NOx Reduction in Diesel Engine Emission Using Adsorption Followed by Nonthermal Plasma Process
(Performances of Three Types of Plasma Reactors)

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ABSTRACT

The NOx reduction performance of three types of nonthermal plasma (NTP) reactors is compared. An experiment using simulated gas is first conducted. Subsequently, those reactors are applied to a NOx aftertreatment system for a stationary diesel engine. Twelve surface discharge elements are placed in a different arrangement in each of those reactors. The results show a possibility that NOx reduction performance depends on the gas flow surrounding the elements. The reactor with the best arrangement shows a NOx reduction energy efficiency of 19.4 g (NO_2)/kWh. The application of one of those reactors to the NOx aftertreatment system indicates that NOx reduction performance is not deteriorated by CO_2. The aftertreatment system shows more than 50% NOx removal efficiency for at least 8 h with a system energy efficiency of 15.4 g (NO_2)/kWh.

Keywords: NOx, Reduction, Nonthermal Plasma, Surface discharge, Desorption, Adsorption

1. INTRODUCTION

Diesel engines are widely used in automobiles, construction machines, electric generators, ships, etc., because these engines are characterized by reduced CO_2 emissions and high thermal efficiencies. However, nitrogen oxides (NOx), mainly nitrogen monoxides (NO) present in exhaust gases, are one of the most critical environmental pollutants. Currently, SCR (selective catalytic reduction) [1-15] and LNT (lean NOx trap) [16,17] are some of the representative and most effective NOx aftertreatment technologies. SCR is the technique in which NOx is reduced by a catalyst with the help of reducing additives such as ammonia [1-6] or hydrocarbons [7-15]. However, the technique that uses ammonia has problems regarding the necessity of infrastructure that supplies the urea solution, ammonia slip, formation of ammonium particles, heavy metal spills, and so on. In the case of the technique that uses hydrocarbons, there are problems such as deactivation of the catalyst by the condensation of fuel on the catalyst surface [15] because fuel is injected as a reducing agent in this type of system. On the other hand, LNT [16,17], in which NOx is stored during the lean condition and reduced during the rich-burn condition, has a problem of sulfur oxides which strongly adhere to the catalyst and deactivate it. Therefore, those technologies are not completely satisfying at this stage, and further study is required to achieve the ideal aftertreatment.

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In our previous studies [18-20], a novel NOx aftertreatment system using adsorption and reduction by nitrogen nonthermal plasma (NTP) desorption was proposed, and the feasibility of the system was observed in laboratory experiments. This system does not use catalysts or special reducing agents but uses an oxygen-deficient gas, which can be produced by pressure swing adsorption or a selective O_2 permeation membrane from the atmosphere or by controlling the engine combustion condition. Another advantage is that NTP is not affected by sulfur in exhaust gas.

For application to a real engine, it is effective to employ thermal desorption by engine waste heat because it can save energy consumption of the NOx aftertreatment system. Figure 1 shows the concept of the system. Hot exhaust gas from the engine first heats adsorption chamber 1, in which the adsorbent is regenerated by thermal desorption, and the gas subsequently enters adsorption chamber 2, where NOx is removed by adsorption. Nitrogen gas flows through adsorption chamber 1 and elutes the desorbed NOx from the chamber. This mixture of the desorbed NOx and N_2 is subsequently reduced with a nonthermal plasma (NTP) reactor. The adsorption and desorption processes are interchanged between the two chambers.

The authors applied this NOx aftertreatment system to a stationary diesel engine generator and demonstrated the system's validity [21]. In a previous report [21], the effect of the configurations of adsorbents (NO and water vapor adsorbent) packed into an adsorption chamber was mainly investigated. However, the performance of this system also strongly depends on the performance of an NTP reactor. The surface discharge elements, which are integrated in the NTP reactor, are ceramic tubes on which surface NTP is generated. There is a possibility that NOx reduction performance is affected by the arrangement of the elements and the cooling method for the element. Therefore, three types of arrangements of the elements are compared with air and water-cooling in this study.
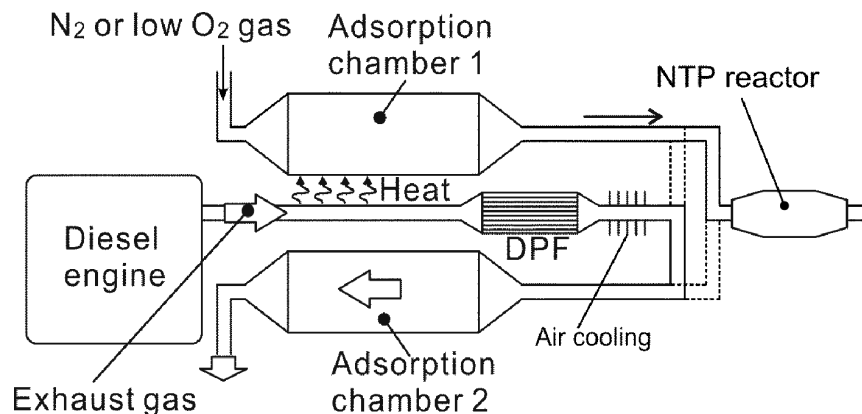

Figure 1. Concept of the NOx aftertreatment system (The adsorption and desorption processes are interchanged between the two chambers 1 and 2)
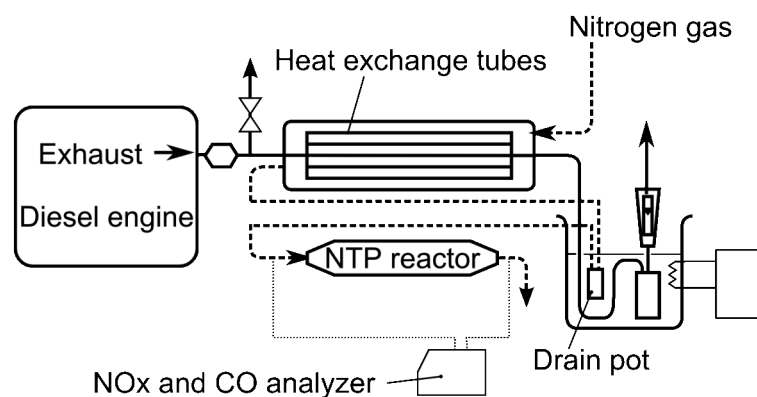
2. EXPERIMENTAL SETUP

Figure 2 shows the schematics of the experimental setup. In the present study, the performances of a single adsorption chamber followed by an NTP reactor are tested. A diesel engine generator (maximum load = 2 kW, displacement = 219 cc, bore diameter = 70 mm, stroke = 57 mm, cylinder number: single, rotating rate = 3600 rpm, exhaust gas flow rate = 300 NL/min, product number: YDG200SS, YANMER Co., Ltd.) is used in the experiment. The exhaust gas contents with 1 kW load are as follows: NO = 200 – 255 ppm, NOx = 230 – 275 ppm, CO = 300 – 350 ppm, CO_2 = 4%, H_2O = 7%. Figure 2(a) shows the layout for the adsorption process of NOx. This process is performed for 60 min. Exhaust gas from the engine at first passes through a DPF (Diesel Particulate Filter) in which PM (Particulate Matters) are removed. Subsequently, the exhaust gas, after a portion is allowed to the atmosphere through a valve in order to adjust the flow rate, passes through a cooling pipe in a water tank, a drain pot, a flow meter, and the adsorption chamber, in which NOx is adsorbed. The temperature of the water is kept at 5°C with a water cooler (LBT-400, As One Corp.). As a result, the dew point of the exhaust gas is reduced to approximately 10°C (water vapor content is 1.2 vol%). NOx and CO concentrations are analyzed with a NOx analyzer (PG-235, Horiba, Ltd) at the upstream and downstream of the adsorption chamber.

Figure 2(b) shows the layout for the desorption process. This process is performed for 45 min. Diesel exhaust whose flow rate is adjusted passes through heat exchange tubes installed in the adsorption chamber. Then the exhaust is discharged into the atmosphere after passing through the water tank and the flow meter. When the hot exhaust gas passes through the heat exchange tubes, the adsorbent pellets packed in the chamber are heated, resulting in thermal desorption of NOx. Simultaneously, nitrogen gas is supplied to the adsorption chamber in the opposite direction of that in the adsorption process. The N_2 gas elutes the desorbed NOx out of the chamber. This mixture of NOx and N_2, after being dehumidified by being cooled in the water tank, flows into the nonthermal plasma (NTP) reactor. NOx and CO concentration is analyzed with the NOx analyzer at the upstream and downstream of the NTP reactor. In the real experiment, the layouts in Figs. 2 (a) and (b) are interchanged with each other with valves not shown in the figure.
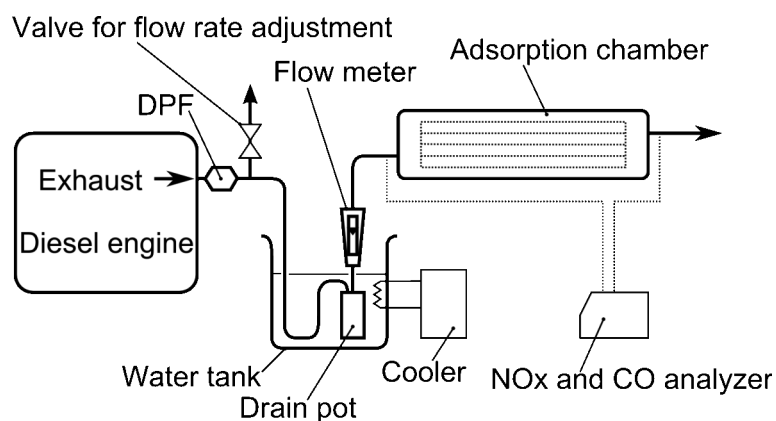
(a) Adsorption process

(b) Desorption process

Figure 2. Experimental setup (a) Layout for adsorption process and (b) Layout for desorption process

The adsorption chamber has an inner volume of 1.3 L where adsorbent pellets can be packed (a detailed description is in our previous paper [21]). The adsorbents tested in this study are zeolite 13X pellets (product name: 13X, 1/16 inch diameter, Union Showa K. K.) for NOx adsorption and silica gel pellets (5-10 mesh spherical pellets, Fuji Silysia Chemical Ltd.) for dehumidification. The exhaust gas passes through those pellets in the order of silica gel and zeolite 13X, so that NOx adsorption performance is not inhibited by water vapor. The packed volume is 466 mL and 933 mL for the silica gel and zeolite 13X pellets, respectively. In addition, the adsorption chamber has ports for thermocouples in order to measure the temperature of the adsorbent pellets.

Figure 3 shows the surface discharge element (OC-002, Masuda Research Inc.), which is assembled in the NTP reactor. The element consists of a tube made of alumina ceramic with outer and inner diameters of 13 mm and 8 mm, respectively. Half of the outer surface has a ground discharge electrode pattern. A high-voltage electrode is covered with an alumina wall. The NTP is generated on the surface by surface discharge. A copper rod with radiator fins is inserted in the center of the ceramic tube and fixed with a silicone insulator to cool the surface discharge element.
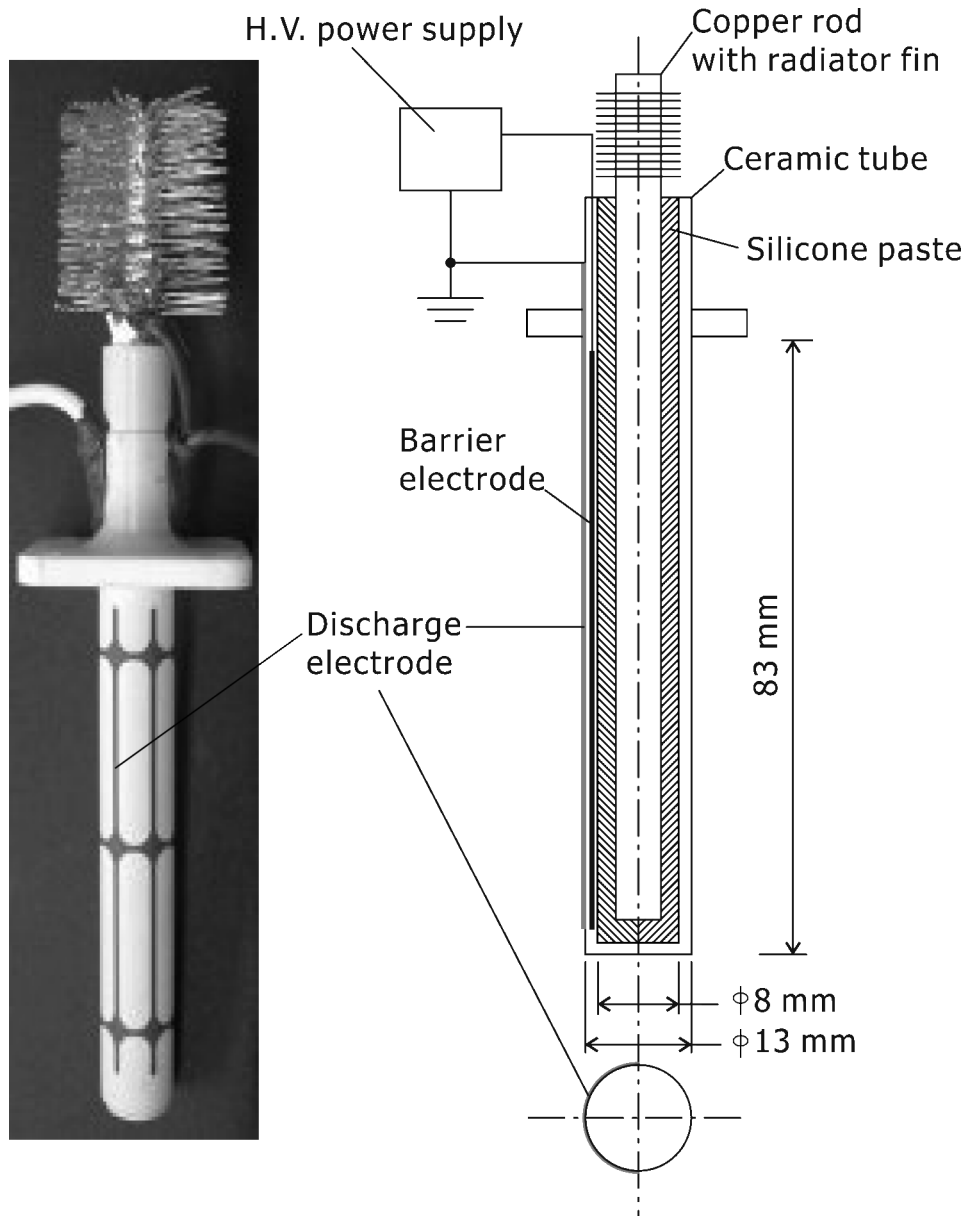

Figure 3. Surface discharge element

Figure 4 shows the three types of NTP reactors (Type I, II, and III) tested in this study. Every reactor is a stainless steel duct that is 600 mm long, 55 mm wide, and 85 mm high, and has 12 surface discharge elements. Figure 4 (a) shows the Type I reactor. Discharge electrode patterns face the center of the duct. Pieces of paper stuffing are placed between the inner wall of the duct and the elements to prevent the gas stream from passing. The gas stream passes through the gaps formed by the electrode patterns. Those gaps are 2 mm, and the pitch of the elements is 40 mm along the gas stream. Figure 4 (b) shows the Type II reactor. Twelve surface discharge elements are placed alternately inside the duct with the discharge electrode patterns facing upstream. The pitch between those elements is 80 mm along the gas

stream. Figure 4 (c) shows the Type III reactor. Each element is surrounded by an aluminum cover (inner diameter = 18 mm) and walls, so that the gas stream is limited near the surface of the element. Figure 4 (d) shows the electrical connection for these reactors. The surface discharge elements are connected in parallel and driven by a pulsed AC high voltage power supply (HC-RK, maximum power = 300 W, maximum peak to peak output voltage = 10 kV, Masuda Research Inc.) that is prepared for the elements with highly matched impedance. The applied high voltage is measured with a voltage divider (P6015A, Tektronix Japan, Ltd.) and a digital oscilloscope (DL1740, Yokogawa Electric Corporation). The sum of the power consumed at these elements is measured by the Lissajous method using a 0.47 μF capacitor inserted between the ground electrode and the ground.

(a) Type I

(b) Type II

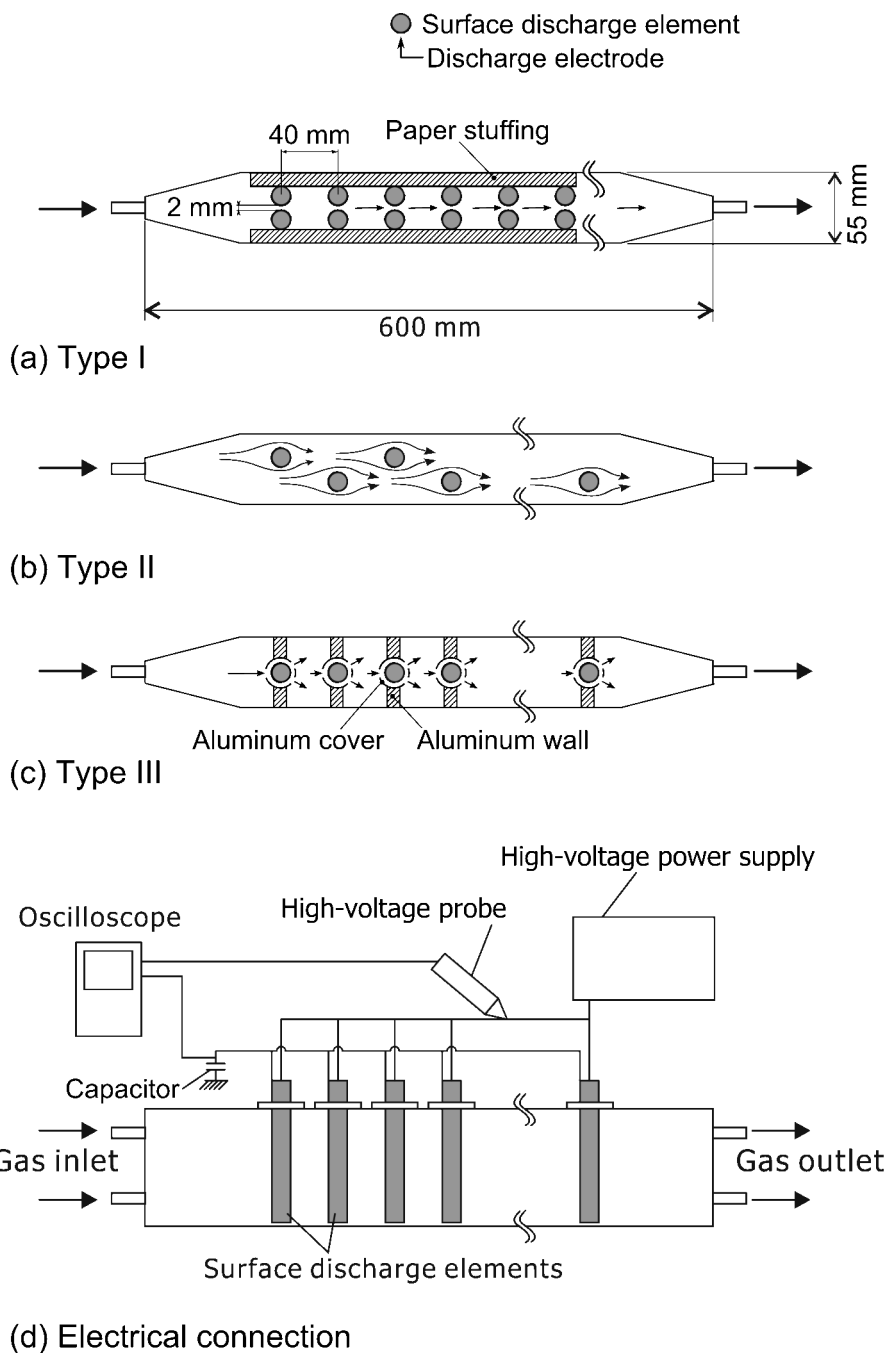(c) Type III

(d) Electrical connection

Figure 4. NTP reactors (a) Type I, (b) Type II, (c) Type III and (d) Electrical connection

Figure 5 shows the water-cooling system applied to the Type I reactor. As for the Type I reactor, the surface discharge elements are attached to the duct by being inserted from both sides. Water tubing is installed over the reactor, and water is sprayed from 1-mm holes formed on the tubing at the position over each radiator of the element. Water is supplied with a water pump at a total flow rate of approximately 3 L/min. The temperature of the water is always kept 10°C below the atmospheric temperature.
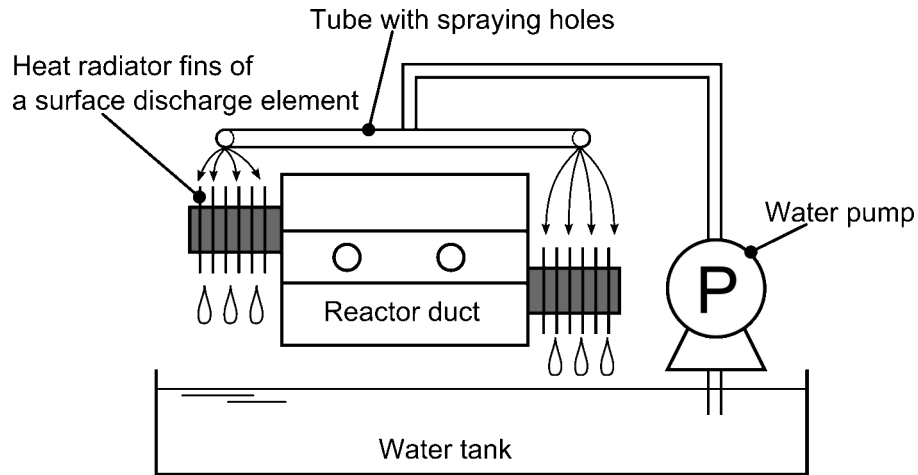

Figure 5. Water-cooling device for Type I reactor

Figure 6 shows a typical waveform of the applied voltage. The waveform has positive and negative peaks with a period of 100 ms. The discharge power is controlled by adjusting the peak voltage of the pulse.
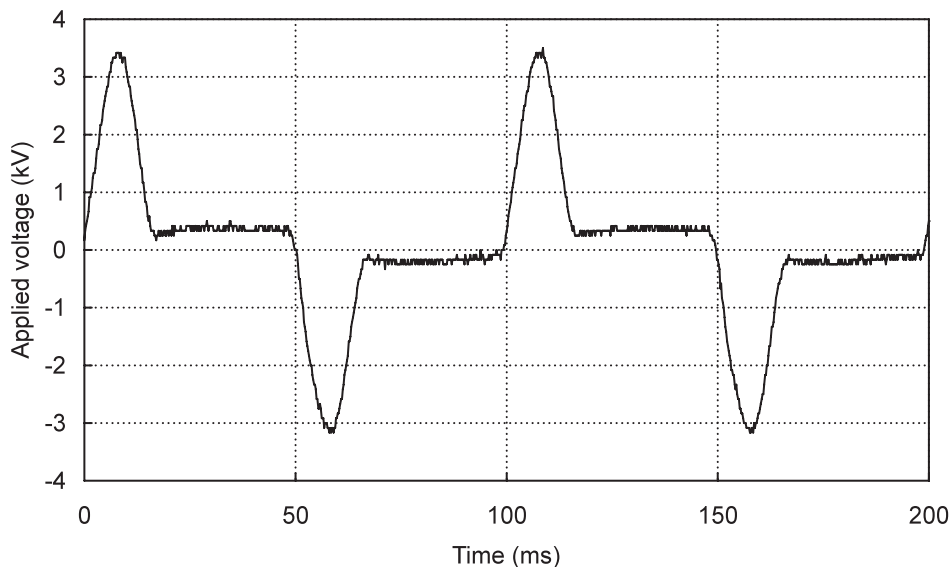

Figure 6. Applied voltage waveform

3. RESULTS AND DISCUSSION
3.1 Comparison of NTP reactors
Initially, comparison of the Type I, II, and III NTP reactors is conducted with simulated gas. The simulated gas composed of 2,000 ppm of NO diluted with nitrogen (purity = 99.999%) flows through the NTP reactors at a flow rate of 18 L/min at 298 K. Space velocities, which is (volumetric flow rate)

/ (volume of the duct – volume of the elements and the other contents), are 577, 454, and 492 h^{-1} for Type I, II, and III, respectively. Figure 7 shows the power consumption at the NTP reactors as a function of applied high voltage, V_{0-p} (zero-to-positive peak voltage). Almost no difference is observed between the water and air-cooling applied to the Type I and II reactors. The Type III reactor shows more power than the others. This larger power may be caused by an easier temperature increase in the elements because the elements are surrounded by aluminum covers. Figure 8 shows the results of the NOx reduction. The vertical axis means the mass flow rate of NOx (NO + NO_2) measured at the downstream of the reactors. In the figure, the NOx mass is shown based on the mass of NO_2 of the same mole number as that of NO + NO_2. This expression using the unit of g/kWh has been traditionally used in automobile emission regulations. Approximately 1 $g(NO_2)$/h corresponds to 492 ppm in 18 L/min at 298 K. A decrease in the NOx mass flow rate can be seen as the power consumption increases. This decrease in NOx could be caused by the reduction to N_2 according to the reactions (1) dissociation of the nitrogen molecule by NTP and (2) reduction of NO by the nitrogen radical [22].
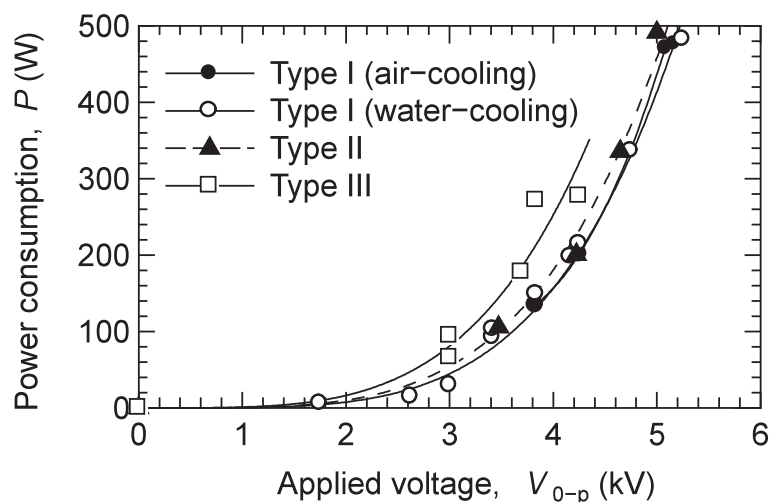
$$N_2 + e \rightarrow 2\,N\bullet + e \tag{1}$$

$$NO + N\bullet \rightarrow N_2 + O\bullet \tag{2}$$

Figure 7. Power consumption at NTP reactors as a function of applied voltage, V_{0-p}
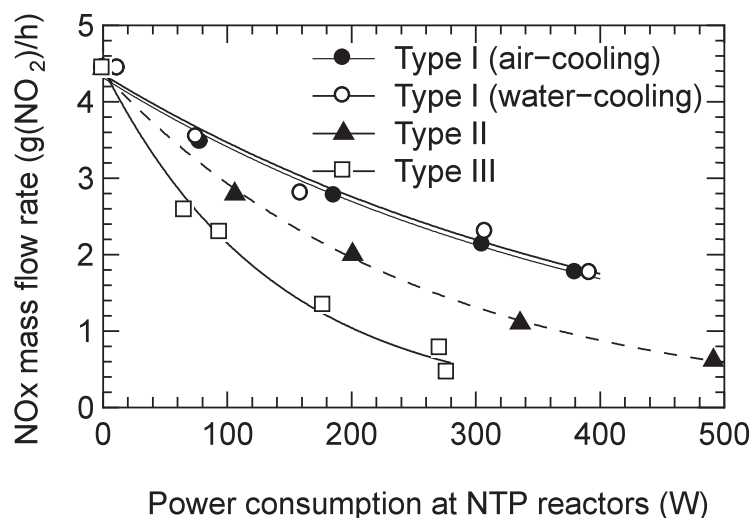

Figure 8. NOx reduction with simulated gas (2,000 ppm NO/N_2, 18 L/min, 298 K)

It is also observed that gradients of the reduction curves in Fig. 8 decrease as the power increases. This suggests an increase in power loss at larger power. Since it was confirmed that almost no concentration dependence exists in the reaction (2) in the range of 500–2,000 ppm of NOx [23], that power loss could be caused by Joule heating loss at the discharge elements. However, water-cooling does not show any effects in the range of this study probably because the water flow rate is too small. Among the reactors with air-cooling, Type III shows the highest performance. It is thought that the main gas stream passes near the region where NTP is generated with the existence of the aluminum covers. The energy efficiency (which is given as the mass of NOx reduced by unit energy consumption at the elements) is calculated as 19.4 g (NO_2)/kWh when approximately 50% NOx at the upstream is reduced. On the other hand, the Type I reactor shows the lowest performance. It is thought that NOx molecules are not sufficiently treated by NTP because a wake formed behind an element may cover a proceeding element. The energy efficiency of the Type I reactor is calculated as 8.9 g (NO_2)/kWh when 50% NOx at the upstream is reduced. In the same way, the energy efficiency of the Type II reactor is calculated as 12.0 g (NO_2)/kWh.

3.2 Application of an NTP reactor to the NOx treatment system

Initially, all the exhaust gas from the engine (300 NL/min) is treated using the system shown in Fig. 2. The load for the engine is always set at 1 kW. In the desorption processes, the exhaust gas temperature is 160°C at the inlet of the heat exchange tubes, and as a result, the maximum temperature of the adsorbent pellets increases up to approximately 90°C. In this experiment, the adsorption and desorption processes are repeated twice, and NTP is applied to the second desorption process. N_2 at 18 L/min flows through the adsorbent pellets and subsequently through the Type I NTP reactor with air-cooling in the desorption process. Figure 9 shows the NOx and CO mass flow rate at the upstream and downstream of the NTP reactor during 45 min of the desorption process. The power consumption of the reactor is set at 160 W. 1 g(NO_2)/h of NOx and 1 g(C)/h of CO correspond to 492 ppm and 1,887 ppm at 298 K, respectively. Since those concentrations of up- and downstream are measured alternately, there are no data of the upstream during the early period. It is clear that the NOx mass flow rate at the downstream of the NTP reactor is smaller than that at the upstream. The energy efficiency of the NOx reduction is calculated as 9.4 g(NO_2)/kWh based on the difference in the NOx mass flow rate between the up- and downstream at 14 min. This value is slightly larger than that obtained in the experiment with simulated gas because the power of 160 W is relatively low as compared to the reduction data in Fig. 8. In addition, it is observed that CO is generated by NTP. The CO is generated from the reduction of the desorbed CO_2. Because zeolite 13X can adsorb CO_2, the desorbing gas contains CO_2. The peak mass flow rate of the CO generated is 1.8 g(C)/h, while CO in exhaust gas ranges from 2.5 – 2.7 g(C)/h. It is considered that the CO generation is not a serious problem because it can be easily burned or oxidized to CO_2 inside the engine by introducing the gas from the NTP reactor to the engine intake. The mass flow rate of CO_2 is 27 g(C)/h at the maximum, though it is not described in Fig. 9. These results suggest that components that may inhibit NOx reduction, such as CO_2 and hydrocarbons, do not strongly affect the reduction of NOx.
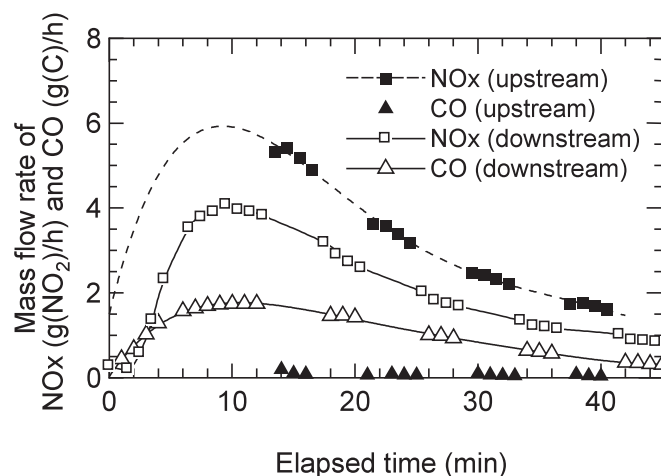

Figure 9. Reduction of NOx by NTP in the NOx aftertreatment system

Secondary, NOx aftertreatment is conducted for 90 NL/min of exhaust gas from the engine. The engine is operated with a 1-kW load during the adsorption processes and is operated with a 2-kW load during the desorption processes in order to enlarge the desorption performance. In desorption processes, the gas temperature is 220°C at the inlet of the heat exchange tubes, and the temperature of the adsorbent pellets is increased to 105°C. It should be noted that 105°C is lower than the exhaust gas temperature of the idling operation. Therefore, the higher temperature needed for desorption will be easily obtained by improvement in the heat exchanger. In this experiment, the adsorption and desorption processes are repeated five times, and then NTP is applied only to the 2nd and 4th desorption processes with the Type III reactor (120 W power consumption at the reactor). The total operation time of the treatment system is 8 h and 45 min. Figure 10 shows the mass of NOx adsorbed and desorbed in each process. It can be seen that the adsorption amount tends to decrease as the number of repetitions increases, while the desorption amount (without NTP) increases more rapidly. Those amounts are assumed to become equal when the NOx treatment system reaches a steady state. Considering the fact that approximately 2.7 $g(NO_2)$ of NOx is exhausted with 90 NL/min of exhaust gas from the engine under investigation for 60 min, it is concluded that the NOx adsorbent keeps adsorption efficiency higher than 50% during this experiment. When NTP is applied, it is observed that most NOx is reduced. According to the previous discussion and based on the energy efficiency of 19.4 g (NO_2)/kWh, the Type III reactor can reduce NOx of approximately 1.7 g (NO_2) during one desorption process. Therefore, even when the NOx treatment system reaches a steady state and the desorption amount becomes larger, it is expected that most NOx desorbed is kept reduced. The total energy efficiency of the NOx treatment system during this experiment (the removed amount of NOx per electrical energy consumption at the NTP reactor) is calculated as 15.4 g (NO_2)/kWh. The value corresponds to about 8.4% fuel consumption (estimation in the case of reducing NOx from 2.0 g (NO_2)/kWh of the 2005 Japan regulation for heavy weight cars to 0.7 g (NO_2)/kWh of the 2009 regulation). A savings in the NTP power and other power loss would reduce the fuel consumption penalty. Another topic for future study is the estimation of the durability of adsorbents and surface discharge electrodes against sulfur components.
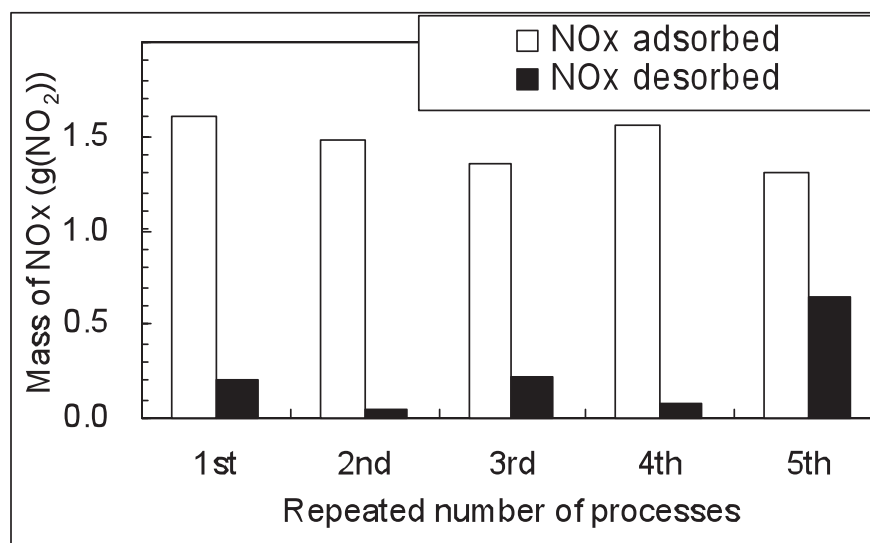

Figure 10. Result of NOx aftertreatment for a diesel engine

4. CONCLUSIONS

The NOx reduction performances of three types of NTP reactors (Type I, II, and III) are compared, and these reactors are combined with a NOx aftertreatment system. The conclusions are summarized below.

    (1)   Performance evaluation using 18 L/min of simulated gas composed of 2,000 ppm NO diluted with N_2 gas is carried out. The energy efficiencies of the Type I, II, and III reactors are calculated as 8.9, 12.0, and 19.4 g (NO_2)/kWh, respectively. The reason for the highest performance of Type III could most likely be the fact that most of the gas stream passes through the region where NTP is generated with the help of aluminum covers.

(2) Water-cooling is applied to the Type I reactor; however, water-cooling does not show any effect on the power consumption or performance of NOx reduction.

(3) The Type I reactor is combined with the NOx aftertreatment system, and reduction of NOx that desorbs from the adsorbent is performed with 160 W power consumption at the reactor. As a result, the energy efficiency of NOx reduction of 9.4 g(NO_2)/kWh is obtained. The result suggests that components that may inhibit NOx reduction, such as CO_2 and hydrocarbons, do not strongly affect the reduction of NOx.

(4) The Type III reactor is combined with the aftertreatment system and NOx treatment for the exhaust gas of 90 NL/min is performed five times with ad/desorption (corresponds to 8 h and 45 min). As a result, the NOx adsorbent keeps the adsorption efficiency higher than 50% during this experiment. When NTP is applied, most NOx desorbed is reduced. According to the previous conclusions, most NOx desorbed is expected to keep being reduced in the system that reaches a steady state of ad/desorption. The total energy efficiency of the NOx treatment system during this experiment (the removed amount of NOx per electrical energy consumption at the NTP reactor) is calculated as 15.4 g (NO_2)/kWh.

ACKNOWLEDGEMENTS

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