

Application of selective non-catalytic reduction of NO_x in small-scale combustion systems

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Abstract

The selective non-catalytic reduction of NO_x has been studied experimentally employing commercial grade urea in a pilot-scale diesel-fired tunnel furnace at 3–4% excess oxygen level and with low ppm of baseline NO_x ranging from 65 to 75 ppm within the investigated temperature range. The furnace simulated the thermal behavior of small-scale combustion systems such as small capacity boilers, water heaters, oil heaters, etc., where the operating temperatures remain within the range of 950–1300 K. NO_x reductions were studied with the variation of injection temperature, residence time, normalized stoichiometric ratio (NSR) of the reagent, etc. With 5% urea solution, at an NSR of 4, as much as 54% reduction was achieved at 1128 K. The result is quite significant, especially for the investigated level of baseline NO_x . The ammonia slip measurements showed that the slip was below 16 ppm at NSR of 4 and optimum temperature of NO_x reduction. Finally, the investigations have demonstrated that selective non-catalytic reduction process is quite applicable to the small-scale combustion applications.

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1. Introduction

Increasing deterioration of atmospheric environment has led to strict regulations to curb the emissions of nitrogen oxides, which consequently stimulated a significant number of researches to derive some effective NO_x abatement technologies. Post-combustion NO_x abatement technologies have been demonstrated to be effective to cope with the stringent NO_x emission standard. Selective catalytic reduction (SCR) and

selective non-catalytic reduction (SNCR) are two major post-combustion technologies, which are being massively used to the large combustion systems such as power plant boilers, industrial boilers, refineries and waste incinerators (Rentz et al., 1996).

Despite the fact that SCR has higher NO_x reduction efficiency, it has several disadvantages as well, which include high capital investment cost, higher operating cost than most other options, limited catalyst life, catalyst poisoning, large space requirement to install and required higher upstream pressure to enable the exhaust gas flow through the catalyst (Caton et al., 1995). In contrast, SNCR has minimized all the problems of SCR. Moreover, it can be used in dirty

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and fouling services (particulates and/or high sulfur) and it is easier to retrofit. As it requires little capital cost and is easy to retrofit, it is best suited to developing countries. Recently, SNCR has been adopted in different industries in South Korea, China, Taiwan and the Czech Republic (Radojevic, 1998).

So many researches based on urea SNCR have already been conducted by different researchers, which demonstrate that NO_x reduction performance and effective temperature window vary based on the geometry of combustion chamber, geometry and performance of the atomizer and types of fuels used (Mansour et al., 1987; Abele et al., 1991; Nylander et al., 1989). Most of the researches were related to the coal and gas burning exhaust and especially with high initial ppm of NO_x . As far as diesel exhaust is concerned, very few documents have been documented using urea SNCR. As most of the small-scale combustion facilities still use the diesel fuel, to fill up the large gap, further studies are warranted in this area. As for low value of baseline NO_x , Teixeira and Muzio (1991) observed that below 125 ppm of baseline NO_x , the performance of NO_x reduction was very insignificant. In their studies, they used a pilot scale combustor and natural gas as fuel. Strechsbier (1998) injected urea in to gas turbine exhaust containing baseline NO_x of 120 ppm. He achieved a very insignificant reduction and in some cases the reduction was inhibited. So, further studies are required with low initial value of NO_x employing different types of fuels' exhaust to get the distinct idea about the performance of urea SNCR as for low ppm of baseline NO_x .

In these perspectives, the present study is aimed to investigate the NO_x reduction characteristics of com-

mercial grade urea in reducing NO_x from diesel burning exhaust, that is containing low ppm of baseline NO_x .

2. Experimental setup

An industrial diesel Burner Riello 40-G10 of capacity of 120 kW was used as a combustion source. A combustion chamber of 390 mm OD and 1765 mm in length was fabricated (Fig. 1). The material was 2.5 mm stainless steel sheet. To reduce the heat transfer in order to get high temperature inside as well as to protect the combustion chamber wall being overheated, 50 mm refractory lining was used. For inserting temperature probe and emission probe, a series of temperature tappings and emission probe were made. The reactor was 815 mm long. The temperature drop along the length of the reactor was about 165–230 K/s. A dual fluid internal mix injector, manufactured by Spraying Systems Co. USA, was used for injecting aqueous urea solution. The injector was mounted on the combustion chamber wall through a hole in such a way that the nozzle tip goes 11 mm inside. The nozzle tip and the body of the injector were made of hastelloy to sustain the high temperatures of the burner. The spray was round type with an angle of 70° . The atomizer was able to produce the droplets in the range of $20 \mu\text{m}$ and above. The compressed air was used as atomizing air. To maintain the sequence of air and urea flow, solenoid valves and timer relays were used. To maintain the constant pressure in air line and urea line, two individual pressure regulators were used. Two flow meters and pressure gauges were used to measure the flow rate and

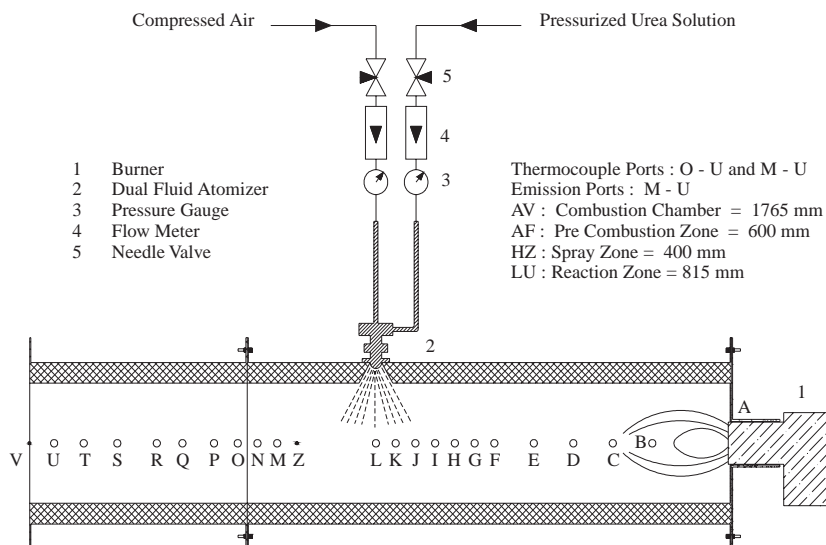


Fig. 1. Layout of the experimental setup.

pressure of urea and air line. To achieve effective droplet sizes with good penetration into the flue gas, the pressure and the flow rate of the atomizing air and the urea were adjusted.

3. Experimental procedure

The burner was operated at 3–4 % of excess oxygen, while the baseline NO_x varied from about 65 to 75 ppm within the operating temperatures, which were in the range of 973 to 1323 K. To study the effect of the variation of injection temperatures on the performance of NO_x reduction, for a particular value of normalized stoichiometric ratio (NSR), the injection temperatures were varied within the investigated temperature range. The pressure of the injection was kept constant during the whole experiment. Before introducing the reagents, the baseline NO_x was noted at different temperatures within the operating temperature range and afterward, while the reagent was injected, the NO_x was measured at the same temperatures. Comparing the two data, NO_x reduction was obtained. To study the effect of the normalized stoichiometric ratio on the NO_x reduction performance, at a certain injection temperature and the different values of NSR the NO_x emission data were taken at point U of the reaction chamber (Figure 1). The NSR was varied in the range of 1–4 by changing the reagent flow rate at a constant pressure of the atomizing air. To investigate the effect of residence time on NO_x reduction, at a particular injection temperature, the emission data were taken at nine different points on the reactor. NO_x emissions during the whole experiment were measured by a chemiluminescent analyzer. The ammonia slip during the investigations was measured using the standard colorimetric method. Firstly, the ammonia slip was measured at the point U of the reactor at different injection temperatures for a particular value of NSR and finally to study the effect of NSR on ammonia slip, at a particular injection temperature the urea solution was injected into the flue gas at varying NSR and the corresponding ammonia slip was recorded.

4. Results and discussion

Fig. 2 shows the variation of NO_x reduction with injection temperature. For all values of NSR, NO_x reduction increased with moderate rate with increasing injection temperature. After a certain value of the injection temperature the reduction decayed very sharply with further increase in temperature. The above NO_x reduction trend with the variation of injection temperatures is in close agreement with the findings reported by some prominent researchers (Jodal et al., 1989; Jones et al., 1989; Sun et al., 1990; Irfan, 1996; Zamansky et

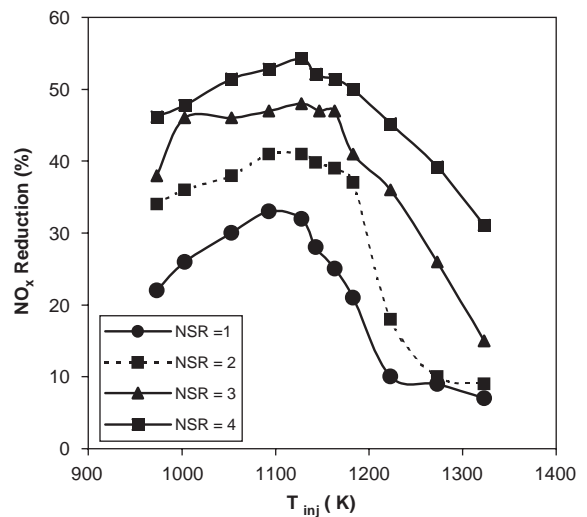


Fig. 2. Effect of injection temperature and NSR on NO_x reduction.

al., 1999). The effective temperature window in their studies was in the range of 1023–1473 K, while the width of temperature window varied from 100 to 300 K. However, in the present study significant reduction was obtained at lower temperatures, i.e., below 1023 K, which indicates that the temperature window has been shifted toward lower temperatures.

In Fig. 2, for an NSR of 1 a maximum reduction of 33% was achieved and it reached as much as 54%, while NSR varied from 1 to 4. One important feature is that with the variation of NSR, the peak temperature of NO_x reduction remained almost unaffected and it became centered around 1128 K. The reduction obtained in the present study is quite significant for the investigated level of baseline NO_x . Fig. 3 compares the reduction of the NO_x concentration among all the four values of NSR with reference to the baseline NO_x . The phenomena related to the improvement of NO_x reduction and widening of effective temperature window in the present studies might be due to the causes as described below.

Firstly, it might have happened due to the use of diesel fuel. The composition of the exhaust gas is very likely to vary with the variation of the fuel used. The exhaust of diesel fuel has contained some organic additives such as unburned hydrocarbon, which might have played an active role as additive to shift the temperature window toward lower temperature (Teixeira and Muzio, 1991; Irfan, 1996; Bowers, 1988). Secondly, the impurities of commercial grade urea might have played the role as additive and contributed to such change. Fig. 2 also shows that the width of the effective temperature window widened with the increasing value of NSR. Between NSR of 2 and 3, for a reduction efficiency of 41%, the effective temperature range was increased by

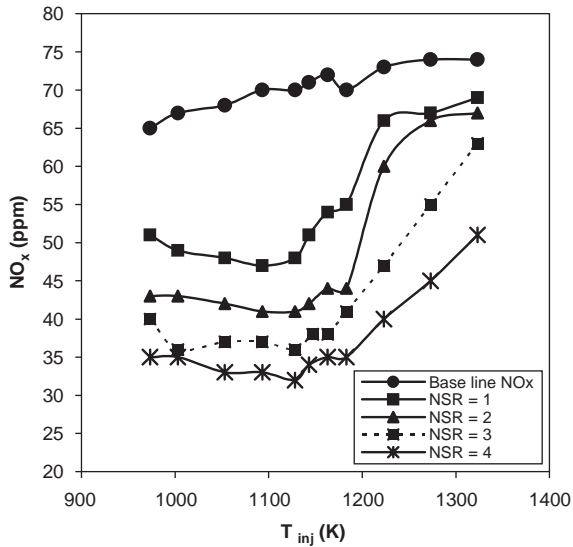


Fig. 3. Effect of injection temperature and NSR on final NO_x concentration.

155 K. This phenomenon of widening of temperature window with the increase in molar ratio (no. of mole of urea/ no. of mole of nitric oxide) is in agreement with the findings of some previous researches (Irfan, 1996; Wenli et al., 1989).

Fig. 4 shows the NO_x reduction as a function of NSR at two different injection temperatures. For optimum temperature, i.e., at 1128 K, the NO_x reduction rose moderately with the increase in NSR, while at 1273 K, which is higher than the optimum value, the rise was not smooth. Between NSR 2 and 3 the improvement was quite dramatic and it was about 26%, whereas at 1128 K, between those two points the improvement was only about 7%. Another significant aspect exhibited in the figure is that with increasing NSR, the slope of the curve tended to be flat which demonstrates that after a certain value of NSR, further increase in NSR influences little on NO_x reduction. This phenomenon is in accordance with the findings of the prior studies reported by Nylander et al. (1989).

Fig. 5 presents the NO_x reduction as a function of residence time. NO_x reduction increased with increases in residence time. For 1173 K case, the reaction rate is higher between the residence time of 262 and 355 ms as in this zone the average reaction temperature was close to the optimum temperature of 1128 K. On the contrary, in 1093 K case, the rate of NO_x reduction is faster up to a residence time of 262 ms, i.e., in the first part of the reactor, as in this case within this part of the reactor temperatures are close to the optimum temperature. For both cases, after a certain value of residence time, with the increase in residence time the rate of NO_x reduction decreased and the profiles gradually straightened up.

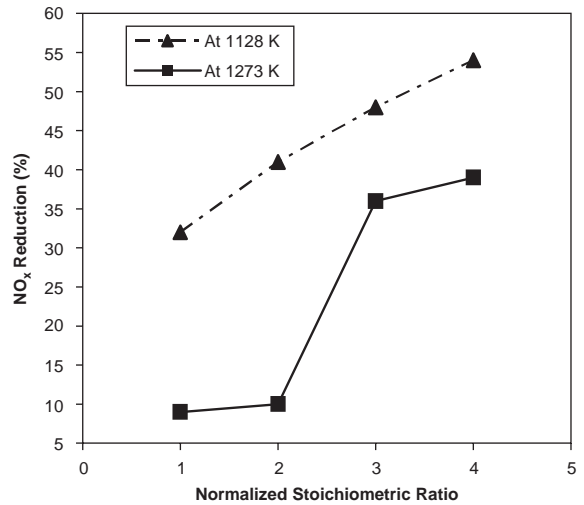


Fig. 4. Effect of NSR on NO_x reduction for 5% urea solution.

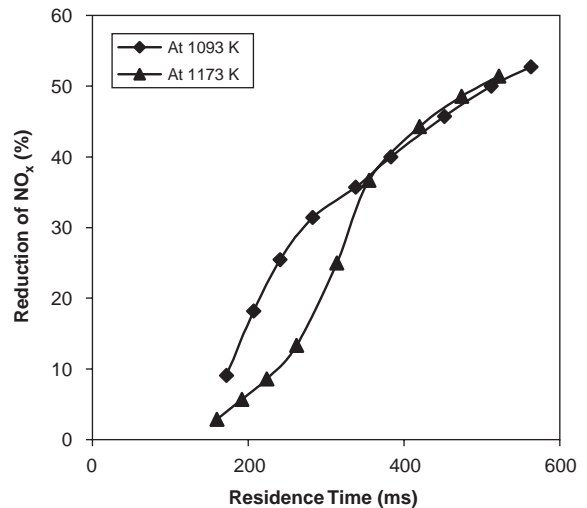


Fig. 5. Effect of residence time on NO_x reduction at NSR of 4.

This is due to the fact that in the present study the reactor temperature was not maintained constant along the length of the reactor. So, with the increase in the residence time the reaction temperature reduced, which caused gradual reduction in reaction rate within the investigated range of the residence time.

Apart from the low-temperature effect, this rate might decrease due to low reagent flue gas ratio. As the residence time increases the amount of active reagent to flue gas ratio decreases along the downstream of the reactor, which may cause poor reaction performance. This trend of reaction profile suggests that, based on the operating conditions, after a certain length, the combustion chamber has no effect on reaction performance.

This phenomenon is in accordance with the findings of the prior studies (Jones et al., 1989; Wenli et al., 1989).

Fig. 6 shows the ammonia slip as a function of injection temperature of the urea. The test was conducted with 5% urea solution at an NSR of 4. The flue gas was collected at the exit section of the reactor. In the figure, the ammonia slip was observed to decrease with the increase in injection temperatures. The maximum ammonia slip was 20 ppm obtained at an injection temperature of 1003 K and the minimum value of 3 ppm was observed at a temperature of 1273 K. This trend of decreasing of ammonia slip with increasing injection temperatures is consistent with the previous studies (Caton et al., 1995; Jodal et al., 1989; Epperly and Broderick, 1988).

Ammonia slip depends upon mixing of reagents with flue gas and rate of vaporization of the reagents. Usually, up to a certain limit, higher temperatures enhance the rate of reagent vaporization as well as mixing of reagents and flue gas and consequently decrease ammonia slip. Apart from that, the above result might be due to the fact that at higher temperatures, some ammonia is oxidized by the oxygen of the flue gas, which increases with the increase in temperature and thus reduces ammonia slip with increasing temperature.

Fig. 7 shows the effect of normalized stoichiometric ratio on ammonia slip at an optimum temperature of 1128 K. The experiment was conducted with 5% urea solution. The ammonia slip was observed to increase with increasing value of NSR over the investigated range of NSR, i.e., 1–4. At an NSR of 1, the ammonia slip was only about 5 ppm and it reached to 12 ppm, while NSR increased from 1 to 4. The trend of increasing of

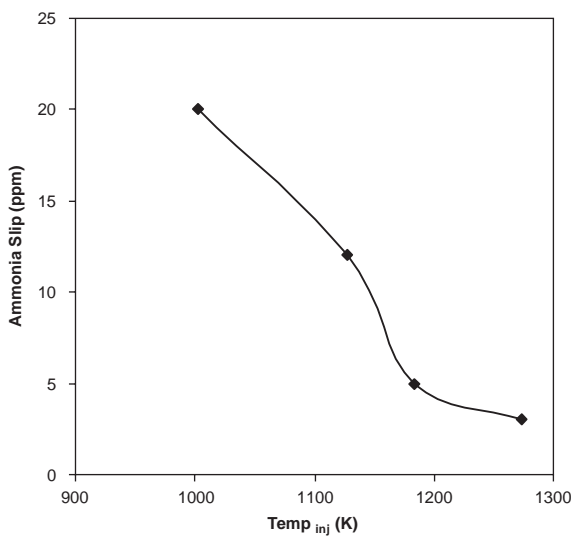


Fig. 6. Ammonia slip vs. injection temperature profile at the exhaust with 5% urea solution at NSR of 4.

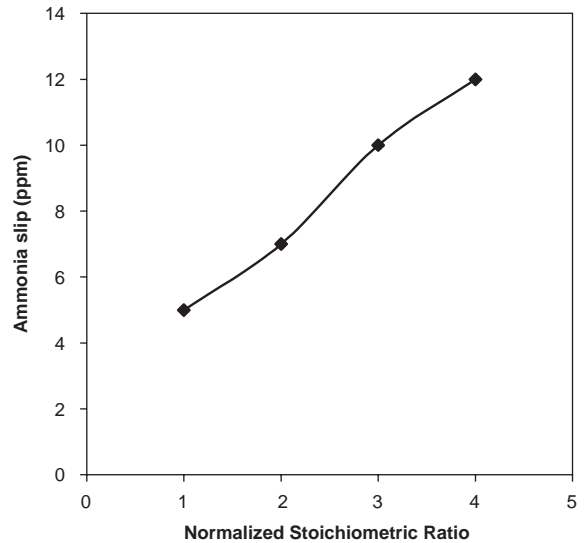


Fig. 7. Effect of NSR on ammonia slip for 5% urea solution at 1128 K.

ammonia slip with the increase in NSR is well demonstrated (Mansour et al., 1987; Jodal et al., 1989; Epperly and Broderick, 1988). This phenomenon could be explained below.

The reagent injection rate increases as NSR increases, which produces coarser droplets. The droplets, which are coarser than the optimum size usually, take longer time to be evaporated and contribute to poor mixing that might cause more ammonia slip at higher NSR. Moreover, at higher NSR, a part of the injected reagent is likely to be unreacted and might increase the ammonia slip. The ammonia slip in the present study is quite low and suggests that the urea SNCR could be used very safely up to an NSR of 4 at the investigated conditions.

5. Conclusions

The SNCR system can achieve significant amount of NO_x reduction from the diesel burning exhaust containing low value of baseline NO_x . At an NSR of 4, a maximum of 54% NO_x reduction was achieved and the effective temperature window was in the range of about 973 K to about 1300 K. The reduction is quite significant as such reduction was not achieved by the previous researchers for the investigated level of baseline NO_x . The effective temperature window is shifted toward lower temperatures as compared to the previous studies, which suggests that the application of commercial grade of urea is effective for low-temperature applications. At optimum temperature of NO_x reduction and NSR of 4, the ammonia slip was around 12 ppm, which is very low and remains within the safe limit. Overall, the

investigations demonstrate that the application of selective non-catalytic reduction of NO_x is quite applicable to small-scale combustion systems.

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