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# LIF imaging of OH radicals in DC positive streamer coronas

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**Abstract**

In this study, the LIF detection of the hydroxyl (OH) radicals was performed in a nozzle-to-plane electrode system having an electrode gap of 30 mm during the steady-state positive streamer corona discharge at atmospheric pressure. For monitoring the ground-state OH radicals, OH transition [$A^2\ \Sigma^+(v'=1) \leftarrow X^2\Pi(v''=0)$] at 282 nm was used. The time relationship between the regular streamer coronas, laser pulse, OH fluorescence and laser-induced streamer was measured. The time dependence of OH radicals between the successive streamers was measured for the evaluation of OH dynamics when the discharge was in a steady-state condition. The two-dimensional OH distribution in the DC streamer corona discharge was observed. The obtained results showed that the ground-state OH radicals were generated mainly in the filamentary part of the streamers. It was found that LIF detectable amount of ground-state OH radicals stayed in the region where streamers propagate during the steady-state DC positive streamer corona in open air.


*Keywords:* DC streamer corona discharge; Laser-induced fluorescence; OH radical; Non-thermal plasma processing

## 1. Introduction

Streamer coronas at atmospheric pressure are widely used in the non-thermal plasma processing of harmful gases. The direct study of the streamer-induced plasma chemical processes in non-thermal plasma reactors is of great importance. Laser-induced fluorescence (LIF) is a useful diagnostic method for in situ observation of the phenomena during the non-thermal plasma processing of gaseous pollutants. The LIF has been already employed by authors [1,2] and other researchers [3–6] for studying NO removal process in various non-thermal plasma reactors. In addition to the behavior of pollutants such as NO and $NO_2$, direct observation of the radical dynamics during the non-thermal plasma process is important not only for understanding the plasma chemical reactions of gas treatment but also supplying the advanced data for modeling of the process. Especially, the hydroxyl (OH) radicals generated in the non-thermal plasma is considered to enhance the chemical reactions in the reactor. Recently, several researchers succeeded in measuring the OH radicals using LIF technique in the pulsed corona discharges [7–9].

For promoting industrial applications of non-thermal plasma technique, DC streamer corona discharge is suitable regarding the initial investment and the operation cost. For example, a corona radical shower system operated by DC streamer coronas is one of the most efficient methods in terms of the energy efficiency and suitable for the retrofit of the existing electrostatic precipitators [10,11]. Although DC streamer coronas have these advantages as the non-thermal plasma for pollution control, there are still no experimental data of OH radicals. This is mainly due to difficulties in the synchronization of the DC streamer corona, LIF signals and observation time. The DC positive streamer corona discharge consists of more of less regular self-repetitive current pulses with pulse duration up to several hundred nanoseconds and repetition frequency in the range of 1 to 100 kHz. On the other hand, the lifetime of OH radicals is very short (up to 1 ms). These complex phenomena make the measurement of OH radicals in the DC streamer corona difficult. In a previous research [12], however, to synchronize the DC corona discharge with the LIF system, DC superimposed pulsed high voltage with slow rise time was used

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and LIF signal was detected after one filamentary discharge in gas mixtures of Ar and air.

In this study, as a first step of the OH radical measurement under DC streamer coronas, we tried to measure ground-state OH radicals presented during the discharge in air, because OH radicals in the excited-states rapidly transfer to the ground-state by quenching [13]. For monitoring the ground-state OH radicals, OH transition at 282 nm (1–0 band) was used. One of the purposes of the present paper is to make clear whether OH radicals can be observed in DC streamer corona or not. According to the measurement of OH radicals in the pulsed corona discharge [7–9], whose peak current is three-order of magnitude higher than the DC streamer corona, higher current density may be responsible for the LIF detectable amount of OH radicals. Moreover, the LIF intensity is often affected by the quenching due to ambient gases. The second purpose is to investigate the spatial distribution of OH radicals during steady-state positive streamer corona discharge if OH radicals are observed by means of LIF.

## 2. Experimental apparatus and methods

The schematic diagram of the experimental apparatus is shown in Fig. 1. In order to observe the ground-state of OH radicals in the reactor using LIF technique, OH [$A^2\Sigma^+(v'=1) \leftarrow X^2\Pi(v''=0)$] system at 282 nm was used. A frequency-doubled-dye laser system was used for the LIF measurement. The laser pulses from an XeF excimer laser (Lambda Physik, COMPex 150, tuned at 351 nm) pumped a dye laser (Lambda Physik, SCANmate) with Coumarin 153 as a dye that generated a laser beam of a wavelength tuned around 564 nm. A BBO crystal ($BaB_2O_4$) pumped by the tuned dye

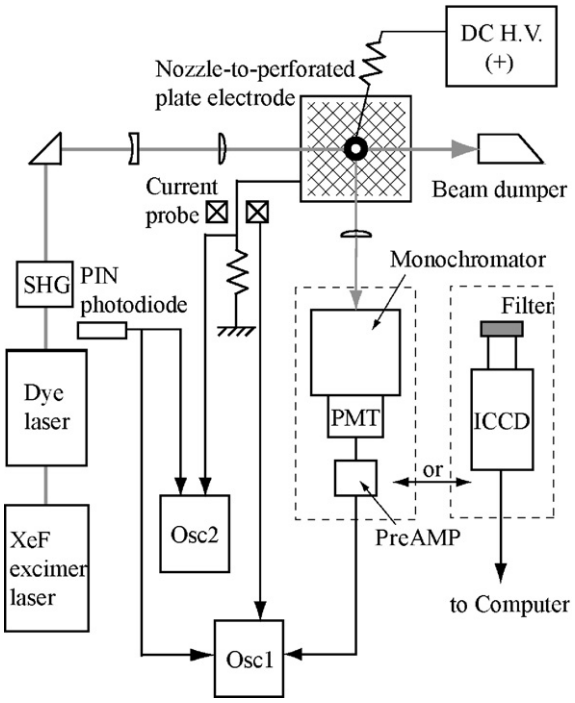


Fig. 1. Schematic diagram of the experimental apparatus (top view).

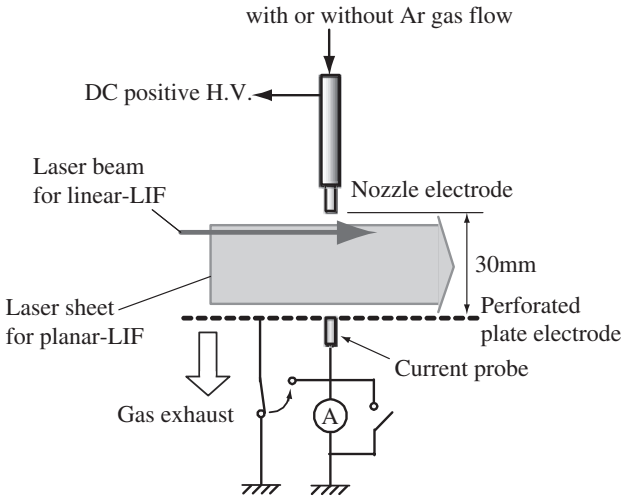


Fig. 2. Schematic diagram of the discharging region and incident laser for LIF measurement (side view).

laser beam produced the second harmonic radiation of a wavelength correspondingly tuned around 282 nm (energy: ~2.5 mJ, duration: 20 ns as FWHM). The 282 nm laser beam with a diameter of 2 mm passed between the electrodes as shown in Fig. 2. Excitation of OH was achieved via the $Q_1(1)+Q_{21}(1)$ line (281.92 nm). This transition was chosen due to the strongest fluorescence signal. LIF signal emitted at 90° to the laser beam was focused onto the entrance slit of a 25 cm monochromator (Nikon, P-250) through a lens. The LIF signal at around 309 nm [$A^2\Sigma^+(v'=0) \rightarrow X^2\prod(v''=0)$] was detected by a photomultiplier tube (PMT). The PMT signal was sent to a digital oscilloscope (Osc1, see Fig. 1) through a preamplifier. In order to observe two-dimensional (2-D) OH profile, a gated ICCD camera (LaVision, Flame Star II) was used instead of the monochromator system. To remove the scattered laser light, the narrow optical filter centered at 310 nm (10 nm as FWHM) was attached to the lens of the ICCD camera. For the 2-D observation of OH radicals between the electrodes with a 30 mm-gap, a laser sheet (1 mm-width and 25 mm-height) was also used.

A stainless-steel pipe with a nozzle (1.0 mm in inner diameter, 1.5 mm in outer diameter) was used as the stressed electrode. An additional gas of Ar can be supplied to the discharge zone through the nozzle. Since the Ar addition changes the discharge structure from branching streamer to filamentary discharge, spatial relationship between the laser beam and streamer passage can be easily optimized for a first step of LIF measurement. Current density is also increased with the injection of Ar due to no branching. The plane electrode (100 mm in square) was a brass plate with an array of holes (1.5 mm in diameter) perforated to allow the gas exhaust. Averaged air flow velocity due to this suction was 1.1 m/s in the gap. DC high voltage with positive polarity was applied through a 10 MΩ resistor to the nozzle electrode. The DC positive streamer corona discharge was realized in open air with and without additional gas flow. The discharge current pulse was measured using a current probe (Pearson Electronics, 2877). Also the potential across a resistor connected between the plane

electrode and the ground was measured. Time averaged current was measured by a current meter as shown in Fig. 2. In order to measure the current density underneath the nozzle electrode, the center of the plane electrode contained a 2 mm diameter probe that was electrically isolated from the plane. The laser shot was monitored using a PIN photodiode placed at 2.6 m in advance of the discharge zone. A time relationship between discharge current and the laser shot was measured by another oscilloscope (Osc2) as shown in Fig. 1. No time synchronization between the discharge and laser shot was made. This means that a laser pulse is irradiated at random between the discharge current pulses. The time relationship between the discharge current pulses, laser pulse, and LIF signal were described in detail elsewhere [14,15]. The experiment was carried out at room temperature under atmospheric pressure. The concentration of water in the room was estimated to be in the range of 1–2 vol.%.

## 3. Experimental results and discussion

Fig. 3 shows the typical time dependence of laser incidence, OH LIF signal and laser induced streamer when the laser beam was irradiated between the successive streamers in humid air during the steady-state discharge condition. In this case, the incidence time of the probe laser beam was 140 μs after the last current pulse of the discharge as shown in Fig. 3(a). The distance between the center of the laser beam and the tip of the nozzle electrode was approximately 5 mm. Taking into account the time delay between the laser pulse detected by the PIN photodiode and OH LIF signal by the PMT as well as the delay due to the circuits and cables, it is considered that the LIF signal appears immediately after the laser pulse. The duration of OH fluorescence is approximately 30–40 ns as shown in Fig. 3(b). When the wavelength of the probe laser was changed to off-resonance, OH LIF signal was disappeared. Consequently, it was confirmed that the ground-state OH radicals were present even in the steady-state DC streamer corona discharge. Additional streamer is induced due to the laser shot for LIF measurement because the photon energy of the probe laser is high enough to trigger the discharge. The emission from the laser-induced streamers is also observed. These peaks due to the laser-induced streamers are attributed to $N_2$ second positive emission. In addition, no OH fluorescence was observed at the same position when the corona discharge was glow mode, because the discharge property of glow mode is much different from that of streamer mode. It is considered that OH radicals are produced in the streamer head and streamer channel.

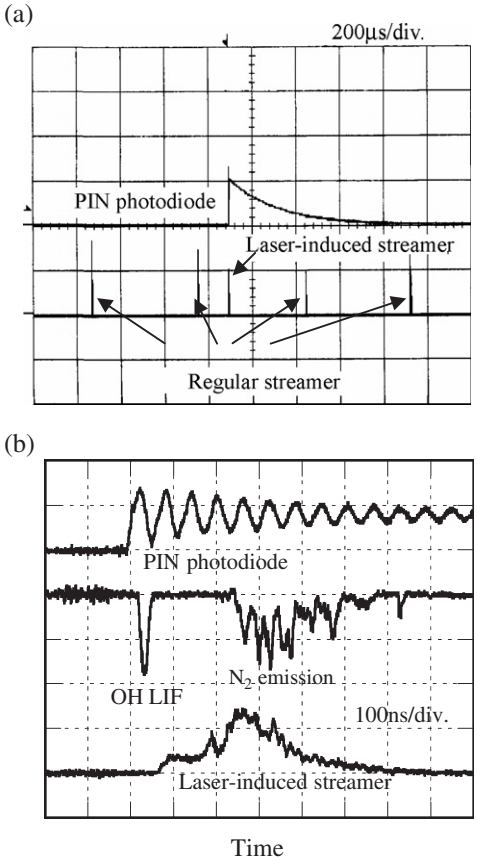


Fig. 3. Timing relationship between laser detected by a PIN photodiode, OH LIF signal and current waveform of laser-induced streamer. The applied voltage is 29 kV and corona current is 185 μA. (a) Regular streamer current pulses and laser-induced current pulse. (b) Typical waveforms of OH LIF signal and laser-induced current pulse. The peak current of laser-induced streamer is 90 mA.

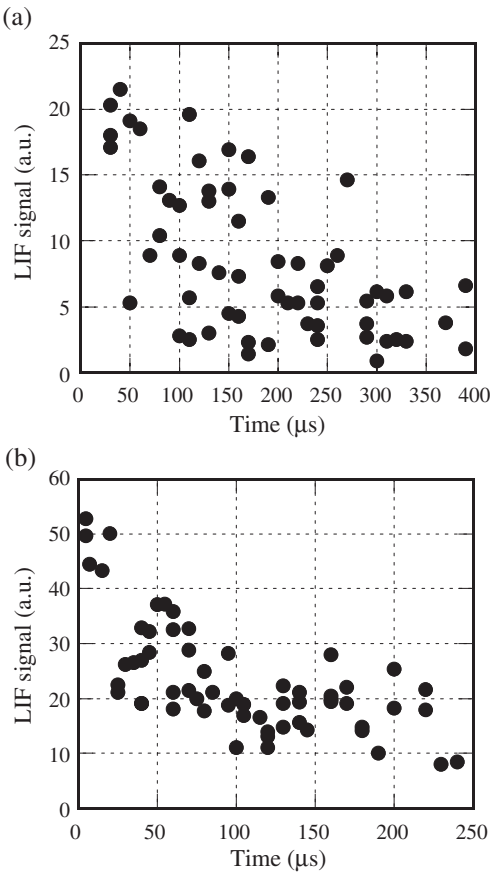


Fig. 4. Time evolution of OH LIF signal after the last streamer before the laser incidence: (a) in open air without Ar gas flow (applied voltage 29 kV, corona current 200 μA); (b) in open air with Ar gas flow (applied voltage 11 kV, corona current 40 μA).

Table 1
Discharge condition in the case of Fig. 4

| | Discharge environment | |
|---|---|---|
| | In open air without Ar gas flow | In open air with Ar gas flow |
| DC applied voltage (kV) | 29 | 11 |
| Time averaged current (μA) | 200 | 40 |
| Average value of current pulse (mA) | 122 | 52 |
| S.D. of current pulse (mA) | 44 | 16 |
| Averaged repetition rate of current pulse (kHz) | 2.1 | 5.1 |
| S.D. of repetition rate (kHz) | 1.2 | 0.9 |

S.D.: Standard deviation.

Fig. 4 shows LIF intensity as a function of postdischarge time in respect to the last current pulse during the steady-state corona streamer discharge with and without Ar gas flow (0.3 L/min). The time interval between the laser shot and the last current pulse of the discharge before the laser beam incidence was measured separately using the oscilloscope (Osc2 in Fig. 1). The LIF signal detected by the fast oscilloscope (Osc1 in Fig. 1) was integrated over the duration of the fluorescence. Table 1 shows the corresponding discharge characteristics. As seen in Table 1, the current waveforms of DC streamer corona discharge show that the averaged streamer repetition frequency for discharge in open air is 2.1 kHz at 29 kV. When an additional gas (Ar) was supplied to the discharge zone through the nozzle electrode with the flow rate of 0.3 L/min, the shape of the streamer discharge was changed drastically (see Figs. 5(a) and 6(a)) and LIF signal became more intensive. Moreover, in the case with Ar gas flow, the transition to spark discharge easily occurred around 15 kV, compared to the spark voltage of over 30 kV without Ar gas flow. The streamer discharge in open air (i.e., humid air) is composed of many branches and occupies much more space, while filamentary type of discharge is generated if Ar is injected into the gap through the nozzle electrode. The averaged current densities underneath the nozzle electrode were about 0.6 μA/mm$^2$ at 26 kV for the discharge without Ar injection and about 4.7 μA/mm$^2$ at 13 kV for the discharge with Ar injection, respectively. The averaged streamer repetition frequency increases up to 5.1 kHz at 11 kV when Ar is present in the discharge. The averaged time interval between two consecutive streamers is about 500 μs for the discharge without Ar gas flow and 200 μs for the discharge with Ar gas flow. Therefore, the OH radicals produced in the one streamer may be still present in the discharge region until the next streamers occur. Moreover, OH LIF signal is highly dispersed in the case of the discharge without Ar gas flow through the nozzle. There are many factors which should be considered concerning to the fluctuation of LIF signals: fluctuation of laser energy, collisional quenching, discharge instability such as fluctuation of the current pulses respect to the time interval and amplitude, branching of the streamers, spatial relationship between the laser and streamers, and electrohydrodynamic (EHD) flow effect. Especially, the discharge characteristics are much different depending on the gas composition existed in the gap as shown in Table 1. In the case of Ar injection, it is considered that not only the energetic electrons but also metastable Ar atoms contribute to generate OH radicals through the dissociation of $H_2O$. From the results shown in Fig. 4, the measurement method based on no time synchronization between the streamer and laser pulse can be applied to the evaluation of steady-state DC streamer coronas. If we average the signals, the steady-state measurement of OH LIF is possible. On the other hand, time synchronized

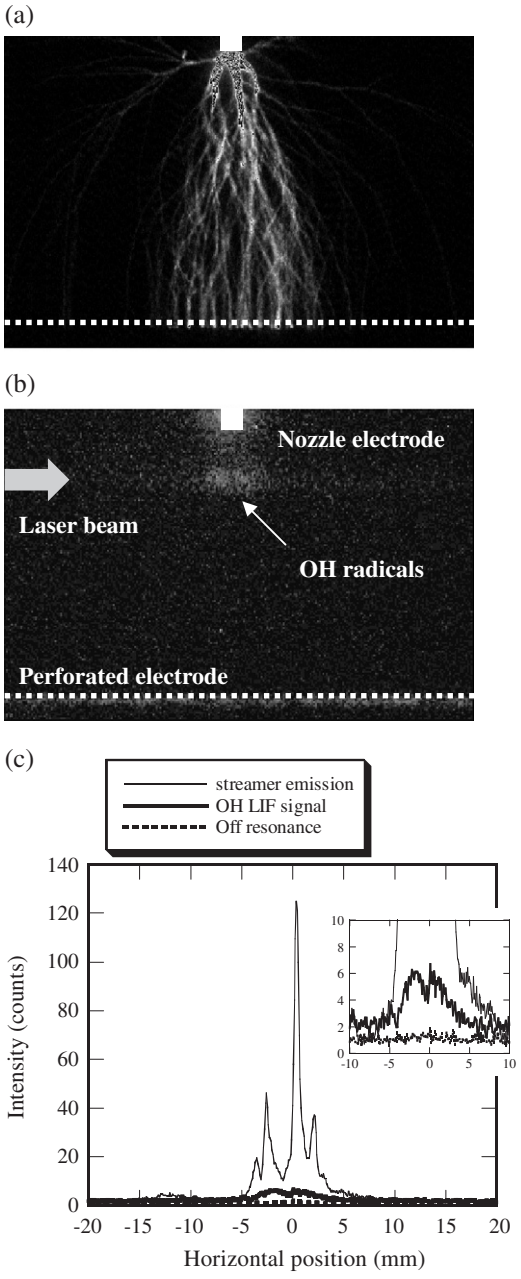


Fig. 5. Comparison of streamer and LIF images in open air. The applied voltage is 29 kV and corona current is 190 μA. (a) streamer emission observed by the ICCD camera with gate time of 900 μs; b) OH LIF near the tip of the stressed electrode (ICCD gate of 100 ns); (c) horizontal profile for both streamer emission and OH LIF. The profile obtained at the laser wavelength of off-resonance is also shown.

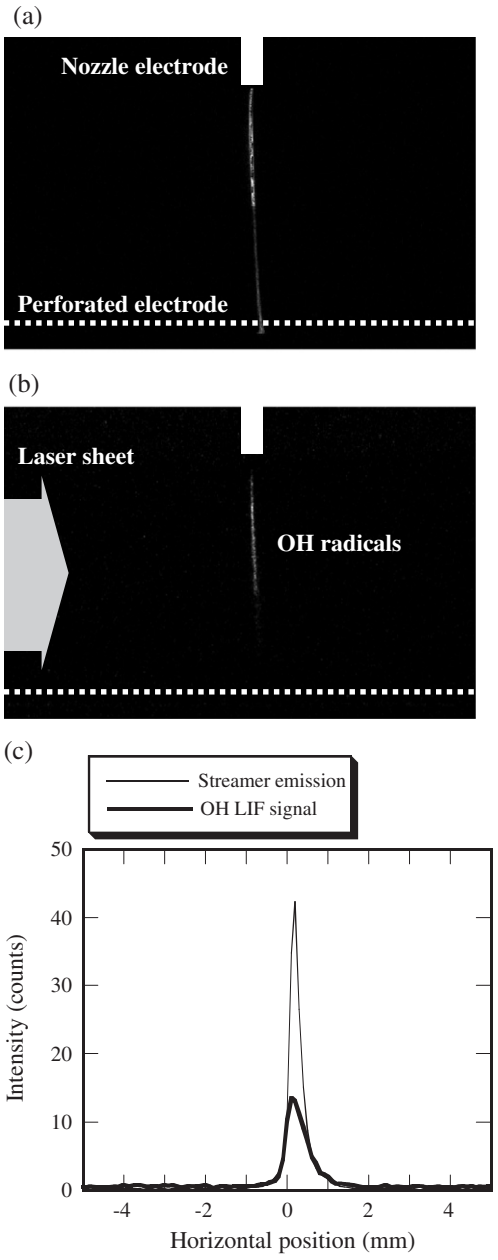


Fig. 6. Comparison of streamer and LIF images in open air. The applied voltage is 14 kV and corona current is 60 μA. (a) streamer emission observed by the ICCD camera with gate time of 900 μs; (b) OH LIF between the electrodes (ICCD gate of 100 ns); (c) horizontal profile for both streamer emission and OH LIF.

measurement has been already performed and evaluated by the authors [12].

Fig. 5 shows the 2-D discharge emission and OH profiles under steady-state DC streamer coronas in open air without Ar gas flow. The images of discharge emission were taken by the ICCD camera without the band pass filter. The discharge emission mainly consists of $N_2$ second positive band and no OH emission was observed [12]. This fact suggests that OH radicals in excited states are rapidly quenched by collisions with ambient gases [13]. In air the streamers are composed of many branches and occupy much more space than filamentary type of discharge with Ar injection. When the discharge was realized in open air, the LIF signal is much weaker and it is insufficient for single-shot 2-D imaging of OH radicals. Therefore, laser beam, which was not expanded to the sheet, was used. In order to increase SN ratio of the image, the image shown in Fig. 5(b) is an average of 50 captured images. Although the streamer emission and LIF images taken separately, it can be seen that OH LIF signal comes mainly from the streamer region as shown in Fig. 5(c).

In contrast, the 2-D discharge emission and OH profiles under the steady-state DC streamer coronas in open air with Ar gas flow (0.3 L/min) through the nozzle electrode is shown in Fig. 6. When Ar was introduced into the discharge region, the shape of the streamers changed to a filament without branches. In a spectroscopic analysis, the spontaneous emission of excited OH radicals was present as well as $N_2$ emission [12]. The OH LIF signal was easily detected even in the case of single-shot LIF imaging. The 2-D OH profile reflects its filamentary discharge structure as seen in Fig. 6. Similarly, OH LIF signal comes from within the streamer volume. These results suggest that OH radical generation occurred inside the streamer. In addition, it is considered that the reason of the difference between the images with and without Ar gas flow is due to the different characteristics of the discharges: the difference in the electric field at the streamer head, electron energy and its density as well as the presence of metastable Ar may affect the production and excitation processes of OH radicals.

## 4. Conclusion

The OH radical generation in DC streamer corona discharge was examined using the LIF method with a frequency-doubled-dye laser system. It was found that the LIF could apply to the measurement of OH radicals stayed in the steady-state DC streamer coronas in open air. The influence of Ar injection on the discharging characteristics and the generation of OH radical were also examined. The streamer discharge in air is composed of many branches and occupies much more space than filamentary type of discharge with Ar injection, resulting in the profiles of OH radicals between the electrodes. 2-D OH images show that OH radicals were generated in the streamers.

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