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Effect of Gap Length of DBD for NO Reduction Using Ammonia Radical Injection Method

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ABSTARCT

The plasma-induced radicals efficiently convert NOx into harmless gases using ammonia radicals. The system consists of two chambers: one used as a radical injector for producing ammonia radicals, and the other as a reaction chamber for decomposing NO gas by mixing it with the ammonia radicals generated in the radical injector. The radicals were produced in a dielectric barrier discharge (DBD) using one-cycle sinusoidal power source. A pair of electrodes for generating DBD is coaxial in configuration with quartz glass tubes as dielectric materials. In this study, the effect of the gap length of DBD on NO reduction was discussed. there is an optimal gap length to bring higher energy efficiency, because the deposited power is appropriately suppressed by prolonging the gap length. The excess energy input causes the decrease of the DeNOx rate probably due to the decomposition of NH_2 *radicals and NO reproduction. For each gap length, the optimal condition of the energy efficiency for NO reduction exists at an applied voltage of 4 kV, which is higher than the initiation voltage.*

KEYWORDS: NOx, Dielectric barrier discharge, radical, Lissajous figure

1. INTRODUCTION

We have developed a radical injection DeNOx system utilizing radical chain reaction for the purpose of efficient NOx removal. Ammonia radicals to be effective for decomposing NO are generated in non-thermal plasma (NTP) in ammonia and argon circumstance. It has been reported that NOx removal is possible to be generated ammonia radicals using NTP in a separate chamber and injected them into the mixing zone of the reaction chamber [1]. DBDs are durable for years so that the apparatus are prolonged, because the dielectric materials, being inert to NOx and the reaction byproducts, can be used. Gap length of DBD plasma is thought to be an influencing factor to DeNOx performance, because the plasma characteristics are changed due to a change of the plasma initiation voltage. In this article, the effect of gap length of coaxial type radical injector for generating NTP was discussed in order to find possibility to improve the energy efficiency of the facility.

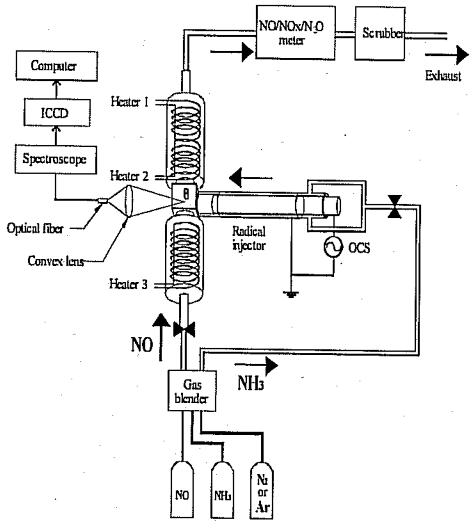


Figure 1 Experimental arrangement.

2. EXPERIMENTAL

Figure 1 shows a schematic diagram of the NOx reduction system using DBD plasma by intermittent dielectric barrier

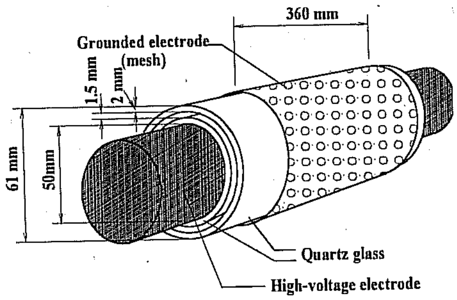


Figure 2 Radical injector.

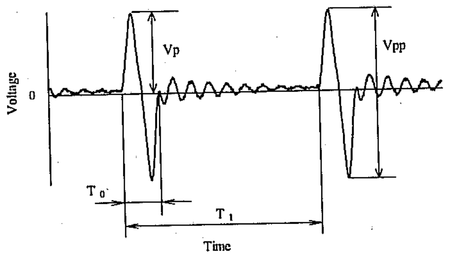


Figure 3 Waveform of one cycle sinusoidal power source.

discharge, by which the ammonia radicals are produced. NO was diluted with nitrogen gas, and ammonia (NH_3) was diluted with argon (Ar) gas. NO flowed in the process chamber, which was externally heated by two electric heaters. The concentrations and flow rates of NO and NH_3 were adjusted 908 ppm, 1.58 l/min and 1000 ppm, 1.23 l/min in the gas blender by mixing with nitrogen and argon, respectively. The adjusted NO gas was fed into the reaction chamber and the adjusted NH_3 gas was fed into the radical injector. The reaction temperature was 600 C. It was measured at the mixing zone in the reaction chamber by a thermocouple.

A schematic diagram of the radical injector (Gap length 1.5 mm) is shown in **Figure 2**. The electrodes are coaxial in configuration with quartz glass tubes as dielectric materials. The outer glass tube is 60 mm in diameter and 2 mm thick, while the inner glass tube is 53 mm in diameter and 2 mm thick. The gap length between the outer and inner glass tube is 1.5 mm, which was used for the previous experiment [1]. Glass tubes with gap lengths of 3, 5 and 7 mm was also used for DeNOx. The outer is 60 mm in diameter and 2 mm thick, while the inner diameter of the tube is varied to set designated gap lengths.

The dielectric barrier discharge was produced at the gap. The grounded outer electrode is made of a mesh steel sheet, and the inner electrode made of stainless steel. **Figure 3** schematically shows a waveform of the applied one-cycle sinusoidal (OCS) voltage. The repetition rate R_R is defined as the reciprocal of the repetition time T_1 of the discharge. T_0 was approximately 10 ìs, and R_R was 10 kHz. Thus, the OCS voltage with a pulse width of 10 ìs was intermittently applied to the gap at a repetition rate of 10 kHz.

The output peak-to-peak voltage of the power supply was 1 to 15 kV. The voltage was stepped up by a pulse transformer (winding ratio of 1:15). The time evolutions of source voltage, current and accumulated charge were simultaneously monitored with an oscilloscope.

The energy input during one cycle of the dielectric barrier discharge was estimated from the accumulated charge and voltage across the output windings of the transformer using the *V-Q* curve of the Lissajous figure. In general, the plasma discharge power *P(t)* is obtained from the product of the instantaneous current *I(t)* and the applied voltage $V_g(t)$ applied to the gap. Thus,

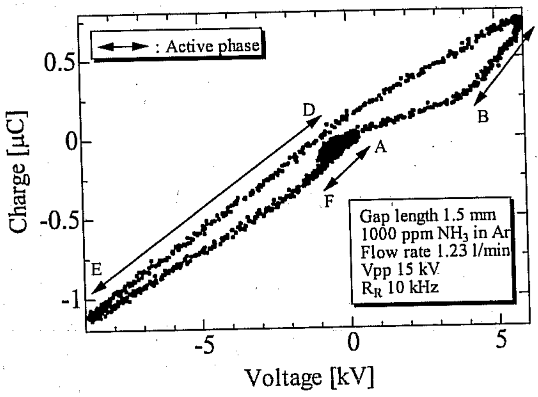


Figure 4 Lissajous figure for an applied voltage of 15 kV.

the energy *E* consumed at the gap from *t*=0 to *t*=T is expressed in Eq.(1),

$$E=\int_{t=0}^{t=T} V_g(t)\times I(t)dt \qquad (1),$$

where T is periodic time in one cycle for arc voltage application. Since *I(t)dt* corresponds to the electric charge *dq(t)* flowed between *t* and *t+dt*. Thus, Eq.(1) is changed as Eq.(2).

$$E=\int_{t=0}^{t=T} V_g(t)\times dq(t) \qquad (2)$$

q(t) can be shown using voltage $V_q(t)$ across the capacitor C_q.

$$q(t)=C_q\times V_q(t) \qquad (3)$$

Therefore,

$$E=C_q\int_{t=0}^{t=T} V_g(t)\times d(V_q(t)) \qquad (4)$$

Thus, *E* can be experimentally estimated by measuring both voltages *Vg(t)* and *Vq(t)*. Concretely, a Lissajous figure is used for obtaining the energy consumed at the gap. When one cycle sinusoidal power source is used for DBD generation, the discharge power can be obtained by *E* multiplied by the repetition rate R_R. The energy efficiency ç [g/kWh] is defined as the removal amount of NO per unit power of 1 kWh and is calculated by Eq.(5).

$$\eta=L\times\frac{N_D}{10^6}\times\frac{\Delta}{100}\times\frac{30}{22.4}\times\frac{1}{P} \qquad (5)$$

where *L* [l/min] is NO flow rate, N_D [ppm] NO concentration in ppm unit. Molecular weight of **NO is 30 g per 22.4 litters for 1 mol (OK??)and** Δ [%] is NO removal rate, *P* [kWh] is the power consumed by discharge.

3. Results

3.1 Electrical characteristics

Figure 4 shows a V-Q curve of the Lissajous figure at a repetition rate of 10 kHz for DRD at an applied voltage of 10 kV and at an ammonia concentration of 1000 ppm diluted with argon. During the cycle starting from A, the DBD plasma occurs only during B-C, D-E, F-A in one cycle. The slope of the curve represents the composite capacitance of both the dielectric material and the gap. Thus, the capacitance becomes large when the discharge occurs, which shows one of the series capacitor at the gap is shortened by the DBD discharge. The figure is not like a parallelogram, which is a typical form of DBD. This is because the voltage waveform is not a stationary ac but intermittent one cycle sinusoid with a period of 10 ìs. The distorted shape of the Lissajous figure was common for gap lengths of 1.5 to 7 mm. **Figure 5** shows a DBD initiation voltage as a function of the gap

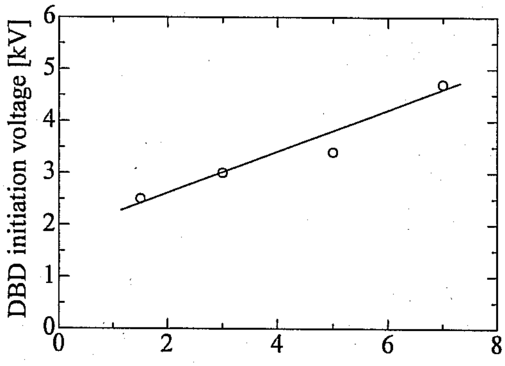


Figure 5 DBD initiation voltage versus gap length of the radical injector.

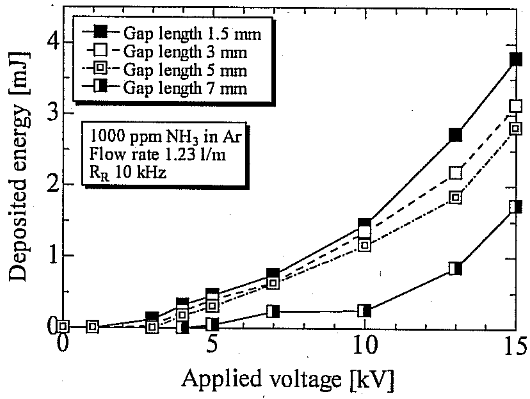


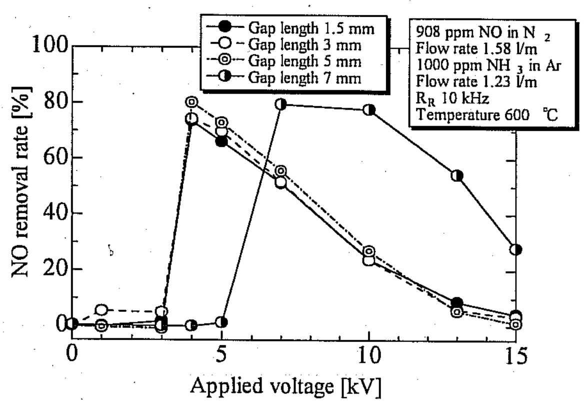
Figure 6 Deposited energies as a function of the applied volatge for gap lengths of 1.5, 3, 5 and 7 mm.

length under the experimental conditions as same as those in **Figure 4**. It can be seen that the initiation voltage linearly increases with the gap length. **Figure 6** shows deposited energies in DBD in one cycle as a function of the applied voltage, *Vpp* at gap lengths of 1.5, 3, 5, and 7 mm. The deposited energy linearly increases with the applied voltage lower than around 10 kV. However, for the applied voltage higher than 10 kV, the energy markedly increases due to transition of the arc-like filament discharges accompanying grow-like DBD discharge. The filament discharges may spend much energy compared with the glow-like discharge. Concerning the effect of gap length, it is seen that shorter the gap length, larger the deposited energy is.

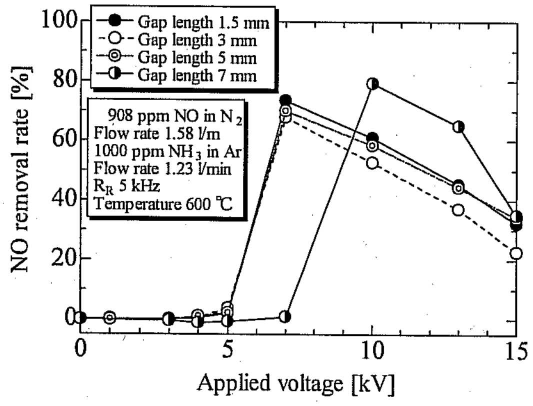
3.2 Applied voltage dependence of DeNOx rate

Figure 7(a) and 7(b) shows DeNOx rates as a function of the applied voltage for gap lengths at repetition rates of 10 kHz and 5 kHz, respectively. The ammonia concentration remains constant at 1000 ppm. DeNOx rates rapidly increase at a threshold of the applied voltage just after which the DeNOx rate becomes maximum. Maximum rates are approximately 80% at an applied voltage of 4 kV for gap lengths of 1.5, 3 and 5 mm at 7 kV for 7 mm. For all of the gap lengths, DeNOx rates become maximum at voltages slightly higher **than that the threshold of the voltage, which corresponds to DBD initiation voltage (check).**

At an applied voltage of 4 kV at a gap length of 7 mm, the energy deposited in DBD plasma is not enough high to make sufficient mount of ammonia radicals, which does not cause NO decomposition. The DeNOx rate gradually decreases at the applied voltages higher than that for showing the maximum DeNOx rate.



(a) R_R=10 kHz



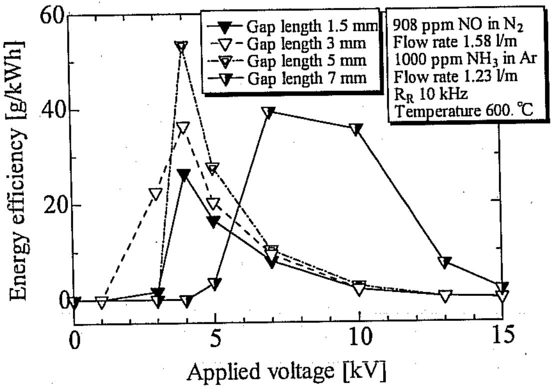
(b) R_R=5 kHz

Figure 7 DeNOx rates as a function of the applied voltage for gap lengths of 1.5, 3, 5 and 7 mm.

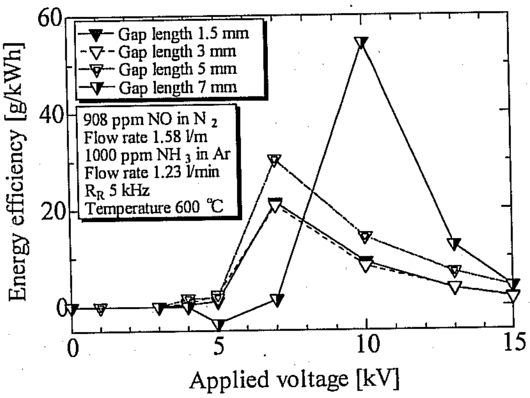
The decrease of DeNOx rate at higher voltage is probably due to production of NH and N radicals from NH_2 radicals by the excess energy input in DBD. NO reproduction may be an additional reason. At the case of a repetition rate of 5 kHz, the applied voltage presenting a maximum DeNOx rate is shifted to a higher voltage compared with the case of 10 kHz. This means that the decrease of power deposited in the DBD is compensated by raising the applied voltage to input more energy in DBD. For gap lengths of 1.5, 3 and 5 mm, the maximum DeNOx rate commonly appears at 7 kV, while 10 kV for a gap length of 7 mm. Both DeNOx results by the difference of the repetition rate show that the influencing factor to DeNOx rate is a power density, but not an energy density deposited in the DBD. An appropriated power density can make an appropriate amount of ammonia radicals so as to removal of NO gas. Thus, the energy efficiency differs by the repetition rate of the power source. If the deposited energy in one cycle is a primary factor to show the DeNOx rate and energy efficiency, repetition dependence to these parameters will not appear. From this viewpoint, deposited power density is thought to influence these parameters due to bringing the change of radical production and NO reproduction. To more clarify this discussion, we must collect more DeNOx data for a wide range of the deposited power.

3.3 Applied voltage dependence of energy efficiency for NO removal

Figure 8(a) and **8(b)** shows energy efficiencies of DeNOx as a function of the applied voltage at repetition rates of 10 kHz and 5 kHz, respectively. It can be seen from **Figure 8(a)** that the

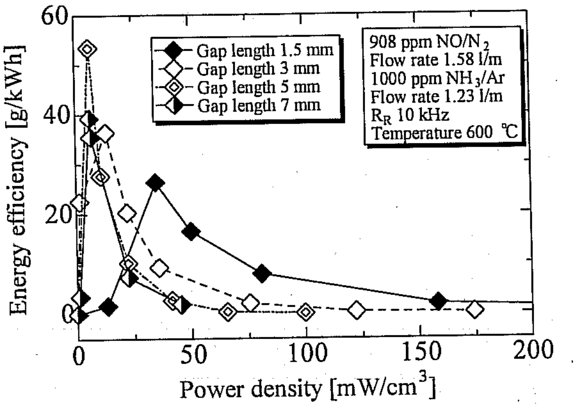


(a) R_R=10 kHz

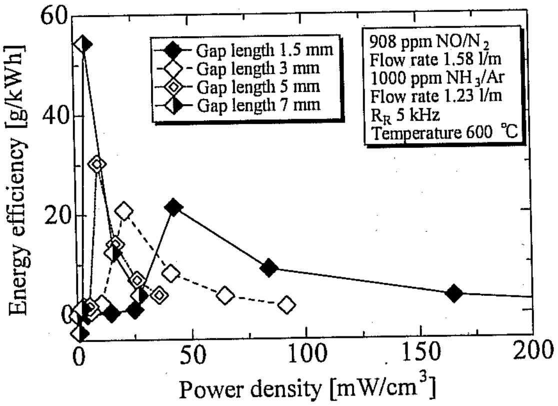


(b) RR=5 kHz

Figure 8 Energy efficiencies as a function of the applied voltage for gap lemgths of 1.5, 3, 5 and 7 mm.



(a) R_R=10 kHz



(b) RR=5 kHz

Figure 9 Energy efficiencies as a function of the power density for gap lemgths of 1.5, 3, 5 and 7 mm.

maximum energy efficiency for varying gap lengths is 58 g/kWh at a gap length of 5 mm. Thus, it is seen that there is an optimum gap length to bring higher energy efficiency, because the deposited power is appropriately suppressed by prolonging the gap length. In previous research for the case of absence of oxygen in the exhaust gas, maximum energy efficiency of 140 g/kWh was obtained at a gap length of 1.5 mm at a repetition rate of 5 kHz, where an ammonia concentration and an applied voltage are 1200 ppm and 3 kV, respectively. Thus, a slight change of the ammonia concentration, for example, brings a change of the optimum condition to find maximum energy efficiency. It is seen in **Figure 8(b)** that the optimum energy efficiency for DeNOx is shifted towards higher voltage region, where the energy efficiency of around 55 g/kWh is obtained at a gap length of 7 mm. At last, to obtain a high energy efficiency of DeNOx, energy deposited in the DBD plasma should be low as possible, of which energy is used only for accelerating electrons in DBD plasma, but is not be used for the heating of the ions. From a view point that the slight change of the experimental parameters brings higher energy efficiency, there is more possibility to obtain higher energy efficiency for DeNOx under the similar experimental conditions.

3.4 Relationship between energy efficiency and deposited energy density

Figure 9(a) and **9(b)** shows a relationship between the energy efficiency and deposited power density for repetition rates of 10 kHz and 5 kHz, respectively. As we can see, if the electrical power deposited in the plasma is enough supplied by increasing the repetition rate of the power source, the energy efficiency may be correlated to the power density except the case for a gap length of 1.5 mm as shown in **Figure 9(a)**. However, for a low power deposited in the plasma at a repetition rate of 5 kHz, the characteristics are not correlated for the investigated entire gap lengths as shown in **Figure 9(b)**. This should be a problem to be solved in a near future. But we can conclude that there is an optimum gap length to obtain a high energy efficiency. In this case, high energy efficiency is brought by the least energy deposition enough to make ammonia radicals in DBD plasma.

4. Summary

The effect of gap length of coaxial type radical injector for generating NTP was discussed on DeNOx rate and its energy efficiency in order to find possibility to improve the energy efficiency of the facility and scale up the facility, as well. The gap length is varied from 1.5 to 7 mm, which causes the change of the NTP initiation voltage. The deposited energy linearly increases with the applied voltage in glow-like NTP. For varying the applied voltage, DeNOx rate rapidly increases at a threshold of the applied voltage and becomes high at a voltage just higher the threshold, which is accompanied by the high energy efficiency of DeNOx. However, both DeNOx rate and energy efficiency show the dependence of the gap length. That is, there is an optimal gap length to bring higher energy efficiency, because the deposited power is appropriately suppressed by prolonging the gap length. The excess energy input causes the decrease of the DeNOx rate probably due to the decomposition of NH_2 radicals and NO reproduction.

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