2nd BSME-ASME International Conference on Thermal Engineering
2-4 January 2004, Dhaka

Application of Urea Based Selective Non-Catalytic Reduction of Nitric Oxide in the Combustion Effluent Containing Low Concentration of NO_x.

Khandoker Abul Hossain[1], Mohammad Nazri Mohd. Jaafar[1], Azeman Mustafa[2] and Farid Nasir Ani[1]
[1] Faculty of Mechanical Engineering, Universiti Teknologi Malaysia
[2] Faculty of Chemical Engineering, Universiti Teknologi Malaysia
81310 Skudai, Johor, Malaysia
E-mail: pgkah@graduan.fkm.utm.my

ABSTRACT
Selective Non-Catalytic Reduction (SNCR) of Nitric Oxide has been studied experimentally using commercial grade of urea in a pilot-scale diesel fired tunnel furnace. The furnace simulated small-scale combustion systems such as low capacity boiler, hot water heater, oil heater etc., where the operating temperature is in the range of about 900 to 1300 K. The experiment was conducted with low ppm of base line NO_x ranged from 65 to 74 ppm. A significant amount of NO_x reduction was achieved which was not pronounced by the previous researchers with urea SNCR for this low ppm of NO_x. At a normalized stoichiometric ratio (NSR) of 4, a maximum of 54% reduction was achieved at 1128 K without adding any additives. The effective temperature window for the reaction was about 973 to 1183 K which was lower than that obtained by the previous researchers. Although the NO_x reduction was improved and effective temperature window widened for higher injection rate of the reagent, the optimum temperature of reduction remained almost unaffected.

Keywords: SNCR, NO_x, Temperature window, NSR, Optimum temperature

1. INTRODUCTION

Over the past 150 years, the emissions of nitrogen oxides have been increasing steadily through out the globe. Their growing presence in atmosphere has tremendous impact on earth's ecology and affects human health. Among the nitrogen oxides, emission of nitric oxide (NO) is more significant. Concern for protecting the environment from pollutants emissions has set some stringent regulations to limit the nitrogen oxides emissions in many countries which has stipulated research and driven the development of numerous NO_x abatement technologies. These technologies can be mainly categorized as process modification and post combustion technologies (Sarofim and Flagan, 1976). Process modification involves the modification of combustion conditions employing air staging, fuel staging, flue gas recirculation, water injecting etc., while post combustion technologies are concerned with the flue gas treatment before exhausting out to the atmosphere (Lyon, 1975; Bowman, 1992). Post combustion technologies have demonstrated substantial reduction of NO_x and as a result, presently these technologies are being retrofitted to different power station and industrial boilers and furnaces to achieve desired NO_X emission levels at optimal cost. Selective Non-Catalytic Reduction (SNCR) is a major post combustion technology, which is very efficient, cost effective and easy to retrofit to the existing industries. So many researches based on urea SNCR are already conducted by different researchers which demonstrates that NO_x reduction performance and effective temperature window vary depending upon the geometry of combustion chamber, geometry and performance of the atomizer and types of fuels used. (Mansour et al., 1987; Nylader 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, no document has been documented using urea SNCR yet. As most of the small-scale combustion facilities still use the diesel fuel, so to fill up the large gap research is strongly required in this area. As for low value of base line NO_x, Teixeira et al. (1991) observed that below 125 ppm of base line 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. 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 base line $NO_{x.}$ In all the previous studies, where urea SNCR was concerned, were conducted using laboratory grade of urea. So far, no document is available using the commercial grade of urea. Laboratory grade is much more expensive than commercial grade. So, to make the urea SNCR cheaper and acceptable to all levels it is essential to conduct some researches to investigate the performance of commercial grade of urea in reducing NO_x.

In these perspectives, the present study is aimed to investigate the NO_x reduction characteristics of commercial grade of urea in reducing NO_x from a diesel burning exhaust that is containing low ppm of base line NO_x.

2. EXPERIMENTAL SETUP AND PROCEDURE

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 1700 mm in length was fabricated. The material was 2.5 mm mild 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 tappings were made. The reactor was 815 mm long. The temperature drop along the length of the reactor was about 230 K, which simulated very closely the practical condition of small-scale boilers and other combustion systems. A dual fluid internal mix injector 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. The liquid urea and airflow were measured precisely by using the needle valves. 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.

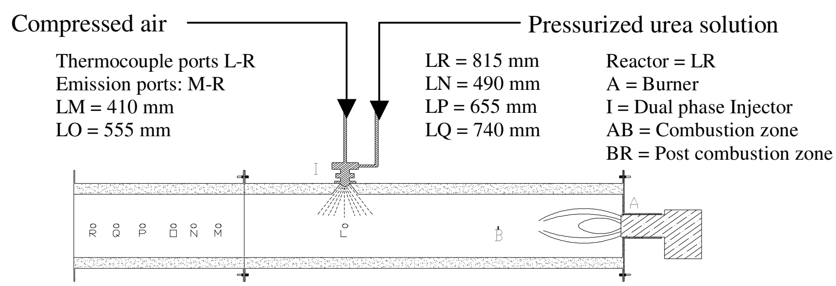

Figure 2.1. Layout of the Experimental Setup

Two flow meters and pressure gauges were used to measure the flow rate and pressure of urea and airline. As urea is highly corrosive, teflon tubing and stainless steel measuring instruments were used. To achieve effective droplet size 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 % of excess oxygen, while the base line NO_x varied from about 65 to 75 ppm within the operating temperatures, which were in the range of 973 K to 1323 K. It is worthy to be mentioned that the fuel contained no fuel bound nitrogen. To study the effect of the variation of the 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. The pressure of the injection was kept constant during the whole experiment. Before introducing the reagents the base line 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 same temperatures. Comparing the two data NO_x reduction was obtained. To study the effect of stoichiometric ratio on the NO_x reduction performance, at a certain injection temperature, for the different values of NSR the NO_x emission data were taken at the exhaust of the combustion chamber. To investigate the effect of residence time on NO_x reduction, at a particular injection temperature, the emission data was taken at the six different points on the reactor. The NO_x emissions were measured by a continuous emission analyzer.

3. RESULTS AND DISCUSSIONS

Figure 3.1 shows the NO_x reduction as a function of 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 sharply with further increase in temperature. The temperatures at which maximum NO_x reduction occurred for different values of NSR, remained in a narrow temperature range of 1093 K to 1128 K. The observed NO_x reduction trend with the variation of injection temperatures is in close agreement with the findings reported by some prominent researchers (Mansour et al., 1987; Jodal et al., 1989; Jones et al., 1989; Sun and Hofmann, 1990; Irfan, 1996; Zamansky et al., 1999). However, the different researchers have got different effective temperature window and different value of peak temperature of NO_x reduction. This variation is very likely to occur, as the experimental conditions in their studies were not same. The effective temperature window in their studies was in the range of 1023 to 1473 K, while the width of the temperature window varied from 100-300 K. In the present study, at a NSR of 1, the effective temperature window was about 973 to about 1200 K, which shifted towards lower temperatures than previous results. Similarly for other three values of NSR, the similar trend was observed.

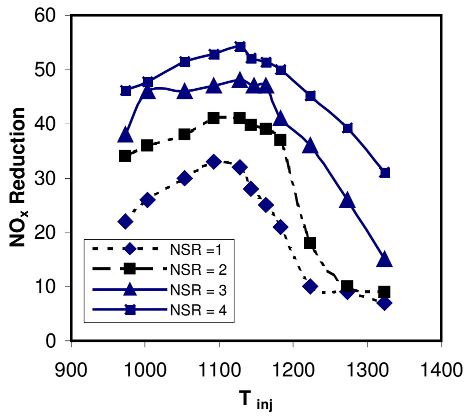

Figure 3.1 Effect of Injection Temperature on NO_x Reduction

In the figure for a NSR of 1 a maximum reduction of 33% was achieved and it reached to as much as 54% while NSR varies 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 about 1128 K. The reduction obtained in the present study is quite significant, which is not pronounced in prior studies for this low ppm of NO_x. Teixeira and Muzio (1991) performed a pilot scale investigation using natural gas fired combustor with a base line NO_x of 125 ppm. They observed that below 125 ppm of initial NO_x, reduction was quite insignificant.

Figures 3.1 and 3.2 show the effect of normalized stoichiometric ratio on the NO_x reduction. Figure 3.2 shows that NO_x reduction increased with the increase in NSR. For optimum temperature i.e. at 1128 K, the NO_x reduction rose moderately with the increase in NSR, whilst at 1273 K, which is higher than the optimum value, the rising was not smooth. Between the 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%. Figure 3.1 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 155 K. This phenomenon of widening of temperature window with increase in molar ratio is in agreement with the findings of Wenli et al. (1989) and Irfan (1996). Another significant aspect is exhibited in the present studies 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 to NO_x reduction. This phenomenon is in accordance with the findings of the prior studies reported by Nylander et al. (1989). Figure 3.3 presents the NO_x reduction as a function of residence time. NO_x reduction increased with increases in residence time. Between the residence time 262 and 355 ms, at 1173 K the reduction was faster. This rise is due to the fact that within that small part of the reactor the average reaction temperature was very close to the peak reduction temperature of 1128 K. On the contrary at 1093 K, the rate of NO_x reduction is faster up to a residence time of 262 ms i.e. at the first part of the reactor, as in this case within the portion of the reactor temperatures are close to the optimum temperature.

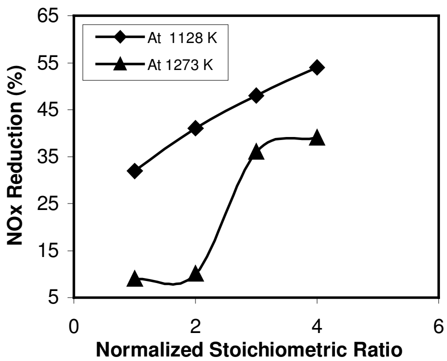

Figure 3.2. Effect of NSR on NO_x Reduction

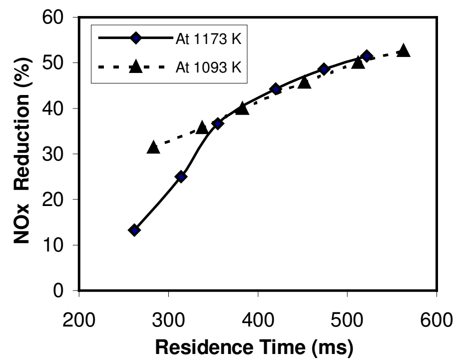

Figure 3.3. Effect of Residence Time on NO_x Reduction

For both cases after a certain residence time, with the increase in residence time the rate of NO_x reduction decreased and gradually tended to be flat. This is due to 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. 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 and this phenomenon is in accordance with the findings of the prior studies reported by Sun et al. (1990) and Irfan (1996).

4. CONCLUSIONS

The selective non-catalytic reduction of NO_x using commercial grade of urea demonstrates substantial NO_x reduction in the diesel burning exhaust that contains low ppm of initial NO_x and that much of NO_x reduction was not pronounced by the previous researchers in their SNCR studies with low ppm of base line NO_x. The commercial grade of urea is a potential NO_x reducing agent and it will make the process cost much lower. The NO_x reduction as well as the width of the effective temperature window increases with increases in NSR. At a NSR of 4, a maximum of 54% NO_x reduction is achieved. At NSR of 4, the effective temperature window is in the range of about 973 K to about 1300 K, which suggests that the selective non-catalytic reduction of NO_x also could be used effectively for low temperature application without using any additives.

REFERENCES

Bowman, C.T. (1992), "Control of Combustion-generated Nitrogen Oxide Emissions: Technology Driven by Regulation", *Proceedings of the Twenty-Fourth Symposium (International) on Combustion,* Combustion Institute, Pittsburgh, pp. 859-878.

Irfan, N. (1996), "Selective Non-catalytic reduction of NO_x", *Unversity of Leeds*, Ph.D Thesis.

Jodal, M., Nielsen, C., Hulgaard, T. and Dam-Johansen, K. (1989), "A Comparative Study of Ammonia and Urea as Reductants in Selective Non-Catalytic Reduction of Nitric Oxide", ACHEMASIA, Beijing.

Jones, D.G., Muzio, L.J., Stocker, E., Nuesch, P.C., Nagrea, S., Ofenbau, K.K, Lautenschlager, G., Wachter, E. and Rose, G. (1989), "Two stage $DeNO_x$ Process Test Data for 300 TPD MSW Inceneration Plant", *The Proceedings of the 82nd APCA Meeting and Exhibition,* Anaheim, CA. No. 8, pp. 21-27.

Lyon. R. K. (1975), "Method for the Reduction of the Concentration of NO in Combustion Effluents Using Ammonia", *U.S. Patent No. 3,900,554*.

Mansour, M.N., Nahas, S.N., Quartucy, G.C., Nylander, J.H., Kerry, H.A., Radak, L.J., Eskinazi, D., and Behrens, T.S. (1987), "Full Scale Evaluation of Urea Injection for NO Removal", *The Proceedings of the Joint EPA/EPRI Symposium on Stationary Combustion NO_x Control*, New Orleans, Louisiana. No. 43, pp. 1-23.

Nylander, J.H., Mansour, M.N. and Douglas, R.B. (1989), "Demonstration of an Automated Urea Injection System at Encina Unit 2", *The Proceedings of the Joint EPA/EPRI Symposium on Stationary Combustion NO_x Control*, San Francisco, No. 9, pp. 35-56.

Sarofim, A.S. and Flagan, R.C. (1976), "NOx Control of Stationary Combustion Sources" *Progress in Energy and Combustion Science*, Vol.2, pp. 1-25.

Sun, W.H., Hofaman, J.E. and Pachaly, R. (1990), "Post Combustion NO_x Reduction with Urea: Theory and Practice", *The Proceedings of the Seventh Annual International Pittsburgh Coal Conference*, Pittsburgh.

Teixeira, D.P. and Muzio, L.J. (1991), "Widening the Urea Temperature Window", *The Proceedings of the Joint EPA/EPRI Symposium on Stationary Combustion NO_x Control*, NTIS, Springfield, VA, Section 6A, pp. 23-41.

Wenli, D., Dam-Johansen, K. and Ostrergaard, K. (1989), "The Influence of Additives on Selective Non-Catalytic Reduction of Nitric Oxide with Ammonia", *ACHEMASIA*, Beijing.

Zamansky, V.M., Lissianski, V.V., Maly, P.M., Ho, L., Rusli, D. and Gardiner, W.C. Jr. (1999), "Reactions of Sodium Species in the Promoted SNCR Process", *Combustion and Flame*, Vol. 117, pp. 821-831.