Streamer Corona Discharge Induced by Laser Pulses During LIF Measurements in a DC Non-thermal Plasma Reactor for NO Oxidation

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Abstract: A possible interference of the light emitted by the laser induced streamer coronas and the laser induced fluorescence (LIF) signal during the measurements of NO concentration in a DC positive corona discharge was experimentally investigated. For *in-situ* NO measurement a LIF system consisting of a XeF excimer laser, dye laser and BBO crystal, generating a tuned laser line at 226 nm was employed. From the measured occurrence timing between the regular streamer coronas, laser pulse, LIF signal and laser induced streamer it was found that the LIF signal appears almost immediately after the laser incidence and lasts over about 30 ns, while the induced streamer starts about 35 ns after the LIF signal and lasts about 350-500 ns. Due to the 5 ns interval between the LIF signal and the laser induced streamer the undisturbed detection of the LIF signal can be possible with a properly adjusted timing of the ICCD camera (the gate opening and exposure time). Two-dimensional distribution of NO molecules concentration in the discharge gap was measured using the LIF technique. The time-resolved evolution of the laser induced streamers was visualized using the ICCD camera with the proper timing adjustment. This resulted in determining the velocity of propagation of the streamer (about 2.5×10^5 m/s) and the averaged diameters of the leader channel and leader streamers (200 μm and 100 μm, respectively).

Introduction

Recently, non-thermal plasma processing has been considered to be one of the most efficient methods for NOx removal (*1*). In order to improve NOx removal performance and optimize the reactor design, it is important to study the discharge-induced plasma chemical reactions, causing NOx removal, in the reactor. The laser diagnostic method including the diagnostics based on Mie scattering, Rayleigh scattering, Raman scattering, laser induced fluorescence (LIF) and particle imaging velocimetry (PIV) is suitable and convenient for basic and applied studies of many transient or turbulent media, including those in the electrical discharges employed in the non-thermal plasma reactors for gaseous pollutant abatement.

The potential of LIF method based on tunable UV laser for diagnostics of transient discharges has been already proved experimentally by Ershov and Borysow (*2*), who investigated time transient density of OH radicals in pulsed discharges. Using LIF method Coogan and Sappey (*3*) have imaged the distribution of the OH radicals within the silent discharge plasma reactor. Recently, Ono and Oda (*4*, *5*) observed spatial and temporal distribution of OH radicals in various kinds of pulsed discharges. While, Hazama et al. (*6*), Roth and Gunderson (*7*), and Tochikubo and Watanabe (*8*) have studied behavior of NO molecules in a pulsed corona discharge by LIF technique. Recently, Fresnet et al. (*9*) also using LIF technique measured NO concentration after a single-pulse discharge. The results obtained by these authors concerned the NO behavior during the post-discharge time interval in either the single transient discharge or low-repetition pulsed discharges in the electrode arrangements with gaps shorter than 1cm.

However, there is an interest in the *in-situ* LIF measurements of various species, such as radicals and molecules in DC corona discharges, which have potential for efficient oxidation of NO molecules, e.g. in a corona radical shower system (*10*).

The typical DC streamer corona discharge is a train of streamers occurring almost regularly with a relatively high repetition rate (about 4 kHz, Figure 1).

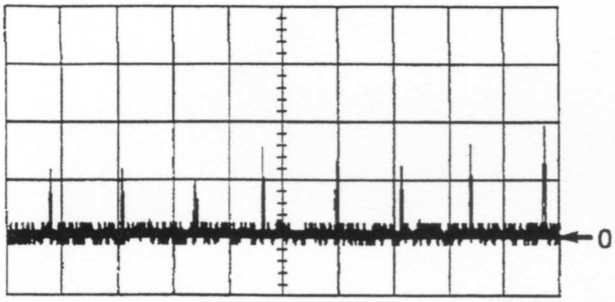

Figure 1. Regular current pulses during the DC positive streamer corona mode in air (applied voltage: 24 kV, time averaged current: 0.16 mA, 0.2 ms/div., 20 mA/div.).

Therefore, the light emitted by these regular streamers can interfere with the LIF measurements around the discharging zone, because LIF signal is weaker than the discharge emission.

Also streamers induced by the UV laser employed to generate the LIF signal may become a real obstacle for the LIF measurements in the DC streamer corona discharges. It is known that high power laser pulses are capable of triggering discharges (*11*). We observed earlier (*12*) that also laser pulses of much lower power, typical of the LIF measurement, induced corona streamers. Such streamers can make the LIF measurement in the DC corona discharge difficult or even impossible.

This work was aimed at experimental testing how the laser induced streamer coronas can affect the LIF measurements of NO concentration in a DC 3 cm-gap positive corona discharge reactor employed for NO oxidation.

Experimental Section

The usefulness of the LIF technique for the monitoring of NO molecules in the high-repetition DC corona discharge was tested employing an experimental apparatus shown in Figure 2. This is a typical setup based on a tunable UV laser, suitable for the LIF monitoring of NO molecules in various vibrational states. The DC corona discharge was realized in open air.

For the monitoring of the NO molecules in the ground state, NO ($A^2\Sigma^+$ (v' = 0) $\leftarrow$ $X^2\Pi$ (v" = 0)) transition at 226 nm was used. The laser pulses from a XeF excimer laser (Lambda Physik, Compex 150, tuned at 351 nm) pumped a dye laser (Lambda Physik, Scanmate) that generated a laser beam of a wavelength tuned around 450 nm. A BBO crystal pumped by the tuned dye laser beam produced the second harmonic radiation of a wavelength correspondingly tuned around 226 nm. The 226 nm laser

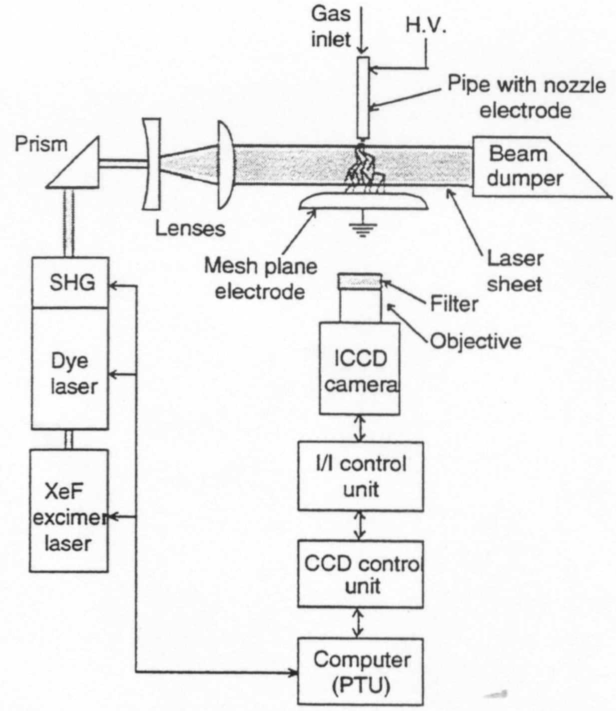

Figure 2. Schematic diagram of the experimental apparatus.

beam pulses of energy up to 2 mJ and constant duration of about 20 ns, transformed by lenses into a 1 mm-thick and 25 mm- height laser sheet passed between the needle-to-plane electrode with a 30 mm gap, in which NOx removal from polluted gas occurred. Two-dimensional images of the corona streamers and LIF signals from the discharging region were recorded by a gated ICCD camera (LaVision, Flame Star II) placed perpendicularly to the laser sheet. A stainless-steel pipe with a nozzle (1.0 mm in inner diameter, 1.5 mm in outer diameter) was used as the needle electrode. The plane electrode was a stainless-steel mesh plate (100 mm in diameter). DC high voltage with positive polarity was applied through a 10 MΩ resistor to the needle electrode. The operation in the streamer corona discharge mode was established by adjusting both the length of the nozzle part of the needle electrode and the operating voltage.

The timing between the regular streamers, laser pulse, LIF signal and induced streamer was investigated. The streamer images and LIF signals were monitored by the ICCD camera. While, the laser pulses were monitored using a biplanar phototube (Hamamatsu, R 1193U-55). The streamer current pulses were measured using a current probe (Pearson Electronics, Inc. Model 2878). The signals from the biplanar phototube and current probe were recorded by a digital oscilloscope (LeCroy, 9362).

The present LIF measurement system also enabled monitoring the NO molecules in the ($X^2\Pi$ (v" = 2) vibrational state using the NO ($A^2\Sigma^+$ (v' = 0) ← $X^2\Pi$ (v" = 2)) transition at 248 nm. In this case, generation of the laser line at 248 nm was attainable from the excimer laser setup only, after changing its working gas from XeF to KrF.

As a test gas, air polluted with NO was introduced into the discharging region through the nozzle electrode, like an additional gas in the corona radical shower reactor (*10*). The gas flow rate was 0.2-1 L/min. The experiment was carried out at room temperature under atmospheric pressure.

Results and Discussion

We found that every laser pulse of energy higher than 0.8 mJ (or energy density of 3.2 mJ/ cm^2) induced a streamer in the gap when the applied voltage was higher than the onset voltage of the regular streamer coronas (Figure 3). In the LIF measurement, we used laser pulses having energies higher than the threshold energy for inducing the streamers in order to obtain an adequate LIF signal intensity. Moreover, the streamer coronas were also induced at the applied voltage that is lower 1-2 kV than the onset voltage. Therefore, the light emitted by both streamers, the regular and the induced by the laser beam may coincide with the LIF signal and hinder the LIF measurements, mainly because the intensity of the light emitted by the streamers is much stronger than that of the LIF.

The current peaks of the laser induced streamers were found to be lower by about 50 % on average than those of the regular streamers. This is likely a result of lowered applied voltage on the stressed electrode in the time interval between two regular streamers, in which the induced streamer occurs. We did not observe any essential differences in the appearances of both regular and induced streamers. However, the laser induced streamers are remarkably alike in appearance.

Figure 4 shows the typical time relationship between the regular streamer coronas, laser pulse, LIF signal and laser induced streamer. The typical time interval between the regular streamers is equal to about 250 μs. Their duration is about 350 ns. Therefore, the probability of coincidence of the regular streamers and a laser pulse having duration of about 20 ns, although much higher than in the pulsed coronas used in the previous investigations (*6-9*), is very low, and in this aspect the regular streamers represent no obstacle for the LIF measurements in the DC corona discharge. On the other hand, the laser induced streamers may essentially interfere with the LIF measurements in the DC corona discharges. The LIF signal appears almost immediately after the laser pulse and lasts over about 30 ns. About 5 ns after the LIF signal, the induced corona streamer appears and it lasts about 350-500 ns. Therefore, due to this 5 ns interval there is no overlapping of the LIF signal and the light emitted by the induced streamer. If the monitoring of the LIF signal lasts longer than about 35 ns, the LIF signal and the light emitted by the induced streamer are recorded as simultaneous signals, and the LIF measurement becomes doubtful. However, the undisturbed detection of the LIF signal

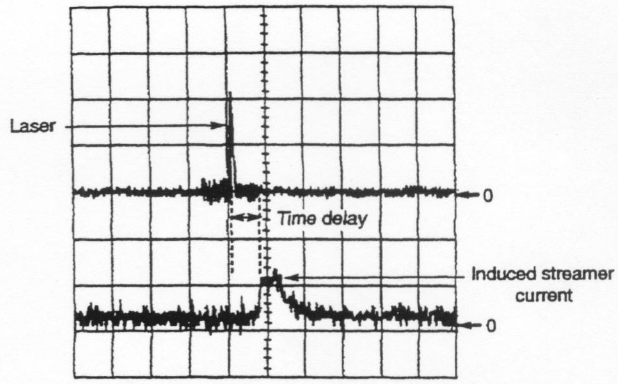

Figure 3. Time relationship between the laser sheet pulse (λ = 226 nm) and induced streamer in air (applied voltage: 25 kV, 200 ns/div, 20 mA/div.).

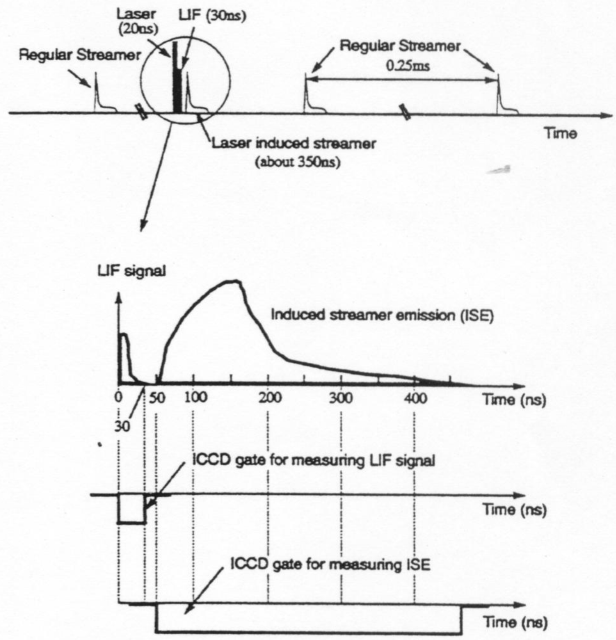

Figure 4. Timing between the regular streamers, laser sheet pulse (λ = 226 nm), LIF signal and laser induced streamer when a laser pulse is shot between the regular streamers. The ICCD gating for the independent measuring of the LIF signal and the induced streamer emission (ISE) is also illustrated.

can be possible with the properly adjusted timing of a fast-gated ICCD camera (the gate should be opened just after the laser pulse and the exposure time should be equal to the duration of the LIF signal, i.e. about 30 ns). Also, as seen from Figure 4, the separate monitoring of the induced corona streamer with the ICCD camera is possible, if appropriate adjusting the recording delay and exposure time of the ICCD camera is set.

Figure 5 shows an example of the two-dimensional LIF image of NO concentration in the flow of NO (200 ppm)/air mixture injected through the nozzle electrode into the discharging region, obtained with an ICCD camera exposure time of 100 ns. This relatively short exposure time prevented the LIF signal to be recorded together with the light emitted by the laser induced streamer. However, if the exposure time was longer, the ICCD camera recorded both the NO LIF signal and the light emitted by the induced streamer corona (Figure 6, ICCD exposure time: 200 ns). The overlapping of both signals, very clearly seen just below the nozzle electrode in Figure 6 makes interpretation of the image obtained very difficult.

The proper timing adjustment of the ICCD camera enabled observing the time-resolved evolution of the laser induced streamers. Different stages of the propagation of the streamer, induced by a laser pulse (λ = 226 nm) having a low energy density of 8 mJ/cm^2, from the needle electrode to the plate electrode are presented in Figure 7. Each image is a result of the accumulation of 30 images of the individual streamers, observed by the ICCD camera with fixed exposure time at 20 ns for different delay times between the streamer onset and the opening of the ICCD camera gate.

Figure 8 shows the time evolution of the streamers in air, induced by a KrF laser sheet pulse (λ = 248 nm). The energy density of the laser pulse was about 1.2 J/cm^2, i.e. it was much higher that that when the laser line λ = 226 nm was used (Figure 7). Differently to the case shown in Figure 7, each image was taken by changing the ICCD camera exposure time. Like in Figure 7, each image actually presents a different streamer. However, as we mentioned above, the laser induced streamers are alike in appearance. It seems that the laser sheet pulse forms an ionization sheet between the electrodes, which is a kind of guide for the induced streamer, affecting the trajectory of its propagation. This may be a reason of similar appearance of the laser induced streamers. Similar behaviour of laser induced streamers was studied in a DC glow corona with much intense laser beam (*13*).

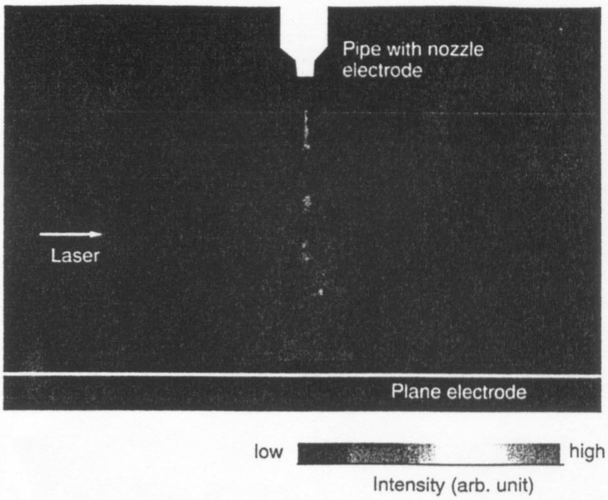

Figure 5. LIF image of NO molecules injected through the nozzle electrode into the discharging region. Laser beam wavelength: 226.19 nm, exposure time of the ICCD camera: 100 ns, working gas: NO (200 ppm)/air, flow rate: 1 L/min, applied voltage: 25 kV, time averaged current: 0.2 mA. NO concentration corresponds to colors in the color bar (in arb. units).

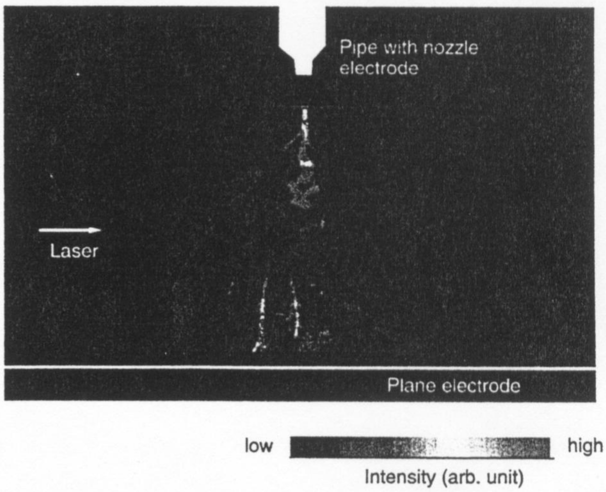

Figure 6. Simultaneous recording of NO LIF signal and light emitted by the laser induced streamer. Laser beam wavelength: 226.19 nm, exposure time of the ICCD camera: 200 ns, working gas: NO (200 ppm)/air, flow rate: 1 L/min, applied voltage: 25 kV. NO concentration corresponds to colors in the color bar (in arb. units).

The velocity of propagation of the streamer, estimated from the time evolution of the streamer shown in Figures 7 and 8 is about 2.5×10^5 m/s. This result is in agreement with our previous results obtained using a photo-multiplier tube (*14*). The averaged diameters of the leader channel and leader streamers measured on the recorded images were 200 μm and 100 μm, respectively. These values are close to those of 150-300 μm reported in the reference (*15*).

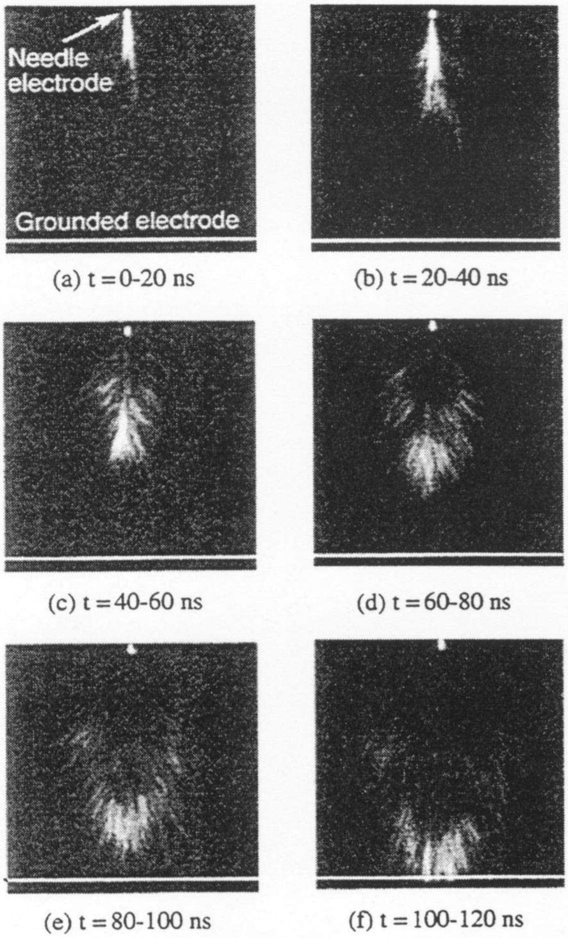

Figure 7. Time evolution of the positive streamer induced by a single laser pulse at λ = 226 nm in air. Each image is an accumulation of 30 individual streamers. Laser pulse energy density: 8 mJ/cm^2, exposure time of the ICCD camera: 20 ns, applied voltage: 23 kV.

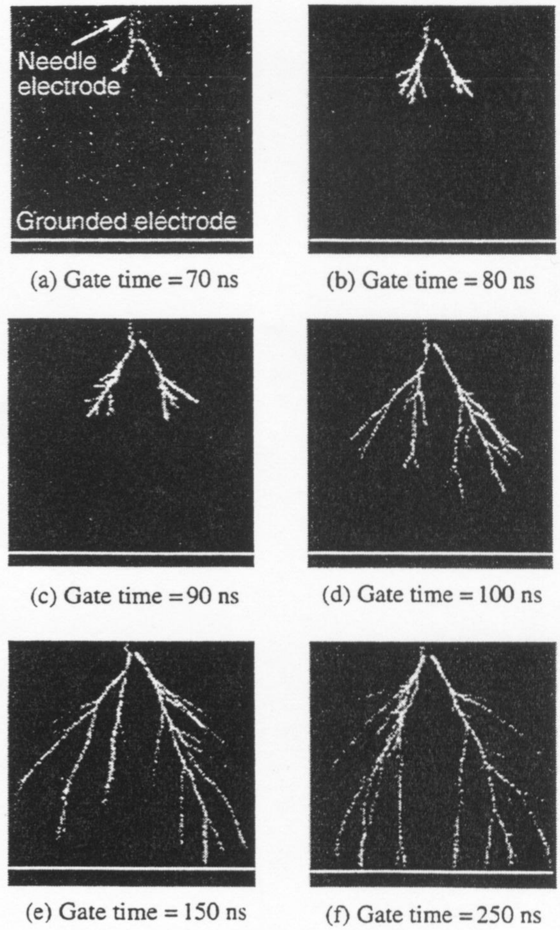

Figure 8. Images of the positive streamer induced by a single KrF laser pulse (λ = 248 nm) for various exposure times of the ICCD camera. Laser pulse energy density: 1.2 J/cm^2, applied voltage: 25 kV.

When KrF laser pulses were shot into the discharge gap, through which NO (1000 ppm)/air mixture flowed, no NO LIF signal was detected. This is comprehensible because the gas had room temperature and the population of the NO molecules in the $X^2\Pi$ (v" = 2) state was very low.

Conclusions

The experimental testing how the laser induced streamer coronas can affect the LIF measurements of NO concentration in the DC 3 cm-gap positive corona discharge employed for NO oxidation was carried out. The results are summarized as follows:

(1) Streamer coronas are induced in the discharge gap by the UV laser pulses, which are shot there for LIF measurement. The light emitted by the laser induced streamer may interfere with the LIF measurement since they occur even at the applied voltage lower than the corona onset.

(2) Occurrence timing between the regular streamer coronas, laser pulse, LIF signal and laser induced streamer was determined. It was found that the LIF signal appears almost immediately after the laser pulse and lasts over about 30 ns, while the induced corona streamer starts about 5 ns after the LIF signal and lasts about 350-500 ns.

(3) Due to the 5 ns interval between the LIF signal and the laser induced streamer the undisturbed detection of the LIF signal can be possible with properly adjusted timing of the ICCD camera (the gate opening and exposure time).

(4) Two-dimensional distribution of NO molecules concentration in the discharge gap was measured using LIF technique.

(5) The time-resolved evolution of the laser induced streamers was visualized using the ICCD camera. This resulted in the determining of the velocity of

propagation of the streamer (about 2.5×10^5 m/s) and the averaged diameters of the leader channel and leader streamers (200 μm and 100 μm, respectively).

Although this experiment clearly showed that the LIF measurement in the DC streamer corona discharge is possible, if the ICCD timing is correctly chosen, coincidence of the laser pulse and regular streamer, hindering the LIF measurement, is theoretically possible, but its probability is very low, even in the case of the high-repetition DC streamer corona discharges.

References

(*1*) Chang, J.-S. *OYO BUTURI*. **2000**, *69*, 268-277 (in Japanese).

(*2*) Ershov, A.; Borysow, J. *J. Phys. D: Appl. Phys.* **1995**, *28*, 68-74.

(*3*) Coogan, J. J.; Sappey, A. D. *IEEE Transactions on Plasma Science*. **1996**, *24*, 91-92.

(*4*) Ono, R.; Oda, T. *IEEE Trans. Ind. Applicat*. **2000**, *36*, 82-86.

(*5*) Ono, R.; Oda, T. In *Proceedings of the IEJ-ESA Joint Symp. on Electrost.*: Kyoto, 2000; pp 286-269.

(*6*) Hazama, H.; Fujiwara, M.; Sone, T.; Hashimoyo, H.; Ishida, M.; Tanimoto, M. In *Proceedings of the Asia-Pacific Workshop on Water and Air Treatment by Advanced Oxidation Technologies: Innovation and Commercial Applications*: Tsukuba, 1998; pp 70-73.

(*7*) Roth, G. J.; Gundersen, M. A. *IEEE Transactions on Plasma Science*. **1999**, *27*, 28-29.

(*8*) Tochikubo, F.; Watanabe, T. In *HAKONE VII International Symposium on High Pressure Low Temperature Plasma Chemistry*: Greifswald, Germany, 2000; pp 219-223.

(*9*) Fresnet, F.; Baravian, G. ; Pasquiers, S. ; Postel, C. ; Puech, V. ; Rousseau, A. ; Rozoy, M. *J. Phys. D: Appl. Phys*. **2000**, *33*, 1315-1322.

(*10*) Ohkubo, T.; Kanazawa, S.; Nomoto, Y.; Chang, J. S.; Adachi, T. *IEEE Trans. Ind. Applicat*. **1996**, *32*, 1058-1062.

(*11*) Sasaki, J.; Kubodera, S.; Ozaki, R.; Uchiyama, T. *J. Appl. Phys*., **1986**, *60*, 3845-3849.

(*12*) Ohkubo, T.; Kanazawa, S.; Nomoto, Y.; Mizeraczyk, J. In *Proc. The First Polish-Japanese Symposium*; Sopot, Poland , 2000; pp 85-88.

(*13*) Yang, J. B.; Izawa, Y.; Nishijima, K. In *2000 National Convention Record IEE Japan*, 2000; p 236. (in Japanese)

(*14*) Yan, K.; Yamamoto, T; Kanazawa, S.; Ohkubo, T.; Nomoto, Y.; Chang, J. S. *J. of Electrostatics*, **1999**, *46*, 207-219.

(*15*) Creyghton, Y. L. M. *Pulsed Positive Corona Discharges: Fundamental Study and Application to Flue Gas Treatment*, Ph.D. Dissertation, Technische Universiteit Eindhoven, 1994.

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