Diagnostics of NO Oxidation Process in a Nonthermal Plasma Reactor: Features of DC Streamer-Corona Discharge and NO LIF Profile

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Invited Paper

***Abstract*—Features of the streamer-corona discharge and the NO processing area in corona radical shower system was studied experimentally. The time evolution of the streamer-corona discharges induced by laser irradiations was measured to understand the discharge characteristics responsible for NO removal. Using the wide-range planar laser-induced fluorescence imaging (image size: 240 mm in width and 35 mm in height), two-dimensional distributions of ground-state NO were observed not only at the discharge zone but also both the downstream and the upstream regions of the reactor. The obtained results showed that the density of NO molecules decreased not only in the plasma region formed by the corona streamers and downstream region of the reactor but also in the upstream region of the reactor. In the present reactor at low main gas flow rate, it was considered that electrohydrodynamic (EHD) flow became to be dominant, and the flow toward the upstream affected the decrease of NO in the upstream region. As the EHD-induced secondary flow is correlated with the current flow from the stressed electrode to grounded electrode, the control of the streamers seems to be an important factor for optimizing the reactors used for NOx removal.**

***Index Terms*—Electrohydrodynamic (EHD) flow, NO removal, planar laser-induced fluorescence (PLIF), streamer.**

I. Introduction

UP TO NOW, various kinds of the nonthermal plasmas generated by electrical discharges at atmospheric pressure have been widely studied for the flue gas treatment [1], [2]. As far as NOx removal is concerned, more than ten kinds of the reactors have been developed and evaluated so far [3]. Specifically, streamer coronas are widely used in the nonthermal plasma reactors. The control of the streamer-corona discharge is a key issue to obtain suitable plasmas for gas treatment with respect to NO removal rate and the energy efficiency. A corona radical shower system [4], [5], which is one of the most efficient methods in terms of the energy efficiency and suitable for the retrofit of the existing electrostatic precipitators, uses a pipe with nozzles electrode as a stressed electrode. Therefore, the shape of the discharge is much more complex than that of the conventional electrode arrangement, such as wire-to-plate or needle-to-plate and is very complex, resulting in the generation of complicated electrohydrodynamic (EHD)-induced secondary flow. Moreover, it was noticed that both the discharge phenomena and the removal process including the chemical reactions were very complicated. In order to establish the proper model consistent with experimental results, it is necessary to understand the behavior of pollutants under the discharge in the reactor.

In recent years, laser-induced fluorescence (LIF) direct measurements of the radicals and molecules concentrations in the plasma reactors have been successfully demonstrated [6]–[10]. LIF can be used for *in-situ* observation of the phenomena in plasma reactors during the NOx removal process [11], [12].

In [11] and [12], we have investigated the NO LIF images for measuring the concentration of NO molecules in a streamer discharge reactor. During the course of the LIF measurement in the dc corona discharge, we have found that an ultraviolet (UV) pulsed laser shot used for LIF measurement induces a streamer. This laser triggering phenomenon allows us to study the characteristics of streamers with precisely spatial and temporal resolutions. It was confirmed that this so-called laser-induced streamer was similar with the normal streamers [12]–[14]. Therefore, the information on streamer and NO distributions in the reactor obtained in the present study seems to be useful for the development of analytical models of streamers as well as NO removal process and the design of the plasma reactor for NOx removal.

In this paper, characteristics of the streamer-corona discharge and related NO concentration profiles in a corona radical shower system are presented. The spatial and temporal dependence of the laser-induced streamer corona was investigated by using a gated-ICCD camera with 5-ns resolution. The planar laser-induced fluorescence (PLIF) technique was used to monitor wide-range two-dimensional (2-D) NO molecule distribution in the reactor during NO treatment.

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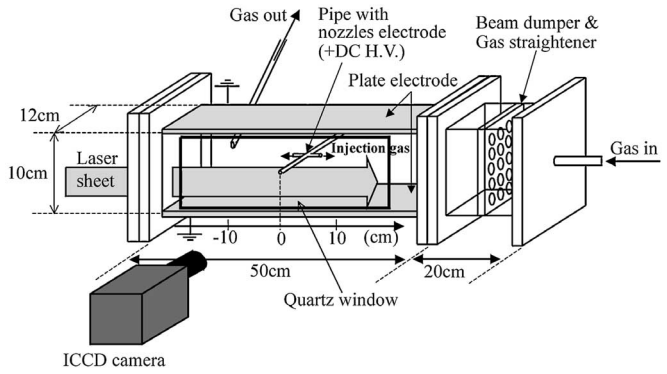

Fig. 1. Schematic of the experimental apparatus.

II. Experimental Method

Fig. 1 shows the schematic diagram of the experimental apparatus used in this study. The apparatus with the laser system and fundamentals of the measuring techniques were described in detail elsewhere [11], [12], [15] and only a brief description is given in this paper. The corona radical shower reactor used in this study consists of a pipe with two nozzles-to-plate electrode system. These electrodes were set inside the acrylic box ($100 \times 120 \times 700$ mm) in which quartz windows were mounted for the laser diagnostics. The gap distance between the nozzles and plate electrodes is 50 mm. DC high voltage with positive polarity was applied through a 10-M Ω resistor to the stressed electrode and the streamer coronas were generated. NO (1000 ppm)/N_2+ air mixture flowed along the reactor with a flow rate of 3 L/min, which corresponds to an average flow velocity of 4.2 mm/s. CO $_2+$ air mixture was injected into the reactor through the nozzles electrode with a flow rate of 0.44 L/min, which corresponds to an average flow velocity of 4.7 m/s. Most experiments were carried out under the initial NO concentration of 100 ppm. The experiment was carried out at room temperature under atmospheric pressure.

In order to observe the ground-state NO profiles in the reactor by PLIF technique, NO $[A^2\Sigma^+(v'=0) \rightarrow X^2\Pi(v''=0)]$ system at 226nm was used. Probe laser pulse of a wavelength of 226 nm was produced by frequency doubling of the laser pulse from a dye laser (Lambda Physik, SCANmate) with Coumarin 47 as a dye, pumped by a XeF excimer laser (Lambda Physik, COMPex 150, tuned at 351 nm). The laser wavelength at 225.547 nm was used for NO PLIF measuring. The laser power was adjusted up to 2 mJ and laser pulsewidth was about 20 ns. A planar laser beam (35-mm height and 1-mm width) was produced using optics, and it passed through central vertical plane between the stressed and grounded electrodes in the reactor. The LIF signal emitted at 90° to the laser sheet was imaged onto a gated intensified charge-coupled device (ICCD) camera (LaVision, Flame Star II) and the 2-D LIF images with the frame size of 240 mm in width and 160 mm in height were observed during the discharge in both the upstream and downstream regions of the reactor.

In the measurement, as the laser pulse induces additional streamers between the regular dc streamer-corona discharges, the light emitted by the laser induced streamer can interfere with the LIF signal during NO PLIF measurement. Fig. 2 shows

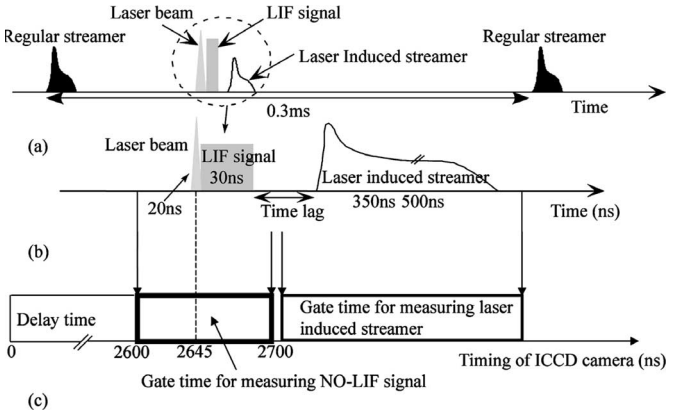

Fig. 2. (a) Schematic illustration of the timing between the regular streamers, laser beam, LIF signal, and laser-induced streamer when a laser pulse is shot between the regular streamer. (b) A detail time relationship between laser beam, LIF signal, and laser-induced streamer. (c) The ICCD gating for the independent measuring of the LIF signal and induced streamer emission. The laser and the ICCD are triggered at "0" ns in axis of "timing of ICCD camera." For capturing the LIF image, an initial delay is necessary.

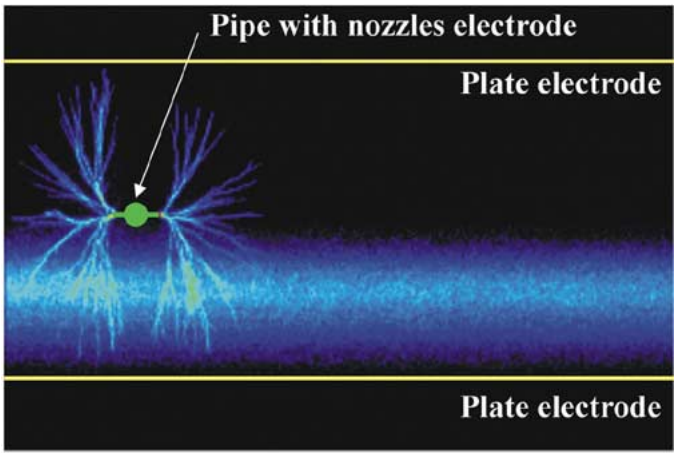

Fig. 3. Simultaneous recording of NO LIF signal and light emitted by the laser-induced streamer. Applied voltage: 27 kV, NO/air 3 L/min. Gate width of ICCD: 490 ns for capturing both the NO LIF signal and the light emitted by the induced streamer corona.

the schematic illustration of the time relationship between the regular streamers, laser pulse, LIF signal, and laser-induced streamer, presented by us in [15]. The LIF signal appears almost immediately after the laser pulse and lasts over about 30 ns, while the laser-induced streamer usually starts later (about 35–300 ns after the laser pulse depending on the position of the laser sheet in the gap) and last over about 350 ns as shown in Fig. 2(b). Consequently, an appropriate adjusting for the delay and exposure time of the ICCD camera enabled to capture the NO image and laser-induced streamer image independently [see Fig. 2(c)]. On the other hand, Fig. 3 shows an example of the simultaneous recording of the NO LIF image and light emitted by the laser induced streamer. The image was obtained with a long exposure time (490 ns). As seen in Fig. 3, as the laser-power density at the center part of the sheet is higher than that of its edges, the LIF signal intensity has a dependence on the location of the laser sheet. To overcome this problem, the LIF image was corrected by dividing the raw LIF image by the beam profile. After that, the absolute concentration of NO

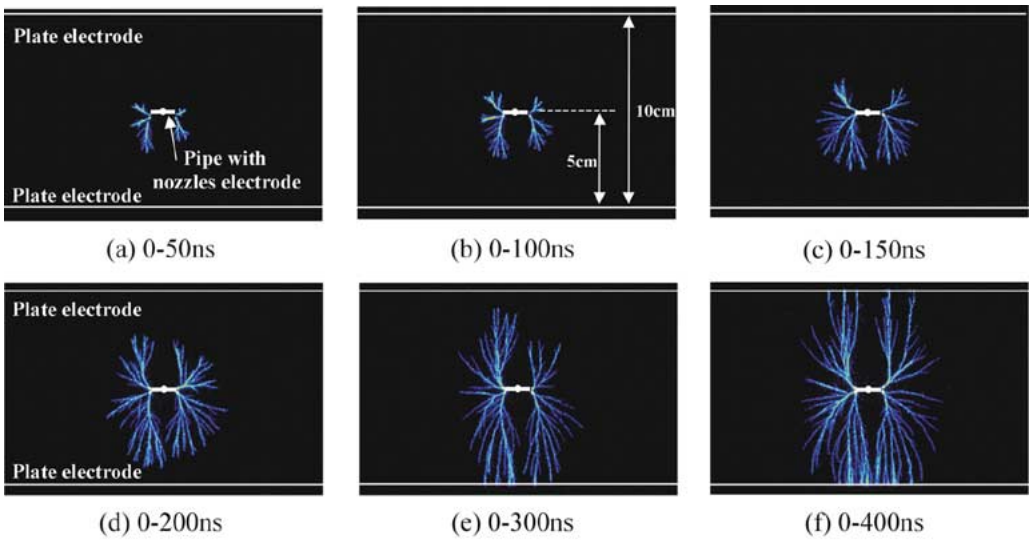

Fig. 4. Images of the streamers induced by a single laser pulse for various exposure times of the ICCD camera. Streamer inception time represented by "0"ns was determined by adjusting the gate time of the ICCD camera. NO(100 ppm)/Air, 3 L/min. Applied voltage: 30 kV. Corona current: 134 μA.

was derived by calibration. The NO LIF signal was calibrated with the measured NO concentrations using a NOx monitor (Hodakatest, Testo 33).

III. Experimental Results and Discussion

A. Streamer-Corona Discharge in Corona Radical Shower System

Fig. 4 shows the time evolution of the streamers in NO/air flow, induced by a laser sheet pulse used for NO PLIF measurement, when there is no additional gas flow through the nozzles electrode. Each image was taken in separate laser irradiation experiments by changing the ICCD camera exposure time. Although each image actually presents a different streamer, similar appearance of the induced streamers was observed with good reproducibility. The streamers propagate from the tip of the nozzles electrode to the plate electrode. They consist of several branches, and the plasma formed by the normal streamers shows a flame-like pattern when we observed the streamer discharge with the naked eye. Similar streamer images were observed even in the case of additional gas (CO_2+air mixture) flow through the nozzles electrode. The volume occupied by the streamers increases with increasing applied voltage. Compared with the induced streamer in the lower part of the reactor, the streamer induced in the upper part of the reactor is delayed. This time delay is due to the distance between the laser irradiation area and streamer generation point on the electrode. The delay time of the induced streamer increases when the distance between the laser sheet and a tip of the nozzles electrode increases [16]. From the time evolution of the streamer discharge shown in Fig. 4, the estimated velocity of the streamer head is about 2.5×10^5 m/s. This result is in a good agreement with our previous results measured by using a photomultiplier tube [17].

Before our first measurement of the NO density profile in the dc needle-to-plate reactor [11], it has been considered that NO removal occurs only in the region of streamer-corona discharge. To clarify the phenomenon in this regard, experimental investigations on this NO removal characteristics by using wide-range NO PLIF were carried out. The results are presented in the next section.

B. 2-D NO Density Profile

When the PLIF technique is used for the NO density distribution monitoring in the NO-seeded airflow without the applied voltage, the obtained NO density profile reflects the treatment gas flow in the reactor. The time evolution of NO density distribution in the reactor after supplying NO(1000 ppm)/N_2 gas into the gas flow is shown in Fig. 5. It is noted that NO(1000 ppm)/N_2 gas was introduced into the established flow of air with injected through the nozzles additional of CO_2+air mixture. The pseudocolor of the images corresponds to NO spatial concentrations as in the NO concentration bar. In order to increase the signal-to-noise (SN) ratio of the image, each image shown in this paper is an average of 50 captured images, which took about 8 s in total. In Fig. 5(a) at $t = 0$, the reactor is not contaminated with NO molecules. As time elapses, the gas flow seeded with NO, moves toward the observation area with a gas flow rate of 3 L/min (an averaged gas velocity of 4.2 mm/s). In this case, the flow field can be evaluated based on the Reynolds number (R_e) defined by

$$R_e = \frac{U_{\rm av} L}{\nu}, \tag{1}$$

where $U_{\rm av}$ is the averaged gas flow velocity, L is the characteristics length based on the reactor or stressed electrode dimension, and ν is the kinematic viscosity of the gas. The Reynolds number (R_e) of the treatment gas flow is 28 and its flow is laminar. The images of NO density distribution in the reactor change gradually with time. Since the residence time of the flowing gas in the reactor is about 2.5 min, the observation area where is in the upstream region of the reactor is uniformly polluted with NO molecules and NO concentration is reached to the initial value of 100 ppm by $t = 4$ minutes [Fig. 5(f)].

On the other hand, Fig. 6 shows the time evolution of NO density distribution in the reactor after supplying NO(1000 ppm)/N_2 gas during the discharge at 27 kV. As the treatment

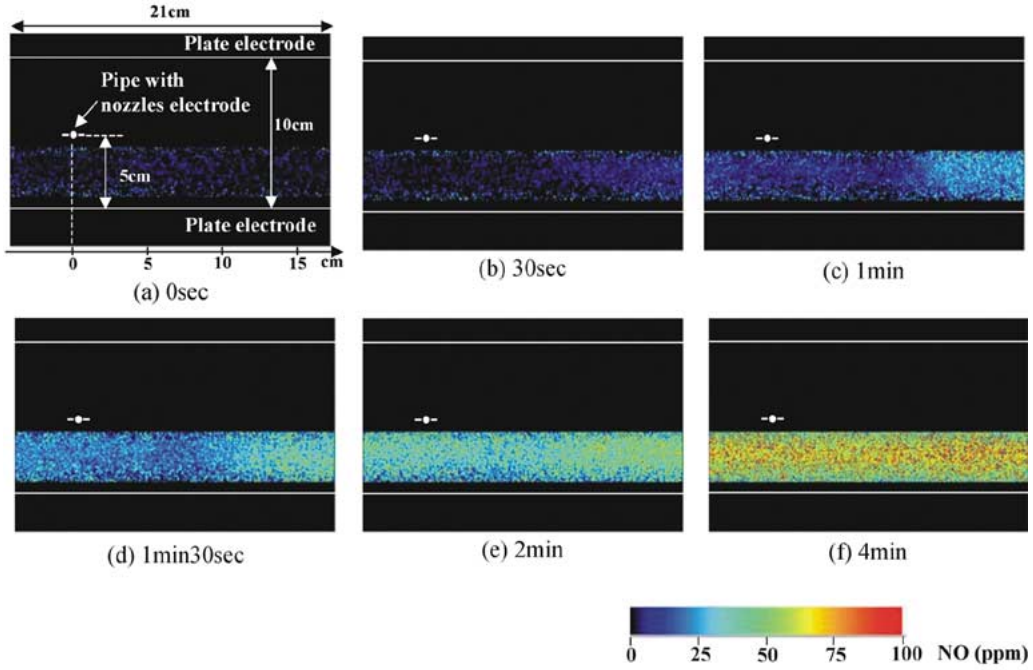

Fig. 5. Wide range 2-D NO concentration profiles under the nondischarge condition as a function of time after NO mixture gas supply.

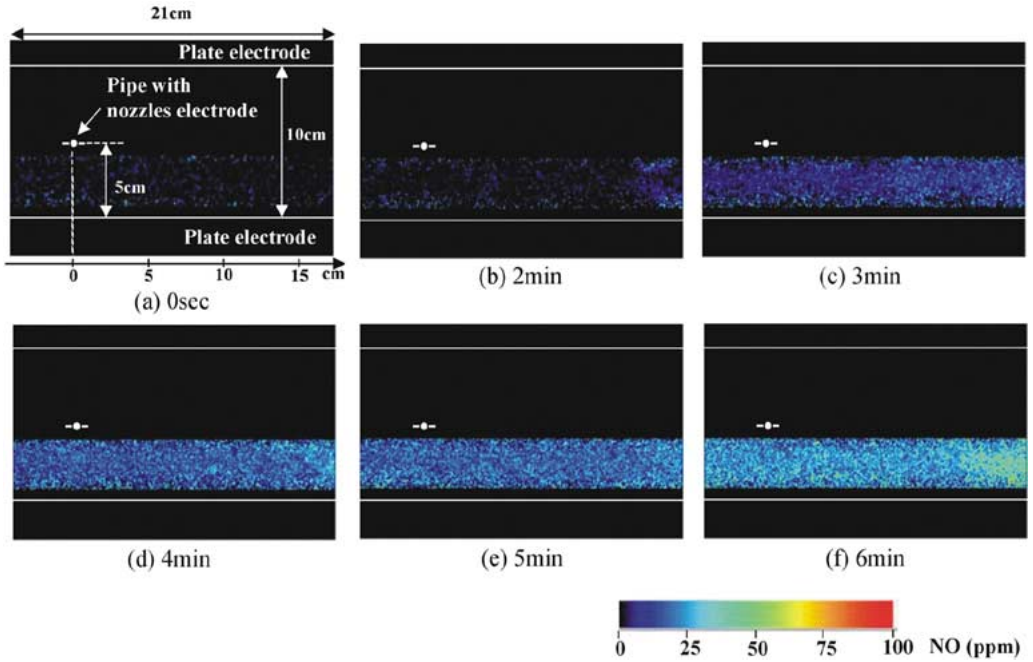

Fig. 6. Wide-range 2-D NO concentration profiles under the discharge condition (27 kV, 83 μA) as a function of time after NO mixture gas supply.

gas flow is disturbed by the discharge-induced secondary flow, the time of NO appearance in the observation area is longer than that of the case without discharge. The secondary flow is generated by the momentum transfer from the ions to neutral gas molecules. In addition, NO concentration became lower due to the NO removal. In this case, NO is mainly converted to NO_2 via the oxidation reactions

$$NO + O + M \rightarrow NO_2 + M, \quad (2)$$

$$NO + O_3 \rightarrow NO_2 + O_2. \quad (3)$$

The increase in NO_2 concentration was confirmed at the reactor outlet by the measurement using the NOx monitor.

Fig. 7(a) and (b) shows NO concentration distributions along the gas flow direction without and with the discharges, respectively. They correspond to the time evolution of NO concentration distribution in the reactor shown in Figs. 5 and 6. It is seen from Fig. 7(b) that the NO depletion region spreads over an upstream region. NO molecule concentration decreased not only in the plasma region created by the streamers but also in the upstream region of the discharge. This result suggests that the EHD flow enhanced the NO treatment in the reactor used in this

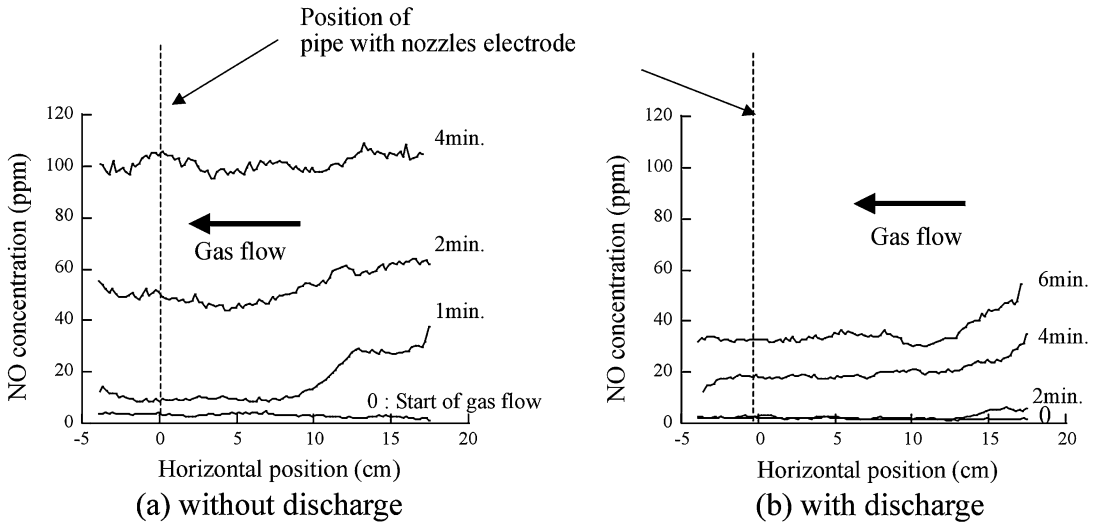

Fig. 7. Spatial NO distribution for various operating times after NO mixture gas supply. (a) Without discharge. (b) With discharge (27 kV, 83 μA). Treatment gas: NO(100 ppm)/air, 3 L/min, Injection gas: mixture of air(0.4 L/min) and CO_2 (40 mL/min).

study. To investigate the influence of the EHD flow, the electrohydrodynamic number (E_{hd}) [18] defined by

$$E_{hd} = \frac{J_p L^3}{\rho_g \nu^2 \mu_i} \tag{4}$$

can be used, where J_P is the current density on the plate electrode, ρ_g is the gas density, μ_i is the positive ion mobility (2×10^{-4} m^2/Vs). The EHD number (E_{hd}) estimated for the case of Fig. 6 (applied voltage: 27 kV, current density on the plate electrode:1.2 μA/cm^2) is 2×10^8. While, Reynolds numbers (R_e) for the treatment gas flow and injection gas flow are 28 and 464, respectively. It is found that since the ratio E_{hd}/R_e^2 is much higher than 1, the gas flow during the discharge is enhanced and the ionic wind (EHD-induced secondary flow) became dominant. The gas flow pattern in this reactor is considered to be more complicated than that in the wire-to-plate electrode or needle-to-plate electrode arrangements. The EHD-induced secondary flow in the present discharge system influences the flow inside the reactor, causing the mixing of the gas in the reactor [19]. This was confirmed by our investigation [20] in which we found ozone far in the upstream region of the reactor, where the ozone was transported by the EHD flow. The ozone transport upstream is a cause of the decrease in NO concentration by reaction (3) in the upstream region of the reactor.

IV. Conclusion

NO density profiles inside the corona radical shower reactor were investigated by using the state-of-the-art planar laser induced fluorescence technique. It was found under the steady-state dc corona discharge condition, that the timing between the streamer discharge pulse, LIF signal, and laser-induced streamer was important for *in-situ* PLIF NO observation. Specifically, separate recording of the LIF signal was possible by an appropriate adjusting of the recording delay and exposure time of the ICCD camera. The images of laser-induced streamers were used to understand the discharge characteristics for NO removal. By comparing the images between the streamer-corona discharge and NO LIF profiles, it was found that NO removal due to oxidation occurred far from the discharge zone in the upstream of the reactor. In the present reactor at a low treatment gas flow rate, EHD flow becomes to be dominant, and ozone transport to the upstream region affects the decrease of NO along the treatment gas flow. Modeling the kinetics of plasma-induced chemical reactions of NOx removal as well as designing the reactors should include the EHD flow effect which may enhance the gaseous pollutant removal in the nonthermal plasma reactor.

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