



Technical Note

Effect of hydrogen generated by dielectric barrier discharge of NH₃ on selective non-catalytic reduction processYoungchul Byun^a, Kyung Bo Ko^a, Moohyun Cho^a, Won Namkung^a, Dong Nam Shin^{b,*}, Dong Jun Koh^b^aSchool of Environmental Science and Engineering, Pohang University of Science and Technology, Hyoja Dong, Pohang, Kyoungbuk 790-784, Republic of Korea^bEnvironmental Research Department, Research Institute of Industrial Science and Technology, Hyoja Dong, Pohang, Kyoungbuk 790-600, Republic of Korea

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ABSTRACT

Plasma-assisted selective non-catalytic reduction (SNCR) has been investigated to clarify which species generated by the plasma play a crucial role in NO reduction. We find that the presence of O₂ is indispensable and only H₂ is observed to be a stable product by dielectric barrier discharge (DBD) of NH₃. As the extent of NH₃ decomposition by DBD increases, the commencement temperature of SNCR processes is lowered and the working temperature window is widened. This propensity may be attributed to the chemical reaction of H₂ with O₂ to generate OH and H radicals which make it possible to yield NH₂ radicals even at low temperature.

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1. Introduction

For several decades, non-thermal plasma (NTP) processes have received much attention as a potential method to remove NO_x and SO₂ simultaneously from flue gases (Penetrante and Schultheis, 1993; Chang, 2003). When NTP technology is applied to removal of NO emitted from various anthropogenic sources, two approaches have typically been employed: (i) the oxidation of NO to NO₂, followed either by neutralization to form ammonium nitrate or by selective catalytic reduction to produce N₂, and (ii) the production of NH₂ from NH₃, followed by selective non-catalytic reduction (SNCR) through the reaction of NO with NH₂ to generate N₂. The reaction mechanism behind the oxidation of NO to NO₂ and then the successive reduction of NO₂ by a catalyst is well understood. In fact, some studies have reported successful NTP applications in pilot and commercial plants (McLarnon et al., 2000; Lee et al., 2003). In contrast to the oxidation process of NO by NTP, however, the role of NTP in the SNCR process for the removal of NO is poorly understood.

It is well-known that the SNCR process relies on the formation of NH₂ primarily via the reaction of NH₃ with OH radicals. The produced NH₂ rapidly reacts with NO even at the room temperature to produce N₂ and H₂O. However, since the formation of NH₂ radicals depends strongly on the temperature of the flue gas, it is hard to attain a high enough NH₂ concentration at temperatures below 800 °C. Therefore, there have been some trials to generate NH₂

by utilizing NTP with the expectation that the working temperature of SNCR would be lowered as compared to the conventional one. Chang and Cheng (1997) reported that as high as 75% removal efficiency of NO was achieved at room temperature. Very recently, Yukimura et al. (2007) reported that the DeNO_x occurs only above the reaction temperature of 480 °C. Both studies have employed NTP to discharge NH₃ before introducing into SNCR reactor for NO_x removal. Some studies have proposed that N₂H₄ produced by NTP is further decomposed to generate NH₂ radicals for NO_x removal (Fateev et al., 2005; Kusano et al., 2005). However, the details behind NO_x removal by plasma-assisted SNCR are still controversial because of the difference in these studies (Chang and Cheng, 1997; Fateev et al., 2005; Kusano et al., 2005; Yukimura et al., 2007). In this study, we have analyzed the stable products formed by dielectric barrier discharge (DBD) of NH₃ and we have explored NO_x removal efficiencies as a function of the reaction temperature with and without NTP of NH₃.

2. Experimental

Fig. 1 shows the schematic diagram of the experimental system which consists of both DBD and SNCR reactors. The SNCR reactor is comprised of two quartz glass tubes (outer diameter: 28 mm, inner diameter: 25 mm, length: 450 mm) set in two tube furnaces. One glass tube is used to heat up the inlet gases to the reaction temperature and the other is used as the reaction chamber. The individual volume of both reactors is 1.10 × 10⁵ mm³. The reformation of NH₃ was carried out using the DBD reactor and then were introduced into the SNCR reactor. The DBD reactor has a quartz glass tube

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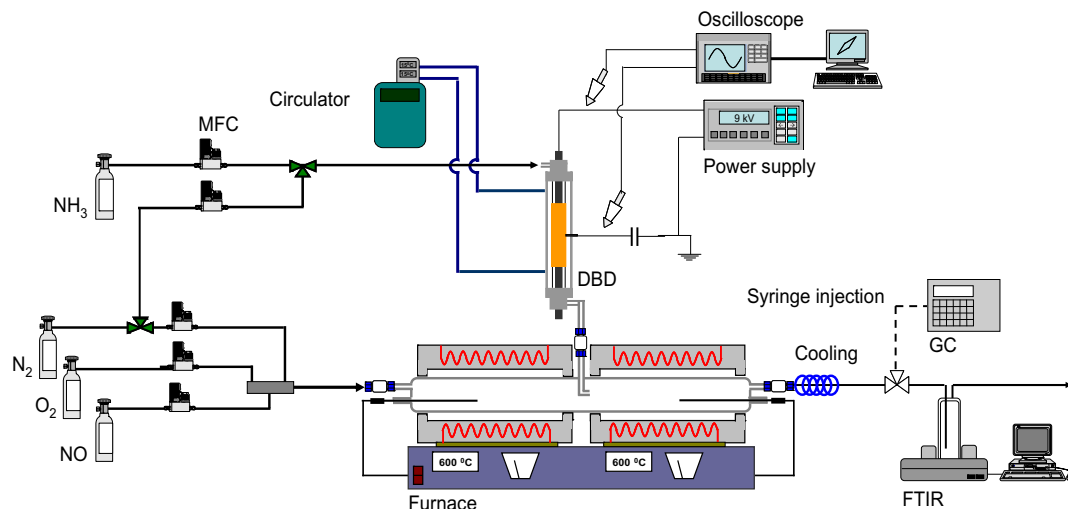


Fig. 1. Schematic diagram of experimental system for the plasma-assisted SNCR.

(outer diameter: 19 mm, inner diameter: 16 mm) with two electrodes. The inner electrode is a stainless steel rod (diameter: 13 mm) placed along the center of glass tube. The outer electrode is Cu tape (length: 100 mm) wrapped around the tube. The DBD reactor was immersed in a pool of cooling water circulated by an electrical chiller (MC-11, Jeio Tech) at a temperature of 15 °C. The high voltage of about 15 kV (peak-to-peak value) ranged from 10 to 18 kHz and was supplied using a high voltage power supply (AP-2 kW, Dawon Sys Co.). Energy introduced to the DBD reactor was measured by a Q-V Lissajou diagram using 1 μ F capacitor and voltage probes (Tektronix P6139A and P6015A) connected to the oscilloscope (Tektronix TDS 620B). The consumed power is determined by integrating the area of Q-V plot and the specific energy density deposited into the plasma reactor is calculated as:

$$\text{Input energy density (J L}^{-1}\text{)} = \frac{\text{Consumed power (J s}^{-1}\text{)}}{\text{Gas flow rate (L min}^{-1}\text{)}} \times 60 \text{ (s min}^{-1}\text{)} \quad (1)$$

The temperature used in Eq. (1) was 15 °C and all the experiments were conducted at atmospheric pressure. Flow rates of NO and NH₃ in N₂ were set to 6.5 and 1.5 L min⁻¹, respectively. The flow rates were controlled by a mass flow controller (MFC; Brooks 5850E) connected to the MFC control box (Brooks Instrument Model 0154E). The residence time of the gases within the SNCR reactor was varied from 0.21 to 0.50 s according to the reaction temperature. The NH₃ concentration passing through the DBD reactor was varied from 1% to 2% and the residence time of NH₃ gases is calculated to be 0.27 s. In all experiments, O₂ and NO concentrations in N₂ were maintained at 10% and 160 ppm, respectively. NO (2000 ppm, N₂ balance), N₂ (99.99%) and O₂ (99.99%) were obtained from BOC GASES. In this study, the NO_x (mainly NO and NO₂) and NH₃ concentration were monitored using a Fourier transform infrared spectrometer (FTIR; ABB Bomem) having a spectral resolution of 0.5 cm⁻¹. The H₂ concentration was quantified by gas chromatography (GC; HP 5890) equipped with a thermal conductivity detector.

3. Results and discussion

Fig. 2 shows the profiles of NO_x concentrations obtained at four different input energy densities as a function of the reaction temperature within the SNCR reactor. The initial NO_x concentration was set to 160 ppm and the input energy density was varied from 0 to 4.7 kJ L⁻¹. It is very important to note that the presence of O₂ in

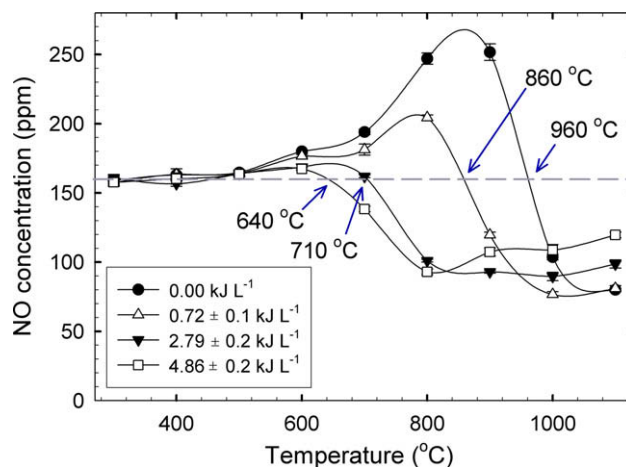


Fig. 2. Effect of NTP on change of NO concentrations obtained at four different input energy densities as a function of the temperature of SNCR reactor. Experimental conditions: total flow rate = 8 L min⁻¹, O₂ concentration = 10%, flow rate of NH₃ in N₂ DBD = 1.5 L min⁻¹, NH₃ concentration = 1.33%, N₂ balance.

the gas mixture is a prerequisite for the plasma-assisted SNCR process (Yukimura et al., 2007). Without O₂, we found no appreciable reduction in NO_x concentrations. In this study, we put a long Teflon tube intentionally between the DBD and SNCR reactors in order to lengthen the residence time of the reformed NH₃ gas stream to 0.50 s. This eliminates the possibility that the electronically excited molecules and radicals generated by DBD of NH₃ take part in SNCR reactions. However, the removal efficiency of NO_x was found to be independent of the length of Teflon tube in this study. This may be due to such transient species having a very short life time, at most, below several hundred micro seconds (Li and Liu, 2000; Fateev et al., 2005; Kusano et al., 2005). It is evident that the partial decomposition of NH₃ by using DBD, before being introduced to the SNCR reactor, leads to a shift in the working temperature to lower values as well as widening the working temperature window. Introducing NH₃ without using DBD, there is an increase in NO_x concentration between 700 and 900 °C with a maximum at 850 °C due to direct NH₃ oxidation in the presence of O₂. The extent of NH₃ oxidation declines with increasing the input energy density and disappears completely over 2.7 kJ L⁻¹. These results suggest that DBD of NH₃ leads to the formation of some species controlling the working temperature window of SNCR.

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