Journal of Electrostatics 67 (2009) 77–83

Contents lists available at ScienceDirect

Journal of Electrostatics

journal homepage: www.elsevier.com/locate/elstat

Honeycomb discharge for diesel exhaust cleaning

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ARTICLE INFO

Article history:
Received 15 September 2008
Received in revised form
22 January 2009
Accepted 1 February 2009
Available online 15 February 2009

Keywords:
Electrical discharge
Non-thermal plasma
Honeycomb catalyst
Diesel exhaust
DeNO$_x$

ABSTRACT

Selectivity and energy efficiency of plasma chemical processes can be significantly improved if plasma is combined with a catalyst. The generation of stable discharge plasma inside the narrow capillaries of an automobile catalytic honeycomb is, however, quite difficult. This paper presents a novel method which utilizes the combination of packed-bed or barrier discharge connected in series with the honeycomb capillaries. By the application of DC voltage across the capillaries, streamers are extended from the packed-bed discharge into the capillaries. With this method, ionization can be made inside fine channels of honeycomb catalyst made of insulating materials. This discharge is designated as "honeycomb discharge". Electrical and optical characteristics of the honeycomb discharge are measured.

1. Introduction

Various types of atmospheric pressure discharges have been developed for electrostatic processes and for plasma chemical processes, such as removing particles, cleaning exhaust gas and volatile organic compounds(VOCs), etc. [1–4]. In order to improve the selectivity and energy efficiency of plasma chemical processes, a combination of plasma and catalyst is effective. For example, catalyst pellets can be used in the packed bed discharge to improve removal efficiency of nitrogen oxides and VOCs [5,6]. Honeycomb is a commonly used geometry of catalysts. It has been difficult to generate electrical discharge evenly inside a honeycomb [3]. If discharge is generated inside a honeycomb, a larger surface area can be obtained with a lower pressure drop for improved chemical reactions.

Several important works have been reported recently. One is the superposition of surface discharge and AC discharge to obtain a large discharge volume [5]. Surface discharge is generated on the inner wall of a cylinder, and AC voltage is applied between the centered electrode and the electrode placed on the inner wall for the surface discharge. The other is a sliding discharge that has been used to cover the large surface area of the wings of airplanes to stabilize airflow [7]. Surface discharge is generated using a pair of electrodes placed between a sheet of insulator film. The other electrode is set apart from the electrode for surface discharge on the film and energized with negative DC. From the surface discharge, streamers are extended by the DC electric field and cover the large surface area.

In order to ionize honeycombs consisting of fine channels, a packed-bed discharge is used in front and a DC electric field is applied across the honeycomb. This electrode configuration enables the ionization of the fine channels (1 mm^2) of a honeycomb made of cordierite.

In this study, conditions for establishing plasma inside a bundle of transparent glass capillary tubes (inner diameter 1 or 2 mm) that simulate the channels of honeycomb catalysts has been studied. This is because the glow associated with the discharge can be visualized in this setup. The characteristics of a honeycomb discharge in a bundle of glass capillaries are reported.

2. Instrumentation

2.1. Plasma reactor

Fig. 1 illustrates the plasma reactor used in this study. A packed-bed discharge was used to form the preceding discharge. The γ-Al_2O_3 pellets (3 mm) were set in a quartz glass tube (inner diameter 26 mm) with a stainless steel rod (diameter 6 mm) at the center as a discharge electrode. The outer surface of the quartz glass tube was covered with an aluminum sheet of 15 mm length as a ground electrode.

In this experiment, bundled thin quartz glass capillary tubes were used to simulate a honeycomb catalyst and to observe the

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doi:10.1016/j.elstat.2009.02.004

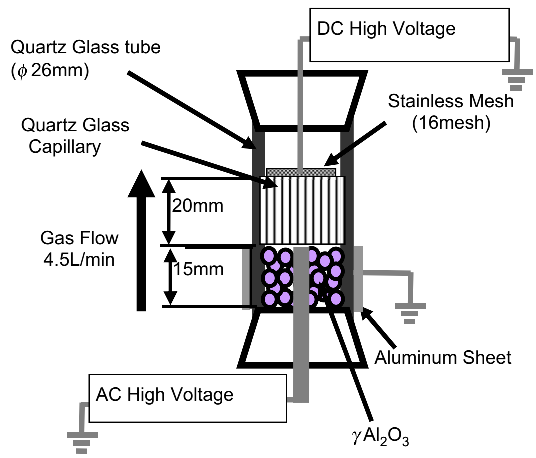


Fig. 1. Schematic diagram of the reactor with bundle of capillary.

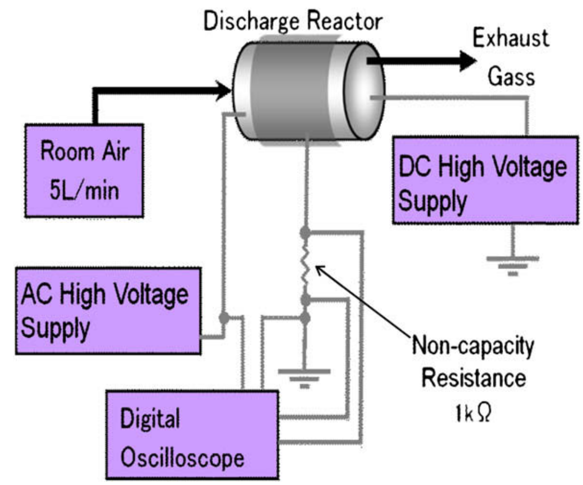


Fig. 2. Experimental setup.

light emission. The inner diameter of the quartz glass capillary was 1 or 2 mm and the length was 20 mm. The diameter of the bundle was 26 mm and the bundle was inserted into the outer quartz glass tube. A stainless steel mesh (16 mesh) was placed on the upper end of the capillaries. This stainless steel mesh was connected to a DC power supply to form the honeycomb discharge in the capillary.

In the packed bed, discharges are generated at contacting points of the γ-Al_2O_3 pellets (3 mm). The packed-bed discharge is used as the preceding discharge. When DC high voltage is applied between the bundle of quartz glass capillaries, the inside of the capillaries can be ionized, as if the AC discharge (packed bed discharge) is sliding into the capillaries by the DC electric field.

2.2. Experimental setup

Fig. 2 illustrates the experimental setup used in this study. The experimental measurements were carried out at room temperature under normal pressures. An air pump placed downstream of this system was used to generate gas flow. The gas flow rate was 5 L/min for room air, or 3 L/min for N_2 gas.

A DC high voltage power supply (Pulse Electronic Engineering Co., Ltd. HDV-50K3SUD) and an AC high voltage power supply (TREK Model20/20C High Voltage Amplifier) equipped with a function generator (Agilent Function/Arbitrary Waveform Generator 33220A) were used. The waveform of the applied voltage was measured using a digital oscilloscope (Tektronix TDS 2014) equipped with a high voltage probe (Tektronix P644A).

The starting voltage of the honeycomb discharge in the capillary is defined as the voltage when the light emission of the discharge is observed inside the capillary.

The maximum voltage is defined as the voltage when sparking takes place and the DC high voltage cannot be applied any more.

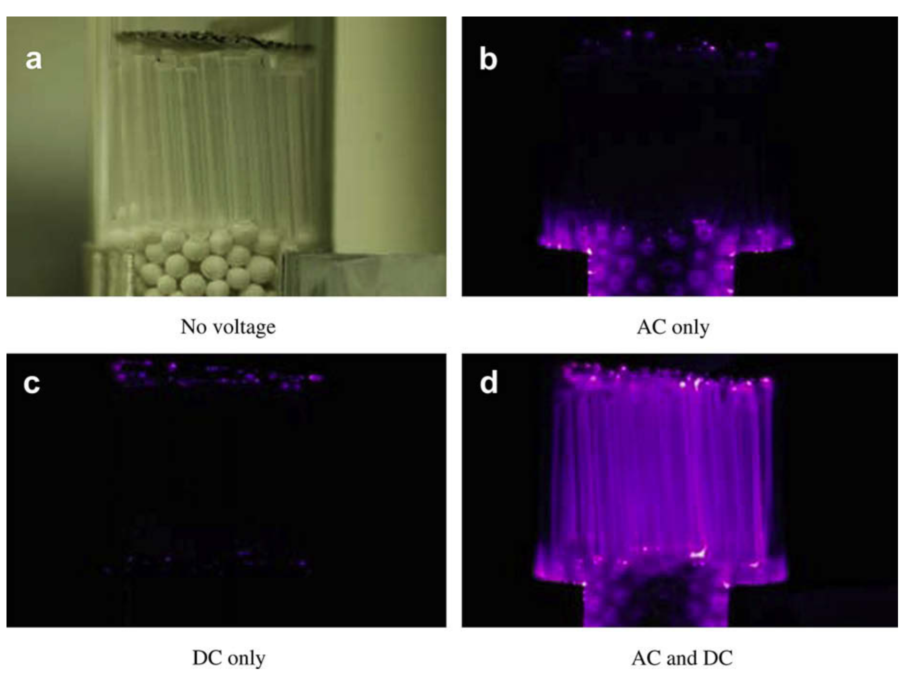


Fig. 3. Photographs of the honeycomb discharge in the bundle of capillary (exposure time of the camera was 6 s). Quartz capillary: 1 mm inner diameter, 20 mm in length, AC 25 $kV_{p\text{-}p}$, 1 kHz, DC −11 kV.

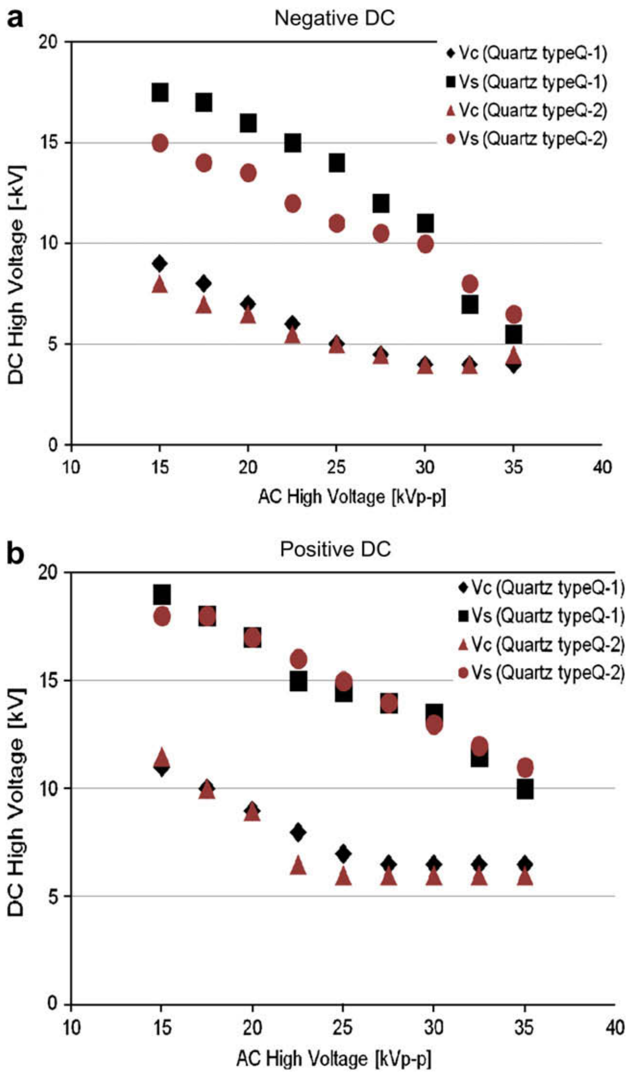


Fig. 4. Relationship between DC and AC high voltage for generation of the uniform discharge plasma. Capillary length, 20 mm; Q-1, 1.0 mm; Q-2, 2.0 mm inner diameter; Vc, the starting voltage; Vs, the maximum voltage (the flashover voltage).

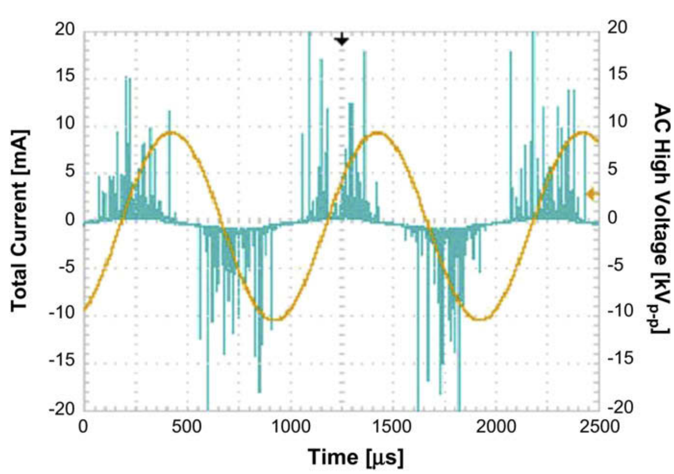


Fig. 5. Waveform of AC high voltage and total current for generation only packed bed discharge (negative DC high voltage 0 kV, 2.0 mm inner diameter, AC high voltage 20 $kV_{p\text{-}p}$, 20 mm capillary).

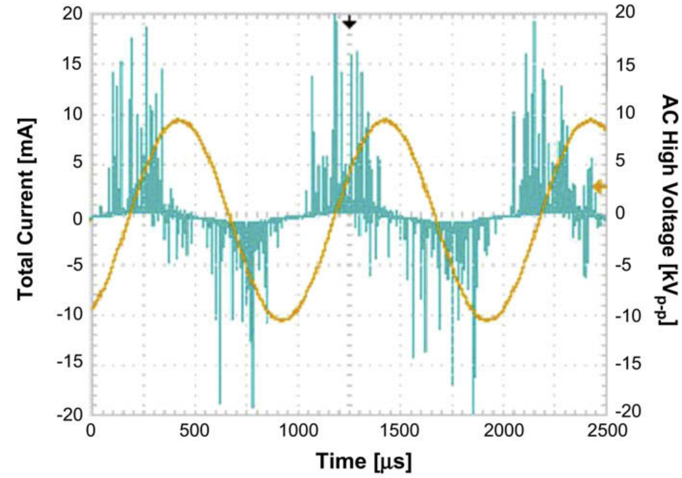


Fig. 6. Waveform of AC high voltage and total current for generation of the uniform slide discharge (negative DC high voltage −12.5 kV, AC high voltage 20 $kV_{p\text{-}p}$, 2.0 mm inner diameter, 20 mm capillary).

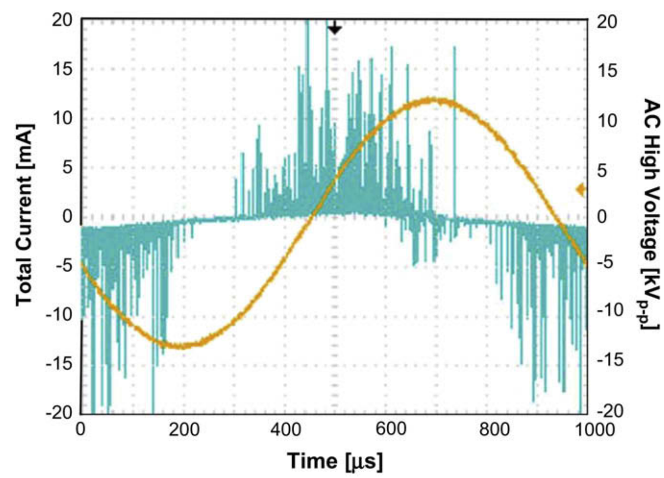


Fig. 7. Waveform of AC high voltage and total current for generation of the uniform slide discharge (negative DC high voltage −10 kV, AC high voltage 25 $kV_{p\text{-}p}$, 2.0 mm inner diameter, 20 mm capillary).

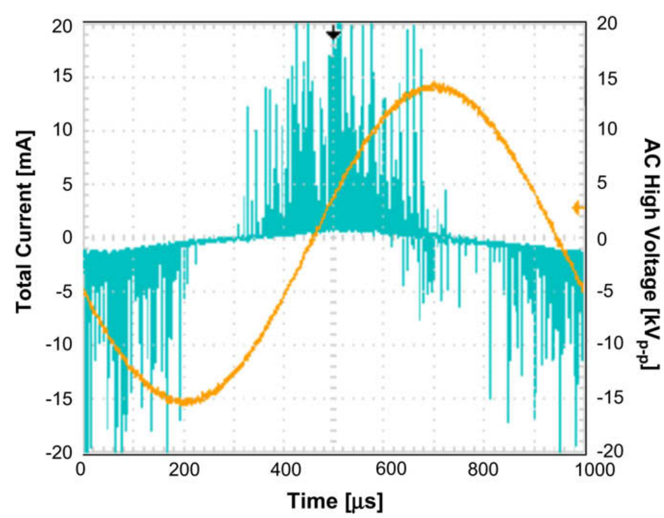


Fig. 8. Waveform of AC high voltage and total current for generation of the uniform slide discharge (negative DC high voltage −8 kV, AC high voltage 30 $kV_{p\text{-}p}$, 2.0 mm inner diameter, 20 mm capillary).

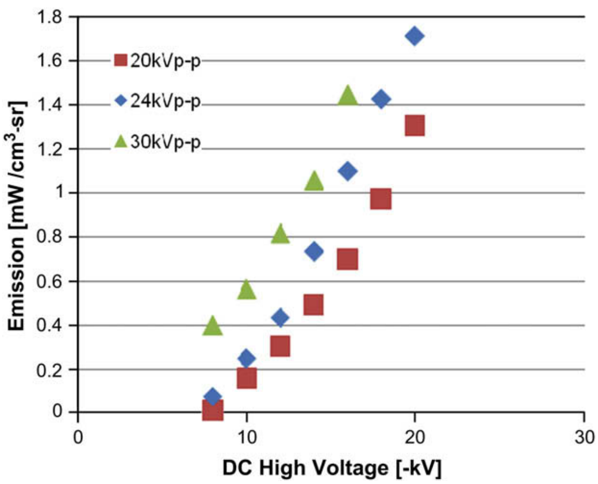


Fig. 9. DC high voltages vs. emission (AC 50 Hz, 2.0 mm inner diameter, 20 mm capillary).

2.3. Measurement of wave form

The waveform of AC high voltage was measured using a digital oscilloscope. The waveform of the total discharge current was measured with a non-inductive shunt resistor. A 1-kΩ resistor was inserted in the circuit between the grounded electrode and the ground.

3. Results and discussion

3.1. Basic characteristics of the honeycomb discharge in the bundle of capillaries

Fig. 3 shows photographs of the discharge in the bundle of quartz glass capillaries of 1 mm inner diameter. Each photo in Fig. 3 is as follows: (a) without applied voltage, (b) AC alone, (c) DC alone, (d) both AC and DC to form a honeycomb discharge in the bundle. The applied AC high voltage was 25 $kV_{p\text{-}p}$, 1 kHz, and the DC high voltage was −11 kV.

In Fig. 3(b) when the AC voltage was applied, the packed bed discharge was observed. In Fig. 3(c) with the DC alone, light emission was observed only in the upper part of the quartz glass tube. In Fig. 3(d) with the AC and DC, the honeycomb discharge in the bundle of capillaries was taking place, and uniform light emission along the capillary tube was observed. The intensity of the light emission increased with the increasing DC voltage until sparking occurred.

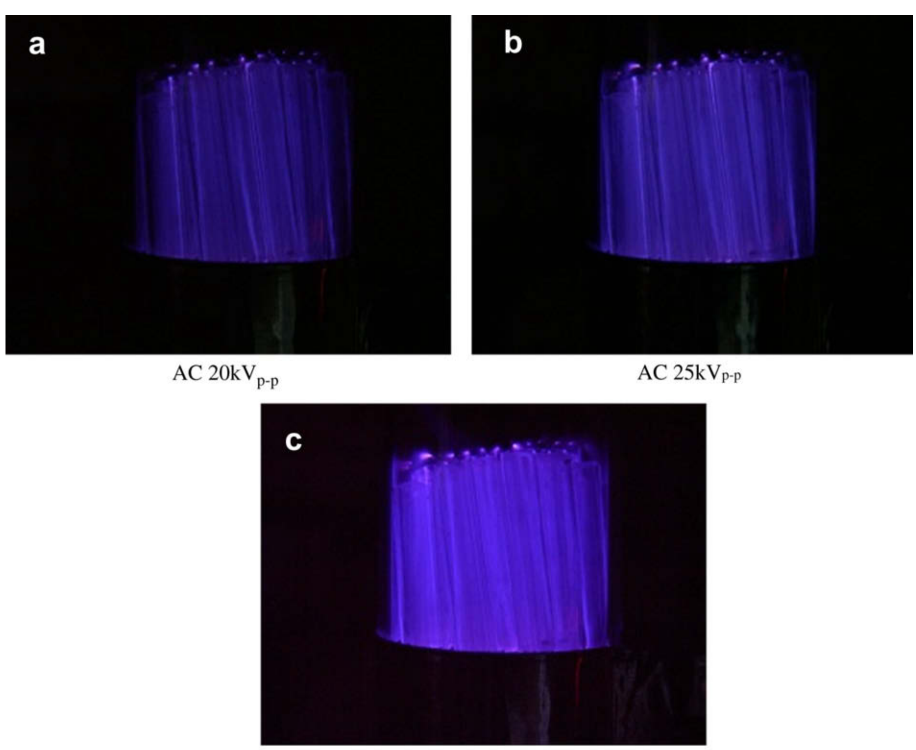
3.2. The relationship between DC and AC high voltage

The relationship between DC and AC high voltages to generate uniform honeycomb discharges in quartz glass capillary was measured. Fig. 4 indicates the starting voltage Vc of the honeycomb discharge in the quartz glass capillary and the maximum voltage Vs (flashover voltage) of the DC voltages when the AC voltage is changed. The DC voltage is negative in Fig. 4(a), and positive in Fig. 4(b).

DC voltage was increased slowly from zero until sparking took place. When it was measured, the AC high voltage and the frequency were fixed. The influence of the inner diameter size of the quartz glass capillary was also measured.

In Fig. 4, Vc decreased with the increasing AC voltage. With negative DC voltage, (Vs − Vc) was larger than that with positive DC. With negative DC, both Vs and Vc values were lower than those with positive DC.

It is well known that positive streamers tend to extend further compared to negative streamers. In this honeycomb discharge in the capillary, ionized gases in the packed-bed discharge act as an electrode, which supply electrons, ions, and photons more easily than metal electrodes. Therefore, more effective streamer generation and propagation could be achieved.



AC 20kV$_{p\text{-}p}$ AC 25kV$_{p\text{-}p}$ AC 30kV$_{p\text{-}p}$

Fig. 10. Photographs of honeycomb discharge (DC high voltage −16 kV, Camera exposure time: 0.5 s).

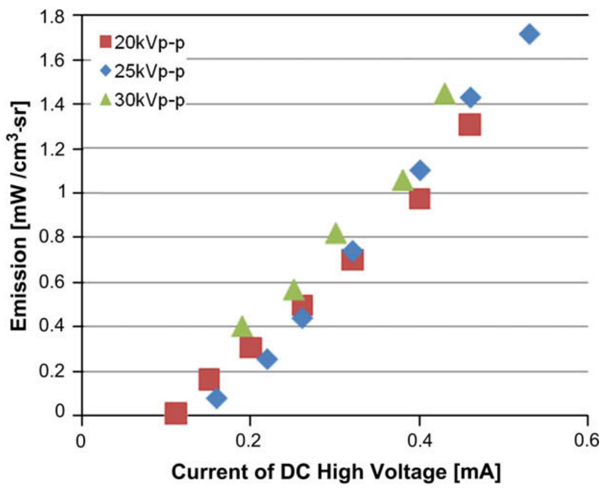


Fig. 11. DC current vs. emission (AC 50 Hz, 2.0 mm inner diameter, 20 mm capillary).

In the experimental conditions, the honeycomb discharge took place roughly with an average DC electric field in the capillary of 3–8.5 kV/cm and depended on the AC voltage value. In the meantime, stronger light emission was observed from the quartz glass tube when negative high voltage was applied.

3.3. Waveform measurement

The waveform of AC high voltage and total current was measured. Fig. 5 shows the waveform of AC high voltage and total current when no DC high voltage was applied. Positive and negative current were observed when the AC voltage was increasing or decreasing with time. The current pulses disappeared when the applied AC voltage reached the maximum and minimum. Because only packed-bed discharge was generated in this case, the current shown in Fig. 5 was typical of barrier discharges.

Fig. 6 shows the waveform of the AC high voltage and the total current when the honeycomb discharge was generated by the DC application to the bundle of capillaries. When the AC voltage reached the maximum in the positive half cycle, a negative current pulse was observed. This result suggests that the negative pulse current can be attributed to the honeycomb discharge.

In Fig. 6, the current pulse at the peak of the AC voltage (at $dV/dt = 0$) was observed only in the positive half cycle of the applied voltage. This is because the capillary tubes were negatively biased and the positive voltage to the packed bed enhances the electric field along the capillary tubes.

Figs. 7 and 8 show the waveforms of the AC high voltage and the total current with higher AC voltage and lower DC voltage. The maximum potential differences were nearly the same in all the experiments (22.5–23 kV). The AC and DC voltages were 25 kV_{p-p} and −10 kV in Fig. 7 and 30 kV_{p-p} and −8 kV in Fig. 8. Figs. 6, 7 and 8 show that amplitude of the current pulse associated with the honeycomb discharge is nearly the same in these figures. The current pulses associated with the honeycomb discharge started at a different value of AC voltages but at nearly the same AC + DC voltages (20 kV). These results suggest that honeycomb discharge is initiated when the electric field exceeds a certain value. The threshold value was about 10 kV/cm in these experimental conditions.

3.4. Emission spectra of the honeycomb discharge in the bundle of capillaries

Fig. 9 indicates the DC high voltage vs. emission of the honeycomb discharge. The emission of honeycomb discharge was measured using a spectrometer (Ocean Optics SD2000 and USB4000). These results were measured 30 s after starting the discharge. The peak value of the measured emission of 337.01 nm (N_2 C-B, 0-0) is plotted.

The inner diameter was 2 mm and the length was 20 mm of the bundle of quartz glass capillaries. AC frequency was set to 50 Hz. The applied AC high voltage was set 20 kV_{p-p}, 25 kV_{p-p}, 30 kV_{p-p}, 50 Hz, and the DC high voltage was varied when measuring the emission of honeycomb discharge.

With the same AC voltage, the emission was stronger with increasing DC voltages. At the same DC voltage, the emission was stronger with increasing AC voltages.

Fig. 10 shows photographs of the honeycomb discharge when the light emission was measured. The DC voltage was −16 kV. The AC voltage of each photograph is as follows: (a) 20 kV_{p-p}, (b) 25 kV_{p-p}, (c) 30 kV_{p-p}, to form a honeycomb discharge in the bundle. Using the transparent quartz glass tube, generation of the ionization inside these tubes was confirmed.

In the case of each AC high voltage, the quartz glass capillaries confirmed that an electric discharge with the emission of light occurs uniformly. In addition, light emission was confirmed that was stronger along AC high voltage values.

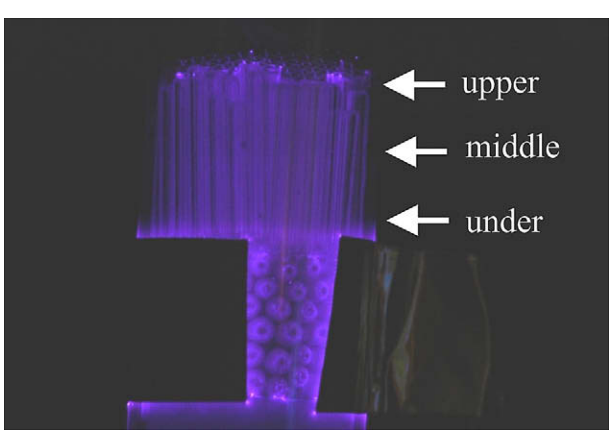


Fig. 12. Photographs of honeycomb discharge (AC 25 kV_{p-p} 1 kHz, DC −15 kV 2 mA, N_2 gas 3 L/min, 2.0 mm inner diameter, 20 mm capillary, camera exposure time 4 s).

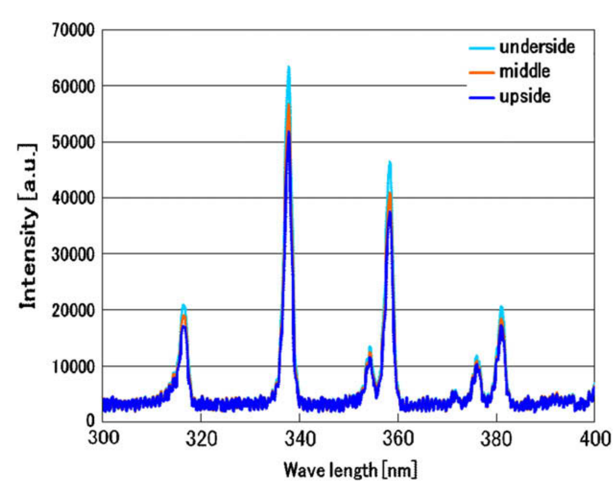


Fig. 13. Wavelength vs. intensity (AC 25 kV_{p-p} 1 kHz, DC −15 kV 2 mA, N_2 gas 3 L/min, 2.0 mm inner diameter, 20 mm capillary).

Table 1
Temperature of the N_2 gas in the honeycomb reactor.

	T_r (K)	dT_r (K)	T_v (K)	dT_v (K)
Packed middle	300	50	2150	150
Honeycomb underside	300	50	2100	200
Honeycomb middle	300	50	2100	150
Honeycomb upside	300	50	2100	150

Fig. 11 shows the light emission intensity vs. DC current. The AC high voltage was set to 20 $kV_{p\text{-}p}$, 25 $kV_{p\text{-}p}$, 30 $kV_{p\text{-}p}$, 50 Hz, and the DC high voltage was changed when measuring the emission. The light emission seems proportional to the DC current.

Unlike the relations between DC high voltage and emission, the DC high voltage current rose with the value of DC high voltage regardless of a value of the AC high voltage. This suggested that the emission spectrum of the honeycomb discharge depended on the current of the DC high voltage.

Fig. 12 shows photographs of the honeycomb discharge when the light emission was measured. The AC high voltage was set to 25 $kV_{p\text{-}p}$, 1 kHz and the DC high voltage was set to −15 kV. The gas flow was set to 3 L/min N_2 gas.

The emission intensity was measured at the upper, middle and under side, as indicated in Figs. 12 and 13, which show the wavelength vs. light emission intensity at those parts. The AC high voltage was set to 25 $kV_{p\text{-}p}$, 1 kHz and the DC high voltage was set to −15 kV. The gas flow was set to 3 L/min N_2 gas.

The emission intensity of the under-side was the strongest. Next was the middle (89% of the under-side) and the lowest was from the upper-side (81%). This result showed the attenuation of the light emission was 10% at 1 cm.

These results suggested that the emission spectrum and intensity of the honeycomb discharge depended on the length of the honeycomb.

Table 1 shows the N_2 gas temperature in the honeycomb reactor. These temperatures were calculated from the emission intensity. Based on the spectral band we also determined rotational (T_R) and vibrational (T_V) temperatures by using Specair software. The typical measured temperatures were $T_R = 300 \pm 50$ K, $T_V = 2100 \pm 150$ K. The plasma generated by the sliding discharge was relatively cold with a high level of non-equilibrium.

3.5. *Generation of honeycomb discharge using AC high voltage and negative pulse*

It is well known that pulse discharge has greater efficiency with chemical reactions than DC high voltage. The DC high voltage was changed to the negative pulse high voltage. The packed-bed discharge was used as generated by AC high voltage. When pulsed high voltage is applied inside the capillaries they can be ionized, as if the AC discharge is sliding into the capillaries.

Fig. 14 shows photographs of the discharge using pulsed high voltage. Each photograph in Fig. 14 is as follows: (a) applied negative pulse high voltage at the positive peak of AC high voltage, (b) applied negative pulse high voltage before 100 µs at the positive peak of AC high voltage, (c) applied negative pulse high voltage after 100 µs at the positive peak of AC high voltage. The applied AC high voltage was 30 $kV_{p\text{-}p}$, 1 kHz and the pulse high voltage was −10 kV, 1 kHz.

In Fig. 14(a) the most shiny honeycomb discharge was observed. The honeycomb discharge in the bundle of capillaries was taking place and uniform light emission along the capillary tube was observed. In Fig. 14(b) before 100 µs, honeycomb discharge was partly observed. In Fig. 14(c) after 100 µs at the peak of AC high voltage, honeycomb discharge was normally observed.

The emission intensity was measured at the middle side (as indicated in Fig. 12). Fig. 15 shows the wavelength vs. light emission intensity. The AC high voltage was set to 30 $kV_{p\text{-}p}$, 1 kHz and the pulse high voltage was set to −10 kV, 1 kHz. The gas flow was set to 3 L/min air.

The emission intensity of the pulse applied at the peak of AC high voltage was the strongest. The next was after 100 µs and the

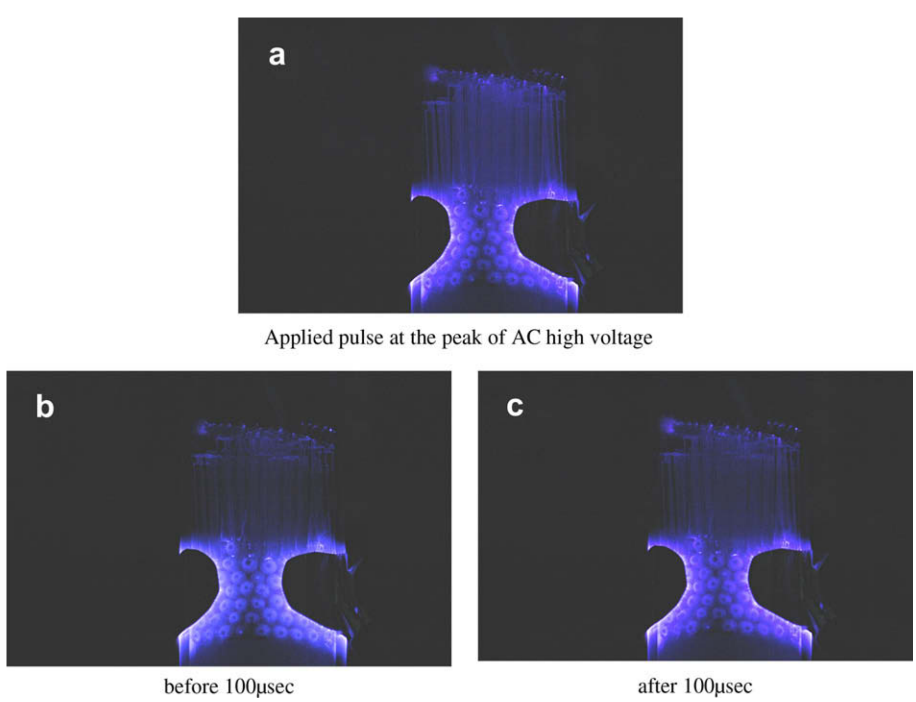


Fig. 14. Photographs of the honeycomb discharge using the pulse high voltage. Quartz capillary: 2 mm outer diameter; 20 mm in length; AC 30 $kV_{p\text{-}p}$; 1 kHz; pulse −10 kV, 1 kHz; camera exposure time 1.6 s.

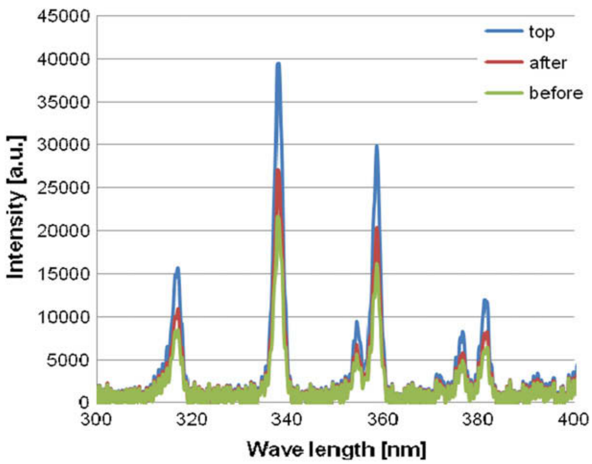


Fig. 15. Wavelength vs. emission intensity (AC 30 $kV_{p\text{-}p}$ 1 kHz, pulse −10 kV, 1 kHz, air 3 L/min, 2.0 mm inner diameter, 20 mm capillary).

lowest was before 100 μs. This result showed the intensity of the light emission was related to the peak of the preceding AC discharge. At 100 μs before the AC high voltage peak, the preceding AC discharge is just generating. As a result, the pulse high voltage cannot expand into the discharge area.

These results suggest that the emission spectrum and intensity of the honeycomb discharge depend on the timing of the pulse and the preceding AC discharge.

4. Conclusion

Using both AC and DC high voltage, generation of plasma inside a honeycomb has become possible. Using a bundle of transparent capillaries simulating honeycomb catalyst, the characteristics of honeycomb discharge have been studied.

AC voltage was applied to the packed bed, and DC voltage was applied to the bundle. To generate a stable honeycomb discharge, the condition of the AC and DC voltages are clarified as the onset and the flashover voltages. The average electric field strength for the onset of the honeycomb discharge was about 10 kV/cm in these experimental conditions. The honeycomb discharge is pulsating, detected as the current pulses at an instant of the maximum of the AC voltage. The light emission intensity from the honeycomb discharge is roughly in proportion to the DC current.

The emission intensity of the under-side was the strongest. Next was the middle (89% of the under-side) and the lowest was from the upper-side (81%). The plasma generated by the sliding discharge is relatively cold with a high level of non-equilibrium.

The emission intensity of the pulse applied at the peak of the AC high voltage was the strongest. The next was after 100 μs and the lowest was before 100 μs. This result showed the intensity of the light emission was related to the peak of the preceding AC discharge.

Acknowledgments

The authors are grateful to Prof. G. Touchard and Prof. E. Moreau of University of Poitiers for the discussions and suggestions given to this work.

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