitude of the applied AC HV @ 1 kHz in (a) dry air and (b) humid air (RH 60%) with flow rate of 2.4 L/min.

3.2 Capillary discharge

Generation of stable and homogeneous streamer discharge plasma inside glass capillary tubes requires assistance of the SBD along with application DC HV across the capillaries. Thus, the mechanism of capillary

discharge formation can be explained by a superposition of the AC powered SBD 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 [6]. When positive DC HV was used, the capillary discharge was formed only during the negative half-period of the applied AC HV and vice-vesa, because of the biggest potential difference across the capillaries. Furthermore, the generation of capillary discharge was performed only using the SBD generated by the ceramic substrate with the air-exposed electrode (S1). When the S2 was used, capillary discharge did not form at all.

The Fig. 4. (a – d) shows the photographs of S1 with bundle of glass capillaries in different conditions: (a) without applied HV; (b) with only AC HV applied, (c) with both AC and DC HV applied, and (d) with only DC HV applied. In the Fig. 4. (b), the 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. 4. (c)). On the other hand, upon application of only DC HV without the assistance of the SBD, the capillary discharge had significantly lower emission intensity and was localised only in the vicinity of the DC powered mesh electrode (Fig. 4. (d)).
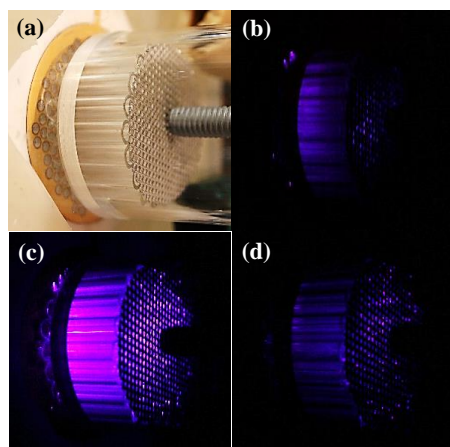

Fig. 4. The ceramic substrate (S1) with glass capillaries: (a) without discharge; (b) with only AC HV applied (4 kV @ 1 kHz); (c) with both AC and DC HV applied (4 kV @ 1 kHz; +14 kV, respectively); (d) with only DC HV applied (+14 kV) (humid air (RH 60%), 2.4 L/min) [Exposure time 8 s, f/5.6, ISO 400].

Evaluation of the capillary discharge quality (stability, activity, homogeneity) was performed 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 N_2 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. 5. (a, b) shows the comparison of emission intensity of the capillary discharge in air with various relative humidity RH (0 – 80%) in the case of negative (Fig. 5. (a)) and positive DC HV (Fig. 5. (b)). The positive effect on the discharge quality was observed with the increasing of air humidity and air flow rate. 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 air was used, a stable discharge dominant in terms of emission intensity was observed. The positive effect of the air humidity on the capillary discharge quality and stability was also reported in [6, 12]. Further, the emission intensity of the capillary discharge was found higher when positive DC HV was applied in contrast to negative DC HV (Fig. 5. (a, b)).
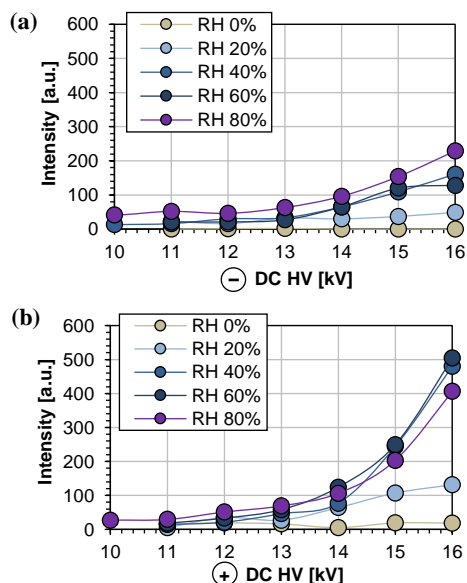

Fig. 5. Emission intensity of the capillary discharge as a function of the applied (a) negative and (b) positive DC HV in air with relative humidity (RH) of 0 – 80% (AC 4 kV @ 1 kHz, 2.4 L/min).

A brief evaluation of the plasma chemical effects of the capillary discharge in the gas phase was performed. It was mainly focused on the production of various gaseous species, e.g. ozone and nitrogen oxides. Production of ozone by capillary discharge was found higher than that of SBD in the same conditions. More details will be presented during the symposium.

4. Conclusion

In this work, 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 (SBD) coupled in series with DC HV applied across the capillaries. Firstly, we investigated basic physical characteristics of the two SBD ceramic substrates. While the discharge power, amplitudes and number of both positive and negative current pulses changed only slightly with the air flow rate, the effect of air humidity was found significantly higher. Secondly, we studied the electrical and optical characteristics of the capillary discharge that was maintained in the streamer discharge regime and positively supported and stabilised by increasing of the air humidity. The effect of polarity of the DC HV, air flow rate and its direction were also studied. Emission intensity of the capillary discharge was found higher when positive DC HV was used in contrast to negative DC HV. In general, we found that air humidity and partially air flow rate have positive effect on homogeneity, quality and stability of the capillary discharge. The results showed, that combination of SBD generated by the ceramic substrate with air-exposed electrode coupled in series with DC HV applied across the honeycomb structure can be potentially used for generation of stable streamer discharge plasma inside honeycomb-shaped catalysts. Brief examination of the plasma chemical effects of the capillary discharge shown an improvement when compared to the plasma chemical effects of the SBD alone. Further investigation focused on long-term stability and chemical activity of the capillary/honeycomb discharge is needed in order to determine the optimal conditions of the discharge operation with the respect to eventual applications.

5. Acknowledgement

6. References

[1] H. Cheng et al., Plasma Process. Polym., **12**, (2015).
[2] Q. Z. Zhang and A. Bogaerts, Plasma Sources Sci. Technol., **27**, (2018).
[3] J. Jansky et al., J. Phys. D: Appl. Phys., **43**, (2010).
[4] J. Jansky et al., J. Phys. D: Appl. Phys., **44**, (2011).
[5] K. Hensel et al., IEEE Trans. Plasma Sci., **36**, (2008).
[6] K. Hensel, Eur. Phys. J. D., **54**, (2009).
[7] S. Sato et al., J. Electrostat., **67**, (2009).
[8] A. Mizuno, Catal. Today, **211**, (2013).
[9] K. Hensel et al., ISPC-19 Ruhr-University Bochum, Germany, (2009).
[10] H. Hayashi et al., Int. J. Plasma Environ. Sci. Technol., **6**, (2012).
[11] S. Sato and A. Mizuno, Int. J. Plasma Environ. Sci. Technol., **4**, (2010).
[12] K. Takashima et al., Int. J. Plasma Environ. Sci. Technol., **7**, (2013).
[13] N. Benard et al., Appl. Phys. Lett., **107**, (2015).
[14] N. Benard et al., J. Electrostat., **88**, (2017).
[15] G. Nayak et al., Plasma Sources Sci. Technol., **26**, (2017).
[16] U. Kogelschatz, Plasma Chem. Plasma Process., **23**, (2003).