

Pilot-Scale NO_x and SO_x Aftreatment Using a Two-Phase Ozone and Chemical Injection in Glass-Melting-Furnace Exhaust Gas

Hashira Yamamoto^{1,2}, Tomoyuki Kuroki², Hidekatsu Fujishima², Masaaki Okubo²

¹Environmental Affairs Office
Nihon Yamamura Glass Co., Ltd.
e-mail: yamamoto_h3@yamamura.co.jp

²Dept. of Mechanical Engineering
Osaka Prefecture University
e-mail: fujisima@me.osakafu-u.ac.jp; kuroki@me.osakafu-u.ac.jp;
mokubo@me.osakafu-u.ac.jp

Abstract—As NO_x and SO_x have significant environmental impacts, advanced treatments are required to remove them from the exhaust gas of a glass melting furnace. Here, we investigate a plasma-chemical hybrid process (PCHP) for this purpose. A pilot-scale experiment of the simultaneous removal of NO_x and SO_x using PCHP combined with the existing semi-dry type desulfurization reactor is conducted on actual high-temperature exhaust gas from a glass melting furnace. NO (the majority of NO_x is NO) in the exhaust gas is oxidized to NO_2 using active oxygen (ozone: O_3) generated by a plasma ozonizer. The exhaust gas must be cooled to less than 150 °C in order to suppress the thermal decomposition of O_3 , while the gas temperature at the outlet of the semi-dry reactor must be kept at 200–250 °C to protect the dry-type electrostatic precipitator (ESP). Therefore, it is important to form a local cooling area for NO oxidation in the reactor. In this study, we use the three-fluid nozzles of O_3 , water, and air to form the local cooling area and effectively oxidize NO to NO_2 . In addition, we spray NaOH aqueous solution for SO_2 absorption downstream of the NO oxidation area to allow sufficient time for NO oxidation. The SO_2 reacts with NaOH to produce Na_2SO_3 , a powerful reducing agent. Subsequently, NO_2 reacts with Na_2SO_3 and is reduced to N_2 , and the Na_2SO_4 generated in this reaction is reused as a clarifier of the raw materials for glass manufacturing. As a result, the ratio of the amount of removed NO and NO_x to the amount of injected O_3 (de-NO/ O_3 and de- NO_x/O_3) is 64% and 78%, respectively; therefore, high efficiency is obtained. This study includes actual examples of the treatment of exhaust gas in a glass melting furnace, using PCHP de- SO_x and de- NO_x technologies along with results from pilot-scale experiments.

I. INTRODUCTION

Glass melting furnaces use the combustion of city gas, heavy oils, and other fuel sources to melt raw materials at approximately 1500 °C. The exhaust gas generated includes nitrogen oxides (NO_x), sulfur oxide (SO_x), and other harmful substances that have significant negative environmental impacts. SO_x and dust in the exhaust gas are removed by an aftreatment system consisting of a chemical reactor using a solution such as sodium hy-

dioxide (NaOH) [1, 2]. The product of this reaction is easily reused as a raw material in the glass melting furnace. Moreover, the flue gas emitted from furnaces includes low melt and adhesive dust such as several alkali metal compounds with a high concentration of SO_x from the raw glass material and fuel. Selective catalytic reduction (SCR) using urea or ammonia is typically used for the treatment of NO_x emissions [3, 4]. However, the catalyst of SCR is clogged, and the ability of the catalyst is reduced by the low melt and adhesive dust in the exhaust gas of glass melting furnace. For this reason, a general denitrification equipment are both difficult to apply to the exhaust gas of glass melting furnaces [5]. Thus, NO_x emission is generally suppressed on the combustion side using a method for reducing the combustion air ratio in glass melting furnaces. Advanced treatments are required to remove NO_x emissions from the exhaust gas.

Substantial research into denitrification technology without the use of a catalyst has been reported [6, 7]. A plasma-chemical hybrid process (PCHP) without catalysts has proven to be suitable for the exhaust gas of glass melting furnaces. The authors previously employed a PCHP and experimented laboratory [8, 9] to conduct a pilot-scale experimental exhaust gas treatment simulating the exhaust gas conditions of a glass melting furnace. PCHP technology without the use of a catalyst can combine a general desulfurization reaction with a denitrification reaction. Moreover, the PCHP can be used for both wet (low-temperature) [2] and dry (high-temperature) [1] exhaust gas treatments; thus, we have developed this method for application to exhaust gas from a glass melting furnace.

In this study, a pilot scale experiment is conducted in an actual semi-dry type after-treatment system of a de- SO_x reactor in combination with a two-phase ozone and a wet chemical process for a glass melting furnace.

II. PRINCIPLE OF AFTERTREATMENT SYSTEM

The plasma combined semi-dry chemical process is composed of two steps: one that oxidizes the nitric oxide (NO) of flue gas in NO_x to nitrogen dioxide (NO_2) by ozone (O_3) (plasma treatment) and one that desulfurizes SO_x to sodium sulfite (Na_2SO_3) by NaOH (chemical process). Then, the generated substances are reacted with each other. The following NO_x removal process is utilized. Plasma induced O_3 gas with gas cooling water, using oxygen as the raw material, is injected into the reactor and decomposed to oxygen radicals. NO generated by combustion in an exhaust gas is oxidized to water-soluble NO_2 using the oxygen radicals at a gas cooling area. SO_x in the exhaust gas is treated to become Na_2SO_3 solution using NaOH solution at the gas cooling area through the desulfurization process. NO_2 is treated to become harmless nitrogen (N_2) by a chemical process using Na_2SO_3 , producing sodium sulfate (Na_2SO_4) in the same area. After the semi-dry state, the Na_2SO_4 is completely dried by the heat of the exhaust gas. Furthermore, the Na_2SO_4 is collected in the dust collector by an electrostatic precipitator (ESP) and bag-filter (BF). The collected dust is reused as a raw material for a glass manufacturing system and NO_x and SO_x are continuously removed.

III. EXPERIMENTAL APPARATUS AND METHODS

The experiment was conducted at the semi-dry type exhaust gas aftertreatment system for the glass melting furnace of Nihon Yamamura Glass Tokyo Plant (Kanagawa, Japan). The furnace can process 245 ton/day of raw glass material by combustion of heavy oil and

LNG at a rate of 1300 L/h and 100 m³/h, respectively. The internal temperature of the furnace is maintained at approximately 1550 °C by controlling the flow rate of fuel and combustion air. A set of combustion burners, located on the north and south side, is installed inside the furnace. Three burners are located on each side (north and south direction) and are alternated every 20 min for heat exchange with the regenerative furnace. The average exhaust gas flow rate and temperature are 30000 m³N/h (N: standard state, 0 °C, and atmospheric pressure) and 450 °C, respectively.

Fig. 1 shows a schematic diagram of the semi-dry type aftertreatment system with the plasma combined chemical process for the glass melting furnace. The pilot-scale experiment was performed during adjustment of the combustion amount at a heavy oil rate of 301 L/h with an LNG rate of 620 m³/h (raw glass material = 153 ton/day). As a result, the exhaust gas of 16800 m³N/h at the reactor inlet of SP1 is then introduced into the semi-dry reactor. The reactor is a cylindrical apparatus ($\phi 3.5 \text{ m} \times 15 \text{ m}$, Asahi Glass Engineering Co., Ltd.) containing three-fluid spray nozzles (GSIM2055S3B, H.IKEUCHI & Co., Ltd.) for gas cooling of O₃ and two-fluid spray nozzles (GSIM6055II, H.IKEUCHI & Co., Ltd.) for desulfurization. The water and compressed air with O₃ in the three-fluid nozzles is sprayed. A water mist is formed by spraying compressed air and soft-water (0.80 m³/h). The three-fluid nozzles are installed in the vertical direction (upward) at 30° and mounted 1.9 m downstream from an inlet of the reactor. O₃ is emitted into the localized gas cooling area formed by water spray. A part of O₃ is absorbed by the sprayed water, then the gas and the water of O₃ is existed as two-phase O₃ in the localized gas cooling area. The NO in the exhaust gas is oxidized to NO₂. The O₃ gas is generated by the air-cooled surface discharge plasma ozonizers. The absorbent aqueous solution containing NaOH (2.0%) and Na₂SO₃ (0–1.0%) and compressed air, used to create a mist at the two-fluid nozzles mounted horizontally, is sprayed. A water mist is formed by spraying compressed air and the NaOH solution (0.69–0.76 m³/h). The two-fluid nozzles are mounted 2.0 m downstream from the three-fluid nozzles. Reaction of SO₂ in the exhaust gas with NaOH produces SO₃²⁻ as a by-product in the localized cooling area. In addition, the NO₂ generated by O₃ injection reacts with SO₃²⁻ and transforms to N₂ and a solution containing SO₄²⁻ ion. The solution containing SO₄²⁻ is dried to a dust of Na₂SO₄ particles by the outlet of the reactor.

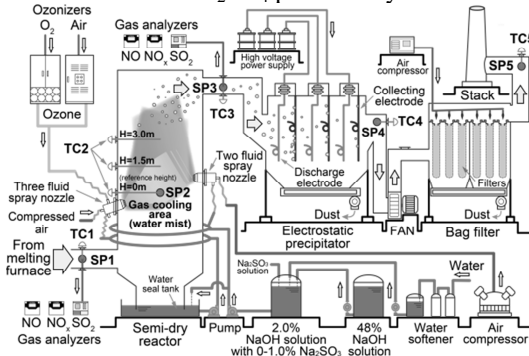


Fig. 1. Schematic diagram of the semi-dry type aftertreatment system with the plasma combined chemical process for the glass melting furnace

Downstream of the reactor, the dried Na₂SO₄ particles contained in the exhaust gas are collected by the dry type ESP (DC voltage = 45 kV, current = 200 mA, Asahi Glass En-

gineering Co., Ltd.) operated with an applied voltage of 23–29 kV with a current control of 61–75 mA. Further downstream of the ESP, the BF (filter = $\phi 155 \text{ mm} \times 6 \text{ m}$, number = 448 filters, Asahi Glass Engineering Co., Ltd.) cleanses the gas of dust to produce a highly purified exhaust gas. The temperature of the exhaust gas is measured at the following points: TC1 is at the inlet of the reactor as SP1, TC2 is at the inside of the reactor (gas cooling area) as SP2 (at 3 heights downstream; $H = 0 \text{ m}$, 1.5 m , 3.0 m), TC3 is at the outlet of the reactor as SP3. The concentrations of the exhaust gas components such as NO , and NO_x are measured at the inlet (SP1) and outlet (SP3) measurement points of the reactor using the NO analyzer (SP1; NOA7000, Shimadzu Corporation) (SP3; PG-202, Horiba, Ltd.), and NO_x analyzer (SP1; URA-4N, Shimadzu Corporation) (SP3; NOA7000, Shimadzu Corporation). The concentration of O_3 generated is measured using an ozone monitor (EG-550, Ebara Jitsugyo Co., Ltd.).

The specifications of the ozonizer A and B are shown in **Table 1**. Ozonizer A (EW-90Z, Ebara Jitsugyo Co., Ltd.) has a pressure swing adsorption (PSA) oxygen (O_2) generator. Ozonizer B is composed of nonthermal plasma (NTP) reactors (HCII-OC70, Masuda Research Inc.) and supplied with O_2 gas from liquid oxygen. A unit of NTP generators consists of 12 NTP reactors and 6 power supplies. For the pilot-scale experiments, three A ozonizers and four B ozonizers are used. The total amount of O_3 generated is 1.4 kg/h with an O_3 gas concentration of 2.0% ($= 43 \text{ g/m}^3\text{N}$) and a maximum power consumption of 22 kWh .

TABLE 1: SPECIFICATIONS OF OZONIZER A AND OZONIZER B

Ozonizer No.	A	B
Manufacturer	Ebara Jitsugyo Co., Ltd.	Masuda Research Inc.
Type	EW-90Z	HCII-OC70 \times 12
Number of ozonizer	3 ozonizers	4 ozonizers
Raw material gas	More than 90% O_2 gas from PSA	O_2 gas from liquid oxygen
O_3 generation	Air-cooled silent discharge plasma	Air-cooled surface discharge plasma
O_3 concentration	$0\text{--}100 \text{ g/m}^3\text{N} = 0\text{--}4.7\%$	$38 \text{ g/m}^3\text{N} = 1.8\%$
O_3 volume flow rate (one device / total)	$0.9 \text{ m}^3\text{N/h} / 2.7 \text{ m}^3\text{N/h}$	$7.2 \text{ m}^3\text{N/h} / 29 \text{ m}^3\text{N/h}$
O_3 mass flow rate (one device / total)	$90 \text{ g/h} / 270 \text{ g/h}$	$275 \text{ g/h} / 1100 \text{ g/h}$
Power consumption (one device / total)	$3.1 \text{ kWh} / 9.3 \text{ kWh}$	$5.4 \text{ kWh} / 22 \text{ kWh}$
Dimension (W \times D \times H)	$740 \text{ mm} \times 925 \text{ mm} \times 1840 \text{ mm}$	$1000 \text{ mm} \times 1600 \text{ mm} \times 700 \text{ mm}$

Fig. 2 shows an example of the measured discharge voltage and the current waveforms for the pulse high-voltage power supply powering the NTP reactors when O_2 gas is passed through them at a flow rate of 36.7 L/min . The discharge voltage and current waveforms are measured using voltage and current probes. In this figure, the input power to the power supply used to drive the two discharge tubes is 560 W . The O_3 concentration at this time is 51 g/m^3 .

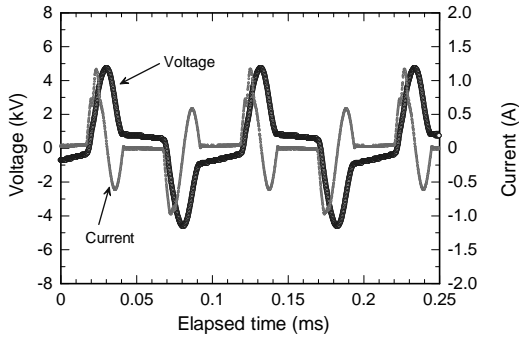


Fig. 2. An example of the measured discharge voltage and current waveforms for two NTP reactors in ozonizer B

IV. RESULTS AND DISCUSSION

Table 2 lists the experimental conditions of T1 to T3 test periods with O_3 gas, water spray, and NaOH solution spray. The conditions are changed between each test period and measured for 120 consecutive minutes. The flow rate of the exhaust gas is $16800 \text{ m}^3\text{N/h}$. The temperature range of the reactor inlet is $490\text{--}493 \text{ }^\circ\text{C}$. The NO , NO_x , and SO_x concentration ranges are $214\text{--}226 \text{ ppm}$, $222\text{--}235 \text{ ppm}$, and $306\text{--}335 \text{ ppm}$, respectively. The average O_2 concentrations at the inlet and the outlet of the reactor are 5.7% and 8.0% , respectively. Hereafter, all gas concentrations are expressed as converted values based on 15% oxygen concentration. O_3 gas is injected with mass flow rates of $1.78\text{--}1.79 \text{ kg/h}$. The Na_2SO_3 concentration of the NaOH spray is changed to 0% , 0.5% , and 1.0% , respectively.

TABLE 2: EXPERIMENTAL CONDITIONS AND EXHAUST GAS CONCENTRATIONS AT REACTOR INLET

Test periods	Exhaust gas volumetric flow rate (dry base) $\text{m}^3\text{N/h}$	Concentration of environmental load substances (average)			Gas temp $^\circ\text{C}$	Injected O_3 mass flow rate kg/h	Water spray flow rate for O_3 m^3/h	NaOH solution spray flow rate m^3/h	NaOH conc. for NaOH spray $\%$	Na_2SO_3 conc. for NaOH spray $\%$	Molar ratio of O_3/NO -
		NO ppm	NO_x ppm	SO_x ppm							
T1	16800	226	235	308	491	1.79	0.80	0.76	2.0	0	0.22
T2	16800	217	227	335	490	1.78	0.80	0.75	2.0	0.5	0.23
T3	16800	214	222	306	493	1.78	0.80	0.69	2.0	1.0	0.23

Fig. 3 (omitted) shows the temperature transition conditions of the exhaust gas at SP1 and SP3. The combustion characteristics of north and south regenerative furnace are different because they have been used for more than 10 years continuously. Therefore, the temperature differs in the north and south. The temperature of SP1 at the reactor inlet before the exhaust gas treatment is $490\text{--}493 \text{ }^\circ\text{C}$. The temperature of SP3 at the reactor outlet cooled by the spray of water for O_3 and NaOH aqueous solution in the reactor is $241\text{--}252 \text{ }^\circ\text{C}$. T3 has a higher outlet temperature of $252 \text{ }^\circ\text{C}$ because the amount of NaOH solution is smaller than for other conditions. The exhaust gas is completely dry.

The gas at height $H = 0 \text{ m}$ of the localized cooling area at SP2 is cooled to an average of $75 \text{ }^\circ\text{C}$. This temperature is cooled to $150 \text{ }^\circ\text{C}$ or lower using cooling water from the three-fluid nozzle. The cooling water with O_3 injected from the nozzle and the reaction field of O_3 and NO are sufficiently cooled. In the upper reaction field, the temperatures of

H = 1.5 m and 3.0 m are 91 °C and 220 °C, respectively. The calculated reaction time of the localized cooling area is 1.9 s. Therefore, conditions are set for oxidizing NO while suppressing the thermal decomposition of O₃.

Fig. 4 shows the time-dependent NO concentrations at the reactor inlet (SP1) and outlet (SP3). For glass melting furnaces, NO_x generally refers to thermal NO_x, which is NO in the combustion of a glass melting furnace. The average ratios of NO to NO_x is more than 90%. The initial NO concentrations at T1, T2, and T3 of SP1 are 226 ppm, 217 ppm, and 214 ppm, respectively. O₃ is injected into the localized cooling area formed by three-fluid nozzles. As a result, the NO at SP3 is oxidized to NO₂ and the NO concentration decreases. The molar ratio in the reaction between NO and O₃ is theoretically 1:1; it decreases with increasing gas temperature. The molar ratio of the injected ozone to the NO of exhaust gas is 0.22–0.23 in shown Table 2. The NO concentrations at T1 to T3 of SP3 are 195 ppm, 186 ppm, and 184 ppm, respectively. A NO oxidation ratio (de-NO/injected O₃) of 64% and NO removal efficiency (de-NO) of 14.5% is achieved for the condition of T2 with 1.78 kg/h of injected O₃. It was confirmed that exhaust gas forms in the localized cooling area by water and O₃ using the three-fluid nozzle and NO is oxidized as a result.

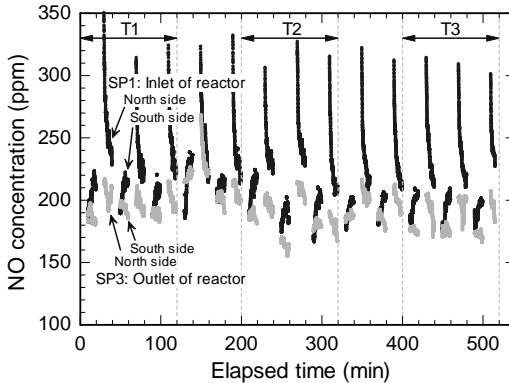


Fig. 4. Time-dependent NO concentration at the reactor inlet (SP1) and outlet (SP3)

Fig. 5 shows the temperature dependence of the ratio of decreased NO on the injected O₃ and the gas temperature at TC2. The result of our previous laboratory study [8] is also shown in this figure. The molar ratio in the oxidation reaction of NO and O₃ is equimolar (NO/O₃ = 1). When the gas temperature is less than 50 °C, de-NO/injected O₃ of more than 90% is achieved. However, the de-NO/injected O₃ gradually decreases with increasing gas temperature until high temperatures such as 300 °C. This is because most of the injected O₃ is decomposed to O₂ due to the high temperature of the gas before reaction with NO. In order to prevent decomposition of O₃, the O₃ must be injected into the localized gas cooling area as a water mist, which is formed by the three-fluid nozzle. In this pilot-scale experiment, 37.0–37.3 mol/h of O₃ is injected into the gas cooling area (height = 0 m) formed by three-fluid nozzles at a temperature of 94–98 °C. As a result, the decreased NO measures 23.7 mol/h and the de-NO/injected O₃ decreases to 64% at T2. In addition, at heights of 0 m and 1.5 m in the gas cooling area the temperature is 96 °C and 89 °C at T2, respectively, into which 37.0 mol/h of O₃ is injected. The gas temperature of the O₃ injection point is cooled in spite with less than 100 °C, NO oxidation ratio is same as at 190 °C. Note that, in this pilot-scale experiment, the actual gas temperature of the reaction zone becomes lower

than that of the exhaust gas in the formed gas cooling area with water spraying. The deterioration in NO removal ratio caused by the high temperature of the gas can be prevented by injecting O_3 into the region cooled by the spray.

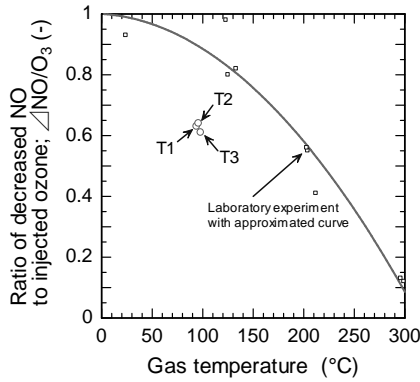


Fig. 5. Temperature dependence of the ratio of decreased NO to injected O_3 (de- NO/O_3)

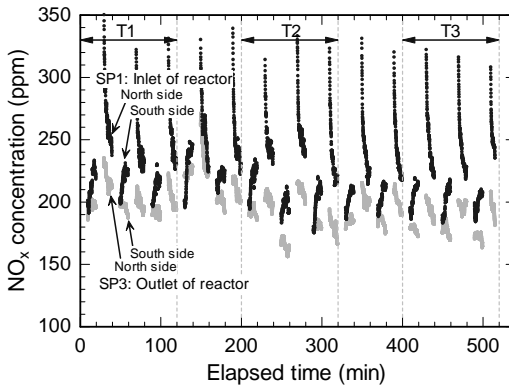


Fig. 6. Time-dependent NO_x concentration at the reactor inlet (SP1) and outlet (SP3)

Fig. 6 shows the time-dependent NO_x concentration at the reactor inlet (SP1) and outlet (SP3). The average NO_x concentrations at T1, T2, and T3 of SP1 are 235 ppm, 227 ppm, and 222 ppm, respectively. O_3 is injected into the localized cooling area formed by three-fluid spray nozzles. In addition, NaOH spray nozzles of different heights are applied to the continuous localized cooling area. SO_x in the exhaust gas is treated to become Na_2SO_3 solution using NaOH solution at the gas cooling area through the desulfurization process. As a result, the NO at SP3 is oxidized to NO_2 and the NO concentration decreases. The average NO_x concentrations at T1, T2, and T3 of SP3 are 201 ppm, 188 ppm, and 185 ppm, respectively. A NO_x removal efficiency (de- NO_x) of 14.6%, 17.0%, and 17.0% is obtained for the condition of T1–3. At this time, the obtained NO_x removal ratios (de- NO_x /injected O_3) of T1, T2, and T3 are 69%, 78%, and 77%, respectively. In addition to the NO_2 oxidized by O_3 , the NO_2 is treated together with the original NO_2 from the exhaust gas of combustion. Also, de- NO_x /injected O_3 increases as Na_2SO_3 concentration is increased. Adding Na_2SO_3 to the NaOH spray solution (in the range of 0 to 1%) can im-

prove NO_2 reduction performance. Thus, it is possible to increase de- NO_x by increasing Na_2SO_3 concentration. De- NO_x is not high because the amount of injected O_3 is insufficient for the existing NO_x (the molar ratio of $\text{O}_3/\text{NO} = 0.22\text{--}0.23$). However, as de- NO_x/O_3 is high, de- NO_x can be increased by increasing the O_3 injection amount.

V. CONCLUSION

A pilot scale experiment for de- NO_x and de- SO_x was conducted using plasma and chemical hybrid processes in an exhaust gas treatment system of a glass melting furnace. Experiments were conducted where the reaction time in the localized cooling area and the amount of NaOH and Na_2SO_3 were changed, and the temperature for de- NO_x was found to improve the performance. The main results are as follows.

(1) With regard to the NO oxidation experiment, the cooling water with O_3 injected from the nozzle and the reaction field of O_3 and NO were sufficiently cooled to 75°C . The initial NO concentrations were 214–226 ppm. O_3 was injected into the localized cooling area by three-fluid spray nozzles. In addition, the outlet of the reactor was maintained above the SO_3 dew-point. As a result, the decreased NO was 23.7 mol/h and the de- $\text{NO}/\text{injected } \text{O}_3$ of 64% was obtained. In addition, no O_3 leaks were observed at the stack because the exhaust gas temperature was higher than the thermal decomposition temperature of O_3 .

(2) The de- NO_x/O_3 was 78%; therefore, high efficiency was obtained. Then, a de- NO_x of 14.6–17.0% was obtained. De- NO_x was not high because the amount of injected O_3 was insufficient for existing NO_x , because the molar ratio of O_3/NO was 0.22–0.23. However, as de- NO_x/O_3 was high, de- NO_x could be increased by increasing the O_3 injection amount. It is necessary to investigate whether efficiencies of de- NO/O_3 and de- NO_x/O_3 can be maintained even when the O_3 injection amount is increased.

Our experiments indicate that PCHP has great potential as a new exhaust gas treatment technology. In this study, we introduced simultaneous denitrification and desulfurization technology using PCHP, which can reduce the environmental impact of exhaust gas emitted from a glass melting furnace.

VI. ACKNOWLEDGMENTS

We would like to thank Mr. H. Shokawa, a graduate student of Osaka Prefecture University, and Mr. R. Tsuji, members of Nihon Yamamura Glass Co., Ltd., for their support in carrying out the experiments.

This paper is a conference paper for the 2018 ESA Joint Conference and provides an overview of our research. A complete report will be submitted to *IEEE Transactions on Industry Applications*.

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