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APPLICATION OF COMMERCIAL GRADE AMMONIA LIQUOR IN THE SELECTIVE NON-CATALYTIC REDUCTION OF NO_X IN COMBUSTED GAS FROM A DIESEL BURNER

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Abstract. A pilot-scale investigation with Selective Non-catalytic Reduction (SNCR) of NO_x were carried out by employing the aqueous solution of commercial grade ammonia liquor as NO_x reducing agent in diesel burning exhaust at 6% excess oxygen and injection temperatures between 923 and 1323 K. The furnace simulated the small-scale combustion systems such as low capacity boilers, water heaters, oil heaters etc., which produce low level of baseline NO_x, usually below 100 ppm and where the operating temperatures remain within the investigated temperature range. With 5% aqueous ammonia solution, at a Normalised Stoichiometric Ratio (NSR) of 4, as much as 57% reduction was achieved at an optimum temperature of 1063 K, which is quite significant for the investigated level of baseline NO_x. The NO_x reduction was observed to increase as the value of NSR increased. The residence time of the reagent has significant effect on the reaction performance, however, most of the reduction occurred within a short range of residence time and after that further increase in residence time showed very insignificant effect. The ammonia slip measurement showed that at optimum temperature and NSR of 4, the slip was about 18 ppm. The investigations demonstrate that commercial grade liquor ammonia is a potential NO_x reducing agent, which could be used effectively and safely considering the health hazards.

Keywords: SNCR, NO_x, NSR, ammonia slip

Abstrak. Satu kajian menggunakan rig skala pandu dengan Penurunan Tanpa-bermangkin Terpilih (SNCR) telah dijalankan menggunakan larutan akuas likur ammonia gred komersial sebagai agen penurunan NO_x di dalam gas ekzos yang membakar diesel pada 6% oksigen lebihan dan dengan suhu pancitan antara 923 dan 1323 K. Relau ini mensimulasikan sistem pembakaran skala-kecil seperti dandang muatan rendah, pemanas air dan pemanas minyak, dan sebagainya, yang menghasilkan NO_x garis dasar yang rendah selalunya di bawah 100 ppm dan di mana suhu kendalian sentiasa berada dalam julat suhu yang dikaji. Dengan larutan 5% akuas ammonia, pada Nisbah Stoikiometrik Ternormal (NSR) 4, sekurang-kurangnya 57% pengurangan telah diperolehi pada suhu optimum 1063 K, yang mana adalah amat ketara bagi aras NO_x garis dasar yang dikaji. Pengurangan NO_x ini didapati meningkat dengan meningkatnya nilai NSR. Masa mastautin regen ini mempunyai kesan yang ketara ke atas prestasi tindak balas, namun, kebanyakan daripada pengurangan berlaku di dalam julat masa mastautin yang pendek dan selepas itu penambahan selanjutnya dalam masa mastautin tidak menunjukkan sebarang perubahan yang ketara. Pengukuran gelincir ammonia menunjukkan bahawa pada suhu optimum dan NSR 4, gelincirnya ialah sekitar

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18 ppm. Kajian ini juga menunjukkan bahawa ammonia likur gred komersial berpotensi sebagai agen penurunan NO_x yang boleh digunakan secara selamat dan berkesan jika dipertimbangkan masalah mudarat kesihatan.

Kata kunci: SNCR, NOx, NSR, gelincir ammonia

1.0 INTRODUCTION

Growing concerns of ambient air quality has led to stringent regulations to curb the emissions of nitrogen oxides, which consequently stimulated a significant number of researches to develop some effective NO_x abatement technologies. Nowadays, post combustion technologies are being widely used throughout the world due to their higher NO_x reduction performance. Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR) of NO_x are two major post combustion technologies, which are being commonly used in large combustion installations such as power plant boilers, industrial boilers, refineries and waste incinerators [1]. Albeit 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 [2]. In contrast, SNCR has mitigated the problems of SCR. Moreover, it can be used in dirty and fouling services and easier to retrofit. Due to these reasons, SNCR is best suited to the developing countries. Recently, it has been adopted in different large installations in South Korea, China, Taiwan, and the Czech Republic [3].

Many researches based on ammonia SNCR have already been conducted by different researchers, which demonstrated 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 [4-6]. Most of the researches were related to 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 ammonia SNCR. As most of the small-scale combustion facilities in the developing countries still use the diesel fuel, so to fill up the large gap, research is strongly required in this area.

As for low value of baseline NO_x, one previous research achieved very insignificant reduction with 120 ppm of baseline NO_x [7], while another research revealed that below 100 ppm initial concentration of NO_x the reduction was prevented [8]. So far, very few experiments were conducted as for low concentration of initial NO_x, therefore, in order to get better understanding on the applicability of SNCR in low initial concentration case, further researches are warranted with different combustion and injection parameters.

So far, no single document revealed the NO_x reduction efficiency of commercial grade liquor ammonia. Since laboratory grade ammonia is much more expensive than commercial grade, comprehensive studies should be warranted to investigate the NO_x reduction characteristics of commercial grade ammonia to make the process running cost cheaper.

In these perspectives, the present studies are aimed to investigate the NO_x reduction characteristics of commercial grade of liquor urea with the variation of injection temperature, NSR and residence time in reducing NO_x from diesel burning exhaust containing low ppm of baseline NO_x.

2.0 EXPERIMENTAL SET-UP

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 (Figure 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 and emission probe, a series of temperature and emission tappings were made. The reactor was 815 mm long. The temperature drop of the flue gas along the length of the reactor was about 165-230 K/sec. A dual fluid internal

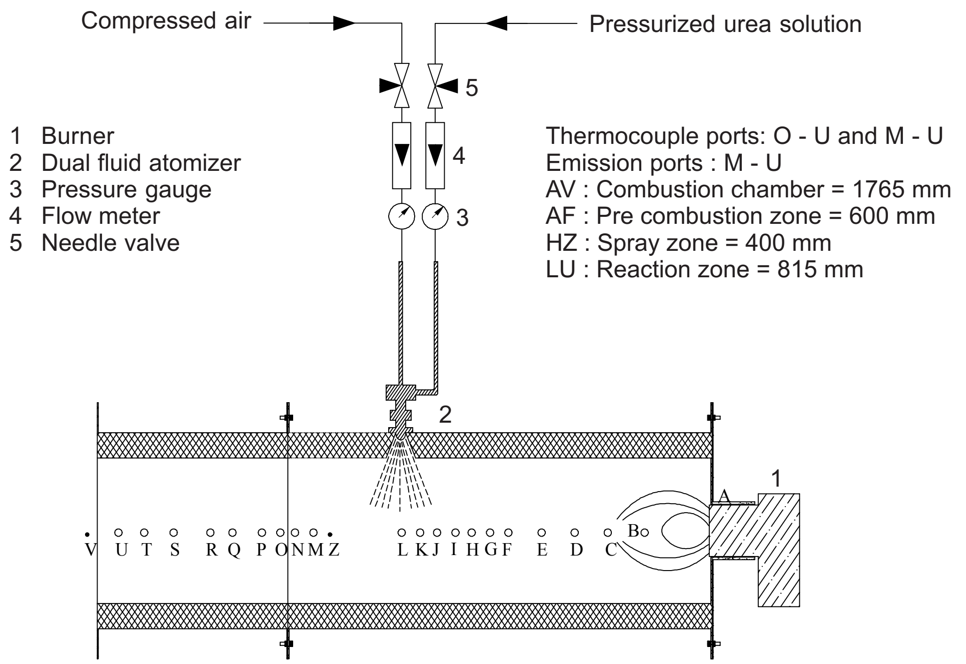

Figure 1 Layout of the experimental set-up

mix injector manufactured by Spraying Systems Co., USA was used for injecting aqueous ammonia 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 an 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 ammonia flow solenoid valves and timer relays were used. To maintain the constant pressure in air line and ammonia line, two individual pressure regulators were used. Two flow meters and pressure gauges were used to measure the flow rate and pressure of ammonia and airline. To achieve effective droplet sizes with good penetration into the flue gas, the pressure and the flow rate of the atomizing air and the ammonia solution were adjusted.

3.0 EXPERIMENTAL PROCEDURE

The burner was operated at 3 - 4% 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. It is worthy to mention 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, 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 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 the normalized 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 just before the exhaust of the reactor i.e. at point U (Figure 1). To investigate the effect of residence time on NO_x reduction, at a particular injection temperature, the emission data was taken at eight different points on the reactor. NO_x emissions during the whole experiment were measured by a continuous chemiluminescent emission analyser. The ammonia slip during the investigations was measured using the standard colorimetric method. Firstly, the ammonia slip was measured at the exhaust 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 plain ammonia solution was injected into the flue gas at varying NSR and the corresponding ammonia slip was recorded. The uncertainty of all the above measurements was less than + 5%.

4.0 RESULTS AND DISCUSSION

4.1 Effect of Injection Temperature

Figure 2 shows the variation of NO_x reduction with the variation of injection temperature. The study was conducted with 5% (wt/vol) aqueous ammonia solution and at an NSR of 4. In the figure the NO_x reduction was observed to increase with the increase in injection temperatures up to a temperature of 1063 K and after that with further increase in temperatures, NO_x reduction was observed to decay drastically. At the optimum temperature of 1063 K, a maximum of 57% of NO_x reduction was recorded.

The above NO_x reduction trend with the variation of injection temperatures is in close agreement with the findings reported by some prominent researchers [9-13]. However, different researchers reported 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 the same. In fact, the performance of SNCR depends on the type of fuels used, injection quality of the reagent, reagent fuel mixing, geometry of the reactor, composition and velocity of the exhaust gas, thermodynamic conditions of the combustion chamber etc.

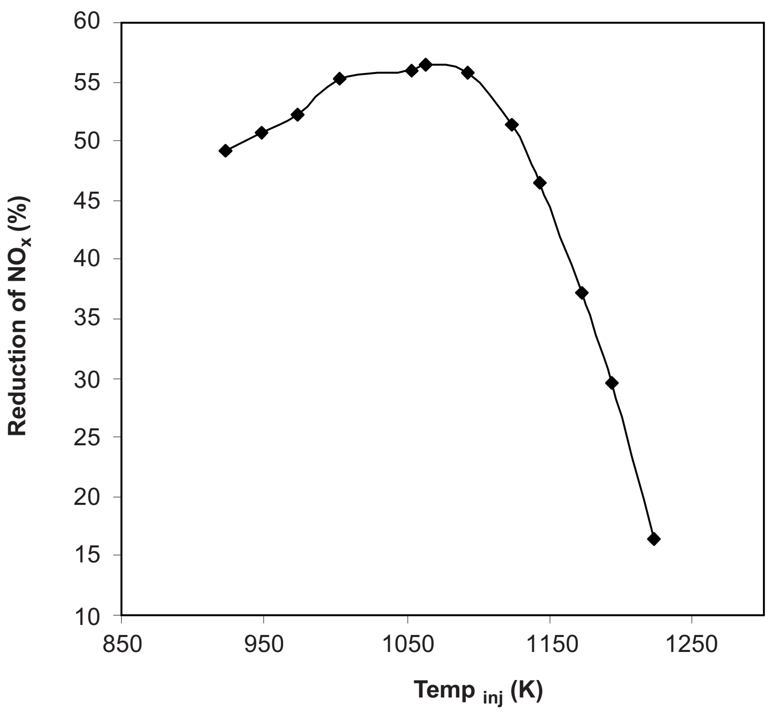

Figure 2 Effect of injection temperature on NO_x reduction for 5% ammonia liquor at NSR of 4

The effective temperature window in their studies was in the range of about 973 to 1373 K, while the width of temperature window and peak temperature of NO_x reduction varied from 100 - 400 K and 1123 - 1323 K respectively. In the present study, at an NSR of 4, the effective temperature window was in the range of about 923 to 1193 K, which shifted towards lower temperature. This might be due to the causes as described below.

Firstly, it might have happened due to the use of diesel fuel as the exhaust of the diesel fuel contains some unburned hydrocarbons, which might have worked as additives and contributed to such change. Secondly, the use of commercial grade ammonia, which contains some impurities such as lead may play the role as additive and contribute to such changes. It is worthy to mention that no previous document is available as for the use of commercial grade ammonia.

4.2 Effect of Normalised Stoichiometric Ratio

Figure 3 shows the NO_x reduction as a function of normalised stoichiometric ratio. The figure shows that NO_x reduction increased with the increase in NSR. The experiment was conducted at an injection temperature of 1093 K. In the figure, the NO_x reduction rose moderately with the increase in NSR. At an NSR of 4, a maximum of 56% NO_x reduction was achieved. The improvement of NO_x reduction with

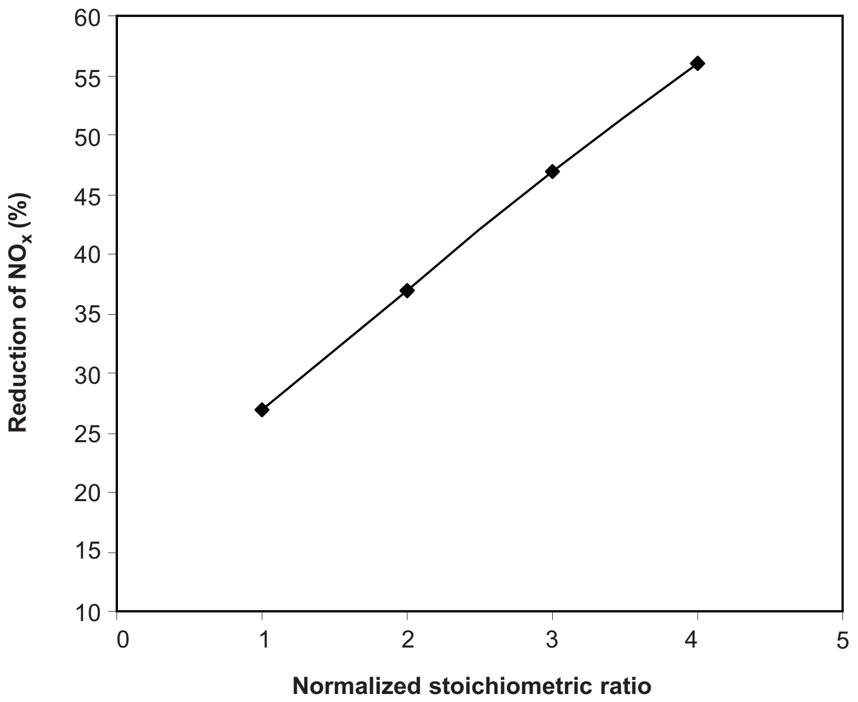

Figure 3 Effect of NSR on NO_x reduction for 5% ammonia liquor at 1093 K

increasing normalised stoichiometric ratio is supported in so many previous studies reported by some prominent researchers [10, 13-14].

One significant feature is that between the NSR of 3 and 4, the reduction efficiency improves slowly than before. This is in close agreement with the findings of some previous researchers [8,10]. They found in their studies that after a certain value of NSR, NO_x reduction increased insignificantly with further increase in NSR.

The good penetration of the reagent, effective reagent-flue gas mixing and long time survival of the reagent in the flue gas greatly depend on the amount of reagent injection. So, when the NSR increases, the amount of reagent increases which achieves better penetration, more effective reagent-flue gas mixing and long time survival of droplets and ultimately results in better NO_x reduction than before.

4.3 Effect of Residence Time

NO_x reaction profile as a function of residence time for 5% aqueous ammonia solution at a temperature of 1093 K is shown in Figures 4 and 5. In Figure 4, it is shown that the concentration of NO_x decreased with the increase in residence time. Up to the residence time of 283 ms the reduction was moderate, however, afterwards with further increase in residence time the reduction rate decreased gradually and tended to be flat. Figure 5 shows that about 43% NO_x reduction was achieved within the

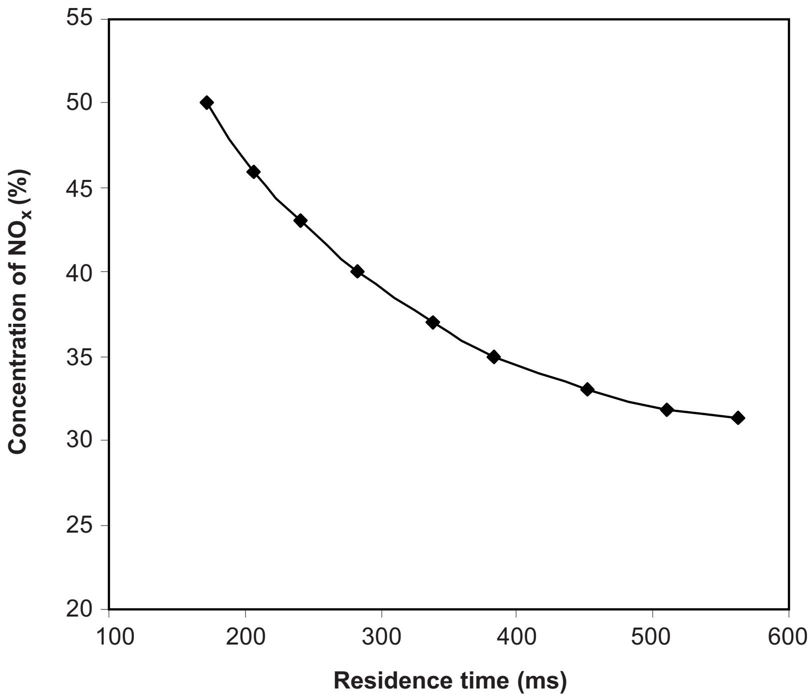

Figure 4 Reaction profile of NO_x concentration for 5% ammonia liquor at 1093 K and NSR of 4

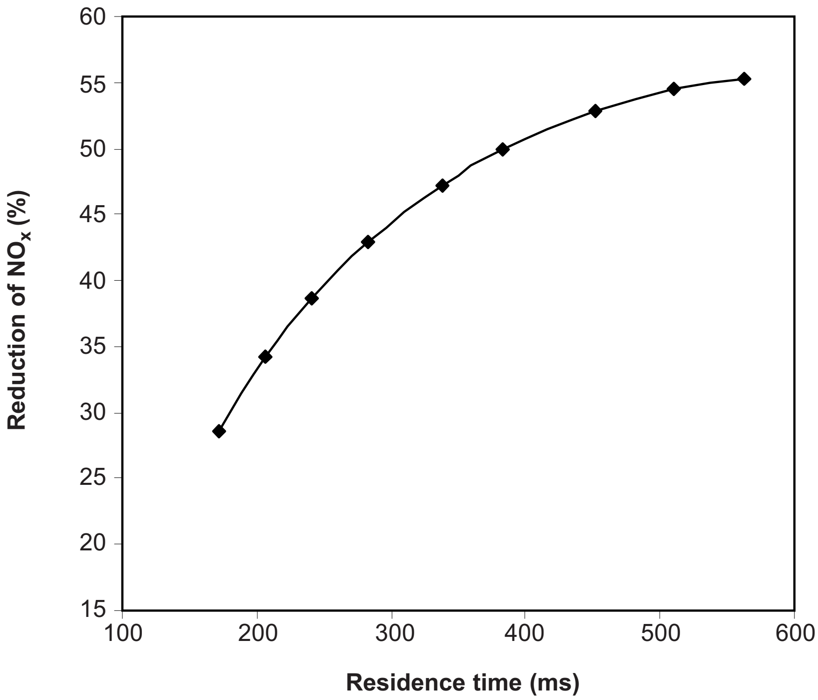

Figure 5 Reaction profile of NO_x reduction for 5% ammonia liquor at 1093 K and NSR of 4

residence time of 283 ms, while in another 280 ms, the reduction was improved by 12% only. The profile obtained in the present study is almost similar to that obtained by Irfan [12], who injected liquor ammonia was in a pilot scale natural gas fired furnace that simulated the power plant conditions.

It has already been discussed in Section 4.1 that injection temperature has significant effect on NO_x reduction performance. Due to the temperature drop along the axial direction of the reactor, the flue gas temperature decreases as the residence time of the flue gas increases which results in decreasing rate of NO_x reduction along the downstream section of the reactor.

Aside from the low temperature effect, this rate might decrease due to low reagent flue gas ratio. The trend of the profile suggests that depending on the operating conditions and geometry of the reactor after a certain length the reactor has no significant role in reducing NO_x.

4.4 Effect of Injection Temperature on Ammonia Slip

Figure 6 shows the ammonia slip as a function of injection temperatures of the ammonia. The test was conducted with 5% ammonia solution at an NSR of 4. The flue gas was collected at the exhaust of the reactor. In the figure, the ammonia slip

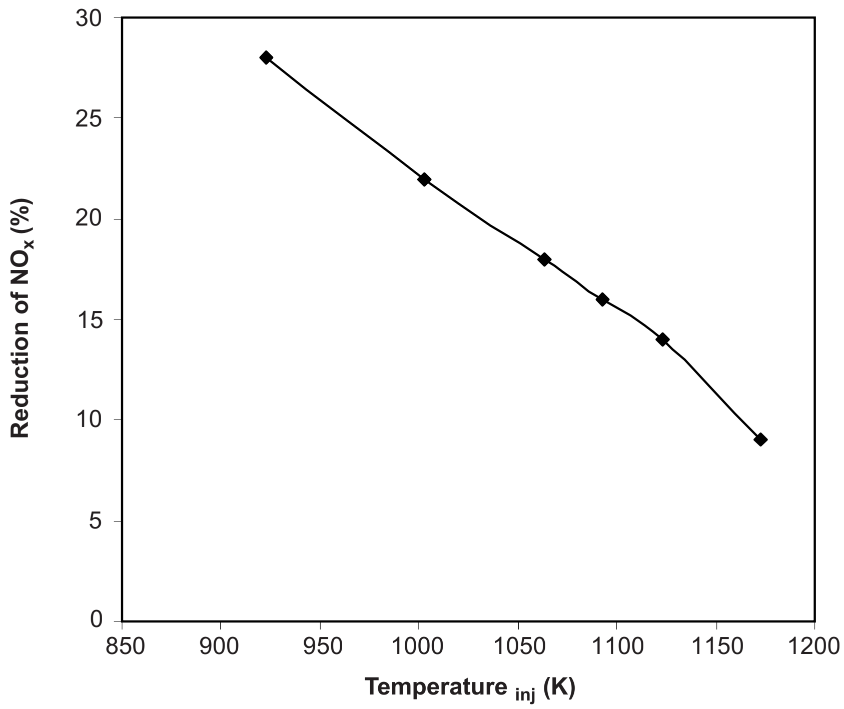

Figure 6 Ammonia slip as a function of injection temperature at NSR of 4

was observed to decrease with increase in the injection temperatures. The maximum ammonia slip was 28 ppm achieved at an injection temperature of 923 K and the minimum value of 9 ppm was observed at a temperature of 1173 K. This trend of decreasing of ammonia slip with increasing temperatures is consistent with the previous studies [2].

Ammonia slip depends upon mixing of reagents with flue gas and rate of vaporisation of the reagents. Usually, up to a certain limit, higher temperatures enhance the rate of reagent vaporisation as well as mixing of the 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 oxidised by the oxygen of the flue gas, which increases with the increase in temperature and thus eventually reduces ammonia slip.

4.5 Effect of Normalised Stoichiometric Ratio on Ammonia Slip

Figure 7 shows the effect of normalised stoichiometric ratio on ammonia slip at an optimum temperature of 1063 K. The experiment was conducted with 5% plain ammonia solution. The ammonia slip was observed to increase with increasing value

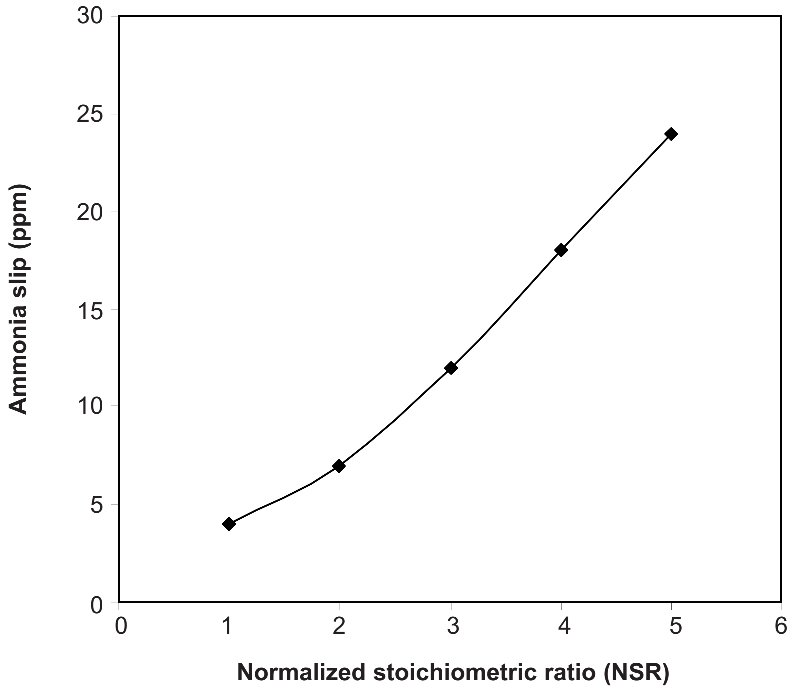

Figure 7 Ammonia slip as a function of NSR at the optimum temperature of 1063 K

of NSR within the investigated range of NSR, i.e. 1 to 5. At an NSR of 1, the ammonia slip was only about 4 ppm and it reached to 24 ppm, while NSR increased from 1 to 5. The trend of increasing of ammonia slip with the increase in NSR is well demonstrated [8, 15]. This phenomenon could be explained below.

The reagent injection rate increases as NSR increases, which produces coarser droplets. Ammonia slip depends to a greater extent on the rate of reagent vaporisation and mixing of the reagents with flue gas. 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 ammonia SNCR could be used very safely up to an NSR of 4 at the investigated conditions.

5.0 CONCLUSIONS

The use of commercial grade of liquor ammonia as NO_x reducing agent in SNCR system achieved significant amount of reduction from the diesel burning exhaust containing low value of baseline NO_x. At an NSR of 4, a maximum of 57% NO_x

reduction was achieved and the effective temperature window was in the range of about 923 K to about 1193 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 residence time profile showed that most of the reductions take place within 400 ms, which suggests that after a certain length of the reactor, further increase in reactor has insignificant effect in reducing NO_x. At optimum temperature of NO_x reduction and NSR of 4, the ammonia slip was around 18 ppm, which is low and remains within the safety limit. Overall, the investigations demonstrate that the application of commercial grade of ammonia liquor is a potential NO_x reducing agent, which could make the SNCR technique more sustainable.

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