STELLINGEN

behorende bij het proefschrift

FUEL STAGING AND ITS IMPLEMENTATION
IN A NOVEL INTERNALLY STAGED BURNER

van

K.J. Knill

Technische Universiteit Delft, november 1990
1. The internal recirculation zone of a swirling flow burner may be utilized as a high temperature, O₂ lean reactor where CH₄ radicals may be formed from reburn fuel and mixed with primary combustion products to reduce NOx.

2. In laboratory experiments on reburning, NOx may be reduced to very low levels by operating at a low reburn zone stoichiometry. However, 25-40% of the NOx is converted to NH₃ which may be reconverted to NOx in the tertiary combustion zone.

3. In pulverized coal-fired utility boilers, the extent that NOx may be reduced is determined by the limits on CO emissions and carbon in ash.

4. In any NOx reduction technique, it is important to ensure that there is no appreciable shift of nitrogen species to N₂O.

5. There is only a narrow window in time to develop NOx reduction methods for pulverized coal fired utility boilers because of deadlines imposed through legislation to attain low NOx emissions.
6. The merit of computational modelling predictions is only as high as the accuracy of experimental data which is generated for verification.

7. For any model on coal combustion, one may always find a coal that is contradictory.

8. The most effective method for the Dutch Government to reduce air pollution in the Netherlands would be to fund the installation of emission control systems on industries and utilities in eastern Europe.

9. Canadians spend half their time convincing Americans that they are not British and the other half of their time convincing the rest of the world that they are not Americans.

10. There is an essential difference in crowd behaviour between the most popular Dutch sport, football, and the most popular Canadian sport, ice hockey. In football, there is widespread violence in the crowd whereas in ice hockey, the crowd enjoys watching the widespread violence amongst the players.
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PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Technische Universiteit Delft, op gezag van de Rector Magnificus, prof. drs. P.A. Schenck, in het openbaar te verdedigen ten overstaan van een commissie aangewezen door het College van Dekanen, op 13 november 1990 te 16.00 uur. door

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Permission is given by the Joint Committee of the International Flame Research Foundation for the use of the Foundation's research results used in the preparation of this dissertation.
to Anna

to Michael, Cory and Christopher
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Summary

There is increasing public concern over the emission of noxious gases from industrial and utility processes. NOx is one of the noxious gases which is formed in industrial and utility systems during the combustion of hydrocarbon fuels. When coal is fired in utility boilers, it is often difficult to meet environmental legislation designed to control NOx emissions. Therefore, the reduction of NOx emissions is of considerable interest to industry and various methods of reduction have been developed.

Among these, fuel staging, or reburning, is a relatively new technique whereby the NOx concentration may be controlled in the furnace. To develop reburning as a viable NOx reduction technique, attention is centered on understanding the process in utility boilers and determining the most economic and reliable equipment arrangement.

This study was undertaken to investigate the process parameters which influenced coal reburning and to apply these principles in designing and testing a coal-fired, fuel staged burner. This burner was intended to be used on wall fired boilers with both coal and natural gas considered as reburn fuels.

The study was conducted in three experimental parts. In the first part (Reburn Zone Investigation), the influence of process parameters on NOx, HCN and NH3 reduction in the reburn zone was considered. The reburn zone temperature, stoichiometry (λRB) and residence time were shown to have the most significant influence on the NOx and total volatile fixed nitrogen (TVFN; sum of NOx, HCN and NH3) reduction in the reburn zone. The NOx concentration could be reduced to less than 50 ppm over a wide range of conditions. However, TVFN could only be reduced to less than 200 ppm when temperature was greater than 1400°C, reburn zone stoichiometry, λRB, was 0.8-0.9 and residence time was greater than 0.5 s.

In the second part (Furnace Investigation), the NOx emissions from a fuel staging furnace, operated with incomplete mixing of reburn fuel and primary combustion products, was evaluated. For a primary zone NOx concentration of 1000 ppm, the NOx emissions could not be reduced to less than 350 ppm. The minimum NOx emission was found to be comparable to the minimum TVFN concentration measured in the Reburn Zone Investigation at the same process conditions. These results demonstrated the
importance of operating a fuel staged burner at conditions which minimized the TVFN concentration at the end of the reburn zone.

In the third part of the study, an Internally Fuel Staged Burner (IFSB) was developed and tested. The design was based on the concept of injecting the coal or natural gas reburn fuel through the center of the burner and reducing primary NOx at the downstream end of the internal recirculation zone.

The NOx emissions were strongly dependent on $\lambda_{RB}$ in the IFSB. Using coal as the reburn fuel, the NOx emissions decreased from 300 to 150 ppm as $\lambda_{RB}$ decreased from 0.82 to 0.72. Natural gas was shown to be a more effective reburn fuel than coal. NOx emissions decreased from 220 to 100 ppm as $\lambda_{RB}$ decreased from 0.85 to 0.7. The improved performance of natural gas reburn fuel was attributed to the increased volatility and lower fuel nitrogen content compared to coal. The results demonstrated that the burner could be operated with low NOx emissions.

It is believed that the IFSB merits further development. The NOx emissions from the IFSB were low compared to existing low NOx burners. Also, burner performance was not adversely affected by operating with low NOx emissions. The CO emission did not exceed 5 ppm and the flame was compact and stable within the quarl for all tests. Additional tests are recommended to improve the design and to establish scaling criteria.
Samenvatting

Er is een toenemende bezorgdheid aangaande de emissies van stikstof bevattende gassen, afkomstig van elektriciteitscentrales en industriële installaties. NOx is een van de stikstof bevattende gassen welke gevormd wordt tijdens de verbranding van fossiele brandstoffen in industriële systemen. In kolen gestookte installaties is het vaak moeilijk om de wettelijk vastgestelde emissie eisen te halen. Daarom is de reductie van de NOx emissies van byzonder belang voor de industrie en verscheidene methoden zijn daartoe ontwikkeld.

Een van deze methoden is gebaseerd op het gefaseerd toevoeren van de beschikbare brandstof aan het verbrandingsproces (eng. "reburning"). Recombusting is een relatief nieuwe techniek waarbij de NOx concentraties kunnen worden gecoreleerd in de verbrandingszone. Het is van belang om het proces te begrijpen en te optimaliseren, teneinde reburning te ontwikkelen tot een betrouwbare en zo economisch mogelijke techniek voor het reduceren van de NOx concentraties in kolenstookte installaties.

Deze studie behelst onderzoek naar de proces parameters die reburning van steenkool beïnvloeden en tevens toepassing van de resultaten (van dit onderzoek) in het ontwerp en de test van een steenkool gestookte brander die gebruik maakt van reburning. De brander is bedoeld om te worden gebruikt als frontbrander, zowel met aardgas als met kolen als brandstof voor reburning.

De studie is uitgevoerd in drie gedeelten. In het eerste gedeelte (onderzoek van de reburn zone) is de invloed van proces parameters op de reductie van NOx en van de totale hoeveelheds stikstof in NOx, HCN en NH3 (TVFN, "total volatile fixed nitrogen") in de reburn zone bestudeerd. Het is gebleken dat de temperatuur, de stoichiometrie ($\lambda_{RB}$) en de verblijftijd de meest belangrijke invloed hebben op de reductie van NOx en van TVFN in de reburn zone. De NOx concentratie kon worden gereduceerd tot minder dan 50 ppm voor een groot aantal proces condities. Alhoewel, de TVFN kon alleen worden gereduceerd tot minder dan 200 ppm bij een temperatuur boven de 1400°C, de stoichiometry in de reburn zone ($\lambda_{RB}$) tussen 0.8 en 0.9 en de verblijftijd groter dan 0.5 s.
In het tweede gedeelte (onderzoek naar menging) zijn de effecten van incomplete menging tussen de primaire verbrandings producten en de reburn brandstof op de NOx emissies geevalueerd. Het bleek dat, bij een NOx concentratie van 1000 ppm in de primaire zone, de NOx emissie niet kon worden gereduceerd tot minder dan 350 ppm. De gevonden minimale NOx emissie was vergelijkbaar met de TVFN waarden, gemeten tijdens het onderzoek van de reburn zone. Deze resultaten demonstremen het belang van het opereren van een brander, gebaseerd op reburning, onder de condities die de TVFN aan het eind van de reburn zone minimaliseren.

In het derde gedeelte van de studie is de IFSB (Internally Fuel Staged Burner) ontwikkeld en getest. Het ontwerp is gebaseerd op het injecteren van steenkool of aardgas als reburn brandstof door het centrum van de brander. Middels deze brandstof kan de primaire NOx gereduceerd worden aan het einde van de interne recirculatie zone.

De NOx emissies waren sterk afhankelijk van $\lambda_{RB}$ in the IFSB. Bij gebruik van steenkool als reburn brandstof daalde de NOx emissie van 300 tot 150 ppm door een reductie van $\lambda_{RB}$ van 0.82 tot 0.72. Aardgas bleek een meer effectieve brandstof voor reburning. De NOx emissie daalde van 200 ppm tot 100 ppm door een reductie van $\lambda_{RB}$ van 0.85 tot 0.7. De grotere effektiviteit van aardgas is te danken aan de grotere vluchtigheid en het lagere gehalte aan gebonden stikstof vergeleken met steenkool. De resultaten toonden aan dat de brander kon worden gebruikt met lage NOx emissies.

Het is geconcludeerd dat de IFSB een verdere ontwikkeling waard is. De NOx emissies van de IFSB waren lager dan verkregen met bestaande lage NOx branders. Bovendien werd het algehele brander gedrag niet negatief beïnvloed door operatie onder de proces condities die lage NOx emissies ten gevolge hadden. Gedurende alle tests was de CO emissie niet hoger dan 5 ppm en was de vlam compact en gestabiliseerd binnen de brander toog. Aanvullende tests zijn aanbevolen teneinde het ontwerp verder te optimaliseren en om criteria voor opschaling vast te stellen.
Preface

The research described in this dissertation was carried out at the International Flame Research Foundation in Umniden. In recent years, a major portion of the research activity at the Foundation was related to the IEA Coal Combustion Sciences Programme: Annex 2. The principle purpose of the program was to investigate NOx reduction in practical coal combustion systems. Fuel staging was an integral part of the program since 1987 and has involved several research projects integrating furnace experiments, mathematical modelling and aerodynamic studies.

There are several people that I would like to thank for their contributions in this research. I am grateful to my promotor, Prof.Dr.Ing. K.R.G. Hein for his valuable comments on the research and this thesis. He explained many of the practical problems associated with NOx reduction in general and fuel staging in particular.

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1. Introduction

There is increasing public concern over the emission of noxious gases from industrial and utility processes. These emissions are regarded as having a negative impact on human health, the environment, vegetation and buildings. NOx is one of the emissions which has been designated noxious and has been subject to environmental legislation. NOx are stable gas species that may react with moisture in the atmosphere to form acidic gases. This, in turn, damages plant matter, acidifies lakes and streams, and erodes concrete structures [1]. The NO may also participate in a set of complex reactions forming ozone in the atmosphere [2,3]. Ozone is a health concern since humans are particularly sensitive to ozone exposure during exercise [3]. As a result, governments in most industrialized countries are legislating strict limits on the concentration and total mass of NOx emissions.

NOx is formed in industrial and utility processes during the combustion of hydrocarbon fuels. In high temperature processes, NOx may be formed from the dissociation of \( \text{N}_2 \) and \( \text{O}_2 \) of the combustion air and recombination as NO (thermal NOx) [4]. The NOx may also be formed from the oxidation of nitrogen containing species of the fuel (fuel NOx) [4]. It is often difficult to meet environmental legislation due to the formation and emission of thermal and, particularly, fuel NOx in coal fired utility boilers. Therefore, the reduction of NOx emissions from pulverized coal fired utility boilers is of considerable interest to industry and several reduction methods have been developed. These methods have been classified according to the stage where they are applied in the combustion process.

One method of lowering emissions is to remove NOx after the combustion process by treating the flue gases. Selective catalytic reduction (SCR) is an industrial technique in which ammonia is injected into the flue gas and passed over a catalyst reducing NOx to \( \text{N}_2 \). Although SCR is effective at lowering NOx, there is a very high capital investment and operating cost associated with this process [5]. There is also concern that the technique may be difficult to retrofit in existing combustion equipment. Another flue gas treatment is to scrub the flue gases using wet chemical cleaning techniques. However, scrubbing processes are still under development and have not been widely applied in industry.
NOx may also be reduced in the heat transfer section of the boiler. Selective non-catalytic reduction (SNCR) processes involve injecting nitrogen based chemicals, such as urea or ammonia, into the furnace product gases [6]. It is difficult to substantially reduce NOx concentrations using SNCR due to operational limitations [7]. Proper injection of the chemicals into the combustion products is a major problem since the process is only effective within a narrow temperature window. Thus, insufficient mixing will cause unreacted chemicals to be emitted in the flue gas.

NOx formation may be controlled directly in the flame by modifying the combustion process [7]. The thermal NOx is minimized by controlling the peak temperature in O2 lean regions of the flame [7]. Fuel NOx formation is prevented by delaying the mixing of nitrogen containing fuel species (fuel-N) with O2 which enhances the reduction of fuel-N to N2 [4]. Most of the in-flame reduction techniques which have been developed affect both the thermal and fuel NOx formation since processes which delay the mixing of fuel and O2 also limit the peak flame temperature.

Low excess air firing and flue gas recirculation (FGR) are techniques which reduce the O2 concentration and temperature in the flame zone. These techniques are primarily used in oil and gas fired boilers although they have been shown to be effective in coal fired boilers in combination with air staging [8]. Air staged burners have been designed to limit the mixing of fuel and air. A portion of the air is added downstream of the flame zone to provide sufficient time for intermediate N-species, generated in the flame, to be reduced to N2. These burners have been shown to reduce NOx emissions by 50% compared to conventional designs.

Fuel staging or "reburning" has been demonstrated to be an effective in-flame reduction technique [9]. In the reburning process, a fuel lean primary combustion zone, \( \lambda_p > 1 \), is formed with most of the fuel, Figure 1.1. The remaining fuel (reburn fuel) is injected into a fuel rich reburn zone, \( \lambda_{RB} < 1 \), where primary zone NOx is reduced to N2. The remaining air is added into the tertiary combustion zone to complete the combustion process. This technique is a promising method of NOx reduction which may be applied directly in the firebox to result in low NOx emissions [10].

Two configurations for reburning have been demonstrated in utility boilers: furnace staging [11] and the use of fuel staged burners [12]. The most extensively tested configuration is furnace staging, Figure 1.2, in which the entire firebox is staged by injecting the reburn fuel above the primary combustion zone. Furnace staging has been
tested in tangential and wall fired boilers with NOx emissions reported to be reduced by up to 50% [7,11].

In fuel staged burners, Figure 1.2, the reburn fuel is injected directly downstream of the primary combustion zone of each burner. Fuel staged burners have only recently been developed and demonstration results are limited. However, fuel staged burners are a promising NOx reduction technique since they combine the advantages of fuel staging with proven performance of low NOx burners.

Figure 1.1 Concept of the reburning process

Figure 1.2 Process configurations for furnace and burner fuel staging
1.1 Purpose of the Study

Existing fuel staged burners have been demonstrated to achieve low NOx emissions. However, the technology is at an early stage of development and existing designs are relatively complex.

This study was undertaken to investigate the process parameters which influenced coal reburning and to apply the results of the investigation in designing and testing a fuel staged burner. The fuel staged burner is intended to be used on wall fired boilers burning pulverized coal with both coal and natural gas considered as reburn fuels.

1.2 Outline of the Study

The study reviews nitrogen chemistry and the published information on reburning development and demonstration tests, Chapter 2. The fundamental chemistry of NOx formation and destruction are detailed with emphasis on the reactions related to reburning and simple kinetic models. Published results relating the effect of process variables on NOx reduction are presented and considered. Large scale demonstration results and experience with fuel staging retrofit are discussed and the importance of fuel staging compatibility with other NOx reduction technologies is reviewed.

A swirl stabilized burner is considered as the basis of the fuel staged burner. The design and operating principles of swirl stabilized burners are also described in Chapter 2. The influence of burner design and flame type on NOx emission reduction using swirl stabilized burners is discussed. In particular, the influence of coal particle trajectories inside the internal recirculation zone (IRZ) on NOx emissions are reviewed.

The experimental method used to evaluate the influence of process variables on NOx reduction and to test the fuel staged burner is described in Chapter 3. The reactor and sampling equipment, experimental procedures, and analytical techniques are presented. The computational model for predicting the flow properties and chemical reactions in the experiments and in the swirl stabilized burner is presented.

The influence of process variables on NOx reduction was evaluated in two experimental programs: the Reburn Zone and Furnace Investigations. In the Reburn Zone Investigation, described in Chapter 4, the change in NOx, HCN and NH₃ concentrations
in the reburn zone were measured. The tests were conducted under rapid mixing conditions to study the chemical processes that were occurring. However, mixing of reburn fuel and primary combustion products may have a significant effect on NOx emissions, and therefore, mixing limited reburning experiments were performed in the Furnace Investigation. Mathematical modelling, flow visualization and tracer studies were conducted to characterize the mixing in the Furnace experiments, Chapter 5. In the Furnace Investigation, described in Chapter 6, the influence of mixing rates, fuel and air injection arrangements on NOx emissions was evaluated. The results of the two Investigations were related to determine the optimum operating conditions necessary to minimize NOx emissions when using a fuel staged burner.

The fuel staged burner was designed based on properties of swirl burners and the results of the two Investigations. The fuel staged burner is presented in Chapter 7 and operating advantages of the burner in a utility boiler are discussed.

Results of experimental tests conducted using the fuel staged burner are detailed in Chapter 8. The NOx emissions are evaluated in light of the findings of the two Investigations, discussed in Chapters 4 and 6. A numerical simulation of the burner is presented and used to interpret the interaction of the reburn fuel and primary combustion products inside and downstream of the IRZ.

Conclusions from the investigations on the reburning process and its application in a fuel staged burner, are given in Chapter 9.
2. Principles and Development of Fuel Staging

The reduction of NOx by fuel staging is affected by the flame chemistry and process conditions. The mechanisms of NOx formation and destruction are reviewed, Section 2.1, and the influence of the most important process parameters on NOx reduction by fuel staging are discussed, Section 2.2.

Reburning has been successfully demonstrated as an NOx reduction technique for utility boilers. These demonstrations are reviewed in Section 2.3. The benefit of installing fuel staging in a utility boiler is based on the level that NOx may be reduced while maintaining acceptable boiler performance and reliability. The impact of installing reburning on the performance of a utility boiler is discussed in Section 2.4.

A swirling flow burner was considered as the basis for designing a fuel staged burner. The principles of swirling flow burners and NOx reduction techniques are presented and discussed, Section 2.5.

2.1 Nitrogen Chemistry

2.1.1 NOx Formation Mechanisms

NOx is partly formed in pulverized coal combustion systems from thermal fixation of combustion air according to the Zeldovich mechanism [13] and partly from oxidation of fuel bound nitrogen. Thermal fixation has a very high temperature dependency due to a high activation energy (78500 cal/mol) and is not significant until flame temperatures exceed 1500°C. It is estimated that only 20% of the NOx formed during coal combustion may be attributed to thermal fixation [14]. Therefore, the majority of NOx is formed from nitrogen bound in the fuel (hereafter referred to as fuel-N), particularly fuel-N contained in the volatile matter [14,15].

When coal is injected into a hot flame, devolatilization occurs at a rate limited by coal heatup and mass transfer of species out of the particle. In utility boiler firing of pulverized coal, most particles have a diameter less than 75 μm and heating rates are on
the order of $10^6 - 10^8$ K/s. Under these conditions, devolatilization is essentially complete in 10 - 30 ms or when the particle temperature exceeds 1000°C [16]. A large fraction of the fuel-N is released with the volatile matter. Pohl and Sarofim [15] and Haussman [17] showed that nitrogen was evolved from the coal at a rate similar to total volatile matter release, with an initial lag in nitrogen evolution in the first 15% of volatile evolution.

Up to 90% of the nitrogen released may be contained in large molecular weight pyridine, pyrrole and aliphatic hydrocarbons in the tar fraction [18,19]. Upon heating, the tar undergoes thermal degradation and the aromatic and aliphatic nitrogen compounds are converted to HCN and NH$_3$, respectively. In pyrolysis studies of pyridine and pyrrole, it was shown that thermal decomposition to HCN could be modelled by the expressions [20]:

$$R_{\text{Pyridine}} = -3.8 \times 10^{12} \exp \left[-35200/RT\right] \text{ s}^{-1}$$  \hspace{1cm} (2.1)

$$R_{\text{Pyrrole}} = -7.5 \times 10^{15} \exp \left[-42800/RT\right] \text{ s}^{-1}$$  \hspace{1cm} (2.2)

Both compounds were stable at 800°C but due to the high activation energy, they totally decomposed to HCN in 2 ms at 1600°C. Therefore, NOx formation from fuel-N may be assumed to proceed from HCN and NH$_3$ precursors. A simplified mechanism for NOx formation from HCN is shown in Figure 2.1. Miller [21] showed that HCN oxidation was dominated by the reactions:

$$\text{HCN} + \text{O} = \text{NCO} + \text{H} \hspace{1cm} \text{(R1)}$$

$$\text{HCN} + \text{O} = \text{NH} + \text{CO} \hspace{1cm} \text{(R2)}$$

Subsequent reactions of NCO and NH with H atoms, producing N atoms, were rapid.

$$\text{NCO} + \text{H} = \text{NH} + \text{CO} \hspace{1cm} \text{(R3)}$$

$$\text{NH} + \text{H} = \text{N} + \text{H}_2 \hspace{1cm} \text{(R4)}$$
Figure 2.1 NOx formation mechanism from volatile fuel-N in coal flames

The reactions of N radical with OH and NO determined the final product distribution:

\[
\begin{align*}
N + OH &= NO + H \\
N + NO &= N_2 + O
\end{align*}
\]  
(R5)  
(R6)

When NH₃ is the precursor, then oxidation to NOx proceeded through a different reaction sequence. Ammonia was converted to NH₂ by hydrogen abstraction:

\[
\begin{align*}
NH_3 + OH &= NH_2 + H_2O \\
NH_3 + O &= NH_2 + OH \\
NH_3 + H &= NH_2 + H_2
\end{align*}
\]  
(R7)  
(R8)  
(R9)

and successively smaller NH₄ radicals were formed by reaction with H atoms:

\[
NH_4 + H(OH) = NH_{4-1} + H_2(H_2O)
\]  
(R10)
Each NH$_4$ radical may oxidize to form NO directly or through the HNO intermediate:

\[
\begin{align*}
\text{NH} + \text{O}_2 & = \text{NO} + \text{OH} \quad \text{(R11)} \\
\text{NH}_2 + \text{O} & = \text{HNO} + \text{H} \quad \text{(R12)} \\
\text{NH} + \text{O}_2 & = \text{HNO} + \text{O} \quad \text{(R13)} \\
\text{NH} + \text{OH} & = \text{HNO} + \text{H} \quad \text{(R14)} \\
\text{HNO} + \text{OH} & = \text{NO} + \text{H}_2\text{O} \quad \text{(R15)}
\end{align*}
\]

2.1.2 NOx Reduction Mechanisms

Hydrocarbon radicals injected into the primary combustion products at substoichiometric conditions may result in significant reduction of NO to molecular nitrogen. The process was first reported by Fenimore [22] who investigated NO reduction in H$_2$ flames doped with HCN and NH$_3$. He proposed the three step mechanism:

\[
\begin{align*}
\text{HCN} + \text{OH} & = \text{NH}_2 + \text{CO} \quad \text{(R16)} \\
\text{NH}_2 + \text{NO} & = \text{N}_2 + \text{H}_2\text{O} \quad \text{(R17)} \\
\text{NH}_2 + \ldots & = \text{NO} + \ldots \quad \text{(R18)}
\end{align*}
\]

to account for the observed NOx reduction. Reaction R18 was a combination of reaction steps leading to NO formation and was related to the competing rate of NO destruction, Reaction R17, by the temperature and water vapour concentration according to:

\[
k_{18} = k_{17} \frac{[\text{H}_2\text{O}]}{[\text{NO}]} 700 \exp \left[ -\frac{50000}{RT} \right] \quad (2.3)
\]
Thorne et al. [23] also studied the mechanism for NO reduction by hydrocarbon radicals using a low pressure, fuel rich C₂H₂ flat flame burner. A model of the dominant reaction sequence was developed which was similar to the reduction sequence shown in Figure 2.2. The dominant reactions and their kinetic constants are listed in Table 2.1. The reduction of NO by hydrocarbon radicals was shown to be initiated by oxidizing C₂H₂ with O radicals according to:

\[ \text{C}_2\text{H}_2 + \text{O} = \text{CH}_2 + \text{CO} \]  \hspace{1cm} (R19)

followed by hydrogen abstraction:

\[ \text{CH}_i + \text{H} = \text{CH}_{i-1} + \text{H}_2 \]  \hspace{1cm} (R20)

The hydrocarbon radicals reduce NO via Reactions R21 - R24:

\[ \text{C} + \text{NO} = \text{CN} + \text{O} \]  \hspace{1cm} (R21)
\[ \text{CN} + \text{H}_2 = \text{HCN} + \text{H} \]  \hspace{1cm} (R22)
\[ \text{CH} + \text{NO} = \text{HCN} + \text{O} \]  \hspace{1cm} (R23)
\[ \text{C}_2\text{H}_2 + \text{NO} = \text{HCN} + \text{OH} \]  \hspace{1cm} (R24)
# Table 2.1 Summary of chemical reactions

<table>
<thead>
<tr>
<th>Number</th>
<th>Reaction</th>
<th>(A)</th>
<th>(\beta)</th>
<th>(E)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>HCN + O = NCO + H</td>
<td>1.4e4</td>
<td>2.64</td>
<td>4980</td>
</tr>
<tr>
<td>2</td>
<td>HCN + O = NH + CO</td>
<td>3.5e3</td>
<td>2.64</td>
<td>4980</td>
</tr>
<tr>
<td>3</td>
<td>NCO + H = NH + CO</td>
<td>5.0e13</td>
<td>0.00</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>NH + H = N + H₂</td>
<td>1.0e14</td>
<td>0.00</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>N + OH = NO + H</td>
<td>3.8e13</td>
<td>0.00</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>N + NO = N₂ + O</td>
<td>3.3e12</td>
<td>0.30</td>
<td>0</td>
</tr>
<tr>
<td>7</td>
<td>NH₃ + OH = NH₂ + H₂O</td>
<td>2.0e6</td>
<td>2.04</td>
<td>566</td>
</tr>
<tr>
<td>8</td>
<td>NH₃ + O = NH₂ + OH</td>
<td>2.1e13</td>
<td>0.00</td>
<td>9000</td>
</tr>
<tr>
<td>9</td>
<td>NH₃ + H = NH₂ + H₂</td>
<td>6.4e5</td>
<td>2.39</td>
<td>10171</td>
</tr>
<tr>
<td>10</td>
<td>NH₂ + H = NH + H₂</td>
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<td>3650</td>
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<tr>
<td>11</td>
<td>NH + O₂ = NO + OH</td>
<td>7.6e10</td>
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<td>1530</td>
</tr>
<tr>
<td>12</td>
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<td>13</td>
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<td>NH + OH = HNO + H</td>
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<tr>
<td>15</td>
<td>HNO + OH = NO + H₂O</td>
<td>3.6e13</td>
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<td>0</td>
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<tr>
<td>16</td>
<td>HCN + OH = NH₂ + CO</td>
<td>1.0e12</td>
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<td>0</td>
</tr>
<tr>
<td>17</td>
<td>NH₂ + NO = N₂ + H₂O</td>
<td>6.2e15</td>
<td>-1.25</td>
<td>0</td>
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<td>18</td>
<td>NH₂ + ... = NO + ...</td>
<td>3.5e15</td>
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<td>50000</td>
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<tr>
<td>19</td>
<td>C₂H₂ + O = CH₂ + CO</td>
<td>1.0e7</td>
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<td>21</td>
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<td>0</td>
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<td>2237</td>
</tr>
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<td>0.00</td>
<td>0</td>
</tr>
<tr>
<td>24</td>
<td>CH₂ + NO = HCN + OH</td>
<td>2.0e13</td>
<td>0.00</td>
<td>0</td>
</tr>
<tr>
<td>25</td>
<td>H₂ + O₂ = OH + O</td>
<td>5.1e16</td>
<td>-0.82</td>
<td>16507</td>
</tr>
<tr>
<td>26</td>
<td>H + NO + M = HNO + M</td>
<td>2.9e22</td>
<td>0.00</td>
<td>-100755</td>
</tr>
<tr>
<td>27</td>
<td>HNO + H = NH + OH</td>
<td>1.0e12</td>
<td>0.00</td>
<td>-13480</td>
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<tr>
<td>28</td>
<td>NH₂ + NO = N₂ + products</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>29</td>
<td>NO + C = CO + 0.5N₂</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>30</td>
<td>2NO + C = CO₂ + N₂</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>31</td>
<td>NO + CO = CO₂ + N₂</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>32</td>
<td>NH + NO = N₂O + H</td>
<td>2.4e15</td>
<td>-0.80</td>
<td>0</td>
</tr>
<tr>
<td>33</td>
<td>N₂O + H = N₂ + OH</td>
<td>7.6e13</td>
<td>0.00</td>
<td>15200</td>
</tr>
</tbody>
</table>

*Rate coefficients are given in the form \( k_r = A \exp(-E/RT) \) where \(A\) (cm³/mol), \(E\) (cal/mol), \(T\) (K).

Coefficients were taken from Thorne et al [23] except R16 and R18 from Fenimore [22].

Once HCN was formed, it was oxidized and subsequently reduced following the dominant pathway:
\[
\begin{align*}
\text{HCN} + \text{O} & = \text{NCO} + \text{H} \quad \text{(R1)} \\
\text{NCO} + \text{H} & = \text{NH} + \text{CO} \quad \text{(R3)} \\
\text{NH} + \text{H} & = \text{N} + \text{H}_2 \quad \text{(R4)} \\
\text{N} + \text{NO} & = \text{N}_2 + \text{O} \quad \text{(R6)}
\end{align*}
\]

Thorne et al. [23] showed good agreement between predicted and measured concentrations of in-flame nitrogen species (NOx, HCN, NH3, N). They determined the sensitivity of NOx reduction in the model by perturbing individual reaction rate constants and observing the influence on NOx concentration. Using this method, it was determined that NOx formation was most sensitive to the OH reaction:

\[
\text{H} + \text{O}_2 = \text{OH} + \text{O} \quad \text{(R25)}
\]

because of its propensity to form oxidizing radicals. Thorne also noted a higher sensitivity of NO reduction to Reaction R21 compared to Reactions R23 and R24. This suggested that the C radical was a better reducing agent than CH or CH2 radicals.

Similarities existed between the NOx reduction models of Thorne et al using hydrocarbons (Reactions R1-R6 and R19-R25) and Fenimore using nitrogenated species (Reactions R16-R18). A sufficient concentration of H, OH and O radicals was necessary in both systems to form the NCO or NH radicals according to Reactions R1-R4. If the O radicals were not present in sufficient quantity, then HCN persisted throughout the reburn zone.

It was not always necessary to inject CH4 and NH radicals to reduce NOx. Roby and Bowman [24] reported the possibility of direct reduction of NO by H2 in atmospheric pressure, fuel rich H2/O2/Ar flames by the mechanism:

\[
\begin{align*}
\text{H} + \text{NO} + \text{M} & = \text{HNO} + \text{M} \quad \text{(R26)} \\
\text{HNO} + \text{H} & = \text{NH} + \text{OH} \quad \text{(R27)} \\
\text{NH} + \text{H} & = \text{N} + \text{H}_2 \quad \text{(R4)} \\
\text{N} + \text{NO} & = \text{N}_2 + \text{O} \quad \text{(R6)}
\end{align*}
\]
However, Miller and Bowman [21] showed, by sensitivity analysis, that NO reduction by H₂ was much less important than Reactions R21-R24 if CH₄ radicals were present.

The mechanisms discussed to this point were developed for gaseous hydrocarbon flames. Glass and Wendt [25] measured the NO, NH₃ and HCN concentrations along the length of a fuel rich laminar pulverized coal flame. They were able to predict NOx reduction using Fenimore’s model. To improve their prediction of experimental results, Wendt and co-workers [26,27] adjusted Fenimore’s model and compared predicted and measured HCN and NH₃ concentrations in the tail of pulverized coal flames. The model made no account for CH₄/NO type reactions but was based on the mechanism:

\[
\begin{align*}
\text{HCN} + \text{O} & \rightarrow \text{NCO} + \text{H} \quad \text{(R1)} \\
\text{NCO} + \text{H} & \rightarrow \text{NH} + \text{CO} \quad \text{(R2)} \\
\text{NH}_i + \text{NO} & \rightarrow \text{N}_2 + \text{products} \quad i=0,1,2 \quad \text{(R3)}
\end{align*}
\]

The concentrations of NHᵢ species, where i may vary from 1 to 3 were assumed to be in partial equilibrium according to the reaction:

\[
\text{NH}_i + \text{OH} \rightarrow \text{NH}_{i-1} + \text{H}_2\text{O} \quad i=1,2,3 \quad \text{(R10)}
\]

The equations which described the rate of NO and HCN decay in the reactor were:

\[
\begin{align*}
\frac{1}{[\text{NO}]} \frac{d[\text{NO}]}{dt} & = -[\text{NH}_3] \phi_1 \quad \text{(2.5)} \\
\frac{1}{[\text{HCN}]} \frac{d[\text{HCN}]}{dt} & = - (\phi_2 + \phi_3) \quad \text{(2.6)}
\end{align*}
\]

where the rate constants, \(\phi_1-\phi_3\), listed in Table 2.2, had the form:

\[
\phi_i = \sum_j A_{ij} T^{nj} \exp(B_{ij}/T) \text{OH}^{mij} \text{H}_2\text{O}^{lj} \quad \text{(2.7)}
\]
Table 2.2 Coefficients of the Bose and Wendt NOx reduction model (Equation 2.7)

<table>
<thead>
<tr>
<th>i</th>
<th>j</th>
<th>A_{ij}</th>
<th>N_{ij}</th>
<th>B_{ij}</th>
<th>M_{ij}</th>
<th>L_{ij}</th>
</tr>
</thead>
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<tr>
<td>1</td>
<td>1</td>
<td>3.97e17</td>
<td>-1.608</td>
<td>5277</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>7.57e15</td>
<td>-0.917</td>
<td>19650</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>3</td>
<td>1</td>
<td>3.2e12</td>
<td>-0.134</td>
<td>39105</td>
<td>3</td>
<td>3</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>9.2e12</td>
<td>0</td>
<td>-7549</td>
<td>1</td>
<td>0</td>
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<tr>
<td>2</td>
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<td>4.8e11</td>
<td>0</td>
<td>-5536</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>3</td>
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<td>1.05e3</td>
<td>2.701</td>
<td>6283</td>
<td>2</td>
<td>1</td>
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<tr>
<td>2</td>
<td>2</td>
<td>1.32e16</td>
<td>-0.695</td>
<td>-1327</td>
<td>2</td>
<td>1</td>
</tr>
</tbody>
</table>

The OH radical concentration was shown to have a significant effect on the rate of NO reduction in Equations 2.5 and 2.6. The OH concentration may be significantly far away from equilibrium conditions in pulverized coal flames where rapid cooling causes an excess of OH radicals downstream of the flame front. Bose et al [27] measured OH concentration in excess of equilibrium values in a laboratory scale flame with a rapid quench rate downstream of the flame front. The concentration of radicals was related to the equilibrium concentration by the expression:

\[ [\text{OH}] = [\text{OH}]_{\text{eq}} \times 5.7837 \times 10^{-4} \exp(13019/T) \]  

(2.8)

Good agreement was obtained between measured and predicted NOx and HCN concentrations. The authors concluded that an extended Fenimore mechanism could be used to predict NOx reduction in pulverized coal flames.

Thus, the reaction mechanisms developed by Fenimore [22] and Thorne [23] are instructive for understanding the influence of process parameters on NOx reduction by reburning. The model, proposed by Bose and Wendt [26], is a simple representation of the complex reburn process based on partial equilibrium assumptions for the NH₃ radical species. The model will be used to evaluate NOx reduction in the Reburn Zone Experiments (Section 4.3.6: p.102).
2.1.3 Char NOx Formation

The nitrogen contained in the char (char-N) may also be oxidized to NOx. The extent that char-N is converted to NOx is dependent on furnace operating conditions but has been shown to be significantly less than from volatile-N [14]. This may be due to reduction of NOx inside the pores of the char after it is formed according to [28]:

\[
\begin{align*}
\text{NO} + 2\text{C}_s & = \text{C}_s(\text{N}) + \text{C}_s(\text{O}) \\
\text{C}_s(\text{N}) + \text{C}_s(\text{N}) & = \text{N}_2 + 2\text{C}_s \\
\text{C}_s(\text{O}) & = \text{CO}
\end{align*}
\]

(R29)  
(R30)  
(R31)

where \( \text{C}_s \) represents a carbon surface and \( \text{C}(\text{N}) \) and \( \text{C}(\text{O}) \) are carbon sites with adsorbed nitrogen and oxygen atoms, respectively.

Glass and Wendt [25] studied the heterogeneous reduction of NOx by coal char to determine the importance of heterogeneous reactions to NOx reduction. The rate of NOx reduction by reaction on coal char could be correlated by:

\[
\frac{d\ [\text{NO}]}{dt} = -500 \ \text{exp} \left[ -37500/(RT) \right] \ P_{\text{NO}} \ \pi \varphi^2 \ (0.21 - P_{\text{O}_2})/0.21 \quad \text{(gmol/s/part.)} \quad (2.9)
\]

where \( P_{\text{NO}} \) and \( P_{\text{O}_2} \) were the NO and \( \text{O}_2 \) mole fractions, respectively, in the bulk gas.

Hahn and Shadman [29] showed that NO reduction occurred on the internal surface of coal particles. They modelled NO reduction using a simple kinetic expression based on internal particle surface area per unit volume, \( A_{\text{int}} \), and a distributed pore size model:

\[
\frac{d\ [\text{NO}]}{dt} = -2250 \ \text{exp} \left[ -34700/(RT) \right] \ P_{\text{NO}} \ A_{\text{int}} \quad \text{(g mol/m}^3/\text{s}) \quad (2.10)
\]

From Equation 2.10 and the pore size model, they were able to predict the reduction in NO during char combustion in a one-dimensional pulverised coal combustion reactor.
The rates of the char/NO reaction were 1–2 orders of magnitude slower than gas phase reactions. Thus, gas phase reactions should dominate NO reduction in the reburning zone of a fuel staged flame.

Although a lower percentage of char-N is converted to NO, it has also been demonstrated that char-N is less amenable to NOx reduction by combustion process modification [30]. This is due to the fact that char NOx is formed in the tertiary combustion zone where it is not possible to create fuel rich conditions since high mixing of tertiary air is necessary to ensure sufficient coal burnout. Therefore, for coals with a high char fraction, the potential to reduce NOx emissions from pulverised coal flames may be limited by the char-N content and its fractional conversion to NOx.

2.2 Process Parameters Influencing Coal Reburning

Results of reburning tests investigating the reduction of NOx emissions in pulverized coal flames were first reported by Takahashi et al [11]. Low NOx emissions were obtained using hydrocarbon gas, kerosene and coal reburn fuels in both laboratory and demonstration scale tests. Based on these results and an interest in NOx reduction, other laboratories have conducted coal reburning experiments. A review of their results indicate that the most important reburn zone process parameters are:

- reburn fuel type,
- primary zone stoichiometry, \( \lambda_P \)
- reburn zone stoichiometry, \( \lambda_{RB} \)
- reburn zone residence time, \( \tau_{RB} \)
- reburn zone temperature, \( T_{RB} \)
- reburn fuel mixing.

In addition, when coal is used as the reburn fuel, the following coal properties may be important:

- fuel nitrogen content and distribution,
- volatile matter content,
- char reactivity.
2.2.1 Reburn Fuel Type

Takahashi et al [11] studied the effect of hydrocarbon fuel type on NO reduction in a laboratory entrained flow combustor. Coal, kerosene and C\textsubscript{1} - C\textsubscript{7} hydrocarbon gases were tested at equivalence ratios of 0 - 1.2, reactor temperature of 1300°C and residence time of 0.2 s. NO\textsubscript{x} decomposition was found to be independent of fuel type with a 90% reduction in NO\textsubscript{x} from 80 ppm initial concentration. The equivalent NO\textsubscript{x} reductions were reported for all reburn fuels despite large differences in their abilities to form CH\textsubscript{4} radicals and their fuel-N contents. The insensitivity of NO\textsubscript{x} reduction to fuel type was confirmed in their demonstration scale tests on a 35 MW boiler firing propane, fuel oil and bituminous coal.

![Graph showing influence of fuel type and $\lambda_{RB}$ on NO\textsubscript{x} emissions](image)

**Figure 2.3** Influence of fuel type and $\lambda_{RB}$ on NO\textsubscript{x} emissions [31]

The results of Takahashi et al [11] were refuted by Chen et al [31]. They used a one-dimensional furnace to test the effectiveness of propane, coal and H\textsubscript{2} to reduce NO\textsubscript{x}. Four coals were studied ranging in rank from lignite (Yallourn coal) to high
volatile bituminous (Utah coal). For all reburn fuels, the NOx reduction was correlated with reburn fuel fraction, $f_{RB}$, and reburn zone stoichiometry, $\lambda_{RB}$, Figure 2.3. Propane was the most effective reburn fuel tested with NOx emissions of 200 ppm at $\lambda_{RB}$ of 0.9. Coal reburning also resulted in reduced NOx with lower emissions measured for higher volatile coals. At $\lambda_{RB}$ of 0.9, the NOx emissions ranged from 220 ppm for Yallourn lignite to 300 ppm for Colstrip hvb coal. H$_2$ was the least effective reburn fuel at $\lambda_{RB}$ of 0.9 with NOx emissions of 350 ppm. Thus, the ability of the reburn fuel to reduce NOx was shown to increase with the number of CH$_4$ radicals which may be formed from the hydrocarbon molecule [31,32]. However, H$_2$ was shown to be effective at reducing NOx for reburn zone stoichiometry less than 0.7 since H$_2$ reduced NOx according to Reaction R26-27 [24].

The results of Takahashi et al [11] were also refuted by Mul holland and Hall [33] who studied NOx reduction in a 0.75 MW boiler using natural gas reburning. The nitrogen content of the reburn fuel was adjusted by doping the natural gas with 0.4% nitrogen as ammonia. Results were interpreted in terms of the NOx reduction which was defined as the ratio of NOx emissions to primary NOx concentration ($\text{NOx}_{\text{em}}/\text{NOx}_{\text{pf}}$). When NO$_{\text{pf}}$ was 155 ppm and $f_{RB}$ was 25%, the NOx reduction was 40% but decreased to only 10% when an equivalent fraction of the nitrogen doped natural gas was used. The effect of fuel quality was more pronounced at low NO$_{\text{pf}}$. At NO$_{\text{pf}}$ of 58 ppm and $f_{RB}$ of 25%, the NOx reduction was 25% but the NOx concentration increased by 125% when the nitrogen doped natural gas was used.

Thus, NOx reduction was strongly dependent on reburn fuel quality. Other studies have shown that hydrocarbon gases are more effective at reducing NOx than coals [34,35]. Specific reasons for the discrepancy in test results are not clear. Therefore, experimental tests are necessary to determine the extent that fuel type influences reburning effectiveness.

### 2.2.2 Reburn Zone Stoichiometry

Reburn zone stoichiometry, $\lambda_{RB}$, has a direct influence on fuel staging reactions by altering the fuel chemistry and CH$_4$, O, H and OH radical concentrations in the reburn zone. As discussed in Section 2.1, the CH$_4$ radicals are essential in reducing primary zone NOx. A sufficient quantity of O and H radicals are necessary in the reburn zone to form CH$_4$ radicals and to convert HCN to NCO.
Figure 2.4 Effect of NOx on fractional NOx reduction [33]

Lanier et al [36] studied NOx reduction in CH₄ flames with λ₀ of 0.8 and concluded that less than 1 ppm of CH radical was necessary to produce their observed reduction of NOx to HCN according to Reaction R23. A 1 ppm concentration of CH radicals is viable in fuel rich flames. Thorne et al [23] measured approximately 1 ppm CH in acetylene flames at 1000°C and λ₀ of 0.6, and Parramayon et al [37] predicted approximately 15 ppm CH radical in methane flames at 2100°C and λ₀ of 0.7.

The fraction of CH radicals appears to directly increase with the reburn fuel fraction, f₀. Roby and Bowman [24] investigated the effect of small hydrocarbon additions into H₂/O₂ flames. They calculated that CH and CH₂ radical concentrations were proportional to reburn fuel fraction in the range 300 - 2000 ppm. Although the concentration of CH₂ was one order of magnitude higher than CH, NOx reduction was sensitive to the
concentration of both radicals.

It has been observed that there is a lower limit of $\lambda_{RB}$ suitable for NOx reduction. This limit is due to a decrease in O concentration at low $\lambda_{RB}$ which inhibits the conversion of HCN to NCO and NH (Reactions R1 and R2). Song and Bartok [38] measured the reduction of total volatile fixed nitrogen (TVFN: the sum of NOx, HCN and NH$_3$) in a reducing environment by injecting a controlled flow of O$_2$ into a preheated flow containing 75 - 150 ppm of TVFN (primarily HCN). At 1575°C, the TVFN decreased by 80% in 100 ms when 0.5% O$_2$ by volume was added. At higher O$_2$ injection concentration, the TVFN increased slightly and the NOx fraction increased sharply. Therefore, a carefully controlled fraction of oxidant in the reducing zone may be essential to maximize NOx reduction to N$_2$.

The $\lambda_{RB}$ necessary to minimize NOx emissions in coal flames is not well defined. In laboratory scale tests, Chen et al [31] showed that the optimum $\lambda_{RB}$ was 0.9 for $f_{RB}$ of 20%. At $\lambda_{RB}$ less than 0.9, there was a small increase in the flue NOx emission due to insufficient HCN oxidation (Reactions R1 and 2). At $\lambda_{RB}$ higher than 0.9, there was excessive oxidation of NCO to NO (Reactions R3-R5).

In contrast, Leikert et al [12] studied the effect of $\lambda_{RB}$ on NOx emissions in a 15 MW pulverized coal combustor. At $f_{RB}$ of 30%, the lowest NOx emissions were measured at $\lambda_{RB}$ of 0.65, Figure 2.5. In these tests, the level of NOx emissions was more sensitive to $\lambda_{RB}$ when a shorter reburn zone was used. At a reducing zone length of 0.776 m, the NOx emission decreased from 400 ppm at $\lambda_{RB}$ of 0.8 to 160 ppm at $\lambda_{RB}$ of 0.65. At a reducing zone length of 1.176 m, the NOx emission decreased from 230 ppm at $\lambda_{RB}$ of 0.79 to 175 ppm at $\lambda_{RB}$ of 0.65.

Experimental configuration may account for the differences in conclusions in the work of Chen et al [31] and Leikert et al [12], respectively. Chen used a one-dimensional furnace where the reburn fuel could rapidly mix with the primary combustion products and attain a relatively homogeneous gas mixture of low stoichiometry. Therefore, an optimum NOx reduction was obtained at an intermediate $\lambda_{RB}$ without excessive HCN or NH$_3$ formation. At the larger scale (15 MW) used by Leikert, the mixing was relatively slower compared to the rate of chemical reactions and the reburn zone may not have been homogeneously mixed. Fuel lean as well as fuel rich conditions may have existed at different positions in the flow. Thus, a lower $\lambda_{RB}$ resulted in fuel rich conditions in a larger portion of the reburn zone.
In both reports, approximately 40% reductions in NOx emissions were possible suggesting that an optimum $\lambda_{RB}$ was not necessary to apply reburning in pulverized coal combustion. Thus, some latitude exist in the choice of $f_{RB}$ and $\lambda_{RB}$ for designing reburning systems.

2.2.3 Residence Time

The residence times in the primary and reburn zones have been shown to influence fuel staging NOx reduction. Longer time in each zone enhances NOx reduction by increasing the fraction of primary fuel-N reacting with reburn fuel CH$_4$ radicals.

Chen et al [31] showed that primary zone residence times must be sufficiently long to ensure the complete combustion of primary fuel. Increasing the primary zone residence time from 70 to 300 ms lowered the NOx emissions from 300-700 ppm to 250-400 ppm.
Complete combustion of the primary fuel lowered the O₂ concentration entering the reburn zone. When primary zone combustion was incomplete, excess O₂ was available to oxidize reburn fuel CH₄ radicals. Complete combustion of the primary fuel was also necessary to ensure that primary fuel-N was completely converted to gas products. If the char-N was not converted entering the reburn zone, then it persisted to the tertiary air injection zone, where it was reoxidized to NOₓ. However, when the char-N was converted to either NOₓ or N₂ in the primary combustion zone, then it could be reduced to N₂ in the reburn zone, thereby lowering direct NOₓ formation from primary char-N in the tertiary zone.

The reburn zone residence time needs to be sufficiently long to ensure adequate mixing and chemical reaction. Lanier et al [36] studied NOₓ reduction in a well mixed reburn zone with a temperature of 1225°C, inlet NOₓ concentration of 49-206 ppm and nitrogen doped natural gas reburn fuel. The time required for 50% NOₓ reduction was 50-75 ms, Figure 2.6. However, the TVFN concentration was only reduced for tests conducted with an inlet NOₓ concentration of 206 ppm. Upon introducing tertiary combustion air after the reburn zone, most of the HCN and NH₃ were converted to NOₓ. Thus, significant conversion of TVFN to N₂ could not be obtained in 75 ms.

Figure 2.6 Total volatile fixed nitrogen, TFN, decay in nitrogen doped methane flames [36]
It has been suggested that the reburn zone residence time should be at least 400 ms to minimize NOx emissions [31,39,40]. A residence time between 75 and 400 ms is necessary for HCN conversion to NH3 and the subsequent reduction to N2. Miller et al [41] studied HCN reduction in laminar H2 flames operating at a temperature of 1000-1125°C, a pressure of 25 Torr and a stoichiometry of 0.67. When HCN was injected at a volumetric fraction of 2%, the concentration decreased by 50% after 50 ms and by 90% after 100 ms.

2.2.4 Reburn Zone Temperature

Reburn zone temperature, $T_{RB}$, influences NOx reduction by its effect on radical species concentrations and reaction rates in the reburn zone. High $T_{RB}$ promotes the formation of O, H and OH radical concentrations that are necessary for the oxidation of volatile fragments and conversion of intermediate N-species to N2. Thorne et al [23] showed that the formation of CHx fragments from acetylene flames due to reaction with O radicals was strongly temperature dependent. The concentration of CH fragments doubled when the temperature in the flame zone increased from 725°C to 925°C.

Higher $T_{RB}$ also increases the rate of NOx reduction in the reburn zone under fuel rich conditions through faster HCN oxidation (Reactions R1 and R2) and O and OH radical formation (Reaction R25). The rates of Reactions R1, R2 and R25 increase by approximately 30% with a temperature increase of 100°C from 1300 to 1400°C.

Farnamayan et al [37] modelled the effect of reburn zone temperature on fuel staging effectiveness in methane flames using the CHEMKIN kinetics model which was also used by Thorne et al [23]. Farnamayan et al showed that the rate of NOx reduction was insignificant at 1425°C, with minimal NOx reduction expected in 400 ms. However, at 1725°C, the rate was significantly higher and an order of magnitude decrease in NOx concentration from 1000 to 100 ppm was calculated in 1.2 s and $\lambda_{RB}$ of 0.75.

High $T_{RB}$ was not always necessary to obtain significant NOx reduction. Takahashi et al [11] demonstrated significant NOx reduction in staged fuel combustion at temperatures below 1200°C. Using propane reburn fuel at $\lambda_{RB}$ of 0.53, the NOx reduction increased from 60% at 900°C to 90% at 1150°C.
Chen et al [31] investigated the effect of reburn zone temperature on NOx reduction using propane and Beulah lignite coal. For propane reburn fuel, a drop in the reburn zone temperature from 1200 to 930°C resulted in a 50 ppm increase in NOx emissions from 200 to 250 ppm, Figure 2.7. This increase was directly related to an increase in TVFN leaving the reburn zone. However, when Beulah coal was injected, the NOx emission decreased from 250 ppm at 1200°C to 80 ppm at 930°C. For this particular coal, a large fraction of the fuel-N evolved as NH3 which reduced the NOx in the tertiary combustion zone. Therefore, results for the effect of reburn zone temperature using hydrocarbon gases may not be generally applicable to coal reburn fuel. In particular, lower reburn zone temperature may be optimum due to reaction of NH3 with NOx. This hypothesis should be tested in laboratory experiments.

![Figure 2.7 Effect of reburn temperature on NOx emissions in propane and coal flames [31]](image)

2.2.5 Mixing Rate

Reburn fuel mixing has also been identified as an important parameter in fuel staging. However, mixing intensities have not been measured in most tests, and therefore, characteristic mixing times cannot be estimated. For example, Chen et al [31] showed that NOx reduction increased from 40% to 60% when natural gas was injected into the reburn zone on a line source rather than a single jet. However, the characteristics of the jets and the mixing pattern were not reported.
At full scale, rapid mixing of reburn fuel with primary combustion products may pose a significant practical problem. Mixing may be enhanced by using recirculated flue gas to inject the reburning fuel. This would allow the secondary fuel momentum to be increased without increasing O₂ availability in the reburn zone. Kiga et al [42] showed that NOx emissions could be reduced from 140 to 110 ppm when the secondary fuel was injected with flue gas instead of air. This process option may be useful in utility boilers which already utilize flue gas recirculation.

2.2.6 Fuel-N content

When a fuel containing nitrogen is injected into the reburn zone, the total concentration of N-species increases. However, disagreement prevails concerning the influence of fuel-N on NOx reduction. Takahashi et al [11] showed that NOx reduction was independent of fuel-N content for gas and solid reburn fuels. This conclusion was disputed by Mulholland and Hall [39] in tests with natural gas doped with ammonia. For all λRB and inlet NOx concentrations tested, the NOx emission increased when ammonia was added to the natural gas, Figure 2.4 (p.30).

Chen et al [31] made a detailed study of the effect of coal nitrogen content on fuel staging NOx reduction in a one-dimensional reactor. The exit NOx concentration was compared with the total available nitrogen (defined as inlet NOx, (NOx)p, plus reburn fuel-N, (N)coal) for four coals, Figure 2.8. At λRB of 0.7 - 0.9, 25% of the total available nitrogen was converted to NOx. The source of the nitrogen in the NOx was indistinguishable and thus emissions could be predicted based on the N-species concentration entering the reburn zone.

Chen et al [31] also showed that fuel staging was ineffective when the inlet NOx concentration to the reburn zone was less than 150 ppm. These results contradicted Takahashi et al [11] who showed significant NOx reduction with primary NOx concentration of 150 ppm. It should be noted that Chen et al used a reburn zone temperature of 1400°C compared to 900 - 1200°C for Takahashi et al. The difference in staging temperature may have influenced the NOx reduction, as discussed in Section 2.2.4 (p.34).
2.2.7 Volatile matter content

High volatile coals tend to be more effective in reducing NOx than low volatile coals. Leikert et al [12] studied NOx reduction in a fuel staged burner using four coals of various rank. They reported a linear correlation of volatile matter with NOx emissions. The effectiveness of brown coal to reduce NOx emissions was a factor of two times higher than Ruhr medium volatile bituminous coal, Figure 2.9. Reburning using two high volatile bituminous coals resulted in NOx emissions between these two extremes. The lower emissions obtained using higher volatile coals may be due to a higher concentration of CH₃ radicals in the reburn zone.

The volatile matter content may affect the fraction of coal nitrogen released in the reburn zone. Pohl and Sarofim [15] showed that the fraction of nitrogen released from a lignite and a high volatile bituminous coal under rapid heating was similar to their respective volatile yields. Approximately 50% of the nitrogen remained in the chars at a stoichiometry of 0.67 and particle temperature of 1225°C but the nitrogen retention in the char decreased in leaner flames where combustion temperatures were higher.
Figure 2.9 Effect of reburn coal volatile matter content on NOx emissions [12]

At 1475°C, and a stoichiometry of 0.67, Peck et al [43] showed that only 10% of the nitrogen remained in a subbituminous coal char compared to 20% retention of nitrogen in the char at a gas temperature of 1300°C and a stoichiometry of 0.33. Although the effect of temperature on nitrogen retention was similar in these two studies, the fraction of nitrogen retained was significantly different.

A detailed study of the effect of coal type on nitrogen retention in pyrolysed chars was conducted by Freihaut and Seery [18,19]. They showed that eastern and western US coals, ranging in rank from subbituminous to medium volatile bituminous, had different nitrogen release yields. The western subbituminous and high volatile bituminous and eastern medium volatile bituminous had nitrogen retentions decreasing from 80% at 600°C to 20% at 1400°C. However, eastern high volatile bituminous coal had a nitrogen retention decreasing from 100% at 600°C to 30% at 1400°C.

In the tertiary combustion zone, the nitrogen retained in the char may be converted directly to NOx. Thus, it is beneficial to minimize the fraction of char remaining after the reburn zone.
2.3 Process Development

The most extensive full scale test on fuel staging for pulverized coal combustion was conducted on the MACT process by Mitsubishi Heavy Industries Ltd. [11]. A tangentially fired twin boiler rated at 23 MW and operating on subbituminous coal was modified to include a fuel staged zone at the top burner row. The process was tested in three independent programs using coal, fuel oil and natural gas as the reburn fuel. The NOx emissions were approximately 100 ppm (at 0% O2) for all reburn fuels, representing a 50% emission reduction. No operational difficulties were reported in the demonstration and the unburned carbon in ash was 0.4% with or without the MACT process operating.

Fuel staging was also tested by Ishikawajima-Harima Heavy Industries Co. Ltd., (IHI) using wall fired 55 MW and 600 MW boilers [40]. The 55 MW boiler was fired with subbituminous coals through nine coal burners. The reburn fuel fraction, f_{RB}, was normally less than 10%. For this low f_{RB}, the primary zone needed to be operated fuel rich in order to achieve significant NOx emission reduction.

For the 600 MW boiler, coal was injected in the second of three elevations of primary burners. The first and the third rows as well as the reburn burners were fired on fuel oil. The reburn fuel oil was injected along a narrow spray angle (30°) in order to penetrate through the entire furnace cross-section and mix with primary combustion products. When fuel staging was used, the furnace could be operated at a higher primary stoichiometry compared to conventional air staging (0.9 vs 0.8) to obtain NOx emissions of 100 ppm (0% O2). It was concluded that fuel staging offered significant advantages over conventional air staging - both in more flexible primary flame operating conditions and lower NOx emissions.

The flexibility of fuel staging to reduce NOx was also demonstrated by Hitachi Zosan in a 17 MW wall fired boiler [44]. The primary combustion zone was operated using standard burners with a stoichiometry of unity and NOx concentration of approximately 1000 ppm (0% O2). The NOx emissions were reduced to 150 - 200 ppm using fuel staging.

Steinmüller have conducted the only reported tests using a coal-fired fuel staged burner [12]. Preliminary demonstration experiments were undertaken in a 15 MW boiler using a burner of the type shown in Figure 1.2 (p.13). The reburn fuel was injected directly
above the quarl and mixed with primary combustion products in the tail of the flame. This method of fuel injection made the fuel staged burner distinct from furnace staging where the reburn fuel was injected from a single elevation above all of the primary burners. It was shown that NOx emissions decreased from 240 to 170 ppm as \( f_{RB} \) was increased from 25 to 40%. A long reburning zone residence time on the order of one second was required to reduce the HCN to N\(_2\) before injecting tertiary air.

Thus, reburning has been demonstrated as a viable method of reducing NOx emissions in coal-fired boilers operated with furnace and burner fuel staging.

2.4 Fuel Staging in Utility Boilers

Although fuel staging has been developed for industrial boilers as a proven method of NOx reduction, it has not been widely applied in new boiler designs nor has the process been retrofit into existing pulverized coal fired boilers. The MACT system has been incorporated into five new pulverized coal utility boilers to the end of 1986 [11] but there are no plans to retrofit any coal fired units. This technology optimally requires a single reburn zone above the primary combustion zone. However, this tends to lower the firing density in the lower half of the furnace and may reduce the coal burnout and the efficiency of the steam cycle. Thus, fuel staging may be more effective in new furnaces with larger volumes where significant NOx reduction and acceptable coal burnout may be attained at the lower firing density.

The retrofit of reburning into existing utility boilers has been studied by Lisauskas et al [45] and Leikert et al [12]. They both concluded that the application of fuel staging and its configuration in a boiler may be site specific, requiring compatibility with the existing furnace design and operation and coal type.

2.4.1 Coal Burnout

The retrofit of fuel staging in pulverized coal fired boilers may be limited by coal burnout requirements. Leikert et al [12] reported that the carbon in ash increased with the rank of the coal. Reburning with Ruhr mvb coal resulted in 16 - 20% carbon in ash compared to 4 - 8% carbon in ash for brown coal. Two high volatile bituminous coals had carbon in ash between these two extremes. The carbon in ash for bituminous coals
was considered unacceptable and more work was planned to improve the combustion efficiency.

In contrast, Miyamae et al [40] showed that, for a given NOx emission, lower carbon in ash was obtained from fuel staging than conventional air staging. The carbon in ash from reburning remained at 3-4% for NOx emissions between 140 and 280 ppm. In air staging experiments, carbon in ash increased from 3 to 6% when NOx emissions were lowered from 450 to 280 ppm.

Lisauskas et al [45] noted that carbon burnout was directly related to the tertiary zone temperature and residence time which were influenced by the primary zone stoichiometry and the quantity of reburn fuel. They concluded that higher reburn fuel fraction may result in higher carbon in ash since a larger percentage of the combustion would occur in the tertiary combustion zone.

Coal burnout could also be improved by using finer reburn coal. Leikert et al [12] showed a slight reduction in NOx emissions from 180 to 160 ppm when the fraction of coal smaller than 75 μm was increased from 80 to 88%. The finer coal appeared to enhance the char combustion rate in the tertiary combustion zone and improved the fuel staging effectiveness by increasing the fraction of CH4 released in the reburn zone.

2.4.2 Boiler operation

Boiler performance criteria often requires that a stable flame is produced for turn-down to one-third load. Murakami [46] reported excellent turn-down performance to one-third load on a tangentially fired MACT process boiler operated with a coal/oil mixture. In this unit, less than 10% of the fuel oil was injected with recirculated flue gas into the reburning zone. The NOx emissions were nominally 60 - 100 ppm (0% O2) and the carbon in ash was 1-3%.

At high reburn fuel fraction, it may be more difficult to maintain acceptable burner performance. Leikert et al [12] noted deteriorated performance when the load of their fuel staged burner was reduced. The primary and reburn zone stoichiometries increased and the NOx emissions increased sharply from 110-175 ppm at 70% burner load to 330-410 ppm at 50% burner load. Under reduced load conditions, the shape of the flame was difficult to maintain. Therefore, it appears that reburning fuel fraction should be
minimized for performance considerations as well as for improved carbon burnout.

2.4.3 Furnace Design

It is preferred that fuel staging burners be compatible with the existing furnace design to ensure successful retrofit. This requires that the fuel and air be delivered through existing supply systems and that the steam cycle should not be adversely affected by altered heat flux profiles.

In furnace fuel staging, retrofit would be best achieved by converting the top elevation of burners to obtain a substoichiometric reburn zone. This method assumes that adequate mixing of the reburn fuel and primary combustion products can be achieved. However, it may not always be possible to obtain complete mixing of the reburn fuel across the furnace section. The size of the forced air fan is normally designed for a transport velocity of 20 m/s and limits the air pressure available for transporting coal to the burner. Reburn fuel may need to be injected at a velocity greater than 20 m/s in order to penetrate across the furnace section and mix with primary combustion products. This, in turn, may require a delivery pressure which exceeds the limits of the forced air fan. Kiga et al [42] stated that fuel spreading limited the potential of NOx reduction by fuel staging in a furnace.

NOx reduction is very sensitive to reburn zone stoichiometry, and thus, the fuel and air distribution to each burner needs to be carefully controlled. This may be difficult to achieve in existing boilers where sophisticated fuel and air distribution systems are not available. The pipe work could be modified and control systems installed. The alternative is to minimize the sensitivity of NOx emissions to reburn zone stoichiometry.

2.5 Swirling Flow Burners

The swirl stabilized burner was considered as the basis for a fuel staged burner. Commonly found in industrial and utility boilers, swirl stabilized burners generate compact and intense flames close to the furnace wall. They may be utilized with a variety of fuels and operating conditions. As well, flame shape and flow patterns may be adjusted to obtain the required heat flux distribution and flame length for low NOx
emissions. Thus, a swirl burner may be adjusted over a wide range of operating conditions. The principles of swirl burner operation and how the operation influences NOx emissions are discussed in this Section.

2.5.1 Swirling Flame Aerodynamics

When air is introduced tangentially into a burner pipe, a spiral flow is established. A balance is then created between the centrifugal force of the spiraling flow and the pressure forces, P, acting on the pipe wall according to:

\[
\frac{dP}{dr} = \frac{\rho_1 w^2}{r}
\]  (2.11)

where \( \rho_1 \) is the gas density, \( w \) is the tangential velocity and \( r \) is the distance from the burner centerline. At the exit of the burner throat, Figure 2.10, the flow expands resulting in a low pressure region in the center of the flow. The negative pressure gradient slowly recovers downstream of the burner and the flow becomes uniform. If the tangential velocity of the air in the burner is sufficiently high, the negative pressure gradient along the burner axis cannot be overcome by the kinetic energy and the flow is reversed. An IRZ is formed which has the shape of a torroidal vortex.

The formation of an IRZ is related to the swirl number of the flow and the burner geometry. In this study, axial pressure gradients are neglected and the modified swirl number, \( S' \), relating the ratio of angular momentum, \( G_\phi \), to axial momentum, \( G_x \), is defined by [47]:

\[
S' = \frac{2G_\phi}{aG_x}
\]  (2.12)
where \( a \) is the diameter of the secondary air duct. For flow in a straight pipe exiting into a free expansion, it has been observed that an IRZ was formed only for \( S' > 0.6 \) when the axial pressure gradient was sufficient to cause recirculation [47]. Beér and Chigier [47] noted that the geometry of the nozzle (quarl) had an influence on the size of the IRZ. A divergent nozzle controlled the expansion of the jet and increased the recirculated mass flow rate.

Hagiwara et al [48] studied the velocity field in cold swirling flows emerging from quars with different expansion ratios, \( b/a \). They observed that the recirculation increased from 0 to 100% as the parameter \( S'(b/a)^2 \) increased from 0 to 16, Figure 2.11. Although recirculation increased with swirl, the strength along the centerline passed through a maximum and decreased. This was attributed to an increasing IRZ diameter with higher swirl.

The recirculated mass flow rate is a measure of the IRZ strength and is an important property of a swirl burner. In a flame, fuel ignition is accelerated by rapid heat transfer from recirculated hot combustion products to the incoming fuel and air. Thus, flames tend to be more stable with higher recirculated mass fraction [49,50].
Fuel is normally injected from a gun centrally located within the secondary air pipe. This gun provides a convenient location for fuel injection and helps to stabilize the flame by reducing the minimum swirl necessary to form an IRZ. Schmid et al [51] studied the flow pattern of primary air jets injected into cold swirling flows for different gun blockage ratios, $D_p/a$, jet momentum, $G_p$, and air injector design. They identified five distinct flow patterns ranging from complete primary jet penetration of the IRZ, Type 1, to rapid entrainment of the primary air into the secondary air flow, Type 2. IRZ strength increased with gun blockage ratio and a Type 1 flow at $D_p/a$ of 0.42 could change to a Type 2 flow at $D_p/a$ of 0.7. For conditions which allow jet penetration to occur, higher $G_p$ increased jet penetration. Physically separating the primary air jet and the annular swirling flow substantially increased the penetration distance, particularly for a blockage ratio of 0.7.

Weber and Dugué [52] studied the influence of the gun blockage ratio on the minimum swirl necessary to form an IRZ. They observed that the minimum swirl decreased from 0.6 to 0.0 as the blockage ratio increased from 0 to 0.7, Figure 2.12. The results of these aerodynamic studies are used in designing the fuel staged burner which is discussed in Chapter 7.
2.5.2 Influence of Combustion

Several studies have investigated the behaviour of swirling pulverized coal and gas flames [54-57]. For any given swirling flow, combustion reduces the mass fraction recirculated in the IRZ. When a swirling fluid parcel begins to combust, it accelerates axially due to the reduced gas density and increases the axially momentum of the flow. This reduces the swirl level and the radial expansion of the flow at the point of combustion. Weber and Dugue [52] studied the influence of combustion on IRZ strength for several fuel injection locations. They showed that, if combustion occurred upstream of the quarl exit, then the recirculation strength and the size of the IRZ were reduced, Figure 2.13. The size and strength of the IRZ in cold and combusting flows could be directly related if the swirl number was corrected to account for the axial acceleration due to increased gas temperature. Because large temperature gradients existed in the combusting flows, no mean temperature could be defined. The flame temperature at the position of the maximum recirculation, $T_m$, was chosen as representative. An effective swirl number, $S_{eff}$, related the cold and combusting swirling flows according to:
\[ S_{\text{eff}} = \frac{T_o S_o}{T_m} \]  

(2.13)

where \( T_o \) and \( S_o \) refer to secondary air inlet temperature and swirl number.

The IRZ strength was related to the burner geometry and \( S_{\text{eff}} \). The maximum recirculation did not change for \( S_{\text{eff}} \) in the range 0-0.5, but increased linearly at higher \( S_{\text{eff}} \). The recirculation strength could be raised by increasing the blockage ratio of the central gun. For \( S_{\text{eff}} \) less than 0.5, the recirculated mass fraction increased from zero with no blockage to 5% and 15% for blockage ratios of 0.42 and 0.7, respectively, Figure 2.12. The increase in IRZ strength with \( S_{\text{eff}} \) greater than 0.5 was more pronounced at the higher blockage ratio.

Figure 2.13 Effect of combustion front position on the swirling vortex [42]
Depending on the gun position and relative momentum of primary and secondary flows, the primary flow may be rapidly entrained into the secondary air (Type 2) or partially or fully penetrate (Type 1) through the IRZ. Weber and Dugué [52] observed the behaviour of annular primary air jets injected into combusting swirling flows. With a blockage ratio of 0.7 and the gas gun positioned at the quarl throat, the primary flow could not penetrate the IRZ for swirl greater than 0.4 and primary momentum of less than 0.4 N, Figure 2.14. They noted that the position of the gun within the quarl influenced the jet penetration. When the injector was moved half the quarl diameter, 100 mm, into the quarl, the transition from Type 1 to Type 2 flows occurred at a swirl level double that observed with the gun at the quarl throat.

Figure 2.14 Criteria for establishing Type 2 flames with annular fuel guns [42]

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Injecting the primary flow on the burner axis increased the potential for the jet to penetrate the IRZ. Central injection had a similar influence on jet penetration as moving an annular injector 100 mm into the quarl. For a 130 mm diameter gun with a 25 mm central opening, Weber [53] observed that cold primary jets penetrated the IRZ for swirl less than 0.4, Figure 2.15. Combustion lowered the recirculation strength and increased the ability of the central jet to penetrate the IRZ. In combusting swirling flows, primary jets, injected at the quarl throat, penetrated the IRZ for swirl less than 0.6.

Hillemans and Leuckel [58] studied the flow patterns in expanding swirling flows for cold and combusting conditions. They found a dramatic change in jet penetration with combustion. A natural gas jet easily penetrated the IRZ due to the axial acceleration of the combusting jet.
2.5.3 NOx Reduction in Coal Flames

Different swirling flow burner designs exist to assist in lowering NOx emissions in the flame. The principle of most swirl stabilized low-NOx burners is to combust the volatiles in an O₂ lean region close to the burner. This is accomplished by controlling the mixing of air and fuel in the flame using mechanical or aerodynamic methods. A variety of burners have been designed to separate fuel and air. These may be classified either as external air staged burners (EASB) or internal air staged burners (IASB).

An EASB has a fuel rich primary combustion zone with a secondary char burnout zone further downstream, Figure 2.16 [61,62]. The secondary air is rapidly mixed with the fuel to form a stable flame and the staged air is mixed with the partial combustion products after the fuel-N has been partially reduced to N₂. The stoichiometry and time

![Diagram of external air staged burner](image)

**External air staged**

![Diagram of internal air staged burner](image)

**Internal air staged**

Figure 2.16 Schematics of external and internal air staged burners

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available in the fuel rich primary zone has a strong influence on the NOx emissions. Phelan and coworkers [63-65] studied the influence of burner operating conditions on NOx emissions in an EASB for a wide range of coals. They showed NOx emissions decreased from 600-800 ppm to 200-400 ppm as $\lambda_p$ was reduced from 1.1 to 0.5, Figure 2.17.

In the IASB, all of the air is injected through the burner quarl and mixing of the fuel and air is controlled aerodynamically, Figure 2.16. By controlling the gun position and jet momentum, the fuel is injected directly into the $O_2$ lean IRZ. The coal is rapidly heated and fuel-N is partially converted to $N_2$ before mixing with the combustion air. Examples of these type of burners are the IFRF Aerodynamically Air Staged Burner (AASB) [66], Foster Wheeler dual throat burner [67] and Babcock Hitachi HTNR [68].

Smart et al [66] studied the influence of operating characteristics and coal type on the NOx emissions from the AASB burner. The trajectory of the coal jet was altered from rapid entrainment in the secondary air (Type 2) to partial or full penetration of the IRZ.
top  Type 2, high NOx flame

bottom  Type 1, low NOx flame

Figure 2.18  Temperature and species concentrations in flames from the AASB [56]

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(Type 1) by inserting the coal gun into the quarl. With the gun positioned at the burner throat and coal injected at 20 m/s, the coal rapidly mixed with the secondary air forming a short intense flame and the NOx emissions were approximately 1000-1300 ppm, Figure 2.18 top. By inserting the gun 0.4-0.6 quarl diameters, a longer, less intense flame was formed and NOx emissions of 400-500 ppm were measured, Figure 2.18 bottom.

The reduction in NOx emissions for the AASB were directly related to the particle paths through the flame. If the particles devolatilized in a fuel rich zone, the fuel-N tended to be reduced. If the particles were directly entrained into the secondary air, NOx was rapidly formed from the fuel-N. These tests highlighted the sensitivity of NOx formation to local stoichiometry during devolatilization and volatile combustion. They also showed that the IRZ may be used to preheat and ignite the fuel without forming NOx. These phenomena are important considerations in the design of the fuel staged burner.

2.6 Concluding Remarks

Reburning has been demonstrated as an effective method of NOx reduction. The processes occurring in fuel staging have been extensively studied using gas flames. However, the applicability of these results to coal reburning is not clear. Contradictory conclusions in the literature regarding the influence of process parameters on NOx reduction need to be considered in depth before designing a fuel staged burner.

In utility boilers, NOx emission reductions of 50% to 85-200 ppm have been demonstrated. However, operational difficulties, such as effective mixing of reburn fuel and primary NOx, burnout of the reburn fuel and retrofit to existing boilers, have been encountered.

Many of these problems may be reduced by using fuel staged burners. The reburn fuel is injected adjacent to each primary flame to optimize mixing of the reburn fuel and primary NOx. As well, burnout problems are minimized since the reburn fuel is injected low in the boiler.

A swirling flow burner is considered as the basis for the fuel staged burner as it has the potential of reliable operation over a wide range of conditions. Significant NOx
reduction may be achieved using swirling flow burners by combusting the fuel in an oxygen lean internal recirculation zone. This behaviour will be utilized in the design of the fuel staged burner.
3. Method

The review of literature related to reburning indicated the process parameters which had the most significant influence on NOx reduction. However, there was no apparent consensus in the literature concerning the influence of these parameters in coal reburning. Optimum process conditions required to operate a fuel staged burner and to minimize NOx emissions could not be readily identified. Thus, a study of the effect of the process parameters on NOx reduction was necessary.

Two investigations were undertaken to determine the influence of process parameters on NOx reduction in coal reburning. The first investigation, Reburn Zone Investigation, was a study of NOx and TVFN reduction in the reburn zone. The Reburn Zone Investigation was conducted in the Isothermal Plug Flow Reactor (IPFR) under well-mixed conditions to determine chemical processes occurring in the reburning process. The equipment design, operation and experimental program are described in Section 3.1.

The second investigation, Furnace Investigation, involved studying NOx reduction for mixing limited conditions. To characterize the mixing in the Furnace Investigation, mathematical modelling, water model flow visualization and CO2 tracer studies were conducted. The furnace arrangement and mixing characterization test methods are described in Section 3.2. The Furnace Investigation was conducted in a 2.5 MW, semi-industrial scale furnace (Furnace No. 1) which was configured to simulate a fuel staged furnace with separate primary, reburn and tertiary combustion zones. Process parameters for each zone were varied independently to determine their influence on NOx emissions. The furnace was operated at approximately the optimum conditions identified for NOx reduction in the Reburn Zone Investigation. The test procedure is described in Section 3.3.

From the results of these two investigations, a burner was proposed, designed and then tested in Furnace No. 1. The influence of primary, reburn and tertiary combustion zone process parameters on NOx emissions using the burner were evaluated. The experimental configuration is described in Section 3.4.

Computational modelling was used throughout the investigations to predict flow
patterns and estimate mixing rates. The flow from the Fuel Staged Burner was modelled to describe flame shape and mixing rates between the primary combustion products and reburn fuel jet. The mathematical model is described in Section 3.5 and details of the computations conducted for the IPFR, Furnace No. 1 and Fuel Staged Burner are given in the pertinent sections.

3.1 Reburn Zone Investigation

Six parameters were considered to have a dominant impact on reduction in the reburn zone:

- coal type,
- primary stoichiometry, \( \lambda_p \)
- reburn fuel fraction, \( f_{RB} \)
- temperature, \( T_{RB} \)
- residence time, \( \tau_{RB} \)
- primary zone NOx concentration, \( NOx_p \).

In addition, coal particle size distribution (PSD) was considered due to the potential that smaller PSD may have on improving coal burnout. The independent and interactive influence of these parameters on NOx and TVFN reduction were studied.

The tests were conducted under well-mixed conditions using the isothermal plug flow reactor, IPFR, Figure 3.1. Primary combustion products were generated in a precombustor with a natural gas flame. The reburn zone was produced by mixing coal with the primary combustion products in the isothermal section of the reactor. The gas composition was measured along the reactor length to determine the concentrations of NOx, HCN and NH3 and the fractional NOx and TVFN reduction in the reburn zone.

3.1.1 Experimental Procedure

The IPFR consisted of three sections: precombustor, main reactor and product collector. The precombustor was a 30 cm diameter by 1 m long refractory lined cylinder with a swirled gas burner at one end and a T-piece and chimney at the other end. Natural gas, air, \( N_2 \), \( O_2 \), and NO were supplied to the precombustor as shown in Figure 3.1.
Combustion products were generated by burning natural gas in air with \( \text{N}_2 \) added to moderate the temperature. The combustion products entered the T-piece where a fraction of the flow was diverted through the main reactor. The combustion products flowed through a 22 mm annulus around the coal feeder probe which was fixed within the T-piece. The feeder probe was 75 mm in diameter and consisted of a water-cooled tube, insulated on the outside to limit cooling of the combustion products.

The combustion products were intended to replicate the gas composition from a pulverized coal burner. It was necessary that the natural gas be completely burned so that the only products entering the furnace were NOx, \( \text{N}_2 \), \( \text{CO}_2 \), \( \text{H}_2\text{O} \), \( \text{O}_2 \) and less than 300 ppm of CO. If the natural gas combustion had been incomplete, primary zone hydrocarbon radicals and soot could have reacted with the NOx. This could have influenced NOx reduction in the reburn zone.
The main reactor consisted of a 117 mm ID by 2 or 5 m long alumina tube heated externally by graphite heating elements. The temperature along the outer face of the tube was measured using S-type thermocouples and controlled by six independent temperature controllers. The tube and heating elements were contained within Safield board insulation and a steel housing. The reactor was connected to the preheater T-piece by a ceramic venturi which had a throat area 2.1 times smaller than the reactor tube area, Figure 3.2. The combustion products flowed into the main reactor around the feeder probe which was positioned on the reactor axis, 17 mm upstream of the venturi throat. The gas flowed out of the reactor through the ejector and was vented. Collection probes could be installed at any position in the tube and were inserted through the bottom of the reactor and fixed with a clamping bracket.

The flow rate through the main reactor, $Q_a$ in Figure 3.2, was set to obtain a velocity of 5 m/s by adjusting a compressed air ejector located at the bottom of the main reactor. For a reactor temperature of 1100°C, the flow rate was 31 Nm$^3$/h and for 1400°C the flow rate was 25.5 Nm$^3$/h. The reactor gas flow rate, measured using a five hole Prandtl tube inserted at the bottom of the reactor, was only measured after adjustments to the furnace temperature, air or gas flow rate were made. A continuous indication of the reactor gas flow rate was provided by a pressure indicator installed between the reactor and the T-piece. For a given set of operating conditions the pressure at the top of the reactor was set during Prandtl tube measurements and maintained throughout the tests.

Coal was transported with 1.0 Nm$^3$/h N$_2$, $Q_i$ in Figure 3.2, and injected into the reactor through the feeder probe at a velocity of 5 m/s. The coal feed rate was controlled using a dual auger screw feeder (KTRON Soder AG). The coal feed rate, $m_{RB}$, was determined by calculating the amount of coal necessary to obtain the required reburn fuel fraction,

--- grid approximation

![Figure 3.2 IPPR reactor geometry around the venturi and grid approximation for the flow simulations](image-url)
$f_{RB}$, in the reactor. This calculation was based on the ratio of O$_2$ in the preheated gas as CO$_2$ and H$_2$O and the O$_2$ requirement for coal combustion according to:

$$m_{RB} = \frac{m_{NG}}{\Phi_c} \frac{f_{RB}}{1 - f_{RB}}$$  \hspace{1cm} (3.1)

where $m_{NG}$ referred to the mass flow rate of combustion products, $f_O$ was the mass fraction of oxygen in the combustion products as CO$_2$ and H$_2$O and $\Phi_c$ was the O$_2$ requirement for complete combustion of the reurn coal. Using Equation 3.1, the coal feed rate normally varied between 0.25 and 1.0 kg/h.

A small amount of NO was added to the gas in order to obtain a predetermined concentration of NOx in the reactor. Initially, the NO was added to the preheated gas in the precombustor section. However, this resulted in excessive consumption of NO due to the venting of a large portion of the NO through the chimney. Subsequently, the NO was injected into the reactor through the feeder probe. Several tests were conducted varying the injection location. It was shown that the injection location had no significant effect on NOx reduction.

The coal and NO were intended to mix rapidly with the combustion products inside and downstream of the venturi to ensure rapid dispersion and heat-up. A study of the mixing and particle heat-up was conducted using numerical modelling and is discussed in Section 3.1.3 (p.62).

The reactor gas temperature was measured using a suction pyrometer downstream of the venturi. During temperature measurement, the feeder probe nitrogen and coal flow were terminated to alleviate the problem of sampling from a cold and hot flow. The reactor gas temperature with the coal and transport nitrogen flowing was calculated from an enthalpy balance. The relationship is demonstrated in the following example. At a reactor temperature of 1400°C, addition of the coal transport nitrogen lowered the gas temperature by 55°C. Therefore, the preheated gas temperature, measured using the suction pyrometer, was set at 1455±20°C with no flow through the feeder probe.
3.1.2 Description of Sampling Probes

The IPFR collection system consisted of a sampling probe, solids filter, sampling pump, condenser and continuous analysers, Figure 3.3. Only one probe could be inserted in the reactor at any given time. Therefore, the permanent gases (NOx, O₂, CO₂ and CO) had to be measured separately from the HCN and NH₃ species. The permanent gases were measured for all tests using a steam heated NOx probe, illustrated in Figure 3.4. The sample was aspirated into the probe and the solids removed in a steam heated filter pot. The sample passed through a sampling pump and into a refrigeration unit which removed moisture. The dried gas flowed through a series of continuous analysers to measure species concentrations (O₂ paramagnetic; CO and CO₂ infrared; NOx chemiluminescence).

![Diagram](image)

*Figure 3.3 Process and instrumentation diagram of the product collection system*
The HCN and NH₃ concentrations were measured using the HCN/NH₃ probe. The sampling probe tip, which was connected to a standard 43 mm OD water-cooled jacket, is shown in Figure 3.5. A detailed description of the probe operation is given in Reference 69. The analysis of HCN and NH₃ were performed according to NEN 6489 and NEN 6472/6644 standards for measurement in waste water [70]. During the HCN and NH₃ analyses, a steam heated NOx probe located at the bottom of the reactor was connected to the continuous analysers for NOx and permanent gas measurement. The HCN/NH₃ probe tended to cool the reactor and all of the gases flowing downstream of the measurement point thereby "fixing" the reaction products. This allowed a comparison of the NOx measured with the NOx probe in the reactor and NOx measured with the HCN/NH₃ probe in the reactor.

---

**Figure 3.4** Schematic of the steam heated NOx probe

**Figure 3.5** Schematic of the HCN/NH₃ probe tip
The H₂ concentration was measured for selected tests using a Perkin Elmer Sigma 1B gas chromatography equipped with a thermal conductivity detector. Solid samples were collected in other selected tests using the IPFR solid sampling probe, Figure 3.6.

3.1.3 Computational Modelling of the Reactor

The flow and thermal characteristics of the reactor were modelled using a finite difference code which is described in detail in Section 3.5, p.76. The equations of continuity, momentum and energy were solved using finite difference analogues. Convective fluxes in the transport equations were obtained using a quadratic upstream differencing scheme and the radiative heat transfer was modelled using the four flux method. Particle devolatilization rate and yield were assumed to be the same as measured in coal characterization experiments [16]. The entrance region, including the venturi and feeder probe were modelled using a fine resolution stepwise grid. The model predictions had been validated in previous cold flow velocity measurements [71].

The input conditions are given in Table 3.1 and a description of the variables may be found in Figure 3.2. The temperature and the preheated gas velocity in the reactor were set to 1400°C and 5 m/s, respectively.
Table 3.1 Process conditions for the IPFR numerical modelling

<table>
<thead>
<tr>
<th>Reactor dimensions*</th>
<th>( d_R ) (mm)</th>
<th>112</th>
</tr>
</thead>
<tbody>
<tr>
<td>venturi diameter,</td>
<td>( d_V ) (mm)</td>
<td>80</td>
</tr>
<tr>
<td>injector diameter,</td>
<td>( d_I ) (mm)</td>
<td>10</td>
</tr>
<tr>
<td>inj. outer dia.,</td>
<td>( d_O ) (mm)</td>
<td>80</td>
</tr>
<tr>
<td>gun offset,</td>
<td>( d_B ) (mm)</td>
<td>16</td>
</tr>
<tr>
<td>reactor length,</td>
<td>( L_R ) (mm)</td>
<td>1300</td>
</tr>
<tr>
<td>wall temperature,</td>
<td>( T_R ) (mm)</td>
<td>1400</td>
</tr>
</tbody>
</table>

Injector Flow

<table>
<thead>
<tr>
<th>temperature,</th>
<th>( T_I ) (°C)</th>
<th>25</th>
</tr>
</thead>
<tbody>
<tr>
<td>flow rate,</td>
<td>( Q_I ) (Nm(^3)/h)</td>
<td>1.3</td>
</tr>
<tr>
<td>velocity,</td>
<td>( V_I ) (m/s)</td>
<td>5.0</td>
</tr>
<tr>
<td>coal mass flow rate,</td>
<td>( m_P ) (kg/h)</td>
<td>0.7</td>
</tr>
<tr>
<td>volatiles</td>
<td>( VM ) (wt%)</td>
<td>64</td>
</tr>
<tr>
<td>NO flow rate,</td>
<td>( Q_{NO} ) (Nm(^3)/h)</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Preheat Gas Flow

<table>
<thead>
<tr>
<th>temperature,</th>
<th>( T_S ) (°C)</th>
<th>1455</th>
</tr>
</thead>
<tbody>
<tr>
<td>flow rate,</td>
<td>( Q_S ) (Nm(^3)/h)</td>
<td>17.8</td>
</tr>
<tr>
<td>velocity,</td>
<td>( V_S ) (m/s)</td>
<td>6.5</td>
</tr>
</tbody>
</table>

* for definition of symbols, refer to Figure 3.2

3.1.4 Description of coals and preparation

In the Reburn Zone Investigation, three bituminous coals were tested to study the effect of volatile matter content on NO\(_x\) reduction. The ultimate and proximate analyses of Hugo, Scotts Branch and Obed Mountain coals are shown in Table 3.2. The volatile matter contents ranged from 25% daf for Hugo coal to 42% daf for Obed Mountain coal.

Three samples of each coal were prepared with different particle size distributions. Normal grind samples with 75% by weight smaller than 75 \( \mu \)m fraction and fine grind samples with 93% by weight smaller than 75 \( \mu \)m fraction were prepared by grinding the air dried coal in a single pass mill. Micronized coal samples, with 100% less than 40 \( \mu \)m, were prepared by milling and classifying the coal and remilling the oversized
particles. An analysis of the particle size distribution for the three fractions of the coals is shown in Table 3.2. A Rosin-Rammler distribution was obtained for the normal and fine grind samples and a typical micronized distribution was obtained for the micronized coal. Oxidation was prevented by storing the samples in sealed containers.

<table>
<thead>
<tr>
<th>Analysis</th>
<th>Obed Mountain</th>
<th>Scotts Branch</th>
<th>Hugo</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>n</td>
<td>f</td>
<td>m</td>
</tr>
<tr>
<td>Ultimate (daf)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>72.7</td>
<td>72.8</td>
<td>72.5</td>
</tr>
<tr>
<td>H</td>
<td>4.6</td>
<td>4.6</td>
<td>4.7</td>
</tr>
<tr>
<td>N</td>
<td>1.8</td>
<td>1.8</td>
<td>1.8</td>
</tr>
<tr>
<td>S</td>
<td>0.6</td>
<td>0.5</td>
<td>0.6</td>
</tr>
<tr>
<td>O</td>
<td>20.3</td>
<td>20.3</td>
<td>20.4</td>
</tr>
<tr>
<td>Proximate (dry)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ash</td>
<td>14.2</td>
<td>14.2</td>
<td>14.2</td>
</tr>
<tr>
<td>VM</td>
<td>36.3</td>
<td>36.3</td>
<td>36.2</td>
</tr>
<tr>
<td>Size Range</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>% &lt;75 (\mu m)</td>
<td>72</td>
<td>92</td>
<td>100</td>
</tr>
<tr>
<td>&gt;150 (\mu m)</td>
<td>2</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>100-150 (\mu m)</td>
<td>11</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>75-100 (\mu m)</td>
<td>15</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>40 - 75 (\mu m)</td>
<td>28</td>
<td>33</td>
<td>8</td>
</tr>
<tr>
<td>20 - 40 (\mu m)</td>
<td>22</td>
<td>33</td>
<td>56</td>
</tr>
<tr>
<td>&lt;20 (\mu m)</td>
<td>22</td>
<td>26</td>
<td>36</td>
</tr>
</tbody>
</table>

\(n = 75\% < 75 \mu m; f = 93\% < 75 \mu m; m = \text{micronized}\)

3.1.5 Data Analysis

The influence of seven process parameters on NOx and TVFN reduction in the reburn zone were studied. Each parameter was studied at three conditions over the range which
would be attainable in industrial processes. The range of parameters studied are listed in Table 3.3 and the test conditions are listed in Table A1 (Appendix A).

<table>
<thead>
<tr>
<th>Process parameter</th>
<th>Test conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>primary stoichiometry</td>
<td>( \lambda_p ): 1.0, 1.075, 1.15</td>
</tr>
<tr>
<td>primary NOx concentration</td>
<td>( \text{NOx}_p ): 200, 400, 600 ppm</td>
</tr>
<tr>
<td>coal type</td>
<td></td>
</tr>
<tr>
<td>particle size distribution</td>
<td>( \text{PSD} ): normal, fine, micronized</td>
</tr>
<tr>
<td>reburn fuel fraction</td>
<td>( f_{RB} ): 10, 20, 30%</td>
</tr>
<tr>
<td>reburn temperature</td>
<td>( T_{RB} ): 1100, 1250, 1400°C</td>
</tr>
<tr>
<td>reburn residence time</td>
<td>( t_{RB} ): 0.1, 0.3, 1.0 s</td>
</tr>
</tbody>
</table>

A Box-Behnkin experimental design [72] was used to minimize the number of tests necessary to determine the effect of the seven process parameters. This technique involved designating a subset of the test conditions in a full three-level factorial design that provided sufficient data to determine the effect of changing individual or any combination of two parameters. The influence of process parameters on NOx and TVFN reduction was modelled using a response surface equation. The response surface equation provides a graphical representation of the influence of process parameters on NOx reduction and is used throughout the results to visualize process effects when complex trends exist. The response surface equation had the form in tensor notation:

\[
y = B_0 + B_i x_i + B_{ij} x_i x_j
\]

(3.2)

where \( i \geq j \) and \( y \) was the relative NOx or TVFN emissions expressed as:

\[
y = 100 \frac{(\text{NOx}_\text{in} - \text{NOx}_\text{out})}{\text{NOx}_\text{in}}
\]

(3.3)

\[
y = 100 \frac{(\text{TVFN}_\text{in} - \text{TVFN}_\text{out})}{\text{NOx}_\text{in}}
\]
The variable, $x$, was a normalized value of each process parameter described by the expression:

$$x = \frac{(2X - X_H - X_L)}{(X_H - X_L)}$$  \hspace{2cm} (3.4)

where $X$, $X_H$ and $X_L$ referred to the actual, maximum and minimum values of each process variable. Using this procedure, $x$ varied from -1 to 1 over the full domain of the process variable. Least squares regression was used to estimate the coefficients $B_i$ and $B_{ij}$ in this model and obtain an empirical equation for the effect of process variables on NOx, HCN and NH3 concentrations.

3.1.6 Additional Reburn Zone Tests

The second Investigation, the Furnace Investigation, was undertaken to determine the effect of mixing on NOx reduction at the optimum conditions identified in the Reburn Zone Investigation. However, it was not possible to attain these optimum conditions in the Furnace Investigation. To aid in comparing the results of the two Investigations, additional reburn zone experiments were conducted and are reported in Chapter 6.

3.2 Mixing Characterization Tests

A 2.5 MW, semi-industrial scale furnace, Furnace No. 1, was used to study the effect of process parameters on NOx emissions under high and low mixing conditions. These tests were designed to simulate the conditions which are likely to be found in a boiler. Significant modifications were made to the furnace in order to create three relatively independent zones representing primary, reburn and tertiary combustion. These modifications are discussed in Section 3.2.1.

Both a high and a low level of mixing between reburn fuel and primary combustion products were required to determine the influence of mixing on NOx reduction by reburning. Before the experiments were conducted, the extent that reburn fuel mixing could be adjusted was not known, and therefore, a series of flow studies was conducted to determine the possibility of varying mixing in the reburn zone. Numerical
computations of the precombustor and reburn zone, described in Section 3.2.2, were conducted to predict the flow properties of the primary combustion products. From the velocity and turbulence fields, optimum injection locations for the low and high mixing levels were proposed. A 1/12th scale water model, Section 3.3.3, was used to verify the predicted flow field in the reburn zone and to visualize the mixing of reburn fuel and primary combustion products. A CO₂ tracer technique was used in Furnace No. 1 to quantify the mixing level of reburn fuel and primary combustion products for the low and high mixing cases, Section 3.2.4.

3.2.1 Experimental Arrangement

Modifications performed on Furnace No. 1 were made to provide adequate combustion zone separation and appropriate primary, reburn and tertiary zone residence times, Figure 3.7. The primary combustion zone was confined to a precombustor installed on the front of the furnace. The precombustor was a refractory lined, water cooled tunnel of 0.65 m inner diameter and 2.4 m length, Figure 3.8. A 2 MW swirled air coal burner was constructed and installed on the front of the precombustor.

Six, 48 mm outer diameter by 1.5 m long, water cooled pipes could be inserted from the front of the precombustor to adjust heat extraction from the flame. The cooling pipes had a measured heat extraction of 300 kW/m² and were capable of removing up to 300 kW from the flame. The precombustor wall heat extraction rate was estimated to be 20 kW/m² providing an additional 100 kW heat extraction.

The precombustor was designed to maintain an exit gas velocity in excess of 9 m/s for all test conditions. This gas velocity was recommended by Smart and Weber [73] to prevent excessive slag deposition on the precombustor walls. At the design conditions, the primary zone residence time was estimated to be 0.3 s. Based on coal characterization studies [16], this residence time was estimated to be sufficient to burn out 95% of the primary fuel prior to injecting the reburn fuel. Maximum burnout of the primary fuel was necessary to separate combustion zones and prevent excess O₂ from entering the reburn zone.
The front section of Furnace No. 1 up to 1.8 m AD was the reburn zone. The reburn zone was separated from the tertiary combustion zone using two refractory choke walls centered at 0.9 and 1.65 m AD. The two chokes had circular openings of 1 m and 1.35 m diameter which approximately matched the predicted flow contour of an expanding jet in a confined chamber [74]. These chokes were intended to split the large external recirculation zone at the front of the furnace and prevent excessive recirculation of tertiary products into the reburn zone. There was sufficient space between the choke walls to allow visual and probe access.
Reburn fuel was injected through four moveable water-cooled guns. The guns were situated around the circumference of the precombustor and were angled inwards converging on the centerline of the furnace at 1.5 m AD. By moving the guns, reburn fuel could be injected up to 1 m into the reburn zone.

The air was injected on the furnace centerline and directed radially outwards at an angle of 75° to the forward flow using the pipes shown in Figure 3.9. The tertiary air pipes were designated as positions 1 through 4 and were located at 1.9, 2.3, 3.5 and 4.5 m axial distance, respectively, Figure 3.7. Tertiary air could be injected through one or any combination of pipes. For most tests in the Furnace Investigation, two combinations of tertiary air injection were chosen. The first combination was to inject all of the tertiary air at pipe positions 1 and 2, both of which were just downstream of the second reburn zone choke. This resulted in a reburn zone residence time of approximately 0.3 s. The second combination was to inject sufficient tertiary air at pipe position 2 to raise the stoichiometry in the flow to unity and to inject the remaining tertiary air at pipe position 4. The injection of the initial tertiary air at pipe position 2 resulted in a reburn zone residence time of approximately 0.5 s. The location of pipe 4, 4.5 m axial distance, was chosen as half the distance between the reburn zone exit and the chimney.

Heat was extracted from the flame through the four water-cooled tertiary air pipes and six cooling loops, located on the outer circumference of the furnace. These cooling loops could extract up to 1000 kW from the flame. The first cooling loop was located downstream of the second refractory choke at 1.9 m AD. Thus, the estimated heat loss in the reburn zone was low (200-400 kW) and was due primarily to heat loss through water-cooled measurement slots doors which were located on the furnace wall.

Figure 3.9 Design of the tertiary air injector
3.2.2 Numerical Computations

Numerical computations were conducted to predict the flow in the precombustor and reburn zone. The two zones were computed separately with the outlet conditions from the precombustor simulation used as the inlet conditions for the reburn zone. The inlet conditions for the precombustor simulation are shown in Table 3.4. The predicted outlet conditions from the precombustor were used as the inlet conditions in a 1500°C, isothermal simulation of the reburn zone and are also shown in Table 3.4.

<table>
<thead>
<tr>
<th>Table 3.4 Inlet conditions for the Furnace Investigation flow simulation</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Primary Air</strong></td>
</tr>
<tr>
<td>Axial velocity (m/s)</td>
</tr>
<tr>
<td>Temperature (°C)</td>
</tr>
<tr>
<td>Turb. kinetic energy (m²/s³)</td>
</tr>
<tr>
<td>Turb. dissipation (m²/s³)</td>
</tr>
<tr>
<td><strong>Secondary air</strong></td>
</tr>
<tr>
<td>Axial velocity (m/s)</td>
</tr>
<tr>
<td>Tangential velocity (m/s)</td>
</tr>
<tr>
<td>Swirl number</td>
</tr>
<tr>
<td>Temperature (°C)</td>
</tr>
<tr>
<td>Turb. kinetic energy (m²/s³)</td>
</tr>
<tr>
<td>Turb. dissipation (m²/s³)</td>
</tr>
</tbody>
</table>

*r refers to precombustor radius

The reburn zone simulation was conducted isothermally and without reburn fuel injection. The isothermal simulation was conducted to reduce computational time as little heat would be extracted or generated in this region during experiments. The reburn fuel was neglected since it would introduce three-dimensional effects in the flow field which could not be accurately modelled. As well, the reburn fuel would flow across the primary flow introducing strong numerical diffusion in the computations.
An initial simulation was made without chokes in the reburn zone to determine the location of the 100% streamline. After the solution had converged, two refractory choke walls were inserted with their orifices located on the 100% streamline. This modification was intended to reduce the strength of the external recirculation zone (ERZ) and prevent tertiary air recirculation. The solution was again converged to obtain a prediction of the flow in the reburn zone.

3.2.3 Water Model Flow Visualization

The water model was a 1/12th representation of Furnace No. 1 modified for the Furnace Investigation. The model consisted of a plexiglass cylinder representing the precombustor attached to a perspex box representing Furnace No. 1, Figure 3.10. Primary combustion products were represented by the flow through the cylinder. The swirling flow from the burner was depicted using an axial vane swirl generator located on an annular injector at the front of the cylinder. The vanes were angled at 30° to produce a flow with swirl number of 0.5.

Reburn fuel injection was represented by eight secondary water nozzles situated on the perimeter of the precombustor cylinder exit. Secondary water could be injected through four of the nozzles angled inward at 12° or through the other four nozzles angled inward at 45°. Tertiary water was injected on the centerline through a pipe inserted from the exit. The pipes could be moved in or out of the model to change the injection locations of the secondary and tertiary flows.

![Figure 3.10 Schematic of the water model flow simulator](image-url)
Tests were conducted with primary to secondary flow rate ratios similar to the range of flow rate ratios envisaged for the Furnace Investigation. The Reynolds number was maintained in order to have similar flow properties and the momentum flux ratio between the reburn jets and the main forward flow was also maintained in order to have representative reburn jet penetration. To satisfy all criteria, the geometric scale similarity had to be relaxed for the secondary water nozzles. The nozzles were 1/6 of the full scale. The relaxing of geometric similarity, when all similarity criteria cannot be satisfied, had been proposed by Thring and Newby [74].

Initial visualization tests were conducted using pH sensitive dyes to determine mixture fraction by colour change. A phenolphthalein dye solution with pH 10 was injected through the secondary nozzles. At pH 10, the solution was red and became colourless when the pH decreased to less than 8.3. This represented a dilution factor of 50 times. However, the tests conducted with a phenolphthalein dye solution were unsuccessful since the high pH of the transport water (8.0) prevented colour change when the dilution was 50 times.

Tests were also conducted with ink dyes injected through the reburn pipes. Mixing was evaluated on the basis that the ink should fill the forward flow between the refractory chokes. Stratified regions of colourless and ink filled water represent a low mixing configuration. The momentum ratio of primary to secondary flow and injection position were varied to obtain the low and high mixing cases.

3.2.4 CO₂ Tracer Studies

Mixing of reburn fuel with the primary combustion products was quantified using a technique suggested by Hawthorne et al [75]. A tracer was injected into the flow at known total concentration and the local concentration measured at several points to determine local dispersion. A mixing factor was defined which was the fraction of the total mass flow with tracer concentration in a specified range according to:

\[
N_f = \frac{\Sigma \rho_i \Omega_i u_i \mid \text{mixed}}{\Sigma \rho_i \Omega_i u_i} \tag{3.5}
\]

where the flow was divided into discrete cells with area, \( \Omega_i \), density, \( \rho_i \), and velocity, \( u_i \). Several suitable tracers existed to measure mixing fraction. In this work, CO₂ and He
were considered. The CO₂ tracer was preferred over He since CO₂ had a density more representative of coal volatile matter. This permitted the existing reburn guns to be used for the tracer work.

The CO₂ was injected through the reburn guns at a momentum flux similar to the planned reburn coal/transport air mixture. The CO₂ mixed with primary air, preheated to 500°C, representing primary combustion products. To determine the dispersion, CO₂ samples were collected on a horizontal and vertical grid at three axial locations: upstream and downstream of the first choke (0.6 and 1.05 m axial distance, respectively) and upstream of the second choke (1.4 m axial distance). For each axial location, the CO₂ concentration was measured at 25 points on a grid of 5 horizontal positions and 5 elevations, between the furnace centerline and the choke opening. The average CO₂ concentration was 1% and a cell was considered to be sufficiently mixed if the CO₂ concentration was in the range 0.5-2.0%. The radial velocity distribution was measured using a Prandtl tube to calculate the mass flux through each cell. The mixture fraction was then determined using Equation 3.5.

3.3 Furnace Investigation

The influence of mixing between reburn fuel and primary combustion products was investigated using the technique developed in the Mixing Characterization Tests. NOx emissions were measured with the mixing adjusted to a high or a low level by moving the reburn guns.

Seven coal and one natural gas reburning flames were studied with different primary zone stoichiometries and reburn fuel fractions. The mixing of reburn fuel with primary combustion products, and tertiary air injection arrangement were varied for each flame.

3.3.1 Experimental Tests

The process conditions for the seven coal reburning flames, designated as flames F1-F7, and the natural gas reburning flame F8 are listed in Table 3.5. Fuel staging was studied for reburn zone stoichiometry, $\lambda_{RB}$, of approximately 0.82 to 0.92. The $\lambda_{RB}$ was set by fixing the primary zone stoichiometry, $\lambda_p$, in the range 0.95 to 1.1 and adjusting the reburn fuel fraction, $f_{RB}$, from 10 to 30%.
Tertiary air was injected through two configurations to determine the influence of staged tertiary air addition on NOx reduction. In the normal air addition pattern (1,2), the tertiary air was injected immediately downstream of the reburn zone at pipe positions 1 and 2, Figure 3.7. In the delayed air staging configuration (2,4), sufficient tertiary air was added at pipe position 2 to raise the stoichiometry to 1.0 and the remaining tertiary air was injected half-way between the reburn zone and the chimney at pipe position 4.

<table>
<thead>
<tr>
<th>Flame No.</th>
<th>Primary stoichiometry</th>
<th>Fuel type</th>
<th>Reburn Zone fraction (%)</th>
<th>Reburn Zone stoichiometry</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.95</td>
<td>coal</td>
<td>10</td>
<td>0.86</td>
</tr>
<tr>
<td>2</td>
<td>0.95</td>
<td>coal</td>
<td>20</td>
<td>0.78</td>
</tr>
<tr>
<td>3</td>
<td>1.04</td>
<td>coal</td>
<td>10</td>
<td>0.96</td>
</tr>
<tr>
<td>4</td>
<td>1.04</td>
<td>coal</td>
<td>20</td>
<td>0.84</td>
</tr>
<tr>
<td>5</td>
<td>1.10</td>
<td>coal</td>
<td>20</td>
<td>0.89</td>
</tr>
<tr>
<td>6</td>
<td>1.10</td>
<td>coal</td>
<td>30</td>
<td>0.78</td>
</tr>
<tr>
<td>7</td>
<td>1.00</td>
<td>coal</td>
<td>20</td>
<td>0.82</td>
</tr>
<tr>
<td>8</td>
<td>1.00</td>
<td>gas</td>
<td>20</td>
<td>0.80</td>
</tr>
</tbody>
</table>

3.3.2 Test Fuels

Experiments were conducted using Saar coal from the Göttelborn pit. The proximate and ultimate analyses of the test coal, as fired, are given in Table 3.6. The coal was received as 20 mm pieces and dried, crushed and pulverized in the IFRF coal preparation facility. The particle size was milled to 75±4% less than 200 mesh throughout the tests. A typical particle size distribution, determined by wet sieve analysis, is given in Table 3.6. The pulverized coal was stored in a fine coal bunker and charged into two KTRON weigh bunkers as required. The primary coal was transported to the burner with 400 kg/h air heated to 60-80°C. The reburn coal was transported in 50-70 kg/h air and divided into four streams using a flow splitter at the furnace.

Natural gas was also used as a reburn fuel and the composition is shown in Table 3.6.
Table 3.6 Analysis of Test Fuels

<table>
<thead>
<tr>
<th>Analyses</th>
<th>Saar coal</th>
<th>Natural gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>Composition (wt%daf)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>80.5</td>
<td>CH$_4$ 85</td>
</tr>
<tr>
<td>H</td>
<td>5.1</td>
<td>C$_2$H$_6$ 6</td>
</tr>
<tr>
<td>N</td>
<td>1.7</td>
<td>C$_3$H$_8$ 1</td>
</tr>
<tr>
<td>O</td>
<td>11.8</td>
<td>CO$_2$ 0.1</td>
</tr>
<tr>
<td>S</td>
<td>0.9</td>
<td>N$_2$ 7</td>
</tr>
<tr>
<td>Ash (wt% dry)</td>
<td>6.8</td>
<td></td>
</tr>
<tr>
<td>VM (wt% dry)</td>
<td>38</td>
<td></td>
</tr>
<tr>
<td>LCV (MJ/kg)</td>
<td>30</td>
<td>44</td>
</tr>
<tr>
<td>Particle Size (wt%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>&gt;150 μm</td>
<td>3</td>
<td></td>
</tr>
<tr>
<td>100 - 150 μm</td>
<td>11</td>
<td></td>
</tr>
<tr>
<td>75 - 100 μm</td>
<td>11</td>
<td></td>
</tr>
<tr>
<td>40 - 75 μm</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>20 - 40 μm</td>
<td>26</td>
<td></td>
</tr>
<tr>
<td>&lt;20 μm</td>
<td>24</td>
<td></td>
</tr>
</tbody>
</table>

3.3.3 Measurement Probes

Gas and solid samples were taken from within the flame and in the chimney using a steam heated Alcan probe. Particles were collected in a sintered bronze filter at the back end of the probe and then the gas sample was drawn in a steam heated line to the gas analysis train. Water was condensed out of sample and the dry gas was analysed for NOx (Thermo Electron chemiluminescent analyzer), O$_2$ (Servomex paramagnetic analyzer), CO and CO$_2$ (Unor infrared analyzers).

HCN and NH$_3$ were sampled in the flame using an HCN/NH$_3$ probe. The sampling procedure was the same as discussed in Section 3.1.2.
3.4 Burner Design and Testing

The fuel staged burner was designed based on requirements for a suitable pulverized coal burner and considerations of the results of the two Investigations. A design was proposed which would be simple to operate and could use both coal and gas as reburn fuel. Important process variables were identified and considered in the design and test program.

The flow pattern was predicted (using the computational model) to estimate the interaction between the reburn fuel and primary combustion products. The simulation was conducted with refractory chokes and tertiary air injection as specified in the Furnace Investigation. In the burner modelling, the full furnace could be modelled using a two-dimensional formulation since all flows were injected axisymmetrically. The predictions were interpreted in light of the two Investigations to suggest whether the design could result in lower NOx emissions than already obtained in the Furnace Investigation.

Burner tests were conducted in Furnace No. 1 using the same refractory choke and tertiary air arrangement as in the Furnace Investigation. A single burner was installed on the front face of Furnace No. 1. The burner is described in detail in Chapter 7. Primary coal was injected through an annulus in the throat of the quarter and reburn fuel was injected with a single reburn coal injector, through the middle of the coal gun. The tertiary air could be injected through any of the four tertiary air pipes.

The NOx emissions were measured with the burner operated at different $\lambda_F$, $f_{RB}$ and $\lambda_{RB}$. These data were compared with the NOx emissions measured in the Furnace Investigation. Reburn fuel injection location and secondary air swirl level were varied to determine the influence of reburn fuel penetration through the IRZ on NOx emissions. Additional tests were conducted with natural gas reburning fuel. The natural gas was injected either radially or axially through a water-cooled probe. The natural gas probe could be inserted up to 1 m into the furnace to obtain complete separation of the primary and reburn zones.

3.5 Computational Modelling of Swirling Flames

A computational model based on the FLUENT code [76] was used to predict swirling
pulverized coal flames by Weber and Visser [77,78]. This computational model was applied in each phase of the research to predict flow patterns and mixing behaviour. A description of the model is presented in this section.

3.5.1 Gas Phase Aerodynamics

The aerodynamics of swirling flows is modelled by mass and momentum balances. The balance equations for mass and flow momentum are written here in cartesian form and tensor notation:

\[
\frac{\partial (\rho U_i)}{\partial x_i} = 0
\]

\[
\frac{\partial (\rho U_i U_j)}{\partial x_i} = - \frac{\partial P_i}{\partial x_j} + \mu_1 \frac{\partial}{\partial x_i} \left[ \frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right] - \frac{\partial (\rho \langle \mathbf{u}_i \mathbf{u}_j \rangle)}{\partial x_i}
\]

where \( U_i \) and \( u_i \) are the mean and fluctuating components of the velocity vector in the direction \( x_i \), and \( P, \rho \) and \( \mu_1 \) are the fluid pressure, density and dynamic viscosity, respectively. The Reynolds stresses \( \langle \mathbf{u}_i \mathbf{u}_j \rangle \) are calculated using a second order closure, the Algebraic Stress Model (ASM) which has been shown to be valid for swirling combusting flows [77].

Turbulence is modelled by a turbulent energy, \( k \), and its dissipation rate, \( \varepsilon \), which are calculated from the transport equations:

\[
\frac{\partial (\rho U_i k)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[ \frac{\mu_1}{Pr_k} \frac{\partial k}{\partial x_i} \right] + \rho (P - \varepsilon)
\]

\[
\frac{\partial (\rho U_i \varepsilon)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[ \frac{\mu_1}{Pr_\varepsilon} \frac{\partial \varepsilon}{\partial x_i} \right] + \frac{\rho C_{\alpha_1} P \varepsilon}{k} - \frac{C_{\alpha_2} \varepsilon^2}{k}
\]
where the effective viscosity, \( \mu_t \) is related to \( k \) and \( \varepsilon \) according to:

\[
\mu_t = \mu_1 + C_\mu \rho k^2/\varepsilon
\]  

(3.10)

and \( c_{e1}, c_{e2} \) and \( c_\mu \) are constants with the values 1.44, 2.09 and 0.09, respectively.

### 3.5.2 Conservation Equations for Species and Enthalpy

It is assumed that the gas phase consists of volatiles, \( O_2 \), \( CO \), combustion products (\( CO_2 + H_2O \)) and \( N_2 \). For each of the gases species, except \( N_2 \), a mass conservation equation is solved for the mass fraction, \( m_x \):

\[
\frac{\partial (\rho U_i m_x)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[ \frac{\mu_t}{Sc_x} \frac{\partial m_x}{\partial x_i} \right] + S_x
\]  

(3.11)

where \( Sc_x \) is the Schmidt number of species \( x \) and was equal to 0.7 for all species. \( S_x \) represents the source or sink of the species \( x \).

The enthalpy conservation equation is as follows:

\[
\frac{\partial (\rho U_i h)}{\partial x_i} = \frac{\partial}{\partial x_i} \left[ \frac{\mu_t}{Pr_h} \frac{\partial h}{\partial x_i} \right] + S_{comb} + S_{rad}
\]  

(3.12)

The source terms due to combustion \( (S_{comb}) \) and radiation \( (S_{rad}) \) are calculated by additional submodels. \( S_{rad} \) is calculated using the six flux method. Since only axisymmetric cases are computed, just four radiative fluxes are sufficient to calculate the source terms in the enthalpy equation. The gas is assumed to be gray, i.e. the absorption and emission coefficients of the gas are assumed to be equal. The value for the absorption coefficient of the coal flame is estimated to be 0.5. This estimation is
based on measurements and calculations for pulverized coal flames in the IFRF semi-industrial scale Furnace No. 1 [79]. The furnace walls are insulated and heat is extracted through cooling loops installed on the inside of the furnace. This is modelled in the computations by assigning an emissivity to the walls of zero and to the cooling loops of 0.6.

3.5.3 Gas Phase Combustion

The rate of gas phase chemical reactions is controlled by the rate of mixing between fuel and oxidant inside a fluid eddy and the kinetic reaction rate of the reactants. In diffusion flames, the chemical kinetic combustion rates are usually an order of magnitude greater than the turbulent mixing rate. The latter is characterized by the ratio between turbulent energy and its dissipation rate \((k/\varepsilon)\). Thus, the reaction rate within a fluid eddy is controlled by the mixing intensity as adapted from Magnusson and Hjertager [80]:

\[
R_{\text{ox}} = \Phi_c \rho A \varepsilon/k \left| [m_f, m_{ox}/S_t] \right|
\]  

(3.13)

where \(R_{\text{ox}}\) is the consumption of oxidant \((O_2)\) per second. The quantities \(m_f\) and \(m_{ox}\) are the mass fractions of fuel and oxidant, respectively, and \(\Phi_c\) is the mass fraction of oxidant required to combust 1 kg of fuel. The operator, \(\left| [\cdot] \right|\), selects the minimum value of the two terms between the brackets. The constant A relates the mixing time to the local turbulent properties. It is assigned a value 0.6 based on computational modelling of pulverized coal flames [81] and CO/air flames [82].

3.4.4 Particle Trajectories

The particle tracking procedure was originally developed for modelling of the particle phase in cyclone separators [83]. The equations of motion for the coal particles are expressed in a Lagrangian frame of reference and the coupling between solid and gas phase is provided by appropriate source terms in the gas phase equations. When only drag force on a particle is taken into account, the equations of motion of particles in a gaseous environment can be expressed as:
\[
\begin{align*}
\frac{dU_p}{dt} &= -\alpha(U_p - U_d) \\
\frac{dV_p}{dt} &= -\alpha(V_p - V_d) + \frac{W_p^2}{r_p} \\
\frac{dW_p}{dt} &= -\alpha(W_p - W_d) - \frac{W_p V_p}{r_p} \\
\alpha &= \frac{18}{p_d d_p^2} \left( C_d \frac{Re}{24} \right) \left( \frac{Re}{\rho_d (U_p - U_d) / \mu} \right)
\end{align*}
\] (3.14) (3.15) (3.16) (3.17)

\( U, V \) and \( W \) are the velocity components in \( x, r \) and \( \theta \)-directions in the cylindrical system of coordinates, while the subscripts \( g \) and \( p \) denote the gas phase and particle phases, respectively. The drag coefficient \( C_d \) is expressed as a function of the relative Reynolds number with the general form:

\[ C_d = a_1 + a_2 \text{Re} + a_3 \text{Re}^2 \] (3.18)

The \( a \) coefficients are given for several ranges of Reynolds number. Stokes regime is obeyed for \( \text{Re} < 0.1 \) and for \( 0.1 < \text{Re} < 1.0 \), the \( a \) coefficients take values of 3.69, 22.73 and .0903 respectively [84].

There are two mechanism controlling coal particle dispersion: gas phase turbulence and the drag force associated with the centrifugal particle motion. In order to account for the effects of turbulence on the trajectories of individual particles, the instantaneous gas velocity is decomposed into time mean and fluctuating components. It is assumed that the fluctuating component prevails for a time period, \( \tau_e \), equal to the mean life time of a fluid eddy. This time can be estimated in terms of the local kinetic energy of turbulence and its dissipation rate [85]:

\[ \tau = .16 \frac{k}{\epsilon} \] (3.19)

The values of the fluctuating velocities are assumed to possess a Gaussian distribution, and are related to the root mean fluctuations by:

\[ u' = \Theta \left( \langle u^2 \rangle \right) = \Theta \left( 2k/3 \right) \] (3.20)

where \( \Theta \) is a normal distributed random number variable and \( u' \) represents the fluctuating component of velocity in each direction.
The coal particle size distribution is represented by fifteen size classes. The mass fraction in each size class may be determined from a sieve analysis of pulverized coal and then represented using the Rosin Rammler equation [86]

\[ R = \exp(-bx^n) \]  

(3.21)

where \( R \) is the mass fraction above particle size, \( x \) (\( \mu \)m), \( n \) is the spread parameter (1.15 in the tests reported here) and \( b \) is a constant related to the mass mean particle size. In the computations, three particles from each size fraction were injected at every 20 iterations of the gas phase equations.

3.5.5 Particle Heating

Coal particles are heated by convection, radiation and combustion. The progressive change in temperature of a single coal particle can be estimated from an energy balance:

\[ m_p C_p dT_p/dt = Q_c + Q_r + Q_h \]  

(3.22)

where \( m_p \) and \( C_p \) are the mass and specific heat of the particle, respectively. \( Q_c \) and \( Q_r \) represent the rates of heat transfer to the particle by convection and radiation, respectively, and \( Q_h \) is the heat generation by combustion.

3.5.6 Devolatilization

The devolatilization model applied in the computational model for swirling pulverized coal flames was based on results from devolatilization experiments of high volatile bituminous (hvb) coals [16,86]. These experiments were carried out in an isothermal plug flow reactor (IPFR) under conditions of rapid particle heating (2x10^5 \( K/s \)). The devolatilization time increased from 10 to 25 ms as the gas temperature was decreased from 1400 to 1000\(^\circ\)C. The experiments indicated that the rate of devolatilization was equal to the particle heating rate predicted using computational modelling. It was found that the high temperature volatile yield could be estimated from the following scheme:
Yield = \frac{0}{\text{VM}_{\text{max}}} \times \frac{T_p}{1000} \quad \text{at} \quad \begin{cases} T_p < 500^\circ C \\ 500^\circ C < T_p < 1000^\circ C \\ 1000^\circ C < T_p \end{cases} \quad (3.23)

where \text{VM}_{\text{max}} was the maximum volatile yield. The only independent parameter in the model was the instantaneous particle temperature, \( T_p \), which was calculated using Equation 3.22.

3.5.7 Char Combustion

The combustion of char occurs predominantly after volatile matter evolution. When the convective flow of \( O_2 \) to the particle surface is greater than the counter flow of volatiles away from the particle, the char burns heterogeneously.

A simple analysis, which assumes that the char reaction may be separated according to bulk diffusion of oxygen to the particle external surface and an Arrhenius type reaction rate incorporating the effects of chemical reaction and pore diffusion, is used to interpret the burnout data. The rate of particle burnout, \( q \), may be expressed by:

\[
q = \frac{-1}{Y} \frac{dY}{dt} \frac{d_{pl} \rho_{pl}}{6} \quad (\text{kg/m}^2/\text{s}) \quad (3.24)
\]

\[
= k_c (P_g - q/k_d)^{1/2} \quad (3.25)
\]

where \( Y \) is the fraction of carbon remaining; \( d_{pl} \) and \( \rho_{pl} \) are the initial particle diameter and the apparent density, respectively, and \( P_g \) is the partial pressure of \( O_2 \). The chemical reaction rate, \( k_c \), is given by:

\[
k_c = A_c \exp \left( -\frac{E_c}{RT_p} \right) \quad (\text{kg/m}^2/\text{s}/\text{Pa}^{1/2}) \quad (3.26)
\]

and \( k_d \) is the bulk diffusion coefficient given by:

\[
k_d = 4.98 \times 10^{-12} \frac{T_m^{7/5}}{d_p} \quad (\text{kg/m}^2/\text{s}/\text{Pa}) \quad (3.27)
\]
In the above expressions, $A_c$ and $E_c$ are the pre-exponential factor and activation energy, respectively, and $T_m$ is the mean temperature between the gas and particle temperature, $T_p$.

The chemical reaction rate is measured for each char of interest. Differences in physical structure, produced during the devolatilization process, result in variable $k_c$. The combustion reactivity of Saar char was determined in coal characterization experiments where it was found that $A_c$ was equal to 5.3 kg/m²/s/Pa and $E_c$ equalled 21000 cal/mol [16].

3.6 Concluding Remarks

An experimental method has been presented to study the influence of process parameters on NOx reduction by fuel staging and to apply these results in designing and testing a novel internally staged burner. The experiments were conducted in three parts: Reburn Zone Investigation, Furnace Investigation and Burner Tests.

The Reburn Zone experiments were conducted in a rapid mixing isothermal reactor (IPFR) in order to determine the maximum NOx reduction possible.

The NOx reduction from reburning under mixing limited conditions was studied in the Furnace Investigation. To quantify the mixing rate of reburn fuel and primary combustion products in the Furnace Investigation experiments, a detailed mixing study was performed on the furnace using computational modelling, water model flow simulations and CO₂ tracer studies.

The results from these two Investigations were implemented in the design of a novel internally fuel staged burner. The burner concept was studied using computational modelling and tested in semi-industrial experiments using a 2.5 MWt burner.
4. Reburn Zone Investigation

The influence of process parameters on NOx reduction in the reburn zone was evaluated in the Reburn Zone Investigation. The effect of:

- reburn coal type,
- coal particle size distribution, PSD
- reburn fuel fraction, \( f_{RB} \)
- primary stoichiometry, \( \lambda_p \)
- NOx concentration, \( NOx_p \)
- reburn zone temperature, \( T_{RB} \)
- residence time, \( \tau_{RB} \)

on NOx and TVFN reduction was evaluated in 59 experimental tests. These tests are designated RB1 to RB59 and the process conditions and experimental results are detailed in Appendix A. Each experimental test was conducted in two parts: the first part to measure NOx and the second part to measure the HCN and NH3 concentrations. During the HCN and NH3 sampling, the concentration of NOx in the product gases was measured and compared to the first NOx measurement to determine experimental repeatability, Section 4.2. Detailed measurements of N-species concentrations up to 0.5 s were made for the three most significant process conditions identified in the experimental tests.

Modelling studies were performed before and after the experiments to interpret the experimental results. Computational modelling was conducted to determine the flow patterns, temperature and mixing rate of reburn fuel and coal volatile matter in the reactor, Section 4.1. The response surface model, described in Section 3.1.5, was used to represent the effect of process variables on NOx, HCN and NH3 concentrations. The ability of the response surface model to estimate the concentrations of NOx, HCN and NH3 was investigated and validated, Section 4.2.

The reduction rate of NOx and HCN, respectively, were compared with predictions made using an extended Fenimore's model for NOx reduction in fuel rich pulverized coal flames (Section 4.3).
4.1 Reactor Flow Simulation

The flow and thermal characteristics of the reactor were modelled using the finite difference computational model described in Section 3.5 (p.77). The inlet conditions of the model were representative of the flow conditions used in tests RB1-RB59, Table 3.1. For the reactor flow simulation, the reactor velocity was set at 5 m/s and a reactor temperature of 1400°C was chosen. The reburn fuel fraction was set to 30% and the volatile matter content of the fuel was 65%. Thus, the total flow contained 2% volatile matter by mass. NO was injected through the feeder probe at 1% of the total mass to demonstrate the effect of jet dispersion.

The predicted streamline contours in the first 0.3 m of the reactor are shown in Figure 4.1. The preheated gas rapidly converged on the central coal jet and expanded after the venturi throat. No negative streamlines were predicted around the coal injector which indicated that a recirculation zone was not formed behind the feeder probe. Thus, the gas residence time could be estimated from the axial sampling position. The impingement of the preheated gas onto the reburn fuel jet caused rapid mixing and the transport $N_2$ was heated to a temperature greater than 1200°C 0.3 m downstream of the venturi throat, Figure 4.2. The heating rate was in the order of $10^4$°C/s.

![Figure 4.1 Predicted streamlines in the IPFR](image)
The mixing between CH₄ radicals in the reburn fuel and the primary NOx was demonstrated by comparing the profiles of NO, injected through the feeder probe, and reburn coal volatile matter, Figure 4.3. It was shown that the NO rapidly dispersed into the primary combustion products. Just downstream of the venturi at 0.1 m axial distance, the peak NO concentration was reduced to 5%, and by 0.3 m axial distance, the peak was reduced to 2%. The volatile matter began to evolve from the coal at 0.1 m axial distance when the NO was already distributed in the primary combustion products. By 0.3 m axial distance, volatile matter was predicted to have mixed to the outside of the forward flow with a peak concentration along the centerline of 4.5%. The 0.3 m axial distance was equivalent to approximately 0.06 s which demonstrated that the reburn fuel radicals and primary NOx were rapidly mixed.

Figure 4.2 Predicted temperature contours in the IPFR

Figure 4.3 Predicted volatile matter and NO concentration in the IPFR
4.2 Method Verification

4.2.1 Repeatability Tests

It was not known at the time of the measurements whether the HCN probe would be acceptable for sampling NOx in the reburn zone. Thus, NOx and HCN/NH₃ measurements were made in two separate parts of the same tests. It was important that the NOx concentration was approximately the same in both parts in order to calculate TVFN. To compare the NOx concentrations in the separate test parts, a gas sampling probe, located at the bottom of the reactor (reactor bottom probe shown in Figure 3.1, p.57) was used to monitor NOx concentration during HCN/NH₃ measurements. It was assumed that the HCN/NH₃ probe quenched the gas as it flowed past the water-cooled jacket thereby fixing the products at the same composition as measured at the tip. Therefore, the NOx concentration measured using the NOx probe and the reactor bottom probe would be the same, assuming all other process variables remained constant. This provided a test of the repeatability of the NOx measurement results in the experiments.

The two NOx measurements, made with the NOx and reactor bottom probes respectively, were compared for all 0.3 s reactor residence time tests. The agreement between the two sets of data was very good as is represented by the solid line in Figure 4.4. The maximum difference between the two measurements was 60 ppm except for one point. If this point is neglected, the average difference was 1 ppm and the

![Figure 4.4 Comparison of NOx measured by the NOx and reactor bottom probes](image-url)

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standard deviation was 40 ppm. The deviation was considered acceptable and it was concluded that the reactor bottom probe could be used to measure NOx for residence time greater than 0.3 s and that the measurement of NOx concentration was valid to ± 40 ppm.

4.2.2 Response Surface Model Verification

The NOx and TVFN concentrations were estimated using the response surface model, Equation 3.2. This involved calculating the equation coefficients $B_0$, $B_1$ and $B_2$ based on the measured results for NOx, HCN and NH$_3$ concentration. The coefficients for NOx and TVFN fractional reduction, based on Equation 3.3, are given in Table 4.1.

The model provided a reasonable estimate of the measured NOx concentration for each test, Figure 4.5. The standard deviation of the difference between measured and predicted NOx was 42 ppm. This value was approximately equal to the standard deviation for the repeatability test described in the previous section (40 ppm). Therefore, the response surface model could be used to graphically represent the effect of reburning process variables on NOx and TVFN concentrations.

![Figure 4.5 Estimation of NOx concentration using the response surface equation](image-url)
### Table 4.1 Response Surface Model Coefficients for NOx and TVFN Reduction

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*Calculated according to Equations 3.2-3.4 where 1 = coal type; 2 = PSD; 3 = \(f_{RB}\); 4 = \(\lambda_{RB}\); 5 = \(T_{RB}\); 6 = \(NOx_p\); 7 = \(\log (\tau_{RB})\).

### 4.3 Effect of Process Variables on NOx Reduction

#### 4.3.1 Coal Type

The effect of coal type on NOx and TVFN concentrations as functions of reburn fuel fraction, \(f_{RB}\), and primary stoichiometry, \(\lambda_p\), are illustrated in Figures 4.6 and 4.7,
respectively. One curve was generated for each coal at a $T_{RB}$ of 1250°C, NOx$_P$ of 400 ppm and $\tau_{RB}$ of 0.3 s.

The trend in NOx concentration was similar for all three coals, Figure 4.6. A maximum NOx concentration of 450 ppm was estimated at $f_{RB}$ of 10% and $\lambda_P$ of 1.15 while a minimum of 150-200 ppm was estimated at $f_{RB}$ of 30% and $\lambda_P$ of 1.0. These response curves may be compared with data listed in Appendix A. In four tests conducted on the effect of coal type on NOx reduction at $f_{RB}$ of 30% and $\lambda_P$ of 1.0, the measured NOx concentration was 200 ppm for Obed Mountain coal (tests RB20, RB21) and 240 and 200 ppm for Hugo coal (tests RB16, RB17).

![Obed Mountain](image1)
![Scotts Branch](image2)
![Hugo](image3)

**Figure 4.6** Effect of reburn fuel fraction, $f_{RB}$, and primary stoichiometry, $\lambda_P$, on NOx concentration for $T_{RB} = 1250^\circ$C, $\tau_{RB} = 0.3$ s and NOx$_P = 400$ ppm.
The trend in TVFN concentration was similar for all three coals, Figure 4.7. Saddle shaped curves were predicted with peak TVFN concentrations at \( f_{RB} \) of 10\%, \( \lambda_p \) of 1.15 and at \( f_{RB} \) of 30\%, \( \lambda_p \) of 1.0. The TVFN concentrations for Scotts Branch and Hugo coals were similar at all conditions but Obed Mountain coal had 50-200 ppm higher TVFN. In the four tests conducted on the effect of coal type on TVFN at \( f_{RB} \) of 30\% and \( \lambda_p \) of 1.0, the measured TVFN concentration was 552 ppm for Obed Mountain coal (tests RB20, RB21) and 442 and 357 ppm for Hugo coal (tests RB16, RB17). Since the NOx concentration was similar for each coal and TVFN was the sum of NOx, HCN and NH3, the differences between Obed Mountain and the other coals was in the concentration of HCN and NH3.

![Graphs showing TVFN concentration for Obed Mountain, Scotts Branch, and Hugo coals.](image)

**Figure 4.7** Effect of reburn fuel fraction, \( f_{RB} \), and primary stoichiometry, \( \lambda_p \), on TVFN concentration for \( T_{RB} = 1250^\circ C \), \( \tau_{RB} = 0.3 \) s and NOx = 400 ppm
These results indicated that reburn coal type and volatile matter content did not have a significant effect on the NOx reduction in the reburn zone. However, the reburn coal type could have some effect on the HCN and NH3 concentrations leaving the reburn zone.

4.3.2 Particle Size Distribution (PSD)

The effect of reburn coal particle size was evaluated in tests comparing normal grind and micronized samples of each coal. The difference in NOx and TVFN concentrations between tests conducted with normal and micronized samples were within experimental error (±40 ppm). At $\lambda_{RB}$ of 0.82, the NOx concentration for Obed Mountain micronized and coarse PSD's was 200 ppm (tests RB20, RB21) and for Hugo micronized and coarse PSD's was 200 and 240 ppm (tests RB17, RB16), respectively. The coefficients used in the response surface equation for PSD ($B_{12}$-$B_{27}$: Equation 3.2) were all less than 8, Table 4.1. The maximum effect of changing the particle size on emissions as a percentage of inlet NOx is the sum of the absolute values of coefficients $B_{12}$, $B_{22}$ to $B_{27}$. This was only 20% and 19% for NOx and TVFN reduction, respectively. Thus, PSD had only a small influence on fractional NOx and TVFN reduction over all test conditions.

The PSD may have had some influence on the devolatilization rate of the reburn coal and the composition of the volatile matter. It has been shown that the devolatilization rate is closely related to particle heating rate which, in turn, is dependent on particle size [16]. Therefore, the heating and devolatilization rates were significantly faster for the micronized coal. Midkiff and Altenkirch [87] have reported that the nitrogen-species evolution from bituminous coal was faster and more complete for 28 μm diameter particles than 52 μm particles. It appeared that process parameters other than particle heating rate and the differences in volatile matter composition for a range of particle size distributions had a much greater influence on NOx reduction.

In further analysis of the reburn zone data, the effect of PSD is neglected.

4.3.3 Reburn Fuel Fraction and Primary Stoichiometry

The response surface curves for NOx reduction at $T_{RB}$ of 1250°C, NOxP of 400 ppm and $\tau_{RB}$ of 0.3 s, Figure 4.6, display the sensitivity of NOx concentration to $f_{RB}$ and $\lambda_F$. The
highest NOx concentration was estimated at \( f_{RB} \) of 10% and \( \lambda_p \) of 1.15, decreasing as \( f_{RB} \) increased to 30% and \( \lambda_p \) decreased to 1.0. The response surface curves reveal that the influence of \( f_{RB} \) and \( \lambda_p \) on NOx reduction could be expressed in terms of a single variable, the reburn zone stoichiometry, \( \lambda_{RB} \). The reburn zone stoichiometry is defined as the amount of O₂ supplied to the reburn zone, \( m_O \), in the form of CO₂, H₂O and O₂, divided by the O₂ that was required to combust the natural gas, \( m_{NG} \), according to:

\[
\lambda_{RB} = \frac{m_O}{m_{NG}} \quad (4.1)
\]

In Figure 4.6, \( \lambda_{RB} \) is constant along diagonal lines and equals 0.9 from the front left-hand corner (\( f_{RB}:10\%, \lambda_p:1.0 \)) to the back right-hand side (\( f_{RB}:25\%, \lambda_p:1.15 \)). Along this line, the NOx concentration was estimated to remain constant for each coal, demonstrating the interdependence of \( f_{RB} \) and \( \lambda_p \).

The lowest NOx concentration did not always coincide with the lowest TVFN concentration. The response surface for TVFN concentration, Figure 4.7, had a saddle shape, with peaks at \( \lambda_{RB} \) of 0.7 (\( f_{RB}:30\%, \lambda_p:1.0 \)) and at \( \lambda_{RB} \) of 1.1 (\( f_{RB}:10\%, \lambda_p:1.15 \)). The lowest TVFN concentration was estimated in the region where \( \lambda_{RB} \) equalled 0.9. At \( \lambda_{RB} \) near 1.0, the high TVFN concentration could be attributed to the large fraction of NOx, whereas at \( \lambda_{RB} \) less than 0.75, the high TVFN concentration could be attributed to large fractions of HCN and NH₃. For example with Scotts Branch coal at 1250°C, the TVFN concentration was 390 ppm, including 380 ppm NOx, at \( \lambda_{RB} \) of 1.15 (test RB51) compared to a TVFN concentration of 620 ppm, including 130 ppm NOx, at \( \lambda_{RB} \) of 0.7 (test RB50). At the optimum condition around \( \lambda_{RB} \) of 0.9, the measured TVFN concentrations for tests RB52 and RB53 were 230 ppm (including 175 ppm NOx) and 280 ppm (including 140 ppm NOx), respectively. Therefore, an optimum reburn zone stoichiometry exists for reducing the TVFN species concentration in the reburn zone.

4.3.4 Temperature

The effect of \( \lambda_{RB} \) on NOx emissions was strongly related to the reburn zone temperature, \( T_{RB} \), Figure 4.8. At 1100°C, 400 ppm NOx and \( T_{RB} \) of 0.3 s, the NOx concentration could not be reduced to less than 250 ppm. At 1400°C, the NOx
concentration was reduced from 360 ppm to 50 ppm as $\lambda_{RB}$ decreased from 1.0 to 0.8.

The effect of $T_{RB}$ and $\tau_{RB}$ on NOx and TVFN concentrations for Scotts Branch coal, with NOx$_{p}$ of 400 ppm and $\lambda_{RB}$ of 0.9, are shown in Figure 4.9. The two response curves had similar shapes. At 1100°C, the NOx and TVFN concentrations remained constant at approximately 250 and 500 ppm, respectively. At higher $T_{RB}$ and longer $\tau_{RB}$, the NOx and TVFN concentrations decreased. An NOx concentration of less than 50 ppm was estimated for $T_{RB}$ greater than 1350°C and $\tau_{RB}$ longer than 0.3 s. The TVFN concentration decreased to a minimum of 200 ppm at $T_{RB}$ of 1400°C and $\tau_{RB}$ of 1.0 s.

The decrease in TVFN concentration at higher temperature was due to the reduction of NOx and HCN, since the NH$_3$ concentration tended to remain constant or rise. For tests using Hugo coal, the NOx, HCN and NH$_3$ concentrations were respectively 320, 100 and 40 ppm at 1100°C (test RB7) compared to 100, 5 and 50 ppm at 1400°C (test RB45). For Obed Mountain coal, the NOx, HCN and NH$_3$ concentrations were respectively 180, 110 and 220 ppm at 1100°C (test RB10), compared to 40, 60 and 270 ppm at 1400°C (test RB47).

![Graph showing NOx concentration vs. reburn zone stoichiometry, $\lambda_{RB}$](image_url)

**Figure 4.8** Effect of reburn zone stoichiometry, $\lambda_{RB}$, and temperature, $T_{RB}$, on the NOx concentration for Scotts Branch coal, $\tau_{RB} = 0.3$ s and NOx$_{p} = 400$ ppm
Figure 4.9 Effect of reburn zone temperature, $T_{RB}$, and residence time, $\tau_{RB}$, on the NOx and TVFN concentrations for Scotts Branch coal, $\lambda_{RB}=0.9$ and NOx$_p=400$ ppm

A reburn zone temperature of 1400°C was necessary to obtain a NOx concentration of less than 100 ppm. NOx was reduced at higher $T_{RB}$ due to the strong temperature dependency of CH$_4$, O, and H radical formation. The high $T_{RB}$ also enhanced the reaction rate of HCN conversion (reactions R1 and R2 in Table 2.1, p.22). Thus, the reburn zone temperature should be maximized to promote radical formation and to accelerate reduction reaction rates.

4.3.5 Residence Time

Residence time, $\tau_{RB}$, had a strong influence on NOx and TVFN reduction, particularly for $T_{RB}$ greater than 1250°C, Figure 4.9. It was generally observed that $\tau_{RB}$ greater than 0.3 s was necessary to obtain absolute NOx concentrations less than 100 ppm. At 1400°C using Scotts Branch coal, the NOx concentration decreased from 280-360 ppm at 0.1 s (tests RB42 and RB43) to 30-50 ppm at 1.0 s (tests RB58 and RB59).

In order to determine the rate of change in NOx, HCN and NH$_3$ concentrations, more detailed measurements were made for three tests representing the most effective conditions for NOx reduction. These tests were designated DRB1, DRB2 and DRB3 to
distinguish them from the RB tests conducted in the parametric study. The test conditions for the detailed flames are given in Table 4.2. DRB1 was measured at 1250°C, $\lambda_{RB}$ of 0.75, 400 ppm inlet NOx and 0% $O_2$. DRB2 was measured at a temperature of 1400°C, $\lambda_P$ of 1.0 and inlet NOx of 400 ppm with $\lambda_{RB}$ set to 0.75 by injecting 25% reburn fuel. DRB3 was measured at a temperature of 1400°C, $\lambda_P$ of 1.075 and 400 ppm inlet NOx with $\lambda_{RB}$ set to 0.82 by injecting 25% reburn fuel.

In DRB1, the NOx concentration decreased from 330 ppm at 0.05 s to 180 ppm at 1.0 s, Figure 4.10. The concentrations of HCN and NH₃ rapidly increased to 200 and 90 ppm, respectively, in the first 0.1 s, then the HCN decayed from 200 to 90 ppm and the NH₃ increased from 90 to 165 ppm at 1.0 s. The CO concentration was approximately 1% for all measurements up to 1.0 s. In DRB2, the NOx concentration decreased from 400 to 30 ppm as the residence time increased from 0 to 1.0 s, Figure 4.11. The HCN concentration decreased from 160 to 30 ppm and the NH₃ concentration increased from 130 to 280 ppm as residence time increased from 0.05 to 0.3 s. In DRB3, the NOx concentration decreased from 400 ppm to 20 ppm over 1.0 s, Figure 4.12. The HCN concentration was less than 20 ppm at each sampling point and the NH₃ concentration increased from 15 ppm at 0.05 s to 110 ppm at 1.0 s.

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<td>$t_{RB}$ (s)</td>
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<td>$\lambda_{RB}$</td>
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Figure 4.10 Gas species profiles for detailed flame 1, DRB1, for Scotts Branch coal, $f_{RB} = 25\%$, $T_{RB} = 1250^\circ C$, $\lambda_f = 1.0$, $\lambda_{RB} = 0.75$ and NOx$_p$=400 ppm

The high concentrations of HCN and NH$_3$ for the first measurement point at 0.05 s in DRB1 and DRB2 could be partly attributed to the devolatilization of N-species, predominantly as HCN, from the reburn fuel [18,19]. To estimate the HCN that could have originated from the reburn fuel, the concentration of N-species devolatilized from Scotts Branch coal was calculated. It has been observed in coal characterization tests for Scotts Branch coal, that the fraction of coal nitrogen in the volatile products was equal to the volatile yield of 61% [89]. Therefore, 61% of the reburn coal nitrogen was assumed to be released as HCN before 0.05 s. This was equivalent to 340 ppm of HCN for 25% reburn fuel fraction. Added to the NOx$_p$ of 400 ppm, this implied that the inlet TVFN concentration was 740 ppm.
The reactions occurring in the first 0.05 s were highly complex since the reburn fuel was simultaneously devolatilizing, forming HCN and CH₃ radicals and reacting with primary NOx to form more HCN. However, the change in N-species concentrations after 0.05 s were less complex and showed clear trends. In DRB1 at 1250°C, the decay in NOx and HCN concentrations were much slower than measured in DRB2 at 1400°C. This suggested that the reaction rate was controlling the reduction process at the lower temperature. For DRB3 at 1400°C, the rate of NOx reduction was much slower and the rate of HCN conversion much faster than measured in DRB2. Thus, it appeared that reburn zone stoichiometry had some influence on the reduction rates.
Figure 4.12 Gas species profiles for detailed flame 3, DRB3, for Scotts Branch coal, $f_{RB} = 25\%$, $T_{RB} = 1400^\circ C$, $\lambda_p = 1.07$, $\lambda_{RB} = 0.82$ and NOx$_p = 400$ ppm

To determine whether these results could be predicted by a known mechanism for N-chemistry reactions, the rates of NOx and HCN reduction were predicted using an extended version of Fenimore's model for NOx decay in staged flames [26,27] (Equations 2.5-2.7, p.24). To separate the complex reactions occurring during devolatilization, the measured concentrations of NOx, HCN and NH$_3$ at 0.05 s were assumed to be initial values in the modelling. The H$_2$O concentration was calculated from the inlet CO$_2$ concentration and from the ratio of CO$_2$/H$_2$O for natural gas combustion.

The model accurately predicted the NOx and HCN concentrations in DRB2 and DRB3 up to the furnace position where their concentrations equalled 25 ppm, Figures 4.13b
and 4.13c. The model predicted that the NOx and HCN concentrations tended to zero whereas a residual concentration of 10-25 ppm was measured in the reactor. Mixing energy was insufficient after 0.5 s in the reactor to attain complete mixing of the species. The predicted rate of NOx reduction in DRB3 was slower than in DRB2 since the concentration of NH₃ was an order of magnitude less, leading to a lower rate for Equation 2.5. However, there was sufficient NH₃ in the reactor to obtain almost complete reduction of NOx in 1 s.

Figure 4.13 Prediction of NOx and HCN decay in the detailed flames using the NOx reduction model of Bose and Wendt [26]
Using the extended Fenimore model, the rate of NOx and HCN reduction in DRB1 was underpredicted by a factor 1.5, Figure 4.13a. It may have been possible that the OH radical concentration was not in equilibrium at these conditions. Bose et al [26] noted that the OH radical concentration was greater than equilibrium in rapidly cooled flames. Calculating the OH radical concentration using Equation 2.8 (p.25) as suggested by Bose et al, overpredicted the rate of NOx and HCN reduction. Thus, the OH concentration may have been between the equilibrium and non-equilibrium concentration.

The success of this relatively simple model to predict NOx and HCN reduction is useful in understanding reburning. It implies that the rate of NOx reduction after 0.05 s may be modelled with a knowledge of the NH3 concentration and the reactor temperature. It is therefore postulated that a sufficient NH3 concentration was necessary to reduce NOx in the reactor after 0.05 s. In coal reburning, this NH3 may be formed from the coal volatile nitrogen products or from the reburning process itself. If the NOx and NH3 concentrations were present in similar concentrations, the NOx and NH3 could be reduced to N2.

4.3.6 Primary NOx Concentration

The influence of the primary NOx concentration, NOxP, on the NOx and TVFN concentrations at the outlet of the reburn zone are shown for different gas temperatures in Figures 4.14 and 4.15, respectively. The points denote measured data and the lines were calculated from the response surface model. At 1100°C, the outlet NOx concentration increased from 200 to 400 ppm as NOxP increased from 200 to 600 ppm, Figure 4.14. At 1400°C, the outlet NOx concentration was approximately 50 ppm and was independent of NOxP in the range 200 to 600 ppm.

The data for TVFN concentration were plotted in Figure 4.15 for Hugo and Scotts Branch coals only. Obed Mountain coal was neglected since it produced 50-200 ppm higher TVFN for all test conditions which would have complicated the analysis. At 1100°C, the TVFN concentration increased from 450 to 700 ppm as NOxP increased from 200 to 600 ppm. At 1400°C, the TVFN concentration decreased from 270 to 200 ppm as NOxP increased from 200 to 600 ppm.
Figure 4.14 Effect of primary NOx, NOxP, on outlet NOx concentration at different temperatures for all coals, λP = 1.0, fRB = 20% and tRB = 0.3 s

To verify that NOx and TVFN were insensitive to NOxP, an additional test was conducted with NOxP of 2050 ppm. This test was designated test RB39a which could be compared directly with NOxP of 600 and 180 ppm from tests RB39 and RB41, respectively. The temperature for all three tests was 1400°C and λRB was 0.8.

The measured NOx concentration increased from 30-50 ppm for NOxP of 180 and 600 ppm, to 250 ppm for NOxP of 2050 ppm, Table 4.3. This suggested that there was a limit to the maximum concentration of NOxP which could be reduced at these conditions. There may have been insufficient CH4 radical species to reduce the NOx by Reactions 21-24 (p.21) or inadequate time to complete the reaction process. Despite the large concentration of NOx reduced in RB39a, the HCN concentration remained approximately constant and NH3 tended to decrease compared to tests RB39 and RB41, Table 4.3. The low concentration of NH3 was particularly surprising since NH3 was the predominant species measured at the end of the reburn zone for most high temperature tests. As such, it was expected that the NH3 concentration would rise for test RB39a.
Figure 4.15 Effect of primary NOx, NOx_p, on outlet TVFN concentration at different temperatures for Scotts Branch and Hugo coals, λ_F = 1.0, f_{RB} = 20% and τ_{RB} = 0.3 s

An alternate mechanism may have significantly affected NH3 reduction for test RB39a. Because of the high concentration of NOx in the reactor, NH3 may have been directly reduced with NO by Reaction 28 (p.24). Reaction 28 also was the basis for NOx reduction in the extended Fenimore model reported by Bose and Wendt [29] which was successfully applied in the Section 4.3.5. Thus, the NH3 and NO could also have been reduced through N2O to N2 under reducing conditions according to:

\[ \text{NH} + \text{NO} \rightarrow \text{N}_2\text{O} + \text{H} \]  \hspace{1cm} (R32)

\[ \text{N}_2\text{O} + \text{H} \rightarrow \text{N}_2 + \text{OH} \]  \hspace{1cm} (R33)

N2O measurements were made for gas samples collected in tests RB23, RB39, RB39a and RB41 to indicate the extent that Reaction R32 was important for NH3 and NO destruction in these tests. The gas samples were dried and collected in Tedlar bags and analyzed using a gas chromatograph equipped with an electron capture detector by Institut Français du Pétrole. The measured N2O concentrations were 0.5 - 4 ppm for all tests, Table 4.3. The N2O concentration increased from 0.5 ppm for test RB39 to
Table 4.3 Influence of Inlet NOx Concentration on N-species Concentration

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<tr>
<td>NOx</td>
<td>200</td>
<td>30</td>
<td>250</td>
<td>50</td>
</tr>
<tr>
<td>HCN</td>
<td>20</td>
<td>30</td>
<td>55</td>
<td>40</td>
</tr>
<tr>
<td>NH₃</td>
<td>8</td>
<td>140</td>
<td>150</td>
<td>220</td>
</tr>
<tr>
<td>N₂O</td>
<td>1.2</td>
<td>0.5</td>
<td>3.8</td>
<td>0.8</td>
</tr>
<tr>
<td>Predicted N₂O (ppm)</td>
<td>0.7</td>
<td>1.7</td>
<td>9.2</td>
<td>2.6</td>
</tr>
</tbody>
</table>

3.8 ppm for test RB39a, Table 4.3. Therefore, N₂O was being formed in low concentrations in the reburn zone.

The N₂O concentration was calculated from Reactions R32 and R33 assuming the rate of formation of N₂O was equivalent to its rate of destruction. It was considered that the NH was in equilibrium with NH₃ and H was in equilibrium with H₂. Therefore, the N₂O concentration was calculated from:

$$[\text{N}_2\text{O}] = \frac{k_{32}^{\text{K}_{\text{NH}}} [\text{NH}_3][\text{NO}]}{k_{33}^{\text{K}_{\text{NH3}}^2 \text{K}_H} [\text{H}_2]^{3/2}}$$

(4.2)

where the values of reaction rates $k_{32}$ and $k_{33}$ were identified from Thorne et al [23], and the values of the equilibrium constants $K_{\text{NH}}$, $K_{\text{NH3}}$ and $K_H$ were available from the Janaf Tables [88]. The equilibrium N₂O concentrations for the tests at 1400°C were consistently three times the measured N₂O, Table 4.3.

These measurements provide some evidence that Reactions R32 and R33 were an important path for NO and NH radical reduction. Therefore, it is suggested that NOx may be reduced to a low level of 30-50 ppm independent of NOxᵢ, provided that sufficient NH radicals are available in the reburn zone.
4.4 Conclusions of the Reburn Zone Investigation

The following conclusions may be made regarding the importance of the measured process variables on NOx and TVFN reduction in the reburn zone.

1. Reburn coal type was not a significant parameter effecting NOx reduction in the reburn zone. Obed Mountain and Scotts Branch hvb coals could be used to reduce NOx over a wide range of operating conditions while Hugo mvb coal could be used to reduce NOx to the same level over a narrower range.

2. Particle size distribution had little influence on NOx or TVFN reduction.

3. NOx reduction was strongly dependent on the reburn zone stoichiometry, λRB. The NOx concentration decreased with λRB, however, TVFN was minimum at λRB of 0.8 - 0.9.

4. Reburn zone temperature, TRB, had a strong effect on NOx and TVFN reduction. At 1100°C, the NOx concentration could only be reduced from an inlet concentration of 400 ppm to a minimum of 260 ppm. At 1400°C, the NOx concentration was reduced from 400 ppm to 30 ppm.

5. The fractional conversion of NOx usually increased as inlet NOx concentration to the reburn zone, NOxF, increased. At 1400°C, the NOx concentration was independent of NOxF and was reduced from 200 - 600 ppm to 50 ppm in 0.3 - 1.0 s. NOxF also had little effect on the concentrations of HCN or NH3. It was concluded that the NOx and NH3 could rapidly react together to form N2 at the conditions studied.

6. Reburn zone residence time, τRB, greater than 0.3 s was necessary to obtain NOx concentrations less than 100 ppm. The residence time necessary to obtain a given NOx reduction was dependent on TRB and the λRB. Less time was necessary to reduce the NOx at a higher TRB and lower λRB.

7. The NOx and HCN reduction occurring after 0.05 s could be accurately predicted using an extended Fenimore model for NOx reduction. This model did not include hydrocarbon/NOx reactions which suggested that NOx reduction was independent of the reactions with hydrocarbons after 0.05 s.

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5. Mixing Characterization Tests

The results of the Reburn Zone Investigation indicated the potential for NOx reduction in the reburn zone. However, these tests were conducted in a laboratory reactor under high mixing conditions. It has been reported that the effectiveness of the reburn process to reduce NOx is dependent on the ability to mix reburn fuel and NOx from the primary combustion products [31,42]. Thus, mixing characterization tests were conducted to develop a technique to study reburning for different levels of mixing between reburn fuel and primary combustion products. This technique was later applied in the Furnace Investigation described in Chapter 6.

The mixing characterization tests were conducted for a 2.5 MW, test furnace, Furnace no. 1. This furnace was sufficiently large to enable the mixing between the reburn fuel and primary combustion products to be varied. To adequately characterize the flow and mixing behaviour in Furnace no. 1, numerical flow computations, water model flow visualization and CO2 tracer studies were performed, and are discussed in Sections 5.1, 5.2 and 5.3, respectively. The method of varying mixing in the reburn zone of the Furnace Investigation is presented in Section 5.4.

The mixing rate of reburn fuel and primary combustion products was estimated using a dimension analysis of the species transport equation in the mathematical modelling, Section 5.5. The dimensional analysis was also applied to computational modelling results of the IPFR, used in the Reburn Zone Investigation, to compare mixing rates in the two sets of experiments.

5.1 Computational Modelling

A numerical simulation of the flow and enthalpy in the precombustor was made using the computational model described in Section 3.5. A fine grid with 40 and 60 grid lines in the radial and axial direction, respectively was used in the computations to accurately model the flow in the burner quarl. During the course of the solution, the grid was refined in the quarl to ensure grid independence. Iteration of the continuity, momentum, energy and species equations was continued until the residual change in velocities, turbulence quantities, enthalpy and species was less than $10^{-3}$. 

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In conducting the simulation, it was important to consider the appropriate boundary condition for particles that collided with the precombustor wall. Because the precombustor had a refractory lining, it would be covered in a molten slag layer and particles would stick on the surface. Therefore, particles that collided with the precombustor wall were assumed to be trapped. At that position, the remaining volatile matter was released and char combusted.

The simulation was conducted for a 2.0 MW$_c$ coal flame from the burner shown in Figure 3.8 (p.68) with the inlet conditions listed in Table 3.4 (p.70). Coal was injected through a central gun into the burner throat where it mixed rapidly with secondary air swirling at a level of 0.6 and a stoichiometry of 1.0. The streamlines up to 1.0 m axial distance are shown in Figure 5.1. The flame was predicted to have a small internal recirculation zone with a long well mixed region to the precombustor exit.

The temperature contours in the precombustor are also shown in Figure 5.1. A peak flame temperature of 1750°C at 0.2 m axial distance was predicted in the internal recirculation zone, decreasing to 1600°C after 1.0 m axial distance. Little heat loss occurred in the precombustor, and therefore, the temperature at the precombustor exit was 1500°C. Angular momentum was maintained and the swirl number at the exit of the precombustor was 0.3.

![Figure 5.1 Predicted streamlines and temperature contours in the precombustor for a 2.0 MW$_c$ coal flame at stoichiometry of 1.0 and swirl number of 0.6](image-url)
From the computations of axial velocity, the average residence time in the precombustor was estimated to be 0.25 s. This residence time should be sufficient to combust the coal before the precombustor exit.

An isothermal simulation was conducted to determine the flow in the reburn zone, shown in Figure 3.7 (p.68). The inlet conditions for velocity, turbulence and temperature were taken from the calculations at the exit of the precombustor and are listed in Table 3.4. The temperature for the simulation was 1500°C and the axial velocity increased from 5 m/s along the centerline to 9 m/s at the precombustor wall. The tangential velocity was set at 4 m/s, and thus the swirl number was 0.3.

The bulk of the forward flow was predicted to follow along the boundary of the chokes with a stagnant or small IRZ zone on the centerline, Figure 5.2. The peak velocity decreased with distance from the precombustor: 8 m/s at 0.6 m axial distance to 4 m/s at 1.4 m axial distance. The velocity along the centerline was between 0 and -1 m/s.

![Figure 5.2 Measured and predicted velocity profiles in the reburn zone.](image)

To verify the predicted flow pattern, a 2.0 MW, gas flame was set in the precombustor and, using a Prandtl tube, velocity measurements were taken in the reburn zone at 0.6, 1.05 and 1.4 m axial distance. The measurements were similar to the predicted velocity profiles, Figure 5.2. At 0.6 m axial distance, a peak velocity of 7 m/s was measured at 70-80 cm horizontal position. The velocity decreased to less than 1 m/s along the centerline. At 1.05 m axial distance, the peak velocity was 5 m/s and decreased to less than 1 m/s at 80 cm horizontal position. Thus, the flow followed along the choke walls and a weak internal recirculation zone formed along the centerline.
These computations indicated that the combustion products exiting the precombustor contained little unburned material and had a simple velocity profile. However, a relatively complex flow existed in the reburn zone due to the expansion of the flow.

5.2 Water model flow visualization

Experiments were then conducted using the water model, Figure 3.10 (p.71), to determine how the reburn fuel would be dispersed in the primary combustion products. The water model was used with primary, secondary and tertiary flows representing the primary combustion products, reburn fuel and tertiary air, respectively. The momentum ratio of primary to secondary flow, $G_p/G_s$, was adjusted to match the conditions expected in the Furnace Investigation (Chapter 6). These conditions are listed in Table 5.1 with reference made to the test numbers (F1, F2, F6 and F7) used in the Furnace Investigation. Momentum ratio was considered the most important criteria to maintain in the water model since it has a dominant influence on jet penetration [47]. The Reynolds number was also matched to simulate turbulent mixing. Temperature and density differences between the primary and reburn flows were not considered in the simulation since the reburn flow was a small fraction of the total flow and the reburn zone was considered isothermal.

The dispersion of black ink from the secondary flow into the primary flow provided a visual indication of the mixing. Regions of the flow which remained clear contained no secondary flow and dark regions were concentrated in secondary flow. It was found that the secondary injector position had the greatest influence on ink dispersion. With the injectors positioned at the front wall of the model, the ink was rapidly deflected away from the forward flow and entrained in the external recirculation zone, ERZ, located upstream of the first choke between the outer radius of the precombustor and the wall of the model. The ink dye in the secondary jets was unable to significantly penetrate the main forward flow due to the high momentum flux ratio of the forward flow relative to the jets. Decreasing the momentum ratio from 11 to 5 did not reduce the entrainment into the ERZ. The forward flow remained relatively clean while the ERZ filled with ink dye.

When the secondary injectors were positioned half way between the front wall and the first choke, there was little entrainment of ink into the ERZ and mixing of the ink with the forward flow was rapid. The ink distribution may be seen in Photographs 1 and 2
Table 5.1 Test Conditions for the Water Model Flow Simulation

<table>
<thead>
<tr>
<th>Test Number</th>
<th>F1</th>
<th>F2</th>
<th>F6</th>
<th>F7</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Furnace Investigation Conditions</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>primary stoichiometry</td>
<td>0.95</td>
<td>0.95</td>
<td>1.1</td>
<td>1.1</td>
</tr>
<tr>
<td>flow rate (kg/h)</td>
<td>2550</td>
<td>2250</td>
<td>2450</td>
<td>2150</td>
</tr>
<tr>
<td>velocity (m/s)</td>
<td>10</td>
<td>9</td>
<td>10</td>
<td>9</td>
</tr>
<tr>
<td>reburn fraction (%)</td>
<td>10</td>
<td>20</td>
<td>10</td>
<td>30</td>
</tr>
<tr>
<td>flow rate (kg/h)</td>
<td>75</td>
<td>100</td>
<td>100</td>
<td>130</td>
</tr>
<tr>
<td>velocity (m/s)</td>
<td>30</td>
<td>30</td>
<td>30</td>
<td>30</td>
</tr>
<tr>
<td>momentum ratio (G_p/G_s)</td>
<td>11</td>
<td>7</td>
<td>8</td>
<td>5</td>
</tr>
<tr>
<td><strong>Water Model Simulation</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>primary flow rate (kg/h)</td>
<td>5500</td>
<td>4900</td>
<td>5400</td>
<td>4700</td>
</tr>
<tr>
<td>velocity (m/s)</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>secondary flow rate (kg/h)</td>
<td>170</td>
<td>190</td>
<td>190</td>
<td>220</td>
</tr>
<tr>
<td>velocity (m/s)</td>
<td>0.9</td>
<td>1.0</td>
<td>1.0</td>
<td>1.1</td>
</tr>
<tr>
<td>tertiary flow rate (kg/h)</td>
<td>1700</td>
<td>1700</td>
<td>1700</td>
<td>1700</td>
</tr>
<tr>
<td>velocity (m/s)</td>
<td>0.6</td>
<td>0.6</td>
<td>0.6</td>
<td>0.6</td>
</tr>
<tr>
<td>momentum ratio (G_p/G_s)</td>
<td>11</td>
<td>7</td>
<td>8</td>
<td>5</td>
</tr>
</tbody>
</table>

where the dark regions represent flow containing secondary flow. The ink dye filled the forward flow region between the two chokes. With the injectors positioned at the edge of the first choke, the mixing was much slower as shown in Photograph 3. The ink dye only started to fill the forward flow at the end of the second choke. Changing the momentum ratio between 5 and 11 had little effect on the observed mixing pattern.

These observations showed that the mixing of primary and secondary flows was dominated by the primary flow behaviour at the position of secondary flow injection. They also showed that the reburn fuel pipes would need to be inserted at least half way to the first choke to obtain sufficient mixing between reburn fuel and primary combustion products.
5.3 CO₂ Tracer Studies

CO₂ tracer studies were conducted in Furnace No. 1 to quantify mixing of reburn fuel and primary combustion products. CO₂ was injected through the reburn guns, Figure 3.7 (p.68), to mix with air exiting the precombustor which was preheated to 500°C. The momentum ratio of the CO₂ to hot air was matched to the conditions designated for flame F7 of the Furnace Investigation, Table 5.1. Based on the results of the water model flow visualization, the reburn gun position was adjusted to vary the mixing level. The reburn guns were positioned either half-way between the front wall and the first choke, 0.35 m axial distance, or at the front edge of the first choke, 0.7 m axial distance. These locations were termed position 1 and position 2, respectively. With the reburn guns at position 1, the CO₂ concentration was measured at three cross-sections of the reburn zone located at 0.6, 1.05 and 1.4 m axial distance. For gun position 2, the CO₂ concentration was measured at two cross-sections of the reburn zone located at 1.05 and 1.4 m axial distance.

With the reburn guns at position 2, there was a large gradient in CO₂ concentration at 1.05 m axial distance from 0.05% at the outer edge of the forward flow (50 cm horizontal position) to 4% along the centerline, Figure 5.3. At 1.4 m axial distance, the gradient was less with CO₂ concentration increasing from 0.24% around the outside of the forward flow to 3% at the centerline. The mixing factor, Nₚ, calculated using

![Figure 5.3 CO₂ concentration profiles with the reburn guns at position 2, low mixing case.](image-url)
Equation 3.5 (p.72), was only 0.4 at 1.4 m axial distance, Figure 5.4, indicating that the reburn fuel would not be well mixed at any location in the reburn zone.

With the reburn guns at position 1, the CO₂ mixed rapidly with the primary flow, Figure 5.5. At 0.6 m axial distance, the position of the reburn jets was apparent from the high concentration of CO₂. After 1.05 m, the CO₂ was well dispersed and gas concentration varied from 0.5% at the outside edge of the forward flow to 1.3% at the centerline. The mixing factor, Nᵢ, increased to a well mixed level (approximately 1.0) at 1.05 m axial distance. These results confirmed the importance of reburn gun position on the mixing between the reburn fuel and primary combustion products.

Figure 5.4 Effect of reburn gun position on the mixing factor
5.4 Summary of the Mixing Predictions

The results of the water model flow visualization and CO$_2$ tracer studies demonstrated the dominance of the primary flow on mixing with the reburn jets. Simply changing the reburn gun position significantly altered the reburn jet trajectories and mixing behaviour.

The results of the computational modelling and CO$_2$ tracer studies provided quantitatively similar information regarding the mixing in the reburn zone. An example of this is a comparison of the measured CO$_2$ concentration from the CO$_2$ tracer studies, and the volatile matter concentration, predicted from computational modelling of reburn fuel injected into the reburn zone, with the reburn guns at positions 1 and 2. In order to compare the CO$_2$ and volatile matter, the concentrations were normalized to the average concentration in the flow. Good qualitative agreement between the normalized CO$_2$ and the volatile matter concentrations was obtained, Figure 5.6. With the reburn guns at position 1, the normalized volatile matter and CO$_2$ concentrations after the first choke were 0.8-1.2. With the reburn guns at position 2, the normalized volatile matter and CO$_2$ concentrations along the centerline after the first choke were 3-4.
Figure 5.6 Comparison of measured CO₂ and predicted volatile matter concentrations in the reburn zone

The three techniques were sufficiently simple to demonstrate that a large change in reburn fuel mixing occurred by varying the gun position. With the reburn guns at position 1, the reburn fuel would be dispersed in the primary combustion products and with the reburn guns at position 2, the reburn fuel would be concentrated around the centerline. Therefore, it was decided to vary only the gun position to obtain the low and high mixing conditions in the Furnace Investigation.
5.5 Characterization of Mixing Rates

The mixing rate in the reburn zone was characterized by dimensional analysis of the transport equation for the time averaged species concentration in a turbulent flow, $C_i$. From the computational model presented in Section 3.4, this transport equation has the form:

$$
\rho U_i \frac{dC_i}{dx_i} = \frac{\mu_e}{Sc} \left[ \frac{\partial^2 C_i}{\partial x_i^2} \right] + S_j
$$

(3.11)

where $U_i$ is the velocity in the $x_i$ direction, $\rho$ is the gas density, $Sc$ is the Schmidt number and $\mu_e$ is the effective viscosity.

The left hand side of Equation 3.11 represents the convective mixing of the species in the flow. A characteristic time scale of convective mixing, $L/U$, may be derived from a dimensional analysis of Equation 3.11. Here, $L$ represents a characteristic distance between reburn fuel and primary combustion products and $U$ is the characteristic velocity of the flow. The first term on the right hand side of Equation 3.11 represents the turbulent diffusion mixing of the species in the flow. A characteristic time scale for the turbulent diffusion mixing is $L^2\epsilon/k^2$ where $k$, the turbulent kinetic energy, and $\epsilon$, the turbulent dissipation, are calculated from the computational modelling. The second term on the right hand side of Equation 3.11 represents a source or sink term for the species due to reaction in the flow. Assuming that the rate of reaction is controlled by mixing of reactants inside a fluid eddy, a characteristic reaction time scale for $S_j$, $k/\epsilon$, may be deduced from the eddy break-up model [80].

The magnitude of these three time scales are compared for the two experiments using length and velocity scales estimated from the experiments. The characteristic distance between the reburn fuel and primary combustion products, $L$, was estimated as the radial distance between the reburn gun and the bulk of the forward flow, Figure 5.7. The characteristic velocity, $U$, was estimated as the average velocity of the primary combustion products at the point of reburn fuel injection. The turbulent energy, $k$, and dissipation, $\epsilon$, were approximated from the calculated values for the forward flow near the point of reburn fuel injection. The mixing times for the Reburn Zone Investigation and low and high mixing cases of the Furnace Investigation are shown in Table 5.2.
The mixing rate of reburn fuel with primary combustion products was significantly faster in the Reburn Zone Investigation, using the IPFR, compared to the Furnace Investigation, using Furnace No. 1. In the IPFR, the convective mixing time was short (0.01 s) due to rapid impingement of primary combustion products on the reburn coal jet. The impingement generated a high level of turbulence and the characteristic reaction time, $k/e$, was estimated at 0.01 s. These mixing times were of a similar order of magnitude to the characteristic times for nitrogen chemistry reactions [90].
Table 5.2 Estimation of Mixing Rates

<table>
<thead>
<tr>
<th>Gun Position (m axial distance)</th>
<th>Furnace no. 1 high mixing</th>
<th>low mixing</th>
<th>IPFR</th>
</tr>
</thead>
<tbody>
<tr>
<td>U (m/s)</td>
<td>0.35</td>
<td>0.3</td>
<td>6</td>
</tr>
<tr>
<td>L (m)</td>
<td>8.5</td>
<td>0.35</td>
<td>0.05</td>
</tr>
<tr>
<td>k (m²/s³)</td>
<td>0.7</td>
<td>6.3</td>
<td>3</td>
</tr>
<tr>
<td>ε (m²/s³)</td>
<td>9</td>
<td>54</td>
<td>300</td>
</tr>
<tr>
<td>μ₀ (kg/m/s)</td>
<td>0.005</td>
<td>0.065</td>
<td>0.001</td>
</tr>
<tr>
<td>L/U (s)</td>
<td>0.01</td>
<td>1</td>
<td>0.01</td>
</tr>
<tr>
<td>L²ε/k² (s)</td>
<td>0.2</td>
<td>0.8</td>
<td>0.08</td>
</tr>
<tr>
<td>k/ε (s)</td>
<td>0.08</td>
<td>0.1</td>
<td>0.01</td>
</tr>
</tbody>
</table>

For the high mixing case in Furnace No. 1, the convective mixing time was short due to the proximity of the reburn fuel in the forward flow. However, the reaction time was estimated to be 0.08 s which was an order of magnitude longer than characteristic times for nitrogen chemistry reactions [90].

For the low mixing case in Furnace no. 1, the convective and turbulent diffusion mixing were estimated at approximately 1 s. This suggests that the large scale mixing between the reburn fuel and the primary combustion products would not be complete at the end of the reburn zone. The characteristic reaction time was estimated at 0.1 s which indicated that NOx could be rapidly reduced in the regions of the flow where reburn fuel and primary combustion products were mixed.

5.6 Concluding Remarks

A simple technique was developed to vary the mixing of reburn fuel and primary combustion products for the Furnace Investigation. By positioning the reburn guns half-way between the front wall and the first choke, position 1, the reburn fuel was dispersed into the primary combustion products. With the reburn guns positioned at the front of the first choke, position 2, the reburn fuel was concentrated along the centerline. The mixing rate was calculated to be significantly slower in the Furnace Investigation than in the Reburn Zone Investigation. Thus, NOx reduction could be compared for a wide range of mixing rates.
6. Furnace Investigation

The purpose of the Furnace Investigation was to determine the influence of process parameters on NOx reduction under mixing limited conditions. The experiments were conducted at the optimum process conditions identified in the Reburn Zone Investigation: reburn zone stoichiometry of 0.8 to 0.95, temperature of approximately 1400°C and residence time greater than 0.3 s.

The influence of mixing on NOx emissions was measured for seven flames with coal reburn fuel and one flame with natural gas reburn fuel. The coal reburning flames were designated as F1-F7 and the natural gas reburning flame was designated as F8. All of the input conditions and flue gas emissions for flames F1-F8 are summarized in Appendix B. The NOx emissions were related to $\lambda_{RB}$, and different tertiary air injection positions, Section 6.2. Natural gas and coal reburn fuels are compared on the basis of NOx emissions and coal burnout.

After completing the Furnace Experiments, it was found that the conditions studied in the Reburn Zone Experiments were not the same. Additional reburn zone tests were conducted in the IPFR to compare the effect of large changes in mixing level on NOx emissions, Section 6.3.

6.1 Evaluation of Combustion Zone Separation

A precombustor and refractory choke walls were added to Furnace No. 1 to control the process conditions in the primary, reburn and tertiary combustion zones. The success of separating the combustion zones is discussed in this Section.

6.1.1 Primary Coal Burnout

To separate the primary and reburn zones, it was preferable to have complete primary coal burnout in the precombustor. Complete coal burnout would ensure that excess $O_2$ did not flow from the precombustor into the reburn zone and that primary zone char did not need to be combusted in the tertiary zone. The burnout could be calculated from the
fraction of coal converted to gas products by comparing the measured and theoretical carbon fraction in the gas product \((\text{CO}_2 + \text{CO})\) exiting the precombustor. The gas composition was calculated from measurements across the precombustor exit at 0.7-1.0 m horizontal position and weighted for the mass flow rate at each measurement position.

For every flame, the mass flow averaged \(\text{CO}_2+\text{CO}\) concentration indicated that the coal was almost entirely converted to gas products, Figure 6.1 top. The solid points, representing the average \(\text{CO}_2+\text{CO}\) concentration measured at the end of the precombustor, lay on the line for 100% coal conversion. The error bars, representing the range in measured \(\text{CO}_2+\text{CO}\) concentration at the end of the precombustor, were greater than the 95% line for every flame.

The coal conversion at the end of the precombustor was also calculated from an ash balance of the coal and char according to:

\[
\text{coal conversion} = 1 - \frac{1 - \text{ash}_{\text{coal}}/\text{ash}_{\text{char}}}{1 - \text{ash}_{\text{coal}}} \quad (6.1)
\]

The coal conversion by ash balance increased from 78 to 87% as the set point stoichiometry increased from 0.95 to 1.0 (Figure 6.1 bottom). There was no observed increase in conversion at stoichiometry greater than 1.0. The low measured conversion may have been due to a selective collection of unburned carbon particles. It is estimated that 50% of the ash exited the precombustor to the reburn zone as a molten slag. Assuming that there was no carbon in the slag, the conversion at the end of the precombustor may have been substantially underpredicted.

Because of the problem of obtaining a representative solid sample at the end of the precombustor, it is estimated that the coal conversion to gas products was between 95 and 100% for the flames. Therefore, the primary and reburn zones were reasonably separated.
6.1.2 Tertiary Air Recirculation Tests

To separate the reburn and tertiary combustion zones, it was preferable to prevent tertiary air recirculation into the reburn zone. Tertiary air recirculation would increase the stoichiometry in the reburn zone. The fraction of air which could be recirculated into the reburn zone was tested using a helium tracer gas injected at tertiary air pipe
 positions 1, 2 and 4 (see Figure 3.7, p.68) at a concentration of 1.3% of the total flow. For each injection position, the helium concentration was measured at the front side of the second choke at 1.4 m axial distance. Recirculated mass fraction, $M_R$, was related to the measured and average helium concentrations by the expression:

$$M_R = \frac{[\text{He}]_{\text{meas}}}{[\text{He}]_{\text{avg}}} \quad (6.2)$$

For each injection location, $M_R$ was highest at the outermost radial position of the forward flow around 40 cm horizontal position and was less than 10% along the centerline, Figure 6.2 top. $M_R$ decreased as the injection point was moved downstream. The value of $M_R$, measured at 40 cm horizontal position and 1.4 m axial distance, decreased from 63%, when helium was injected at tertiary pipe position 1, to 17% when helium was injected at tertiary pipe position 2.

The recirculation tests indicated that the chokes were only partly effective in eliminating tertiary air recirculation into the reburn zone. The tertiary air was entering the reburn zone from the outside of the forward flow. Recirculated tertiary air probably increased the reburn zone stoichiometry, $\lambda_{RB}$, by increasing the $O_2$ supplied. As the fraction of recirculated air increased with distance from the centerline, the tertiary air was mixed with the primary combustion products on the perimeter of the reburn zone.

An effective reburn zone stoichiometry, $\lambda_{\text{eff}}$, could be estimated from the calculated reburn zone stoichiometry, $\lambda_{RB}$. The value of $\lambda_{\text{eff}}$ was calculated as the air supplied in the primary combustion and reburn zones divided by the air required to burn the coal, $\Phi_c$. The air supplied was the sum of the primary and reburn air, $\Phi_c\lambda_{RB}$, plus the recirculated tertiary air from the first and second tertiary air injection locations according to:

$$\text{air available} = \Phi_c\lambda_{RB} + M_i(\Phi_c-\Phi_c\lambda_{RB}) + M_i(0.15\Phi_c) \quad (6.3)$$

and therefore:

$$\lambda_{\text{eff}} = \lambda_{RB} + M_i(1-\lambda_{RB}) + M_i(0.15) \quad (6.4)$$
Figure 6.2 Calculation of an effective reburn zone stoichiometry, $\lambda_{\text{eff}}$, due to tertiary air recirculation.

where $M_i$ and $M_f$ are the fractions of tertiary air recirculated from the initial and final tertiary air injection pipe. $M_R$ was assumed to accurately represent tertiary air recirculation since differences in molecular diffusion of helium tracer and air should not be important in these high convection flows. The fraction of air recirculated from each
pipe, \( M_1 - M_4 \), could thus be calculated. The relationship between \( \lambda_{\text{eff}} \) and \( \lambda_{RB} \) is shown in Figure 6.2 bottom. Along the centerline, \( \lambda_{\text{eff}} \) was insensitive to the tertiary air injection location, increasing from 0.78 to 0.96 as \( \lambda_{RB} \) increased from 0.75 to 0.95. In the bulk of the forward flow at 50 cm horizontal position, \( \lambda_{\text{eff}} \) was sensitive to tertiary air injection location. With tertiary air injected equally at pipe positions 1 and 2, \( \lambda_{\text{eff}} \) increased from 0.89 to 1.01 as \( \lambda_{RB} \) increased from 0.75 to 0.95. With tertiary air injected equally at pipe positions 2 and 4, \( \lambda_{\text{eff}} \) increased from 0.84 to 0.99 as \( \lambda_{RB} \) increased from 0.75 to 0.95.

These calculations show that a distribution in reburn zone stoichiometry probably existed at the reburn zone exit independent of the extent of mixing. As the bulk of the flow passed through the reburn zone at 50 cm horizontal position, the stoichiometry calculated at that position was assumed to be \( \lambda_{\text{eff}} \).

6.2 Effect of Process Parameters on NOx Emissions

Several general observations concerning the Furnace Investigation can be made. Specifically, the NOx concentration from the precombustor and the mixing of reburn fuel and primary combustion products are reported.

The gas concentration exiting the precombustor was measured at 0.1 m axial distance for each flame. The concentration profiles across the precombustor exit were flat for each flame. This result supported the conclusion, made from the computational modelling of the precombustor, discussed in Section 5.1, that the precombustor products were well-mixed. Thus, a single measurement could be used to describe the gas concentration exiting the precombustor.

The NOx concentration exiting the precombustor was generally high for all tests (770-1000 ppm) and relatively insensitive to changes in operating conditions. This was partially the result of the high mixing in the precombustor which caused rapid oxidation of the fuel nitrogen and partially the result of thermal NOx which could be significant in the precombustor due to high wall temperatures (1400-1600°C) and relatively low heat extraction (25% of the thermal input).

The behaviour of reburn coal jets with the reburn guns at position 1 (see Figure 5.6: high mixing condition, p.115) is shown in Photographs 4 and 5. The coal, exiting the reburn guns, was rapidly ignited by the hot primary combustion products. By the end of
the first choke, a significant amount of the jets appeared to be entrained in the primary combustion products and by the end of the second choke, the jets were distributed across the entire cross-section of the forward flow. With the reburn guns at position 2, (low mixing condition), the four reburn jets remained distinct and concentrated in the center of the flow up to the end of the second choke. This observation supported the conclusions made in the mixing characterization studies, Chapter 5, that moving the gun from position 1 to position 2 substantially lowered the mixing of reburn fuel and primary combustion products.

There remained an apparent unmixedness at the end of the second choke for both the high and low mixing cases. This is characterized by non-luminous and luminous flows as evident in Photograph 5 taken between the first and second choke. Judging reburn jet mixing based on luminosity criteria may be misleading since volatile reburn fuel components may mix with primary combustion products and appear non-luminous. However, the luminous jets indicate the general character of reburn fuel particles in the forward flow. Turbulent mixing of the two streams was probably not complete before tertiary air addition even under high mixing conditions (as discussed in Section 5.4).

6.2.1 Reduction Behaviour in the Reburn Zone

To investigate the reduction of NOx in the reburn zone, detailed in-flame gas composition measurements were performed for flame F7 with $\lambda_P$ of 1.0 and $f_{RB}$ of 20%. The reburn guns were located at position 1 and the tertiary air was injected at positions 2 and 4. Gas samples were collected upstream and downstream of the two chokes and the first tertiary air injector at 0.6, 1.05, 1.4, 1.8, 2.2 and 3.0 m axial distance.

The gas temperature profile in the reburn and tertiary combustion zones was measured at 0.6, 1.05, 1.4, 1.8, 2.2 and 3.0 m axial distance. The primary combustion products exited the precombustor at 1500°C and decreased to 1400°C by the end of the reburn zone, Figure 6.3. There was a steady decrease in the gas temperature along the tertiary combustion zone due to heat extraction from the six water-cooled pipes located along the furnace wall. By the chimney, the gas temperature had decreased to 1000°C. An average gas residence time was calculated by integrating the temperature dependent velocity from the gas flow through the furnace. The residence time was approximately 0.5 s in the reburn zone and 4.5 s in the tertiary combustion zone.
The primary combustion products contained 1% O₂, 1% CO and 900 ppm NOx. The gas concentration profiles upstream and downstream of the second reburn zone choke are shown in Figures 6.4a and 6.4b, respectively. Before the first choke at 0.6 m axial distance, a sharp peak in O₂ of 15% was measured at 60 cm horizontal position which coincided with the trajectory from one of the reburn jets. Outside of this narrow region of the flow, the NOx concentration was near 900 ppm which indicated that no reduction had occurred at that position. Downstream of the first choke at 1.05 m axial distance, the O₂ peak was absent and the CO concentration was greater than 1% across the entire forward flow with a peak of 7.5% at 60 cm horizontal position. The CO peak corresponded to a minimum NOx concentration of 280 ppm, although the HCN and NH₃ concentrations were less than 50 ppm. Upstream of the second choke at 1.45 m axial distance, the NOx started to decrease across the entire reburn zone from 780 ppm along the centerline to 440 ppm at 40 cm horizontal position. Downstream of the second choke at 1.8 m axial distance, the NOx concentration decreased substantially to 350 ppm at 40-60 cm horizontal position. The HCN and NH₃ concentrations continued to rise with peak concentrations of 415 and 150 ppm, respectively, at 80 cm horizontal position.
Figure 6.4a Gas concentration profiles upstream of the second choke for flame F7: $\lambda_p = 1.0$, $f_{RB} = 20\%$ and $\lambda_{RB} = 0.8$.

Upstream of the first tertiary air injection position at 2.2 m axial distance, the NOx concentration continued to decrease to 300 ppm across the entire flow. Peaks in HCN and NH$_3$ were again measured at 80 cm horizontal position, although, the values were less than measured downstream of the second choke. This was the first measurement location where the CO concentration decreased and the O$_2$ concentration increased.
Figure 6.4b Gas concentration profiles downstream of the second choke for flame F7: $\lambda_p = 1.0$, $f_{RB} = 20\%$ and $\lambda_{RB} = 0.8, 20\%$ and $\lambda_{RB} = 0.8$.

which indicated the presence of tertiary air. Downstream of the first tertiary air injection at 3 m axial distance, the O$_2$ and CO concentrations were 1-2% and 0-0.3%, respectively. In the flue gas, the CO was only 9 ppm and the NOx concentration was 300 ppm. The change in CO concentration from 0.3-2.2% upstream of the tertiary air injector to less than 0.1% downstream demonstrates the rapid oxidation of CO in these
tests. This was due to the high velocity of tertiary air, 50 m/s, injected at pipe position 1 which promoted fast mixing and the high temperature, 1300-1400°C, which enhanced reaction rates.

These measurements demonstrated that with the reburn gun at position 1, the reburn zone was O₂ lean after the first choke. NOx reduction occurred gradually up to the point of the first tertiary air injection, at which point, HCN, NH₃ and CO were rapidly converted to final products.

6.2.2 Reburn Zone Mixing

The effect of reburn zone mixing on NOx reduction was studied for flames F1-F6 by measuring the gas emissions with the reburn guns at position 1 and 2. For most of the flames, the concentration of NOx, HCN, NH₃, CO and O₂ was also measured at the end of the reburn zone around 1.8 m axial distance. The concentration profiles and emissions data for flames F1 to F6 are presented in Figures 6.5 to 6.10, respectively. In the top part of each figure, the gas concentration profiles at the end of the reburn zone are presented. In the bottom part of each figure, the average process conditions at the end of the precombustor and emissions in the flue gas are listed in tabular form.

Flame F1 was established at a primary stoichiometry, λₚ, of 0.95 and a reburn fuel fraction, f_RB, of 10%. Primary combustion products exited the precombustor at a temperature of 1460°C and contained 0.6% O₂, 1% CO and 850 ppm NOx, Figure 6.5. With the reburn guns at position 1, the high mixing case, the O₂ concentration was reduced to 0% at the end of the reburn zone while the CO concentration rose to 1-2% with a peak observed at 70 cm horizontal position, Figure 6.5 top. The NOx concentration at the end of the reburn zone increased from 520 ppm at 40 cm horizontal position to 740 ppm along the centerline. NOx emissions were similar to the concentration measured at the end of the reburn zone. With tertiary air injected at pipe positions 1 and 2, the NOx emission was 500 ppm and with tertiary air injected at pipe positions 2 and 4, the NOx emission was 460 ppm. The CO was effectively combusted in the tertiary combustion and concentrations of 1-4 ppm were detected.

With the reburn guns at position 2, the low mixing case, the shape of the O₂, CO and NOx concentration profiles at the end of the reburn zone were similar to those measured with the reburn guns at position 1. The O₂ concentration was reduced to 0.1% and the
CO concentration rose to 0.5-2.5% with a peak observed at 70 cm horizontal position, Figure 6.5 bottom. The NOx concentration increased from 550 ppm at 40 cm horizontal position to 760 ppm along the centerline. In the bulk of the forward flow around 40-60 cm horizontal position, the NOx concentration was 30-100 ppm higher with the reburn guns at position 2 than at position 1. Accordingly, the NOx emissions in the flue

![Graphs showing NOx and CO concentrations at reburn gun positions](image)

**top** reburn zone exit gas concentrations

<table>
<thead>
<tr>
<th>location</th>
<th>reburn gun position</th>
<th>tert. air pipe pos.</th>
<th>O2 (%)</th>
<th>CO (ppm)</th>
<th>NOx (ppm)</th>
<th>burnout (%)</th>
<th>temp. (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>precomb.</td>
<td></td>
<td></td>
<td>0.6</td>
<td>1</td>
<td>850</td>
<td></td>
<td>1460</td>
</tr>
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<td>1</td>
<td>1,2</td>
<td>3</td>
<td>4</td>
<td>500</td>
<td></td>
<td>1000</td>
</tr>
<tr>
<td>flue</td>
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<td>2,4</td>
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<td>1</td>
<td>460</td>
<td></td>
<td>1000</td>
</tr>
<tr>
<td>flue</td>
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<td>1,2</td>
<td>3</td>
<td>0</td>
<td>540</td>
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<td>1000</td>
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<tr>
<td>flue</td>
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<td>3</td>
<td>1</td>
<td>450</td>
<td></td>
<td>1000</td>
</tr>
</tbody>
</table>

**bottom** precombustor and flue gas emissions

Figure 6.5 Gas concentration profiles at the end of the reburn zone and data on the precombustor and flue gas emissions for Flame F1: $\lambda_F = 0.95$, $f_{RB} = 10\%$ and $\lambda_{RB} = 0.87$
gas were approximately 40 ppm higher with the reburn guns at position 2 than at position 1. Thus, mixing had only a small effect on the gas concentration profiles in the reburn zone and the NOx emissions for flame F1.

Flame F2 was conducted at the same $\lambda_p$ as flame F1, 0.95, but with a higher $f_{RB}$ of 20%. Primary combustion products exited the precombustor at a temperature of 1490°C and contained 0.2% O$_2$, 1.8% CO and 780 ppm NOx, Figure 6.6. With the reburn guns at position 1, the O$_2$ concentration was reduced to 0% at the end of the reburn zone while the CO concentration rose to 0.5-3% with a peak observed at 80 cm horizontal position. HCN and NH$_3$ were detected across the entire reburn zone exit at concentrations in the range 10-100 ppm. The NOx concentration was 400 ppm over most of the reburn zone exit with 600 ppm measured at a single point on the centerline. The NOx emission in the flue gas was lower than measured at the reburn zone exit. With tertiary air injected at pipe positions 1 and 2, the NOx emission was 360 ppm and with tertiary air injected at pipe positions 2 and 4, the NOx emission was 250 ppm. As was found in flame F1, the CO was effectively combusted in the tertiary combustion and concentrations of 2-7 ppm were detected.

With the reburn guns at position 2, the shape of the O$_2$, CO HCN, NH$_3$ and NOx concentration profiles at the end of the reburn zone were substantially different. The O$_2$ concentration was higher at 0.2-0.5% and a peak CO concentration of 4.5% was measured at 80 cm horizontal position. The HCN and NH$_3$ had similar concentration profiles as CO with peaks of 230 and 120 ppm, respectively, at 80 cm horizontal position. The HCN and NH$_3$ concentrations decreased rapidly with radial distance and neither species was detected at 40 cm horizontal position where the flow was oxygen rich. The NOx concentration increased from 320 ppm at 80 cm horizontal position to 540-570 ppm in the bulk of the forward flow around 40-60 cm horizontal position. The NOx concentration at the end of the reburn zone was 140-170 ppm higher with the reburn guns at position 2 than at position 1. Accordingly, the NOx emissions in the flue gas were approximately 70-90 ppm higher with the reburn guns at position 2 than at position 1. Thus, mixing had a significant effect on the gas concentration profiles in the reburn zone and the NOx emissions for flame F2.

Flame F3 was conducted at $\lambda_p$ of 1.04 and $f_{RB}$ of 10%. Few measurements were made for this flame because of the poor NOx reduction. Without reburn fuel injection and tertiary air injected at pipe position 1, the NOx emission in the flue gas was 910 ppm, Figure 6.7. The NOx emissions could be reduced to 810 ppm by injecting the tertiary air
top reburn zone exit gas concentrations

<table>
<thead>
<tr>
<th>location</th>
<th>reburn gun position</th>
<th>tert. air pipe pos.</th>
<th>$O_2$ (%)</th>
<th>CO (ppm)</th>
<th>NOx (ppm)</th>
<th>burnout (%)</th>
<th>temp. (°C)</th>
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<td>0.2</td>
<td>1.8%</td>
<td>780</td>
<td>78</td>
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<td>2.4</td>
<td>2.9</td>
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<td>360</td>
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<td>1000</td>
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<tr>
<td>flue</td>
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<td>250</td>
<td>92</td>
<td>1000</td>
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<td>2.7</td>
<td>4</td>
<td>430</td>
<td>97</td>
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<td>3.4</td>
<td>33</td>
<td>340</td>
<td>92</td>
<td>1000</td>
</tr>
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</table>

bottom precombustor and flue gas emissions

Figure 6.6 Gas concentration profiles at the end of the reburn zone and data on the precombustor and flue gas emissions for Flame F2: $\lambda_p = 0.95$, $f_{RB} = 20\%$ and $\lambda_{RB} = 0.78$
**top** reburn zone exit gas concentrations

**bottom** precombustor and flue gas emissions

<table>
<thead>
<tr>
<th>location</th>
<th>reburn gun position</th>
<th>tert. air pipe pos.</th>
<th>( \text{O}_2 ) (%)</th>
<th>CO (ppm)</th>
<th>NOx (ppm)</th>
<th>burnout (%)</th>
<th>temp. (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>precomb.</td>
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<td>1</td>
<td>4.2</td>
<td>0</td>
<td>900</td>
<td>880</td>
<td>1490</td>
</tr>
<tr>
<td>flue</td>
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<td>4</td>
<td>3.6</td>
<td>0</td>
<td>810</td>
<td>900</td>
<td>880</td>
</tr>
<tr>
<td>flue</td>
<td>1</td>
<td>1.2</td>
<td>3.9</td>
<td>1</td>
<td>620</td>
<td>940</td>
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<td>flue</td>
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<td>1.3</td>
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<td>620</td>
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<tr>
<td>flue</td>
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<td>2.4</td>
<td>3.6</td>
<td>0</td>
<td>610</td>
<td>880</td>
<td></td>
</tr>
</tbody>
</table>

Figure 6.7 Gas concentration profiles at the end of the reburn zone and data on the precombustor and flue gas emissions for Flame F3: \( \lambda_P = 1.04, \text{f}_{RB} = 10\% \) and \( \lambda_{RB} = 0.96 \)

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at pipe position 4. Upon injecting reburn fuel at gun positions 1 or 2, the NOx emissions decreased to 610-620 ppm. There was little influence of tertiary air pipe position on the NOx emissions or carbon burnout. This was due to the high O₂ concentration, 2%, at the end of the reburn zone. Thus, there was insufficient reburn fuel to obtain substantial NOx reduction.

Flame F4 was conducted at the same λₚ as flame F3, 1.04, but with a higher f_RB of 20%. Primary combustion products exited the precombustor at a temperature of 1490°C and contained 1.0% O₂, 0.4% CO and 860 ppm NOₓ, Figure 6.8. With the reburn guns at position 1, the O₂ concentration was only slightly reduced at the end of the reburn zone to 0.5%. However, the NOₓ concentration was reduced across the entire reburn zone from 400 ppm at 40 cm horizontal position to 650 at the centerline. With tertiary air injected at pipe positions 1 and 2, the NOₓ emission was 420 ppm which was similar to the level at the reburn zone exit. With tertiary air injected at pipe positions 2 and 4, the NOₓ emission decreased to 300 ppm. However the burnout decreased from 98% to 96% with the later air addition.

With the reburn guns at position 2, peak CO, HCN and NH₃ levels coincided with the minimum NOₓ at 80 cm horizontal position. In the bulk of the forward flow between 40 and 60 cm horizontal position, the NOₓ concentration was 60-150 ppm higher with the reburn guns at position 2 than at position 1. Accordingly, the NOₓ emissions in the flue gas were approximately 70-90 ppm higher with the reburn guns at position 2 than at position 1.

Flame F5 was conducted at the same f_RB as flame F4, 20%, but with a higher λₚ of 1.1. The primary combustion products were oxygen rich with a gas composition of 2.5% O₂, 0.2% CO and 1000 ppm NOₓ, Figure 6.9. Fuel rich conditions could not be generated when the reburn fuel was added at either gun position and the O₂ concentration was only reduced to 0.5-2.0% at the end of the reburn zone. However, the NOₓ concentration was reduced across the entire reburn zone to 600-850 ppm. Despite the different N-species concentration profiles for the two flames, the NOₓ emissions were independent of reburn gun position. With tertiary air injected at pipe positions 1 and 2, the NOₓ emissions were 590 ppm and 630 ppm for reburn gun positions 1 and 2, respectively.

Flame F6 was conducted with the highest λₚ of 1.1 and the highest f_RB of 30% and it demonstrated the effect of reburn fuel fraction on the gas concentration in the reburn zone. With the reburn guns at position 1, the O₂ concentration was reduced from 3.5% at
Figure 6.8 Gas concentration profiles at the end of the reburn zone and data on the precombustor and flue gas emissions for Flame F4: $\lambda_p = 1.04$, $f_{RB} = 20\%$ and $\lambda_{RB} = 0.85$
Figure 6.9 Gas concentration profiles at the end of the reburn zone and data on the precombustor and flue gas emissions for Flame F5: $\lambda_p = 1.10$, $f_{RB} = 20\%$ and $\lambda_{RB} = 0.90$
the precombustor exit to 0.2% at the reburn zone exit, Figure 6.10. NOx was reduced to 400 ppm across the entire reburn zone. With the reburn guns at position 2, peak CO, HCN and NH₃ levels coincided with the minimum NOx at the furnace centerline. In the bulk of the forward flow between 40 and 60 cm horizontal position, the O₂ concentration was 1-1.5% and the NOx concentration was 550-600 ppm or 150-200 ppm higher with the reburn guns at position 2 than at position 1. Accordingly, the NOx emissions in the flue gas were approximately 90-140 ppm higher with the reburn guns at position 2 than at position 1.

Coal burnout was adversely affected by the high reburn fuel fraction. With tertiary air injected at pipe positions 2 and 4, the burnout was only 90% which was the lowest for all flames.

The data from these flames indicate that the effect of mixing on NOx reduction may be classified into two groups. For flames F1, F3 and F5, with λ_{RB} of 0.87-0.94, mixing had little effect on NOx reduction. However, for flames F2, F4 and F6, with λ_{RB} of 0.77-0.84, NOx emissions were 50-130 ppm lower at the higher mixing level (reburn gun position 1).

Differences in the influence of mixing for the two groups of flames could be partly related to the availability of O₂ in the reburn zone. Process conditions which resulted in O₂ concentration of less than 0.5% throughout the reburn zone had lower NOx emissions with the higher mixing. In flames F3 and F5, it was not possible to attain O₂ concentrations less than 0.5% with either reburn gun position. Thus, the mixing could not influence the reduction of NOx emissions. In flame F6 with the reburn guns at position 1, O₂ concentrations of 0.1-0.5% were measured across the reburn zone and lower NOx emissions. In contrast with the reburn guns at position 2, 0.8-1.4% O₂ was measured in the bulk of the forward flow.

Mixing could also be important for flames with low O₂ concentrations in the reburn zone. In flame F2, 0-0.5% O₂ was measured throughout the reburn zone for low and high mixing and NOx was reduced from 430 to 360 ppm with the higher mixing. However, in flame F1, 0-0.5% O₂ was measured throughout the reburn zone for low and high mixing but there was no effect of reburn fuel mixing on NOx reduction. The main differences between flames F1 and F2 were f_{RB} (10 and 20%, respectively) and λ_{RB} (0.87 and 0.78, respectively).
Figure 6.10 Gas concentration profiles at the end of the reburn zone and data on the precombustor and flue gas emissions for Flame F6: $\lambda_F = 1.10$, $f_{RB} = 30\%$ and $\lambda_{RB} = 0.79$
Thus improved NOx reduction with higher mixing could be attributed to at least two effects. Flames where the improved mixing lowered O\textsubscript{2} concentrations, also had reduced NOx emissions. In the case where the O\textsubscript{2} concentration was already low exiting the precombustor, improved mixing could lower NOx if there was 20% reburn fuel fraction.

6.2.3 Reburn Zone Stoichiometry

Primary stoichiometry, $\lambda_P$, and reburn fuel fraction, $f_{RB}$, had a strong influence on NOx reduction. In general, these two variables could be considered a single variable, the reburn zone stoichiometry $\lambda_{RB}$, and directly related to the NOx emissions. $\lambda_P$ was not a sensitive parameter as long as $f_{RB}$ was adjusted to obtain a given $\lambda_{RB}$. When tertiary air was injected at pipe positions 1 and 2, the NOx decreased from 1050 to 350 ppm as $\lambda_{RB}$ decreased from 1.1 to 0.77, Figure 6.11. The NOx emissions were steadily decreasing with $\lambda_{RB}$ suggesting that lower emissions may be obtained at lower stoichiometry.

![Figure 6.11 Effect of reburn zone stoichiometry on NOx emissions for low and high reburn fuel mixing.](image)

6.2.4 Delayed Tertiary Air Addition

The effect of tertiary air injection position on NOx emissions was studied for several arrangements with flame F7. The NOx emissions decreased from 800 ppm at the
precombustor exit, to 410 ppm with air injected at pipe positions 1 and 3, and to 280 ppm with air injected at pipe positions 3 and 4, Figure 6.12. In all cases, there was no effect of delaying the tertiary air injection on the CO emissions which were less than 10 ppm. However, the coal burnout decreased from 99% with air injected at positions 1 and 3 to 95% with tertiary air injected at positions 3 and 4, Figure 6.13. This was due to the shortening of the residence time available to burn out the char and lower flame temperatures closer to the chimney. The gas temperatures at the final tertiary air injection location were 1200°C and 1130°C for pipe positions 3 and 4, respectively, Figure 6.3. The average residence times between the final tertiary air addition location and the chimney were 3 s and 2 s for pipe positions 3 and 4, respectively.

The differences in NOx emissions due to delayed tertiary air addition were relatively small considering the wide range in reburn zone residence time and the temperature at the point of injection. The reburn zone residence time up to the first tertiary air injection position could be adjusted from 0.4 to 2.0 s, Figure 6.3.

Given the strong correlation of NOx emissions with $\lambda_{RB}$, it was possible that the delayed air addition may have lowered NOx emissions due to reduced tertiary air recirculation into the reburn zone. This would influence the effective reburn zone stoichiometry, $\lambda_{eff}$, calculated according to Equation 6.1. The NOx emissions are plotted as a function of $\lambda_{eff}$ for flames F1 to F7 in Figure 6.14. There was little distinction between tertiary air injection arrangements. The correlation of NOx emissions with $\lambda_{eff}$ suggests that extending the reburn zone beyond the second choke did not have a significant influence on NOx reduction in these experiments.

6.2.4 Natural Gas Reburning

Natural gas was used as the reburning fuel in flame F8 at the same conditions as flame F7. The $\lambda_p$ was set at 1.0 and $f_{RB}$ at 20%. The NOx concentration in the flue gas decreased from 290 ppm, with tertiary air injected at positions 1 and 2, to 160 ppm, with tertiary air injected at positions 3 and 4, Figure 6.12. The coal burnout decreased from 98%, with tertiary air injected at positions 1 and 2 to 96% with tertiary air injected at positions 3 and 4, Figure 6.13.

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Figure 6.12 Effect of tertiary air injection position on NOx emissions for coal reburning, flame F7, and natural gas reburning, flame F8.

Figure 6.13 Effect of tertiary air injection position on coal burnout for coal reburning, flame F7, and natural gas reburning, flame F8.
The NOx in the flue was consistently 100-120 ppm less than in flame F7 with coal as the reburn fuel. The lower emission could be attributed to better NOx reduction potential of natural gas or higher mixing of the reburn fuel with the primary products. The reduction potential of natural gas reburning fuel was probably higher due to its high volatliity and low N-species content and not due to improved mixing. If the mixing had been important, it would be expected that the difference between NOx emissions for natural gas and coal would become less as the tertiary air was delayed. However, this was not the case.

Due to its higher volatility, natural gas would tend to consume more O2 than the coal in the reburn zone resulting in lower $\lambda_{eff}$. For coal reburning, a difference in stoichiometry of approximately 0.05 could result in a 100 ppm difference in NOx emissions, Figure 6.14. As the volatility was 100% for natural gas compared to 60% for the coal, a reduction in $\lambda_{eff}$ of 0.05 due to the use of natural gas was possible.
When tertiary air was injected after tertiary pipe position 2, the coal burnout was consistently 1% higher when natural gas was used as the reburn fuel. These data show that incomplete burnout may be attributed to primary as well as reburn fuel. Thus, natural gas may be used to significantly reduce NOx emissions, although, coal burnout may remain a problem.

6.3 Comparison to Reburn Zone investigation

The Furnace Investigation involved mixing at only two levels: low and high mixing. However, the tests were able to show that mixing was significant for reducing NOx emissions at low reburn zone stoichiometry. To increase the comprehension of the relationship between mixing and NOx reduction, the data from the Furnace Investigation were compared with additional tests conducted on the IPFR where mixing rates were significantly faster (as discussed in Section 5.5, p.116).

The additional tests were conducted in the IPFR to simulate the conditions found in the reburn zone of the Furnace Investigation. Thus, tests were conducted at 1450°C and with a primary stoichiometry of 1.0 to 1.05, reburn fuel fraction of 10-30% and primary NOx concentration of 900 to 1200 ppm. NOx, HCN and NH3 concentrations were measured after 0.2-0.5 s in the reactor. In total, five tests were conducted with coal reburning and one with natural gas reburning.

To simulate the O2 concentration in the primary combustion products, an assumption was made relating \( \lambda_p \) to the O2 concentration. In the IPFR, primary combustion products were generated by burning natural gas and, because combustion was complete, \( \lambda_p \) could be directly related to the exit O2 concentration. However, in the precombustor of Furnace no. 1, O2 concentrations of approximately 1% were measured for \( \lambda_p \) of 0.95-1.0. Therefore, in the IPFR tests, the inlet O2 was set to obtain a primary stoichiometry of 1.0 or 1.05 and then an additional 1% O2 was added to account for incomplete O2 consumption. For calculations of the reburn zone stoichiometry, the additional 1% was neglected although the additional 1% O2 would increase \( \lambda_{RB} \) by 0.05. The gas composition, including HCN and NH3, was measured in the reactor after 0.2 to 0.5 s. The NOx and TVFN concentrations were corrected to 0% O2 by multiplying the measured concentration by \( \lambda_{RB} \).
The NOx, HCN and NH3 concentrations for the six tests are shown in Figure 6.15a-6.15f. At $\lambda_P$ of 1.0 and $f_{RB}$ of 10%, Figure 6.15a, the NOx concentration could be reduced from 1050 ppm to 320 ppm after 0.3 s with no further reduction measured up to 0.5 s. Only trace concentrations of HCN, 5 ppm, and NH3, 40 ppm, were measured in the reburn zone. When $f_{RB}$ was increased to 20%, Figure 6.15b, the NOx concentration was reduced to 70 ppm in 0.3 s. The HCN concentration was still low, however, NH3 increased from 280 ppm at 0.2 s to 360 ppm at 0.5 s and was the predominant final species. The TVFN concentration could not be reduced to less than 450 ppm.

At $\lambda_P$ of 1.05 and $f_{RB}$ of 10%, Figure 6.15c, the NOx concentration could only be reduced from 900 to 550 ppm. However, when $f_{RB}$ was increased to 20%, Figure 6.15d, the NOx concentration was reduced to 175 ppm at 0.3 s. At 0.5 s, the TVFN concentration was 380 ppm of which 250 ppm was NH3. Increasing the reburn fuel fraction to 30%, Figure 6.15e, lowered the NOx concentration to 90 ppm after 0.3 s, but raised the NH3 concentration to 310-410 ppm and the TVFN concentration to approximately 500 ppm.

Reburning with natural gas at $\lambda_P$ of 1.05 and $f_{RB}$ of 13%, Figure 6.15f, resulted in a rapid NOx reduction to 60 ppm after 0.3 s. There was again a large NH3 concentration of 240 ppm measured after 0.5 s, and thus, the TVFN concentration was 260 ppm after 0.5 s.

The NOx concentration was strongly dependent on $\lambda_{RB}$ in agreement with findings from the Reburn Zone Investigation. The NOx concentration decreased from 560 ppm to 25-60 ppm as $\lambda_{RB}$ decreased from 0.94 to 0.80, Figure 6.16. For each test, the NOx concentration remained constant between 0.2-0.5 s. TVFN was relatively insensitive to $\lambda_{RB}$ and $t_{RB}$, Figure 6.16, and varied from 240 to 550 ppm with a minimum measured at $\lambda_{RB}$ of 0.9. For residence time greater than 0.3 s, the TVFN concentration remained constant with a redistribution of N-species. Thus, the residence time between 0.2 and 0.5 s did not have a significant effect on the NOx and TVFN concentrations in the reburn zone of the IPFR.

The correlation of NOx emissions as a function of $\lambda_{eff}$, measured in the Furnace Experiments and the IPFR, is shown in Figure 6.16. The NOx emissions from the Furnace Experiments were much higher than in the IPFR particularly at $\lambda_{eff}$ less than
Figure 6.15 Influence of reburn zone process conditions on NOx reduction in the IPFR at high inlet NOx.
Figure 6.16 Comparison of NOx and TVFN concentrations in the IPFR with NOx emissions in the Furnace Experiments for coal and natural gas reburning.

0.9. However, the furnace staging NOx emissions were similar to the TVFN over the entire range in $\lambda_{eff}$ for both coal and natural gas burn fuels.

6.3.1 Mixing Rates

The mixing rate of reburn fuel with primary combustion products was significantly faster in the IPFR compared to Furnace No. 1. The characteristic times for mixing by convection, $L/U$, turbulent diffusion, $L^2\varepsilon/k^2$, and reaction, $k/\varepsilon$, are shown in Table 5.2 (p.118) for the computational modelling performed in the Reburn Zone Investigation. The convective mixing time in the venturi region was predicted to be short, on the order of 10 ms, due to rapid impingement of primary combustion products with reburn coal. The impingement generated a high level of turbulence and the reaction mixing time was estimated as 10 ms. This implied that the mixing times were of a similar order of magnitude to the characteristic times for nitrogen chemistry reactions [90].
Based on this comparison, it is postulated that unmixedness in the Furnace Experiments favoured early oxidation of HCN and NH₃ to NOₓ. In Furnace No. 1, the characteristic times for convective mixing and reaction were 1 to 2 orders of magnitude longer than in the IPFR. Under these conditions, a significant portion of the flow may not have had sufficient time, or the stoichiometry required, to reduce nitrogen containing species to N₂, and thus, they were oxidized to NOₓ. The low concentration of HCN and NH₃ species measured in the reburn zone from the Furnace Experiments supports this argument. As a result, the TVFN concentration measured in the IPFR provided a better estimate of the NOₓ concentration in the Furnace Investigation.

The implication of these results is that turbulent mixing in the reburn zone would need to be very fast in order to obtain low NOₓ concentrations. HCN and NH₃ species, measured as products in the Reburn Zone Investigation, could be rapidly reconverted to NOₓ under mixing limited conditions. This would result in the same TVFN concentration but with a higher fraction of NOₓ.

6.4 Conclusions of the Furnace Investigation

The effect of mixing on coal reburning was studied for reburn fuel fraction of 10-30% and primary stoichiometry of 0.95-1.1. Comparative tests were conducted with natural gas reburn fuel.

1. The effect of reburn fuel mixing on NOₓ emissions was sensitive to λRB. For λRB of 0.87-0.95, there was little influence of reburn fuel mixing since there was sufficient O₂ to rapidly oxidize N-species intermediates. At λRB of 0.75-0.84, the NOₓ emission was 50-130 ppm lower for the high mixing case. The NOₓ reduction was most sensitive to mixing level at a reburn fuel fraction of 30%. This was attributed to the large concentration of reburn fuel which could be utilized more effectively at the higher mixing level.

2. The reburn zone stoichiometry had a dominant influence on the NOₓ emissions which decreased from 1000 to 250 ppm as λRB was reduced from 1.1 to 0.75.
3. Delayed tertiary air addition lowered NOx emissions, however the effect was attributed to a decrease in tertiary air recirculation into the reburn zone. When this effect was considered, reburn zone residence time longer than 0.3 s did not appear to have a significant influence on NOx emissions.

4. Natural gas was shown to be a better reburn fuel than coal. The NOx emissions were approximately 100 ppm lower for natural gas reburn fuel.

5. Much lower NOx emissions were obtained in the IPFR at a given $\lambda_{\text{eff}}$. The more efficient NOx reduction in the IPFR was attributed to an order of magnitude increase in convective, turbulent diffusion and reaction mixing rates. It was concluded that NOx concentrations less than 100 ppm were not attained at the end of the reburn zone in Furnace no. 1 because of the unmixedness of the reburn fuel and primary combustion products for all test conditions. The TVFN concentrations measured in the IPFR provided a reasonable estimate of the NOx emissions in Furnace no. 1.
7. Proposed Burner Design

Various independent and conceptually integrated aspects of the literature review, Reburn Zone and Furnace Investigations were used to design a fuel staged burner and to identify operating guide-lines for the burner. Discrepancies noted in the literature concerning the influence of reburning process variables on NOx reduction were addressed in the Reburn Zone and Furnace Investigations and the optimum process conditions to operate a fuel staged burner were proposed.

The burner concept and its technical specifications are presented in Section 7.1. The preferred operational configuration of the fuel staged burner in a utility boiler is detailed in Section 7.2. The effect of using a fuel staged burner on the boiler steam cycle and the furnace availability are discussed, Section 7.2.

7.1 Burner Concept

A simple swirl stabilized burner was designed which operates based on the fuel staging process. The burner forms a primary flame with 70-80% of the thermal input. Primary coal is injected and mixed with sufficient swirling secondary air to result in complete combustion of the coal. The reburn fuel is injected through the middle of the burner and is intended to mix with primary combustion products downstream of the primary flame zone. To complete the combustion process, tertiary air is then added after the reburn zone to complete the combustion process. Since the reburn fuel is injected through the middle of the flame, the burner is designated as an Internally Fuel Staged Burner (IFSB).

7.1.1 Physical Specifications

The burner consists of a primary coal gun centered in a secondary air pipe which is attached to a quarl. The quarl is designed with a length to diameter ratio, L/a, of 1.0 and an expansion ratio, b/a, of 2.0 (see Figure 2.10, p.44, for a description of burner dimensions). The quarl is formed with zero inlet and outlet angles to confine the forward flow and prevent slagging inside the quarl and on the adjacent wall.
The primary coal gun consists of an annular fuel injection opening at the outer edge. Coal is injected through an annulus to enhance mixing with the secondary air. This is accomplished by minimizing the primary coal jet thickness and the separation distance between the primary coal and the secondary air. The primary coal gun is designed with a high blockage ratio of the secondary air pipe, $D_v/a$, to ensure recirculation at all secondary air swirl levels [51].

The reburn fuel is injected through the reburn gun centered in the primary coal gun. To limit the fraction of reburn fuel which may be entrained into the secondary air inside of the quarl, the separation distance between the reburn gun and secondary air is maximized. With the reburn gun positioned at the quarl throat, the separation distance is equal to the radius of the quarl throat, $a/2$, Figure 7.2. By inserting the reburn gun to the quarl exit, the separation distance is increased to a value equal to the throat diameter, $a$. There is little increase in separation distance if the reburn gun is inserted further since the secondary air flow does not expand outside of the quarl unless swirl is excessively high ($>2.0$) and wall jet type flames are formed [51].

A 2.5 MW, IFSB was designed for tests on Furnace No. 1, Figure 7.3. The coal gun and secondary air pipe diameters are 130 and 210 mm, respectively, representing a bluff body ratio of 0.6. The reburn gun is a 48 mm water cooled gun with a 25 mm inner
diameter. The gun may be inserted 0.2 m past the quarl throat, flush with the furnace front wall.

7.1.2 Operating Conditions

Primary coal is transported by air (2 kg air/kg coal) and injected through an annulus in the coal gun at a velocity of 17 m/s. The coal injection velocity is sufficient to prevent settling in the coal gun but minimized to ensure the coal is rapidly entrained by the
secondary air. Secondary air is injected through the annulus at a forward velocity of 30-40 m/s and a swirl number of 0.6-1.5. The coal/air mixture is rapidly heated by convection from recirculated hot combustion products in the IRZ. The primary zone stoichiometry, $\lambda_p$, is maintained at 0.95-1.05 which is adequate to combust the primary coal and limit the formation of NOx [63,64].

The reburn fuel is transported with sufficient air to prevent settling in the pipe (1 kg air/kg coal) and injected into the middle of the IRZ at a velocity of 30-50 m/s. By injecting the reburn fuel through the center of the burner, devolatilization occurs in the O$_2$ lean IRZ. CH$_4$ radicals, generated from the volatile matter, react with NOx in the IRZ to form intermediate N-species. With sufficiently high momentum, the reburn jet penetrates the downstream end of the IRZ [51]. The jet, containing CH$_4$ radicals, N-species and char, flows out of the downstream end of the IRZ where it then mixes with primary combustion products.
7.1.3 Operation under Optimum Process Conditions

The most important operating conditions necessary to obtain low NOx emissions were identified in the Reburn Zone and Furnace Investigations as:

- reburn zone temperature, \( T \),
- reburn zone stoichiometry, \( \lambda_{RB} \),
- reburn zone residence time, \( \tau_{RB} \),
- mixing rate of combustion products and reburn fuel.

The reburn zone temperature should be greater than 1400\(^\circ\)C to form a high concentration of CH\(_4\) radicals and to promote rapid reduction of NOx and TVFN. The temperature inside and downstream of the IRZ are the highest in the flame. Thus, with the reburn fuel injected through the IRZ, volatile matter is evolving in a high temperature flow and the concentration of CH\(_4\) radicals is maximized. The high temperature downstream of the IRZ is also favourable for the rapid reduction of TVFN before the introduction of tertiary air.

It was shown in the Furnace Investigation that NOx emissions decreased at low \( \lambda_{RB} \) since reoxidation of nitrogen containing intermediates to NOx was minimized. High mixing was also observed to lower NOx emissions because of the improved contact between primary NOx and CH\(_4\) radicals. The possibility of forming O\(_2\) rich regions in the reburn zone is minimized for the IPSB by aerodynamically separating the reburn fuel and primary combustion products until they mix inside and downstream of the IRZ. The turbulent energy in the flame is highest at the downstream end of the IRZ [78]. The high energy promotes rapid mixing of reburn fuel and primary combustion products.

As a result of the Furnace Investigation, it was proposed that the TVFN in the reburn zone should be minimized to obtain low NOx emissions. From the Reburn Zone Investigation, TVFN was shown to be reduced to less than 200 ppm only in flames where the reaction of NO and NH\(_2\) radicals occurred. This reaction was relatively slow, and residence time on the order of 1 s and \( \lambda_{RB} \) less than 0.85 were necessary. In the fuel staged burner, reburn fuel is injected directly after the primary flame and thus provides the longest possible reburn zone residence time before the injection of tertiary air.

Staged tertiary air injection was shown to have little impact on the NOx emissions in the Furnace Investigation. A measured NOx emission reduction was attributed to reduced
recirculation of air into the reburn zone. Without a method of physically separating combustion zones in a utility boiler, recirculation would be more difficult to prevent than in the Furnace Investigation. In the IFSB, the separation distance between the reburn and tertiary combustion zones is maximized which limits the detrimental effect of tertiary air recirculation.

7.2 Application in a Utility Boiler

7.2.1 General Requirements

Application of the IFSB in a utility boiler would be assessed in terms of its performance in a full scale furnace configuration under continuous duty. The following boiler operating criteria are considered before installing a new burner:

- maximum boiler availability
- maximum boiler efficiency
- minimum carbon in ash
- low CO and NOx emissions
- minimum maintenance requirements.

The boiler should be available to raise steam continuously, and therefore, a new burner should be able to function continuously during normal operation. This criteria is closely related to the maintenance requirements of the boiler. If the burner increases the requirement for scheduled and unscheduled maintenance, then the boiler availability would decrease. The efficiency of the boiler is calculated in terms of the quantity and quality of superheated steam generated and the sensible heat loss in the flue gas. To maintain the boiler efficiency, it is preferable that the high heat transfer rate in the firebox does not change when a new burner type is installed. The burner should also not increase the by-products of the combustion process, namely carbon in ash, CO and NOx emissions, due to the cost of disposal and the environmental impact.

7.2.2 Reburn Fuel Mixing

The influence of flame interactions due to multiple burner arrangements on the performance of the IFSB should be considered. In a 400 MW_e wall fired boiler, there
may be several 50-85 MW, burners located on opposite walls and arranged in rows. Each flame is stabilized within its quarl and is deflected upwards by the convective flow out of the boiler and the buoyancy of the hot gases. Depending on the boiler arrangement, the flow may be highly stratified, Figure 7.4. The flow from the lowest burner elevation penetrates furthest toward the middle of the boiler with decreasing penetration at higher elevation [91].

In a furnace with reburn fuel injected from a single elevation, it is difficult to mix reburn fuel with primary combustion products across the entire furnace section [42]. Because of the flow stratification, the reburn fuel jet may be deflected upwards unless sufficient momentum is imparted to the jet.

This problem should be reduced with the IFSB. Reburn fuel penetrating the IRZ could mix with primary combustion products throughout the firebox, Figure 7.4. If the reburn jet momentum is too high, then it would mix with the primary combustion products from a burner at a lower elevation.

7.2.3 Burner Operation

At present, the distribution of fuel and air to each burner has been difficult to set and maintain [91]. Burners may operate fuel lean or fuel rich and thus, adversely affect NOx reduction. Maldistribution of fuel and air may be overcome by increasing the reburn fuel injection velocity to improve mixing with primary combustion products. However, this option may require higher reburn fuel transport air pressure than is normally available. Air and fuel controllers may be installed on the boiler, however, these control systems tend to be costly and are difficult to install on older boilers.

The IFSB would be relatively insensitive to maldistribution of fuel and air since the reburn fuel is injected through the IRZ. The potential for early oxidation of CH₄ radicals due to fuel lean burner operation is limited and some NOx reduction could occur at \( \lambda_{RB} \) approaching 1.0.

By injecting the reburn fuel through the IRZ, some of the mixing energy is supplied by the turbulent kinetic energy of the swirling secondary air at the IRZ closure. Therefore, adequate mixing of reburn fuel and primary combustion products may be attained without high reburn fuel injection velocity.
Figure 7.4 General flow patterns in a wall fired utility boiler showing stratified flow.

7.2.4 Combustion Intensity

The burner is designed to operate with a high primary zone combustion intensity. This ensures that sufficient heat is generated in the firebox to maintain the steam flow. If too large a fraction of the heat is released when the flow is near the superheater section, then
the efficiency of the steam cycle will decrease. The superheat steam temperature may rise [92] and affect the rating of the boiler and the control of the steam turbine [93].

7.2.5 Deposit Formation and Corrosion

Operation of a boiler at substoichiometric conditions may aggravate deposit formation [94]. Coal ashes that have low melting points over a broad temperature range are difficult to remove from metal surfaces [93]. This is related to the oxidation state of the iron and sulphur in the slag. Strongly reducing conditions in the reburn zone favour lower oxidation states of the coal ash which lowers the slag viscosity and may cause severe slagging.

Substoichiometric conditions also accelerate corrosion [94]. In the presence of CO and H₂, the protective oxide layer on steel tube may react with Cl and S-containing flue gases thereby exposing the metal surface to attack by the corrosive gas components. Raask [94] noted that corrosion increased substantially with low O₂ and high CO concentrations in the gas above a metal surface.

The IFSB would limit the O₂ lean conditions along the boiler wall since all of the fuel is injected through the quarl and the primary flame is operated fuel lean.

7.3 Concluding Remarks

An internally fuel staged burner was designed based on an investigation of reburning process parameters and fundamental aspects of swirl burner aerodynamic and combustion phenomena. The design was evaluated in terms of boiler compatibility and performance and it is concluded that the burner has potential for application in utility boilers.
8. Fuel Staged Burner Tests

The 2.5 MW, IFSB, presented in Chapter 7, was extensively studied. The operating characteristics and the burner capabilities in operating with a low NOx emission were investigated by computational modelling and furnace experiments. Computational modelling was conducted to predict the flow, temperature and gas composition in the burner and to determine a suitable method of varying the reburn fuel mixing with the primary combustion products, Section 8.1. In the Furnace Investigation, mixing in the reburn zone was shown to have a strong influence on NOx emissions. Thus, mixing rates in the Burner Tests are compared with the rates obtained in the Mixing Characterization Tests (discussed in Chapter 5).

Experimental tests were conducted in Furnace No. 1 using the 2.5 MW, IFSB, Section 8.2. The tests concentrated on studying the influence of burner and furnace operating parameters on NOx emissions, Section 8.2. The NOx emissions are related to results from the Reburn Zone and Furnace Investigations at similar test conditions.

8.1 Computational Modelling

Computations were performed for the first case, Case 1, with the reburn gun positioned at the burner throat and $\lambda_p$ set at 1.0 and $f_{RB}$ set at 30%, Table 8.1. A fine grid, with 122 grid lines in the axial direction and 66 lines in the radial direction, was used in the computations to accurately model the flow in the quarl, around the chokes and at the point of tertiary air injection. The grid was refined in the quarl and in the region of tertiary air injection to the extent that a finer grid did not change the model predictions.

After converging Case 1 with the reburn gun positioned at the quarl throat, the reburn gun was then positioned flush with the furnace front wall at 0.0 m axial distance, Case 2. The solution rapidly reconverged (800 iterations) indicating that the reburn gun position did not have a major influence on the overall flow properties. The flame properties for the two Cases are described below.

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Table 8.1 Inlet Conditions for Burner Computational Modelling

<table>
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<th>Primary</th>
<th>Secondary</th>
<th>Reburn</th>
<th>Tertiary</th>
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<td></td>
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<td></td>
<td>90</td>
<td></td>
</tr>
<tr>
<td>air</td>
<td>400</td>
<td>1520</td>
<td>60</td>
<td>1200</td>
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<tr>
<td>Velocity (m/s)</td>
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<td>17</td>
<td>31</td>
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<td>12</td>
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<td>v</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>58</td>
</tr>
<tr>
<td>w</td>
<td>0</td>
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<td>0</td>
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<td>10</td>
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</tr>
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<td>Temperature (°C)</td>
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<td>70</td>
<td>350</td>
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<tr>
<td>Injector Position (m axial distance)</td>
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<td>-0.2</td>
<td>-0.2 or 0.0</td>
<td>2.3</td>
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8.1.1 Case 1

The contours of stream function for Case 1 are shown in Figure 8.1. The region up to the end of the second choke was called the flame zone since a luminous flame was observed throughout this region. As in the Furnace Investigation, the region downstream of the second choke was called the tertiary combustion zone. The 0% and 100% streamlines define the boundaries of the region of unrecirculated flow from the burner. The 100% streamline followed from the edge of the quarl and along the choke walls. After the second choke, the streamline expanded rapidly to the furnace wall due to the momentum of the tertiary air. The 0% streamline, outlining the boundary of the IRZ, expanded out of the quarl and returned to the centerline at a distance approximately equal to the quarl diameter. Outside of the forward flow, a strong external recirculation zone, equal to 1.5 times the total flow formed between the front wall and the first choke. In the burnout zone, the forward flow was rapidly deflected to the furnace walls by the momentum of the tertiary air. A large recirculation zone formed behind the tertiary air injector and extended to the chimney.

The temperature, O₂ and CO contours in the flame zone are shown in Figure 8.2. Gas temperature exceeded 1000°C at the end of the quarl and reached a peak of 1800°C on
the outside edge of the IRZ between the front wall and the first choke. At the end of the reburn zone, the temperature had decreased to 1600°C across the entire flame zone. The secondary air was rapidly consumed in the flame zone and the O₂ concentration was less than 2% by the end of the first choke and less than 1% by the end of the second choke. Because of the O₂ lean conditions, a high CO concentration was measured throughout the flame zone with a peak concentration of 9% in the quarl and approximately 1% throughout most of the flame zone. These values indicate that a stable flame was predicted.

The trajectories of 60 μm primary and reburn coal particles demonstrate the behaviour of the flow in the flame zone, Figure 8.3. To demonstrate the influence of gas phase turbulence on particle trajectories, five primary and five reburn coal particles are shown. The primary coal particles mixed rapidly with the secondary air inside of the quarl and flowed along the boundary of the IRZ until the first choke, Figure 8.3 top. After the first choke, the primary coal particles started to disperse throughout the forward flow. The reburn coal particles easily penetrated into the front of the IRZ, Figure 8.3 bottom. However, the reburn jet had insufficient forward momentum to displace the internal recirculation zone from the centerline, and thus, the 60 μm particles were deflected radially outward. Only the 250 μm particles could penetrate through the IRZ but they represented only 2% of the reburn fuel mass fraction and did not have a strong influence on the flame properties.
Figure 8.2 Predicted temperature, O$_2$ and CO concentration profiles in the flame zone for Case 1.
The residence time in the reburn and tertiary combustion zones was estimated using the particle tracks. A numerical sampling probe was inserted in the computational grid behind the second choke at 2.0 m axial distance and between the 0% and 100% streamlines. For both primary and reburn coal particles, the average particle residence time was 0.6 s with a large scatter from 0.3 to 1.2 s, depending on the individual particle track. The particle size had little influence on the average residence time of the particles collected.

To determine the residence time in the tertiary combustion zone, twenty 40 µm particles were injected by computer simulation at the front of the tertiary combustion zone
(2.2 m axial distance: 0 cm horizontal position) and collected in the chimney. The average residence time was 2 s. Particles that were entrained into the large internal recirculation zone behind the tertiary air pipe had residence times on the order of 6 s. Thus, there could be a wide distribution of residence times in the tertiary combustion zone.

8.1.2 Case 2

With the reburn gun inserted 0.2 m into the quarl, the flow pattern in the flame zone differed from that calculated for Case 1, Figure 8.4. The 0% streamline was located downstream of the quarl up to the end of the first choke but had been displaced from the furnace centerline by the momentum of the reburn jet. The 100% streamline followed along the choke walls with a strong external recirculation zone between the front wall and the first choke.

The calculated temperature, O₂ and CO composition fields are shown in Figure 8.5. Gas temperature exceeded 1000°C at the end of the quarl and reached a peak of 1800°C outside of the IRZ. At the end of the reburn zone, the temperature had decreased to 1600°C across the entire flame zone. The peak O₂ concentration decreased to 4% by the front of the first choke and 1% at the front of the second choke. CO was predicted throughout the forward flow, with a concentration decreasing from 9% in the quarl to 0.5% at the outside of the forward flow around 50 cm horizontal position.

![Diagram of flame zone and tertiary combustion zone](image)

*Figure 8.4 Predicted flow streamlines in the flame zone for Case 2.*
Particle trajectories for five tracks of the primary and reburn fuel differed from those calculated in Case 1, Figure 8.6. Primary coal was entrained into the secondary air inside of the quarl and flowed around the boundary of the IRZ. There was also a tendency for some of the primary coal particles to follow along the reburn gun where they were entrained into the reburn jet. The entrainment was predicted for all particle sizes and was attributed to a low velocity formed around the side of the reburn gun. The reburn coal penetrated through the IRZ, Figure 8.6. Little spread in the jet was predicted up to the end of the first choke, and then, the particles dispersed rapidly, some entrained in the IRZ and some flowing directly into the tertiary combustion zone. However, the radial dispersion was not sufficient to fill the forward flow and the reburn fuel tended to remain near the centerline.

8.1.3 Comparison to Reburn Zone and Furnace Investigations

To determine whether the IFSB could be operated under conditions sufficient to produce low NOx emissions, the calculated flame zone properties were compared with the optimum conditions observed in the Reburn Zone and Furnace Investigations. For both Case 1 and Case 2, O2 lean conditions were calculated throughout the flame zone after the first choke. This was considered necessary in the Furnace Investigation to prevent reoxidation of HCN and NH3 to NOx in the reburn zone, Section 6.3 (p.143). The predicted flame zone temperature was in the range 1400-1700°C which was in excess of the temperature range studied in the Reburn Zone Investigation, but was similar to the reburn zone temperature studied in the Furnace Investigation. The high temperature generally promotes the formation of CH radicals to reduce NOx and enhance N-species reduction rates. In the Reburn Zone Investigation, a residence times of 0.3 s was considered to be sufficiently long to promote rapid NOx reduction at temperature greater than 1400°C. From the calculated particle residence times in the IFSB, there is sufficient residence time to reduce NOx before tertiary air injection. Thus, it was possible to operate the IFSB under the optimum conditions for temperature, stoichiometry and residence time.

An important parameter for NOx reduction, identified in the Furnace Investigation, was the mixing between reburn fuel and primary combustion products. Mixing needed to be sufficiently fast and complete to form O2 lean conditions throughout the reburn zone and to prevent reoxidation of HCN and NH3 to NOx. The mixing rates in the IFSB were estimated using the dimensional analysis described in Section 5.5 (p.116).
Figure 8.5 Predicted temperature, \( O_2 \) and CO concentration profiles in the flame zone for Case 2.
Characteristic times were calculated for convective, \( L/U \), diffusive, \( L^2 \epsilon/k^2 \), and reaction, \( k/\epsilon \), mixing in the flame zone for both computed cases. The characteristic length, \( L \), was estimated as the radius of the quarl exit (0.2 m) since this was equal to the radial separation distance between the reburn fuel and the primary combustion products (see Figure 7.2 p.151). Values for the absolute velocity, \( U \), kinetic energy, \( k \), and dissipation, \( \epsilon \), were calculated in the computational modelling.

The mixing times in the flame zone were similar for both flames and are shown for Case 2 in Figure 8.7. For Case 1, the reburn fuel mixed with the primary combustion
Figure 8.7 Predicted characteristic convective, diffusive and reaction mixing times in the flame zone of the IFSB, Case 2.
products just downstream and at the outer edge of the quarl. At that position, the characteristic convective, diffusive and reaction mixing times were 0.05, 0.04 and 0.04 s, respectively. For Case 2, the reburn fuel and primary combustion products mixed downstream of the IRZ at 1 m axial distance. At that position, the characteristic convective, diffusive and reaction mixing times were 0.05, 0.04 and 0.08 s, respectively.

These values suggest that the reburn fuel was mixed with the primary combustion products in times approaching those estimated for the Reburn Zone Investigation. Therefore, it may be concluded that the mixing was sufficiently fast in the IFSB to promote NOx reduction.

8.2 Experimental Results

The 2.5 MW, IFSB was tested in Furnace no. 1 over a wide range of process conditions for six flames with coal reburning and three flames with natural gas reburning. The coal reburning tests are designated FSB1-BSB6 and the natural gas reburning tests are designated FSB7-FSB9. \(\lambda_F\) was set between 0.95 and 1.1 and \(f_{RB}\) in the range 20-30% resulting in a reburn zone stoichiometry, \(\lambda_{RB}\), of 0.72 to 0.86. For most of the flames, secondary air swirl level, reburn gun and tertiary air injector positions were varied. Test conditions and measured emissions are summarized in Appendix C.

8.2.1 Helium recirculation tests

In the Furnace Investigation, it was concluded that tertiary air recirculation into the reburn zone influenced NOx emissions. Therefore, to determine the fraction of tertiary air which could be recirculated into the flame zone, helium tracer measurements were made with the IFSB. Helium (He) tracer was injected with tertiary air at pipe positions 2 and 4 (see Figure 3.7 p.68) at a total concentration of 1.3%. Gas samples were collected before the second choke at 1.4 m axial distance and 40-100 cm horizontal position. To determine the influence of secondary air swirl level on tertiary air recirculation, tracer measurements were performed with the swirl set at 0.7 and 1.2.

When swirl level was 0.7 and tertiary air injected at pipe position 2, recirculation increased from 12% along the centerline to 35% at 40 cm horizontal position, Figure 8.8. With tertiary air injected at pipe position 4, the fraction of helium
recirculated increased from 8% along the centerline to 17% at 40 cm horizontal position. When the secondary air swirl level was increased to 1.2, only 3–8% of the helium was recirculated from pipe position 2 and no helium was recirculated from pipe position 4.

For swirl level at 0.7, the fraction of recirculation was comparable to that found in the Furnace Investigation (see Figure 6.2 top on p.123) with tertiary air recirculated from the outside of the forward flow into the flame zone. However, tertiary air recirculation could be reduced by increasing the secondary air swirl level to 1.2. The higher swirl level increased the tangential momentum of the primary combustion products and the pressure in the region between the chokes, thereby reducing the recirculation.

As a result of the helium tracer measurements, most burner tests were conducted with a swirl level of 1.2 for which recirculation was low. However, adjustment of swirl level provided a convenient method of determining the effect of tertiary air recirculation on the NOx emissions from the IFSB.

![Figure 8.8 Helium recirculation into the flame zone measured upstream of the second choke.](image-url)
8.2.2 Coal reburning

For all flames studied, there was a characteristic flame shape described by the luminous flame zones. The flame structure without reburn fuel may be seen in Photograph 6 which shows the primary flame seated inside of the burner quarl. The flame was compact with a luminous length approximately equal to the quarl diameter. When reburn coal was added, the flame changed to a dull luminous hue, Photograph 7, which extended up to the position of first tertiary air injection.

The behaviour of the reburn jet in the flow could be varied by adjusting the position of the reburn gun. With the reburn gun positioned at the quarl throat, the black reburn coal could be seen penetrating into the IRZ but was not observed exiting from the downstream end. With the reburn gun positioned flush with the furnace front wall (20 cm insertion), the reburn fuel jet was clearly visible penetrating the downstream end of the IRZ. The observed penetration verified the computational modelling predictions which indicated that the reburn jet trajectory was altered by inserting the reburn gun into the quarl.

Flame FSB3 was performed at a primary stoichiometry, $\lambda_P$, of 1.0 and a reburn fuel fraction, $f_{RB}$, of 20%, Table 8.2. With tertiary air injected at positions 2 and 3, there was little effect of swirl level (in the range 0.7 to 1.2) or reburn gun position (0 to 1 quarl diameter) on emissions. The NOx and CO emissions were in the range 320-370 ppm and 0-3 ppm, respectively, for these cases. The position of tertiary air injection did have an influence on NOx emissions which decreased from 440 ppm with air injected at positions 1 and 3 to 320 ppm with tertiary air injected at positions 2 and 3. For a single measurement, coal burnout was 98%.

For flame FSB2, $\lambda_P$ was decreased to 0.95 and $f_{RB}$ was maintained at 20%. Despite the lower $\lambda_{RB}$ compared to flame FSB3, the NOx emissions were unchanged at 330-350 ppm, Table 8.2. As was observed for flame FSB3, reburn gun position had little influence on NOx emissions.

Flame FSB4 was performed with similar process conditions to flame FSB3 but with $f_{RB}$ increased to 30%. With tertiary air injected at pipe positions 2 and 3, the NOx emissions were 240-260 ppm. The NOx emissions decreased substantially to 150 ppm when the tertiary air was injected at positions 2 and 4. CO emissions were not detected in these tests and the burnout was in the range 98-99%.
<table>
<thead>
<tr>
<th>Swirl number</th>
<th>reburn gun position (quartil dia.)</th>
<th>tert. air pipe pos.</th>
<th>(O_2) (ppm)</th>
<th>CO (ppm)</th>
<th>NOx (ppm 0% (O_2))</th>
<th>burnout (wt %)</th>
<th>flue gas temp (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flame FSB1: (\lambda_p = 1.1, f_{RB} = 30%, \lambda_{RB} = 0.80)</td>
<td>1.2</td>
<td>1</td>
<td>2.3</td>
<td>2.7</td>
<td>0</td>
<td>280</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>1</td>
<td>2.4</td>
<td>2.9</td>
<td>0</td>
<td>200</td>
<td>97.7</td>
</tr>
<tr>
<td>Flame FSB2: (\lambda_p = 0.95, f_{RB} = 20%, \lambda_{RB} = 0.75)</td>
<td>0.7</td>
<td>0</td>
<td>2.3</td>
<td>3.1</td>
<td>0</td>
<td>350</td>
<td>1010</td>
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<tr>
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<td>1.2</td>
<td>0</td>
<td>2.3</td>
<td>2.9</td>
<td>0</td>
<td>330</td>
<td>1060</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>0.5</td>
<td>2.3</td>
<td>2.7</td>
<td>0</td>
<td>340</td>
<td>1060</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>1</td>
<td>2.3</td>
<td>2.8</td>
<td>0</td>
<td>330</td>
<td>98.7</td>
</tr>
<tr>
<td>Flame FSB3: (\lambda_p = 1.0, f_{RB} = 20%, \lambda_{RB} = 0.80)</td>
<td>0.7</td>
<td>0</td>
<td>2.3</td>
<td>3.5</td>
<td>1</td>
<td>360</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>0.7</td>
<td>0.25</td>
<td>2.3</td>
<td>3.2</td>
<td>3</td>
<td>350</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>0.7</td>
<td>0.5</td>
<td>2.3</td>
<td>3.3</td>
<td>2</td>
<td>370</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>0.7</td>
<td>1</td>
<td>1.2</td>
<td>2.8</td>
<td>3</td>
<td>410</td>
<td>97.7</td>
</tr>
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<td></td>
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<td>1</td>
<td>1.3</td>
<td>2.9</td>
<td>2</td>
<td>440</td>
<td>97.7</td>
</tr>
<tr>
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<td>1</td>
<td>1.4</td>
<td>3.0</td>
<td>3</td>
<td>390</td>
<td>97.7</td>
</tr>
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<td></td>
<td>0.7</td>
<td>1</td>
<td>2.3</td>
<td>3.2</td>
<td>3</td>
<td>320</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>0.7</td>
<td>1</td>
<td>2.4</td>
<td>3.2</td>
<td>1</td>
<td>340</td>
<td>97.7</td>
</tr>
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<td></td>
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<td>1</td>
<td>2.3</td>
<td>3.6</td>
<td>0</td>
<td>370</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
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<td>1</td>
<td>2.3</td>
<td>3.5</td>
<td>0</td>
<td>350</td>
<td>97.7</td>
</tr>
<tr>
<td>Flame FSB4: (\lambda_p = 1.0, f_{RB} = 30%, \lambda_{RB} = 0.70)</td>
<td>0.7</td>
<td>1</td>
<td>2.3</td>
<td>2.5</td>
<td>0</td>
<td>270</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>0.5</td>
<td>2.3</td>
<td>3.1</td>
<td>0</td>
<td>260</td>
<td>97.7</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>1</td>
<td>1.2</td>
<td>2.5</td>
<td>1</td>
<td>260</td>
<td>98.2</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>1</td>
<td>2.3</td>
<td>2.7</td>
<td>0</td>
<td>240</td>
<td>98.8</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>1</td>
<td>2.4</td>
<td>2.8</td>
<td>1</td>
<td>150</td>
<td>97.6</td>
</tr>
<tr>
<td>Flame FSB5: (\lambda_p = 1.07, f_{RB} = 20%, \lambda_{RB} = 0.87)</td>
<td>0.7</td>
<td>1</td>
<td>2.3</td>
<td>3.2</td>
<td>3</td>
<td>500</td>
<td>97.7</td>
</tr>
<tr>
<td>Flame FSB6: (\lambda_p = 1.05, f_{RB} = 30%, \lambda_{RB} = 0.75)</td>
<td>0.7</td>
<td>1</td>
<td>2.3</td>
<td>3.0</td>
<td>1</td>
<td>340</td>
<td>98.0</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>0</td>
<td>2.3</td>
<td>3.3</td>
<td>0</td>
<td>300</td>
<td>98.0</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>1</td>
<td>2.3</td>
<td>3.0</td>
<td>0</td>
<td>250</td>
<td>98.5</td>
</tr>
</tbody>
</table>
Flames FSB6 and FSB1 were also performed with $f_{RB}$ of 30% and $\lambda_{RB}$ of 1.05 and 1.1, respectively. With swirl number of 1.2 and the reburn gun positioned at 1 quart diameter, the NOx emissions were 250 ppm and 280 ppm for FSB6 and FSB1, respectively.

In general, the gun position and swirl level had little effect on the NOx emissions from the IPSB, Figure 8.9. This observation was interesting in that the gun position was observed to have a substantial influence on the reburn particle trajectories, and the swirl level affected the tertiary air recirculation into the reburn zone. Thus it was concluded that NOx emissions from the IPSB were not sensitive to reburn fuel trajectories or tertiary air recirculation. This was probably due to the rapid mixing of reburn fuel and primary combustion products downstream of the quart for all reburn gun positions as discussed in Section 8.1.

The effect of $\lambda_P$ and $f_{RB}$ on NOx emissions for the IPSB is shown in Figure 8.10. At $f_{RB}$ of 20%, the NOx emissions decreased from 500 ppm at $\lambda_P$ of 1.07 to 330 ppm at $\lambda_P$ of 0.95. At $f_{RB}$ of 30%, there was little influence of $\lambda_P$ on NOx emissions which were in the range 240-280 ppm.

In the Furnace Investigation, it was concluded that the effects of $\lambda_P$ and $f_{RB}$ could be combined and expressed in terms of the reburn zone stoichiometry, $f_{RB}$. This was not the case using the IPSB as demonstrated by the NOx emission curves in Figure 8.11. At a given $\lambda_{RB}$, NOx emissions were 70 ppm lower when $f_{RB}$ was increased from 20% to 30%. Coal burnout and CO emissions were not adversely affected by operating with a high reburn fuel fraction. Thus, there was a significant advantage in operating the IPSB with a high reburn fuel fraction.

The influence of delayed tertiary air injection on NOx emissions was similar for flames FSB1, FSB3 and FSB4, Figure 8.12. The NOx emissions decreased by approximately 50 ppm when the first tertiary air injection was moved from position 1 to position 2. NOx emissions decreased by a further 80 ppm when the final tertiary air injection was moved from position 3 to position 4. Thus, two stages of air injection had a positive influence on NOx reduction with the IPSB.
Figure 8.9 Influence of reburn gun position on NOx emissions in the IFSB

Figure 8.10 Effect of primary stoichiometry, $\lambda_p$, and reburn fuel fraction, $f_{RB}$, on NOx emissions from the IFSB
Figure 8.11 Influence of reburn zone stoichiometry, $\lambda_{RB}$, on NOx emissions from the IFSB for coal and natural gas reburning.

Figure 8.12 Influence of tertiary air injection position on NOx emissions from the IFSB.
8.2.3 Natural gas reburning

Flames FSB7-FSB9 were created using natural gas reburn fuel. The complete flame conditions are listed in Appendix C and the emissions are summarized in Table 8.3. Two types of gas injectors were tested. A water-cooled, radial injector was used to inject reburn gas radially outward from the burner centerline at 0.5, 1.0 and 2.0 m axial distance. An axial injector was used to inject natural gas along the burner axis in a similar way as the reburn coal jet. The secondary air swirl level was maintained at 1.2 for the tests with natural gas reburning.

<table>
<thead>
<tr>
<th>Swirl number</th>
<th>reburn gun position (quarl. dia.)</th>
<th>tert. air pipe pos.</th>
<th>O_2 (ppm)</th>
<th>CO (ppm)</th>
<th>NOx (ppm 0% O_2)</th>
<th>burnout (wt %)</th>
<th>flue gas temp (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flame FSB7: ( \lambda_p = 1.15 ), ( f_{RB} = 30% ), ( \lambda_{RB} = 0.85 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.2</td>
<td>2.5</td>
<td>2,3</td>
<td>2.7</td>
<td>3</td>
<td>330</td>
<td>250</td>
<td>1090</td>
</tr>
<tr>
<td>1.2</td>
<td>3.7</td>
<td>2,3</td>
<td>2.4</td>
<td>1</td>
<td>270</td>
<td>270</td>
<td>1090</td>
</tr>
<tr>
<td>1.2</td>
<td>5.0</td>
<td>2,3</td>
<td>2.4</td>
<td>2</td>
<td>100</td>
<td>100</td>
<td>1090</td>
</tr>
<tr>
<td>Flame FSB8: ( \lambda_p = 1.0 ), ( f_{RB} = 30% ), ( \lambda_{RB} = 0.70 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.2</td>
<td>3.7R</td>
<td>2,3</td>
<td>2.1</td>
<td>10</td>
<td>140</td>
<td>98.7</td>
<td>1100</td>
</tr>
<tr>
<td>1.2</td>
<td>3.7R</td>
<td>2,4</td>
<td>3.5</td>
<td>1</td>
<td>110</td>
<td>97.6</td>
<td>1100</td>
</tr>
<tr>
<td>1.2</td>
<td>1.0A</td>
<td>2,4</td>
<td>2.2</td>
<td>0</td>
<td>100</td>
<td>100</td>
<td>1100</td>
</tr>
<tr>
<td>Flame FSB9: ( \lambda_p = 1.0 ), ( f_{RB} = 20% ), ( \lambda_{RB} = 0.80 )</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1.2</td>
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<td>2,4</td>
<td>2.3</td>
<td>0</td>
<td>200</td>
<td>98.0</td>
<td>1100</td>
</tr>
</tbody>
</table>

Flame FSB9 was performed with \( \lambda_p \) of 1.0 and \( f_{RB} \) of 20\%. The NOx emissions were in the range 170-220 ppm. There was no significant effect of gas injection arrangement or injector position on the NOx emissions. As well, CO emissions were low, in the range 0-13 ppm, and coal burnout was 98%.

The \( f_{RB} \) was increased to 30\% for flame FSB8. The NOx emissions decreased by approximately 100 ppm to 100-140 ppm. It was difficult to discern the effect of natural
gas injector arrangement because of the limited number of tests and the low NOx values. However, with tertiary air injected at pipe positions 2 and 4, the NOx emissions were 100 and 110 ppm for axial and radial reburn gas injection, which was not considered a significant difference.

The $\lambda_F$ was increased to 1.15 for flame FSB7 resulting in a reburn zone stoichiometry of 0.85. The NOx emissions increased substantially to 250-330 ppm. Thus, $\lambda_{RB}$ had a significant effect on NOx emissions for gas reburning in the IFSB.

NOx emissions were 70-100 ppm lower than measured for coal reburning with $f_{RB}$ of 30%, Figure 8.11. The lower NOx emissions were probably due to the reduced fuel-N content when a fraction of the fuel was natural gas.

8.2.4 Comparison to the Furnace Investigation

The influence of reburn zone stoichiometry, $\lambda_{RB}$, on NOx emissions was similar for the Furnace Investigation and Burner Tests, Figure 8.13. The NOx emissions lay on a single curve decreasing from 1000 ppm at $\lambda_{RB}$ of 1.1 to 250 ppm at $\lambda_{RB}$ of 0.72. The coal burnout for the two experiments was also equal and lay in the range 97.5-99%, Figure 8.13.

![Figure 8.13](image_url) **Figure 8.13** Comparison of NOx emissions and coal burnout from the IFSB and the Furnace Investigation.
The NOx emissions for the IFSB and Furnace Investigation were similar despite large differences in heat extraction. With the IFSB, there was 450 kW less heat extraction in the first 2 m due to the absence of the precombustor. The temperature at the reburn zone exit was calculated to be 200°C hotter than obtained in the Furnace Investigation. Temperature differences in the reburn zone and at the point of tertiary air injection may have an influence on NOx reduction and formation. Higher temperature in the reburn zone would tend to promote rapid reduction whereas higher temperature at the point of tertiary air injection would tend to result in higher NOx. Either these processes balanced each other in the flames studied or the effect of temperature was much less significant than the effect of $\lambda_{RB}$.

8.3 Concluding Remarks

The flow characteristics and combustion performance of an internally fuel staged burner (IFSB) was tested. Computational flame modelling demonstrated that a reburn fuel jet could penetrate into or through the internal recirculation zone, thus allowing the reburn fuel to rapidly mix with primary combustion products. In the experimental tests with coal reburn fuel, NOx emissions could be reduced to 150-250 ppm by injecting 30% of the fuel through the reburn gun. With natural gas reburning, the NOx emissions could be reduced to 100 ppm using 30% reburn fuel. The low sensitivity to operating conditions demonstrated that burner settings and process conditions do not need to be precisely controlled in order to obtain low NOx and CO emissions.
9. Conclusions and Recommendations

A comprehensive study was conducted to determine the influence of fuel staging process parameters on NOx and TVFN reduction. The process parameters were studied in two investigations. The Reburn Zone Investigation was conducted to determine the influence of process parameters on NOx and TVFN reduction in the reburn zone. Mixing of reburn fuel and primary zone NOx was maximized in order to focus on the chemical process. Mixing limitations were introduced in the Furnace Investigation to extend the findings to conditions representative of industrial furnaces.

The results of these two investigations lead to the design of a novel internally fuel staged burner, IFSB. A 2.5 MWt IFSB was tested and, as proposed in the design, resulted in low NOx emissions. The conclusions from the Reburn Zone and Furnace Investigations and the testing of the IFSB are presented below.

9.1 Reburn Zone Investigation

The parameters which had the greatest influence on NOx reduction were the reburn zone temperature, stoichiometry, and residence time. Reburn coal type, particle size distribution and the inlet NOx concentration to the reburn zone were found to have a lesser influence on NOx reduction.

NOx reduction was substantially enhanced by operating the reburn zone at high temperature. At 1100°C, the NOx concentration was reduced from 400 ppm to a minimum of 260 ppm. At 1400°C, the NOx concentration could be reduced from 400 ppm to 30 ppm.

NOx reduction was also enhanced by operating the reburn zone at a low stoichiometry. At 1400°C, the NOx concentration exiting the reburn zone decreased from 350 to 50 ppm as the reburn zone stoichiometry decreased from 1.04 to 0.8. However, the TVFN concentration was independent of the reburn zone stoichiometry below 0.9. Thus, NOx was reduced to HCN and NH₃ at lower stoichiometry.
Reburn zone residence time greater than 0.3 s was necessary to obtain NO\textsubscript{x} concentrations less than 100 ppm. However, the residence time necessary to obtain a given NO\textsubscript{x} reduction was dependent on the temperature and the reburn zone stoichiometry. Less time was necessary to reduce the NO\textsubscript{x} at a higher temperature and lower reburn zone stoichiometry. The NO\textsubscript{x} and HCN reduction occurring in the reburn zone after 0.05 s could be accurately predicted using a model for NO\textsubscript{x} reduction by NH\textsubscript{3} radicals. This suggested that reactions between CH\textsubscript{3} radicals and NO\textsubscript{x} were complete before 0.05 s.

9.2 Furnace Investigation

The effect of the mixing between reburn fuel and primary combustion products was considered in the Furnace Investigation. NO\textsubscript{x} emissions were measured for two levels of mixing. At low mixing, the reburn fuel flowed through the reburn zone without completely mixing with the primary combustion products. At high mixing, the reburn fuel was well dispersed in the primary combustion products.

The reburn fuel fraction and primary stoichiometry had a dominant influence on the NO\textsubscript{x} emissions. It was concluded that these two variables were dependent and could be expressed in terms of the reburn zone stoichiometry. The NO\textsubscript{x} emissions decreased from 1000 to 250 ppm as reburn zone stoichiometry was reduced from 1.1 to 0.75.

NO\textsubscript{x} emissions were only sensitive to mixing level at low reburn zone stoichiometry. For reburn zone stoichiometry of 0.87-0.95, there was little influence of reburn fuel mixing on NO\textsubscript{x} emissions and for reburn zone stoichiometry of 0.75-0.84, the NO\textsubscript{x} emissions were 50-130 ppm lower for the high mixing case. The improved NO\textsubscript{x} reduction with higher mixing was attributed to two effects. Lower NO\textsubscript{x} emissions were measured in flames where the higher mixing reduced the O\textsubscript{2} concentration throughout the reburn zone. In cases where O\textsubscript{2} concentration was low at both mixing levels, higher mixing promoted more complete NO\textsubscript{x} reduction in the reburn zone.

Natural gas was shown to be a better reburn fuel than coal. The NO\textsubscript{x} emissions were approximately 100 ppm lower for natural gas reburn fuel over a wide range of conditions. The better performance of natural gas was attributed to its higher volatility and lower nitrogen content.
A comparison was made between NOx emissions in the Furnace Investigation and the NOx and TVFN concentrations measured in the Reburn Zone Investigation. Much lower NOx emissions, obtained in the Reburn Zone Investigation, were attributed to a 1-2 order of magnitude difference in convective, turbulent diffusion and reaction mixing times. It was concluded that the NOx concentration at the end of the rebum zone could not be reduced to less than 200 ppm because the mixing rate of rebum fuel and primary combustion products was an order of magnitude slower than characteristic nitrogen chemistry reaction times. Thus, intermediate CH₄, HCN and NH₃, could be oxidized before the mixing process was completed.

It was not certain that improved mixing would have lowered NOx emissions. The TVFN concentrations measured in the Reburn Zone Investigation, provided a reasonable estimate of the NOx emissions in the Furnace Investigation. Since the TVFN concentration was insensitive to rebum zone operating conditions, it may have been difficult to obtain further NOx emission reduction.

9.3 Burner Tests

The results of the Reburn Zone and Furnace Investigations were applied in designing a fuel staged burner for pulverized coal. Reburn fuel was injected through the internal recirculation zone to mix with primary combustion products further downstream. The burner could be operated at the optimum process conditions for rebum zone temperature, stoichiometry, residence time and mixing.

The NOx emissions were strongly dependent on rebum zone stoichiometry and decreased from 300 to 150 ppm as rebum zone stoichiometry decreased from 0.82 to 0.72. Burner operating conditions, including rebum jet penetration into the IRZ and secondary air swirl level, had little influence on the NOx emissions. Thus, the burner did not need to be carefully controlled in order to obtain low NOx emissions.

Natural gas was shown to be a more effective rebum fuel with the fuel staged burner. NOx emissions decreased from 220 to 100 ppm as rebum zone stoichiometry decreased from 0.85 to 0.7. The method of gas injection was not important and similar NOx emissions could be attained with rebum fuel injected axially in the quarl or radially at the end of the IRZ.
At a given reburn zone stoichiometry, the NOx emissions in the IFSB and Furnace Investigation were similar. This similarity demonstrated that the mixing in the IFSB was relatively fast and that reburn zone stoichiometry had a dominant influence on NOx emissions.

The NOx emissions from the IFSB were low compared to existing commercial coal-fired burners. The IFSB may be operated over a wide range of process conditions without adversely affecting NOx and CO emissions and coal burnout.

9.4 Further Development

The IFSB has demonstrated significant positive potential for operating with low NOx emissions. However, additional tests are recommended to provide information on improvements to the design and to allow scaling criteria to be established:

1. Computational modelling of the IFSB demonstrated that the reburn fuel could penetrate through the IRZ to mix with primary combustion products. Measurements of temperature, gas composition and coal burnout in the flame are recommended to verify the model predictions.

2. Lower NOx emissions were measured using natural gas as the reburn fuel than with coal as the reburn fuel. This may have been due to the higher volatility of natural gas which lowered the effective stoichiometry in the flame zone, or to the lower nitrogen content which reduced the quantity of fuel nitrogen in the furnace. Tests with ammonia doped natural gas reburn fuel are recommended to investigate the reason for low NOx emissions with natural gas reburn fuel.

3. Only trace concentrations of CO were measured in the flue gas despite low NOx emissions. The efficient combustion of CO was attributed to the high mixing and temperature in the tertiary combustion zone at the point of air injection. Further tests with lower mixing rates between flame zone products and tertiary air are recommended to determine whether there is an influence of burner operation and NOx emissions on CO emissions.
4. The burner tests were conducted with little heat extraction in the reburn zone. This increased the rate of reduction reactions but may have resulted in excessive oxidation of TVFN in the tertiary combustion zone. The dependence of NOx emissions on tertiary air injection location supports this conclusion. Tests with higher heat extraction in the reburn zone may result in lower NOx emissions.

5. The IFSB operates with the reburn fuel injected directly into an O₂ lean zone. Since the burner was tested with reburn coal transported in air, which is an oxidizing gas, lower NOx emissions may be obtained if the reburn fuel is transported in recirculated flue gas. This technique has been demonstrated as an effective NOx reduction technique in air and fuel staged utility boilers. Thus, tests with flue gas recirculation are recommended.

6. The importance of reburn zone stoichiometry on NOx emissions was demonstrated in the Furnace Investigation and in the Burner Tests. However in the IFSB, lower emissions were measured when the reburn fuel fraction was increased from 20 to 30%. The results suggest that lower NOx emissions may be obtained with a higher reburn fuel fraction as long as the flame shape is maintained.
## List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Units</th>
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<tbody>
<tr>
<td>A</td>
<td>mixing rate constant</td>
<td></td>
</tr>
<tr>
<td>A&lt;sub&gt;c&lt;/sub&gt;</td>
<td>preexponential factor</td>
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<tr>
<td>a</td>
<td>secondary air pipe diameter</td>
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<tr>
<td>B&lt;sub&gt;1&lt;/sub&gt;</td>
<td>response surface equation coefficients</td>
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<tr>
<td>b</td>
<td>quarl exit diameter</td>
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<tr>
<td>C&lt;sub&gt;t&lt;/sub&gt;</td>
<td>time averaged species concentration</td>
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<td>C&lt;sub&gt;P&lt;/sub&gt;</td>
<td>specific heat</td>
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<td>C&lt;sub&gt;e1&lt;/sub&gt;</td>
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<td>C&lt;sub&gt;e2&lt;/sub&gt;</td>
<td>turbulence dissipation constant (2.09)</td>
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<td>effective viscosity constant (0.09)</td>
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<td>particle diameter</td>
<td>(m)</td>
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<tr>
<td>f&lt;sub&gt;RB&lt;/sub&gt;</td>
<td>reburn fuel fraction</td>
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<tr>
<td>G&lt;sub&gt;p&lt;/sub&gt;</td>
<td>primary air momentum</td>
<td>(kg·m/s²)</td>
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<td>k&lt;sub&gt;s&lt;/sub&gt;</td>
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<td>k&lt;sub&gt;d&lt;/sub&gt;</td>
<td>diffusion rate</td>
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<td>quarl length</td>
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<td>helium recirculation factor</td>
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<td>mass flow rate of primary combustion products</td>
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<td>flow rate of O₂ in precombustor products</td>
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<td>CO₂ mixing factor</td>
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<td>primary zone NOₓ concentration</td>
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<td>gas pressure</td>
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<td>O₂ partial pressure</td>
<td>(Pa)</td>
</tr>
<tr>
<td>P&lt;sub&gt;NO&lt;/sub&gt;</td>
<td>NO partial pressure</td>
<td>(Pa)</td>
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<td>Q&lt;sub&gt;c&lt;/sub&gt;</td>
<td>heat transferred to particle by convection</td>
<td>(W)</td>
</tr>
<tr>
<td>Q&lt;sub&gt;h&lt;/sub&gt;</td>
<td>heat generated in particle due to combustion</td>
<td>(W)</td>
</tr>
<tr>
<td>Q&lt;sub&gt;r&lt;/sub&gt;</td>
<td>heat transfer to particle by radiation</td>
<td>(W)</td>
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<td>q</td>
<td>char combustion rate</td>
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<tr>
<td>R&lt;sub&gt;ox&lt;/sub&gt;</td>
<td>consumption rate of O₂</td>
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<td>R&lt;sub&gt;u&lt;/sub&gt;</td>
<td>universal gas constant</td>
<td>(J/mol/K)</td>
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<tr>
<td>r</td>
<td>radial distance</td>
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<tr>
<td>S&lt;sub&gt;eff&lt;/sub&gt;</td>
<td>effective swirl number</td>
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<td>S&lt;sub&gt;n&lt;/sub&gt;</td>
<td>swirl number at air temperature, T&lt;sub&gt;n&lt;/sub&gt;</td>
<td>(kg/m³/s)</td>
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<td>S&lt;sub&gt;comb&lt;/sub&gt;</td>
<td>combustion source terms in Equation 3.12</td>
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<td>radiation source terms Equation 3.12</td>
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<td>species source terms Equation 3.11</td>
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<td>S&lt;sub&gt;'&lt;/sub&gt;</td>
<td>modified swirl number</td>
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<td>T&lt;sub&gt;g&lt;/sub&gt;</td>
<td>mean gas flame temperature</td>
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<td>T&lt;sub&gt;n&lt;/sub&gt;</td>
<td>inlet gas temperature</td>
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<td>T&lt;sub&gt;RB&lt;/sub&gt;</td>
<td>reburn zone temperature</td>
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<td>U</td>
<td>time mean axial gas velocity</td>
<td>(m/s)</td>
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<td>u</td>
<td>fluctuating axial gas velocity</td>
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<td>V</td>
<td>time mean radial gas velocity</td>
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<td>v</td>
<td>fluctuating radial gas velocity</td>
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</table>
\( W \)  
\( W \) time mean tangential gas velocity  
\( w \)  
\( w \) fluctuating tangential gas velocity  
\( X \)  
\( X \) dependent process variable (Equations 3.2-3.4)  
\( x \)  
\( x \) normalized process variable (Equations 3.2, 3.4)  
\( Y \)  
\( Y \) fraction of char remaining after partial combustion  
\( y \)  
\( y \) relative NO\(_x\) or TVFN concentration (Equation 3.3)

**Greek**

\( \alpha \)  
\( \alpha \) particle drag coefficient  
\( \epsilon \)  
\( \epsilon \) turbulent dissipation rate  
\( \Theta \)  
\( \Theta \) normal distributed random number variable  
\( \lambda_{\text{eff}} \)  
\( \lambda_{\text{eff}} \) effective reburn zone stoichiometry  
\( \lambda_p \)  
\( \lambda_p \) primary stoichiometry  
\( \lambda_{RB} \)  
\( \lambda_{RB} \) reburn zone stoichiometry  
\( \mu_l \)  
\( \mu_l \) laminar dynamic viscosity  
\( \mu_t \)  
\( \mu_t \) turbulent dynamic viscosity  
\( \rho_A \)  
\( \rho_A \) particle apparent density  
\( \rho_g \)  
\( \rho_g \) gas density  
\( \phi_c \)  
\( \phi_c \) O\(_2\) requirement for complete combustion  
\( \phi \)  
\( \phi \) rate functions used in Equations 2.5 and 2.6  
\( \tau_e \)  
\( \tau_e \) fluid eddy lifetime  
\( \tau_{RB} \)  
\( \tau_{RB} \) reburn zone residence time  
\( \nu_l \)  
\( \nu_l \) laminar kinematic viscosity  
\( \nu_t \)  
\( \nu_t \) turbulent kinematic viscosity  
\( \Omega \)  
\( \Omega \) cross-sectional area in Equation 3.5

**Subscripts**

\( g \)  
\( g \) gas  
\( p \)  
\( p \) particle  
\( RB \)  
\( RB \) reburn zone

**Abbreviations**

AASB  
\( \text{aerodynamically air staged burner} \)  
ERZ  
\( \text{external recirculation zone} \)  
hvB  
\( \text{high volatile bituminous} \)  
IFS\(_B\)  
\( \text{internally fuel staged burner} \)  
IPFR  
\( \text{isothermal plug flow reactor} \)  
IRZ  
\( \text{internal recirculation zone} \)  
mvB  
\( \text{medium volatile bituminous} \)  
PSD  
\( \text{particle size distribution} \)  
TVFN  
\( \text{total volatile fixed nitrogen (NO}_x + \text{HCN + NH}_3 \)  
VM  
\( \text{volatile matter} \)
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Appendix A

Reburn Zone Investigation Test Data

The influence of seven process parameters:

- coal type
- particle size distribution
- reburn fuel fraction
- primary stoichiometry
- reburn zone temperature
- reburn zone residence time and
- inlet NOx concentration

on NOx and TVFN reduction in the reburn zone was studied in 59 experimental tests. These tests were designated RB1-RB59 to represent the Reburn Zone Investigation. Each parameter was studied at three conditions over the range which would be attainable in industrial processes. The range of parameters studied are listed in Table 3.3 and the inlet conditions for each test are given in Table A1.

The test results are summarized in Table A2. At the time of the measurements, it was not known whether a single probe would be suitable for both NOx and HCN/NH3 measurements. Therefore, these measurements were conducted separately in two parts of the same test. Since inlet conditions could vary slightly from one part of a test to the second part, the measured NOx concentration and calculated reburn zone stoichiometry are reported for both parts.

Table A.1 Summary of inlet test conditions for the Reburn Zone study

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1SB = Scotts Branch; Obed = Obed Mountain
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1SB = Scotts Branch; Obed = Obed Mountain

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2 Data measured with HCN/NH<sub>3</sub> probe in reactor
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<sup>1</sup> Data measured with NOx probe in reactor

<sup>2</sup> Data measured with HCN/NH<sub>3</sub> probe in reactor
Appendix B

Input/Output Data from the Furnace Investigation

The influence of mixing on NOx reduction by fuel staging was studied in the Furnace Investigation. Seven coal reburning flames, F1-F7, and one natural gas reburn flame, F8, were studied. The results of these tests are tabulated in this appendix. Each flame was designated as having a particular primary zone stoichiometry and reburn fuel fraction. Two reburn gun positions, designated 1 and 2, were studied for each flame and the tertiary air injection arrangement was varied, designated as position 1 through 4.

The flow rates, velocity and temperature are given for each input and the stoichiometry at the end of the reburn zone, tertiary combustion zone and in the flue gas is listed. The flue gas temperature and gas emissions are also listed. The NOx concentration in the flue gas is reported at 0% O2, whereas, all other gas species are reported on a dry basis.
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### INPUT CONDITIONS

- **coal (kg/h)**: 210, 209, 211, 207, 215, 216, 215, 218, 217, 191, 193, 186, 193
- **prim air (kg/h)**: 400, 400, 400, 400, 400, 400, 400, 400, 400, 400, 400, 400
- **temp (°C)**: 57, 58, 56, 57, 57, 56, 58, 58, 55, 39, 45, 52, 52
- **velocity (m/s)**: 18, 18, 18, 18, 18, 18, 18, 17, 17, 17, 17, 17

- **temp (°C)**: 280, 274, 280, 280, 276, 281, 283, 283, 280, 275, 282, 279, 280
- **velocity (m/s)**: 37, 37, 38, 38, 42, 42, 42, 42, 37, 38, 36, 37

- **exit temp (°C)**: 1495, 1495, 1482, 1482, 1400, 1449, 1449, 1458, 1429, 1462, 1453, 1450, 1450
- **exit vel. (m/s)**: 10, 10, 10, 10, 10, 10, 10, 11, 10, 9, 9, 9, 9
- **exit stoich**: 4, 1.04, 1.04, 1.04, 1.04, 1.04, 1.11, 1.1, 1.10, 1.10, 1.09, 1.1, 1.1

### REBURN ZONE

- **coal (kg/h)**: 62, 62, 56, 55, 0, 60, 57, 60, 58, 88, 89, 85, 88
- **gas (kg/h)**: 62, 62, 56, 55, 0, 60, 57, 60, 58, 88, 89, 85, 88

- **reb. air (kg/h)**: 60, 60, 70, 60, 70, 60, 60, 60, 60, 60, 60, 60, 60
- **temp (°C)**: 57, 58, 60, 56, 56, 59, 57, 57, 57, 80, 49, 46, 58, 58
- **velocity (m/s)**: 36, 31, 36, 31, 32, 32, 32, 32, 31, 32, 32, 32
- **gun position**: 2, 1, 1, 2, 2, 2, 2, 1, 1, 0.30, 2, 2
- **exit stoich**: 0.84, 0.84, 0.86, 0.85, 1.14, 0.89, 0.89, 0.89, 0.89, 0.77, 0.76, 0.80, 0.77

### TERTIARY ZONE 1

- **air flow (kg/h)**: 924, 867, 504, 504, 713, 677, 279, 240, 602, 1012, 1009, 602
- **temp (°C)**: 240, 240, 244, 249, 245, 245, 245, 245, 251, 251, 251, 251
- **velocity (m/s)**: 43, 43, 50, 50, 35, 19, 16, 24, 61, 50, 51, 61
- **inj. position**: 1, 1, 2, 2, 1, 1, 1, 2, 2, 1, 1, 2
- **exit stoich**: 1.16, 1.16, 1.04, 1.04, 1.14, 1.15, 1.14, 1.00, 0.98, 0.98, 1.14, 1.16, 0.98

### TERTIARY ZONE 2

- **air flow (kg/h)**: 352, 436, 414, 418, 417, 417
- **temp (°C)**: 244, 249, 245, 245, 251, 251
- **velocity (m/s)**: 35, 44, 42, 42, 42, 42
- **inj. position**: 4, 4, 4, 4, 4, 4
- **exit stoich**: 1.16, 1.16, 1.18, 1.18, 1.14, 1.15, 1.14, 1.13, 1.13, 1.14, 1.14, 1.15

### CHIMNEY

- **temp (°C)**: 928, 936, 923, 928, 911, 983, 987, 992, 894, 924, 957, 991, 924
- **O2 (%)**: 4, 4, 3, 3, 4, 3, 3, 3, 4, 4, 2, 3, 3
- **CO2 (%)**: 16, 16, 16, 16, 15, 15, 15, 16, 16, 16, 16, 15, 16
- **CO (ppm)**: 3, 3, 7, 4, 1, 0, 0, 4, 0, 0, 2, 1, 1
- **NOx (ppm 0% O2)**: 480, 420, 300, 390, 1050, 590, 630, 510, 510, 260, 330, 470, 350
- **burnout (%)**: 98.2, 97.7, 95.7, 95.4, 97.7, 97.5, 97.0, 94.7, 93.3, 90.4, 95.8, 95.5, 90.2
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Appendix C
Input/Output Data from the Burner Tests

The internally fuel staged burner was studied in six coal reburning flames, FSB1-FSB6 and three natural gas reburning flames, FSB7-FSB9. The results of these tests are tabulated in this appendix. Each flame was designated as having a particular primary zone stoichiometry and reburn fuel fraction. For coal reburning, the reburn gun position was varied between 0 and 1 quarl diameters downstream of the quarl throat. For natural gas reburning, the gas was injected either radially (designated R in the Table) or axially (designated A in the Table) up to 5 quarl diameters downstream of the burner throat. The tertiary air injection arrangement was varied between position 1 through 4.

The flow rates, velocity and temperature are given for each input and the stoichiometry at the end of the reburn zone, tertiary combustion zone and in the flue gas is listed. The flue gas temperature and gas emissions are also listed. The NOx concentration in the flue gas is reported at 0% O₂, whereas, all other gas species are reported on a dry basis.
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<td>350</td>
<td>374</td>
<td>374</td>
<td>374</td>
</tr>
<tr>
<td>velocity (m/s)</td>
<td>38</td>
<td>39</td>
<td>39</td>
<td>39</td>
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<tr>
<td>exit stoich</td>
<td>1.10</td>
<td>1.14</td>
<td>1.14</td>
<td>1.14</td>
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</table>

**CHIMNEY**

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<table>
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<tbody>
<tr>
<td>temp (°C)</td>
<td>1105</td>
<td>1092</td>
<td>1097</td>
<td>1097</td>
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<tr>
<td>burnout (%)</td>
<td></td>
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<td>98.0</td>
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<tr>
<td>O₂ (%)</td>
<td>2</td>
<td>2</td>
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<tr>
<td>NOₓ (ppm 0%O₂)</td>
<td>100</td>
<td>220</td>
<td>170</td>
<td>190</td>
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<tr>
<td>CO (ppm)</td>
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<td>0</td>
<td>6</td>
<td>13</td>
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<tr>
<td>CO₂ (%)</td>
<td>14</td>
<td>15</td>
<td>15</td>
<td>16</td>
</tr>
</tbody>
</table>

- 206 -
CURRICULUM VITAE

Kevin J. Knill

15 November 1959
born at London, Canada

1977 - 1982
University of Waterloo, Waterloo, Canada
BASc Mechanical Engineering.

1982 - 1987
Alberta Research Council, Edmonton, Canada
Research Engineer: conducted research on coal
hydropyrolysis, coal and char combustion.

University of Alberta, Edmonton, Canada
M.Sc. Mechanical Engineering, June 1987,
Thesis: Evaluation of the Combustibility
of Hydropyrolysis Chars

1987 - present
International Flame Research Foundation, IJmuiden
Research Engineer: Studied NOx reduction by fuel and air
staging in coal, oil and gas fired systems.
Investigated coal devolatilization and char combustibility.
Studied direct iron ore reduction processes.
Photograph 1

water model flow simulation:
high mixing case.

Photograph 2

water model flow simulation:
high mixing case.

Photograph 3

water model flow simulation:
low mixing case.
Photograph 4
Furnace Investigation - reburn coal flow between the front wall and the first choke: high mixing case.

Photograph 5
Furnace Investigation - reburn coal flow between the first and second chokes: high mixing case.

Photograph 6
IFSB - without reburn fuel injection:
\( \lambda_p = 1.05, \, \text{swirl no.} = 1.2 \)

Photograph 7
IFSB - with reburn fuel injector positioned at one quarter diameter:
\( \lambda_p = 1.05, \, f_{RB} = 30\%, \)
\( \lambda_{RB} = 0.75 \)
\( \text{NOx} = 250 \, \text{ppm} \)