Low temperature SNCR by photochemical activation of ammonia

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Abstract

To broaden and lower the temperature window of the selective non catalytic reduction (SNCR) of nitric oxide (NO), the use of activated ammonia was examined. A wavelength of 172 nm was employed as the excitation source for molecular ammonia. Activated ammonia was injected into a model flue gas ($NO/O_2/N_2$) at room temperature. The effects of reaction temperatures, oxygen concentrations, and NH_3/NO molar ratios on NO removal were investigated in a lab-scale plug flow reactor. Reaction temperatures ranged from 500 °C to 850 °C. A temperature window enlargement of 150 °C was achieved at the lower boundary of the temperature window. Above 600 °C, NO removal was effected by injection of activated ammonia, while around 750 °C, conventional SNCR by injection of molecular ammonia was effective. An approximate 80% NO removal was attained at 700 °C with an MR = 2.0 and 8.3% O_2. The formation of nitrous oxide (N_2O) using activated ammonia SNCR technology was also investigated and was found to be strongly affected by O_2 concentrations, while the concentration of N_2O increased with an increase in NO removal.

1. Introduction

Selective non catalytic reduction (SNCR) techniques are a conceptually simple process that involves injecting molecular ammonia into the furnace without using a catalyst [1]. SNCR systems seem to be a promising technology because of their cost-effectiveness, although critical issues regarding their application still exist.

In SNCR systems, NO_x reduction occur at temperatures between 850 °C and 1175 °C (temperature window), however, high enough NOx reduction was not obtain in large-scale combustors [2]. To improve NOx reduction efficiency, the expansion of the temperature window is desired.

It has been reported that chemical additives together with molecular ammonia can lower and widen the temperature window. Various additives have been studied, including hydrogen, hydrogen peroxide, hydrocarbons, and carbon monoxide [3], all of which are effective. However, utilization of chemical additives increases the cost

of NO reduction. A recognized research goal is to expand the temperature window without the need for additives. The aim of the research presented in this study was to find an alternative method of producing effective chemical species for NO removal without the use of argon gas.

2. Experimental section

The experimental setup is shown in Fig. 1. The apparatus consists of two gold furnaces with quartz tubes, the gas mixing and flow control systems, the photochemical reactor, and the gas analyzers. The quartz tubes were connected via the mixing chamber.

An $NO/O_2/N_2$ gas mixture was prepared as the model flue gas, and fed into the pre-heater quartz tube. Ammonia gas diluted with nitrogen was used as the NO removal agent, which was fed into the photochemical reactor at room temperature. Molecular ammonia is excited by photons emitted from the excimer lamp in the photochemical reactor. In this paper, the chemical species generated by VUV radiation will be termed “activated ammonia”. Activated ammonia was introduced into the mixing chamber.

The reaction temperature was varied from 500 °C to 850 °C. The total gas flow rate of the $NO/O_2/NH_3/N_2$ gas mixture was fixed at 3.0 L/min for all experimental conditions. The gas composition of the output stream was continuously measured by gas analyzers for NO_x, O_2, and N_2O.

Fig. 2 depicts the configuration of the photochemical reactor. The excimer lamp (USHIO Inc.) was placed on top of the center of the power unit. An aluminum cylindrical chamber coaxial in configuration to the excimer lamp was fitted around the excimer lamp. An NH_3/N_2 gas mixture was fed into the gap between the excimer lamp and the inside wall of the cylindrical chamber. The radiation power of the VUV ray was 26 mW/cm^2 on the quartz glass surface of the excimer lamp.

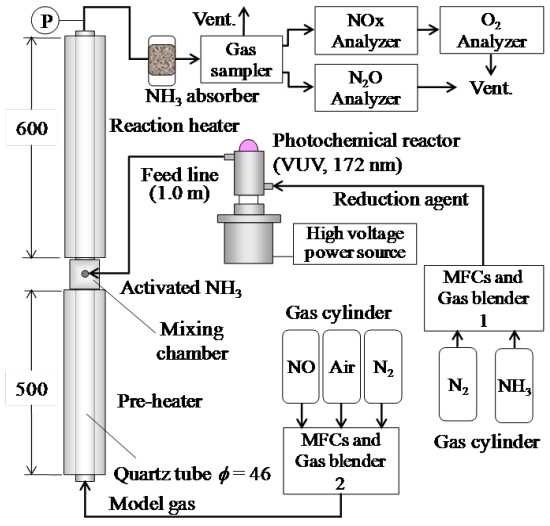

Fig. 1. Experimental setup.

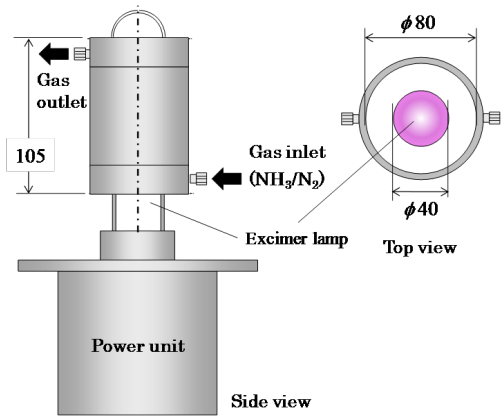

Fig. 2. Configuration of the photochemical reactor.

3. Results and Discussion

3.1. Effect of reaction temperatures

The relationship between reaction temperatures and NO removal in both SNCR systems is presented in Fig. 3. In conventional SNCR system, it can be observed that an NO removal of 3–4% was effected at 750 °C and increased significantly above 800 °C for both molar ratios of 1.0 and 1.5. In the activated ammonia SNCR, slight NO removal began at the reaction temperature of 600 °C and increased almost proportionally with a further increase in the reaction temperature. It clearly indicates that the injection of activated ammonia broadened the temperature window and lowered its starting temperature. The temperature shift was 150 °C at an NO removal of 20%. This result suggests that effective chemical species for NO removal were formed from activated ammonia generated by VUV radiation at the reaction temperature above 600 °C.

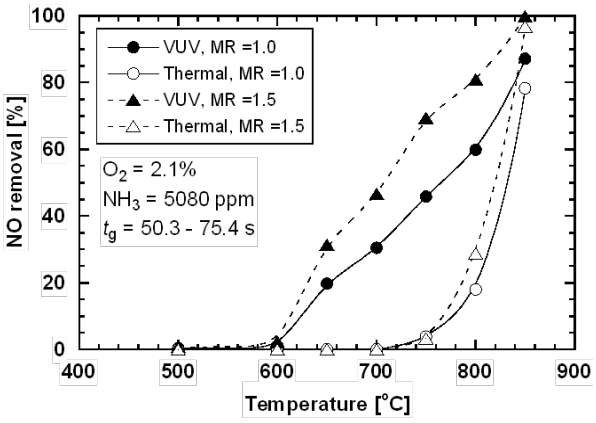

Fig. 3. NO removal performances of conventional SNCR (Thermal) and activated ammonia SNCR.

3.2. Effect of molar ratios and oxygen concentrations

Fig. 4 shows the variation in NO removal with the variation of NH_3/NO molar ratios in the range of 1.0–3.5 at 700 °C. With the activated ammonia SNCR, NO removal proportionally increased with increasing the molar ratios up to *MR* = 2.0, beyond which a gradual increase in NO removal was observed. With conventional SNCR, because the reaction temperature of 700 °C was outside the temperature window, slight NO removal was observed even at high molar ratios. Approximately 80% NO removal was obtained at *MR* = 2.0 at 700 °C using the activated ammonia SNCR technology.

The effect of the oxygen concentration on NO removal in the *MR* range of 1.0 to 2.0 is also shown in Fig. 4. A slight increase of about 5% NO removal was observed at 8.3% O_2 compared to that at 2.1% O_2 for the activated ammonia technology. With the conventional SNCR, the O_2 concentration had a weak effect on NO removal, although NO removal increased monotonically with O_2 concentration. This suggested that the reaction mechanisms for NO removal in the activated ammonia SNCR was caused by similar elemental reaction pathways to the conventional SNCR, although the reaction temperatures differed for both SNCR systems.

3.3. N_2O formation

In conventional SNCR systems, N_2O is usually generated as a byproduct during the

reactions that effect NO removal. Fig. 5 shows the N_2O formation with the activated ammonia SNCR as a function of NO removal with the different oxygen concentrations and NH_3/NO molar ratios. It can be seen that N_2O concentrations increased with increasing NO removal for all experimental conditions. The increase in the O_2 concentration significantly promoted N_2O formation, while the molar ratio did not have much effect on N_2O formation. The maximum N_2O concentration of 17 ppm was detected at 80% NO removal with 8.3% O_2 and MR = 1.5.

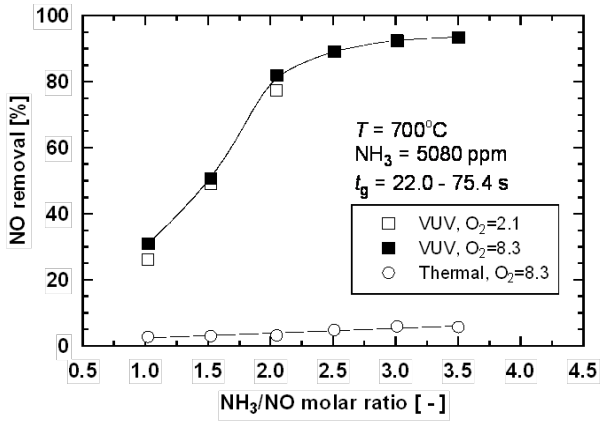

Fig. 4. Variation in NO removal with NH_3/NO molar ratios in activated ammonia SNCR and conventional SNCR at temperature of 700 °C.

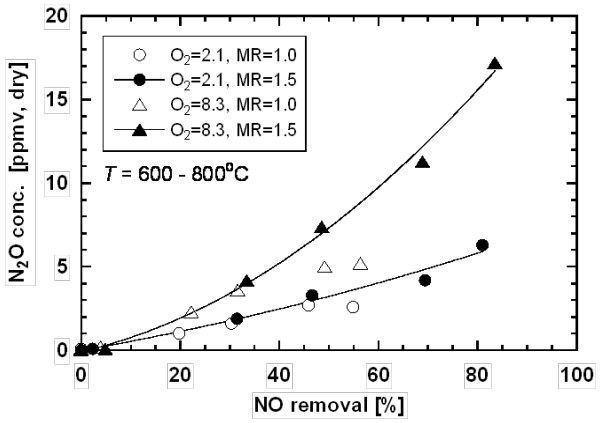

Fig. 5. N_2O formation with activated ammonia SNCR as a function of NO removal with different O_2 levels and NH_3/NO molar ratios.

4. Conclusions

With the activated ammonia SNCR, NO removal began at a reaction temperature of 600 °C, and NO removal almost proportionally increased with a further increase in reaction temperature. Approximately 80% NO removal was obtained at a molar ratio of 2.0 at 700 °C. Oxygen concentrations had little influence on NO removal: an increase of about 5% NO removal was observed with an increase from 2.1% to 8.3% O_2. However, an increase in the oxygen concentration caused the formation of N_2O. The molar ratios had a large impact on NO removal.

Acknowledgment

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