

# Optimum Conditions of Low Temperature SNCR by Hydrogen Injection

Yohei Tsuji<sup>1</sup>, Toko Kawaoka<sup>1</sup>, Shinji Kambara<sup>1,\*</sup>, and Megumi Masui<sup>2</sup>

<sup>1</sup>*ERES, Gifu University, 1-1 Yanagido, 501-1193, Japan*

<sup>2</sup>*Actree Co., Ltd, 375 Misumimachi, 924-0053, Japan*

*\*E-mail: kambara@gifu-u.ac.jp*

## Abstract

Selective non-catalytic reduction (SNCR) of NO<sub>x</sub> using activated ammonia generated by pulsed plasma has been developed to expand the narrow temperature window for deNO<sub>x</sub>. The temperature window enlargement of 150 °C was achieved at the lower boundary of the temperature window by activated ammonia injection. Molecular hydrogen in activated ammonia played a key role in the enlargement of the temperature window.

The purpose of the present study is to investigate optimum conditions for the low temperature SNCR. DeNO<sub>x</sub>, which were examined by using an NH<sub>3</sub>/H<sub>2</sub> gas mixture at the temperature ranges of 650–750 °C. The optimum H<sub>2</sub>/NH<sub>3</sub> molar ratio changed by the reaction temperature. The optimum value of H<sub>2</sub>/NH<sub>3</sub> molar ratio was decreased with an increase in the reaction temperature; the ranges of optimum H<sub>2</sub>/NH<sub>3</sub> ratios at NH<sub>3</sub>/NO = 1.0 were 0.5–0.2 at the temperature range of 650–750 °C. It found that injected hydrogen contributed production of OH radical, which was available for deNO<sub>x</sub> reactions at the low temperatures. The deNO<sub>x</sub> level was approximately 80% at NH<sub>3</sub>/NO = 1.0 under the optimum H<sub>2</sub>/NH<sub>3</sub> ratio at the temperature ranges of 650–750 °C.

## 1. Introduction

Selective catalytic reduction (SCR), an efficient treatment technology, has been used worldwide for NO<sub>x</sub> removal in large-scale combustors such as coal-fired power plants. In SCR systems, nitric oxide (NO) reacts with injected molecular ammonia in the presence of a catalyst and oxygen at a temperature of around 350 °C at which NO converts to molecular nitrogen and water. A drawback of SCR systems in application to incinerators and ship diesel engines is that they are particularly costly because frequent replacement of the catalyst is required owing to catalyst poisoning by sulfur dioxide, plugging and erosion by ammonium bisulphate, and deposition of ash, amongst others.

Selective non-catalytic reduction (SNCR) is a conceptually simple process that involves the injection of molecular ammonia into a furnace without using a catalyst. SNCR systems offer a promising technology because of their cost-effectiveness, although critical issues regarding their application still exist. In SNCR systems, NO<sub>x</sub> reduction occurs at a high temperature range of 850 °C–1175 °C: it is called temperature window<sup>(1)</sup>. However, a sufficiently high efficiency of NO<sub>x</sub> reduction cannot be obtained in combustors, because the gas residence time in the temperature window is very short for the deNO<sub>x</sub>. It's desirable to install the SNCR equipment to incinerators, but temperature of the flue gas is around 750 °C, therefore it is impossible to apply the incinerators.

We have been developed an original SNCR using dielectric barrier discharge<sup>(2,3)</sup>. The advantages of the SNCR system are low temperature reaction. The purpose of this research is to investigate deNO<sub>x</sub> reaction mechanisms and to determine the optimum conditions.

## 2. Methodology

Fig. 1 shows experimental setup for SNCR with hydrogen addition. Equipment consists of model gas supply system, deNO<sub>x</sub> gas supply system, preheating department, gas mixture office, reaction department, gold furnace and NO<sub>x</sub>, N<sub>2</sub>O, analysis installation. Gold furnace

can do temperature control of a preheating department and a reaction independently respectively.

The gas to which the oxygen density (8.2%), the NO density (200, 500ppm), the NH<sub>3</sub> density (160 – 500 ppm) and the general flow rate (3.0 L/min fixing) were adjusted by a mass flow controller with a gas blender was supplied from the preheating part side. H<sub>2</sub> was injected in the gas mixture part, and after mixing with model gas, a Thermal reaction has been caused in the reaction part. The temperature of the reaction part was heated from 600 °C to 750 °C and the temperature was made regular. The pressure in the system was controlled in the atmospheric pressure neighborhood (103.1 ± 0.1 kPa) by the style pressure equipment.

After removing slip ammonia by an adhesion pill during dense fog so as not to affect an analyzer, the gas treated with hydrogen addition introduced into a NO<sub>x</sub> meter and N<sub>2</sub>O meter by a gas sampler with a pump and analyzed continuously. NO<sub>x</sub> removal efficiency was asked by gauging the density of NO in the reaction part temperature 500 °C ([NO]<sub>in</sub>) and the density of NO of whole exit gas in each experimental conditions ([NO]<sub>out</sub>) with NO<sub>x</sub> total (1).

$$\text{NO removal} = ([\text{NO}]_{\text{in}} - [\text{NO}]_{\text{out}}) / [\text{NO}]_{\text{in}} \times 100 \quad (1)$$

### 3. Results

#### 3.1 Low-temperature SNCR by hydrogen addition

Fig. 2 shows characteristics of NO<sub>x</sub> removal by hydrogen addition SNCR when [NH<sub>3</sub>] / [NO] ( $M_{R1}$ ) = 1.0. NO<sub>x</sub> removal efficiency rose by addition of hydrogen. [H<sub>2</sub>] / [NO]<sub>0</sub> ( $M_{R2}$ ) = 0 shows the condition that is not the addition of hydrogen altogether the NO<sub>x</sub> removal by thermal deNO<sub>x</sub> method. NO<sub>x</sub> removal as to increase the  $M_{R2}$  is increased, it was a constant at 80% NO<sub>x</sub> removal efficiency at reaction temperature is 650 °C in about  $M_{R2}$  = 0.49, at 700 °C in about  $M_{R2}$  = 0.25, at 720 °C in about  $M_{R2}$  = 0.22. Therefore it was clear that H<sub>2</sub> promotes thermal deNO<sub>x</sub> reaction and play a role to shift the temperature window to low temperature side. At lower reaction temperature, to obtain 80% of the NO<sub>x</sub> removal was found that it is necessary to increase the value of  $M_{R2}$ .

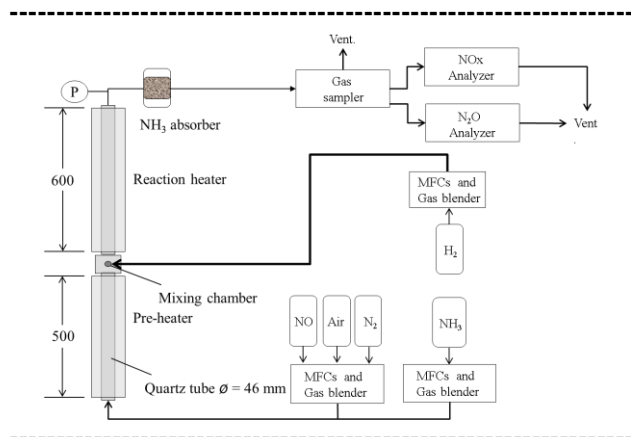


Fig. 1. Schematic diagram of experimental apparatus.

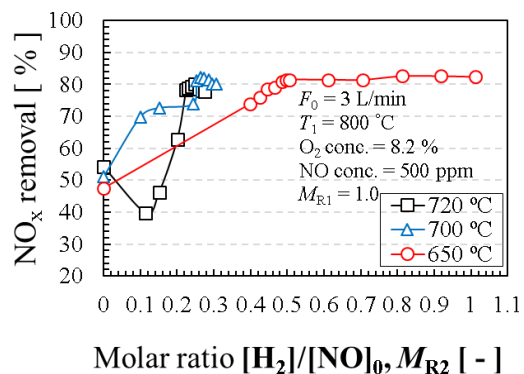


Fig. 2. Characteristics of NO<sub>x</sub> removal by hydrogen addition.

### 3.2 Optimum hydrogen conditions

From the results of 3.1, it was able to determine the optimum hydrogen concentration in each reaction temperature at several NO concentration and  $M_{R1}$ . Fig.4 plot the optimum  $M_{R2}$  value against reaction temperature when changing the  $M_{R1} = 0.8-1.0$  in  $F_0 = 3.0$  L / min,  $[NO]_0 = 500$  ppm,  $[O_2] = 8.2\%$ . In any  $M_{R1}$ , optimum  $M_{R2}$  value decreases by increasing the reaction temperature, and it was found to exhibit a sharp decrease between particular 650-700 °C. When the reaction temperature 700 °C,  $M_{R1} = 1.0$ , the optimum  $M_{R2}$  value was 0.25, and when the  $M_{R1} = 0.8$ , the optimum  $M_{R2}$  value was 1.2. Thus optimum  $M_{R2}$  value was found to be greatly influenced by  $M_{R1}$ .

Fig.4 is a diagram showing  $M_{R1}$  the effect of the initial NO concentration on the optimum  $M_{R2}$  values as parameters. When  $M_{R1}$  is constant, optimum  $M_{R2}$  were found to be tends not changed substantially value even so  $[NO]_0$  was different value. But only if the reaction temperature 650 °C and  $M_{R1} = 1.0$ , the optimum  $M_{R2}$  value is increased at the lower  $[NO]_0$  value

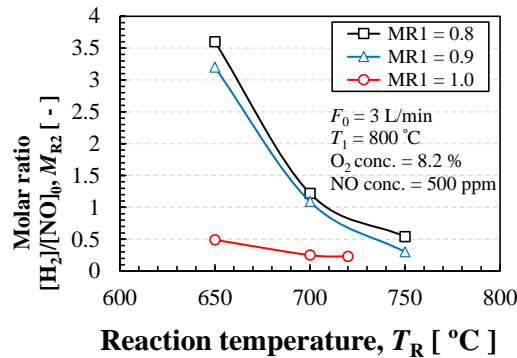


Fig. 3. Optimum  $M_{R2}$  in various reaction temperatures and  $M_{R1}$ .

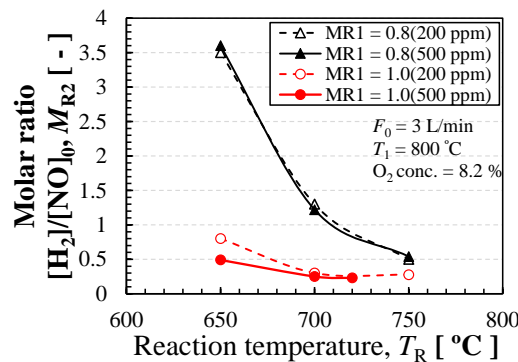


Fig. 4. Effect of temperature and  $M_{R1}$  on optimum  $M_{R2}$

### 3.3 Reaction mechanism

Fig. 5 shows the comparison of experimental and simulation results of de $NO_x$  by hydrogen injection in  $F_0 = 3.0$  L/min,  $T_1 = 800$  °C,  $NO = 500$ ppm,  $O_2 = 8.2$  %, and  $M_{R1} = 1.0$ .

NO<sub>x</sub> removal also increased as the  $M_{R2}$  increases both the results of the experimental results and simulation by CHEMKIN-PRO. Also two of the graph showed a very similar behavior.

It was thought to be able to discuss the mechanism from the simulation than this thing. Reaction rate of generation of NH<sub>2</sub> to promote the deNO<sub>x</sub> than simulation results by the injection of H<sub>2</sub> was found to be fast. This is the generation of OH is a chemical species involved in the NH<sub>2</sub> generation is considered because promoted by hydrogen injection. It compared the reaction of OH generated by thermal deNO<sub>x</sub> and hydrogen injection deNO<sub>x</sub>. Eq. (2) – (4) shows the formation reaction of OH by H<sub>2</sub> injection. Eq. (5) – (7) shows showing the generation reaction of thermal deNO<sub>x</sub>.

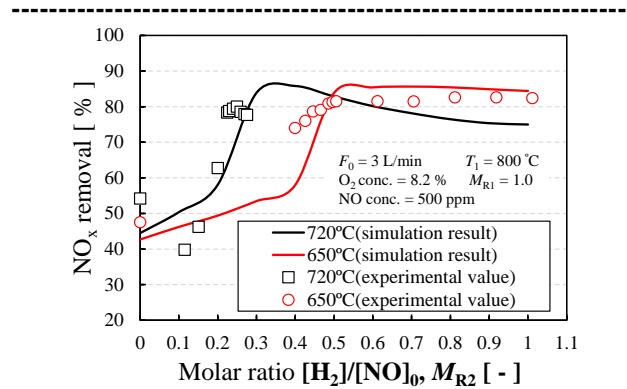
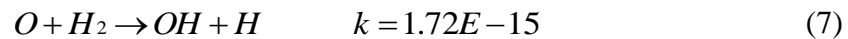
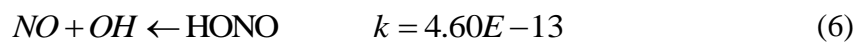
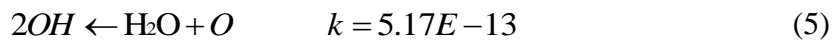
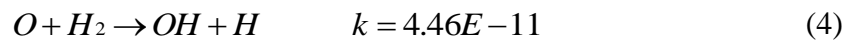
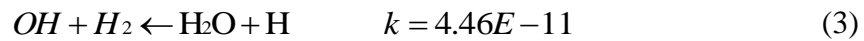
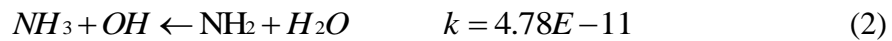


Fig. 5. Comparison of NO<sub>x</sub> removal between experimental result and simulation.

Eq. (2) – (4) shows the formation reaction of OH by H<sub>2</sub> injection. Eq. (5) – (7) shows showing the generation reaction of thermal deNO<sub>x</sub>.



Reaction rate of OH generation of hydrogen deNO<sub>x</sub> conditions is about 10<sup>2</sup>-10<sup>4</sup> faster. This is increasing the amount of H<sub>2</sub>O by hydrogen injection, occur reaction of Eq. (2), (3), think it's because the generation of OH is promoted. Since the OH generation is facilitated by the hydrogen injected from these results, it was found that the NO<sub>x</sub> removal is increased.

## 4 Conclusion

Experiments were conducted to determine the optimum conditions for the low-temperature SNCR using hydrogen, and to determine the optimum amount of hydrogen injection in  $M_{R1} = 0.8 - 1.0$ ,  $T_R = 650 - 750$  °C, NO = 200 ppm, 500 ppm. When the value of  $M_{R1}$  is constant, the optimal hydrogen addition amount decreases as the temperature increases. In the same temperature range, the optimal amount of hydrogen addition is reduced to the value of  $M_{R1}$  was high. When  $M_{R1}$  is constant, optimum  $M_{R2}$  were found to be tends not changed substantially value even so [NO]<sub>0</sub> was different value.

## References

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