MODELING THE NO$_x$ EMISSIONS IN A LOW NO$_x$ BURNER
WHILE FIRED WITH PULVERIZED COAL AND DAIRY
BIOMASS BLENDS

A Thesis

by

HARI KRISHNA UGGINI

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

May 2012

Major Subject: Mechanical Engineering
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Approved by:

Chair of Committee, Kalyan Annamalai
Committee Members, Timothy Jacobs
Yassin Hassan
Head of Department, Jerry Caton

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ABSTRACT

Modeling the NO\textsubscript{x} Emissions in a Low NO\textsubscript{x} Burner While Fired with Pulverized Coal and Dairy Biomass Blends. (May 2012)

Hari Krishna Uggini, B.E (BITS-Pilani, India)

Chair of Advisory Committee: Dr. Kalyan Annamalai

New regulations like the Clean Air Interstate Rule (CAIR) will pose greater challenges for coal fired power plants with regards to pollution reduction. These new regulations plan to impose stricter limits on NO\textsubscript{x} reduction. The current regulations by themselves already require cleanup technology; newer regulations will require development of new and economical technologies.

Using a blend of traditional fuels and biomass is a promising technology to reduce NO\textsubscript{x} emissions. Experiments conducted previously at the Coal and Biomass Energy Lab at Texas A&M reported that dairy biomass can be an effective reburn fuel with NO\textsubscript{x} reduction of up to 95%; however little work has been done to model such a process with feedlot biomass as a blend with the main burner fuel. The present work concerns with development of a zero dimensional for a low NOx burner (LNB) model in order to predict NO\textsubscript{x} emissions while firing a blend of coal and dairy biomass. Two models were developed. Model I assumes that the main burner fuel is completely oxidized to CO, CO\textsubscript{2}, H\textsubscript{2}O and fuel bound nitrogen is released as HCN, NH\textsubscript{3}, N\textsubscript{2}; these partially burnt product mixes with tertiary air, undergoes chemical reactions specified by
kinetics and burns to complete combustion. Model II assumes that the main burner solid fuel along with primary and secondary air mixes gradually with recirculated gases, burn partially and the products from the main burner include partially burnt solid particles and fuel bound nitrogen partially converted to N₂, HCN and NH₃. These products mix gradually with tertiary air, undergo further oxidation-reduction reactions in order to complete the combustion. The results are based on model I. Results from the model were compared with experimental findings to validate it.

Results from the model recommend the following conditions for optimal reduction of NOₓ: Equivalence Ratio should be above 0.95; mixing time should be below 100ms. Based on model I, results indicate that increasing percentage of dairy biomass in the blend increases the NOₓ formation due to the assumption that fuel N compounds (HCN, NH₃) do not undergo oxidation in the main burner zone. Thus it is suggested that model II must be adopted in the future work.
ACKNOWLEDGEMENTS

I would like to thank my committee chair, Dr. Annamalai, and my committee members, Dr. Jacobs, and Dr. Yassin, for their guidance through the course of this research.

Thanks also to my friends in the Coal and Biomass Energy Laboratory who helped me along the development of my model. Also, I am thankful to Ben Lawrence for providing me experimental data required for validation of this model. I wish to express my thanks to my supervisor Ms. Yasuko Sakurai & the Utilities and Energy Management Texas A&M for partially funding my graduate studies.

And finally I dedicate this research to my parents & Viveka who have been unflinching in their support throughout.
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<th>Description</th>
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<tr>
<td>BTU</td>
<td>British Thermal Unit</td>
</tr>
<tr>
<td>CO</td>
<td>Carbon monoxide</td>
</tr>
<tr>
<td>CO2</td>
<td>Carbon dioxide</td>
</tr>
<tr>
<td>DB</td>
<td>Dairy Biomass</td>
</tr>
<tr>
<td>EGU</td>
<td>Electricity Generating Unit</td>
</tr>
<tr>
<td>EPA</td>
<td>Environmental Protection Agency</td>
</tr>
<tr>
<td>HCN</td>
<td>Hydrogen Cyanide</td>
</tr>
<tr>
<td>HHV</td>
<td>Higher Heating Value</td>
</tr>
<tr>
<td>LNB</td>
<td>Low NOx Burner</td>
</tr>
<tr>
<td>PRB</td>
<td>Powder River Basin</td>
</tr>
<tr>
<td>TAMU</td>
<td>Texas A&amp;M University</td>
</tr>
<tr>
<td>CABEL</td>
<td>Coal And Biomass Energy Laboratory</td>
</tr>
<tr>
<td>ER</td>
<td>Equivalence Ratio</td>
</tr>
<tr>
<td>VM</td>
<td>Volatile Matter</td>
</tr>
<tr>
<td>FC</td>
<td>Fixed Carbon</td>
</tr>
<tr>
<td>OFA</td>
<td>Over Fired Air (tertiary air)</td>
</tr>
<tr>
<td>RB</td>
<td>Reburn Zone</td>
</tr>
<tr>
<td>MB</td>
<td>Main Burner</td>
</tr>
<tr>
<td>PPM</td>
<td>Parts Per Million</td>
</tr>
<tr>
<td>EIA</td>
<td>Environment Information Agency</td>
</tr>
<tr>
<td>EPA</td>
<td>Environmental Protection Agency</td>
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<tr>
<td>DOE</td>
<td>Department Of Energy</td>
</tr>
<tr>
<td>SCR</td>
<td>Selective Catalytic Reduction</td>
</tr>
<tr>
<td>SNCR</td>
<td>Selective Non Catalytic Reduction</td>
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CHAPTER I
INTRODUCTION

Coal consumption in the power generation industry is a norm since it represents a steady supply in lieu of the vast reserves in the USA and it is also the cheapest available fossil fuel. According to EIA, Coal accounts for 43.1% of the total energy consumed for power generation. In the year of 2010 coal consumption in the power sector was to the tune of 1085.3 million short tons which is around 92% of the total coal consumption in the USA [1].

The combustion of coal, a solid fuel poses many challenges due to the pollution it creates. Coal combustion releases CO₂ to the tune of 90kg/GJ which aids in the phenomena of global warming EPA reports that nitrogen oxides are one of the major pollutants generated in the USA and a large fraction of it comes from coal fired power plants [2]. During the combustion of coal, there is formation of various pollutants like nitric oxides (NOₓ), sulfur dioxide (SO₂), mercury, fly ash and particulate matter which are released to the environment. Concerns over increased levels of air pollution and its harmful effects have resulted in stringent emission laws. In the year 2005 phase III of EPA’s CAIR program limited the NOₓ and SOₓ 0.11 lbm/MMBTU).

This thesis follows the style of Fuel.
The implementation of the Clear Air Interstate rule lowers the acceptable limits for average NO\textsubscript{X} to 0.11 lbm/MBtu by the year 2015 [3]. NO\textsubscript{X} is particularly harmful since they are one of the main ingredients involved in the formation of ground level ozone which can lead to serious respiratory problems. Besides they contribute to the formation of acid rains, deterioration of water quality and global warming [4]. Formation of NO\textsubscript{X} depends on a wide variety of factors like the type of fuel, stoichiometry, temperatures and time of residence. There are three main mechanisms of NO\textsubscript{X} formation in the gas phase namely Thermal NO\textsubscript{X}, Fuel NO\textsubscript{X} and prompt NO\textsubscript{X}.

Thus, extensive research has been conducted over several decades on understanding the formation and destruction of NO\textsubscript{X} and SO\textsubscript{X} & on control of emissions through used of various technologies. NO\textsubscript{X} reduction can be done through various processes like Selective Catalytic Reduction (SCR), Selective Non Catalytic Reduction (SNCR), Reburn with natural gas as reburn fuel and use of low NO\textsubscript{X} burners (LNB). The reburn process typically involves firing a small amount of natural gas at the end of combustion of fuel from the main burners. Utilization of natural gas as reburn fuel aids in the reduction of NO\textsubscript{X}.

Previous research at Coal and Biomass Energy Laboratory (CABEL) reported that CO2 neutral cattle biomass (CB) can be an effective reburn fuel for reduction of NO\textsubscript{X} [5]; literature review performed by Carlin has revealed that most of the boilers were replaced by low NO\textsubscript{X} burners due to the high cost of natural gas. In a low NO\textsubscript{X} burner air can be staged to reduce NO\textsubscript{X}. However, modeling has been limited for the reduction of NO\textsubscript{X} in a LNB and almost none while using a blend of dairy biomass (DB)
and coal as main burner fuel. Concentrated animal feeding operations result in accumulation of feedlot and dairy biomass which causes environmental concern. Cattle biomass can be beneficially used for energy extraction and NO\textsubscript{X} reduction. Thus it is of interest to study the effects of firing blended fuels in LNB to achieve NO\textsubscript{X} reduction.

**Description of a coal burner**

In primitive coal fired burners all the coal and air were injected together, this lead to high NO\textsubscript{X} emissions and hence were abandoned. In the modern day coal burner fuel is injected with carrier air (15-20\% of total air). Rest of the air required for combustion and to stabilize the flame is called secondary air. It is preheated to around 500K and is injected in a swirling motion to improve mixing characteristics. Modern burners incorporate a slightly rich combustion zone in the main burner to aid the reduction in formation of NO\textsubscript{X} and then Tertiary air (also called overfired air) is injected to complete combustion. A schematic of the modern day coal burner is as shown in Figure 1.1.
Figure 1.1 Schematics of a coal burner
CHAPTER II
LITERATURE REVIEW

This chapter shall provide a literature review of the main aspects of this work. Firstly, a general review on the theories about NO\textsubscript{X} formation is presented. Various NO\textsubscript{X} reduction techniques are presented next. Attention shall be paid to the various important parameters of NO\textsubscript{X} reduction such as equivalence ratios, mixing time and oxidation-reduction kinetics.

NO\textsubscript{X} formation

Combustion of hydrocarbons with air leads to the formation of many pollutants like oxides of nitrogen, carbon and Sulphur amongst many others. Of these oxides of nitrogen are particularly harmful. These oxides might be:

- Nitric oxide (NO)
- Nitrous oxide (N\textsubscript{2}O)
- Nitrogen dioxide (NO\textsubscript{2})

And they are collectively called with the generic term of NO\textsubscript{X}.

NO\textsubscript{X} formation can take place anywhere in the furnace but a majority of it is often formed in a small region. 10\% of the flame volume could account for almost 90\% of the NO\textsubscript{X} formation.

Coal combustion leads to insignificant amount of N\textsubscript{2}O and NO\textsubscript{2} and both represent small fractions of the NO\textsubscript{X} emissions. NO constitutes the largest fraction. Once in the atmosphere, NO combines with oxygen in air to readily form NO\textsubscript{2}. For
standardized reporting of NO\textsubscript{X} emissions on mass basis (g/GJ), EPA requires the use of molar mass of NO\textsubscript{2}.

There are a variety of factors which influence NO\textsubscript{X} formation. Some of them include amount of fuel burned, the stoichiometry, the temperatures, the mixing and the residence time. In the gas phase: Thermal NO\textsubscript{X} Fuel NO\textsubscript{X} and Prompt NO\textsubscript{X} are the three mechanisms for NO\textsubscript{X} formation.

Fuel NO is formed from the nitrogen contained in the fuel, and in the case of coal it can account for 75% of the total NO formed [6]. It is formed more readily than thermal NO as the bonds of nitrogen with coal or in the molecules emitted from coal (mainly HCN and ammonia) is much weaker than the triple bond of the molecular nitrogen present in the gas stream. Thus formation of fuel NO can be considered almost temperature independent.

Fuel nitrogen is normally emitted as molecular nitrogen, ammonia or HCN. Especially the last two species are the most significant, and their amount in the gas stream is a strong function of the kind of fuel [7]. In general high rank coals tend to emit most of their nitrogen as HCN, while low rank coals have also a significant fraction of ammonia [7]. It has been found that biomass emits a very large fraction of FN as ammonia [8]. These species then react in the gas phase and they could either decay to NO or N2, depending on the local stoichiometry, with more NO produced in the case of lean mixture.

Thermal NO\textsubscript{X} originates from the reaction of oxygen in the gas stream with nitrogen at high temperatures [7]. This pathway has a very strong dependence on the
temperature and on the oxygen concentration. This pathway can be described by the widely accepted two-step Zeldovich mechanism:

\[ N_2 + O \leftrightarrow NO + N \]  
(I.1)

\[ N + O_2 \leftrightarrow NO + O \]  
(I.2)

\[ N + OH \leftrightarrow NO + H \]  
(I.3)

The third reaction is particularly important under rich flame conditions where the OH radicals are present in higher concentrations than atomic hydrogen or oxygen.

At mean temperatures below 1800 K, thermal NO formation is very slow [9]. Figure 2.1 represents the thermal NOx equilibrium calculation for the combustion of methane according to the excess air provided [10]. It is noted that if the excess air is low, the NOx formation becomes significant only for temperature roughly above 1800 K.

Figure 2.1 Thermal NOx equilibrium calculations for methane at different excess air
In the case of coal flames, as flame temperature is normally below this threshold due to radiation and other heat losses, the thermal NOx formation is not very significant.

In the case of prompt NOx, nitric oxide can be formed when hydrocarbon fragments (CH, CH$_2$) resulting from devolatilization process attack molecular nitrogen near the reaction zone of the flame [9] particularly for rich mixtures.

The main reaction in this process is:

$$N_2 + CH \rightarrow HCN + N \quad \text{(I.4)}$$

$$CH + NO \rightarrow HCN + N_2 \quad \text{(I.5)}$$

Then HCN reacts with oxygen to create NO. Prompt NO is more significant in fuel rich flames since it needs hydrocarbon to initiate the chain of NO formation. Prompt NOx is normally considered the most significant in the case of clean fuels (that contain no nitrogen). In the case of coal combustion it is normally ignored [9]. Reaction I.b shows the Reverse prompt NOx mechanism which results in the decrease of NO since the CH fragments react with NO and reduce it to harmless N$_2$.

**Dairy biomass**

Dairy biomass (DB) is the waste from dairy animals which when not handled properly can impact the environment adversely. Concentrated animal feeding operations result in accumulation of nutrients in the ground which causes environmental concern. Diary biomass on a dry ash free basis has a heating value around 8500 BTU/lbm which is pretty close to heating values for a low grade coal (e.g. Texas lignite). Also diary
Biomass can be considered a clean fuel because the formation of food consumed by animals involves photosynthesis which reduces the amount of CO$_2$ in the air.

Thien [11], Carlin [5] and Lawrence [12] at CABEL have conducted extensive experiments on the effects of manure pollution and processing of dairy biomass for cofiring in a coal fired burner. The composition of animal feed, collection & storage of manure waste and their associated problems was extensively studied by [11]. Carlin [5] discusses the methods in use to dispose DB and reports problems like lagoon overflow and water source contamination due to the nutrients in manure leaching into the ground water.

Pennsylvania State University has conducted extensive experiments on cofiring blends of different bio-fuels on a circulating fluidized bed and concluded that when appropriate non fouling methods would not pose problems [13].

**NO$_X$ regulation**

Reduction of NO$_X$ emissions and providing cleaner air is one of the top motives for EPA. In March 2005 a new rule called Clean Air Interstate rule (CAIR) imposed tight regulation on NO$_X$ emission by electricity generating units (EGU’s) in 23 states including the state of Texas. CAIR regulates the NOx emissions from a particular state by placing a cap on the total NOx emissions. NOX generating units have a choice of alternatives to limit the NOx production. Some of which include: installing pollution control equipment, switching fuels and buying credits from sources which have reduced their pollution [14]. The targets for the state of Texas through the year 2015 have been
listed below in table 2.1. By 2015 CAIR will help Texas sources reduce emissions of nitrogen oxides (NOx) by 52,000 tons or 25% [3]

<table>
<thead>
<tr>
<th>NOX emissions (thousand tons)</th>
<th>2003</th>
<th>2009</th>
<th>2015</th>
</tr>
</thead>
<tbody>
<tr>
<td>Texas NOX emissions without CAIR</td>
<td>211</td>
<td>186</td>
<td>179</td>
</tr>
<tr>
<td>Texas NOX emissions with CAIR</td>
<td>N/A</td>
<td>167</td>
<td>159</td>
</tr>
</tbody>
</table>

The existing EPA limits on NOx emission [3] have been indicated in the Table 2.2 below.

<table>
<thead>
<tr>
<th>Unit built on or after Feb 2005</th>
<th>NOx emission limit (lbm/MMBTU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>New unit</td>
<td>1 lbm/MWH (gross energy output basis)</td>
</tr>
<tr>
<td>Reconstructed unit</td>
<td>0.11</td>
</tr>
<tr>
<td>Modified unit</td>
<td>0.15</td>
</tr>
</tbody>
</table>

One of the alternatives suggested is switching fuels. The use of dairy biomass as a constituent in Cofiring is one such option.
**NOX emission control**

The techniques to reduce NOx emissions can be in general divided into two categories: combustion control and post combustion control. In the combustion control the parameters of the combustion are optimized in order to avoid the formation of NOx.

One such technique is to lower the flame temperature as in this way the thermal NOx formation is directly affected. Another possible configuration is to create a fuel rich zone in the region with the maximum flame temperature: reducing the oxygen available the NOx formation can be directly reduced. Alternatively, NOx reduction can be achieved by lowering the residence time under oxidizing conditions. Combustion control systems such as fuel staging, reburning, flue gas recirculation, over-fire air and water / steam injection can provide substantial NOx reduction [15]. In the case of post combustion techniques, there is a dedicated cleanup process that takes place after the combustion [16]. These techniques can be further divided into Selective Catalytic Reduction (SCR, operational temperature 650 K) and Selective Non-Catalytic Reduction (SNCR, operational temperature 1100K). Clearly the difference between the two is the presence or not of a catalyst. Using SCR it is possible to achieve NOx reductions up to 90% [16]. The problem with SCR is the cost of catalysts, which have pushed the research to find new ways to gain high NOx reduction at lower costs.

Since the current study focuses on a Low NOX burner, we shall discuss it in further detail.
Low NO\textsubscript{X} burner

Latest power plants as well as retrofits of existing power plants utilize the design of Low NO\textsubscript{X} burners (LNB) to meet stricter emission regulations [17] describes its concept which focuses on staging air into two different paths, i.e. splitting the air into primary and secondary air flow to reduce to formation of NO\textsubscript{X} due to mixing of fuel bound nitrogen with more air at the top of the burner. This staging, delays the formation of NOx from fuel nitrogen. Also, this kind of control reduces the peak temperatures, which leads to reduction in the formation of thermal NO\textsubscript{X} within the flame. In an aerodynamically staged LNB the mixing of fuel with a portion of the required air for complete combustion is delayed to produce a flame with a relatively large fuel rich flame area within the flame[18]. Example of a LNB has been indicated in figure 2.2.

![Diagram of an advanced low NO\textsubscript{X} burner](image)

Figure 2.2 LNB example (adapted from [16])
A lower peak operating temperature is maintained in order to reduce NO\textsubscript{X} formation by limiting the amount of air available. This is also defined as staging. In the primary zone of combustion a fuel rich zone is established. Introduction of secondary air demonstrates well known usage of incomplete combustion to retard production of NO\textsubscript{X} from the burner [19]. The reduction in NO\textsubscript{X} formation takes place due to high levels of CO, CO\textsubscript{2}, H\textsubscript{2}O and fuel in the initial stage. The reaction rates for CO and CO\textsubscript{2} are much higher than the NO\textsubscript{X} formation kinetics due to the fact that NO\textsubscript{X} kinetics are slow, hence NO is formed in ppm; Thus enabling us to treat it as a trace species [10].

The formation of NO\textsubscript{X} from fuel nitrogen is based on the competition between the formation of NO\textsubscript{X} and the formation of N\textsubscript{2} from the nitrogenous species in the fuel volatiles. The staged combustion carefully controls the mixing of fuel and air which drives the reactions to form N\textsubscript{2} [18]. The low amount of oxygen available in the fuel rich zone leads to the formation of more N\textsubscript{2} in lieu of its faster kinetics. This increased nitrogen formed does not react with the oxygen provided in the secondary and tertiary zones due the high amount of energy required to break the triple bonds in the nitrogen molecule.

In some modern LNB burners operate the burners in slightly rich conditions, these burners also utilize over fired air (tertiary air) at a lower stage to complete combustion. This kind of setup is also known as globally staged combustion as shown in figure 2.3 [18].
Figure 2.3 NOx reduction area in LNB (adapted from [18])
Modeling of NOₓ emissions

A 1-D mixing model to predict reburn performance with natural gas as reburn fuel has been developed by [20]. This is a 1-D chemistry-mixing model that predicts reburning performance with natural gas as reburn fuel. A schematic of the facility modeled has been indicated in the Figure 2.4 below.

![Figure 2.4 Schematic of facility used in modeling [21]](image)

This model takes into account a detailed chemical mechanism of reactions in the reburning zone and simplified description of two mixing processes: reburning jets with flue gas and overfired air jets with flue gas. The model was also updated to include biomass fuels such as furniture waste. Previous modeling studies of reburning with solid
fuels demonstrated that relatively good results could be achieved using a homogeneous model with assumption of rapid decomposition of fuel particles into simple hydrocarbons. Previous studies also demonstrated that the presence of Na, K, and Fe compounds in the reburning zone significantly improved NO\textsubscript{x} control.

Though there is a lot of experimental data available on this setup, there has not been much modeling effort in the area of LNB with blended fuels. The current research is focused towards developing a zero dimensional model to determine the composition of gaseous species and temperature vs. time for globally staging combustion.
CHAPTER III

OBJECTIVE AND TASKS

The current research at Coal and Biomass Energy Laboratory (CABEL) is focused on the use of dairy biomass in a blend with (Permian River Basin) PRB coal. Previous experimental studies have shown that dairy biomass leads to extensive reduction of NO\textsubscript{X}. Dairy biomass being easily available near dairy farms has the economic advantages.

The objective of this current research is to develop a simplified zero dimensional model for the NO\textsubscript{X} emission in a LNB while firing a blend of coal & dairy biomass under rich conditions in the main burner of LNB and tertiary or overfired air introduction downstream of the main burner to complete the combustion. The following tasks shall be performed in order to achieve the overall objective

1. Obtain thermo chemical characteristics of Coal, Dairy biomass and the blends of both fuels.
2. Modify the existing reburn model for application to a LNB.
3. Determine species profiles for quantities of CO,CO\textsubscript{2},H\textsubscript{2}O,O\textsubscript{2},N\textsubscript{2},NH\textsubscript{3},HCN & NO as a function of time.
4. Conduct parametric studies to evaluate the effects of equivalence ratio of main burner, overall equivalence ratio, staging percentage of overfired air and percentages of DB in the blend effect the NO\textsubscript{X} emission.
5. Comparing the predicted NO emission predicted using the model with experimental findings to validate the results.
CHAPTER IV
MODELING

In this Chapter, typical operation of LNB will be described first followed by the experimental facility at Coal and Biomass Energy Laboratory (CABEL), TAMU developed by [17] shall be discussed first, while giving out details about its schematic, thermal rating etc. Next, modifications that have been made to the reburn model [15] to simulate the combustion in a LNB are detailed along with an explanation of the code.

Typical LNB

In actual LNB process, the fresh stream C with cold fuel particles of given size distribution and primary and secondary air enters the furnace and it gradually mixers with recirculating gases (RG; stream D) within recirculation zone (RZ). Thus the stream C which follows around the RZ gradually heats up the cold air along with solid particle, ignite and burn. Typically the ratio of mass flow of RG (stream D) to main air flow is a function of swirl number and is approximately 1 at Swirl # ≈1 [22]. This process has been indicated as shown in the Figure 4.1 below. Typically the main burner is fuel rich in order to reduce N from fuel to harmless N2 and as such there is incomplete combustion. Thus fresh premixed coal and primary and secondary air enter the control volume (CV1) and partially burnt gases leave as stream E. The stream E and stream F and tertiary or arm air (stream F) enters the CV2 and burnt products leave CV2 as stream G.
Figure 4.1 Actual LNB burner
Description of TAMU LNB facility

The experimental facility is a laboratory-scale, down-fired furnace, providing a rated output of 100,000 Btu/hr (29.3 kW), based on the higher heating value (HHV) of the fuel. This facility is used for testing the NO\textsubscript{x} formation while firing various solid fuels. A schematic of the lab scale facility has been indicated in figure 4.2 below.

![Lab scale facility schematic](image)

Figure 4.2 Lab scale facility schematic
Typical flow rates in the lab set up are

- Coal around 5 kg/hr
- Primary air around 100 lpm (15-20%)
- Secondary air around 500 lpm (75-85%)

The dimensions of the Low NO\textsubscript{X} burner air flow inlets are as shown in figure 4.3 below.

Figure 4.3 LNB design with primary, secondary and tertiary air flow
For a detailed description and dimensions of the Low NO\textsubscript{X} burner please refer to [17]. Fuel plus carrier or primary gas enters from the top along with secondary air which can be preheated. There are 11 thermocouples placed vertically down the furnace beginning 6 inches below the nozzle and spread 6 inches apart along the length of the Low NO\textsubscript{X} burner. An exhaust sampling port is located 66 inches below the nozzle which is connected to a gas analyzer. This analyzer measures the composition and provides a digital printed summary for amounts of different gases in the exhaust.

The main burner fires fuel in a rich atmosphere. Downstream, the product gases enter the overfired zone. Here the overfired air is injected in the furnace. The amount of air in the overfired zone can be varied to study its effects on the performances. The facility is equipped with extensive diagnostics to keep track of the temperature along the furnace and to measure the gas composition at the exit of the furnace. The LNB model schematic has been indicated in figure 4.4 below.
Reburn process

Reburn is one of the promising techniques for NO\textsubscript{X} reduction. During this process there are two different zones for combustion Main burner zone (typically lean) and reburn zone (typically rich). “In reburning, additional fuel (typically natural or gas, about 15 % of total fuel) is injected downstream from the primary combustion zone to create a fuel rich reburn zone where NO\textsubscript{X} is reduced through reactions with hydrocarbons (called reverse prompt NO\textsubscript{X} i.e. Reverse of reactions of HC fragments with N\textsubscript{2} to NO\textsubscript{X}). The nitrogen in the reburn fuel if any then recombines with oxygen to form NO\textsubscript{X}, or combines with N to form N\textsubscript{2}. After the reburn zone, additional air called over fire air is injected in the burnout zone to complete the combustion process”[10]. Reburn process has been indicated using the Figure 4.5 below.
Reburn model

Giacomo [15] developed a reburn model to predict NOx reduction when reburn process is used. The main burner gas temperature and compositions are known and this stream will be called Stream B while the reburn stream will be called stream A. Stream A in Giacomo model consists of a distributed solid fuel particles of different sizes and transported by carrier gas of known composition (e.g. air). This cold stream A mixes
gradually with hot stream B and the fraction of mass added from B to A is modeled with using an exponential relation.

Fraction mixing with stream A= \{1-\exp (-t/t_{mix})\}, where \(t_{mix}\) is the mixing time.

At \(t=0\) zero amount of stream B mixes with stream A; as \(t \to \infty\), whole stream B mixes with stream A and at this time mass flow in stream A increase to a sum of stream A and stream B and gases produced by combustion of fuel particles. As stream B mixes with A, stream A heats up which in turn hats the particles leading to pyrolysis, char oxidation and combustion. Reactions are considered in homogenous gas phase including NOx reactions. The results are report for change in concentration of species, temperature of mixed stream, burnt fraction etc. with time.

The species tracked are CO, CO\(_2\), H\(_2\), HCN, H\(_2\)O, N\(_2\), NH\(_3\), NO, and O\(_2\). All the species are tracked on the total mass basis and at each temporal step, the molar and mass concentration of the gas are computed knowing the total mass of each species. Events are tracked using a Lagrangian frame of reference; this means that the observer travels with the gas.

The choice of setting the observer as traveling with the stream A is called inverse mixing approach; alternatively it would have been possible to set the observer traveling with stream B. The choice of the inverse mixing approach depends on the fact that it was reported that this approach gives a more realistic description of the experimental data than the regular mixing [23] [20]. In the reburn model, energy conservation is used to solve for local temperature of gas stream. Assuming all the different gases to be ideal, the enthalpy function is a nonlinear function of the temperature alone. Knowing the
value of the enthalpy at some temperatures it is possible to set up enthalpy functions that interpolate the value of the enthalpy between the successive intervals, once the temperature of the gas species is specified. The values used are from [10]. The reburn model described above will be modified to predict combustion behavior of LNB. A schematic of the reburn model is as shown in figure 4.6 below.

![Schematic of Giacomo's reburn model](image)

Figure 4.6 Schematic of Giacomo's reburn model
During the mixing with the hot gases, the overfired air is heated up. This overfired air reacts with main burner gases and undergoes homogenous reactions in the gas phase.

The reactions include four homogeneous reactions involving NO, three homogeneous reactions for the oxidation of CO, H₂ and main burner fuel.

The code based on the model uses the following inputs:

- Main burner heat input, fuel characteristics, excess air, inlet temperature of fuel and air.
- Inlet temperature and composition of the carrier gas, homogeneous kinetics parameters, FN products composition and overall equivalence ratio.
- Percentage of total air staging.

Output of the code:

- Composition (Xₖ) of the gas phase in the free stream.
- The concentration of NO versus time.

**Reburn model simplification**

The model developed by [15] for the reburn process shall be simplified to match with the LNB model in the following manner. First primary and secondary air shall be clubbed together as carrier air which is injected along with the blend of coal and diary biomass. The tertiary air shall be used as an input for the reburn air in the reburn model. The product gas temperature can either be computed assuming a certain heat loss percentage or given as an input to the code. The NOx containing gases from the main burner, then gradually mix with the secondary air where a set of oxidation and reduction
reactions take place which lead to complete combustion and reduction in NO\textsubscript{X}. During the mixing with the hot gases, the overfired air is heated up. This overfired air reacts with main burner gases and undergoes homogenous reactions in the gas phase.

The reactions include four homogeneous reactions involving NO, three homogeneous reactions for the oxidation of CO, H\textsubscript{2} and main burner fuel.

The code based on the model uses the following inputs:

- Main burner heat input, fuel characteristics, excess air, inlet temperature of fuel and air.
- Inlet temperature and composition of the carrier gas, homogeneous kinetics parameters, FN products composition and overall equivalence ratio.
- Percentage of total air staging.

Output of the code:

- Composition (X\textsubscript{k}) of the gas phase in the free stream.
- The concentration of NO versus time.

The species tracked are: Main burner fuel, CO, CO\textsubscript{2}, H\textsubscript{2}, HCN, H\textsubscript{2}O, N\textsubscript{2}, NH\textsubscript{3}, NO, and O\textsubscript{2}. All the species are tracked on the total mass basis and at each temporal step, the molar and mass concentration of the gas are computed knowing the total mass of each species. Events are tracked using a Lagrangian frame of reference; this means that the observer travels with the gas from the overfired air zone, and the mass tracked increases as the flow from the main burner mixes with the flow from the overfired zone, and the composition of the different species changes according to the various reactions taking place.
The choice of setting the observer as traveling with the overfired zone air is called inverse mixing approach; alternatively it would have been possible to set the observer traveling with the main burner products. The choice of the inverse mixing approach depends on the fact that it was reported that this approach gives a more realistic description of the experimental data than the regular mixing [23]. In the model, energy conservation is used to solve for local temperature of gas stream. Assuming all the different gases to be ideal, the enthalpy function is a nonlinear function of the temperature alone. Knowing the value of the enthalpy at some temperatures it is possible to set up enthalpy functions that interpolate the value of the enthalpy between the successive intervals, once the temperature of the gas species is specified. The values used are from [10].

**Zero D model for LNB**

The reburn model by [15] will be modified to create a Zero Dimensional model to predict combustion behavior in a LNB. In order to do this a list of assumptions made are listed below.

**Assumptions**

The assumptions are summarized as follows:

**Main burner:**

- NO in the main burner is generated only by the decomposition of fuel bound nitrogen.
- The combustion at the main burner is complete and no dissociation is considered among its products.
Overfired zone:
- The mixing between air in the overfired zone and the main burner gases is described by an exponential model.

Gas phase:
- All the gases are treated as ideal gases.
- The species are constantly perfectly mixed at any given time.

Chemical reactions:
- All the reactions are described by simplified kinetics.
- In case detailed kinetics are not available for biomass, lignite kinetics are valid for biomass.
- Gases coming from the particle mix instantaneously with the free stream of gas at each temporal step.

Energy conservation:
- Energy transfer is at quasi steady state.
- Gas mixing processes are isenthalpic.

General:
- The interior of the furnace is at atmospheric pressure.

In order to simulate the Low NOx burner (LNB) two different approaches have be thought of, named LNB model I and LNB model II. LNB model I is used extensively, while model II can be used to more accurately predict NOx in a LNB.
LNB model I

The control volume (CV 2) considered in this model has been indicated in the Figure 4.7 below.

Figure 4.7 Stratified Burner for NOx reduction for gas fired combustors: Fuel jet in fuel gun surrounded by primary, secondary and tertiary air (adapted from [18])

In order to use reburn model, main burner gas in reburn model will be termed as stream A in LNB model and the reburn gases in reburn model will be termed as stream B in LNB model. Mass flow in the reburn gases will gradually increase due to mixing of main burner gases.

In this model the modifications we need to make are: the stream-B will be same as main burner gases in reburn model and gas composition corresponds to incomplete
combustion of all solid fuels to CO, CO\textsubscript{2} and H\textsubscript{2}O but with production of all fuel bound nitrogen into HCN, NH\textsubscript{3} and N\textsubscript{2} from FB nitrogen and arbitrarily assigned NO. The stream A for LNB model will be same as tertiary air; It is the same as reburn air and negligible fuel in reburn model [15]. The mass flow in stream A gradually increases due to mixing of stream B using the Lagrangian frame of reference discussed earlier. However such an assumption seems to be oversimplified since there is insufficient tie between main burner and tertiary port in LNB and thus combustion may be incomplete.; Figure 4.8 shows a schematic of the overfired zone.

![Figure 4.8 Schematic of the overfired zone LNB model I](attachment://image.png)
Hence model II is also thought of as an improvement over the model I to predict NOx emissions.

**LNB model II**

So in LNB model II, the first run-Run I simulates the Control volume CV 1 and will assume the stream C in LNB will be stream B of the reburn model and solid fuel flow is carried by stream C. Stream D, the recirculating gases will be stream A in the reburn model. Thus stream C entrains gases from stream D and hence heats up. Gas composition corresponds to combustion of all solid fuels to CO, CO₂, H₂O, N₂, and O₂ (if any). At end of \( t = \frac{L}{V} \), \( v \): velocity of gases) the time of travel for stream C to reach tertiary air port. The mass flow of stream C gradually increases and the mass flow just before end of recirculation point E will be sum of gaseous mass of stream C+ gases produced by solid particles and mass of recirculated gases. The mass flow at port E will be sum of gaseous mass of stream C+ gases produced by solid particle since apart of the gaseous mass has been recirculated.

This will be followed by run II to simulate the control volume CV 2. For run II, stream E in LNB will be stream A in reburn model with input of gas composition, the unburnt particles with remaining VM, FC and fuel N. Stream F will be stream B in reburn model with very small equivalence ratio so that fuel input is reduced to almost zero. Completely burnt products leave a stream G.

The output of this model will now be able to give us a better picture of the volatiles remaining, temperature distribution of the different size groups and FC remaining. However results in the current thesis will cover model I only.
LNB model I: Main burner

The main burner fuel is assumed to be represented by the formula $CH_xO_yN_z$ which is burned along with some NH$_3$ to simulate the desired amount of NO. The amount of ammonia to be fired with the fuel is adjusted in order to achieve the desired amount of NO.

The solution for incomplete combustion of a general fuel is:

$$CH_xO_yN_z + w \cdot NH_3 + \left(1 + \frac{a}{100}\right) \left(1 + \frac{x}{4} - \frac{y}{2}\right) \cdot O_2 + 3.76 \cdot N_z \rightarrow \left((-a/50)^* (1 + x/4 - y/2) - 1.95^* z\right)CO + ...$$

$...((1 + a/50)^*(1 + x/4 - y/2) + 1.35^* z)CO_2 + \left(\frac{x}{2} + \frac{3w}{2}\right)H_2O + (0.6^* z)HCN + (0.3^* z)w \cdot NO + ...$

$$...\left[0.1^* z/2 + 3.76 \cdot \left(1 + \frac{a}{100}\right) \left(1 + \frac{x}{4} - \frac{y}{2}\right)\right]N_2$$

where ‘$a$’ is the percentage excess of air based on the main burner stoichiometric air requirement. ‘$w$’ is the finite NOx for the reburn application and is now set to an arbitrary value for the LNB application. The fuel bound nitrogen is assumed to split into HCN, NH$_3$ and N$_2$ in the ratio 60:30:10 respectively [24]. The coefficients of the empirical formulae of the blend $x$, $y$, $z$ change as one blends biomass with coal. Also the split of biomass N is assumed to be HCN, NH$_3$, and N$_2$ in the ratio of 30:60:10 respectively. Thus amount of HCN, NH$_3$ and N$_2$ released with depend on the percentage of biomass in the blend.

This formula has been obtained with the atom balance of the species of the products and reactants. With this formula, it is possible to know the composition of the gas leaving the main burner zone.
Now the amount of air to be injected in the main burner fuel can be calculated and also the composition of the products coming from the main burner is known. Using the overall equivalence ratio and the staging percentage we can determine the amount of air to be injected into the overfired zone.

As the thermal power coming from the main burner is fixed (29.3 KW), it is possible to compute the mass flow of the main burner fuel:

\[
\dot{m}_{fuel MB} = \frac{Thermal Rating_{MB}}{HHV_{fuel MB}} \begin{bmatrix} kg \\ s \end{bmatrix}
\]

The components of the various species from the main burner are represented in vector form as:

\[
\vec{m}_{MB} = \frac{\dot{m}_{fuel MB}}{M_{fuel MB}} \begin{bmatrix} 0 \\ ((-a/50)*(1+x/4-y/2)-1.95z)*M_{CO} \\ (1+(a/50)*(1+x/4-y/2)+1.35z)\cdot M_{CO} \\ 0 \\ (0.6* z)_*M_{HCN} \\ (x/2-0.75* z + 3w/2)_*M_{H_2O} \\ [0.1* z/2 + 3.76\cdot((1+a/100)\cdot(1+x/4-y/2))]\cdot M_{N_2} \\ (w+0.3* z)_*M_{NH_3} \\ w\cdot M_{NO} \\ 0 \end{bmatrix} = \begin{bmatrix} \dot{m}_{MB, f} \\ \dot{m}_{CO} \\ \dot{m}_{CO_2} \\ \dot{m}_{H_2} \\ \dot{m}_{HCN} \\ \dot{m}_{H_2O} \\ \dot{m}_{N_2} \\ \dot{m}_{NH_3} \\ \dot{m}_{NO} \\ \dot{m}_{O_2} \end{bmatrix} \begin{bmatrix} kg \\ s \end{bmatrix}
\]

The temperature of the gases leaving the main burner zone can be computed by applying the energy conservation equation between the products and the reactants and considering a fraction of heat to be lost, proportional to the heating value of the main burner fuel.
\[ \dot{H}_{in,MB} = \dot{H}_{out,MB} + Q_{lost} \]

where the total enthalpy \( H \) is rate per unit time.

\[
\frac{\dot{H}_{in,MB}}{\dot{m}_{fuel}} = h_{f,\text{fuel}} + h_{r,\text{fuel}} + \frac{\dot{m}_{air,MB}}{\dot{m}_{fuel}} \cdot h_{r,air} \left[ \begin{array}{c} kJ \\ \text{kg}_{MB,\text{fuel}} \end{array} \right]
\]

and

\[
H_{out,MB} = \sum_{i=1}^{\text{species}} \frac{\dot{m}_{i}}{\dot{m}_{fuel}} \cdot (h_{f,i} + h_{r,i}) \left[ \begin{array}{c} kJ \\ \text{kg}_{MB,\text{fuel}} \end{array} \right]
\]

The enthalpies of formation are fixed while the thermal enthalpies are non-linear functions of the products’ temperature; therefore this equation needs to be solved in implicit form. The enthalpy of formation of the fuel is computed from its heating value and considering its complete combustion with air:

\[
h_{f,\text{fuel}} = \frac{\bar{h}_{f,CO_2} + x/2 \cdot \bar{h}_{f,H_2O}}{MW_{fuel}} + HV_{MB} \left[ \begin{array}{c} kJ \\ \text{kg}_{MB,\text{fuel}} \end{array} \right]
\]

It is difficult to quantify the heat loss in the main burner; if the temperature of the products of combustion of the main burner fuel is known, it is possible to specify it directly: this is the case used in this study, the previous case has been taken in consideration in order to make the model more general and usable also in case the temperature was not known.

The composition of the products is known, so also the mass flow rate of every species is known. It is noted that model I presumes that there are no particles at the end of combustion in the main burner zone and the products contain CO, CO_2, HCN, NH_3 and N_2.
LNB model I: Overfired zone modeling

The mass of each species varies over time as some species are produced and others are consumed; therefore the data of the masses of the gas phase is stored in a matrix, in which the rows correspond to the species \( i \) and the columns correspond to a certain temporal step \( t \).

\[
\text{Time} = \left\{ 0, \ldots, T, \ldots, T_{\text{fin}} \right\}
\]

\[
\text{Total mass} = \left\{ \begin{array}{ccc}
  m_{\text{CH}_4} & m_{\text{CH}_4} & m_{\text{CH}_4} \\
  m_{\text{CO}} & m_{\text{CO}} & m_{\text{CO}} \\
  m_{\text{CO}_2} & m_{\text{CO}_2} & m_{\text{CO}_2} \\
  m_{\text{H}_2} & m_{\text{H}_2} & m_{\text{H}_2} \\
  m_{\text{H}_2\text{O}} & m_{\text{H}_2\text{O}} & m_{\text{H}_2\text{O}} \\
  m_{\text{N}_2} & m_{\text{N}_2} & m_{\text{N}_2} \\
  m_{\text{NH}_3} & m_{\text{NH}_3} & m_{\text{NH}_3} \\
  m_{\text{NO}} & m_{\text{NO}} & m_{\text{NO}} \\
  m_{\text{O}_2} & m_{\text{O}_2} & m_{\text{O}_2}
\end{array} \right\}
\]

LNB model I: Mixing model

The mixing of the overfired air with the main burner exhaust is a very important part of the staging process; therefore it must be modeled carefully. Assuming the mixing to be instantaneous is far from reality, as this process takes time to be completed; besides, previous work [20] has shown that the assumption of instantaneous mixing is a bad depiction of reality and leads to poor results. In this case the mixing of the overfired air with the main burner gas is described using an exponential model [23]; an alternative finite mixing model would be the linear mixing [20]. More in details, an inverse mixing model (main burner gases into overfired air: which means setting the observer traveling
with the overfired air) is used as it has been shown [20] that it leads to better results than direct mixing (overfired air into main burner gases).

With respect to an observer traveling with the air in overfired zone, the total mass will be composed of the overfired air and a fraction of the main burner mass that is added gradually over time, and will approach a total mass equal to the sum of the overfired air and main burner gases.

Considering exponential mixing model, the mass flow in the overfired zone due to mixing with main burner gases is:

$$
\dot{m}_{OFZ,t} = \dot{m}_{OFZ,t=0} + \dot{m}_{prod,MB} \cdot \left(1 - \exp\left(\frac{-t}{\tau_{mix}}\right)\right)
$$

The equation above satisfies the initial (t→0) and final (t→∞) condition. The mixing time \(\tau_{mix}\) depends on the geometry of the furnace and the overfired air velocity. It is estimated from experimental data for the furnace and air injection configuration used for the experiment. \(\tau_{mix}\) is estimated to be around 40ms [25]. In the discussion of the results from the simulation, it is shown that reasonable variations of this constant will not affect significantly the NO\(_x\) reduction, which is the most important parameter of this simulation and, most importantly, will hardly change the qualitative trend.

This is in agreement with what found by Lissanski [20]. The value of the mixing time is most critical at small values (close to the transition between instantaneous mixing and finite – rate addition of reagents); at higher values of \(\tau_{mix}\), its variations affect less the NO reduction. It is clear that as \(t\) increases the total mass seen by the observer increases.
The elemental amount of mass coming from the main burner that will be added over a period of time \( dt \) is given as:

\[
dm_{MB} = \frac{\dot{m}_{prodMB}}{\tau_{mix}} \cdot \exp\left( -\frac{t}{\tau_{mix}} \right) \cdot dt
\]

The term \( dm_{MB} \) is a vector and contains the contribution of every gas species, and as well as contributes thermal energy to RB gases; the elemental mass \( dm \) decreases as time progresses as less and less mass is left to be mixed.

Since the composition of the gas coming from the main burner is known, it is possible to determine the quantity of each species at each temporal step of integration (considering only the contribution from the mixing process).

\[
\dot{m}_{OFA,t+1} = \dot{m}_{OFA,t} + \dot{m}_{MB,t} = \begin{pmatrix}
\dot{m}_{MB,t} \\
\dot{m}_{CO} \\
\dot{m}_{CO_2} \\
\dot{m}_{H_2} \\
\dot{m}_{HCN} \\
\dot{m}_{H_2O} \\
\dot{m}_{N_2} \\
\dot{m}_{NH_3} \\
\dot{m}_{NO} \\
\dot{m}_{O_2}
\end{pmatrix}_{OFA,t} + \begin{pmatrix}
\dot{m}_{MB,t} \\
\dot{m}_{CO} \\
\dot{m}_{CO_2} \\
\dot{m}_{H_2} \\
\dot{m}_{HCN} \\
\dot{m}_{H_2O} \\
\dot{m}_{N_2} \\
\dot{m}_{NH_3} \\
\dot{m}_{NO} \\
\dot{m}_{O_2}
\end{pmatrix}_{MB} \cdot \exp\left( -\frac{t}{\tau_{mix}} \right) \cdot dt
\]

**LNB model I: Chemical reactions**

In order to reduce the computational effort, a simplified kinetics model has been adopted. The homogeneous reactions are the reactions that take place in the gas phase; for these reactions the species concentrations are directly computed knowing the composition of the gas phase stream.
**NO reactions**

A widely used model, for reduced NO reactions in the reburn process, is the one formulated by De Soete [26]; However, the simulations based on his kinetics have brought unsatisfactory results, especially with pure biomass or a blended fuel with a high content of biomass. Further the De Soete’s kinetics have been formulated based on data points at temperature mostly above 2000 K, while in this work, the temperatures are of the order of 1500 K. So the two reaction rates from De Soete regarding ammonia will be substituted with the recent data by [27], which have been developed to describe the oxidation of volatile nitrogen in biomass combustion. The two reaction rates by De Soete regarding HCN will be substituted with the ones by [28] that are a very slight modification on De Soete’s ones.

\[ \text{I}_N \quad \text{Ammonia oxidation [27].} \]

\[ \text{NH}_3 + O_2 \rightarrow NO + H_2O + 0.5 \cdot H_2 \]

\[ \dot{w}_{\text{NH3},i} = -1.21 \cdot 10^{11} \cdot T_g^2 \cdot [\text{NH}_3] \cdot [O_2]^{3/5} \cdot [H_2]^{2/5} \cdot \exp\left(\frac{-8000}{T_g}\right) \left[ \text{kmol} m^{-3} \cdot s \right] \]

\[ \text{NH}_3 + NO \rightarrow N_2 + H_2O + 0.5 \cdot H_2 \]

\[ \dot{w}_{\text{NH3},ii} = -8.73 \cdot 10^{14} \cdot T_g \cdot [\text{NH}_3] \cdot [NO] \cdot \exp\left(\frac{-8000}{T_g}\right) \left[ \text{kmol} m^{-3} \cdot s \right] \]

\[ \text{III}_N \quad \text{HCN oxidation [28]} \]

\[ \text{HCN} + O_2 \rightarrow NO + CO + 0.5 \cdot H_2 \]

\[ \dot{w}_{\text{HCN,iii}} = -10^{11} \cdot X_{\text{HCN}} \cdot X_{O2}^b \cdot \frac{P}{R \cdot T_g} \cdot \exp\left(\frac{-280328}{R \cdot T_g}\right) \left[ \text{kmol} m^{-3} \cdot s \right] \]
IV_{N} \quad \text{HCN reduction [28]}

\begin{align*}
\text{HCN} + \text{NO} & \rightarrow \text{N}_2 + \text{CO} + 0.5 \cdot H_2 \\
\dot{w}_{\text{HCN,IV}_N} & = -3 \cdot 10^{12} \cdot X_{\text{HCN}} \cdot X_{\text{NO}} \cdot \frac{P}{R \cdot T_g} \cdot \exp \left( \frac{-251000}{R \cdot T_g} \right) \left[ \frac{\text{kmol}}{m^3 \cdot s} \right]
\end{align*}

The $b$ exponent (used in reaction IV_N) is calculated by a curve fit from the experimental data from [26].

$$b = \begin{cases} 
0 & \text{if } \ln X_{O_2} \geq -3 \\
233 \cdot \exp \left( \frac{-28}{0.5 + \ln X_{O_2}} \right) & \text{if } -5.67 < \ln X_{O_2} < -3 \\
1 & \text{if } \ln X_{O_2} \leq -5.67
\end{cases}$$

**Gas phase homogenous oxidation reactions**

These are other homogenous reactions that are taking place in the gas phase, in which NO is not involved.

I_{G} \quad \text{CO oxidation [29].}

\begin{align*}
\text{CO} + 0.5 \cdot O_2 & \rightarrow \text{CO}_2 \\
\dot{w}_{\text{CO,I}_G} & = -1.3 \cdot 10^{17} \cdot [\text{CO}] \cdot [O_2]^{0.5} \cdot [H_2O]^{0.5} \cdot \exp \left( \frac{-125580}{R \cdot T_g} \right) \left[ \frac{\text{kmol}}{m^3 \cdot s} \right]
\end{align*}

II_{G} \quad \text{H}_2 \text{ oxidation [30].}

\begin{align*}
\text{H}_2 + 0.5 \cdot O_2 & \rightarrow \text{H}_2O \\
\dot{w}_{\text{H}_2,II_G} & = -0.68 \cdot 10^{19} \cdot \left( \frac{Y_{H_2}}{2} \right)^{0.25} \cdot \left( \frac{Y_{O_2}}{32} \right)^{1.5} \cdot \rho_g^{1.75} \cdot \exp \left( \frac{-20130}{R \cdot T_g} \right) \left[ \frac{\text{kmol}}{m^3 \cdot s} \right]
\end{align*}

From the stoichiometry of the reactions, it is possible to compute the reaction rates of each species $k$:

$$\dot{n}_{\text{homoreact}} = \sum_{k=1}^{\text{homoreact}} v_{i,k} \cdot w_i \cdot \frac{R_{tk} \cdot m_{\text{TOT,gas}} \cdot T_g}{p} \left[ \frac{\text{kmol}}{s} \right]$$
Where $\nu_{i,k}$ is the stoichiometric coefficient of species $i$ in homogeneous reaction $k$, and it is positive if the species is being produced and negative if the species is being consumed. It is zero if the species $i$ does not appear in the reaction $k$. Knowing the molecular weight of each species, it is possible to compute the mass variation rate.

$$\dot{m}_{\text{hom},i} = \dot{n}_{\text{hom},i} \cdot M_i \quad \left[ \frac{kg}{s} \right]$$

More details on heating up of particles, kinetics of pyrolysis, heterogeneous oxidation of carbon and homogenous reactions are provided in [15].
CHAPTER V

RESULTS AND DISCUSSION

This chapter presents the results from the LNB model I. All input parameters are first listed. The output of the code shall be used to compare it with experimental data on overfiring/arm firing obtained from experiments conducted on Cofiring PRB coal and diary biomass. Also parametric studies are conducted on various parameters like mixing time ($t_{mix}$), overall equivalence ratio ($\Phi$), chemical kinetics constants, main burner equivalence ratios, overall equivalence ratio and percentage overfired air.

Fuel properties

The Ultimate and Proximate analysis for Powder River basin (Coal) and Dairy biomass (DB) coal adapted from [12] are as indicated in the Table 5.1 below

<table>
<thead>
<tr>
<th></th>
<th>Dairy Biomass</th>
<th>PRB coal</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Proximate Analysis</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry loss (% moisture)</td>
<td>25.26</td>
<td>32.88</td>
</tr>
<tr>
<td>Ash</td>
<td>14.86</td>
<td>5.64</td>
</tr>
<tr>
<td>FC</td>
<td>13.00</td>
<td>32.99</td>
</tr>
<tr>
<td>VM</td>
<td>46.88</td>
<td>28.49</td>
</tr>
<tr>
<td><strong>Ultimate Analysis</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon, C</td>
<td>35.21</td>
<td>46.52</td>
</tr>
<tr>
<td>Hydrogen, H</td>
<td>3.71</td>
<td>2.73</td>
</tr>
<tr>
<td>Nitrogen, N</td>
<td>1.93</td>
<td>0.66</td>
</tr>
<tr>
<td>Oxygen, O</td>
<td>18.60</td>
<td>11.29</td>
</tr>
<tr>
<td>Sulfur</td>
<td>0.43</td>
<td>0.27</td>
</tr>
<tr>
<td>HHV (kJ/kg) as received</td>
<td>12844.17</td>
<td>18193.02</td>
</tr>
<tr>
<td>HHV (kJ/kg) Dry</td>
<td>17185.90</td>
<td>27106.57</td>
</tr>
</tbody>
</table>
HHV(kJ/kg) DAF | 21449.85 | 29508.00
---|---|---
Chemical formula | $\text{CH}_{1.255424}\text{N}_{0.046999}\text{O}_{0.396524}\text{S}_{0.004573}$ | $\text{CH}_{0.699206}\text{N}_{0.02165}\text{O}_{0.18217}\text{S}_{0.002174}$
FN distribution $\text{N}_2$:HCN:NH$_3$ | 1:3:6 | 1:6:3

**Data input for model I**

Tables 5.2 and 5.3 below show the input data for the main burner zone and the overfired zone.

**Table 5.2 Main burner input**

<table>
<thead>
<tr>
<th>Fuel</th>
<th>PRB coal, PRB and DB blend</th>
</tr>
</thead>
<tbody>
<tr>
<td>Main Burner Power</td>
<td>29.3kW (100000 BTU/hr)</td>
</tr>
<tr>
<td>HHV (PRB Coal)</td>
<td>29809 kJ/kg</td>
</tr>
<tr>
<td>Percentage of excess air in main burner</td>
<td>-33.81 to 3.81</td>
</tr>
<tr>
<td>Inlet temperature of primary air and fuel</td>
<td>300 K</td>
</tr>
<tr>
<td>Temperature of gases from main burner</td>
<td>1500 K</td>
</tr>
</tbody>
</table>

Input data in the overfired zone

**Table 5.3 Overfired zone input**

<table>
<thead>
<tr>
<th>Overall equivalence ratio</th>
<th>0.85-1.05</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inlet temperature of overfired air</td>
<td>300 K</td>
</tr>
<tr>
<td>Mixing time</td>
<td>50 ms</td>
</tr>
</tbody>
</table>
The relevant kinetics data are illustrated in table 5.4 below

Table 5.4 Homogenous reaction kinetics

<table>
<thead>
<tr>
<th>Reaction</th>
<th>A(m³-kmol-s)</th>
<th>E(kJ/kmol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I_N</td>
<td>1.21*10¹¹</td>
<td>66500</td>
</tr>
<tr>
<td>II_N</td>
<td>8.73*10²⁰</td>
<td>66500</td>
</tr>
<tr>
<td>III_N</td>
<td>10¹¹</td>
<td>28000</td>
</tr>
<tr>
<td>IV_N</td>
<td>3*10¹²</td>
<td>251000</td>
</tr>
<tr>
<td>I_G</td>
<td>6.8*10¹⁸</td>
<td>20130</td>
</tr>
<tr>
<td>II_G</td>
<td>5.74*10¹⁰</td>
<td>60000</td>
</tr>
</tbody>
</table>

The reactions are numbered as indicated previously in the modeling section.

Algorithm of the model

Given below is an algorithm on the mixing model. After the above data has been entered gases at the exhaust of the main burner are computed. Integration over time can then start, which determines the composition and temperature of gas particles. The loop ends when the simulation time is reached. Algorithm of the model has been shown in figure 5.1
Where $t_{res}$ is the prescribed residence time.
The choice of temporal step plays an important role in results, a large time step might lead to errant results while reducing the computational time. On the other hand a small computation step would lead to increase in run time. A statistical way to determine if a particular time step is small enough is to compute the difference between the solutions from the current time step and its half. If the variation is small then our solution has converged, else we need to pick a smaller time step. This study has been conducted and found that at 0.025 seconds, the difference in the results becomes negligible [15]. Hence this value has been used in the model for all cases.

**LNB model I results**

The NO (ppm) at end of the burner for Pure PRB coal have been presented in figure 5.2 below. The NO (ppm) at the end of the burner reduces greatly as overall equivalence ratio is increased to a value greater than 0.95, this observation is on par with theoretical expectations that rich conditions lead to lower NO.
Figure 5.2 Pure PRB NO vs. overall ER
The oxygen concentration along the burner for pure PRB at 10% overfired air while varying equivalence ratio has been indicated in Figure 5.3 below.

![Oxygen concentration Vs. Time step](image)

Figure 5.3 Oxygen concentration along the burner for pure PRB vs. overall equivalence ratio at 10% staging

As we can observe the oxygen concentration along the furnace reduces with increasing overall equivalence ratio, since there is lesser amount of air available.
LNB model I validation

Ben Lawrence had conducted experiments with overfired/arm fired air using PRB coal & blends of PRB coal and DB at CABEL, TAMU. The experimental data uses the term arm-firing which is the same as overfiring used in the current thesis. The base case for this validation has been chosen at 10% staging for the pure PRB firing while varying the equivalence ratio. NO at the end of the burner observed during the experiments and those obtained from the model have been compared in the figure 5.4 below.

![Comparison of Experimental & Model results](image)

Figure 5.4 Comparison of experimental and model results for pure PRB & 10% staging
As we can see from the above graph, model results show that NO\textsubscript{X} formation reduces as we increase the overall equivalence ratio. The results follow a trend similar to the ones obtained from the experiment, but values for the lean mixtures are higher than those experimentally observed. Both results indicate that NO\textsubscript{x} produced is at its lowest when we have an overall equivalence ratio greater than 1.

**NO concentration along the burner**

NO and O\textsubscript{2} mass at each time step along the burner for pure PRB coal at ER=0.95 and 10% staging have been plotted in figure 5.5 below.

![NO and O\textsubscript{2} mass at each time step](image)

Figure 5.5 NO and O\textsubscript{2} concentration along the burner for pure PRB at ER=0.95 and 10% staging
To observe the phenomena closely the profiles for CO\textsubscript{2} and HCN are indicated in the Figure 5.6 below.

![Figure 5.6 CO\textsubscript{2} and HCN concentration for pure PRB 10\% staging ER=0.85](image)

Figure 5.6 CO\textsubscript{2} and HCN concentration for pure PRB 10\% staging ER=0.85

From graphs 5.5 and 5.6 we can see oxygen is being primarily consumed for oxidation of CO. Since we assumed fuel nitrogen release as HCN and NH\textsubscript{3} in the partially burnt products of the main burner the NO gradually increases due to the oxidation of NH\textsubscript{3} and HCN. The presence of NH\textsubscript{3} and HCN is in trace ammounts and hence it does not affect the profile of O\textsubscript{2}.
**Blended fuel results**

Model was run for PRB-DB 95-5 blend at various equivalence ratios for fixed staging percentages. The results obtained are as shown in Figure 5.7 below.

![Variation of PRB-DB NO vs Eq. ratio at 10% staging](image)

Figure 5.7 PRB vs. ER for different staging percentages
The dissociation of fuel bound nitrogen into species HCN, NH₃ and N₂ is different for both fuels. This has been indicated in the Table 5.5 below.

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Empirical Formula</th>
<th>N₂:HCN:NH₃ Percentage Dissociation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dairy biomass</td>
<td>CH₁.₂₁₅₄₂₄N₀.₀₄₆₉₉₉O₀.₃₉₆₅₂₄S₀.₀₀₄₅₇₃</td>
<td>10:30:60</td>
</tr>
<tr>
<td>PRB coal</td>
<td>CH₀.₆₉₉₂₀₆N₀.₀₂₁₆₅O₀.₁₈₂₁₇S₀.₀₀₂₁₇₄</td>
<td>1:60:30</td>
</tr>
</tbody>
</table>

As observed, more ammonia is released from Dairy biomass 60% than PRB coal 30%. Ammonia readily oxidizes into NO due to its faster kinetics and hence more NO is formed when blend percentage with Dairy biomass is increased.
For the 90% PRB coal-10% Dairy biomass blend while varying the equivalence ratios from 0.85-1.05 the following results have been found as shown in Figure 5.8 below.

![Variation of NO for PRB-DB 95-5 blend vs Eq. ratio](image)

**Figure 5.8 Variation of NOx vs. ER for 95-5 PRB-DB blend**

There are two effects: temperature and O\textsubscript{2}. When equivalence ratio is increased, less air is sent to main burner and as well as cold tertiary air; thus temperature decreases more slowly as the main burner gases mix with tertiary air. Thus temperature effect is dominant from 0.85 to 0.9; further increase increases the oxygen concentration to decrease and the oxygen effect seems to be dominant for Overall ER > 0.9.
The higher percentage of fuel bound nitrogen in Dairy biomass which can be observed in the Table 5.6 below:

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Empirical Formula</th>
<th>Fuel bound nitrogen</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dairy biomass</td>
<td>CH$<em>{1.255424}$N$</em>{0.046999}$O$<em>{0.396524}$S$</em>{0.004573}$</td>
<td>0.04699</td>
</tr>
<tr>
<td>PRB coal</td>
<td>CH$<em>{0.699206}$N$</em>{0.02165}$O$<em>{0.18217}$S$</em>{0.002174}$</td>
<td>0.02165</td>
</tr>
</tbody>
</table>

As we can see from the empirical formulae Dairy biomass has a higher level of FB nitrogen 0.04699 than PRB coal 0.02165. Further it is expected that the NO level should increase as we increase the blend percentage of Dairy biomass. These results have been presented next.
Comparison of NO emission for pure coal and blend

NO emission results obtained for all blends 95-5, 90-10 and 85-15 have been computed using the model and have been indicated in figure 5.9 below.

![Graph showing NO vs ER for all fuels](image)

Figure 5.9 Comparison of NO emissions for blended to unblended fuel

We can see that blended fuel gives a higher NO emission throughout the range of equivalence ratios; this can be attributed to the fact that FB nitrogen in biomass dissociates into 60% ammonia which has faster oxidation kinetics that FB nitrogen in coal which dissociates into 30% ammonia. This extra amount of ammonia generated from biomass leads to higher NO emission. The results are also verified by experimental data which shows more NO emission for blended fuels.
**Parametric studies**

**Effect of mixing time**

To observe the effect of mixing time on the results of the model, mixing time has been varied from 10-50-80ms and the following NO emission results have been captured in figure 5.10.

![NO vs ER graph](image)

**Figure 5.10 Parametric evaluation of mixing time on NO**

From the above graph it can be observed that when mixing time is 80ms, the model gives a higher value at ER=0.95. But at the same time, we can see that a mixing time of 50ms or 10ms follow the same pattern and return close values. Mixing time is directly related to the time taken by the gases to cool down. In reality the mixing times is dictated by the experiment.
**Effect of pre exponential factors**

The pre-exponential factors for NO oxidation reactions have been reduced by a factor of 10 in one case & reduction reactions were increased by a factor of 10. The results have been presented in Figure 5.11 below.

It can be observed how changing kinetics by a factor of 10 has a profound impact on the NO emission. It shows the sensitivity of the model to kinetics and arriving at results close to experimental results would require kinetics specific to the application. Other iterative procedures could be used for arriving at the NO results predicted by the experiments but that would require much higher computational efforts.
Comparison of NO vs Staging percentage

Contrary to theoretical and experimental findings, NO emission increases with staging. As observed in the Figure 5.12 below

![Pure PRB NO vs percentage staging](image)

Figure 5.12 NO vs. percentage staging for pure PRB

This deviation from previous literature is due to an assumption made in making the model. Increase in NO observed is due to fuel bound nitrogen released as ammonia, hydrogen cyanide and molecular nitrogen. This hydrogen cyanide and ammonia produced are oxidizing in the overfired zone in excess of air to produce more NO. In an actual burner, there is some fuel left in the MB which consumes oxygen available in the overfired zone and prevents fuel bound nitrogen components from oxidizing. Thus leading to reduction in NO
To correct this discrepancy in results obtained from the model for increased staging, a new method- LNB model II to modify the original reburn model has been thought of. The changes to be made in this model have been discussed earlier in the modeling chapter.
CHAPTER VI

CONCLUSIONS

The zero dimensional LNB model I though simplistic is able to predict the NOx emission trend and results close to experimental values when the ER>1. Since kinetics from literature closest to the operating parameters has been used there is a difference in the level of NO emissions between model results and experimental data. Iterative procedures could be used for making the model results much closer but that would require huge computational efforts, reducing the ease of use of this model. Below is a list of conclusions

1. NOx emission is at its minimum for overall rich conditions for both pure PRB and blends.
2. Richer ER leads to lesser NO, but optimizing the amount air in both zones is essential to achieve maximum power which emitting least permissible NO
3. Firing blends releases around 15% more NO than pure PRB firing.
4. Accuracy of results is strongly dependent on the selection of applicable kinetics.
5. Mixing time of 50ms is a good approximation for this kind of application.
6. Assumption that all fuel goes to incomplete combustion products leads to deviant results when staging percentage is increased.
CHAPTER VII

FUTURE WORK

Future work to the modeling currently done would be:

1. Including swirl number, particle size distributions and as a parameter to the model and observe the effect of it on NO reduction using LNB model II suggested.

2. Creating a 3D model using commercial software like fluent.

3. Calculation the mixing time in the LNB burner using an experimental procedure.

4. Conduct experiments on Cofiring blends of (coal+torrefied) biomass and energy crops like (coal+sorghum) and validate the same using this model.
REFERENCES


   http://www.epa.gov/air/nitrogenoxides/

   http://www.epa.gov/cair/tx.html

   http://www.epa.gov/oaqups001/nitrogenoxides/health.html


http://www.epa.gov/cair/


# VITA

<table>
<thead>
<tr>
<th>Name:</th>
<th>Hari Krishna Uggini</th>
</tr>
</thead>
<tbody>
<tr>
<td>Address:</td>
<td>Department of Mechanical Engineering</td>
</tr>
<tr>
<td></td>
<td>Texas A&amp;M University</td>
</tr>
<tr>
<td></td>
<td>College station, TX 77843-3123</td>
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<td>Email Address:</td>
<td><a href="mailto:haribits150@gmail.com">haribits150@gmail.com</a></td>
</tr>
<tr>
<td>Education:</td>
<td>B.E. (Hons) Mechanical Engineering BITS-Pilani, India</td>
</tr>
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