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The influence of mixing between NH_3 and NO for a De-NOx reaction in the SNCR process

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Abstract

The objective of this study was to determine the mixing effect on De-NOx in the selective non-catalytic reduction (SNCR) process using NH_3 as a reductant. A bench scale reactor set-up was used to perform experimental investigations on the mixing in the SNCR process, using an injection of NH_3, with or without an air injection, into the bulk gas. Mixing between NH_3 and NO is a very important parameter, as are kinetic parameters, such as the NSR and reaction temperature. The type of NH_3 with NO mixing method caused different De-NOx results, in which for the case of NH_3 injected with air was more effective for NO reduction than under NH_3 premixing conditions. For the case of air injection conditions, the NO reduction efficiency increased with increasing NSR, initial NO concentration, and retention time. The temperature for the maximum NO removal efficiency was 900 °C with ammonia premixing conditions, but was around 800 °C when ammonia was premixed and air was injected at the same time. That is, the optimization of the reductant mixing and injection conditions are very useful for improving the NO removal efficiency, as well as lowering the reaction temperature to that were optimal efficiency is observed. The optimum momentum ratio (J), which refers to the mixing characteristics, was found to be about 100 in this study.

Keywords: NOx removal; SNCR; Mixing

1. Introduction

Continuous increases in the generation of waste and energy consumption have been caused by rapid industrialization and economic growth in Korea. In the process of incineration for the treatment of wastes, fossil fuels are combusted to obtain energy, with the emission of many pollutants (SOx, NOx, CO) into the atmospheric environment. Recently, both the increasing deterioration of air quality and stringent emission limits in most countries require significant reductions of nitrogen oxides (NOx) emissions from various industrial boilers and incineration plants. The two most important oxides of nitrogen, with regard to air pollution, are nitric oxide (NO) and nitrogen dioxide (NO_2), jointly referred to as NOx. These gases play important roles in the environment through acidification, forest damage, smog formation, damage to human health, depletion of the stratospheric ozone layer, and the greenhouse effect.

NOx control technologies can be broadly divided into ‘‘Pre-combustion’’, ‘‘Combustion modification’’ and ‘‘Post-combustion’’, which refer to the use of low nitrogen fuels, modifying design and operating features of the combustion unit, and flue gas treatment after the combustion process, respectively [1]. Of these technologies, post-combustion NOx control techniques have been reported as an effective way to meet stringent NOx emission regulations. Two major post-combustion NOx control techniques are selective catalytic reduction (SCR) and selective non-catalytic reduction (SNCR), which are widely used in the large combustion units, such as various boilers, refineries, and waste incinerators.

SCR is most widely used for the control of NOx emissions from stationary sources, due to its efficiency and selectivity. This process involves the reduction of the NOx in flue gas by

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the injection of NH_3 into the catalyst bed at a temperature of 350 °C. However, it has several disadvantages, including the high capital investment cost, higher operating cost, limited catalyst life, catalyst poisoning by particulate matter or SO_2, higher oxygen contents in stationary sources, large space requirement for installation, reheating of the exhaust gas to maintain the optimum reaction temperature, and the need for higher upstream pressure to enable the exhaust gas to flow through the catalyst bed [1–3]. As SNCR has minimized all the problems of SCR, it is now considered a viable alternative.

SNCR is a useful method for the control of NOx emissions by the injection of reducing agents, such as ammonia, urea, and cyanuric acid into flue gases [4,5]. This is a relatively simple process for the reduction of NOx to nitrogen and water at relatively higher temperature ranges of 850–1100 °C. The performance of the SNCR process is influenced by several parameters; including, (i) the flue gas temperature within the reducing agent injection zone [6], (ii) the oxygen content in the flue gas [7], (iii) additives [8], (iv) the NH_3/NOx molar ratio [9], (v) the residence time of the flue gas in the relevant temperature range [10], (vi) the initial NOx concentration [7,10], (vii) the water content of the flue gas [6], and (viii) the mixing conditions [11,12]. Since the study of Lyon [4], there have been many investigations on the effect of parameters on the performance of SNCR in laboratory, pilot, and full-scale plants, but these have mostly focused on the kinetic parameters. Despite the mixing conditions, known as engineering parameters, being essential to obtain the proper performance of SNCR, few studies have reported these facets.

The objective of this experimental study was to investigate the influence of the mixing conditions on the NOx removal efficiency in the SNCR process, using an injection of ammonia, in a lab-scale reactor.

2. Theory

2.1. SNCR process with ammonia injection

The principles of NOx removal by the SNCR method is based on the thermal conversion of nitrogen oxides by the injection of a reducing reagent (NH_3) into the flue gas stream at an appropriate temperature ranges of 850–1100 °C. Many studies have reported the removal of NOx on the basis of the kinetic mechanism of chain reactions. The mechanism of NOx removal by the SNCR technique can be explained by combining Eqs. (1)–(8) listed below [1,6,13–15].

$$NH_3 + OH \leftrightarrow NH_2 + H_2O \tag{1}$$

$$NH_3 + O \leftrightarrow NH_2 + OH \text{ (in the absence of water)} \tag{2}$$

$$O + H_2O \leftrightarrow OH + OH \text{ (in the presence of water)} \tag{3}$$

$$NH_2 + NO \leftrightarrow N_2 + H_2O \tag{4}$$

$$NH_2 + NO \leftrightarrow NNH + OH \tag{5}$$

$$NNH + NO \leftrightarrow N_2 + HNO \tag{6}$$

$$HNO + M \leftrightarrow H + NO + M \tag{7}$$

$$H + O_2 \leftrightarrow OH + O \tag{8}$$

The chain reaction mechanism of the NOx removal process using ammonia is described by Dean et al. [13].

2.2. Mixing in the reaction zone

Mixing between bulk gas containing NOx and the fluid injected into the bulk gas is greatly influenced by the velocity of the bulk gas, the injection velocity and the direction of the injected fluid. The mixing characteristics caused by the injected fluid depend on the momentum ratio (J), as given in Eq. (9), where the properties with the subscript j refer to the injection fluid, and those with the subscript o refer to the bulk gas flow [16].

$$J = \left(\frac{\rho_j}{\rho_o}\right)\left(\frac{U_j^2}{U_o^2}\right) \tag{9}$$

3. Experiments and methods

Experiments were performed in a laboratory scale reactor (120-mm inner diameter and 700-mm height), which was able to be operated at temperature up to 1100 °C, as shown in Fig. 1. The reactor temperature was controlled by an electric heater, designed to maintain the reaction temperature at the desired value, which was measured by three K-type thermocouples located inside the reactor. The simulated bulk gas and injected air were taken from gas cylinders and a compressor, with their individual flow rates measured and regulated via mass flow controllers (MFC). Nitrogen (N_2) and nitric oxide (NO), with or without ammonia (NH_3), were mixed and introduced into a pre-heater prior to entering the reactor. A perforated gas distributor (45 holes, diameter 1-mm and interval 2-mm) was placed inside the reactor to distribute the bulk gas. The initial concentration of NO was varied within the ranges of 100–500 ppm, and that of oxygen at either 4 or 21 vol.%. The normalized stoichiometric ratio (NSR) of the reducing agents ($[NH_3]/[NO]$) was varied from 1 to 3. The concentration of NO was measured using an ND-IR type NOx analyzer (Model: 42C, USA). The experimental parameters and conditions are listed in Table 1.

4. Results and discussion

4.1. Characterization of the reactor and basic experiment

Fig. 2 shows the formation of NO according to the temperature when 300–1200 ppm (NSR = 1–4) NH_3 was injected into the bulk gas where the NO concentration was 0 ppm. The formation of NO increases in proportion to the amount of NH_3 injected and the rise in temperature, but the difference was only small at low temperatures. When the NH_3 concentration was 1200 ppm, the production of NO sharply increased from 900 °C, with 22 ppm NO produced at 1000 °C.