Wide-Range Two-Dimensional Imaging of NO Density Profiles by LIF Technique in a Corona Radical Shower Reactor

Seiji Kanazawa, *Member, IEEE*, Tomoyoshi Sumi, Naruaki Sato, Toshikazu Ohkubo, *Member, IEEE*, Yukiharu Nomoto, *Member, IEEE*, Marek Kocik, Jerzy Mizeraczyk, and Jen-Shih Chang, *Senior Member, IEEE*

***Abstract*—Planar laser-induced fluorescence was employed during the process of NO_x removal in a corona radical shower system. Using the wide-range imaging (image size: 240 mm in width and 160 mm in height), two-dimensional distributions of ground-state NO could be observed not only in the discharge zone but also both in 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 the downstream region of the reactor but also in the upstream region of the reactor. The effect of the gas injection through the nozzles electrode on the NO profile in the reactor was negligible. The NO removal rate was almost the same for both cases with and without the injection gas once the streamer discharge was produced. In the present reactor at low main gas flow rate, it was considered that electrohydrodynamic flow became to be dominant, and the flow toward the upstream affected the decrease of NO in the upstream region. This fact is important for optimizing the performance of the nonthermal plasma reactor.**

***Index Terms*—Corona radical shower system, electrohydrodynamic (EHD) flow, planar laser-induced fluorescence (PLIF), streamer, wide-range two-dimensional NO concentration distribution.**

I. Introduction

THE nonthermal plasma generated by electrical discharge at atmospheric pressure is widely studied for the flue gas treatment. In the case of NO_x removal, the reactor developed has been evaluated so far in terms of the NO_x removal rate and energy efficiency. However, it is noted that the removal process is complicated. In order to establish the proper model consistent with experimental results, it is necessary to understand the behavior of pollutants 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 [1]–[8]. LIF can be used for *in-situ* observation of the phenomena in plasma reactors during the NO_x removal process [6], [7].

We have investigated the NO LIF images for measuring the concentration of NO molecules in a corona radical shower reactor without injection gases [8]. As a result, the effect of the induced EHD flow on the NO removal process was considered to be significant. In addition, the gas injection through the nozzle electrode in a corona radical shower system plays the important role of stabilizing the streamer discharge and improving the NO removal efficiency [10].

In this study, the planar laser-induced fluorescence (PLIF) technique was used to monitor wide-range two-dimensional NO molecule distribution in the reactor during NO treatment. The effect of the injection gas on the NO removal and NO distribution in the reactor was also investigated.

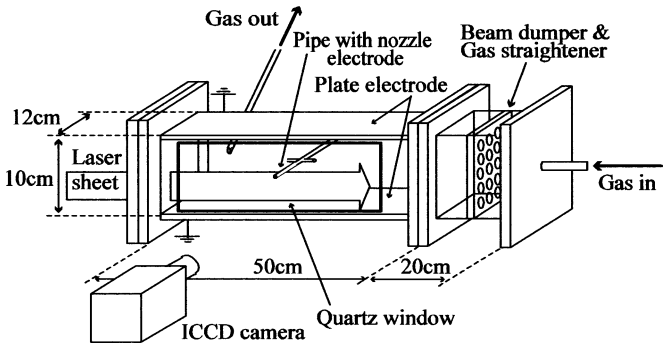

Fig. 1. Schematic diagram of the experimental apparatus.

II. Experimental Apparatus and Methods

Experiments were performed using a corona radical shower reactor. Fig. 1 shows the schematic diagram of the reactor. The apparatus and fundamentals of the measuring techniques have been described in detail elsewhere [7]–[9] and only a brief description is given in this paper. The corona radical shower reactor used in this study consists of two nozzles-to-plate elec-

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S. Kanazawa, T. Ohkubo, and Y. Nomoto are with the Department of Electrical and Electronic Engineering, Oita University, Oita 870-1192, Japan (e-mail: skana@cc.oita-u.ac.jp; tohkubo@cc.oita-u.ac.jp; nomoto@cc.oita-u.ac.jp).

T. Sumi and N. Sato were with the Department of Electrical And Electronic Engineering, Oita University, Oita 870-1192, Japan. They are now with Kyushu Electric Power Company Ltd., Fukuoka 810-8720, Japan (e-mail: Tomoyoshi_Sumi@kyuden.co.jp; Naruaki_Satou@kyuden.co.jp).

M. Kocik was with the Department of Electrical And Electronic Engineering, Oita University, Oita 870-1192, Japan. He is now with the Centre for Plasma and Laser Engineering, Institute of Fluid Flow Machinery, Polish Academy of Sciences, 80-231 Gdańsk, Poland (e-mail: kocik@imp.gda.pl).

J. Mizeraczyk is with the Centre for Plasma and Laser Engineering, Institute of Fluid Flow Machinery, Polish Academy of Sciences, 80-231 Gdańsk, Poland (e-mail: jmiz@imp.gda.pl).

J.-S. Chang is with the Department of Engineering Physics, McMaster University, Hamilton, ON L8S 4M1, Canada (e-mail: changj@mcmaster.ca).

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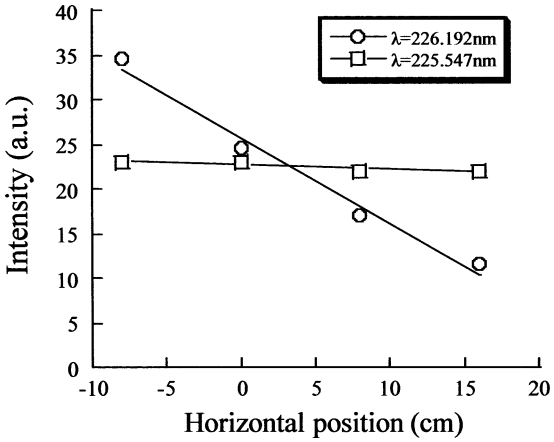

Fig. 2. Laser beam absorption characteristics for different probe laser lines.

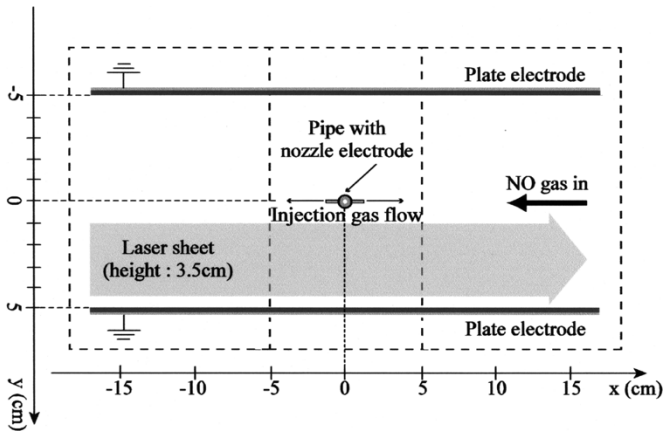

Fig. 3. Schematic of LIF observation area. The dashed rectangle indicates the image capturing area by the ICCD camera for the upstream and downstream regions of the reactor. The laser sheet is about 35 mm in height and 1 mm in width.

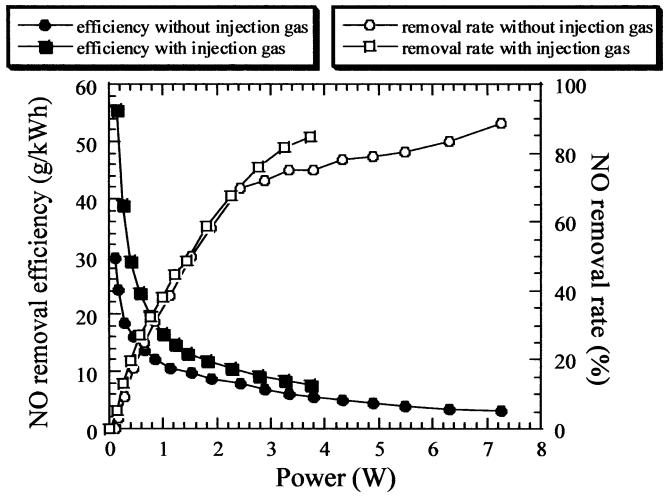

Fig. 4. NO_x removal and energy efficiency characteristics (primary gas: NO (200 ppm)/dry air, gas flow rate: 3 L/min, injection gas: $CO_2(9\%)$/dry air, gas flow rate: 0.44 L/min).

trode geometry, having an electrode gap of 50 mm. The nozzles (1.5 mm outer diameter, 1 mm inner diameter, 5 mm length) and plate (120 mm × 500 mm) electrodes were set inside the acrylic box (100 mm × 120 mm × 700 mm) in which quartz windows were mounted for the laser diagnostics. DC high voltage with positive polarity was applied through a 10-MΩ resistor to the stressed electrode and the streamer coronas were generated. NO (1000 ppm)/N_2 + dry air mixture flowed along the reactor with a flow rate of 3 L/min. As an injection gas, CO_2 + dry air mixture was injected into the reactor through the nozzles electrode. The experiment was carried out at room temperature under atmospheric pressure.

In order to observe ground-state NO molecules by LIF technique, a probe laser with a wavelength of 226 nm [$A^2\Sigma^+(v' = 0) \rightarrow X^2\Pi(v'' = 0)$] was produced by frequency doubling of the laser beam from a dye laser (Lambda Physik, SCANmate) with Coumarin 47 dye, pumped by an XeF excimer laser (Lambda Physik, COMPex 150, tuned at 351 nm). A planar laser beam (35 mm height and 1 mm width) was produced using optics, and it passed through the 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 ICCD camera (LaVision, Flame Star II) with the image size of 240 mm in width and 160 mm in height and the two-dimensional LIF images were observed by a monitor and recorded on the hard disk for detailed analysis. In order to obtain a wide-range two-dimensional NO image, the wavelength of the probe laser beam (i.e., NO excitation line) was evaluated before the measurements. Fig. 2 shows the characteristics of laser absorption on the beam path for two different probe laser wavelengths: 226.19 nm and 225.55 nm. The incident laser beam with 226.19 nm is attenuated along the laser beam path in the reactor. This laser beam is not suitable for wide-range NO observations, while the laser beam absorption for the wavelength of 225.55 nm is negligible. It was confirmed that the LIF intensity using this laser beam was proportional to the NO concentration until ca. 300 ppm. Since the laser power density within the sheet was not uniform, the image was corrected by dividing the raw image by the beam profile. Signal averaging was performed to increase the SNR of the image. Moreover, in order to obtain the two-dimensional NO density profiles in the reactor, LIF images in both the upstream and downstream regions of the reactor (Fig. 3) were connected by using an image processing software (Adobe Photoshop).

III. Experimental Results and Discussion

A. Relation Between the NO Removal and Discharge-Induced Plasma

Fig. 4 shows NO removal rate and energy efficiency as a function of discharge power with and without injection gas. NO (200 ppm)/dry air mixture was introduced to the reactor at the gas flow rate of 3 L/min. Concentrations of NO and NO_2 were measured at the outlet of the reactor using an NO_x monitor (Hodakatest, Testo 33). In this experiment, NO removal means that NO is oxidized to NO_2 by the streamer discharge-induced plasma reaction. By injecting additional gases of CO_2 (40 mL/min) and dry air (0.4 L/min) into the discharge zone through the nozzles electrode, corona discharge is stabilized and stable streamers can be produced [10]. Although CO_2 injection plays a dominant role in generating streamers, CO_2 itself does not have

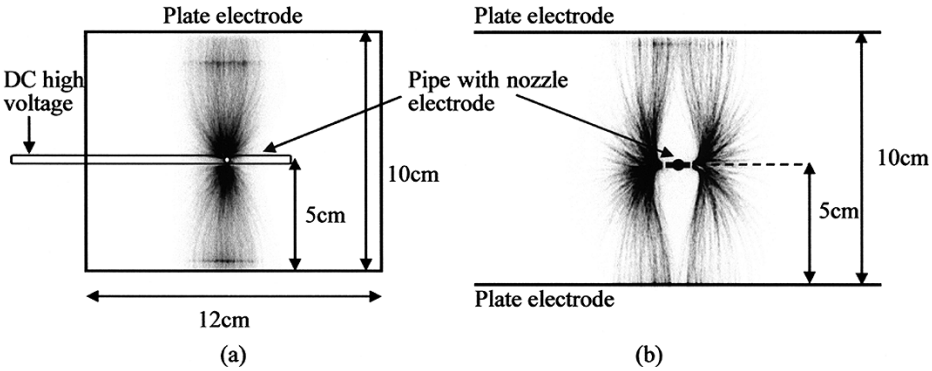

Fig. 5. Streamer corona images at 27 kV (3.3 W). (a) Front view. (b) Side view. The image is an average of 100 images observed by the ICCD camera with the gate width of 0.6 ms.

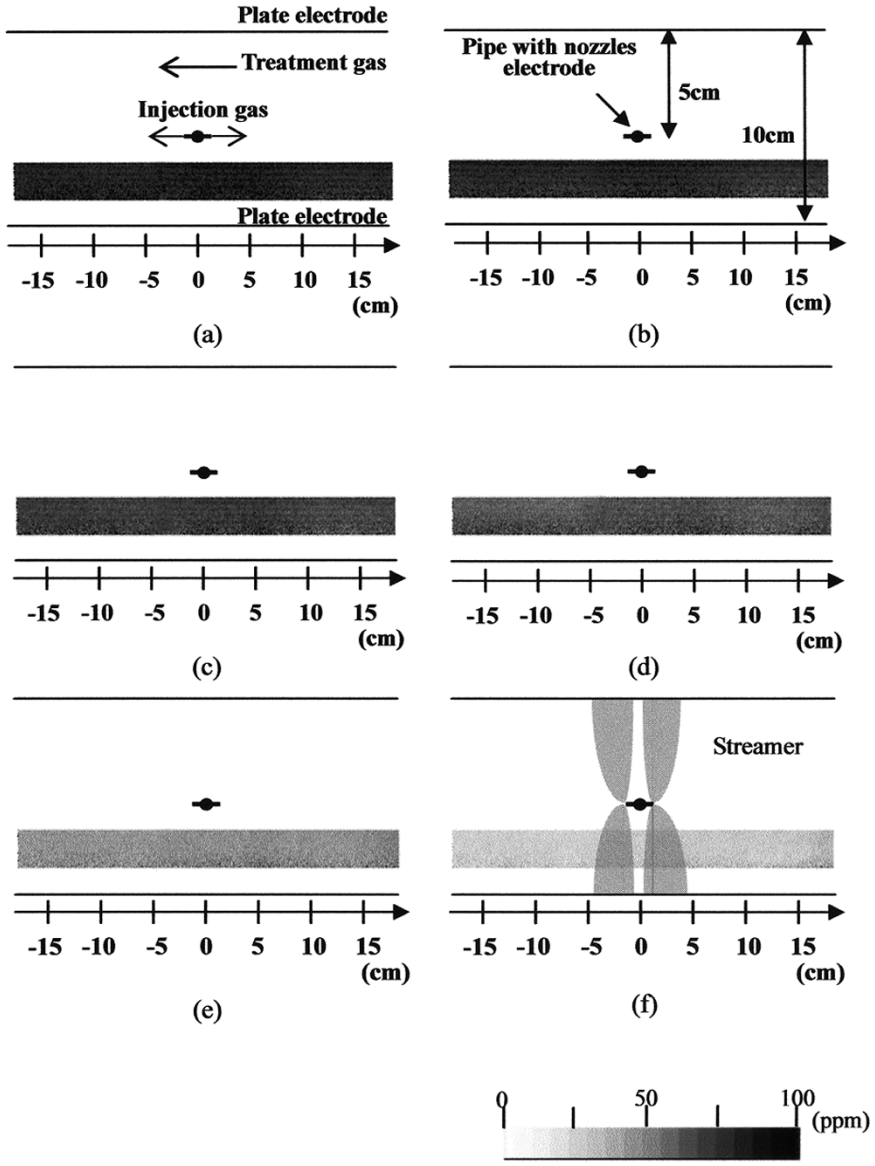

Fig. 6. Wide-range two-dimensional NO concentrations inside the reactor for various applied voltages (primary gas: NO (100 ppm)/dry air, gas flow rate: 3 L/min, injection gas: $CO_2(9\%)$/dry air, gas flow rate: 0.44 L/min). The pipe with nozzles electrode is not shown in real scale. (a) $V = 0$ kV, $(P = 0$ W). (b) $V = 16$ kV, $(P = 0.14$ W). (c) $V = 18$ kV, $(P = 0.40$ W). (d) $V = 20$ kV, $(P = 0.76$ W). (e) $V = 24$ kV, $(P = 1.80$ W). (f) $V = 28$ kV, $(P = 3.72$ W).

an effect on the NO removal. Therefore, once streamers are produced not only in the case with gas injection but also without gas injection, the NO removal rate and its energy efficiency have a similar tendency for both cases as shown in Fig. 4.

A streamer consists of a streamer head and streamer channel. The streamer head can be observed by detecting its optical emission. Fig. 5 shows the trace of propagating streamer head taken with ICCD camera (average of 100 images captured with a long

exposure time of 0.6 ms). As shown in Fig. 5, the streamer head splits into parts generating several branches during the propagation from the tip of the nozzles electrode to the plate electrode, and the plasma formed by the streamers shows a flame-like pattern. In this discharge using the pipe with two nozzles electrode, however, the volume occupied by the streamers is limited as shown in Fig. 5(a), but the NO removal rate is higher than 80% as shown in Fig. 4. This means that the influence of gas sneakage on NO treatment is negligible at lower gas flow rate. On the other hand, positive ions (space charges) flow in the discharge gap after streamer discharge and momentum transfer between positive ions and gas molecules occurs. This is the cause of ionic wind generation. The ionic wind-induced electrohydrodynamic (EHD) flow is considered to be an influence for NO removal in this experiment. To clarify the phenomenon in this regard, experimental investigations on the NO removal characteristics observed by planar LIF are presented in the next section.

B. Wide-Range Two-Dimensional NO Density Profile

Fig. 6 shows wide-range two-dimensional images of NO concentration in the reactor for several applied voltages. Initial NO concentration is about 100 ppm. The images were taken after the NO concentration in the reactor reached a steady state. Each image is an average of 50 captured images. The intensity of the images corresponds to NO spatial concentrations. As the applied voltage increases, the NO LIF signal becomes weaker compared with that of the nonapplied voltage, indicating the decrease of NO concentration. Fig. 7(a) and (b) shows the NO density profiles along the treatment gas flow with and without additional gas injection, respectively. They correspond to the NO concentration along the middle line of the laser sheet.

It is seen from the images in Fig. 6 and spatial NO distribution in Fig. 7 that NO molecule concentration decreased not only in the plasma region created by the streamers but also in the upstream region of the discharge. In this experiment, the primary gas flow velocity is 4.2 mm/s, while the injection gas velocity at the tip of the nozzles is 4.7 m/s. The gas flow in the reactor without the discharge is laminar by the estimation using the Reynolds number (R_e), which is also confirmed by the LIF gas flow visualization using air containing NO. However, the flow structure during the discharge results from the interaction between the primary flow, additional flow through the nozzles, and secondary flow due to the ionic wind. The velocity flow pattern in this reactor is more complicated than that in the wire-to-plate electrode or needle-to-plate electrode arrangements. Although the injection gas jet penetrates deep into the narrow upstream region due to a low primary gas flow velocity [11], the influence of the injection gas for spatial NO distribution is almost negligible under the operation of streamer discharge mode as shown in Fig. 7. The NO removal area spreads over an upstream region for both cases with and without injection gas. From these results, it is considered that not only energetic electron-induced plasma reaction occurs at the streamer head but also the produced active radicals play an important role for NO removal in the reactor used in this study. In particular, the EHD flow is responsible for the enhanced NO removal far in the upstream region of the reactor. The strong vortexes are formed in both the upstream and downstream regions around the nozzle electrode

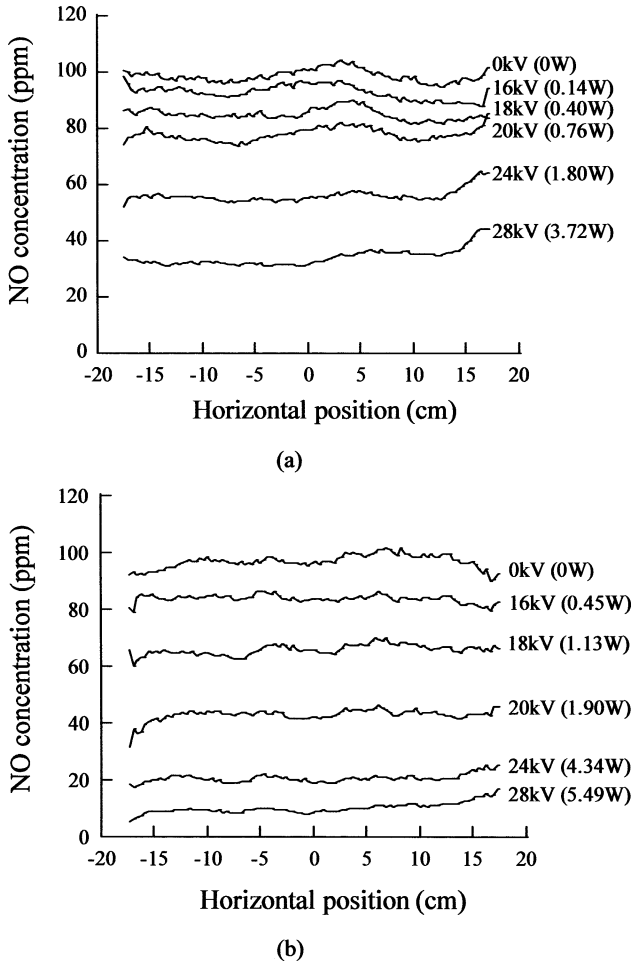

Fig. 7. Spatial NO distribution for various applied voltages. (a) With injection gas. (b) Without injection gas. (Primary gas: NO (100 ppm)/dry air, gas flow rate: 3 L/min, injection gas: $CO_2(9\%)$/dry air, gas flow rate: 0.44 L/min.)

as a result of the strong EHD secondary flow [11], [12]. The influence of EHD flow can be estimated using an electrohydrodynamic number E_{hd} [13], [14] defined by

$$E_{hd} = \frac{J_p L^3}{\rho_g \nu^2 \mu_i} \tag{1}$$

where J_p is the current density on the plate electrode, L is a characteristic length based on the reactor or stressed electrode dimension, ρ_g is the gas density, ν is the kinematic viscosity of the gas, and μ_i is the positive ion mobility. The EHD number (E_{hd}) estimated for the typical operating condition is in the range of 10^6–10^8 depending on J_p and L, while the Reynolds number based on the reactor height is 40. Since the ratio E_{hd}/R_e^2 is much higher than 1, the ionic wind (EHD-induced secondary flow) becomes dominant [13], [14]. Therefore, the EHD-induced secondary flow in the present discharge system significantly influences the flow pattern inside the reactor (i.e. , mixing of the gas occurs in the reactor) [11], [15]. In particular, ozone molecules produced by the streamer corona discharge in the corona radical shower reactor are observed not only in the downstream region but also in the upstream region of the reactor [11]. The EHD secondary flow is responsible for the ozone transport upstream of the discharge region. These facts

are the cause of the decrease of NO concentration measured in the upstream region of the reactor.

IV. Conclusion

Planar laser-induced fluorescence was used for *in-situ* observation inside the corona radical shower reactor for NO removal. The influence of the injection gas containing CO_2 for spatial NO distribution in the reactor is negligible under the operation using streamer discharge mode. NO removal due to oxidation occurs far from the discharge zone in the upstream of the reactor. In the present reactor at a low primary gas flow rate, EHD flow becomes dominant, and the flow toward the upstream affects the decrease of NO along the primary gas flow.

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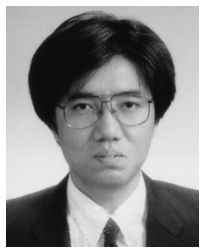

Seiji Kanazawa (M'91) was born in Oita, Japan, in 1961. He received the B.E., M. E., and Ph.D. degrees from Kumamoto University, Kumamoto, Japan, in 1985, 1987, and 1990, respectively.

Since 1990, he has been with Oita University, Oita, Japan, where he is presently an Associate Professor in the Department of Electrical and Electronic Engineering. His research interests are in the areas of applied electrostatics, air pollution control, plasma technologies, plasma diagnosis, and eco-material processing using lasers.

Dr. Kanazawa is a Member of the Institute of Electrical Engineers of Japan, Institute of Electronics, Information, and Communication Engineers of Japan, Institute of Electrostatics Japan, and Japan Society of Applied Physics.

Tomoyoshi Sumi was born in Oita, Japan, in 1978. He received the B.E. and M.E. degrees from Oita University, Oita, Japan, in 2002 and 2004, respectively.

He is currently with Kyushu Electric Power Company Ltd., Fukuoka, Japan.

Mr. Sumi is a Member of the Institute of Electrostatics Japan.

Naruaki Sato was born in Nagasaki, Japan, in 1977. He received the B.E. and M.E. degrees from Oita University, Oita, Japan, in 2001 and 2003, respectively.

He is currently with Kyushu Electric Power Company Ltd., Fukuoka, Japan.

Toshikazu Ohkubo (M'86) was born in Beppu, Japan, in 1948. He received the B.E., M.E., and D.E. degrees from Kyushu University, Fukuoka, Japan, in 1972, 1975, and 1986, respectively.

From 1975 to 1987, he was a Research Associate in the Department of Electrical Engineering, Oita University, Oita, Japan. From 1988 to 1989, he was a Visiting Research Fellow at McMaster University, Hamilton, ON, Canada. He became an Associate Professor in 1987 and a Professor in 1995 in the Department of Electrical and Electronic Engineering, Oita University. His research interests include NO_x removal by corona-discharge-induced plasmas, electrohydrodynamics, NO measurement by laser-induced fluorescence, and thin-film preparation by pulsed-laser deposition for fuel cells.

Dr. Ohkubo is a Member of the Institute of Electrical Engineers of Japan, Institute of Electrostatics Japan, and Japan Society of Applied Physics.

Yukiharu Nomoto (M'86) was born in Shinyo, China, 1942. He received the B.E., M.E., and D.E. degrees from Kyushu University, Fukuoka, Japan, in 1965, 1967, and 1982, respectively.

In 1970, he became a Research Associate at Kyushu University. He joined Oita University, Oita, Japan, as an Associate Professor in 1973 and became a Professor in 1985. His research interests include low-temperature plasmas, plasma chemistry, electrostatic applications, and ozonizers.

Dr. Nomoto is a Member of the Institute of Electrical Engineers of Japan, Institute of Electrostatics Japan, and Japan Society of Applied Physics.

Marek Kocik received the M. Sc. degree in experimental physics in the field of atomic spectroscopy from the University of Gdańsk, Gdańsk, Poland, in 1996, and the Ph.D. degree from the Institute of Fluid Flow Machinery, Polish Academy of Sciences, Gdańsk, Poland, in 2002.

From 2003 to 2004, he held a postdoctoral position with the Department of Electrical and Electronic Engineering, Oita University, Oita, Japan. He is currently an Associate Researcher at the Polish Academy of Sciences. He has authored 15 published refereed papers and presented more than 40 conference papers. His research interests include laser applications to micromachining, laser-flow diagnostics, and laser spectroscopy.

Jerzy Mizeraczyk received the M.Sc. degree from the Technical University of Gdańsk, Gdańsk, Poland, in 1966, and the Ph.D. degree from the University of Gdańsk, Gdańsk, Poland, in 1975.

He was a Fellow of the Japan Society for the Promotion of Science at Nagoya University, Nagoya, Japan, and of the A. von Humboldt Foundation and H. Hertz Foundation at the Ruhr University Bochum, Germany. He was a Visiting Senior Researcher at Chalmers University of Technology, Göteborg, Sweden, and at McMaster University, Hamilton, ON, Canada. He also was a Full Professor at Oita University, Oita, Japan. He is currently the Head of the Centre for Plasma and Laser Engineering, Institute of Fluid-Flow Machinery, Polish Academy of Sciences, Gdańsk, Poland. He was a Co-Director of two European Community Copernicus Projects and one NATO "Science for Peace Programme" Project. He has worked in the areas of plasma physics, dc, pulsed, RF, and MW discharges, gas lasers and their applications, and plasma chemistry for environmental technologies. He has authored more than 100 published refereed papers and presented more than 100 conference papers on these topics.

Jen-Shih Chang (M'90–SM'96) received the B. Eng. and M. Eng. degrees in electrical engineering from Musashi Institute of Technology, Tokyo, Japan, and the Ph.D. degree in experimental space sciences from York University, Toronto, ON, Canada.

During 1973–1974, he was a Researcher at the Centre de Recherches en Physique de l'Environment, CNRS, France. From 1975 to 1979, he was a Project Scientist/Assistant Professor with the Department of Physics and Center for Research in Experimental Space Sciences, York University. From 1979 to 1986, he was an Assistant Professor/Associate Professor with the Department of Engineering Physics, McMaster University, Hamilton, ON, Canada. From 1985 to 1996, he was a Visiting Professor at Musashi Institute of Technology, Tokyo Denki University, The University of Tokyo, University of Seville, Joseph Fourier University, University of Poitiers, Oita University, and Tokyo University of Agriculture and Technology. Since 1987, he has been a Professor at McMaster University, and is involved in research on applied electrostatics, lightning, air pollution control, and solid and liquid waste destruction plasma technologies.

Prof. Chang is currently Chair of the Electrohydrodynamics Technical Committee of the IEEE Dielectrics and Electric Insulation Society.