Effects of Fly Ash on NO_x Removal by Pulsed Streamers

Shunsuke Tsukamoto, Takao Namihira, Douyan Wang, Sunao Katsuki, Reuben Hackam, *Fellow, IEEE*, Hidenori Akiyama, *Fellow, IEEE*, Akihiro Sato, Yoshitaka Uchida, and Masami Koike

***Abstract*—NO_x removal methods using plasma chemical reactions in nonthermal plasmas have been widely studied. In this paper, the effects of the addition of fly ash on NO_x removal using short-pulsed discharge plasmas are described. Fly ash which had been collected from a coal-burning thermal electrical power plant was used. Experiments were performed using four different mixtures of gases which included NO. These were (N_2 + NO), (N_2 + NO + O_2), (N_2 + NO + H_2O), and (N_2 + NO + O_2 + H_2O). These gas mixtures were used either with or without the addition of fly ash. The initial concentration of NO was fixed at 200 ppm (NO parts per million of the gas mixture). The study of the NO_x (NO + NO_2) removal was performed with the fly ash, as it is relevant to real situations in coal power plants. The results show that the presence of fly ash decreased the NO_x removal rate slightly in the case of dry gas mixtures while it increased the NO_x removal rate substantially in the case of wet gas mixtures. These results suggest that the presence of fly ash in the flue gases, which also contain a few percentages of moisture, would be advantageous to the treatment of flue gases emitted from thermal power plants for the removal of nitrogen oxides.**

***Index Terms*—Flue gases, fly ash, NO_x removal, particle size distribution, pulsed streamers, pulsed waveforms.**

I. Introduction

NITROGEN oxides and sulfur oxides emitted from thermal power plants, factories, and automobiles cause acid rain which is detrimental to the environment. At the present time, ammonia-catalytic De-NO_x and calcium-gypsum De-SO_x methods are used for the treatment of combustion flue gases emitted from thermal power plants. Although these processes are effective and reliable, both the initial and the running costs are very high. On the other hand, it has been shown that discharge plasmas can decompose volatile organic gases, nitrogen oxides and sulfur oxides [1]–[16].

The use of short-duration high-voltage pulsed power in a coaxial cylindrical configuration at atmospheric pressure results in the formation of a nonthermal plasmas [1]–[4], [6]–[12]. Under these conditions, high-energy electrons are created while the temperatures of the ions and the neutral remain relatively unaffected [17], [18]. This reduces the energy consumption because most of the energy is used to create energetic electrons. The high-energy electrons produce chemical radicals, which decompose the pollutant molecules of nitrogen oxides [7]–[9], [19]–[21].

Clements *et al.* reported on the effect of fly ash on SO_2 removal in an experiment using high-voltage pulsed energized discharge reactor [1]. Chang *et al.* discussed the relevant corona discharge processes in an electrostatic precipitator [7]. Lowke *et al.* analyzed theoretically the removal of NO_x (NO + NO_2) and SO_2 in an electrostatic precipitator operating in a typical flue gas [14]. Recently Daito *et al.* reported on the removal of NO in a mixture of N_2, O_2, H_2O, and NO and also measured the growth of NH_4NO_3 [22]. However, it appears that, with the exception of a preliminary report by the authors [23], there have been little or no reports on the effects of the presence of the fly ash on the removal of NO_x. A combined removal of SO_2, NO_x, and fly ash particles was performed using pulse streamer and a dc bias voltage [1]. The presence of fly ash improved the removal efficiency of SO_2 but the synergetic effects of the fly ash on the removal of NO_x were not reported [1]. It was shown by Tokunaga *et al.* that the removal efficiencies of both NO_x and SO_2 were increased in the presence of powdery silica [24]. This is because the SO_2 and NO_x, which had been absorbed on the surface of the dust, reacted with water, and converted into acids after further oxidation [16]. A study on the collection of fly ash was reported using a combination of pulsed and dc bias energization but without NO_x removal [25]. The removal of NO_x and SO_2 from a thermal power plant using coal with electron beam and therefore in the presence of fly ash was reported [26]. However, the comparison on the removal of NO_x with and without fly ash was not studied.

In this work, fly ash was injected into the discharge region to study its influence on the NO_x removal. Fly ash was mixed with different mixtures of NO, nitrogen, oxygen, and with and without water vapor.

II. Experimental Setup and Procedures

A. Apparatus

Fig. 1 shows a schematic diagram of the NO_x removal system. The system comprised a pulsed power generator, gas bottles for simulating flue gases, a humidity source, a discharge reactor and a fly ash mixing arrangement, simulated gases, and measuring instruments.

A three-stage Blumlein generator was used as the pulsed power source. Each stage of the generator consisted of two

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S. Tsukamoto is with the Department of Electrical Engineering, Ariake National College of Technology, Fukoka 836-8585, Japan.

T. Namihira and S. Katsuki are with the Department of Electrical and Computer Engineering, Kumamoto University, Kumamoto 860-8555, Japan.

D. Wang is with Hitachi, Ltd., Ibaraki 317-8511, Japan.

R. Hackam is with the Department of Electrical and Computer Engineering, University of Windsor, Ontario, N9B 3P4 Canada.

H. Akiyama is with the Graduate School of Science and Technology, Kumamoto University, Kumamoto 860-8555, Japan.

A. Sato, Y. Uchida, and M. Koike are with the Research Laboratory, Kyushu Electric Power Co., Inc., Fukuoka 815-0032, Japan.

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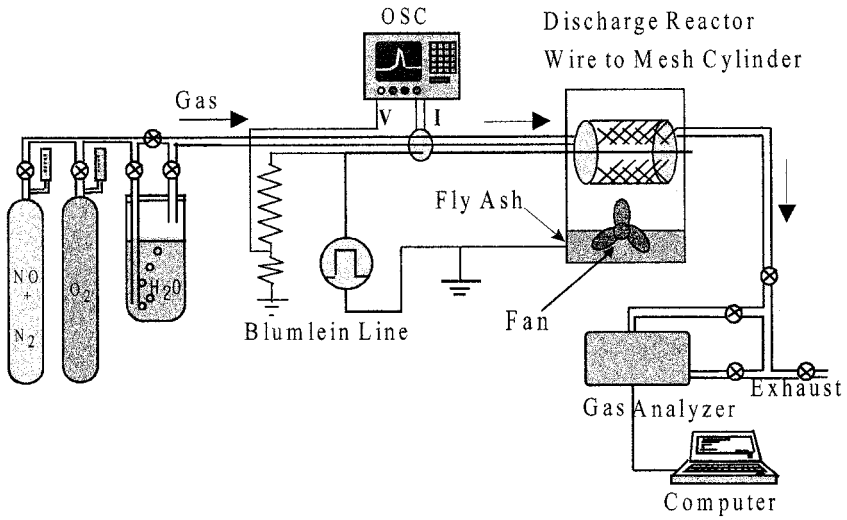

Fig. 1. Experimental setup for removal of NO in the presence of fly ash using a concentric coaxial reactor.

coaxial cables, which had a characteristic impedance of 50 Ω (RG-213/U, Mitsubishi Densen, Japan). A schematic diagram of the generator used was reported in [27] where further details may be obtained. The length of the coaxial cable defines the pulsewidth and, in the present work, a length of 10 m, which produced a pulsewidth of 100 ns was used. The width of the pulse is defined as the full width at half maximum voltage (FWHM). The applied voltage from the Blumlein generator was measured using a resistive voltage divider, which was connected between the central electrode and the ground electrode. A positive voltage pulse was employed, as it was more effective than a negative polarity in removing NO_x [8]–[10]. In the positive wire-plane geometry, there were more streamer channels compared to the negative wire and the former had more branching [27]. Furthermore, the positive streamer corona propagates into longer distances in the electrodes gap than the negative streamer [7]. The current was measured using a Rogowski coil (Pearson current monitor, model 2878, Pearson Electronics). A Hewlett-Packard digital oscilloscope (HP54542A) with a maximum bandwidth of 500 MHz and a maximum sample rate of 2 G samples was used to record the signals. The current probe was located on the high-voltage end of the reactor as shown in Fig. 1. The oscilloscope was located inside a shielded room (70 dB) to reduce the transient interference. The energy ($\int vi\,dt$) was determined from the digitized signals of the current (i), the voltage (v), and time (t), which were stored on a computer (Fig. 1).

Fig. 2 shows the discharge reactor and the fly ash mixing arrangement. Both were placed in a nylon box. The fly ash was blown in all directions within the box from the bottom by a motor fan (3 W). The outer cylindrical electrode was made of stainless steel mesh having an internal diameter of 80 mm to enable the fly ash to enter in and mix readily with the gases in the discharge region. The mesh size was 4×4 mm^2. The length of the reactor was 100 mm. The central electrode was made of copper rod 2 mm in diameter.

A source of NO gas of 1000 ppm mixed with N_2 was diluted to 200 ppm with pure N_2 to simulate the flue gases from coal thermal power plants. 5% of O_2 and 3.1% of water vapor

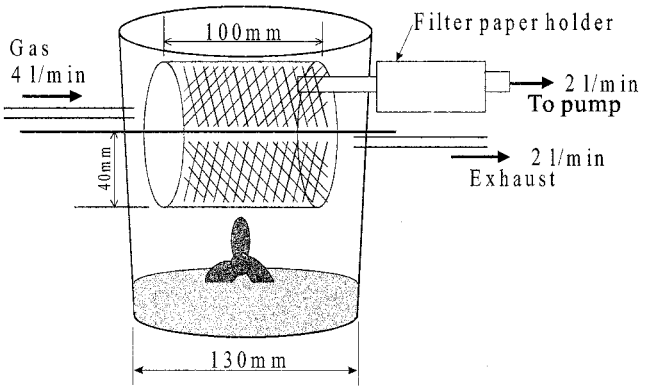

Fig. 2. Setup for measurements of the fly ash concentration.

(23.9±2.8 Torr at $25 \pm 2\,^{\circ}$C) were also added to the mixture. The total pressure of the gas mixture was 1.01×10^5 Pa and the temperature of the reactor was $25 \pm 2\,^{\circ}$C.

The NO and NO_2 concentrations were measured using a gas analyzer (Testoterm, Model Testo33, Germany). The method of measurements of nitrogen oxides was based on potentiostatic electrolysis. This method relies on the dependence of the current passing along the surface of a gold electrode immersed in sulfuric acid on the reactions of oxidation and deoxidation in the presence of various gases. The instrument was calibrated by the manufactures. The O_2 concentration was measured using the galvanic battery method (Testoterm, Model Testo33, Germany). The concentration of the fly ash in the gas mixture was determined by extracting the fly ash using an air pump (2 l/min) across a Whatman silica filter (0.3 μm) with a combination of a sensitive electric balance (10^{-3} g, type MJ-300 made by YMC Company, Japan). This procedure was repeated several times to determine the reproducibility of the measurements.

B. Measurement of Fly Ash Particle Size Distribution and Concentration

The particle size distribution of the fly ash was measured using a laser-diffraction method (Shimazu SALD-3000 S, Japan). The particle size distribution was determined from the

intensity ratio of the diffraction light to the scattering light of the incident laser beam into a suspension of fly ash and pure water. The wavelength of the laser was 690 nm and the range of the measurement of the fly ash was 0.08 to 3000 μm.

C. Measurement of Fly Ash Concentration in Simulated Gas

Fig. 2 shows the setup for the measurements of the fly ash concentration in the simulated mixture of gases. A filter was employed to collect the particles using an air pump in accordance with the Japanese Industry Standard (JIS Z 8808). The collection of the fly ash was not affected by the presence of NO_x or SO_x since the filter was made of silica. Typically, a quantity of 80 g of fly ash was placed in the discharge reactor as shown in Fig. 2. A suction filtration was used to collect the fly ash particles as shown. For the purpose of measuring the fly ash concentration, a gas flow rate of 4 L/min was used in the reactor, which was split equally into the analyzer where the fly ash was prevented from reaching it by filtering it out, and into the air suction. The fly ash was completely removed during 2 min at a flow late of 2 L/min by the suction pump. The fly ash concentration (g/Nm^3, N, normalized to 1.01×10^5 Pa and 0 °C) was calculated from the measured weight and the known volume. The measurements of the fly ash concentration were repeated three times under dry and wet conditions.

D. NO and NO_x Removal Experiment

NO removal was performed using the system shown in Fig. 1. Four gas mixtures at 1.01×10^5 Pa and 25 ± 2 °C that have different compositions were used with and without fly ash.

1) N_2 and NO (200 ppm).
2) N_2, NO (200 ppm) and O_2 (5%).
3) N_2, NO (200 ppm) and H_2O (3.1%).
4) N_2, NO (200 ppm) O_2 (5%) and H_2O (3.1%).

Initially the study of the NO and NO_x removal was done without fly ash using the four gas mixtures. The gas flow rate in the reactor was fixed at 2 L/min in the NO removal experiments. The Blumlein generator was charged at 25 kV dc and a peak pulse voltage of about 78.9 ± 1.2 kV was applied to the discharge electrodes. The pulsed voltage was impressed at 1 pulse per second (pps) until saturation in the reduced NO concentration was attained. This took typically from 3 to 4 min. After the saturation of the NO concentration, the repetition rate was increased gradually from 1 pps to 10 pps.

III. Results and Discussions

A. Fly Ash Particle Size Distribution and Concentration in Simulated Gases

The particle size was found to be distributed in the range from 0.08 to about 3000 μm. Fig. 3 shows the cumulative percentage of particles of fly ash from a coal burning power plant against their diameter size. The median size was in the range of 15 μm (Fig. 3). The fly ash was mainly consisted of 38.6% silica (SiO_2), 29.5% alumina (Al_2O_3), 7.6% calcium oxide, 5.7% Fe_2O_3, 1.9% TiO_3, 1.8% Na_2O and small quantities of other oxides.

Fig. 4 shows the distribution of the particle size of the fly ash against the diameter size. Typically the concentrations of the fly ash for the diameters 8.2, 10.2, 12.6, 15.5, 19.2, and 23.6 μm were 4.8%, 5.1%, 5.2%, 5.3%, 5.3%, and 5.1%, respectively. For 0.08 and 3000 μm, the concentrations were 0.05% and 0.06% of the total, respectively (Fig. 4). The present measurements of the particle size are consistent with the previously reported measurements of the particles formed by De-NO_x and De-SO_x process in the combustion gases in a coal thermal power plant [29]. In that work, the particle sizes were reported to vary from tenths of micrometer to about 40 μm.

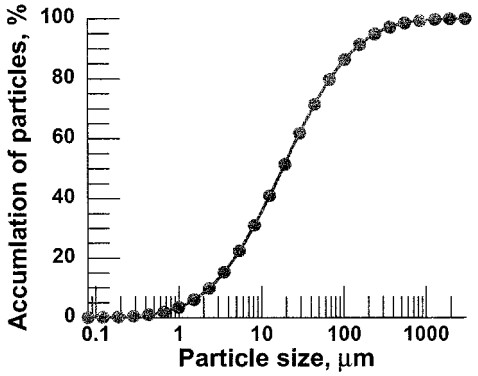

Fig. 3. Accumulation of particles of fly ash collected from a coal power plant against their size.

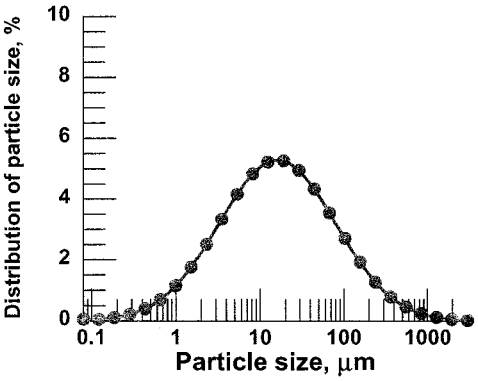

Fig. 4. Distribution of particle size of fly ash collected from a coal power plant against their size.

The concentration of the fly ash in the mixture of gases without an addition of moisture was measured and found to be 11.25 ± 0.75 g/Nm^3. These measurements were taken after about 15 min when a steady-state condition was reached. This is consistent with a reported fly ash concentration of about 20 g/Nm^3 at the air preheater of a thermal power plant [30]. The concentration of the fly ash when a moisture of 3.1% was added to the mixture of gases was found to be 0.4 ± 0.1 g/Nm^3.

B. Voltage, Current, and Energy Characteristics

Fig. 5 shows the voltage and the current waveforms for different the gas mixtures. Fig. 5(a) shows the voltage and current waveforms applied to a mixture of N_2 and 200 ppm of NO without fly ash and without water vapor. Fig. 5(b) shows the waveforms for the case of N_2 and 200 ppm of NO with the addition of fly ash of 11.25 g/Nm^3. It can be observed from Fig. 5(b) that the values of the peak currents and voltages are similar, and, therefore, the fly ash did not appear to influence the waveforms.

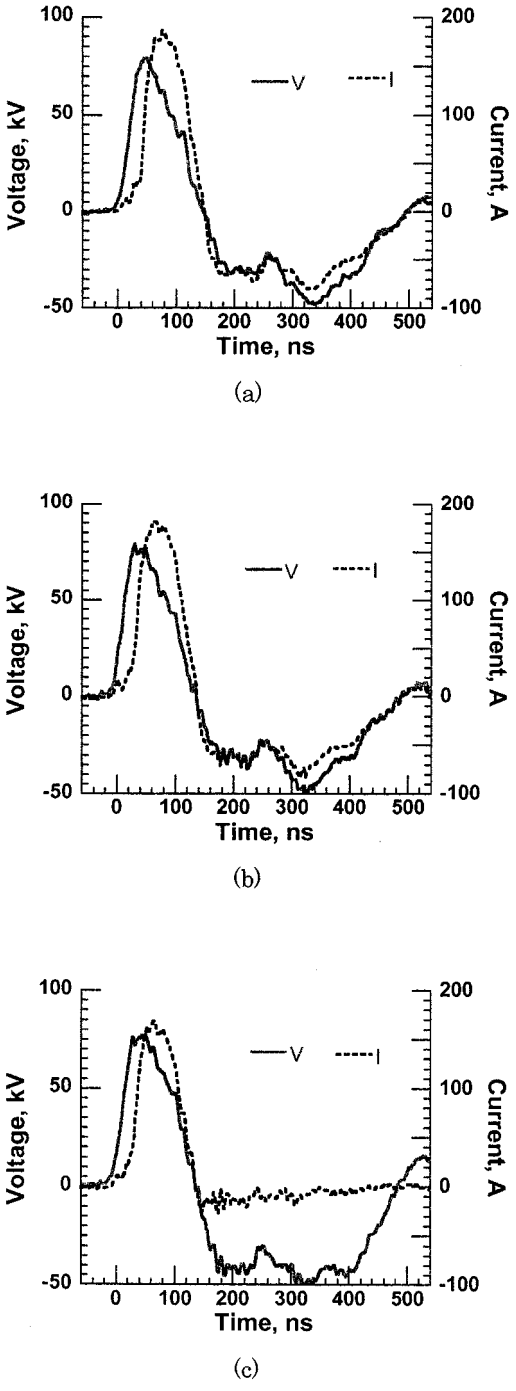

Fig. 5. Voltage and current waveforms for different gas mixtures. (a) N_2, NO (200 ppm) without fly ash, (b) N_2, NO (200 ppm) with fly ash (11.25 g/Nm^3), and (c) N_2, NO, 0_2 (5%), H_20 (3.1%) and without fly ash. Condition, 1.01×105 Pa; temperature 25 ± 2 °C; pulse rate 1 pps.

In all cases, the pressure was fixed at 1.01×10^5 Pa and the temperature at 25 ± 2 °C. Fig. 5(c) shows the waveforms for a mixture of N_2, NO (200 ppm), O_2 (5%), and H_2O (3.1%) without fly ash. Here the current peak decreased from about 181 A [Fig. 5(a) and (b)] to about 166 A [Fig. 5(c)]. This was due to the presence of both O_2 and H_2O in the mixture, which promoted electron attachments to form negative ions. Although the peaks of the pulse voltage in all three cases were the same at 78.9 ± 1.2 kV, the shapes of the pulse voltage and current changed due to the impedance mismatch between the pulsed power source and the reactor varied with the changing conductivity of the discharge.

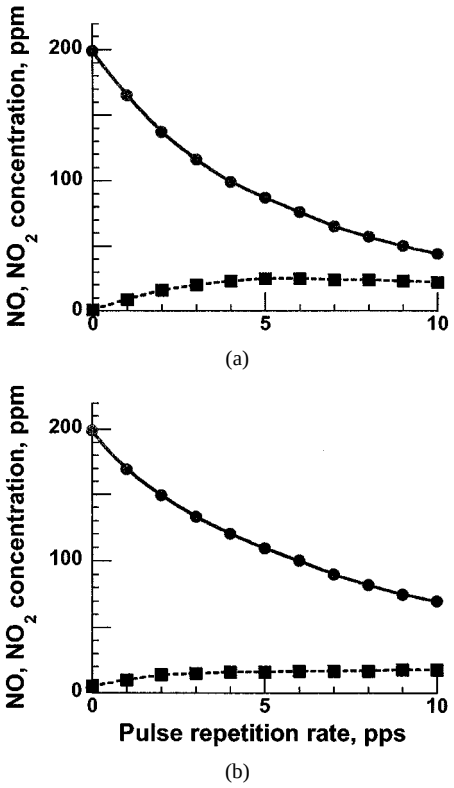

Fig. 6. Final concentration of NO and NO_2. (a) Without the addition of fly ash and (b) with fly ash as a function of pulse repetition rate. Conditions: dry mixture of N_2 with initial NO concentrations of 200 ppm; fly ash, 11.25 g/Nm^3; pressure, 1.01×105 Pa; temperature 25 ± 2 °C; pulse width, 100 ns; peak voltage, 78.9 ± 1.2 kV; peak current 173.5 ± 7.5 A, • NO, ■ NO_2.

The energies of the pulse were calculated and were found to vary from 0.84 to 1.4 J/pulse depending on the type of mixture used and whether ash was present or absent in the mixture.

C. NO_x Removal Characteristics

Fig. 6 shows the concentrations of NO and NO_2 as a function of pulse repetition rate in a dry mixture of NO having an initial concentration of 200 ppm and N_2 with and without fly ash. It can be observed that the concentration of NO steadily decreased with increasing pulse rate. For example, the NO concentration decreased from the initial value of 200 ppm to 44 ppm without the addition of fly ash [Fig. 6(a)] and to 70 ppm with fly ash [Fig. 6(b)]. Coincidentally, the concentration of NO_2 increased from 0 to 22 ppm [Fig. 6(a)] and from 0 to 18 ppm [Fig. 6(b)], without and with fly ash, respectively. The reduction in the removal of NO in the presence of the fly ash [Fig. 6(b)] might be caused by the increase of electrons, which are caught by the fly ash. A loss of electrons by the fly ash decreases the atomic nitrogen dissociated from the nitrogen molecules. Therefore, it is suggested that in the presence of fly ash less radicals N are produced compared to the case without fly ash and consequently less NO is removed (reaction 1) as observed in Fig. 6. Possible reactions for the decomposition of NO have been suggested [5], [9]:

$$\mathrm{NO} + \mathrm{N} \rightarrow \mathrm{N_2} + \mathrm{O} \qquad k_1 = 5.9 \times 10^{-11}\ \mathrm{cm^3/s}\ [5]. \quad (1)$$

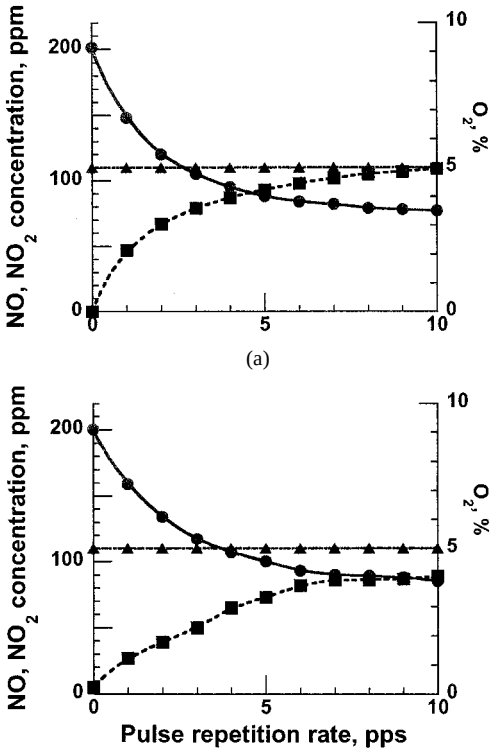

Fig. 7. As for Fig. 6 except the gas mixture was N_2 (95%), NO (200 ppm) and 0_2 (5%): (a) without fly ash and (b) with fly ash (11.25 g/Nm^3). Other conditions of pressure and temperature were as in Fig. 6, • NO, ■ NO_2, ▲ O_2.

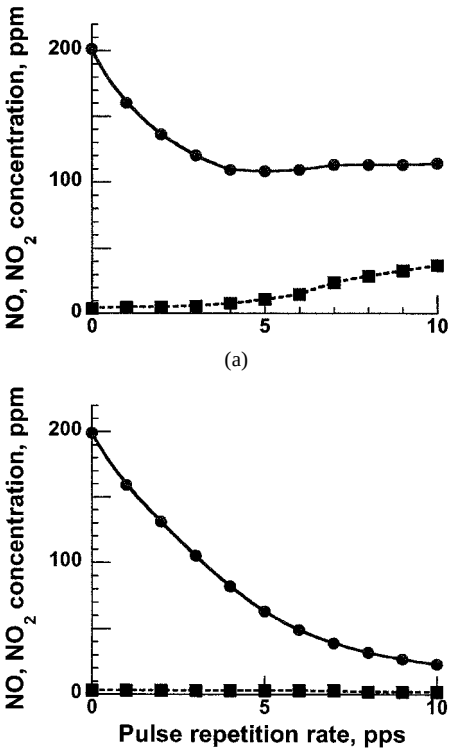

Fig. 8. As for Fig. 6 but with a mixture of N_2 (96.9%), NO (200 ppm) and H_2O (3.1%): (a) without fly ash and (b) with fly ash (0.4 g/Nm^3). Other conditions of pressure and temperature were as in Fig. 6, • NO, ■ NO_2.

where N is a radical. The constant k_1 shows the reaction rate for (1) and [19]

$$\mathrm{NO + O + M \rightarrow NO_2 + M} \qquad k_2 = 6.9 \times 10^{-32}\ \mathrm{cm^6/s}\ [31] \tag{2}$$

where M is a third body such as N_2.

Fig. 7 shows the concentrations of NO, NO_2, and O_2 as a function of the pulse rate in a dry mixture of NO (initial concentration 200 ppm), N_2 and 5% of O_2 at 1.01×10^5 Pa. It can be observed that the concentration of NO decreased from 200 to 70 ppm and the NO_2 increased from 0 to 110 ppm [Fig. 7(a)] at 10 pps without fly ash. When the fly ash was added, the corresponding concentrations of NO and NO_2 at 10 pps were 85 ppm and 89 ppm [Fig. 7(b)], respectively.

With fly ash the concentration of O_2 remained at about 5% [Fig. 7(b)]. Additional reactions for this mixture [32], [33] are

$$\mathrm{N + O_2 \rightarrow NO + O} \qquad k_3 = 1.0 \times 10^{-16}\ \mathrm{cm^3/s}\ [14] \tag{3}$$

and [7]

$$\mathrm{NO + O_3 \rightarrow NO_2 + O_2} \qquad k_4 = 1.8 \times 10^{-14}\ \mathrm{cm^3/s}\ [5]. \tag{4}$$

Fig. 8 shows the concentrations of NO and NO_2 as a function of the pulse repetition rate in a mixture of N_2, NO, and H_2O. The concentration of NO decreased substantially in the presence of fly ash and water vapor, from 200 to 21 ppm while the concentration of NO_2 for 10 pps remained negligibly small [Fig. 8(b)]. Without fly ash, NO decreased from 200 to 122 ppm while NO_2 increased from 0 to 36 ppm [Fig. 8(a)]. A comparison between Figs. 6(a) and 8(a) shows that when O_2 was not present in the mixture the removal of NO decreased with the addition of water vapor while it increased with addition of fly ash under the same conditions [Figs. 6(b) and 8(b)]. The presence of H_2O generates the OH radicals by dissociation. Possible reactions for the removal of NO and NO_2 have been suggested [21]:

$$\mathrm{NO + OH + N_2 \rightarrow HNO_2 + N_2} \tag{5}$$

$$\mathrm{NO_2 + OH + N_2 \rightarrow HNO_3 + N_2} \tag{6}$$

and for generating NO and NO_2 [19]

$$\mathrm{HO_2 + NO \rightarrow NO_2 + OH} \tag{7}$$

and [14]

$$\mathrm{HNO_2 + OH \rightarrow NO_2 + H_2O} \tag{8}$$

$$\mathrm{2HNO_2 \rightarrow NO + NO_2 + H_2O.} \tag{9}$$

The presence of water could result in the production of NO (Figs. 8 and 9) according to reactions (11) and (12) [19]

$$\mathrm{H + OH \rightarrow NO + H.} \tag{10}$$

The radical H reacts with O_2 (Figs. 8 and 9) to form water vapor and by collisions with NO_2 [21]

$$\mathrm{NO_2 + H \rightarrow NO + OH.} \tag{11}$$

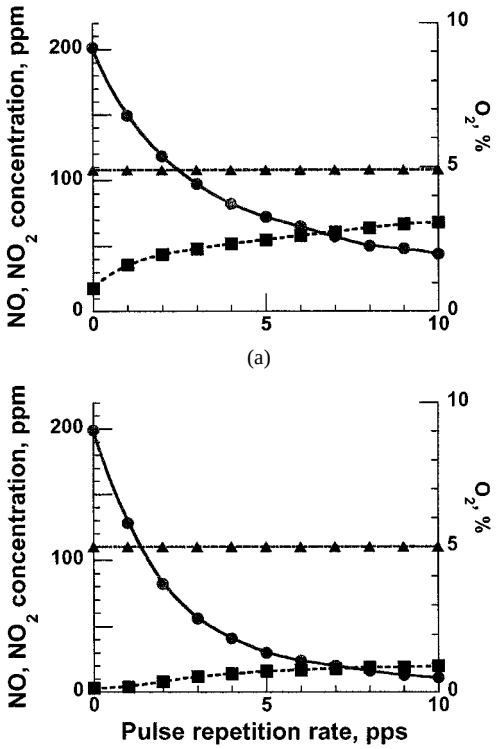

Fig. 9. As for Fig. 6 except the gas mixture was N_2 (91.9%) NO (200 ppm), O_2 (5%), and H_2O (3.1%): (a) without fly ash and (b) with fly ash (0.4 g/NM^3). Other conditions of pressure and temperature were as in Fig. 6, • NO, ■ NO_2, ▲ O_2.

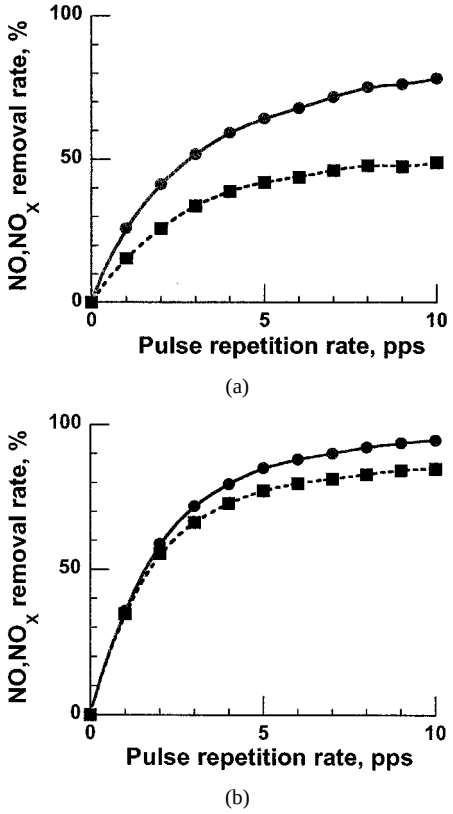

Fig. 10. Removal rates of NO and NO_x for a mixture of N_2 (91.9%), NO (200 ppm), O_2 (5%) and H_2O (3.1%): (a) without fly ash and (b) with fly ash (0.4 g/NM^3). Other conditions of pressure and temperature were as in Fig. 6, • NO, ■ NO_x.

NO_2 may also be removed by

$$NO_2 + N \rightarrow N_2O + O \qquad k_{12} = 3.0\times10^{-12}\ \text{cm}^3/\text{s}\ [14]. \quad (12)$$

When solid particles are present in the mixture it is thought that the H_2O, which may be attached on the surface of the fly ash, absorbed the NO_2 molecules [Fig. 8(b)]. The presence of fly ash when water vapor was present enhanced the removal of NO_2 [Fig. 8(b)] because the absorbed NO_2 on the surface of the fly ash reacted with water and was converted into acid [25], [16].

Fig. 9 shows the concentrations of NO, NO_2 and O_2 as a function of the pulse repetition rate for a mixture of N_2, NO (initial 200 ppm), O_2 (5%), and H_2O (3.1%). Fig. 9(a) shows that before energizing the reactor (pps = 0) the concentration of NO_2 was 18 ppm. This was due to the presence of O_2 (5%) and NO (200 ppm) in the initial mixture, which produced NO_2. It can be observed that at 10 pps, the concentration of NO decreased from 200 to about 10 ppm with the addition of fly ash [Fig. 9(b)] and to 44 ppm without fly ash [Fig. 9(a)]. This shows that the presence of fly ash promotes the removal of NO in general agreement with a previous study when silica powder was added to the gas mixture [24]. Fig. 9 shows that the concentration of O_2 remained unchanged at about 5% with and without the addition of fly ash.

A comparison between Figs. 7 and 9 shows that the removal of NO was enhanced in the presence of O_2 when H_2O was added to the mixture for both with and without fly ash. Our measurements without fly ash are consistent with a previous study,

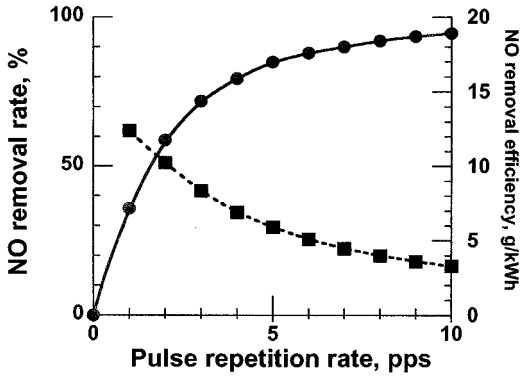

Fig. 11. NO removal rate (•) and NO removal energy efficiency (■) against pulse repetition rate in the presence of fly ash. Conditions are as in Fig. 10.

which showed that humid air was more effective than dry air in removing NO [8].

Fig. 10 shows the removal rates of NO and NO_x for a mixture of N_2, NO (200 ppm), O_2 (5%), H_2O (3.1%) with (0.43 g/Nm^3) and without the addition of fly ash. The removal rate of NO is defined as the rate of the measured NO to the initial concentration of NO (200 ppm). It will be observed that when fly ash was added to the mixture a removal rate of 94.4 and 84.5% could be achieved for NO and NO_x, respectively [Fig. 10(b)]. The corresponding rates without fly ash were 77.5 and 48% for NO and NO_x, respectively [Fig. 10(a)]. The improvements in

the removal rate of NO and NO_x with the addition of fly ash were thus 21.8% (from 77.5 to 94.4%) and 76% (from 48 to 84.5%), respectively. These are significant improvements to the removal of the pollutant gases.

D. NO Removal Energy Efficiency

Fig. 11 shows the NO removal energy efficiency against pulse repetition frequency. The pulse energy of 0.92 J/pulse was used to calculate the energy efficiency. It will be observed that the energy efficiency decreases from 12.8 g/kWh at 1 pps to 3.2 g/kWh at 10 pps. It is rather difficult to make meaningful comparison with other work because the removal energy efficiency strongly depends on the additives to the simulated flue gas mixture, the initial concentration of NO, the flow rate, the geometry of the reactor, the applied pulsed voltage, and pulse length. Dinelli *et al.* [2] reported a yield of 25 g (NO)/kWh at 50% NO_x removal efficiency in a mixture of flue gases from a coal burning plant. The gas mixture contained 500–550 ppm of NO_x, 350–400 ppm of SO_2, 80–120 mg/Nm^3 solid particles and 0.8 of (NH_3/(NO_x+SO_2). The present work shows that at 50% removal rate of 11 g (NO)/kWh (Fig. 11). In a mixture of 91% N_2, 5% O_2, 4% H_2O, and an initial concentration of 200 ppm NO, the energy yield varied from 45 g/kWh to 15 g/kWh at 50% removal efficiency of NO when the length of the applied pulse voltage varied, respectively, from 40 ns to 120 ns [27].

IV. Conclusion

The influence of the fly ash on the NO and NO_x removals using pulsed streamer discharges were studied.

1) Particle sizes of the fly ash in a thermal coal electrical power plant ranged from 0.08 to 3000 μm. The median size was 15 μm.
2) The waveforms of the voltage and current remained unchanged by the addition of fly ash to the gas mixtures containing, N_2, NO, O_2, and H_2O. The injected energy of the pulse also did not change with the addition of the fly ash.
3) In the presence of moisture, the fly ash increased significantly the removal rates of NO and NO_x.
4) In dry mixture of gases, the presence of fly ash decreased slightly the removal rates of NO and NO_x.

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Reuben Hackam (M'76–SM'76–F'88) received the B.S. degree from the Technion, Israel Institute of Technology, Israel, in 1960 and the Ph.D. and D.Eng. degrees from the University of Liverpool, Liverpool, U.K., in 1964 and 1988, respectively.

From 1964 to 1968, he was with General Electric-English Electric Company, Stafford, U.K., from 1969 to 1978, he was with the University of Sheffield, Sheffield, U.K., and since 1979, he has been a Professor of Electrical Engineering at the University of Windsor, Windsor, Ontario, Canada, where he holds the position of University Distinguished Professor. He was President of IEEE Dielectrics and Electrical Insulation Society in 1997–98 and was on sabbatical leave at Kumamoto University, Kumamoto, Japan in 1998/99.

Shunsuke Tsukamoto was born in Kumamoto, Japan, in 1954. He graduated from the Department of Electrical Engineering of the Ariake National College of Technology, Fukuoka, Japan, in 1975, and received the M.S. degree from Kumamoto University, Kumamoto, Japan in 1998, where he is currently pursuing the Ph.D. degree. He is also currently an Assistant Professor at Ariake National College of Technology.

Hidenori Akiyama (M'87–SM'99–F'99) was born in Ehime, Japan, in 1951. He received the B.S. degree in electrical engineering from the Kyushu Institute of Technology, Fukuoka, Japan, in 1974, and the M.S. and Ph.D. degrees from Nagoya University, Nagoya, Japan, in 1976 and 1979, respectively.

From 1979 to 1985, he was a Research Associate at Nagoya University he joined the faculty at Kumamoto University in 1985, Kumamoto, Japan, where he is currently a Professor.

Takao Namihira was born in Shizuoka, Japan, in 1975. He received the B.S. and M.S. degrees from Kumamoto University, Kumamoto, Japan, in 1997 and 1999, respectively.

Since 1999, he has been a Research Associate at Kumamoto University.

Akihiro Sato was born in Oita in 1960. He graduated from Department of Industry Chemistry of the Oita Technical High School. In the same year, he joined Kyushu Electric Co., Inc., Fukuoka Japan. From 1995 he has been researched as a Research Engineer of Power Plant Engineering Group in Research Laboratory.

Douyan Wang was born in Beijing, China, in 1975. She received the B.S. and M.S. degrees from Kumamoto University, Kumamoto, Japan, in 1998 and 2000, respectively.

In 2000, she joined Hitachi, Ltd. 3-1-1 Saiwai, Hitachi, Ibaraki 317-8511, Japan.

Yoshitaka Uchida was born in Fukuoka, Japan, in 1960. He received the M.E. degree in material science from Kyushu University, Fukuoka, Japan, in 1985. In the same year, he joined Kyushu Electric Co., Inc., Fukuoka Japan, and is now a Senior Research Engineer of Power Plant Engineering Group in the research laboratory.

Sunao Katsuki was born in Kumamoto, Japan in 1966. He received the B.S., M.S., and Ph.D. degrees from Kumamoto University, Kumamoto, Japan, in 1989, 1991, and 1998, respectively.

From 1991 to 1998, he was a Research Associate at Kumamoto University. Since 1998, he has been an Associate Professor at Kumamoto University.

Masami Koike was born in Fukuoka, Japan, in 1952. He received the M.E. degree in applied chemistry from Kyushu University, Fukuoka, Japan, in, 1976. In the same year, he joined Kyushu Electric Power Co., Inc., Fukuoka, Japan, and is now a Principal Research Engineer of Power Plant Engineering Group in the Research Laboratory.