Small-Scale Application of Urea Based Selective Non-Catalytic Reduction of Nitric Oxide in Diesel Burning Effluent

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ABSTRACT

Selective Non-Catalytic Reduction (SNCR) of nitric oxide has been studied experimentally injecting different concentrations of aqueous urea solution in a pilot-scale diesel fired tunnel furnace, which was set to run at 3-4% excess oxygen level and was producing low ppm of baseline NO_x ranged from 65 to 74 ppm within the investigated temperature range. The furnace simulated small-scale combustion systems such as low capacity boilers, hot water heaters, oil heaters etc., where the operating temperatures are usually in the range of about 900 to 1300 K and baseline NO_x emission level remains within 100 ppm. One significant aspect of the study is that it employed commercial grade of urea as NO_x reducing agent. The test data showed a significant amount of NO_x reduction, which was not pronounced by the previous researchers with urea SNCR for this low ppm of NO_x. Variation in concentrations of urea solution has effect on NO_x reduction characteristics with residence time, injection temperatures and normalized stoichiometric ratio (NSR). At a particular value of NSR, NO_x reduction as well as effective temperature window was observed to be affected at different concentrations of urea flow. At an NSR of 3, a maximum of 51 % reduction was observed using 10% aqueous urea solution at 1128 K, while 25% urea solution achieved only a maximum of 26% reduction. Although the NO_x reduction and width of the effective temperature window varied significantly with the variation in solution concentration, the peak temperature of the reduction varied trivially.

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

INTRODUCTION

Today, control of NO_x emission is becoming increasingly important because of their tremendous adverse effect on health and environment. The industrial revolution is the central cause for the increase of NO_x in the atmosphere. Emissions are rapidly increasing in the areas such as East Asia and Southeast Asia, where increases in fossil fuel use by transport are helping to push emissions up. Rapid increases in global air travel are also a concern with greater potential for increased emission of NO_x directly to the troposphere. Due to the growing concern of the NO_x some stringent regulations have been applied to limit the NO_x emission and as a result, to date, so many technologies have been invented, which can be mainly categorized as process modification and post combustion technologies (Sarofim and Flagan, 1976). Post combustion

technologies have demonstrated better performance as compared to process modification

technologies. The major post combustion technologies are Selective Catalytic Reduction
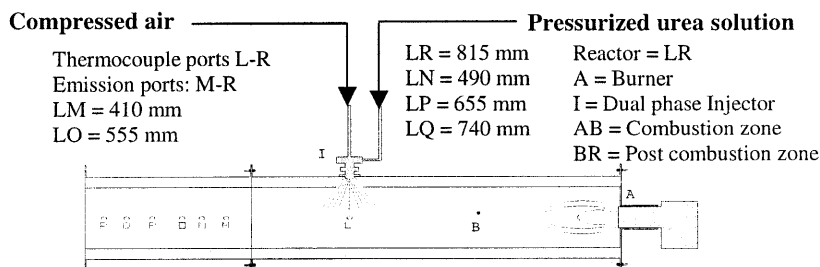
Compressed air ———————————— **Pressurized urea solution**

Thermocouple ports L-R
Emission ports: M-R
LM = 410 mm
LO = 555 mm

LR = 815 mm   Reactor = LR
LN = 490 mm   A = Burner
LP = 655 mm   I = Dual phase Injector
LQ = 740 mm   AB = Combustion zone
                 BR = Post combustion zone

Fig. 1 Layout of the Experimental Setup

(SCR) and Selective Non-Catalytic Reduction Process (SNCR). 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. In contrast, SNCR has minimized all the problems of SCR. Moreover, it can be used in dirty and fouling services (Particulates and/or high sulfur) and it is easier to retrofit. So many researches based on urea SNCR are already conducted by different researchers, which demonstrated that NO_x reduction performance and effective temperature window vary depending on 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 natural gas as fuel. So, further studies are required employing different types of fuels' exhaust to get a 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 study the SNCR performance with commercial grade of urea. In these perspectives, the present study is aimed to investigate the NO_x reduction characteristics of SNCR using different concentrations of commercial grade of aqueous urea solution from a diesel burning exhaust containing low ppm of baseline NO_x.

EXPERIMENTAL SETUP AND ROCEDURE

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. 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 varied 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. 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 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 % of excess oxygen, while the base line NO_x was 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 injection temperatures on the performance of NO_x reduction, at 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. To study the influence of reagent concentration, firstly a certain concentration of urea solution was injected at a

particular value of NSR over an investigated range of injection temperature and the NO_x data was recorded. After that another concentration of the urea solution was injected with the same NSR and at same injection temperatures. Similar procedure was repeated for other concentrations of the urea solution.

RESULTS AND DISCUSSIONS

Figure 2 shows the effect of dilution of the urea solution on the NO_x reduction vs. injection temperature profile. In the figure the NO_x reduction has been plotted as a function of the injection temperature. The aqueous solution of urea of three different concentrations i.e. 5%, 10% and 25% were studied, while the NSR during the experiment was set to 3. The graphs show that maximum NO_x reduction for 5%, 10% and 25% cases was 48%, 51% and 32% respectively, while the peak temperature of reduction was 1128 K, 1103 K and 1093 K respectively. So, for 10% case the reduction was quite significant and for 25% case reduction was rather low.
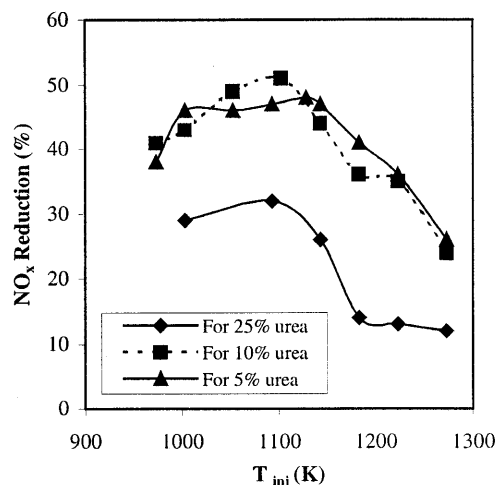

Fig. 2 Effect of Dilution on NO_x Reduction at NSR of 3

Another significant feature is apparent that the peak temperature of NO_x reduction shifted slightly towards lower temperatures as the concentration of the solution increased. As far as temperature window is concerned, figure 2 exhibits that apart from shifting the peak temperature of NO_x reduction, dilution has a significant effect on widening the effective temperature window as

well. For 25% concentration case the width of the temperature window is insignificant, however, for other two cases the temperature window was widened to a certain extent. Between 5% and 10% cases it seems that the temperature window is slightly wider for 10% case. For 5% case temperature window shifted slightly towards higher temperatures as compared to 10% case. On the other hand for 10% case, the window shifted a bit towards lower temperatures than 5% case. The above results of dilution effect qualitatively support the previous studies reported by Irfan (1996).

The above results are due to the variation in droplet sizes produced during the experiment. For 10% case with NSR of 3, at a particular pressure and flow rate of the carrier gas the reagent injection rate was quite good in producing proper droplets sizes to have better mixing and penetration of the reagent in the effluent and thus resulted in highest NO_x reduction performance. As for 25% solution to maintain the same value of NSR, the required reagent injection rate was less, which resulted in finer droplet size that contributed to poor mixing and penetration within the flue gas. As far as 5% urea solution is concerned, the droplet size was too coarse to give better output around the temperature 1103 K but afterwards with increase in temperature, 5% urea solution resulted in better NO_x reduction. Similarly regarding widening of temperature window, for 10% urea, the droplet sizes were finer than 5% urea, which were more effective for lower temperatures and thus the temperature window was widened towards lower temperatures and narrowed towards higher temperatures than 5% urea. The above results and explanations are consistent with the claim of Bowers (1988), who pronounced that at higher effluent temperatures, the more dilute solution is the more effective. This phenomenon is due to the fact that more dilute regent produces relatively coarser droplets. Apart from the good mixing and penetration within the effluent, coarser droplets, which contain more water, help to protect the active reagent from the extreme heat until it is delivered to the preferred location for reaction. Again if the droplet size is too coarse with respect to a certain temperature then water takes time to be evaporated, which impedes the active reagent to be converted to

gaseous phase and thus mixing with NO and that ultimately results in poor reduction performance.

The reduction obtained in the present study for 5 and 10% cases 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 insignificant.

In the figure for 5 and 10% cases the width of the temperature window was widened greatly compared to 25% case. The observed effective temperature window for above two cases is about 973 to 1250 K. However, the different researchers have got different effective temperature window and different value of peak temperature of NO_x reduction (Mansour et al., 1987; Jodal et al., 1989; Jones et al., 1989; Sun and Hofmann, 1990; Irfan, 1996; Zamansky et al., 1999). This variation is very likely to occur, as the experimental conditions in their studies were not same. Actually, the performance of urea SNCR is subject to the type of fuels used, injection quality of the reagent, geometry of the reactor, composition and velocity of the exhaust gas, thermodynamic conditions of the combustion effluent etc. The effective temperature window in their studies was in the range of 1023 to 1473 K, while the width of temperature window varied from 100-300 K. So, In the present study the observed temperature window shifted towards lower temperatures. The phenomena related to the improvement of NO_x reduction and shifting of effective temperature window towards lower temperatures might be due to the following causes:

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 might have contained some organic additives such as unburned hydrocarbon, which might have played an active role as additive to shift the temperature window towards lower temperatures. Some previous studies demonstrated that a range of organic compounds could be used as additives to improve the NO_x reduction performance as well as to shift the effective temperature window

(Bowers, 1988; Teixeira and Muzio, 1991; Irfan, 1996).

Secondly, the use of commercial grade of urea, which contains some impurities such as biuret may play the role as additive and contribute such change. It is worthy to be mentioned that no previous document is available as for the use of commercial grade of urea.

Figure 3 exhibits the effect of dilution on the NO_x reduction vs. NSR profile. The test was conducted using two different concentrations of aqueous urea solution i.e. 5 and 25% at a range of NSR from 1 to 4 and at 1093 and 1183 K. The figure shows that at both temperatures 5% urea solution achieved better NO_x reduction within the investigated range of the NSR. Another significant feature revealed in the figure is that the rate of NO_x reduction with the increase in NSR is higher for 5% urea solution for both the temperatures throughout the NSR range. The result is evidently due to the fact that 25% urea solution always produced the droplets finer to get optimum result at tested injection temperatures.
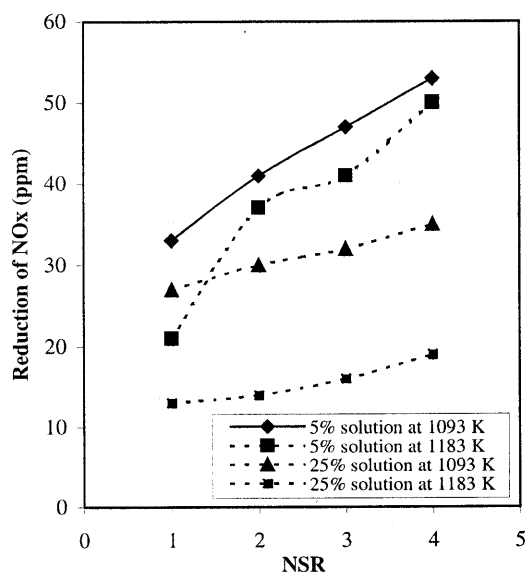

**Fig. 3 Effect of Dilution on NOx
Reduction Vs. NSR Profile**

Figure 4 presents the NO_x reduction as a function of residence time for 5% and 25% concentrations of the aqueous urea solution at 1173 K, NO_x reduction increased with increases in residence

time. Although the NO_x reduction at different residence time for the two cases was observed to be different, however the trend of the both curves are similar. It is evident that 5% urea solution achieved higher reduction corresponding to at any residence time throughout the investigated range, which suggests that to achieve a particular NO_x reduction value more dilute solution requires smaller reactor. For both cases, in the first zone of the reactor the reaction rate was observed to be faster but afterwards the reaction rate decayed with the increase in residence time 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.
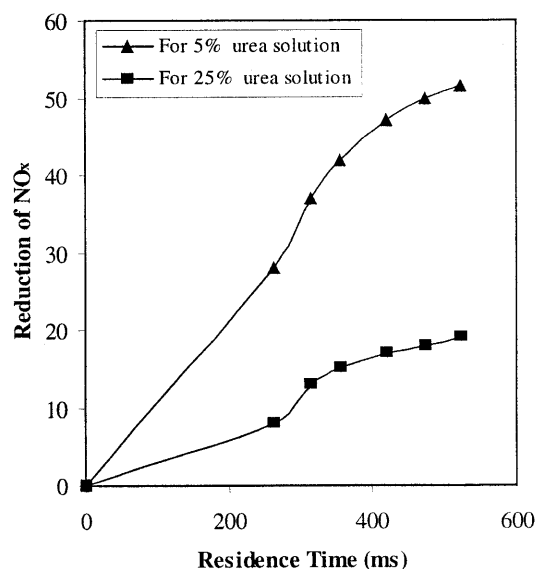

**Fig 4. Effect of Dilution on the NO_x
Reduction Vs. residence Time
Profile at NSR = 4**

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 reduction performance and this phenomenon is in accordance with the findings of the prior studies reported by Sun et al. (1990) and Irfan (1996).

CONCLUSIONS

The selective non-catalytic reduction of NO_x employing commercial grade of urea demonstrates to be suitable method for NO_x reduction in terms of the NO_x reduction value and the width of the effective temperature window in diesel burning effluent containing low initial ppm of NO_x. As much as 51% reduction was achieved at an NSR of 3 for 10% aqueous urea solution, which is quite significant as such level of reduction was not achieved by the previous researchers for low ppm of base line NO_x. The study also demonstrates that dilution of reagent has significant effect in reducing NO_x as well as widening the effective temperature window of NO_x reduction. Besides, it also affects the NO_x reduction vs. residence time profile and NO_x reduction vs. NSR profile. In the present studies it is observed that 5% and 10% solution achieved overall better NO_x reduction performance than 25% case. Between 5% and 10% cases, 5% case achieved overall wider temperature window, on the other hand 10% case achieved better NO_x reduction performance, which suggests that dilution of the reagent is evidently effective, however it does not mean that the more dilute solution is more effective. The effective dilution is subject to the operating conditions of the combustion system and the target NO_x reduction value from the particular SNCR system.

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