Promoted Selective Non-Catalytic Reduction: Prospective Technology for Nitrogen Oxide Reduction

Khandoker Abul Hossain
Mohammad Nazri Mohd-Jaafar
Farid Nasir Ani
Faculty of Mechanical Engineering
Universiti Teknologi Malaysia
81310 UTM, Skudai, Johor, MALAYSIA

Promoted selective non-catalytic reduction (SNCR) of nitric oxide (No_x) has been studied experimentally by injecting aqueous urea solution with and without additive 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 tests were carried out using commercial grade urea as NO_x-reducing agent and commercial grade sodium carbonate (Na_2CO_3) as additive. The furnace simulated the small-scale combustion systems where (a) operating temperatures are usually within 973–1,323 K and (b) NO_x-emission level remains below 100 ppm. In the SNCR process with 5% urea solution, at normalized stoichiometric ratio (NSR) of 4, as much as 54% reduction was achieved at 1,128 K; while in the promoted SNCR process using Na_2CO_3 additive, NO_x reduction improved to as much as 69% at 1,093 K. In addition, the effective temperature window as well as peak temperature of NO_x reduction shifted towards lower temperatures in promoted SNCR. These results were significant especially for the investigated level of baseline NO_x. The ammonia slip measurements showed that in both cases the slip was below 16 ppm at an NSR of 4 and an optimum temperature of NO_x reduction. The investigations demonstrated that urea-based promoted SNCR may be used for small-scale combustion applications and that commercial grade Na_2CO_3 is a potential additive.

Keywords: Nitrogen oxide (No_x) reduction, normalized stoichiometric ratio (NSR), optimum temperature, selective non-catalytic reduction (SNCR), temperature window, and urea additive.

INTRODUCTION

Increasing deterioration of ambient air has led to stringent regulations to limit the emissions of nitrogen oxide (NO_x), which stimulated a significant number of researches to devise some effective NO_x-abatement technologies. Of these methods, postcombustion technologies are the most widely used throughout the world due to their high NO_x-reduction performance.

Selective catalytic reduction (SCR) and *selective non-catalytic reduction* (SNCR) of NO_x are the two major postcombustion technologies being used in the large combustion installations, such as power plant boilers, industrial boilers, refineries, and waste incinerators (Rentz et al. 1996).

Although SCR has a higher NO_x-reduction efficiency, it has its disadvantages, namely: high capital investment cost, higher operating cost than most options, limited catalyst life, catalyst poisoning, large space requirement to install, and higher upstream pressure required to enable the exhaust gas flow through the catalyst (Caton et al. 1995). In contrast, SNCR has mitigated the problems of SCR. Moreover, it can be used in

dirty and fouling services and is easier to retrofit. Due to these reasons, SNCR is best suited to the needs of developing countries. Recently, it has been adopted by large installations in South Korea, China, Taiwan, and the Czech Republic (Radojevic 1998).

Despite its being inexpensive, the capabilities of SNCR are limited by its: narrow temperature window of NO_x control, higher ammonia slip, and low utilization of NO_x reducing agent. These limitations can be greatly minimized by using *promoted* SNCR, which makes use of additives to the NO_x-reducing agent to promote the reaction and thus, (a) minimize ammonia slip and (b) achieve higher NO_x-reduction efficiency.

It is already demonstrated in a number of researches that additives in urea solution play a significant role in improving the NO_x-reduction performance as well as shifting or widening the effective temperature window of reduction. Many studies which were conducted to find suitable additives to urea in SNCR application demonstrated that certain organic and inorganic compounds may be used as additives.

Among the more common organic compounds are methane, various combinations of hydrocarbons, ethylene glycol, furfural, series of sodium acrylamide co-polymers, and alkaline oxide co-polymers;.while the inorganic compounds are hydrogen, carbon monoxide, hydrogen peroxide, calcium phosphate, and sodium nitrate, among others (Diep et al. 1996, Lyon and Hardy 1986, Burton 1989).

In a recent study, Zamansky et al. (1999) found sodium carbonate (Na_2CO_3) to be a very effective inorganic additive. However, they used laboratory grade Na_2CO_3. In order to reduce the operating cost of the SNCR process some more, further researches are essential to investigate the reduction performance of commercial grade Na_2CO_3.

Thus, the present study is aimed at (a) investigating the NO_x-reduction characteristics of the promoted SNCR process using commercial grade Na_2CO_3 as additive and (b) comparing the results with those of the SNCR process.

EXPERIMENTAL SETUP AND PROCEDURE

An industrial diesel Burner Riello 40–G10 with a 120-kW capacity was used as the combustion source. A combustion chamber of 390-mm OD and 1765-mm in length was fabricated (see Figure 1). The material was 2.5-mm stainless steel sheet. To reduce the heat transfer in order to get high temperature inside
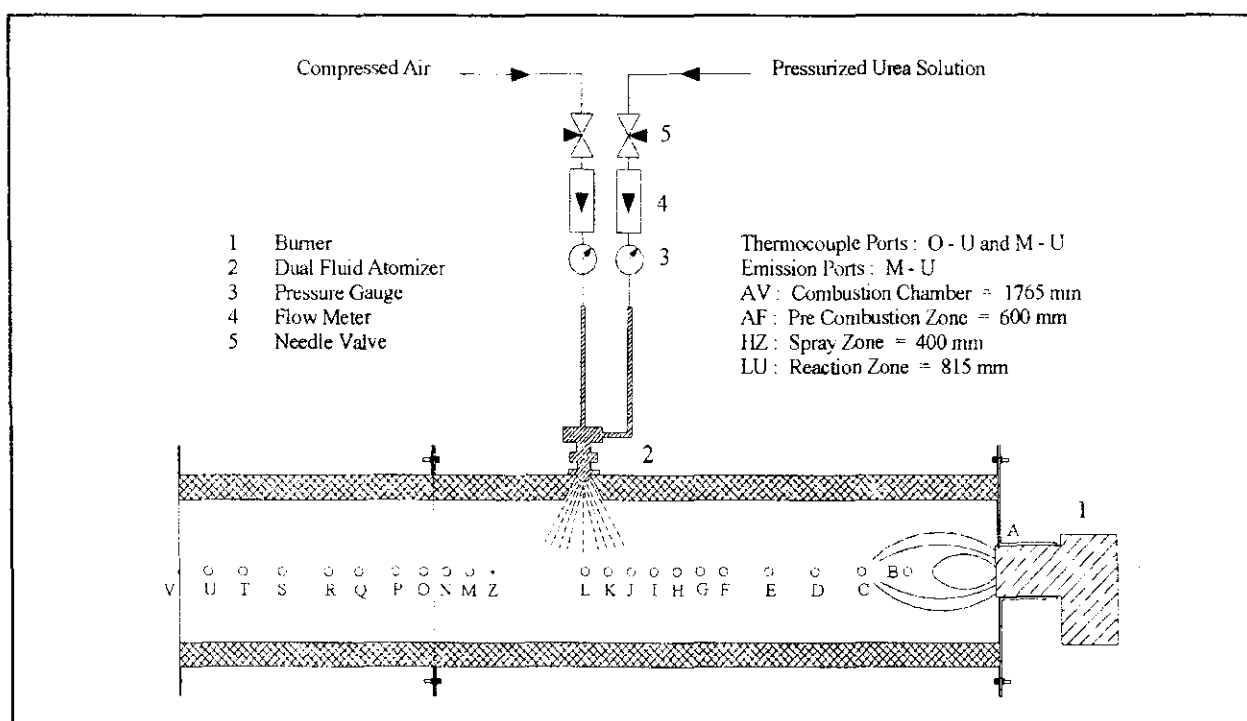

Figure 1. Layout of the Experimental Setup

as well as to protect the combustion chamber wall from overheating, a 50-mm refractory lining was used. For inserting temperature probe and emission probe, a series of temperature tappings and emission tappings were made. The reactor was 815 mm long. The temperature drop along the length of the reactor was about 165-230 K/sec. 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 the hole in such a way that the nozzle tip goes 11 mm inside. The nozzle tip and the body were made of hastelloy to sustain the high temperatures of the burner. The spray was round type with angle of 70°. The atomizer was able to produce the droplets in the range of 20 micron 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 pressure of urea and airline. To achieve effective droplet sizes with good penetration in to the flue gas, the pressure and the flow rate of the atomizing air and the urea were adjusted.

The burner was operated at 3–4 % excess oxygen, while the baseline NO_x varied from about 65 to 75 ppm within the range of operating temperatures, which were in the range of 973 K 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 NSR, the injection temperatures were varied within the investigated temperature range. Before introducing the reagents the baseline NO_x were noted at different temperatures within the operating temperature range and afterward, while the reagent was injected, the NO_x was measured at the corresponding temperatures. Comparing the two data NO_x reduction was obtained. NO_x emissions during the whole experiment were measured at the point U as shown in Figure 1 by using a chemiluminescent analyzer. To investigate the NO_x reduction characteristics of promoted SNCR process a certain amount of additive i.e. sodium carbonate was added to a particular concentration of urea solution and then several

tests were conducted. To investigate the effect of residence time on NO_x reduction at a particular injection temperature, the emission data was taken at six different points on the reactor.

The ammonia slip during the investigation was measured using the standard colorimetric method. The measurement was taken at the exhaust of the reactor at different injection temperatures for a particular value of NSR. The same experiment was repeated for additive case as well. The uncertainty of all the above measurements was less than ± 5%.

RESULTS AND DISCUSSION

Figure 2 shows the NO_x-reduction characteristics of promoted SNCR and compares the data with those of the SNCR process. Urea was used as the NO_x-reducing agent in the SNCR process. In promoted SNCR, Na_2CO_3 was added to the urea solution. The concentration of urea was 5% (wt/vol) and that of Na_2CO_3 was 5% of urea (wt/wt) in the aqueous urea solution. The value of the NSR was 4 throughout the experiment. The figure shows that in promoted SNCR process, the maximum reduction was about 69% at 1,093 K, while with same initial conditions in SNCR process only a maximum of 54% reduction was observed at 1,128 K. Thus, the use of additive in promoted SNCR increased
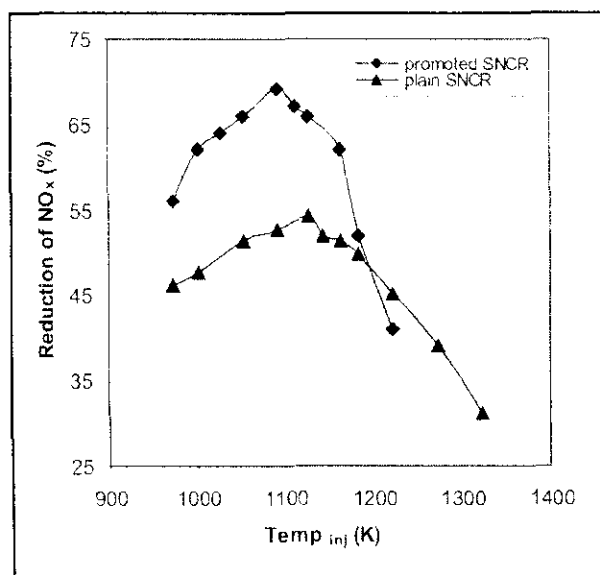

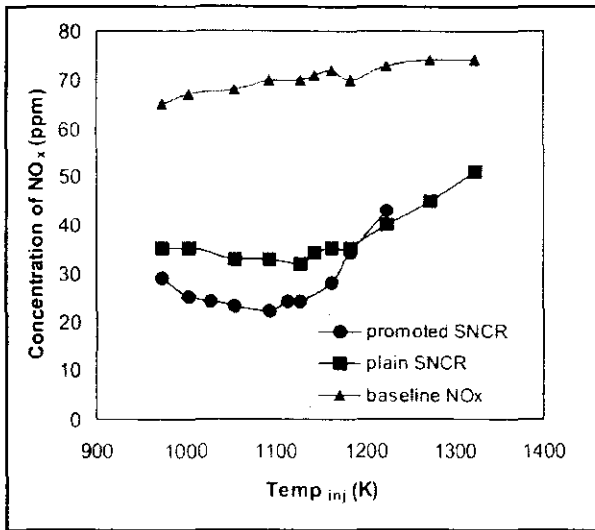**Figure 2. Reduction and Injection Temperature Profiles (SNCR vs. Promoted SNCR)**

Figure 3. NO_x Concentration and Injection Temperature Profiles (SNCR vs. Promoted SNCR)

Figure 4. NO_x Reduction and Residence Time Profiles at 1,093 K and NSR of 4 (SNCR vs Promoted SNCR)

the NO_x reduction by 15%, nevertheless, shifted the peak temperature of NO_x reduction by 35 K towards lower temperatures as compared to SNCR process. This improvement is obviously due to the use of additive as additives play some significant roles in refining the droplets size. Another important feature is that promoted SNCR process the temperature window widened towards lower side of the peak temperature of NOx reduction but the window narrowed towards the higher temperatures, even it was narrower than that obtained without additive case.

The above trends are consistent with the findings of Zamansky et al. (1999), who studied promoted SNCR in a pilot-scale study with laboratory grade Na_2CO_3 in a coal-fired boiler simulator facility. In their studies at optimum temperature, the improvement of NO_x reduction using additive was about 8%, which is lower than the value obtained in the present studies. For the peak temperature-reduction efficiency, in promoted SNCR case the effective temperature window was widened by about 158 K, whilst Zamansky et al. (1999) got only a shifting of about 75 K.

The Figure 2 also shows that between the temperatures of about 1,193 and 1,223 K, in promoted SNCR process, the NO_x reduction decreased more than that achieved in SNCR; and, afterwards some inhibiting effect to the NO_x-reduction performance was observed. These
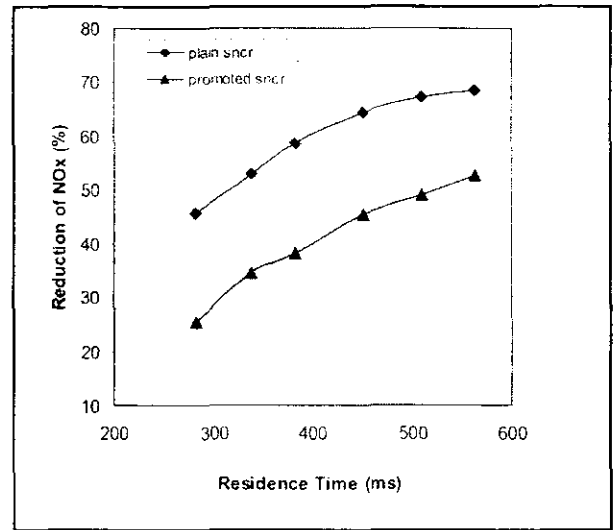
phenomena might be due to the effect of impurities in commercial grade Na_2CO_3, which were sodium chloride (0.7%), sodium sulfate (0.03%), and iron (0.004%).

Figure 3 shows the effect of injection temperatures on the concentration of NO_x and compares the results obtained in promoted SNCR and SNCR cases referring the initial NO_x value of the flue gas as baseline. The baseline NO_x was in the range of 65–75 ppm. The figure shows that up to a certain temperature the concentration of the NO_x in the exhaust gas decreased with increasing injection temperatures for both cases. For the SNCR process, the minimum concentration of NO_x in the flue gas was 32 ppm, observed at 1,128 K, while in promoted SNCR, a minimum of 22 ppm of NO_x was recorded at 1,093 K. So, due to the use of additive in promoted SNCR process, the variation in both final NO_x concentration and peak temperature of reduction occurred as compared to SNCR process.

Figure 4 compares the reaction profiles obtained in SNCR and promoted SNCR process. The experiment was conducted at an NSR of 4 and at 1,093 K. Both profiles exhibited that that NO_x reduction varied as residence time was increased. In the promoted SNCR process, the variation is more uniform and follows a regular trend line more nicely as compared to the SNCR process.
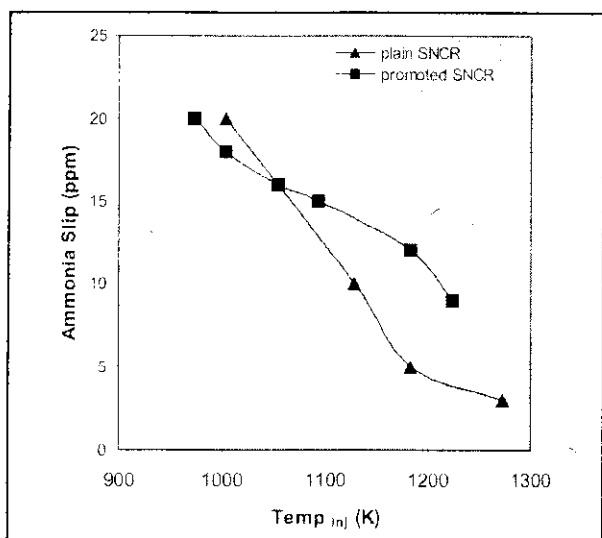
Figure 5. Ammonia Slip at NSR of 4
(SNCR vs. Promoted SNCR)

The above phenomena are due to use of additives as additives have a significant role in refining the droplets, which include the increase of droplets size and reduce the deviations of droplets size and thus play a vital part enabling uniform mixing of the NO_x reducing agent with the effluent and penetration of the reagent sufficiently along the internal path of the reactor to achieve better NO_x reduction performance. So, when additive is added in the urea solution, it produced more uniform droplets, which were more effective in achieving improved NO_x reduction through a regular reaction profile throughout the reactor. In contrast, in SNCR process, the deviation in droplet size is more which achieved overall lower and non-uniform reduction rate. Furthermore, the droplet size of the urea is finer than the additive case and thus resulted in higher reduction rate at the low temperature zone of the combustor compared to the additive case.

Figure 5 shows the ammonia slip as a function of injection temperature in SNCR and promoted SNCR cases. In both cases the ammonia slip was observed to decrease with the increase in injection temperature. The figure shows that in promoted SNCR case, only in a narrow temperature range i.e. 973-1053 K, the ammonia slip was observed to be lesser as compared to SNCR process; otherwise, the SNCR process got lesser ammonia slip through

out the investigated temperature range. This lowering of ammonia slip in promoted SNCR process suggests that it is suitable to be used at further lower temperatures safely as compared to the urea SNCR case. In the present study, for both cases at optimum temperatures the ammonia slip was below 16 ppm. Note that the ammonia slip in the present experiment was measured at 40 mm before the exit plane of the reactor. So, when it mixes with room air it becomes more diluted. As to the limit of ammonia exposure, The Occupational Safety and Health Administration (OSHA) have set a limit of 50 ppm over an 8h-workday, while the American Conference of Governmental Industrial Hygienists (ACGIH) recommended the limit of ammonia gas at 25 ppm in room air. Therefore, the ammonia slip obtained in the present study is quite low and has no hazardous effect on working environment.

CONCLUSIONS

The investigations demonstrated that the promoted SNCR process could achieve significant NO_x reduction compared to the SNCR process. The use of commercial grade Na_2CO_3 as additive in promoted SNCR noticeably improved the NO_x reduction value as well as shifted the effective temperature window towards lower temperatures.

These results suggest that commercial grade Na_2CO_3 is a potential additive that could make the promoted SNCR process more suitable and effective for use at further lower temperatures as compared to the SNCR process.

Finally, the ammonia slip measurements showed that in promoted SNCR, the ammonia slip was lower and leaned towards lower temperatures compared to that in SNCR, which ascertains that promoted SNCR process could be used safely at further lower temperatures than SNCR process.

ACKNOWLEDGMENTS

the Faculty of Mechanical Engineering of Universiti Teknologi Malaysia.

REFERENCES

Burton, A. A. (1989). U.S. Patent 4,842,834.

Bowers, W. E. (1988). U.S. Patent No. 4,719,092.

Caton, J. A., Narney, J. K., Cariappa, H. C., and Laster, W. R. (1995). *The Canadian J. Chem. Eng.*, *73*, 345–50.

Diep, D. V., Linda, L. M., and Cheristiansen, P. B. (1996). U.S. Patent 5,536,482.

Lyon, R. K., and Hardy., J. E. (1986). *Ind. Eng. Chem. Fundam.*, *25*, 19–24.

Radojevic, M. (1998). *Environ. Pollution, 102*, 685–89.

Rentz, O., Schleef, H. J., Dorn, R., Sasse, H., and Karl, U. (1996). *Emission control at stationary sources in the Federal Republic of Germany, vol. 1: Sulfur oxide and nitrogen oxide emission control*, French-German Institute for Environmental Research, University of Karlsruhe, Karlsruhe.

Zamansky, V. M., Lissianski,V. V., Maly, P. M., Rusli, L., Ho, D., and Gardiner, W. C. (1999). *Combust. Flame, 117*, 821–31.