

**STUDY OF UREA BASED SELECTIVE NON-CATALYTIC REDUCTION OF
NO_x IN SMALL SCALE COMBUSTION APPLICATIONS**

KHANDOKER ABUL HOSSAIN

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To my beloved parents, who sacrificed a lot

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ABSTRACT

Selective Non-Catalytic Reduction (SNCR) of nitric oxide was studied experimentally by injecting different concentrations of aqueous urea solution in a pilot-scale diesel fired tunnel furnace at 3-4 % excess oxygen level and with low ppm of baseline NO_x ranged from 65 to 75 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 973 to 1323 K and NO_x emission level remains below 100 ppm. In order to investigate the influence of additive on reduction characteristics, different concentrations of commercial grade sodium carbonate (Na_2CO_3) were added to urea solution. The significant aspects of the studies are that it employed commercial grade urea as NO_x reducing agent and commercial grade Na_2CO_3 as additive to urea solution to minimize the cost of the SNCR operation. NO_x reductions were studied with the variation of different parameters such as injection temperature, residence time, Normalized Stoichiometric Ratio (NSR) of the reagent, carrier gas pressure, etc. 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 . With 5% plain urea solution, at an NSR of 4 as much as 54% reduction was achieved at 1128 K, whilst in the additive case the NO_x reduction was improved to as much as 69% at 1093 K. Apart from this improvement, in the additive case, the effective temperature window as well as peak temperature of NO_x reduction shifted towards lower temperatures. The ammonia slip measurements showed that in both cases the slip was below 16 ppm at NSR of 3 and optimum temperature of NO_x reduction. Finally, the investigations demonstrated that urea based SNCR is quite applicable to small-scale combustion applications and commercial grade urea and sodium carbonate are potential NO_x reducing agent and additive respectively.

ABSTRAK

Satu kajian mengenai Penurunan Bukan Bermangkin Terpilih (SNCR) terhadap nitrik oksida telah dijalankan secara ujikaji dengan menyembur larutan akues urea yang berbeza kepekatan di dalam sebuah terowang relau disel berskala kecil pada paras lebih oksigen sebanyak 3-4% serta pada nilai emisi NO_x yang rendah iaitu antara 67-75 ppm dalam julat suhu yang dikaji. Relau yang digunakan mewakili sebuah sistem pembakaran berskala kecil seperti dandang berkapasiti rendah, pemanas air, pemanas minyak dan sebagainya, di mana suhu operasi relau pada kebiasaannya adalah di dalam lingkungan 973-1323 K manakala paras emisi NO_x adalah di bawah 100 ppm. Dalam mengkaji kesan penambahan larutan urea bagi mengurangkan emisi, pelbagai kepekatan natrium karbonat (Na_2CO_3) bergred komersial telah ditambah ke dalam larutan urea tersebut. Aspek-Aspek penting yang difokuskan dalam kajian ini adalah penggunaan Na_2CO_3 , juga bergred komersial, sebagai bahan tambahan kepada larutan urea, untuk mengurangkan kos operasi SNCR. Kajian ke atas pengurangan NO_x dijalankan dengan mengubah beberapa parameter seperti suhu semburan, masa bermastautin, nilai Nisbah Stoikiometri Ternormal (NSR) bagi reagen, tekanan gas pembawa dan sebagainya. Pengurangan nilai NO_x yang ketara telah diperolehi dalam kajian ini yang mana belum pernah dilaporkan oleh mana-mana penyelidik sebelum ini bagi bahan urea SNCR untuk kadar ppm NO_x yang rendah. Pada nilai NSR bersamaan 4 dengan 5% larutan urea, sebanyak 54% pengurangan NO_x telah diperolehi pada suhu 1128 K, manakala bagi kes penambahan bahan penambah pula, pengurangan NO_x telah meningkat sehingga 69% pada suhu 1093 K. Selain dari pembaikan ini, bagi kes bahan penambah, julat suhu efektif dan suhu maksimum pengurangan NO_x telah berubah ke nilai yang lebih rendah. Dalam kedua-dua kes, didapati nilai ammonia yang tidak bertindakbalas yang telah diukur menunjukkan satu nilai yang rendah iaitu di bawah 16 ppm pada NSR 3 serta pada suhu pengurangan NO_x yang optimum. Akhir sekali, kajian yang telah dijalankan menunjukkan bahawa SNCR berasaskan urea boleh digunakan bagi kegunaan pembakaran berskala kecil, manakala bahan urea serta natrium karbonat yang bergred komersial juga berpotensi sebagai agen pengurangan NO_x serta agen penambah.

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LIST OF SYMBOLS

$^{\circ}\text{C}$	-	Degree Celsius
$^{\circ}\text{F}$	-	Degree Fahrenheit
<i>ASME</i>	-	American Society of Mechanical Engineers
<i>CN</i>	-	Cyanide
<i>CO</i>	-	Carbon monoxide
<i>CO₂</i>	-	Carbon oxide
<i>d</i>	-	Diameter
<i>FGR</i>	-	Flue Gas Recirculation
<i>gm</i>	-	Gram
<i>GPH</i>	-	Gallon per Hour
<i>H₂</i>	-	Hydrogen
<i>H₂O₂</i>	-	Hydrogen peroxide
<i>H₂SO₄</i>	-	Sulfuric acid
<i>HCN</i>	-	Hydrogen cyanide
<i>hr</i>	-	Hour
<i>K</i>	-	Kelvin
<i>kJ</i>	-	Kilo Joule
<i>Kmol</i>	-	Kilo mole
<i>kW</i>	-	Kilowatt
<i>LNB</i>	-	Low NO _x Burner
<i>m³</i>	-	Cubic meter
<i>ml</i>	-	Milliliter
<i>mm</i>	-	Millimeter
<i>MW</i>	-	Megawatt
<i>N₂O</i>	-	Nitrous oxide
<i>Na₂CO₃</i>	-	Sodium carbonate

NH_3	-	Ammonia
NO	-	Nitric oxide
NO_2	-	Nitrogen dioxide
NO_x	-	Nitrogen oxides
NSR	-	Normalized Stoichiometric Ratio
O_2	-	Oxygen
O_3	-	Ozone
OFA	-	Over Fire Air
ppm	-	Parts Per Million
SCR	-	Selective Catalytic Reduction
$SNCR$	-	Selective Non-Catalytic Reduction
SO_2	-	Sulfur dioxide
T	-	Temperature
VOC	-	Volatile Organic Compound
vol	-	Volume
wt	-	Weight

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CHAPTER 1

INTRODUCTION

Over the past 150 years, the emissions of nitrogen oxides have been increasing steadily through out the globe. Their growing presence in the atmosphere has tremendous impact on earth's ecology and affects human health. The origin of these emissions is mostly anthropogenic and mainly attributed to the combustion of fossil fuels and biomass. Continuing industrial revolution and growing numbers of traffics increase the use of fossil fuels and as a result, these oxides are becoming increasing. Rapid increases in global air travel are also a concern with great potential for increased emission of nitrogen oxides directly to the troposphere. All oxides of nitrogen are commonly referred as NO_x but most common are NO, NO_2 and N_2O . Among the nitrogen oxides, emission of nitric oxide (NO) is the most significant. Over 90% of the NO_x from the combustion sources is in the form of NO and eventually it forms NO_2 reacting with atmospheric oxygen and only for this reason most of the NO_x reduction efforts are related to the abatement of NO produced during combustion. Concerns for protecting the environment from pollutants emissions have set some stringent regulations to limit the nitrogen oxides emissions in many countries.

This chapter initially provides a brief overview of the environmental and health concerns of NO_x . This is followed by the importance of the present studies. Finally, the objectives and scopes of the present work and the thesis layout are described at the end of the chapter.

1.1 Detrimental Effects of NO_x

Concerns related to NO_x emissions are becoming increasing because of their tremendous adverse effect on health and environment. NO_x are directly involved in producing photochemical pollutants and acid rain. Besides, N₂O is regarded as green house gas, which causes global warming.

1.1.1 Impact of NO_x on the Environment

1.1.1.1 Photochemical Smog

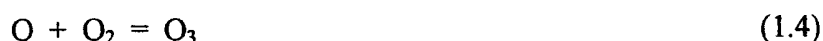
Photochemical smog is a condition, which develops when the primary pollutants such as oxides of nitrogen and volatile organic compounds (VOC) interact under the influence of sunlight at a temperature greater than 291 K (Grennfelt *et al.*, 1984). Development of photochemical smog is typically connected with the particular climatic conditions and most likely to occur in the cities of high population density. Cities like Los Angeles, New York, Vancouver, Sydney and London frequently experience the episodes of photochemical smog. Photochemical smog consists of hundreds of different hazardous chemicals. It reduces the visibility due to its glowing brown colour (Elsom, 1992). The major constituents of photochemical smog are NO_x, volatile organic compounds, ozone and peroxyacetyl nitrates (PAN). Nitrogen oxides might be formed by one of the following reactions:



R is the hydrocarbon radical that is produced from volatile organic compounds. Sunlight can break down the nitrogen dioxide (NO_2) to form nitrogen oxide (NO).



The atomic oxygen of the above equation reacts with atmospheric oxygen and eventually forms ozone (O_3).



NO_2 also reacts with the hydrocarbon radicals in a series of reaction and forms PAN.

All the components of photochemical smog have harmful impact on environment and human health. NO and NO_2 are harmful if inhaled in high concentration. NO_2 can reduce the photosynthesis and CO_2 fumigation and thus suppresses the plant growth (Hill and Bennett, 1970). Volatile organic compounds (VOC) causes eye irritation, respiratory irritation and decreases visibility due to its blue-brown-haze. Ozone has numerous detrimental effects on human health such as it causes bronchial constriction, coughing, wheezing, respiratory irritation and eye irritation. It has also bad impact on environment as it retards plant growth, damages plastics and breaks down rubber. Peroxyacetyl nitrates (PAN) cause eye irritation, respiratory irritation and damages proteins. Like other constituent of photochemical smog it is also toxic to plants.

1.1.1.2 Acid Deposition

Acidic deposition, or "acid rain," describes any form of precipitation, including rain, snow, and fog, with a pH of 5.5 or below. Two common air pollutants are responsible for acid rain: sulfur dioxide (SO_2) and nitrogen oxides (NO_x). When

nitrogen oxides dissolve in water and decompose with water they form nitric acid (HNO_3) and nitrous acid (HNO_2). Acid rain causes damages to plantation and vegetation. It increases the acidity of lake water and severely affects the aquatic species. Acidification can also affect vertebrate species other than fish. For example, studies show acidic deposition can affect the diet, foraging, distribution, and reproduction of bird species that depend on the aquatic environment (Longcore *et al.*, 1993). Acidic deposition affects terrestrial wildlife species by damaging habitat or contaminating food sources (Schreiber and Newman 1988).

1.1.1.3 Global Warming

Global warming is a consequence of green house effect, however, green house effect is due to the effect of some trace gases in the atmosphere such as nitrous oxide, carbon dioxide, ground level ozone and methane. They absorb the longer wavelength of heat energy or reflect it back to earth surface and thus provide resistance to heat energy to be radiated to the space. As a consequence the globe is getting heated gradually. If such trend is allowed to continue then forests, agriculture, water resources, natural ecosystem and human as well as animal health will be severely affected. Besides, sea level is predicted to rise, which will submerge plenty of low lands.

N_2O contributed about 6% of the green house effect in 1980s. Between 1880 and 1980, the concentration of N_2O was changed from 285 to 300 ppb, which was theoretically responsible for 0.02 K temperature change (Houghton *et al.*, 1990). Although NO and N_2O are not directly related to the green house effect, they contribute indirectly in producing the ground level ozone.

1.1.1.4 Ozone Layer Depletion

Stratospheric ozone is found in a broad band, usually extending from about 15 to 35 kilometers above the earth. Although it makes up to one-millionth of the volume of the atmosphere, Ozone plays very important role to absorb the UV-B and UV-C rays from the sun, so it is essential to the existence of most life on earth (Hengeveld, 1995). Ultraviolet radiation is extremely harmful to living tissue and UV-B and UV-C rays are particularly damaging. Ozone absorbs almost all of the UV-C and prevents more than 70% of the UV-B radiation from entering into the earth's surface. (Hengeveld, 2000). The ozone layer is continuously depleting by some direct or indirect effects of some substances such as chlorine (Cl), bromine (Br), nitrogen oxides (NO_x), and hydrogen oxide radicals (HO_x). Among these only direct as well as indirect activity of NO_x has been attributed to contribute to almost half of the stratospheric ozone layer depletion (Sloss et al, 1992).

The Aircrafts are the major sources of nitrogen oxides' emissions to the stratosphere. For instance, nitrogen oxide emissions (NO_x) from aircraft account for around 3% of anthropogenic NO_x emissions, however 25-30% of total NO_x in the upper troposphere is due to aircraft emissions (Lamarque *et al.*, 1996). Ultraviolet ray splits relatively unstable O_3 molecules into O_2 and atomic O. Most of the time, the O atom created by ozone break up and recombines with one of the plentiful O_2 molecules to form O_3 again. This ozone-creation process is constantly at work and producing more ozone. But N_2O reacts with that free O atom to form NO. Ozone then oxidizes the NO to form NO_2 that further reduces by the O atom and eventually forms more NO. So, plenty of ozone atoms are destroyed by this chain reaction. If N_2O is made double it could result in a 12% increase in total stratospheric ozone (Sloss *et al.*, 1992).

1.1.2 Impact of NO_x on Health

The oxides of nitrogen have severe adverse effect on human as well as animal health. Both NO and NO₂ contribute to heart and lung problems. Exposure of low levels of NO₂ can affect the function of kidneys, liver, spleen, red blood cells and cells of the immune system (Sloss *et al.*, 1992). Besides, NO and NO₂ may encourage the spread of cancer. Most common disease related to NO₂ exposure is respiratory illness. An intensive survey conducted in some areas in USA showed that an increase in respiratory illness occurred after six months for an average concentration of NO₂ from 0.109 to 0.062 ppm. The same survey reported that the infant acute bronchitis cases also increased for NO₂ concentrations in the range from 0.063 to 0.083 ppm over a six- month period (Shy *et al.*, 1970).

NO_x have severe bad impact on animal health as well. It was observed to be fatal to most exposed mammals at a concentration higher than 100 ppm (US EPA, 1993). Short-term non-lethal exposure of NO₂ can change the pulmonary function in the lungs of monkey (Henry *et al.*, 1965). Exposure of 15 to 50 ppm for two hours caused damage to lungs, heart, liver, kidneys and pulmonary changes of monkeys (US EPA, 1993). A 12-minute exposure of mice to 2500 ppm of NO was observed to be lethal (Flury and Zernick, 1931).

1.1.3 Impact of NO_x on Materials

The effects of air pollution are also extended to man made items such as fabrics, metals and cultural properties. Loss of color has been observed in cotton as well as rayon fabrics due to the effect of only 0.6 to 2 ppm of NO_x generated from natural gas heated domestic dryers (McLendon and Richardson, 1965). Cotton and nylon textile fibers can deteriorate from exposure to elevated ambient NO_x

concentrations (Morris et al, 1964). Increased metal failures might occur due to the elevated particulate nitrate and NO_x concentrations (Hermance *et al.*, 1970). Nitrogen oxides can also accelerate corrosion to nickel, aluminum and pewter.

1.2 The Importance of the Present Studies

Today, control of NO_x emission is becoming increasingly important because of their tremendous adverse effect on health and environment as mentioned in Section 1.1. The industrial revolution is the central cause for the increase of NO_x in the atmosphere. All types of combustion sources are contributing the NO_x. Boilers, furnaces and industrial burners are the major combustion sources. The continuous rising trend of NO_x is becoming increasingly alarming for the human health and environment. Due to the growing concerns of the NO_x some stringent regulations have been applied to limit the NO_x emission and as a result, emission from the developed world is remained relatively constant over the last few years. So application of NO_x abatement technologies is very important for developing countries. A great numbers of researches are required to develop suitable NO_x abatement technologies in perspective of these areas.

In order to reduce NO_x, both primary and secondary measures are employed. (Sarofim and Flagan, 1976; Rosenberg *et al.*, 1980). Primary measures modify the combustion conditions by employing different techniques, such as fuel rich combustion, lowering the primary air temperature, multistage combustion, flame cooling, flue gas recirculation etc. However, such measures tend to produce undesirable levels of nitrous oxide and carbon monoxide and their NO_x reduction efficiency is not so remarkable. There is no known primary method that can reduce both NO_x and carbon monoxide to an acceptable level without serious economic drawbacks.

As a consequence of continuous extensive investigations, some economically attractive as well as efficient methods in aspect of NO_x reduction have come in to face and among these most common and widely accepted methods are Selective Catalytic Reduction (SCR) and Selective Non-Catalytic Reduction (SNCR). Both are involved in combustion gas treatment before it comes out to atmosphere (Lyon, 1975; Bowman, 1992).

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 and fouling services (Particulates and/or high sulfur) and it is easier to retrofit. For this, nowadays a number of SNCR installations have been adopted in coal, oil and gas fired power station boilers, industrial boilers, refineries and waste incinerators (Rentz *et al.*, 1996). As it requires little capital cost and easy to retrofit, it is best suited to the developing countries. Recently, SNCR has been adopted in different industries in South Korea, China, Taiwan and the Czech Republic (Redojevic, 1998).

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 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; 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 urea. So far, no document is available using the commercial grade urea. Laboratory grade is much more expensive than commercial grade. So, in order 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 urea in reducing NO_x .

It is already demonstrated in a number of researches that additives in urea solution have some significant roles in improving the NO_x reduction performance as well as shifting or widening the effective temperature window of reduction. A lot of studies were conducted in order to find some suitable additives for urea SNCR application, which demonstrated that certain organic and inorganic compounds could be used as additives. The organic compounds are commonly methane, various combinations of hydrocarbons, ethylene glycol, furfural, series of sodium acrylamide co-polymers and alkaline oxide co-polymers, while inorganic compounds are hydrogen, carbon monoxide, hydrogen peroxide, calcium phosphate, sodium nitrate etc. (Daniel *et al.*, 1996; Lyon and Hardy, 1986; Burton, 1989). In a recent study, Zamansky *et al.* (1999) demonstrated the sodium carbonate (Na_2CO_3) to be a very effective inorganic additive. However, they used laboratory grade Na_2CO_3 , so, in order to reduce the operating cost of the SNCR process more, further researches are essential to investigate the reduction performance of the commercial grade Na_2CO_3 .

In these perspectives, the present studies are aimed to investigate the NO_x reduction characteristics of commercial grade urea in reducing NO_x from a diesel burning exhaust that is containing low ppm of base line NO_x and also to study the effect of commercial grade additive such as Na₂CO₃ on the urea based SNCR process.

1.3 Objectives and Scopes of the Present Studies

1.3.1 Objectives

- 1) To study the NO_x reduction behavior of urea based SNCR in diesel burning exhaust gas containing low ppm of initial NO_x.
- 2) To study the effect of inorganic additives on the urea based SNCR in terms of NO_x reduction performance.

1.3.2 Scope of the Studies

- 1) Design and fabrication of a pilot-scale combustion chamber for a small capacity industrial diesel burner in order to study the urea based SNCR.
- 2) Design and fabrication of an injection system for the aforesaid combustion chamber, which is capable of producing a wide range of droplet sizes varying the injection pressure and flow rate of the NO_x reducing agent.

- 3) Experimental studies so as to know the effect of commercial grade urea instead of laboratory one on the NO_x reduction characteristics with varying injection temperatures, atomizing pressures, concentrations of reagent, reagent injection rate and residence time.
- 4) Study the performance of the urea SNCR in the diesel burning exhaust and comparing the results obtained by previous researchers for the different fuels exhaust.
- 5) Study the performance of the urea SNCR in the diesel exhaust containing low value of initial NO_x .
- 6) Experimental studies to understand the effect of commercial grade inorganic additives such as Na_2CO_3 on the NO_x reduction performance of urea SNCR with varying concentrations of Na_2CO_3 in aqueous urea solution, temperatures of injection and residence time.
- 7) Determination of the level of ammonia slip formed as a byproduct during the SNCR application.

1.4 Thesis Outline

The thesis is completed in subsequent five chapters, which are organized as follows.

Chapter two gives a literature review, which includes the information about different NO_x formation mechanisms and different pre-combustion and post combustion technologies of NO_x control, while chapter three provides a review of present status of Selective Non-Catalytic Reduction of NO_x using urea and ammonia as NO_x reducing agents, effect of different operating parameters on NO_x reduction

efficiency and temperature window of SNCR and review of the effect of additives on NO_x reduction characteristics of SNCR process. Chapter four describes the details of the experimental set up and test procedures adopted. Results of the present studies are plotted in chapter five. This chapter also includes elaborate discussion of all the results comparing with the work of previous researchers. Finally, in chapter six, the conclusions of the findings of present studies are provided along with the proposal for future work necessary to be investigated in this particular area of research.

should be studied. This could be carried out by developing a kinetic modeling employing CHEMKIN or SENKIN chemical kinetic code.

Reagent-flue gas mixing has a significant effect on the NO_x reduction performance. In the present studies no such investigation was carried out. In the future some studies could be conducted in this potential area of SNCR application.

The present studies were concerned with low ppm of baseline NO_x , however the performance of commercial grade urea should also be studied in the effluent containing high ppm of baseline NO_x .

N_2O emission is a harmful byproduct of SNCR operation, which is usually more while urea is injected as reducing agent. In present studies it could not be studied due to the limitation of the gas analyzer, however in future it is strongly recommended that such test could be carried out in the present investigated conditions.

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