Development of super-clean diesel engine and combustor using nonthermal plasma hybrid aftertreatment

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Abstract. One of important and successful environmental applications of atmospheric-pressure corona discharge or plasma is electrostatic precipitator (ESP), which have been widely used for coal- or oil-fired boilers in electric power plants and particulate matter control emitted from industries such as glass melting furnace system, etc. In the ESPs, steady high voltage is usually applied to a pair of electrodes (at least, one of these has sharp edge). Unsteady pulsed high voltage is often applied for the collection of high-resistivity particulate matter (PM) to avoid reverse corona phenomena which reduce the collection efficiency of the ESPs. It was found that unsteady high voltage can treat hazardous gaseous components (NO\textsubscript{x}, SO\textsubscript{x}, hydrocarbon, and CO, etc.) in the exhaust gas, and researches were shifted from PM removal to hazardous gases aftertreatment with unsteady corona discharge induced plasmas. In the paper, recent results on diesel engine and industrial boiler emission controls are mainly reviewed among these our research topics.

1. Introduction
One of important and successful environmental applications of atmospheric-pressure corona discharge or plasma is electrostatic precipitator (ESP), which have been widely used for coal- or oil-fired boilers in electric power plants and particulate matter control emitted from industries such as glass melting furnace system, etc. In the ESPs, steady high voltage is usually applied to a pair of electrodes (at least, one of these has sharp edge). Unsteady pulsed high voltage is often applied for the collection of high-resistivity particulate matter (PM) to avoid reverse corona phenomena which reduce the collection efficiency of the ESPs. It was found that unsteady high voltage can treat hazardous gaseous components (NO\textsubscript{x}, SO\textsubscript{x}, hydrocarbon, and CO, etc.) in the exhaust gas, and researches were shifted from PM removal to hazardous gases aftertreatment with unsteady corona discharge induced plasmas [1-3].

In our laboratory, we have tried to develop nonthermal-plasma environmental applications technology since 1998. Main research topics are exhaust gas cleaning as follows: diesel engine emission control, boilers emission control, NO\textsubscript{x} removal for glass melting furnace, VOC (volatile organic compounds) control from factories, odor and dioxin controls from garbage incinerators, CO\textsubscript{2} and PFC (perfluoro compounds) emission and moisture controls in various environments, and indoor electric air cleaner. All our papers are listed in [4]. In the paper, recent results on diesel engine and industrial boiler emission controls are mainly reviewed among these our research topics.
2. Marine Diesel Engine Emission Control [5-8]

A photograph of the target diesel engine (6DK-20e, Daihatsu Diesel MFG Co. Ltd., Japan) is shown in figure 1. In marine vessels, diesel engines are used not only as the main propulsion power sources but also for electrical sub power generation. In this study, the target is the latter of these. The specifications of the engine are as follows: a maximum or 100% output power of 1071 kW, four cycles, six cylinders with a cylinder bore of 200 mm and a stroke of 300 mm, a constant rotation rate of 900 rpm, and the total mass of the engine and dynamo of 16 ton. Marine diesel oil (A-heavy oil, sulfur = 0.075 mass%, nitrogen = 0.01 mass%, heat quantity = 45.4 MJ/kg) is used as the fuel. The exhaust flow rate is 3920 Nm$^3$/h for 50% load or output power, 5526 Nm$^3$/h for 75%, and 6815 Nm$^3$/h for 100% (N denotes the standard state of 0°C, 0.1 MPa).

Figure 1. Photograph of the targeted diesel engine (6DK-20e, max. power = 1071 kW) [5]

Figure 2 shows the experimental setup for the aftertreatment system in the marine diesel engine (6DK-20e, output power = 1071 kW, Daihatsu Diesel MFG Co., Ltd, fuel: A-heavy oil or marine diesel oil). In this system, the combination of adsorption and the NTP (nonthermal plasma) is used for reduction of NO$_x$. The exhaust gas emitted from the engine is first separated and approximately 13% of the bypassed gas the flow rate of which is controlled by the flow regulation valve passes through a set of ceramics diesel particulate filters (DPFs). More than 95% of the PM in the exhaust gas is

Figure 2. Schematic diagram of experimental setup for the aftertreatment system in the marine diesel engine [5]
captured in the DPFs. The accumulated PM in the DPF is treated by nonthermal plasma-induced ozone (O\(_3\)) injection technology \([6, 8]\). After this process of the PM removal, NO\(_x\) in the exhaust gas is treated by NTP combined with repeated adsorption and desorption processes. In the adsorption process, for an effective NO\(_x\) adsorption to adsorbents, the exhaust gas is cooled from 270°C down to approximately 45°C with the water-cooling type cooler. An adsorption chamber that contains adsorbents (1.2–2.4 mm-sized granular pellets of MnO\(_x\)–CuO compound) adsorbs NO\(_x\) of 92% in maximum. Finally, the exhaust gas is flowed out from a stack. In the desorption process, after the valves are controlled, N\(_2\) gas of lower flow rate of 11.1 ~ 22.2 Nm\(^3\)/h supplied from liquid N\(_2\) cylinder passes through the chamber. The direction of the N\(_2\) gas is opposite to that of the exhaust gas in the adsorption. Simultaneously, the chamber is heated up by the waste heat of the exhaust gas. As a result, high-concentration (typically 1% = 10000 ppm) NO\(_x\) flows out from the chamber, and introduced to the NTP reactors of total 21.6 kW in maximum.

**Figure 3** shows a time-dependent NO\(_x\) emission before and after the gas passes through the aftertreatment system for operation cycles 2 ~ 4, 9, and 10. Exhaust gas is continuously treated during these repeated cycles. In this graph, the mass flow rate of NO\(_x\) is taken in the vertical axis evaluated by NO\(_x\) analyzers based on the molecular mass of NO\(_2\) with the unit of g(NO\(_2\))/h. Furthermore, the untreated NO\(_x\) is indicated by white circles with lines, and treated NO\(_x\) is indicated by black circles.

**Figure 3.** Time-dependent NO\(_x\) emission from the aftertreatment system for the marine diesel engine

![Figure 3](image_url)

**Figure 4.** IMO emission standards and the required NO\(_x\) reductions for Tiers I, II, and III \([5]\)

![Figure 4](image_url)
with lines. NTP is applied only in the desorption processes, and the input power to the NTP generator is 21.6 kW in the cycles 2 ~ 4 and 12.0 kW in the cycles 9 and 10. It is known this graph that large amount of NO\textsubscript{x} is treated. At 10th cycle, the highest system energy efficiency among these cycles of \( \eta_{\text{system}} = 161 \text{ g(NO}_2\text{/kWh} \) is achieved. Application of the highest recorded energy efficiency to the requirement of the IMO (International Maritime Organization) emission standards from Tier II to III corresponds to only 4.3% (= 6.9 / 161 \times 100) of the engine output power as explained in figure 4.

3. Industrial Boiler Emission Control [9-13]

The number of small boilers using city natural gas, heavy oil, and waste oils has been increasing year by year in Japan, and more stringent regulations for NO\textsubscript{x} emission are being anticipated in order to reduce environmental NO\textsubscript{x} concentration. A suitable flue gas treatment system for small boilers will be required soon. Several studies have been conducted on laboratory-scale nonthermal plasma–chemical hybrid processes for the removal of NO\textsubscript{x} from gases emitted from various stationary sources. The authors propose a plasma–chemical hybrid process consisting of an indirect nonthermal plasma process and wet-chemical treatment. The principle of NO\textsubscript{x} removal is as follows:

\[
\begin{align*}
\text{NO} + \text{O}_3 & \rightarrow \text{NO}_2 + \text{O}_2 \quad (1) \\
2\text{NO}_2 + 4\text{Na}_2\text{SO}_3 & \rightarrow \text{N}_2 + 4\text{Na}_2\text{SO}_4 \quad (2)
\end{align*}
\]

Figure 5 shows an overview of the plant. The flue tube boiler (Takao Iron Works Co., Ltd.) has an original rotary burner for gas and/or oil and is operated by using city natural gas (13A) at 157 Nm\textsuperscript{3}/h and A-type heavy oil at 171 L/h and waste oils. The boiler has a steam generation rate of 2.5 t/h. Two sets of silent discharge-type ozonizers (EW-90Z, Ebara Jitsugyo Co., Ltd.) equipped with a pressure swing adsorption (PSA) oxygen generator are employed to generate O\textsubscript{3}.

Figure 6 shows the structure of the discharge section of the plasma reactor in the ozonizer. A coaxial NTP reactor or silent discharge-type reactor is used. O\textsubscript{3} gas is generated after O\textsubscript{2} gas by PSA passes through the 30 plasma reactors inside the ozonizer and then injected into a flue gas duct for NO oxidation. The O\textsubscript{3} gas flow rate is constant, 0.9 Nm\textsuperscript{3}/h. When the discharge power is 1.5 kW and the PSA power is 1.6 kW, 90 g/h of O\textsubscript{3} is generated in maximum with a concentration of 4.7% and the energy efficiency of 29 g/kWh. High-concentration O\textsubscript{3} gas is injected into the duct and diluted in order to oxidize almost all the NO to NO\textsubscript{2} according to reaction (1). The flue gas is then introduced into the scrubber in order to reduce NO\textsubscript{2} to N\textsubscript{2} according to reaction (2).

Experimental results and discussion are shown next. In order to confirm NO\textsubscript{x} removal performance, the mixed oil of WVO and heavy oil and city natural gas are tested as the boiler fuels.
Figure 6. Structure of the discharge section in the ozonizer

Figure 7 shows time-dependent NO and NOx concentrations at MP1 (measurement point 1 at the exit of the boiler) and MP3 (measurement point 1 at the exit of the exhaust gas cleaning) [13]. The NOx levels are measured by NOx analyzers. The experiment is carried out with daily start and stop operation. The average flow rate of city natural gas is 52 Nm$^3$/h and that of flue gas is 718 Nm$^3$/h at an O$_2$ concentration of 3.3–4.0%. The average rates of O$_3$ injection and Na$_2$SO$_3$ supply are 86 g/h and 63 mol/h. NOx concentrations at MP1 are within 45–50 ppm. This small difference of NOx each day is due to the difference in the O$_2$ concentrations at MP1 between 3.3% and 4.0%. The NOx concentrations at MP3 are within 5–8 ppm, which are equivalent to a NOx removal efficiency of more than 85% over an operating time of 23 h. The NOx removal performance achieved is satisfactory and stable. Concerning the specific gravity of the scrubbing solution, it increases gradually and is stable at approximately 1.08 which is an allowable value as a control index.

Figure 7. Time-dependent NO and NOx concentrations at MP1 (boiler exit) and MP3 (exit of aftertreatment) during a four-successive-day operation in firing city natural gas.
4. Conclusion
Recent results on the development of super-clean diesel engine and combustor using nonthermal plasma hybrid aftertreatment are reported. We achieved highest removal efficiency of NOx reduction greater than 100 g(NO$_2$)/kWh when the nonthermal plasma is combined with other chemical processes (adsorption or wet chemical scrubbing). The efficiency is approximately ten-times higher than that in the case with plasma application only.

An adsorption-plasma combined NOx aftertreatment system for marine diesel exhaust gas is developed. An experiment using a pilot-scale aftertreatment system for a marine diesel engine with an output power of 1 MW has been carried out using an NTP generator with a power of 21.6 kW. The characteristics of NOx adsorption/desorption and NOx reduction by NTP are analyzed using the experimental data. Finally, the energy efficiency of NOx reduction by NTP is evaluated. This system demonstrates an excellent system energy efficiency for NOx removal of 161 g(NO$_2$)/kWh. Because high-concentration NOx is treated by NTP with the help of NOx adsorption and desorption, the present aftertreatment system can achieve higher energy efficiency.

Further, the pilot-scale investigation of a plasma–chemical hybrid NOx removal process is carried out using a low emission boiler system comprising multi-fuel boiler and a chemical scrubber. The amount of NO removed is almost the same as the amount of the corresponding ozone concentration to oxidize NO to NO$_2$ (1:1 stoichiometric ratio) regardless of the flue gas flow rate. NOx removal efficiency of more than 85% is achieved over an operating time of 23 h when firing city natural gas.

It is confirmed that the present nonthermal plasma hybrid aftertreatment is very effective to these targets and applicable for industrial use.

Acknowledgments
The author thanks Dr. H. Fujishima, Dr. T. Kuroki (Osaka Prefecture University), Dr. T. Kuwahara (Nippon Institute of Technology), and Dr. K. Yoshida (Osaka Institute of Technology) for their support in the researches. The author also thanks kind reproduction permission for Figures 1, 2 and 4 from Springer Science and Business Media. This work was supported by the Regional R&D Resources Utilization Program in JST and JSPS KAKENHI Grant Number 24246145 and 249848.

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