Two-Dimensional Imaging of NO Density Profiles by LIF Technique in a Pipe With Nozzles Electrode During NO Treatment

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***Abstract*—Two-dimensional NO concentration distribution was studied by a planar laser-induced fluorescence (PLIF) technique in nonthermal plasma during NO treatment. A pipe with a nozzles-to-plate electrode system, having an electrode gap of 50 mm, was used. A stable dc streamer corona discharge was generated in an NO/air mixture at atmospheric pressure. Laser pulses in the form of a sheet were shot between the electrodes during the discharge. LIF signal emitted at 90° to the laser sheet was imaged onto a gated-ICCD camera and two-dimensional distributions of NO concentration in the reactor were measured as a function of time during NO treatment. NO concentration was also monitored at the reactor outlet. The images of NO concentration covering almost the whole length of the reactor show that the density of NO molecule decreased not only in the plasma region formed by corona streamers but also in the upstream region of the reactor. This information is important for modeling and optimizing the plasma processes and designing the nonthermal plasma reactors.**

***Index Terms*—Electrohydrodynamic (EHD) flow, planar laser-induced fluorescence (PLIF), streamer corona discharge, two-dimensional NO concentration distribution.**

I. Introduction

VARIOUS kinds of nonthermal plasma reactors have been proposed in order to remove gaseous pollutants, such as NO_x, SO_2, and volatile organic compounds (VOCs), from exhaust gases [1]–[4]. In order to optimize the reactor and develop the proper model for numerical simulation, it is important to study directly the discharge induced plasma chemical processes responsible for the removal of gaseous pollutants in the reactor. However, in the experimental investigations of NO_x removal, NO concentrations have been measured using conventional NO_x monitors based on the methods of chemi-luminescence, infrared absorption, or potentiostatic electrolysis. In these cases, the information obtained concerns the concentration of NO molecules at the outlet of the reactor.

Laser-induced fluorescence (LIF) can be considered to be a useful diagnostic method for *in-situ* observation of the phenomena in plasma reactors during the NO_x removal process. In recent years, LIF direct measurements of the radicals and molecules concentrations inside the plasma reactors have been successfully demonstrated [5]–[9]. However, these LIF measurements are restricted to the observation after the pulsed discharges in short gap ($<$1 cm) electrode arrangements. Characteristics of the streamer corona discharge and related LIF images when measuring the concentration of NO molecule in the corona discharges with larger gap ($>$3 cm) electrode arrangement by LIF technique were presented [10], [11]. For the industrial application of nonthermal plasma in the field of flue gas emission control, larger electrode gap arrangement is necessary for a scale-up or a retrofit. Recently, a corona radical shower system, which is suitable for the retrofit of the existing electrostatic precipitators, has been proved to be one of the most efficient nonthermal plasma reactors used for NO_x removal [12]. Besides this, the overall pressure drop for long gap arrangement is much smaller compared with that of short gap type reactor. For an industrial scale, we have to minimize plasma reactor overall pressure drop.

In the corona radical shower reactor, an electrode with one or several nozzles is used for additional gas injection across the corona discharge zone into the flue gas [13], [14]. By using a pipe with nozzles electrode as a stressed electrode, the discharge mode is much different from that of the conventional electrode arrangement, such as wire-to-plate or needle-to-plate. The differences concern distributions of the electric field and current density, as well as the secondary flow. In particular, when NH_3 gas is injected into the reactor through the nozzles, the discharge mode changes with operational time due to the deposition of aerosol particles (NH_4NO_3) onto the electrodes and reactor wall [13]. In this paper, as one of the fundamental studies on NO LIF measurement in steady-state streamer discharge using the pipe with nozzles electrode, NO LIF images for measuring the concentration of NO molecules in a corona radical shower re-

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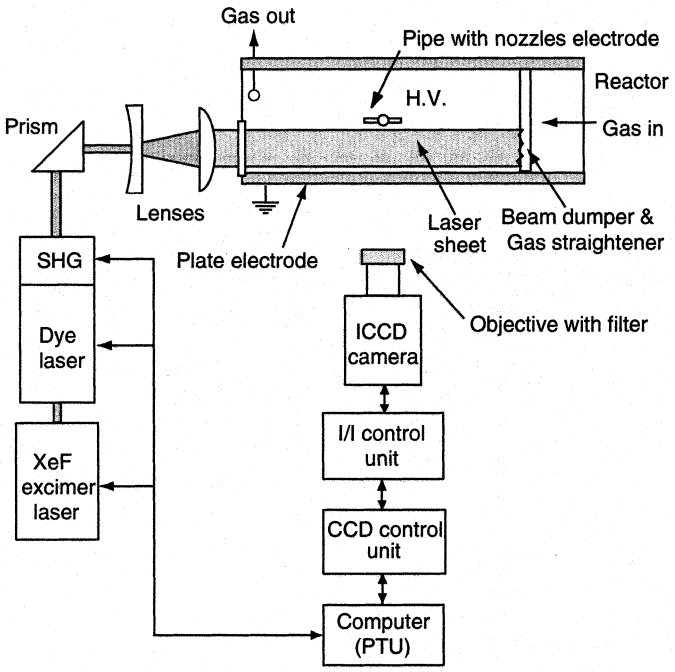

Fig. 1. Schematic diagram of the experimental apparatus. In the ICCD camera control unit, I/I means an image intensifier and PTU a programmable timing unit.

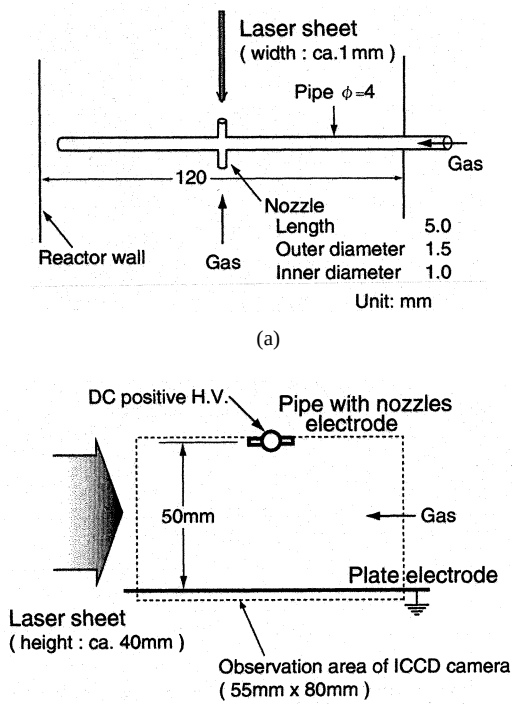

Fig. 2. Schematic diagram of the discharging electrode and its arrangement. (a) Top view. (b) Side view. The dashed rectangle indicates the typical area of LIF measurements. The laser sheet is about 35 mm in height and 1 mm in width.

actor without injection gases are presented. For this purpose, the planar LIF (PLIF) technique was used to monitor two-dimensional NO molecule distribution in the reactor during NO treatment.

II. Experimental Apparatus and Methods

The schematic diagram of the experimental apparatus is shown in Fig. 1. In order to detect the ground-state NO molecules, NO $(A^2\Sigma^+(v'=0) \leftarrow X^2\prod(v''=0))$ system at 226 nm was used [15]. The laser pulses from a XeF excimer laser (Lambda Physik, COMPex 150, tuned at 351 nm) pumped a dye laser (Lambda Physik, SCANmate) with Coumarin 47. The dye laser generated a laser beam of a wavelength tuned around 450 nm. A BBO crystal (BaB_2O_4) pumped by the tuned dye laser beam produced the second harmonic radiation of a wavelength correspondingly tuned around 226 nm. The 226-nm laser beam pulses of energy of 0.8–2 mJ and duration of about 20 ns, transformed into the form of a 1–mm-width and 40-mm-height laser sheet passed between the electrodes with a 50–mm gap, in which NO oxidation by the streamer corona discharge occurred. To suppress the light scattering, the laser pulses were absorbed in a set of two ceramic plates with small holes (a beam dumper) placed at rear of the reactor as shown in Fig. 1. The reactor was an acrylic box (100 mm $\times$ 120 mm $\times$ 700 mm) in which quartz windows were mounted for the transmission of the laser pulses and the LIF signal detection. As a discharging electrode, a pipe with nozzles electrode was used and mounted in the middle between two grounded parallel plate electrodes. The pipe with nozzles electrode consisted of a stainless pipe (4 mm in diameter) and nozzles (1.5 mm outer diameter, 1 mm inner diameter), as shown in Fig. 2.

The LIF signal emitted at 90° to the laser sheet was imaged onto a gated ICCD camera (LaVision, Flame Star II) and two-dimensional NO concentration profile data under the steady-state corona operation were acquired and processed by a computer. In order to determine a proper laser wavelength for NO LIF measurements, the laser wavelength was scanned through NO $[A^2\Sigma^+(v'=0) \leftarrow X^2\prod(v''=0)]$ transitions in the range of 225–227 nm and the corresponding NO excitation spectrum was monitored. The laser wavelength at 226.19 nm, at which NO LIF signal was strongest, was chosen for NO LIF measuring. The procedure of setting for the delay and exposure time of the ICCD camera has been described in a previous paper [10], [11]. It was possible to observe the NO LIF signal under the steady-state discharges by adjusting the delay time and gate time of the ICCD camera. DC high voltage with positive polarity was applied through a 10-MΩ resistor to the pipe with nozzles electrode, and the streamer corona discharge was generated.

Dry NO(200 ppm)/air mixture was introduced to the reactor. The gas flow rate was 1–4 L/min. At this stage of the experiment there was no additional gas (such as NH_3 or CH_4) injected through the nozzle into the main gas stream, unlike in the typical corona radical shower discharges [13], [14]. Concentrations of NO and NO_2 were measured using an NO_x monitor (Hodakatest, Testo 33). NO LIF at 226.19 nm was calibrated as a function of NO concentrations in the gas sampled at the LIF observation area as shown in Fig. 2(b). Absolute concentration of NO has been derived by the calibrated LIF data [10]. It was confirmed that the LIF signal was proportional to the NO concentration up to about 300 ppm. The experiment was carried out at room temperature under atmospheric pressure.

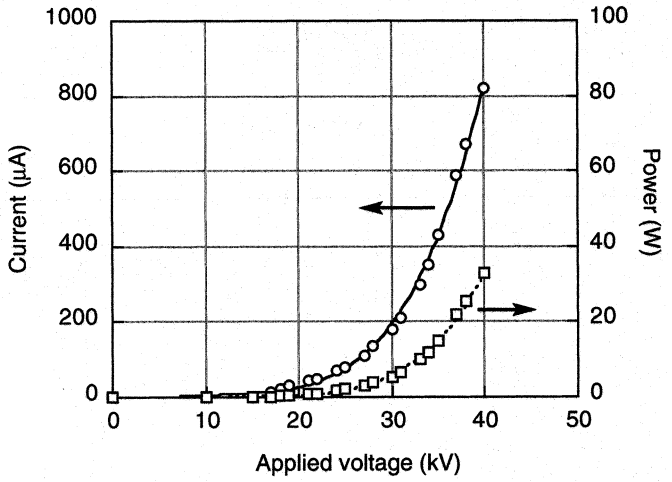

Fig. 3. Time-averaged current and power–voltage characteristics for corona radical shower in NO/air mixture at 1 L/min.

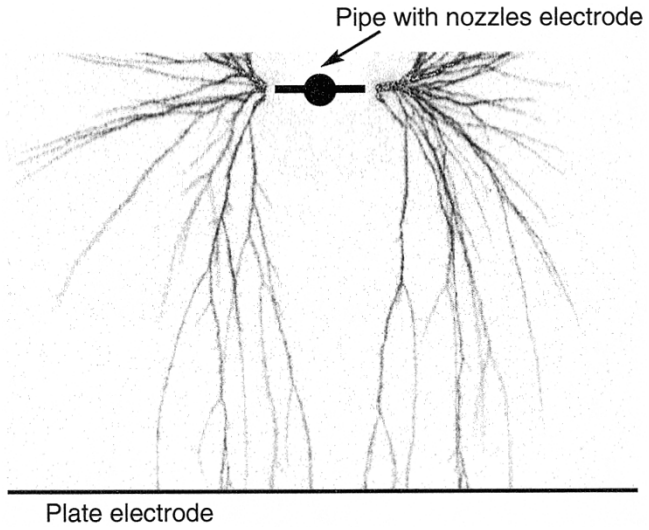

Fig. 4. Typical streamer corona image in NO/air mixture observed with the gate width of the ICCD camera 500 ns. The applied voltage to the nozzle electrode is 30 kV and corona current is 200 μA.

III. Experimental Results and Discussion

A. Discharge Characteristics

Fig. 3 shows time-averaged discharge current and corresponding power–voltage characteristics of the streamer corona discharge in the two-nozzles-to-plate electrode system shown in Fig. 2. When the applied voltage is increased to about 15 kV, the corona discharge occurs. Then, the current increases with increasing voltage. The dc streamer corona discharge consists of tiny pulsed discharges (peak current 10–50 mA, pulse width 350–500 ns) with a relative high repetition rate (about 2–7 kHz). Fig. 4 shows an image of the streamers taken with the ICCD camera. The streamers consist of several branches and propagate from the tip of the nozzles electrode to the plate electrode, bridging the gap between the electrodes. The volume occupied by the streamers increases with increasing applied voltage. Also, the streamer repetition rate increases with increasing applied voltage. In the corona radical shower discharge, the plasma formed by the streamers shows a flame-like pattern when we recorded with a long ICCD exposure time or observed with the naked eye.

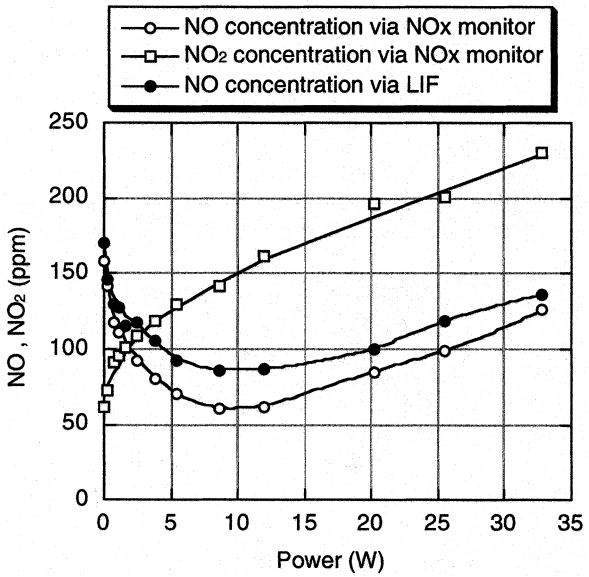

Fig. 5. NO and NO_2 concentrations as a function of the discharge power. Initial concentration of NO is 200 ppm at the inlet of the reactor (operating gas: NO (200 ppm)/air, gas flow rate: 1 L/min).

B. NO Treatment by Streamer Coronas

NO and NO_2 concentrations as a function of applied discharge power are shown in Fig. 5. Dry air with NO (200 ppm) at a flow rate of 1 L/min was introduced to the reactor with the averaged gas velocity of 2 mm/s. The dc discharge power was calculated by multiplying the applied voltage and time-averaged corona current. NO concentration was measured by both NO LIF detected near the stressed electrode as shown in Fig. 2(b) and the NO_x monitor placed at the reactor outlet. The measurement of NO LIF was carried out after the NO concentration measured by the NO_x monitor reached a steady state. From the LIF images in the vicinity of the discharging electrode, corresponding to the area shown in Fig. 2(b), we derived the NO concentration in parts per million of NO. As the oxidation of NO occurs even downstream of the discharging region, NO concentration measured by the LIF is higher than that by the NO_x monitor placed at the reactor outlet. The natural oxidation of NO also occurs when the gas flows through the reactor. As seen in Fig. 5, the difference in NO concentrations between the NO measured by the LIF and that of the NO_x monitor at 0 W indicates the amount of naturally oxidized NO molecules when they travel with the flowing gas from the LIF observation area (around the pipe electrode) to the reactor outlet. For a discharge power lower than 10 W, NO is oxidized to NO_2 by the streamer discharge, while both NO and NO_2 concentrations increase with increasing discharge power above 10 W, indicating the formation of NO_x.

C. Time-Dependent NO Concentration and Two-Dimensional NO Distribution

In order to investigate the NO treatment process, LIF data were also collected as a function of time after a high voltage was applied to the electrode. We applied the high voltage at $t = 0$ s

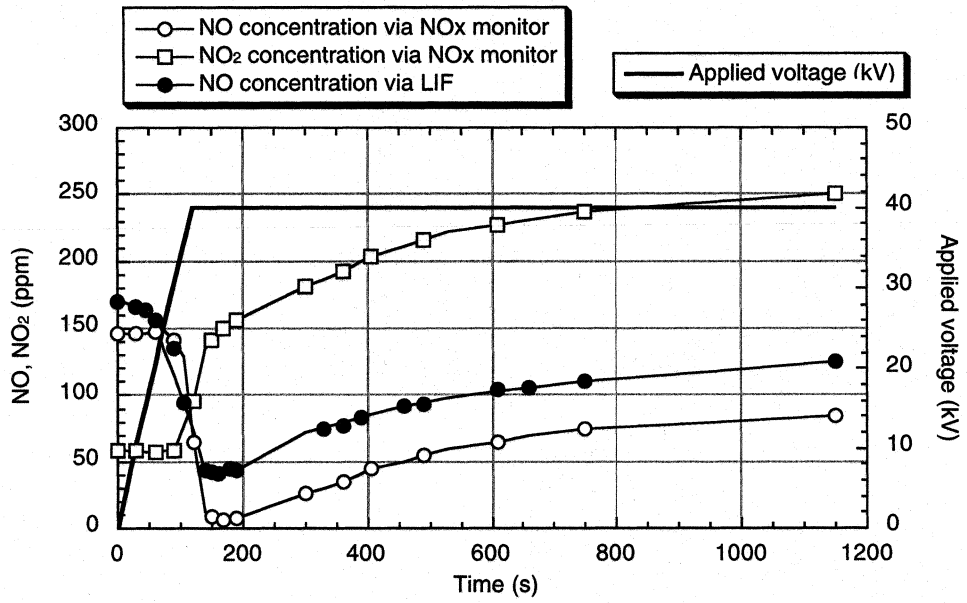

Fig. 6. Time-dependent characteristics of the NO treatment process: the high voltage was applied at t = 0s (operating gas: NO (200 ppm)/air, gas flow rate: 1 L/min).

and increased it linearly until 40 kV at a constant rate as shown in Fig. 6. NO concentration was measured by both the NO LIF detected near the stressed electrode as shown in Fig. 2(b) and NO_x monitor placed at the reactor outlet. As the time elapsed after the discharge start, NO concentration decreased and NO_2 concentration increased as shown in Fig. 6. Due to the low gas velocity (2 mm/s), the gas residence time in the reactor was relatively long (about 5 min) and, therefore, NO converted gradually with time to NO_2 by the oxidation. It is considered that the NO signal measured by LIF corresponds to the real-time NO concentration in the discharge region, while NO concentration measured by the NO_x monitor has a time lag due to both the delay time for the gas sampling and the response time of the NO_x monitor. Fig. 6 also shows that NO concentration decreased at the beginning of the discharge and then increased with time. After that, NO concentration reached a steady state as the discharge further continued.

Fig. 7 shows two-dimensional images of NO concentration in the discharge and surrounding areas as a function of time during the NO treatment when the operating voltage was varied as shown in Fig. 6. The intensity of the images corresponds to NO spatial concentrations. Since the laser power density within the sheet was not uniform, the image was corrected by dividing the raw image by the beam profile. Moreover, in order to increase SN ratio of the image, each image presented in Fig. 7 is an average of ten captured images. After the improvement of the images, however, there is still a vertical nonuniformity of NO density due to a lack of vertical uniformity of the laser sheet. As the time elapsed, the NO LIF signal became weaker compared to that of the earlier time period, indicating the decrease of NO concentration. After 170 s from the start, the spatial NO distribution reached minimum density, then, NO concentration increased with time.

To further investigate the NO density distribution inside the reactor, wider LIF images were acquired that had an LIF observation area corresponding to the 25-cm length by 5-cm height. Fig. 8 shows images of NO density profiles for different gas flow rates during the discharge. The images were taken after the NO concentration in the reactor reached a steady state and averaged over ten images. It is seen from the images in Fig. 8 that 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 may suggest that the electrohydrodynamic (EHD) flow enhanced the NO treatment in the reactor used in this study. Since the gas flow velocity was low, the ionic wind (EHD-induced secondary flow) became dominant. For the conventional electrode arrangement such as wire-to-plate electrode or needle-to-plate electrode with a present discharge current level (100–800 μA), the EHD flow velocity reaches by several meters per second [16], [17]. In a hollow needle-to-plate electrical discharge, which resembles the corona radical shower discharge, the velocity flow pattern [18] is even more complicated than that in the wire-to-plate electrode or needle-to-plate electrode arrangements due to the additional flow through the hollow needle. The EHD-induced secondary flow in the present discharge system influences the flow pattern inside the reactor (i.e., mixing of the gas occurs in the reactor). This is a cause of the decrease of NO concentration measured in the upstream region of the reactor. In Fig. 8(a), which corresponds to the gas flow rate of 2 L/min and an averaged gas flow velocity of 4 mm/s, the NO depletion region spreads over an upstream region with a distance that is a few times of the gap distance. When the gas flow rate was increased to 4 L/min, NO depletion region reduced as shown in Fig. 8(b). The experimental results of Figs. 7 and 8 are particularly important for the modeling of the nonthermal plasma processes and the designing of the reactors. In order to investigate the flow structures affecting the NO density profile in detail, flow visualization inside the corona radical shower reactor is currently in progress.

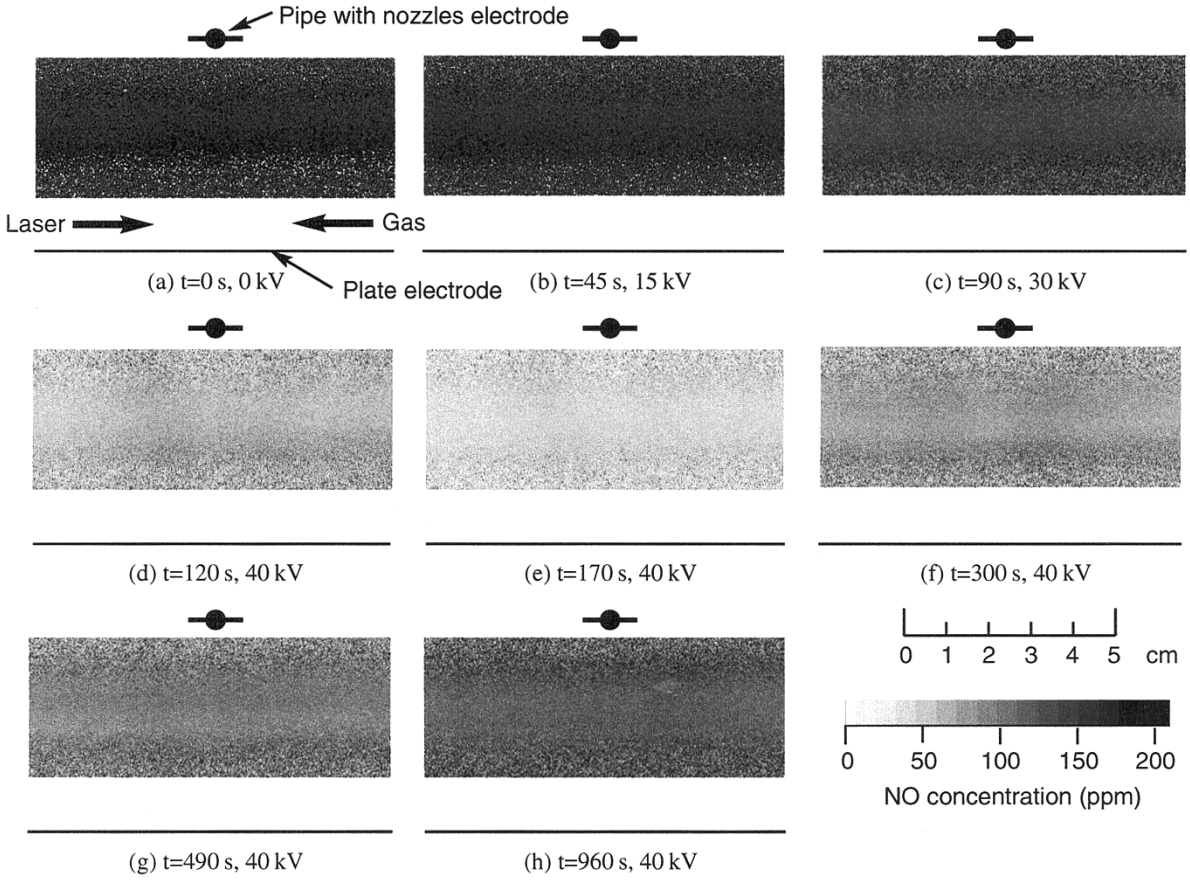

Fig. 7. Two-dimensional time-dependent concentrations of the ground-state NO molecules. Parameters as in Fig. 6.

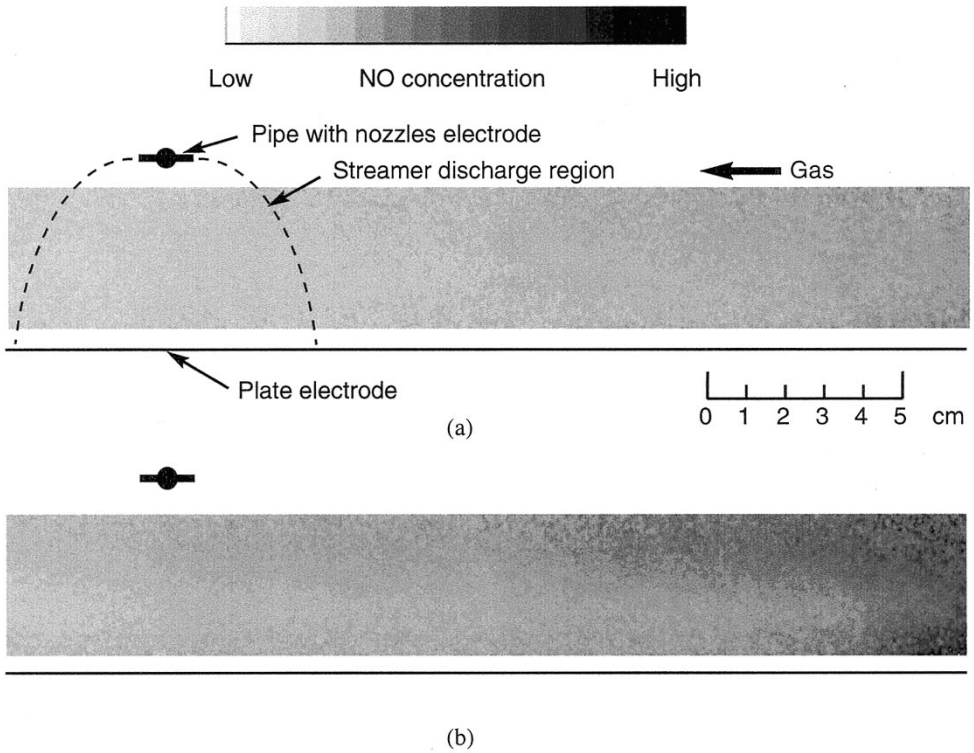

Fig. 8. The wide-space two-dimensional LIF observation in the upstream direction of the gas flow inside the reactor during the discharge. (a) Gas flow rate 2 L/min, applied voltage 37 kV, and corona current 500 μA. (b) Gas flow rate 4 L/min, applied voltage 35 kV, and corona current 550 μA. The pipe with nozzles electrode is not shown in real scale.

IV. Conclusion

Two-dimensional NO concentration profile measurements were conducted during NO treatment in the discharge with larger electrode gap in the pipe with nozzles-to-plate electrode system using the LIF diagnostic system.

The results are summarized as follows.

1) The streamer coronas were visualized with the gated-ICCD camera. The detailed features of the streamer propagation in the reactor using the pipe with nozzles electrode were clearly observed.
2) The two-dimensional distribution of the ground-state NO molecule concentration between the electrode gap could be seen directly on the display of the LIF system. The LIF images provided dynamics of NO distribution inside the reactor in real time. The NO LIF measurements were compared to the measured NO concentration using the NO_x monitor. The time evolution of NO concentration in the reactor was studied using both methods. It was found that LIF measurements provided the NO concentration as well as its distribution in the reactor.
3) The NO molecule concentration decreased not only in the plasma region created by the streamers but also in upstream region of the reactor. The wide space LIF monitoring showed that NO molecule depletion due to their oxidation occurs far from the discharge zone in the upstream.

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