Influence of NO Concentration on Removal Efficiency in Multipoint-to-Plane Electrode Dielectric Barrier Discharge Reactor

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An influence of initial nitrogen monoxide (NO) concentration on removal efficiency with a multipoint-to-plane electrode dielectric barrier discharge (DBD) reactor was investigated experimentally. Several hundreds ppm NO was diluted with nitrogen and oxygen gas mixture (N_2:O_2=9:1) and was fed to the DBD reactor as a diesel exhaust simulated gas. The experimental results were evaluated using a simple model, which relates the change of NO concentration with input energy density (IED: J/L). About 50% of NO was removed with energy efficiency of 11 g/kWh at 200 ppm initial NO concentration. The energy efficiency at 100 ppm NO removal decreased from 11 to 6 g/kWh with increasing initial NO concentration from 200 to 500 ppm at sinusoidal applied voltage. The energy consumed in the reactor during one cycle of the applied voltage decreased from 8.5 to 2 mJ with increasing NO initial concentration from 50 to 300 ppm at 6.5 kV_{pp} sinusoidal applied voltage. This energy decrease depended on the applied voltage waveform shape. The decrease of the consumed energy changed from 6.5 to 2 mJ by changing the applied voltage waveform shape from the sinusoidal to the pulse. The experimental results were also evaluated for the system performance of the reactor using the energy constant k_E (L/J). The energy constant decreased exponentially with increasing of NO initial concentration.

Keywords: Dielectric barrier discharge, NOx removal, Multipoint electrode, Pulse voltage, Non-thermal plasma, Gas treatment, Energy constant.

1. Introduction

Air pollutants like nitrogen oxides (NO_x, NO plus NO_2) by various sources have been recognized as a serious environmental problem. NOx is the main cause of acid rain, urban smog, etc [1]. The exhausts of gasoline engines are cleaned very efficiently with the three-way-catalyst. For diesel engines, the three-way-catalyst does not operate because the high oxygen content in the exhaust gasses prevents the reduction of NO [2].

Nonthermal plasma is one of the purifying technologies for NOx containing exhaust gas. A dielectric barrier discharge (DBD) can produce non-thermal plasmas at atmospheric pressure [3]. For most exhaust gasses, NO is the main component of NO_x. NO_2 is easily absorbed into water and/or an alkali solution. Therefore, the DBD performance of NOx removal depends on NO removal [4].

NO removal with high-energy efficiency in wide range of initial NO concentration is important issue for industrial applications. However, NO removal efficiency changes by initial NO concentration [5]. This paper describes the influence of initial NO concentration and applied voltage waveform on NO removal efficiency investigated experimentally using a multipoint-to-plane geometry DBD reactor.

2. Experimental Procedure

Figure 1 shows a schematic diagram of experimental apparatus consisting of the DBD reactor. The high voltage and the ground electrode were set to upper and lower sides, respectively, in the rectangular prism DBD reactor with capacity of 100 × 260 × 50 mm (width × length × height). The multipoint electrode was used as the ground electrode. The employment of the multipoint electrode which produces strongly distorted high electric field around the tip of the projections has advantages such as low-voltage operation, high energy yield compared with a plane electrode, etc [6]. The multipoint electrodes have pyramids of 528 points with tip angle of 45° in an area of 132 cm^2 (6 × 22 cm). The height of the pyramid is 5 mm. The plane

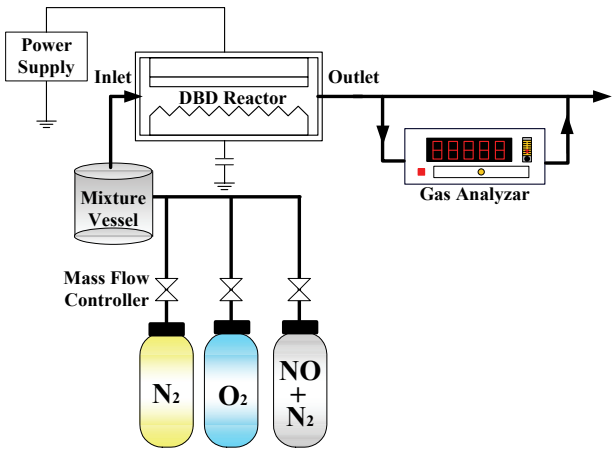

Fig.1 Schematic diagram of experimental apparatus.

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electrode having 6 × 22 cm dimension was also used as the high voltage electrode, and was set with the gap length of 1 mm. The plane electrode was coated with the alumina (Al_2O_3) dielectric barrier of 0.5-mm thickness via a powder flame spray. The gap length was defined as the distance from the projection point to the dielectric barrier.

Sinusoidal voltage of 10 kHz or pulse voltage of 1 kHz was generated by a power supply and was applied to the high voltage electrode. The applied voltages were measured using a Tektronix P6015 probe, and were recorded with a digital storage oscilloscope (Tektronix TDS 3024B). The consumed energy was obtained with a V-Q Lissajous figure. Total charge moved by the discharges was measured using a 0.05-μF capacitor inserted between the DBD reactor and the ground.

Several hundreds ppm NO was diluted with N_2 and O_2 (N_2:O_2=9:1) gas mixture and was fed to the DBD reactor as a diesel exhaust simulated gas with the gas flow rate of 5.0 L/min. The residence time in the reactor was calculated to be 773 msec. In this system, initial NO concentration was defined as the concentration when the diesel exhaust simulated gas was not fed to the DBD reactor. The gas flow was controlled by mass flow controllers (NO: CMQ9500, N_2 and O_2: CMQ0005), and was measured with a NOx analyzer (BEST Sokki BCL-511) at the outlet of the DBD reactor.

3. DBD Driven by Sinusoidal Voltage

3.1 NO Removal Efficiency

Figure 2 shows NO removal as a function of input energy density for various initial NO concentrations. The sinusoidal voltage of 10 kHz was applied to the high voltage electrode using a high-voltage AC power supply [6]. NO removal decreases with the increasing initial NO concentration, and NO was not completely removed when initial NO concentration exceeded 200 ppm. Under this condition, NO removal was approximately up to 200 ppm.

Figure 3 shows energy yield as a function of NO

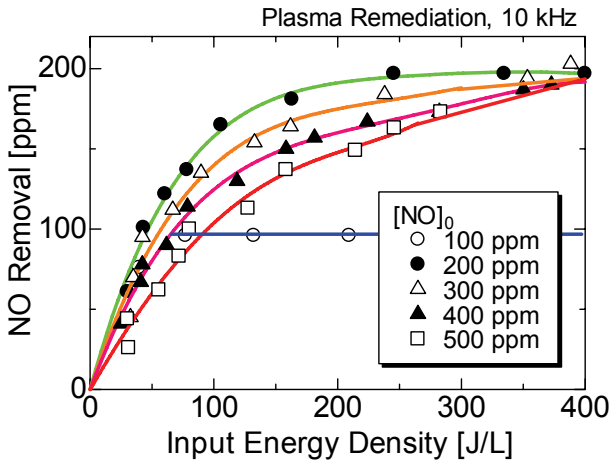

Fig.2 NO removal as a function of input energy density for various initial NO concentrations.

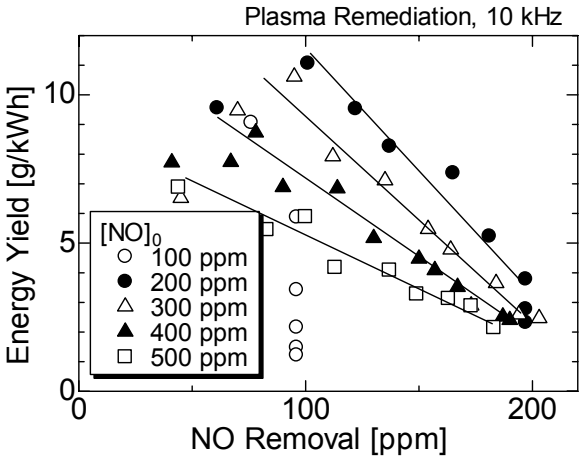

Fig.3 Energy yield as a function of NO Removal for various initial NO concentrations.

removal for various initial NO concentrations. Energy yield for NO removal is determined by the following equation:

$$\eta = \frac{60 \cdot q \cdot ([NO]_0 - [NO]) \cdot 10^{-6} \cdot M}{N \cdot U} \times 10^3 \quad \text{[g/kWh]} \quad (1)$$

where q[L/min] means flow rate; $[NO]_0$[ppm] initial NO concentration; [NO][ppm] NO concentration after DBD plasma treatment; M[g/mol] molecular weight; N[L/mol] volume of gasses per mol; U[W] consumed power. Energy yield decreases with increasing initial NO concentration and NO recombination was maybe activated because the amount of NO_2 increased more than that of NO [4]. These results suggest that initial NO concentration affects to NO removal in case of employing a sinusoidal voltage as DBD drive power source.

3.2 Energy Constant for Sinusoidal Voltage

In nonthermal plasma treatment of gasses, energy yield depends on initial gas concentration, gas flow rate, gas components, etc. In this section, energy constant is employed to evaluate performance of the DBD reactor [5]. As shown the previous works, NO removal can be expressed by a function of input energy density as following equation [5]:

$$\frac{d[NO]}{dIED} = -k_E \cdot [NO] \quad \text{.....(2)}$$

where IED[J/L] and k_E [L/J] mean input energy density and energy constant, respectively. The integration of Eq.(2) is expressed by the following equation:

$$-\ln \frac{[NO]}{[NO]_0} = k_E \cdot IED \quad \text{.....(3)}$$

Therefore, the energy constant k_E represents the magnitude of NO removal. The large number of k_E means the NO can be removed with high-energy efficiency.

Figure 4 shows logarithm NO concentration as a function of input energy density for various initial NO concentrations. In the semi-log plot of Eq. (3), the slope of

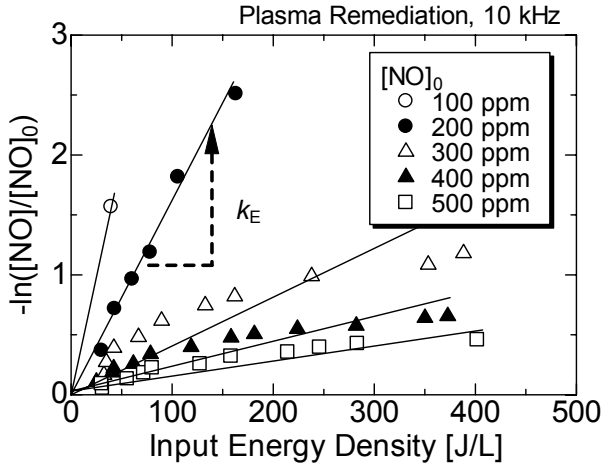

Fig.4 Logarithm NO concentration as a function of input energy density for various initial NO concentrations.

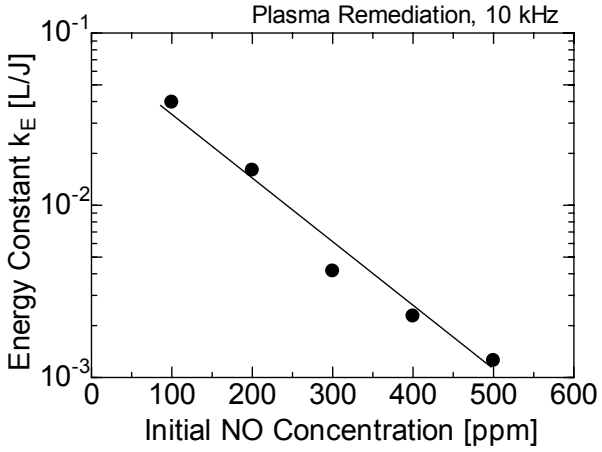

Fig.5 Energy constant for NO removal as a function of initial NO concentration.

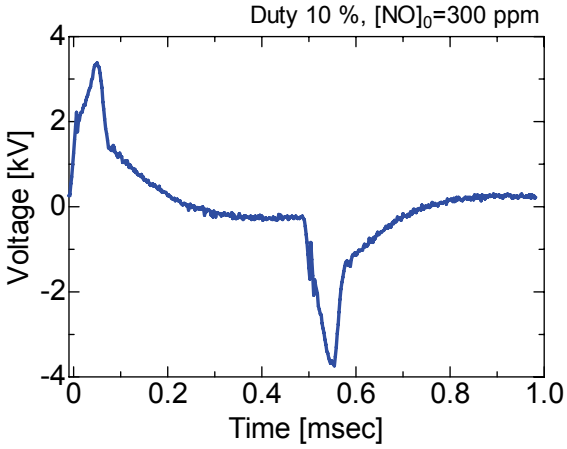

Fig.6 Typical waveform of applied voltage (initial NO concentration = 300 ppm, duty cycle = 10 %).

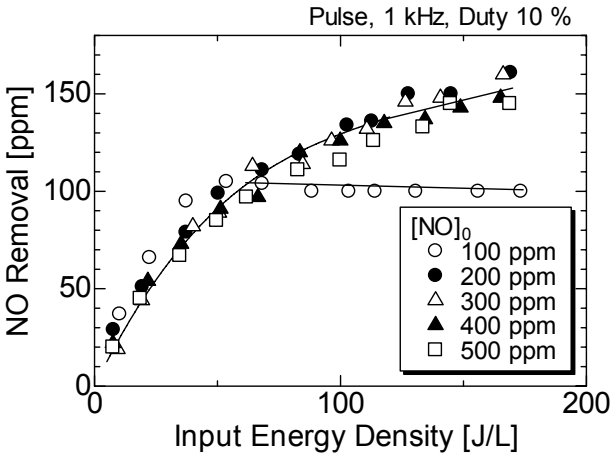

Fig.7 NO removal as a function of input energy density for various initial NO concentrations at pulse applied voltage.

the linear line is equal to the energy constant k_E. Therefore, the removal efficiency of NO is then given as

$$\frac{[NO]_0 - [NO]}{[NO]_0} = 1 - \exp(-k_E \cdot IED) \quad \text{.....(4)}$$

Eq.(4) indicates that performance of the DBD reactor can be expressed with the energy constant k_E.

Figure 5 shows energy constant for NO removal as a function of initial NO concentration. Energy constant decreases exponentially with increasing initial NO concentration. This result suggests that the more input energy density is required to remove the higher initial NO concentration (the lower energy constant k_E).

4. DBD Driven by Pulse Voltage

4.1 NO Removal Efficiency

The pulse voltage of 1 kHz was applied to the high voltage electrode using a pulse modulator to compare with the NO removal at sinusoidal applied voltage. The pulse modulator was driven by MOS-FET switches and was set the 10 % duty cycle, which is defined by a ratio of a voltage-on time to an inverse number of the applied voltage frequency. The experimental conditions were the same as chapter 3 except the applied voltage waveform. Figure 6 shows the typical pulse applied voltage waveform.

Figure 7 shows NO removal as a function of input energy density for various initial NO concentrations at pulse applied voltage. NO removal was almost independent of initial NO concentration. This tendency is quite different from that at sinusoidal applied voltage.

Figure 8 shows energy yield as a function of NO removal for various initial NO concentrations at pulse applied voltage. Although the values of the energy yield slightly decreases with increasing NO initial concentration, the influence of the NO initial concentration on the energy yield for NO removal is much smaller than that at sinusoidal applied voltage shown in Fig. 3.

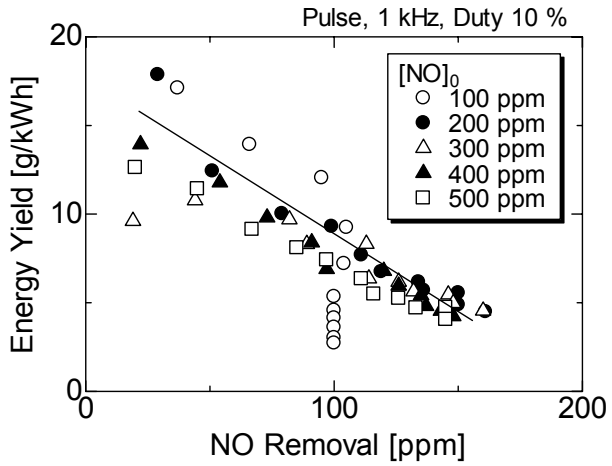

Fig.8 Energy yield as a function of NO removal for various initial NO concentrations at pulse applied voltage.

4.2 Influence of applied voltage waveform

Figure 9 shows NO removal as a function of initial NO concentration for two different applied voltage waveforms. The consumed power was fixed to 7.0 and 13.3 W, in the case of applying pulse voltage and sinusoidal voltage, respectively. For applying pulse voltage, NO removal has almost constant value of 110 ppm at larger of initial NO concentration than 110 ppm. On the other hand, for applying sinusoidal voltage, NO removal decreases gradually with increasing initial NO concentration at larger of initial NO concentration than 200 ppm. These results indicate that the initial NO concentration slightly affects NO removal at pulse applied voltage. This NO removal independency of initial concentration was reported in a pulse corona reactor by Jani [7]. The transfer efficiency from NO to NO_2 was changed by changing the applied voltage waveform from sinusoidal to pulse [7]. This result means that the composition of radical spices such as N, O(^{1}D), O(^{3}P) changes with the applied voltage waveform.

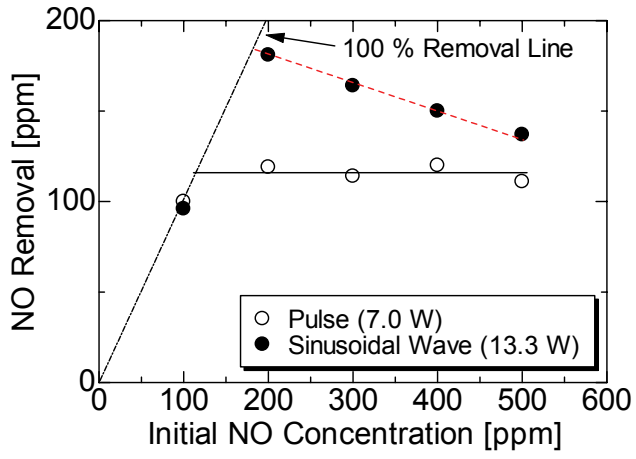

Fig.9 NO removal as a function of initial NO concentrations for two different applied voltage waveforms.

Figure 10 shows energy constant for NO removal as a function of initial NO concentration for two different applied voltage waveforms. The energy constants at pulse applied voltage show higher values than those at sinusoidal applied voltage for various initial NO concentrations. This result shows that the performance of the DBD reactor can be changed with the applied voltage waveform.

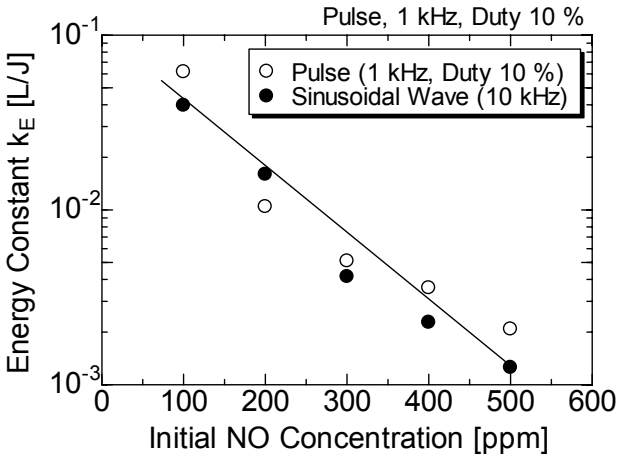

Fig.10 Energy constant for NO removal as a function of initial NO concentration for two different applied voltage waveforms.

5. Conclusion

The influence of initial NO concentration on NO removal was investigated experimentally using two different applied voltage waveforms. NO removal depends on initial NO concentration at sinusoidal voltage, whereas NO removal shows the same value for various initial NO concentrations at pulse voltage. The energy constants at pulse applied voltage show higher values than those at sinusoidal applied voltage for various initial NO concentrations.

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