## RANS Simulation of Oxy-Natural Gas Combustion

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Board of Studies of Energy Aalborg University

Author:

Lars Christian Riis JOHANSEN

Supervisor:

Chungen YIN



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Lars Christian Riis Johansen

have been used as the test case for the CFD simulations. Symmetry has been used to create quarter meshes of the furnace for CFD. Oxy-natural gas chemistry has been investigated with equilibrium calculations. The WD mechanism has been used. The JL mechanism failed in FLU-ENT due to program error. A numerical fuel has been developed for the WD mechanism. A WSGGM applicable to oxy-fuel conditions is used in the simulations. CFD simulations with various models have been performed. All simulations provided good predictions of velocity and turbulence and poor predictions of temperature. Species were predicted to varying accuracies. Inlet turbulence intensity has been found to have the greatest effect on simulation accuracy.

The accuracy of CFD simulations using the RANS approach has been investigated

OXYFLAM-2 experiments by the IFRF

The

for oxy-natural gas combustion.

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By signing this document, each member of the group confirms that all participated in the project work and thereby all members are collectively liable for the content of the report.

# Preface

This report has been written in the 2010 spring semester. The report is submitted as documentation for the project work performed during the 4th semester of the M.Sc. program in Thermal Energy and Process Engineering (TEPE4) under the Board of Studies of Energy at Aalborg University. Associate Professor Chungen Yin has supervised during the project period.

The Chicago style guide citation system has been used. Citations are stated with the surnames of the authors or organisation and date of publication i.e. Surname #1, Surname #2, Surname #3 et al. [Year of publication]. Figures and tables are numbered according to the number of the chapter in which they are placed and a separate sequential number for figures and tables. The first figure in chapter 2, for example, is referred to as Figure 2.1. A nomenclature of symbols and abbreviations precedes the table of contents and a bibliography follows after the main body of the report. Relevant material not included in the report is placed in the appendix following the bibliography. A DVD with the report and CFD case files is included with the report.

# Nomenclature

## Abbreviations

Abbreviation	Description
ASU	Air separation unit
CARS	Coherent anti-Stokes Raman spectroscopy
CCS	Carbon capture and storage
CEA	Chemical Equilibrium with Applications
CFD	Computational fluid dynamics
CPR	Constant pressure reactor
DO	Discrete ordinates
ECBM	Enhanced coal bed methane
ED	Eddy dissipation
EDC	Eddy dissipation concept
EOR	Enhanced oil recovery
EPR	Electron paramagnetic resonance
EWBM	Exponential wide band model
FGR	Flue gas recirculation
FID	Flame ionization detector
GHG	Green house gas
IFRF	International Flame Research Foundation
IGCC	Integrated gasification combined cycle
JL	Jones-Lindstedt
LDV	Laser Doppler velocimetry
LHV	Lower heating value $\begin{bmatrix} kJ \\ kg \end{bmatrix}$
MEA	Mono ethanol amine
MDEA	Methyl diethanol amine
MILD	Moderate to intensive low oxygen dilution
Mtons	Million metric tons
NDIR	Non dispersive infrared sensor
NG	Natural gas
NS	Navier Stokes
PCF	Pulverized coal fired
PSR	Perfectly stirred reactor
RANS	Reynolds averaged Navier Stokes
UDF	User defined function
VCRF	Vertical Combustor Research Facility

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Abbreviation	Description
WD	Westbrook-Dryer
WSGGM	Weighted sum of grey gases model

## Symbols

Symbol	Description
a	Absorption coefficient $[m^{-1}]$
$a_{arepsilon,i}$	WSGGM weighting factor function for grey gas $i$
$c_p$	Specific heat at constant pressure $\begin{bmatrix} J \\ kg \cdot K \end{bmatrix}$
$c_v$	Specific heat at constant volume $\begin{bmatrix} J \\ kg \cdot K \end{bmatrix}$
e	Specific internal energy $\left\lfloor \frac{\mathbf{J}}{\mathbf{kg}} \right\rfloor$
g	Gravitational acceleration
h	Specific enthalpy $\left\lfloor \frac{kJ}{kg} \right\rfloor$
$ar{h}$	Mole specific enthalpy $\left[\frac{kJ}{kmol}\right]$
k	Thermal conductivity $\begin{bmatrix} W \\ \overline{K \cdot m} \end{bmatrix}$
k	Turbulent kinetic energy $\left[\frac{m^2}{s^2}\right]$
$k_{b,r}$	Backward reaction rate of reaction $r$
$k_{f,r}$	Forward reaction rate of reaction $r$
l	Turbulence length scale [m]
m	Mass [kg]
$\dot{m}$	Mass flow rate $\left\lfloor \frac{\text{kg}}{\text{s}} \right\rfloor$
n	Moles [kmol]
$\dot{n}$	Mole flow rate $\left[\frac{\text{kmol}}{\text{s}}\right]$
p	Pressure [Pa]
$\dot{q}$	Heat flux $\lfloor \frac{W}{m^2} \rfloor$
r	Reaction r
$\mathbf{r}_{co/xf_i}$	Cell centroid to cell face vector [m]
r	Incident radiation direction vector
s	Specific entropy $\left  \frac{J}{kg \cdot K} \right $
$\mathbf{S}$	Radiation direction vector
t	Time [s]
u	x component of the velocity vector $\left\lfloor \frac{\mathbf{m}}{\mathbf{s}} \right\rfloor$
v	y component of the velocity vector $\begin{bmatrix} m \\ s \end{bmatrix}$
w	z component of the velocity vector $\left\lfloor \frac{\mathbf{m}}{\mathbf{s}} \right\rfloor$
x	x coordinate [m]
y	y  coordinate  [m]
z	z coordinate [m]
$A_r$	Pre-exponential factor
$\mathbf{A}_i$	Cell face vector [m <sup>4</sup> ]
$D_k$	Binary diffusion coefficient of species $k \left\lfloor \frac{\mathbf{m}^2}{\mathbf{s}} \right\rfloor$
$E_A$	Activation energy $\begin{bmatrix} J \\ kmol \end{bmatrix}$

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$\mathbf{Symbol}$	Description
F	Body force $\left[\frac{N}{m^3}\right]$
Ι	Total radiation intensity $\left[\frac{W}{m^2 \cdot sr}\right]$
$K_r$	Equilibrium constant of reaction $r$
M	Mixture mole mass $\left[\frac{\text{kg}}{\text{kmol}}\right]$
$M_i$	Mole mass of species $i \left[ \frac{\text{kg}}{\text{kmol}} \right]$
$N_{ heta}$	Angular subdivisions in the $\theta$ coordinate
$N_{\phi}$	Angular subdivisions in the $\theta$ coordinate
P	Sum of partial pressures of absorbing species [bar]
Pr	Prandtl number
$\dot{Q}$	Heat rate [W]
R	Specific gas constant $\begin{bmatrix} J \\ \overline{K \cdot kg} \end{bmatrix}$
$R_u$	Gas constant. $R_u = 8314.472 \begin{bmatrix} J \\ \overline{K \cdot kmol} \end{bmatrix}$
$R_{i,r}$	Mass reaction rate of species <i>i</i> in reaction $r \left[\frac{\text{kg}}{\text{m}^3 \cdot \text{s}}\right]$
$\overline{R}_{i,r}$	Mole reaction rate of species <i>i</i> in reaction $r \left[\frac{\text{kmol}}{\text{m}^3 \cdot \text{s}}\right]$
$Re_h$	Reynolds number based on hydraulic diameter
S	Beam length [m]
Sc	Schmidt number
$S_e$	Energy source $\left[\frac{J}{m^3 \cdot c}\right]$
T	Temperature [K]
U	Velocity $\left[\frac{m}{s}\right]$
$U'_{\rm rms}$	Root mean square fluctuating velocity $\left[\frac{m}{s}\right]$
X	Mole fraction
Y	Mass fraction
$\mathbf{U}$	Velocity vector $\left[\frac{\mathbf{m}}{\mathbf{s}}\right]$
[X]	Mole concentration $\left[\frac{\text{kmol}}{\text{m}^3}\right]$

## Greek Symbols

Symbol	Description
$\beta_r$	Temperature exponent
$\delta_{ij}$	Kronecker delta function
$\epsilon$	Turbulent energy dissipation rate $\left[\frac{m^2}{s^3}\right]$
ε	Emissivity
$\eta$	Rate exponent
$\kappa$	Absorption coefficient $\left[ bar^{-1} \cdot m^{-1} \right]$
$\mu$	Dynamic viscosity $\left[\frac{\text{kg}}{\text{m}\cdot\text{s}}\right]$
ν	Stoichiometric coefficient
$\varphi$	Scalar variable
ρ	Density $\left[\frac{\text{kg}}{\text{m}^3}\right]$
$\sigma$	Stefan-Boltzmann constant $\sigma = 5.6704 \cdot 10^{-8} \left[ \frac{W}{m^2 \cdot K^4} \right]$
$\sigma_s$	Scattering coefficient $[m^{-1}]$

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Symbol	Description
$ au^*$	Fine scale time scale [s]
Ω	Solid angle [sr]
$\Phi$	Equivalence ratio
Θ	Viscous work $\left[\frac{J}{m^3 \cdot s}\right]$
$\nabla$	Gradient operator
$ abla \cdot$	Divergence operator
abla  imes	Curl operator

## Superscripts

Symbol	Description
0	At reference state temperature and pressure
*	Fine scale property
/	Fluctuating component
/	Reaction reactant
//	Fluctuating component including the effect of density fluctuation
//	Reaction product

## Subscripts

Symbol	Description
f	Formation property
i	Species $i$
j	Species $j$
r	Reaction $r$
s	Derivative with respect to constant entropy
s	Turbulent quantity
F	Fuel
0	Oxidizer

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# Chapter 1

# Introduction

This chapter is a revised and updated version of work by the author first presented in Johansen et al. [2009].

The increasing focus on climate change and the role of greenhouse gases (GHG) in climate change has brought focus on the use of fossil fuels for power generation. It is widely accepted that anthropogenic greenhouse gas emission is the leading cause of climate change and the main source of anthropogenic greenhouse gases is the use of fossil fuels as stated by International Energy Agency [2006] and International Energy Agency [2009]. Coal and natural gas, fossil fuels, are widely used in power generation and are sources of the greenhouse gases  $CO_2$ ,  $SO_x$  and  $NO_x$ . At present all coal and natural gas fired power plants exhaust the  $CO_2$  produced from combustion to the atmosphere. This is in contrast to the emission of other pollutants such as sulphur oxides  $SO_x$  and nitrous oxides NO<sub>x</sub> which are regulated to some degree in most countries and are removed from the flue gas. In 2007, 28962.4 Mtons of  $CO_2$  were emitted from anthropogenic sources with an increase of 38% from 1990 to 2007 according to figures given by International Energy Agency [2009]. Coal accounted for 12228.1 Mtons and natural gas for 5733.8 Mtons of the 28962.4 Mtons of CO<sub>2</sub> emitted representing a combined 62.02% of emissions according to International Energy Agency [2009]. International Energy Agency [2009] report that the main activity electricity and heat production sector accounted for 67.12% of the CO<sub>2</sub> emissions from the use of coal and natural gas respectively in 2007. A 76.9% and 59.2% increase respectively has occurred between 1990 and 2007 according to International Energy Agency [2009]. Viewed in relation to the total production of greenhouse gasses the main activity electricity and heat production sector using coal and natural gas represented a combined 23.7% of greenhouse gas production in 2007. It can be concluded that electricity and heat generated from coal and natural gas represent a significant source of greenhouse gas emissions with large increase in emissions from this sector occuring over the last two decades.

### 1.1 Electricity Production CO<sub>2</sub> Emissions

One of the avenues pursued to reduce  $CO_2$  emission from power generation is the use of renewable energy sources. Renewable energy sources meet an increasing share of the world energy demands. However, as pointed out by Buhre et al. [2005], until these sources can reliably produce a significant share of the energy demands the most

accessible energy source will continue to be fossil fuels. Coal is an abundant and cheap fossil fuel in many regions and is already a well established method of power generation. Natural gas is also widely used to produce electricity. As an example, in 2009 coal and natural gas accounted for 44.6% and 23.3% respectively of electrical energy produced in the USA according to figures from U.S. Energy Information Administration [2010]. Over 85% of electricity generated in Australia is produced by pulverised coal fired (PCF) power plants according to Buhre et al. [2005]. There is great interest in continuing development of clean coal technology due to the stable and economic supply of coal and the ability of new coal utilisation technology to meet stricter emission controls according to Beér [2006]. Methods for reducing CO2 emission from PCF and natural gas fired power plants are therefore of interest. The following methods to achieve this aim are listed by Buhre et al. [2005]:

- Improving efficiency of power plants.
- Capture and storage of CO2.

It has been concluded by Beér [2006] that efficiency improvement is the most predictable and lowest cost method to reduce all emissions. This approach to emission reduction is aided by power generation efficiency increasing steadily as reviewed in Beér [2006]. Until the introduction of commercially viable plants incorporating CCS, the most cost effective emission reduction approach is to construct power plants with the highest efficiency permitted by cost and the availability of required technology. Ber concludes that future "zero emission" power plants will include carbon capture and storage. The last option, carbon capture and storage (CCS), eliminates  $CO_2$  emissions due to combustion entirely. This makes the option attractive for a drastic reduction in  $CO_2$  emissions from existing PCF and natural gas fired power plants as well as future plants. Several methods for CCS are presented by Buhre et al. [2005]:

- CO<sub>2</sub> capture by scrubbing of the flue gas.
- $\bullet\,$  Integrated gasification combined cycle (IGCC) with  $\mathrm{CO}_2$  capture for PCF power plants.
- Oxy-fuel combustion with external recycling.
- Oxy-fuel combustion with internal recycling.
- Chemical looping.

Of these options the first three are considered closest to commercial application by Buhre et al. [2005]. Scrubbing of the flue gas using mono ethanol amine (MEA) or methyl diethanol amine (MDEA) is costly according to Buhre et al. [2005].  $CO_2$  capture is made easier with higher concentrations of  $CO_2$  in the flue gas. This can be achieved with oxy-fuel combustion where pure oxygen is used rather than air as the oxidizer. The major constituents of the flue gas from oxy-fuel combustion are  $H_2O$  and  $CO_2$  and the flue gas is ready to be compressed and sequestered without separation of the  $CO_2$ from the flue gas beforehand. With flue gas recycling the  $CO_2$  concentration is raised to above 90% according to Beér [2006]. A possible flow chart for an oxy-PCF power plant is shown in 1.1 from Buhre et al. [2005].



Figure 1.1: Flow chart of a oxy-PCF combustion power plant for CCS as presented in Buhre et al. [2005].

### 1.2 Oxy-Fuel Combustion

During normal air firing of coal and natural gas the  $CO_2$  produced is diluted by the 79%  $N_2$  present in air on a volume basis. Using 95% pure  $O_2$  and recycling the flue gas a significant increase in the volume fraction of  $CO_2$  in the flue gas can be achieved. Using oxygen as the oxidizer instead of air raises the constant pressure adiabatic temperature. Flue gas recycling is used to control the flame temperature and to compensate for the missing  $N_2$  gas volume. The following differences between air firing and oxygen firing are pointed out by Buhre et al. [2005].

- The  $O_2$  proportion passing through the burner is typically 30% and a recycle ratio of 60% is employed for a similar adiabatic temperature to air firing.
- The higher proportion of  $\rm CO_2$  and  $\rm H_2O$  in the furnace gas leads to a higher gas emissivity.
- The higher proportion of CO<sub>2</sub> and H<sub>2</sub>O leads to a higher specific thermal capacity.
- The higher proportion of CO<sub>2</sub> leads to a higher gas density.
- The furnace gas volume is reduced slightly. The flue gas volume after recycling is reduced by 80%.
- The SO<sub>x</sub> concentration in the flue gas is higher due to reduced flue gas volume.

A number of laboratory and pilot scale studies have been performed to gain a better understanding of oxy-fuel combustion. A review of laboratory and pilot scale studies can be found in Buhre et al. [2005]. A review of technology and economic investigations is also found in Buhre et al. [2005] and Beér [2006]. The review of pilot scale studies concludes that oxy-PCF combustion presents no prohibitive technical barriers. Oxy-fuel combustion can, as mentioned previously, be implemented as a retrofit to existing power plants. Further,  $NO_x$  and possibly mercury emissions are reduced as discussed in Section 1.2.2. The review of technology and economic investigations concludes that oxy-fuel combustion is economically feasible for retrofitting. Oxy-fuel combustion for CCS is a competitive power generation technology despite the associated efficiency and cost penalties when compared with MEA and MDEA CCS technology. The highest cost is associated with the air separation unit (ASU) required to produce the oxygen used for combustion. This cost may be offset by the reduced cost of flue gas treatment due to the reduced flue gas volume and elimination of DeNO<sub>x</sub> and DeSO<sub>x</sub> equipment. The difference in flowcharts between air-firing and oxy-firing for CCS is illustrated in 1.2.



Figure 1.2: Flow charts for a PCF power plant using as oxidizer (a) air and (b) oxy-flue gas as presented by Beér [2006].

Currently there are a number of projects with oxy-fuel combustion running. The Schwarze Pumpe power station in north-east Germany is the first pilot scale power plant to implement carbon capture and storage as reported by Harrabin [2008]. The pilot project is led by Vattenfall. Another pilot scale power plant project is also underway in Australia as reported by Beér [2006]. Development of a CCS demonstration project by Vattenfall at the Nordjylland Power Station was announced in 2008 as reported by Vattenfall [2010].

#### 1.2.1 Sequestration

The subject of  $CO_2$  sequestration is still under investigation. There are as of present a number of possibilities being investigated some of which are presented by Buhre et al. [2005]. Oxy-fuel combustion initially attracted interest in the 1980s as a method of producing  $CO_2$  for enhanced oil recovery (EOR).  $CO_2$  can be used in depleted oil and gas reservoirs to acquire remaining deposits. Enhanced coal bed methane (ECBM), similar to EOR, is a method to acquire the methane locked in unmineable coal seams by injection of  $CO_2$  into the seam. Oceans are natural carbon sinks and as such have been investigated for  $CO_2$  sequestration. However, there is at present concern about ocean acidification due to increased atmospheric  $CO_2$  and the effect of  $CO_2$  sequestration is not fully understood. Deep underground saline aquifers and sandstone formations have been investigated due to their ability to absorb large amounts of  $CO_2$  and their widespread occurrence. The CCS demonstration project by Vattenfall at the Nordjylland Power Station intends to use nearby sandstone formations for sequestration as reported by Vattenfall [2010].

#### 1.2.2 Emissions

A review of findings on emissions from oxy-fuel combustion is given by Buhre et al. [2005]. Studies suggest that it is possible to obtain  $\rm CO_2$  flue gas concentrations higher than 95% with oxy-fuel combustion. Oxy-fuel combustion has garnered a large amount of interest in the USA because of the reduction in formation of  $\rm NO_x$  documented in several studies.  $\rm NO_x$  reductions to less than a third of the levels seen under air firing conditions have been obtained. This is thought to be due to the low concentration of  $\rm N_2$ , reburning of the recycled  $\rm NO_x$  and reduction of the recycled NOx in the area of volatile matter release.  $\rm SO_x$  emissions have similarly found to be reduced in oxy-fuel combustion. A reduction ranging from 91% to 64% of the coal bound sulphur reacting to form  $\rm SO_2$  was observed. The reduced flue gas volume leads to higher  $\rm SO_x$  concentrations which may cause problems with corrosion in the combustion equipment. This may necessitate  $\rm DeSO_x$  depending on the conditions in spite of the reduced quantities of  $\rm SO_x$ .

#### 1.2.3 Heat Transfer

As mentioned in Section 1.2 there are a number of differences between air firing and oxygen firing of coal and natural gas. The change in heat transfer characteristics in a furnace when oxygen firing is employed has been previously mentioned. The main reasons for this change are due to the change in gas thermal capacity and gas radiative properties as pointed out by Buhre et al. [2005]. The higher proportion of  $CO_2$  and  $H_2O$  increases the emissivity and absorptivity. The  $N_2$  present under air firing conditions does not absorb or emit any significant amount of thermal radiation.  $CO_2$  and  $H_2O$  have numerous absorption bands in the infrared spectrum and this increases the gas absorbtivity and emissivity as discussed by Siegel and Howell [2002]. This increases the magnitude of radiative heat transfer in the furnace in proportion to convection heat transfer.

### **1.3** Oxy-Fuel Experiments

Oxy-fuel combustion experiments to investigate conditions in furnaces and provide data for subsequent modelling and simulation efforts can be found in the literature. Oxy-coal combustion at the CANMET Vertical Combustor Research Facility (VCRF) is designed to study oxy-fuel combustion and has been used to investigate both oxy-natural gas and oxy-coal combustion as discussed by Chui et al. [2003]. The International Flame Research Foundation (IFRF) has conducted a series of experiments in oxy-natural gas combustion experiments in a water cooled and refractory lined furnace under the OXYFLAME project to gather detailed in furnace data as documented by Lallemant et al. [1997]. The temperature data were found to be several hundred degrees lower than the true gas temperature as reported by Lallemant et al. [1997]. Breussin et al. [2000] pointed out that this was due to the double venturi pyrometer which was difficult to calibrate. New temperature measurements were performed by Breussin et al. [2000] in light of the deficiencies in the data reported by Lallemant et al. [1997]. The IFRF OXYFLAME furnace has also been used to conduct experiments with staged combustion of natural gas under moderate to intensive low oxygen dilution (MILD) combustion conditions as described by Breussin et al. [2000]. MILD combustion is the use of preheated air to combust fuel in the presence of large quantities of flue gas as explained by Mancini et al. [2002]. MILD combustion resembles oxy-fuel combustion where  $CO_2$  and  $H_2O$  are also present in large quantities during combustion.

### 1.4 Oxy-Fuel Modelling and Simulations

Chui et al. [2003] conducted CFD simulations of oxy-coal combustion for verification of CFD models based on the data gathered at the VCRF facility. Chui et al. [2003] used a coal combustion model developed for combustion with air and report that radiative properties should automatically adjust to give the correct values. The veracity of the assumption of applicability to oxy-coal combustion are not investigated. The CFD simulations are found to yield results that are reasonably accurate engineering predictions of temperature, CO,  $O_2$  and NO. More recently Chui et al. [2004] investigated several oxy-coal burner designs using the same approach.

Bollettini et al. [1997] conducted CFD simulations of the oxy-natural gas combustion measured by Lallemant et al. [1997] as part of the OXYFLAME project. Bollettini et al. [1997] employed the eddy dissipation (ED) model with a two step global reaction scheme to simulate the chemistry and the chemistry-turbulence interaction. A constant absorption coefficient determined using the EWBM model for average furnace conditions was used. Models for the thermal and prompt formation mechanisms of NOx were used. Good agreement was found for the fluid dynamics and main species. Further work on the prediction of  $H_2$  and CO was found to be required and NOx was poorly predicted.

Breussin et al. [2000] expanded upon the work by Bollettini et al. [1997] and performed CFD simulations of oxy-natural gas combustion and MILD combustion of natural gas. The simulations used the ED model used by Bollettini et al. [1997] and an eddy dissipation concept model coupled with equilibrium chemistry. A reburn NOx formation mechanism model was added to the thermal and prompt formation mechanisms. There is no mention of the radiation model employed and it is assumed to be the same as that used by Bollettini et al. [1997]. Good agreement with the experimental data has been achieved for the fluid dynamics, temperature and main species and NO prediction has been improved. CO is poorly predicted. It has been found that the equilibrium approach yielded the most accurate results of the two chemistry models used.

Andersen et al. [2009] modified the Westbrook-Dryer (WD) two step global reaction scheme for methane and the Jones-Lindstedt (JL) four step global reaction scheme for hydrocarbons for oxy-natural gas combustion conditions. The initiating reactions are retained for both schemes and the H<sub>2</sub>-CO-CO<sub>2</sub> reactions have been modified. Effort was concentrated on correct prediction of CO. CFD simulations of a oxy-propane combustion were performed using the P-1 radiation model, only applicable to optically thick mediums as discussed by Siegel and Howell [2002], the realizable k- $\epsilon$  turbulence model and the EDC model. No mention of the absorption coefficient model used is made. The modified WD scheme yielded improved temperature and CO predictions in the post flame region. The modified JL scheme provided slightly better CO predictions in the flame zone.

Currently CFD simulations often employ the weighted sum of gray gases (WSGG) model in order to determine gas emissivity. The standard WSGG model has been developed for air firing conditions. As evident from the preceding discussion, the standard WSGG model is unlikely to yield accurate results for gas radiative properties under oxy-fuel combustion. Johansson et al. [2010] investigated the accuracy of the WSGGM, the spectral line weighted sum of grey gases and a two grey gas approximation for the gas emissivity. New coefficients for the WSGGM were developed for oxy-fuel conditions. Yin et al. [2010] developed WSGGM models for oxy-fuel conditions by fitting the WSGGM to emissivity data obtained from the exponential wide band model (EWBM). This has been incorporated in to a UDF file for use in the CFD software Fluent.

## Chapter 2

# **Investigation Subject**

The design of oxy-fuel combustion systems for efficient and low emission operation requires the ability to determine furnace conditions. This can be achieved through experiments and measurements with actual systems or through modelling and CFD simulations. The former is costly when compared to CFD simulations where multiple designs and operating conditions can be investigated with no additional cost. The accuracy of CFD simulations is dependent on the validity and accuracy of the models employed as discussed by Casey et al. [2000]. Models are employed for the discretization of equations, turbulence, chemistry, chemistry-turbulence interaction, radiation properties, radiation transfer solvers and a number of other flow properties. From the discussion in Section 1.4 it is apparent that CFD simulations of oxy-fuel combustion have so far either employed radiative property models developed for combustion with air or have employed constant properties. Only Andersen et al. [2009] has so far employed reaction schemes developed for oxy-fuel combustion and are assumed to lack validated radiative property modelling. Bollettini et al. [1997] and Breussin et al. [2000] both use either a two step reaction scheme or equilibrium chemistry. Thus all work so far on oxy-fuel combustion has lacked either validated chemistry modelling or radiative property modelling. The preceding discussion leads to the following question.

#### How accurate is RANS simulation of oxy-natural gas combustion?

The effort of this report has been to answer the above question. The investigation has focused on the effect of various models on the accuracy of CFD simulations. Natural gas has been chosen as the subject of investigation because there are fewer uncertainties associated with natural gas combustion as compared to coal combustion. Modelling of multiphase flow, char oxidation and the determination of the coal composition are not required with natural gas combustion simulation. As discussed in Section 1.3, Lallemant et al. [1997] present detailed in furnace data gathered for oxy-natural gas combustion during the OXYFLAME project. The data on fluid dynamics, temperature and main species provides a means of validating CFD simulation accuracy. The furnace and operating conditions described by Lallemant et al. [1997] have therefore been chosen as the test case for investigation.

Investigation of NOx formation has been neglected. NOx formation in combustion systems is of concern as it causes photochemical smog and acid rain as discussed by Turns [2006]. Reduction of NOx is therefore of great interest to designers and, as discussed in Section 1.2.2, oxy-fuel combustion has been shown to lower NOx emissions. CFD simulations are often concerned with predicting NOx formation in order to design lower NOx emissions. NOx prediction has been a primary goal in CFD simulations by Bollettini et al. [1997], Breussin et al. [2000] and Mancini et al. [2002]. NOx concentrations are usually small when compared to the major species, ranging from the order of 100 ppm (dry) in MILD combustion as determined by Mancini et al. [2002] to 1000 ppm (dry) as measured by Lallemant et al. [1997] for oxy-natural gas combustion measurements. Because of the small fraction of NOx, there is a negligible change in the major species  $O_2$ ,  $N_2$  and hydrocarbons involved in the NOx formation reactions and the enthalpy of the flow. Neglection of NOx formation therefore does not introduce any significant error in the determination of the furnace conditions. Further evidence of this fact is the practice of simulating NOx formation after all other flow properties have been determined and using fixed flow conditions for NOx calculations as discussed by ANSYS Inc. [2009]. Accurate determination of furnace conditions is therefore assumed to be imperative to NOx determination. With the accurate simulation of flow properties, NOx can be determined subsequently.

## Chapter 3

# Furnace

This chapter details the IFRF OXYFLAM-2 furnace with burner A chosen as the test case for CFD simulations. The burner, operating conditions and the measurements performed are discussed. The meshes constructed for the CFD simulations based on the furnace and burner are then described.

### 3.1 Furnace

The IFRF conducted the OXYFLAM project, in collaboration with industry partners, with the objective of producing engineering information on oxy-natural gas combustion as outlined by Lallemant et al. [1997]. The purpose of the gathered information has been for use in optimizing heat transfer and NOx emission in oxy-natural gas combustion. Specifically outlined objectives of the OXYFLAM project were to characterize oxy-natural gas flames of 1 to 2 MW thermal input through detailed in flame measurements. The characterization focused on the effect of various parameters on flame structure, NOx formation and heat transfer. NOx reduction methodology was investigated through measurements of mixing, species and temperature.

Measurements of oxy-natural gas combustion were performed in the horizontal IFRF furnace no. 2 in two configurations, OXYFLAM-1 and OXYFLAM-2. Details of the furnaces and experiments conducted are presented by Lallemant et al. [1997], Lallemant et al. [2000]. In the OXYFLAM-1 configuration the furnace had bare walls. In the OXYFLAM-2 configuration a refractory lining was used. The furnace in both configurations consisted of 13 water cooled segments 300 mm wide. The OXYFLAM-1 configuration was operated at a thermal input of 1 MW while the OXYFLAM-2 furnace operated at 0.78 MW. The lower operating power of the OXYFLAM-2 configuration was necessary to lower temperatures in the furnace as necessitated by the refractory lining increasing the temperature compared to the OXYFLAM-1 configuration. Both furnaces were operated at a pressure of  $3 \text{ mm H}_2O$  to prevent air leaking in. The OXYFLAM-2 configuration has been chosen as the test case in this report. The dimensions of the OXYFLAM-2 furnace are given in Table 3.1 and the properties of the refractory lining are given in Table 3.2. Sources disagree on the length of the furnace. Lallemant et al. [1997] and Bollettini et al. [1997] report a length of 3.74 [m] while Breussin et al. [2000] and Lallemant et al. [2000] report 3.44 [m]. The latter has been used. A cross section diagram of the OXYFLAM-2 furnace is shown in Figure 3.1. A

	Dimension [mm]
Internal length	3440
Nearly square section	$1050 \ge 1050$
Chimney contraction diameter	500

Table 3.1: Dimensions of the refractory lined OXYFLAM-2 furnace as given by Lallemant et al. [1997].

Magnesite stone thickness	114 [mm]
Heat loss	$35\left[\frac{kW}{m^2}\right]$
Estimated hot face temperature	1656 [°C]
Cold face temperature	$38 [^{\rm o}C]$
Apparent porosity	8%
Bulk density	$2950 \left[\frac{\text{kg}}{\text{m}^3}\right]$
Conductivity	$6\left[\frac{W}{m\cdot K}\right]^2$
Melting temperature of ceramic bricks and binder	$\approx 1750  [^{\rm o}{\rm C}]$

Table 3.2: Specifications for the refractory lining in the OXYFLAM-2 furnace as given by Lallemant et al. [1997].

horizontal slot spans the first 1700 mm from the burner for measurement instrument access. For the remainder of the furnace access is provided by slots in the middle of each water cooled segment. The slots are illustrated in Figure 3.1.



Figure 3.1: Cross section diagram of the OXYFLAM-2 furnace reproduced from Lallemant et al. [1997].

Four burners were designed based on the same generic design shown in Figure 3.2. The natural gas was fed through the central inlet with diameter Dg in most experiments while the oxygen was fed through the annular coaxial inlet with inner diameter D1 and outer diameter D2. An experiment with the streams swapped was also performed. The separation of fuel and oxidizer until the furnace produces non premixed combustion. Dimensions for burners A, B and C are given in Table 3.3. The burners had increasing inlet diameters respectively the same gas and oxygen inlet separation of 6 mm. The

Burner	Momentum	$D2 \ [mm]$	D1 [mm]	$Dg \ [mm]$
Burner A	High	36	28	16
Burner B	Medium	45	33	21
Burner C	Low	60	42	30
Burner 2	Medium	54	45	21

Table 3.3: Dimensions of the burners used in the OXYFLAM furnace as shown by Lallemant et al. [1997].

fourth burner, burner 2, had a separation of 12 mm to study the effect of stream separation distance. All burner were found to be quiet according to Lallemant et al. [1997] and Lallemant et al. [2000]. Burner A has been chosen as the test case for the CFD simulations.



Figure 3.2: Diagram of OXYFLAM-2 burner from Lallemant et al. [1997].

The mass flow rates of natural gas and oxygen, inlet area and Reynolds number are given in Table 3.4. Pipe flow can be assumed to be turbulent for a Reynolds number based on hydraulic diameter of  $Re_H > 4000$  as stated by Munson et al. [2006]. Jets are subject to inviscid instability and will amplify long wavelength disturbances at  $Re_H > 10$  and thus become turbulent as discussed by Versteeg and Malalasekra [2007]. Further, for  $Re_H > 10^4$  the interface between the jet and surrounding fluid contains both large and small scale eddies whereas there is a lack of small scale eddies below this Reynolds number as discussed by Davidson [2004]. As evident from the Reynolds number for both streams, the natural gas and oxygen pipe flows are fully turbulent and the jets issuing from the burner are also fully turbulent. The turbulent flow of natural gas and oxygen gives rise to turbulent non premixed combustion. Reaction takes place at the interface between the natural gas and oxygen jets where mixing occurs. The natural gas used during the OXYFLAM-2 experiments is primarily composed of CH<sub>4</sub> and the composition is presented in Table 3.5 The oxygen feed is reported as being 99.9% pure by Lallemant et al. [1997].

#### 3.1.1 Measurements

The following description of observations and measurements are based on Lallemant et al. [1997], Lallemant et al. [2000] and Lallemant et al. [2003] where additional details can be found. During observations the flame could not be distinguished from background emissions. A faint yellow glow was observed 1-1.5 [m] downstream of the burner. This indicated the presence of soot. As no soot deposits were found on probes

	Natural Gas	Oxygen
Mass flow rate	0.01764	0.06298
Injection velocity (NTP) $\left\lceil \frac{m}{s} \right\rceil$	105.4	109.7
Momentum [N]	1.86	6.9
Reynolds number	128600	62400
Inlet area $[m^2]$	$2.011\cdot 10^{-4}$	$4.021\cdot10^{-4}$
Density at NTP $\left[\frac{\text{kg}}{\text{m}^3}\right]$	0.8325	1.428

Table 3.4: Operating conditions for the natural gas and oxygen streams used in the OXYFLAM-2 furnace with Burner A as given by Lallemant et al. [1997]. Velocities given at natural temperature and pressure (NTP) 273 [K] and 1 [atm].

Natural Gas Property	Units	Quantity
$CH_4$ fraction	% Vol	86
$C_2H_6$ fraction	%Vol	5.4
$C_3H_8$ fraction	%Vol	1.87
$C_4H_{10}$ fraction	%Vol	0.58
$C_5H_{12}$ fraction	%Vol	0.14
$\rm CO_2$ fraction	% Vol	1.79
$N_2$ fraction	% Vol	4.01
$O_2$ fraction	% Vol	0.21
Molecular Weight	$\left[\frac{\text{kg}}{\text{kmol}}\right]$	18.661
Density at $273.15[K]$ , $1[atm]$	$\left[\frac{\text{kg}}{\text{m}^3}\right]$	0.8325
LHV	$\left[\frac{kJ}{kg}\right]$	44454

Table 3.5: Properties of the natural gas used in the OXYFLAM-2 experiments as reported by Lallemant et al. [1997].

used during measurements and the yellow glow was faint the soot was determined to exist in trace amounts. A thin layer of soot was found on the burner at the conclusion of experiments. The total heat transfer rate to walls was determined from the flow rate of cooling water and the measured temperature rise. The burner head reached a temperature of 550 [°C]. The side and top wall temperatures were measured with 15 type B thermocouples at various axial distances from the burner. The temperatures measured on the top wall were 170 [°C] higher compared to the side wall temperatures. This indicates that the flame was inclined upwards, likely due to buoyancy. The flue gas temperature and composition were measured at the points indicated in Figure 3.1. The natural gas and oxygen flow rates, furnace heat extraction, wall and flue gas temperature and the flue gas composition were measured at a rate of 1 [Hz] and averaged over 60 [s].

In flame measurements were performed for velocity, velocity fluctuation, temperature and volume fractions of  $CH_4$ ,  $O_2$ , CO,  $H_2$ ,  $N_2$ ,  $CO_2$ , NO and NOx. Measurements were made in the radial direction from the furnace centerline at 0.22, 0.82 and 1.42[m] downstream of the burner. Species measurements were also performed at 2.21 [m] downstream of the burner. The measurements were in general made at radial distances up to 0.45 [m] at intervals increasing from 0.01 [m] at the centerline to 0.05 [m] at the radial distance 0.45 [m]. Additionally, a few measurements close to the centerline were made in the opposite radial direction. Measurements of velocity and velocity fluctuations were performed with laser Doppler velocimetry (LDV). The required seeding for LDV measurements was performed with zirconium oxide particles with a size range of 2-8 [ $\mu$ m]. Zirconium oxide has a melting point of 2980 [°C] and is therefore suited for high temperature flows. In flame temperature measurements were performed with a double venturi suction pyrometer equipped with a type B thermocouple. The suction pyrometer was water cooled. This caused the temperature of the gas sampled to drop before the temperature measurement. The suction pyrometer therefore required calibration. This was done with coherent anti-Stokes Raman spectroscopy (CARS) temperature measurements. The calibration curve developed extends to approximately 1650 [K] for the suction pyrometer measurement corresponding to a CARS temperature of approximately 2200 [K]. The measurements presented by Lallemant et al. [1997] are not corrected. Bollettini et al. [1997] presents corrected temperature measurements and these measurements have been used in this report for comparison. Bollettini et al. [1997] extrapolated the calibration curve for use at higher temperatures. This likely introduces errors in the temperature. Bollettini et al. [1997] also noted that the double venturi suction pyrometer is difficult to use and the accuracy of the instrument is difficult to assess. Gas species measurements were performed with a gas sampling probe. The water vapor was condensed and analysis of the gas sample was performed with non dispersive infrared sensor (NDIR) and electron paramagnetic resonance (EPR) spectroscopy, chemiluminescence and flame ionization detector (FID) depending upon the gas species to be determined. The specific analyzer used, operating range and accuracy can be found in Lallemant et al. [1997]. Further details of measurement techniques used can be found in Lallemant et al. [2003].

### 3.2 Mesh

Computational meshes of the interior furnace volume have been constructed for use in the CFD simulations. The dimensions used for the meshes are given in Table 3.1. As no data is available on the length of the chimney a length of 0.8 [m] has been used. Similarly the end of the burner has been assumed to be flush with the wall due to the lack of data. From the diagram of the furnace in Figure 3.1 and the dimensions in Table 3.1 it is apparent that the vertical and horizontal midplanes and the diagonal planes of the furnace are planes of geometrical symmetry as illustrated in Figure 3.3. If flow and other conditions in the furnace are also symmetrical, simulation of conditions in the furnace can be reduced to simulation of a region in the furnace bounded by symmetry planes and the furnace walls. Conditions in the rest of the furnace are determined by mirroring the results obtained in the simulated region. This approach reduces the dimensions of the mesh and thereby the number of cells and the computing time required to obtain simulation results. Conversely it is possible to increase the cell number density of the mesh of the delimited furnace region and obtain the same computation time as for a mesh of the entire furnace with a lower cell number density. The flow in the furnace is not symmetric with respect to the horizontal midplane due to the upward tilting of the flame discussed in Section 3.1.1. The horizontal asymmetry is assumed to be small enough to employ the horizontal symmetry plane with minimal error being introduced in the simulation results as a consequence. This is based on the reported difference of 170 [K] between the side and top wall which is small relative to the wall temperatures ranging between 1700-1900[K] as reported by Lallemant et al. [1997]. Differences in other conditions are assumed to minimal. Due to the limited number of measurements in the opposite radial direction, it is not possible to determine definitively whether furnace conditions are symmetrical with respect to the vertical midplane. The measurements available suggest symmetry exists and this is therefore assumed to be the case. Diagonal symmetry is dependent upon both horizontal and vertical symmetry. Both the horizontal and vertical symmetry of furnace conditions assumed are therefore employed and simulation has been limited to a quarter of the furnace. The top left quarter has been used in CFD simulations. The error introduced by the symmetry assumption can be determined with simulation using a mesh of the entire furnace with buoyancy included. The assumption of symmetry has not been validated. Breussin et al. [2000] and Mancini et al. [2002] have used the horizontal and vertical symmetry plane assumption in CFD simulations of the IFRF MILD combustion experiments. This is taken as further indication of the small error associated with the symmetry assumption.

The mesh with fewest cells constructed for this furnace quarter, with 304044 cells and designated q, is shown in Figure 3.4. All other meshes constructed have the same structure and the same boundary names shown in Figure 3.4. The mesh centerline is aligned with the z axis. The meshes have been constructed using ANSYS ICEM CFD 12.0.1. All meshes are constructed with the block structured approach and consist of hexahedral cells. Hexahedral cells can be aligned with the flow, as opposed in general to tetrahedral cells, and this minimizes numerical diffusion. The block structured approach consists of subdividing the geometry in to blocks. The blocks are then subdivided with hexahedral cells. Blocks can viewed as the first level geometry discretization. A central block is used for the chimney and extends to the bottom wall. This can be seen in Figure 3.5 showing the bottom wall. The largest circle in the mesh has the



Figure 3.3: Furnace geometrical symmetry planes.

chimney diameter. The burner inlets are visible in the bottom right corner. Three blocks are used for the surrounding region of the furnace. The central block is further subdivided with four ogrid blocks for the burner. The mesh burner inlets and wall are shown in Figure 3.6. The cells are concentrated at the centerline where the flame is located and, as a consequence, large gradients in flow properties exist. More cells are therefore concentrated here in order to more accurately resolve the gradients. The growth ratio of the distance between cell nodes as a rule of thumb should be smaller than 1.2. Large growth ratios cause errors in the evaluation of gradients as discussed by Casey et al. [2000]. The largest growth ratio exists in the radial direction of the circular portion of the mesh shown in Figure 3.5 for the meshes constructed. The growth ratio in this region is greatest for mesh q where the ratio is 1.07. The ratio decreases in this region for the meshes constructed in order of increasing mesh cell number.



Figure 3.4: Mesh of the top left quarter of the OXYFLAM-2 furnace. Labels indicate the boundary names.

In general the accuracy of simulations increases as the number of mesh cells increases. As described in Section 6.8, a mesh independence study is required in order to determine



Figure 3.5: Mesh of the furnace bottom with the burner inlets visible in the bottom right of the mesh. Labels indicate the boundary names.



Figure 3.6: Close up of the burner nozzle mesh. Labels indicate the boundary names. The nozzle boundary is a wall. The lowest quality cells are highlighted in light blue.

Mesh	Cells	Min. angle	Max. cell squish	Max. aspect ratio
q al	$304044 \\ 397854$	$45^{\circ}$ , 2 cells $45^{\circ}$ , 2 cells	$0.210 \\ 0.197$	$29.3 \\ 29.3$
$q^2$	510804	$45^{\circ}, 2$ cells	0.197	21.1
q3	600132	$45^{\circ}, 2$ cells	0.214	24.5
q4	799632	$45^{\circ}, 2 \text{ cells}$	0.214	24.5

Table 3.6: Mesh cell number and mesh quality.

a mesh which yields a mesh independent simulation. Meshes with increasing number of mesh cells have been constructed for the purpose of the mesh independence study. The meshes are designated q through q4 in order of increasing number of mesh cells. The number of mesh cells for each mesh is given in Table 3.6. All meshes use the same number of radial nodes for the natural gas and oxygen inlets and the nozzle. The number of nodes in the axial, radial and angular directions have been varied. Node increase in the angular direction has the greatest effect on the number of mesh cells. The quality of the meshes constructed have been evaluated by minimum angle, cell squish and aspect ratio for the mesh cells. The value corresponding to the lowest quality cell found in the mesh and number of cells with this value are given in Table 3.6. The minimum angle is determined from the minimum internal angle. Casey et al. [2000] recommends that cell angles should not be less than 40°. The minimum angle found for all meshes is 45° and the location of these cells are highlighted in Figure 3.6. The aspect ratio is computed as the ratio of the maximum value to the minimum value of the distances between the cell centroid and face centroids, and the distances between the cell centroid and nodes as stated by ANSYS Inc. [2009]. The maximum aspect ratio varies from 21.1 to 29.3. ANSYS Inc. [2009] recommends that the aspect ratio not exceed 5 with a ratio smaller than 35 recommended for the energy equation. The cells are aligned along the centerline. The cells with the maximum aspect ratios are located in the chimney where the flow is of least interest. As the flow is predominantly oriented along the centerline, the large cell aspect ratios with respect to the recommendations are assumed to be acceptable. The cell squish is determined from Eq. 3.1.

Cell squish index = max 
$$\left(1 - \frac{\mathbf{A}_i \cdot \mathbf{r}_{co/xf_i}}{|\mathbf{A}_i| |\mathbf{r}_{co/xf_i}|}\right)$$
 (3.1)

where  $\mathbf{A}_i \begin{bmatrix} \mathbf{m}^2 \end{bmatrix}$  is the cell face area vector and  $\mathbf{r}_{co/xf_i}$  [m] is the cell centroid to cell face centroid vector. The lowest quality cell has a value of 1 and the highest quality a value of 0. ANSYS Inc. [2009] states that the maximum cell squish should be lower than 0.99. This is the case for all meshes constructed.

# Chapter 4

# Chemistry

In this chapter the combustion chemistry is discussed. Equilibrium calculations of adiabatic flame temperatures and composition of oxy-natural gas combustion with dissociation are discussed. The reaction mechanism investigated and used in the CFD simulations are presented. Finally, the modelling of natural gas is explained.

### 4.1 Oxy-Natural Gas Equilibrium Calculations

As discussed in Section 1.2, oxy-fuel combustion involves using oxygen instead of air as the oxidizer. The reaction for stoichiometric combustion of CH<sub>4</sub> with air is given in Eq. 4.1. From an energy balance of this reaction the constant pressure adiabatic flame temperature without dissociation is determined to be 2329 [K]. The adiabatic flame temperature for the stoichiometric combustion of CH<sub>4</sub> with oxygen given in Eq. 4.2, is determined in the same manner to be 5241 [K]. This illustrates the significantly higher temperatures reached in oxy-fuel combustion. This is due to the lack of essentially inert N<sub>2</sub> which is heated in air combustion thus lowering the temperature. In addition, CO<sub>2</sub> and H<sub>2</sub>O are the only products in oxy-fuel combustion which alters the constant pressure specific heat capacity  $c_p \left[\frac{kJ}{kg\cdot K}\right]$  from 1.5230  $\left[\frac{kJ}{kg\cdot K}\right]$  for CH<sub>4</sub>-air combustion at the adiabatic flame temperature.

$$CH_4 + 2(O_2 + 3.71N_2) \rightarrow 2H_2O + CO_2 + 7.43N_2$$
 (4.1)

$$CH_4 + 2O_2 \rightarrow 2H_2O + CO_2 \tag{4.2}$$

#### 4.1.1 Adiabatic Flame Temperature

The adiabatic flame temperatures determined by energy balance of the reactions in Eq. 4.1 and Eq. 4.2 do not take into account the effect of dissociation that becomes increasingly important as the temperature increases. The ideal stoichiometric combustion products  $CO_2$  and  $H_2O$ , and  $N_2$  in the case of air combustion, dissociate to yield species that are compounds of the elements present in the reaction products. The

dissociation species are typically CO, O<sub>2</sub>, H<sub>2</sub>, OH, H, O, N, NO and possibly others depending on the conditions. Dissociation is a consequence of the second law of thermodynamics which requires dS > 0. The ideal stoichiometric reaction does not yield the maximum system entropy. The maximum entropy exist for system comprised of both ideal reaction products and dissociation species as discussed by Turns [2006]. The composition of the combustion products will shift towards the maximum entropy and once maximum entropy is reached no further changes can take place due to the second law of thermodynamics and the system is at a state of equilibrium. The adiabatic flame temperature with dissociation included is lower compared to the adiabatic flame temperature where dissociation is neglected. This is a consequence of the species of dissociation, in general, having a higher enthalpy of formation than the ideal products. The equilibrium adiabatic flame temperature for a specified furnace inlet flow rate of species provides an estimate of the maximum flame temperature and the species and mole fractions at this maximum temperature. The actual flame temperature is lower than the adiabatic flame temperature and the equilibrium composition for the specified furnace inlet flow rate of species as a function of temperature can be used as more accurate estimate of the species and mole fraction conditions in the furnace. Equilibrium calculations do not include reaction kinetics and therefore fail to take into account the time required for the equilibrium composition to be reached. Species that are products of dissociation reactions with slow reaction rates are therefore likely to be over predicted and, as a consequence, the dissociation reaction reactants are under predicted.

The constant pressure adiabatic flame temperatures at 1 [bar] for air-NG and oxy-NG combustion as a function of equivalence ratio  $\Phi$ , determined from equilibrium calculations using the Chemical Equilibrium with Applications (CEA) software, are shown in Figure 4.1. All species are included in the equilibrium calculations. Details of the calculations performed by CEA are presented by Gordon and McBride [1994] and the software manual and program structure is provided by McBride and Gordon [1996]. The equivalence ratio  $\Phi$ , given in Eq. 4.3, is the air-fuel ratio relative to the stoichiometric air-fuel ratio.

$$\Phi = \frac{\frac{m_F}{m_O}}{\left(\frac{m_F}{m_O}\right)_{\text{stoich}}} = \frac{\left(\frac{m_O}{m_F}\right)_{\text{stoich}}}{\frac{m_O}{m_F}}$$
(4.3)

where  $m_O [kg]$  is the oxidizer mass and  $m_F [kg]$  is the fuel mass. From this definition it follows that  $\Phi = 1$  corresponds to stoichiometric conditions,  $\Phi < 1$  to fuel lean conditions and  $\Phi > 1$  to fuel rich conditions. The oxy-NG adiabatic flame temperatures as a function of  $\Phi$  with equilibrium calculation species limited to the species in the Westbrook-Dryer (WD) and Jones-Lindstedt (JL) reaction mechanism respectively, are also shown in Figure 4.1. The WD mechanism reactions include the species CH<sub>4</sub>, O<sub>2</sub>, H<sub>2</sub>O, CO<sub>2</sub>, and CO. The JL mechanism includes the same species as the WD mechanism with the addition of H<sub>2</sub> for the case of CH<sub>4</sub> combustion. The composition of the natural gas used in the OXYFLAM-2 experiments is used and is given in Table 3.5. For reference, the OXYFLAM-2 experiments operated at  $\Phi = 0.9871$  based on calculations using the operating conditions given in Table 3.4 and the natural gas composition.

From Figure 4.1 it is evident that the adiabatic flame temperature of oxy-NG combustion is approximately 1000 [K] higher than that of air-NG combustion with the



Figure 4.1: Adiabatic flame temperature at various  $\Phi$  for air-NG combustion, oxy-NG, oxy-NG with WD species and oxy-NG with JL species.

difference being smallest between a  $\Phi$  value of 1-1.05. The adiabatic flame temperature for air-NG is at a maximum at the same  $\Phi$  interval and drops markedly for fuel lean and fuel rich conditions. The adiabatic flame temperature of oxy-NG is at a maximum for  $\Phi$  in the interval of 1.05-1.15 and remains relatively constant at all  $\Phi$  unlike the air-NG adiabatic flame temperature. The adiabatic flame temperature for oxy-NG combustion with the species limited to those present in the Westbrook-Dryer mechanism shows a significant error compared to oxy-NG with all species included in the equilibrium calculation. The temperature at  $\Phi = 0.6$  is 500 [K] higher for the oxy-NG WD calculations and the difference increases with  $\Phi$  to 1000 [K] at  $\Phi = 1.35$ . The adiabatic flame temperature for oxy-NG lacks the drop in temperature exhibited at fuel rich conditions for oxy-NG with all species. The oxy-NG adiabatic flame temperature with species limited to those present in the Jones-Lindstedt mechanism also exhibits higher temperatures in the whole range of  $\Phi$ . The adiabatic flame temperature for the oxy-NG JL calculations is markedly lower compared to the oxy-NG WD case. The inclusion of  $H_2$  lowers the difference between the oxy-NG JL and oxy-NG temperatures to 250 [K] and the difference is relatively constant. The corollary of the constant difference is that the oxy-NG JL temperatures exhibit the drop in temperature at fuel lean and rich conditions shown by the oxy-NG results.

It can be concluded that limiting the species to those in the WD mechanism leads to an over prediction of the adiabatic flame temperature of 1000[K]. A better prediction of the adiabatic flame temperature is obtained when adding H<sub>2</sub>, included in the JL mechanism. This indicates dissociation of H<sub>2</sub> is significant. However, the JL mechanism still does not account for other significant dissociation species and therefore still predicts a higher adiabatic flame temperature compared to the actual adiabatic flame temperature. This

indicates that both the WD and JL schemes are likely to over predict the furnace temperature, and major species. Both schemes neglect significant dissociation species.

#### 4.1.2 Adiabatic Equilibrium Composition

The significant species mole fractions for adiabatic oxy-NG combustion as a function of  $\Phi$  are shown in Figure 4.2. All species are included in the equilibrium calculations. H<sub>2</sub>O and CO<sub>2</sub> are main species for the entire  $\Phi$  range as is expected from the ideal reaction of oxy-NG. The H<sub>2</sub>O mole fraction,  $X_{H_2O}$ , is relatively constant ranging from 0.3 to 0.4.  $X_{CO_2}$  decreases steadily from 0.15 to 0.08 between  $\Phi = 0.6$  to 1.4. The O<sub>2</sub> mole fraction is large in the fuel lean range as expected. In the fuel rich range  $X_{O_2} = 0.082$  at  $\Phi = 1$  where the ideal reaction predicts  $X_{O_2} = 0$  and at  $X_{O_2} = 0.013$  at  $\Phi = 1.4$ . OH is a significant dissociation species and remains relatively constant decreasing from  $X_{OH} = 0.093$  at  $\Phi = 0.6$  to  $X_{OH} = 0.056$  at  $\Phi = 1.4$ .  $X_{CO}$ ,  $X_{H_2}$  and  $X_H$  increase with  $\Phi$ .  $X_{CO}$  becomes a dominant species for  $\Phi > 0.85$  and is significant at all  $\Phi$ . The radical O is most significant in fuel lean conditions and decreases significantly along with with O<sub>2</sub> in fuel rich conditions. The mole fractions of N<sub>2</sub> and NO are the smallest with N<sub>2</sub> increasing and NO decreasing in the transition from fuel lean to fuel rich conditions. It can be concluded that for the adiabatic equilibrium at  $\Phi = 1$ , H<sub>2</sub>O, CO, CO<sub>2</sub>, OH, O<sub>2</sub>, H<sub>2</sub>, H and O, in order of decreasing mole fraction, are the main species.



Figure 4.2: Adiabatic equilibrium composition of oxy-NG combustion as a function of  $\Phi$ .

#### 4.1.3 Stoichiometric Equilibrium Composition

As the temperature in the furnace is lower than the adiabatic temperature, the equilibrium composition of stoichiometric oxy-NG combustion as a function of temperature has been calculated with CEA and is shown in Figure 4.3. This provides information on the significant species present at equilibrium for the lower temperatures encountered in the furnace. The peak temperature measured, as reported by Bollettini et al. [1997], is 2770 [K] and the lowest temperature measured is 1800 [K] at the side walls. The furnace temperatures are therefore assumed to be within this range.



Figure 4.3: Equilibrium composition of stoichiometric oxy-NG combustion as a function of T.

In general it is found that dissociation increases with temperature. This causes the two main species  $H_2O$  and  $CO_2$  to dissociate and  $X_{H_2O}$  and  $X_{CO_2}$  decrease.  $X_{H_2O}$  and  $X_{CO_2}$  remain constant up to 2500 [K]. From 2500-3500[K],  $X_{H_2O}$  and  $X_{CO_2}$  decrease from 0.58 and 0.27 to 0.14 and 0.03 respectively. The mole fractions of the species CO,  $O_2$ , OH and  $H_2$  all increase with temperature and show the same mole fraction gradient with respect to temperature in the entire temperature range.  $X_{CO}$ ,  $X_{O_2}$ ,  $X_{OH}$  and  $X_{H_2}$  are on the order of magnitude of  $10^{-3}$  below 2000 [K] and increase to 0.01 in the temperature interval 2000-2500[K].  $X_{CO}$  increases to 0.1 at 2750[K]. This is the same order of magnitude as  $X_{H_2O}$  and  $X_{H_2O}$  at the same temperature. Above 3000 [K]  $X_{CO}$  is greater than  $X_{CO_2}$  and this also becomes the case for  $X_{O_2}$ ,  $X_{OH}$  and  $X_{H_2}$  in the temperature interval 3000-3500[K]. The mole fraction of the free radical H and O also increase with temperature and show the same gradient with respect to temperature.  $X_{H}$  and  $X_{O}$  become equal  $10^{-2}$  at 2750[K] increasing above this temperature to equal the other dissociation products.  $X_{N_2}$  is relatively constant in the whole temperature interval.  $X_{NO}$  approaches  $X_{N_2}$  with increasing temperature. Based on relative mole

fractions in the temperature interval 1500-2500[K] the species  $H_2O$ ,  $CO_2$ , CO,  $H_2$ ,  $O_2$ , OH and  $N_2$  are found to be significant.

### 4.2 Reaction Mechanism

Combustion occurs through a number of elementary reactions that together lead to the overall reaction. Elementary reactions together constitute a reaction mechanism. Detailed mechanisms for the combustion  $CH_4$  can include thousands of elementary reactions as discussed by Turns [2006]. Detailed mechanisms are in general therefore not applicable to CFD simulations due to the computation time involved to solve the large system of differential equations associated with such a mechanism when reaction kinetics are included. Global and quasi global mechanisms seek to model the overall behaviour of the detailed reaction mechanism with a reduced set of reactions. The Westbrook-Dryer (WD) and Jones-Lindstedt (JL) are multi step reaction mechanisms often used in combustion modelling of hydrocarbons. The WD mechanism, given in Table 4.1, is a two step reaction mechanism for air combustion of  $CH_4$  with the final reaction being reversible. The JL mechanism, given in Table 4.5, is a four step reaction for air combustion of  $CH_4$ ,  $C_2H_6$ ,  $C_3H_8$  and  $C_4H_{10}$  with the final two steps being reversible. Both mechanisms model the combustion reaction kinetics. Andersen et al. [2009] modified both the WD and JL mechanisms in order for the mechanisms to better model oxy-fuel combustion as both mechanisms were originally developed for combustion in air. The main effort was to improve model predictions of the  $X_{\rm CO}$ trend and equilibrium value. Andersen et al. [2009] retained the initiating reactions involving hydrocarbons and modified the  $H_2$ -CO-CO<sub>2</sub> reactions. The modified WD mechanism was found to provide a better prediction of CO and temperature while the modified JL mechanism provided slightly better CO predictions in the flame according to Andersen et al. [2009]. Based on the equilibrium calculations and discussion of results in Section 4.1 the JL mechanism is expected to yield more accurate predictions of temperature and species compared to the WD mechanism. Both mechanisms are expected to over predict the temperature. The WD, modified WD have been used in the CFD simulations performed. It has not been possible to use the JL and modified JL mechanisms due to an error in the chemistry solver in the CFD solver FLUENT used to perform the CFD simulations.

Equilibrium calculations are the limiting case for reaction kinetic calculations. The residence time of the furnace species is finite and the equilibrium composition is therefore not reached. Modelling of the reaction rates takes into account the time dependency of the reaction mechanism. The mole reaction rate of species *i* due to reaction *r* is proportional to reactant concentrations, the reation rate constant and the stoichiometric coefficient of the species. The mole reaction rate,  $\overline{R}_{i,r}$  [ $\frac{\text{kmol}}{\text{m}^3 \cdot \text{s}}$ ] is determined from Eq. 4.4.

$$\bar{R}_{i,r} = \frac{d\left[X_{i,r}\right]}{dt} = \left(\nu_{i,r}'' - \nu_{i,r}'\right) \left(k_{f,r} \prod_{j=1}^{N} \left[X_{j,r}\right]^{\eta_{j,r}'} - k_{b,r} \prod_{j=1}^{N} \left[X_{j,r}\right]^{\eta_{j,r}''}\right)$$
(4.4)

where  $[X_{i,r}]$   $\left[\frac{\text{kmol}}{\text{m}^3}\right]$  is the mole concentration of species *i* in reaction *r*,  $\nu'_{i,r}$  and  $\nu''_{i,r}$  are the reactant and product stoichiometric coefficients of species *i* in reaction *r* re-

spectively,  $\eta'_{j,r}$  and  $\eta''_{j,r}$  are the rate exponents of the reactant and product species j in reaction r respectively,  $[X_{j,r}] \left[\frac{\text{kmol}}{\text{m}^3}\right]$  is the mole concentration of spieces j in reaction r and N is the number of species in reaction r. The forward and backward reaction rate constants  $k_{f,r}$  and  $k_{b,r}$  are usually evaluated with the Arrhenius equation given in Eq. 4.5.

$$k_{f,r} = A_r T^{\beta_r} \exp\left(-\frac{E_A}{R_u T}\right) \tag{4.5}$$

where  $A_r$  is the pre-exponential constant of reaction r, T [K] is the temperature,  $\beta_r$  is the temperature exponent of reaction r,  $E_A \begin{bmatrix} \frac{J}{\text{kmol}} \end{bmatrix}$  is the activation energy of recation r and  $R_u \begin{bmatrix} \frac{J}{\text{kmol}\cdot\text{K}} \end{bmatrix}$  is the gas constant. The mass reaction rate of species i in reaction r,  $R_{i,r} \begin{bmatrix} \frac{\text{kg}}{\text{m}^3\cdot\text{s}} \end{bmatrix}$  is determined with Eq. 4.6.

$$R_{i,r} = \overline{R}_{i,r} M_i \tag{4.6}$$

If the reaction is reversible  $k_{b,r}$  is determined from the equilibrium constant  $K_r$  for the reaction r using Eq. 4.7 unless a distinct reverse reaction is specified. Determination of  $K_r$  is discussed by Turns [2006].

$$k_{b,r} = \frac{k_{f,r}}{K_r} \tag{4.7}$$

Jones and Lindstedt [1988] states that a requirement for reaction mechanisms is mathematically tractability. Rate exponents that are negative should be avoided entirely and exponents should be close to unity or higher. Negative rate exponents of concentrations approaching zero will cause reaction rates to increase rapidly and approach infinity. The requirements for rate exponents for mathematical stability is often in conflict with the required exponents for accuracy. The WD and modified WD mechanisms both have reaction exponents less than unity and the modified WD2r reaction exponent for  $[O_2]$  is -0.25, a negative exponent less than absolute unity. The JL3 reaction  $H_2O$ exponent -1 is also negative. Jones and Lindstedt [1988] provides an alternative, JL3 alternative, for the reaction JL3 where the exponents are all positive. An alternative WD2r reaction with a  $O_2$  exponent of 0 has been used to define the modified WD B mechanism to investigate the effects of the negative exponent. All other parameters for the modified WD B mechanism are taken from the modified WD mechanism.

FLUENT employs the SI unit system. The values given in Table 4.5, 4.6, 4.1 and 4.2 are given in the units [cm], [s], [cal] and [mol] must therefore be converted. The activation energy  $E_A$  given by Jones and Lindstedt [1988] is given in  $\left[\frac{\text{cal}}{\text{mol}}\right]$  and not in  $\left[\frac{\text{cal}}{\text{kmol}}\right]$  as is stated by Jones and Lindstedt [1988]. This has been determined from the Andersen et al. [2009] and Kim et al. [2008]. The conversion factor for  $E_A$  is 4184  $\left[\frac{\text{J/kmol}}{\text{cal/mol}}\right]$ . The unit for the pre-exponential factor  $A_r$  depends on the reaction order, determined from the sum of rate exponents, in Eq. 4.4 and is a unit such that Eq. 4.4 yields the unit  $\left[\frac{\text{kmol}}{\text{m}^3\text{s}}\right]$ . SI units are used for  $A_r$  in the JL mechanism given in Table 4.5 and no conversion is required. The units [cm], [s], [cal] and [mol] are used for the WD, modified WD and modified JL mechanisms and unit conversion factors are not equal as the reaction orders for the reactions in the mechanisms are not equal. The
unit conversion factors must therefore be determined individually for each reaction. As an example the unit conversion calculation for the modified WD2r reaction  $A_r$  is presented in Eq 4.8. For the modified WD2r  $A_r$  has the units  $\left[\left(\frac{\text{mol}}{\text{cm}^3}\right)^{-0.25} \cdot \text{s}^{-1} \cdot \text{K}^{0.97}\right]$  in order for Eq. 4.4 to have the units  $\left[\frac{\text{mol}}{\text{cm}^3 \cdot \text{s}}\right]$ .  $A_r$  must therefore be converted to the unit  $\left[\left(\frac{\text{kmol}}{\text{m}^3}\right)^{-0.25} \cdot \text{s}^{-1} \cdot \text{K}^{0.97}\right]$  for use in FLUENT.

$$6.16 \cdot 10^{13} \left[ \left( \frac{\text{mol}}{\text{cm}^3} \right)^{-0.25} \cdot \text{s}^{-1} \cdot \text{K}^{0.97} \right] \cdot \left( \frac{1}{1000} \left[ \frac{\text{kmol}}{\text{mol}} \right] \right)^{-0.25} \\ \cdot \left( 100^3 \left[ \frac{\text{cm}^3}{\text{m}^3} \right] \right)^{-0.25} = 1.095 \cdot 10^{13} \left[ \left( \frac{\text{kmol}}{\text{m}^3} \right)^{-0.25} \cdot \text{s}^{-1} \cdot \text{K}^{0.97} \right]$$
(4.8)

The same approach has been used for the remaining  $A_r$  unit conversions. The results of the unit conversions of  $E_A$  in the JL mechanism and the parameter values in Table 4.1, 4.2 and 4.6 are given in Table 4.5, 4.3, 4.4 and 4.7 respectively.

Reaction no.	Reaction	$A_r$	$\beta_r$	$E_A\left[\frac{\mathrm{cal}}{\mathrm{mol}}\right]$	Reaction orders
WD1	$\mathrm{CH}_4 + \tfrac{3}{2}\mathrm{O}_2 \rightarrow \mathrm{CO} + 2\mathrm{H}_2\mathrm{O}$	$1.59\cdot 10^{13}$	0	$47.8\cdot 10^3$	$[\rm{CH}_4]^{0.7}  [\rm{O}_2]^{0.8}$
WD2	$\mathrm{CO} + \frac{1}{2}\mathrm{O}_2 \to \mathrm{CO}_2$	$3.98\cdot 10^{14}$	0	$40.7\cdot 10^3$	$[CO] [O_2]^{0.25} [H_2O]^{0.5}$
WD2r	$CO_2 \rightarrow CO + \frac{1}{2}O_2$	$5.0\cdot 10^8$	0	$40.7\cdot 10^3$	$[\mathrm{CO}_2]$

Table 4.1: Westbrook-Dryer mechanism given by Andersen et al. [2009]. Quantities given in [cm], [s], [cal] and [mol].

Reaction no.	Reaction	$A_r$	$\beta_r$	$E_A\left[\frac{\mathrm{cal}}{\mathrm{mol}}\right]$	Reaction orders
Mod. WD1	$\mathrm{CH}_4 + \tfrac{3}{2}\mathrm{O}_2 \rightarrow \mathrm{CO} + 2\mathrm{H}_2\mathrm{O}$	$1.59\cdot 10^{13}$	0	$47.8\cdot 10^3$	$\left[\mathrm{CH}_4\right]^{0.7} \left[\mathrm{O}_2\right]^{0.8}$
Mod. $WD2$	$\mathrm{CO} + \frac{1}{2}\mathrm{O}_2 \to \mathrm{CO}_2$	$3.98\cdot 10^8$	0	$10.0\cdot 10^3$	$[CO] [O_2]^{0.25} [H_2O]^{0.5}$
Mod. WD2r	$\mathrm{CO}_2  \mathrm{CO} + \frac{1}{2}\mathrm{O}_2$	$6.16\cdot10^{13}$	-0.97	$78.4 \cdot 10^3$	$[CO_2] [H_2O]^{0.5} [O_2]^{-0.25}$

Table 4.2: Modified Westbrook-Dryer mechanism given by Andersen et al. [2009]. Quantities given in [cm], [s], [cal], [K] and [mol].

Reaction no.	Reaction	$A_r$	$\beta_r$	$E_A \left[ \frac{\mathrm{J}}{\mathrm{kmol}} \right]$	Reaction orders
WD1	$\mathrm{CH}_4 + \tfrac{3}{2}\mathrm{O}_2 \rightarrow \mathrm{CO} + 2\mathrm{H}_2\mathrm{O}$	$5.028\cdot 10^{11}$	0	$2.001\cdot 10^8$	$[\rm{CH}_4]^{0.7}  [\rm{O}_2]^{0.8}$
WD2	$\mathrm{CO} + \frac{1}{2}\mathrm{O}_2 \to \mathrm{CO}_2$	$2.238\cdot10^{12}$	0	$1.704\cdot 10^8$	$[CO] [O_2]^{0.25} [H_2O]^{0.5}$
WD2r	$CO_2 \rightarrow CO + \frac{1}{2}O_2$	$5 \cdot 10^8$	0	$1.704\cdot 10^8$	$[\mathrm{CO}_2]$

Table 4.3: Westbrook-Dryer mechanism given by Andersen et al. [2009] in SI units. Quantities given in [m], [s], [J], [K] and [kmol].

Reaction no.	Reaction	$A_r$	$\beta_r$	$E_A \left[ \frac{\mathrm{J}}{\mathrm{kmol}} \right]$	Reaction orders
Mod. WD1	$\mathrm{CH}_4 + \tfrac{3}{2}\mathrm{O}_2 \rightarrow \mathrm{CO} + 2\mathrm{H}_2\mathrm{O}$	$5.028\cdot10^{11}$	0	$2.001\cdot 10^8$	$[CH_4]^{0.7} [O_2]^{0.8}$
Mod. $WD2$	$\mathrm{CO} + \frac{1}{2}\mathrm{O}_2 \to \mathrm{CO}_2$	$2.238\cdot 10^6$	0	$4.186\cdot 10^7$	$[CO] [O_2]^{0.25} [H_2O]^{0.5}$
Mod. WD2r	$\mathrm{CO}_2 \rightarrow \mathrm{CO} + \frac{1}{2}\mathrm{O}_2$	$1.095 \cdot 10^{13}$	-0.97	$3.282\cdot 10^8$	$[CO_2] [H_2O]^{0.5} [O_2]^{-0.25}$

Table 4.4: Modified Westbrook-Dryer mechanism given by Andersen et al. [2009] in SI units. Quantities given in [m], [s], [J], [K] and [kmol].

Reaction no.	Reaction	n	$A_r$	$\beta_r$	$E_A \left[\frac{\mathrm{cal}}{\mathrm{mol}}\right]$	Reaction orders
JL1	$\mathbf{C_nH_{2n+2}} + \tfrac{n}{2}\mathbf{O_2} \rightarrow n\mathbf{CO} + (n+1)\mathbf{H_2}$	1	$0.44\cdot 10^{12}$	0	$1.2558\cdot 10^8$	$\left[\mathrm{C_{n}H_{2n+2}}\right]^{\frac{1}{2}}\left[\mathrm{O_{2}}\right]^{\frac{5}{4}}$
		2	$0.42\cdot 10^{12}$	0	$1.2558\cdot 10^8$	$\left[ C_{n}H_{2n+2} \right]^{\frac{1}{2}} \left[ O_{2} \right]^{\frac{5}{4}}$
		3	$0.40\cdot 10^{12}$	0	$1.2558\cdot 10^8$	$\left[ C_{n}H_{2n+2} \right]^{\frac{1}{2}} \left[ O_{2} \right]^{\frac{5}{4}}$
		4	$0.38\cdot 10^{12}$	0	$1.2558\cdot 10^8$	$\left[C_{n}H_{2n+2}\right]^{\frac{1}{2}}\left[O_{2}\right]^{\frac{5}{4}}$
JL2	$\mathrm{C_nH_{2n+2}} + n\mathrm{H_2O} \rightarrow n\mathrm{CO} + (2n+1)\mathrm{H_2}$	1-4	$0.30\cdot 10^9$	0	$1.2558\cdot 10^8$	$\left[\mathrm{C_{n}H_{2n+2}}\right]\left[\mathrm{H_{2}O}\right]$
JL3	$H_2 + \frac{1}{2}O_2 \rightleftharpoons H_2O$	1	$0.25\cdot 10^{17}$	-1	$1.6744\cdot 10^8$	$[{\rm H}_2]^{\frac{1}{2}}  [{\rm O}_2]^{\frac{9}{4}}  [{\rm H}_2{\rm O}]^{-1}$
		2	$0.35\cdot 10^{17}$	-1	$1.6744\cdot 10^8$	$[\mathrm{H}_2]^{\frac{1}{2}} [\mathrm{O}_2]^{\frac{9}{4}} [\mathrm{H}_2\mathrm{O}]^{-1}$
		3	$0.30\cdot 10^{17}$	-1	$1.6744\cdot 10^8$	$[\mathrm{H}_2]^{\frac{1}{2}} [\mathrm{O}_2]^{\frac{9}{4}} [\mathrm{H}_2\mathrm{O}]^{-1}$
_		4	$0.28\cdot 10^{17}$	-1	$1.6744\cdot 10^8$	$[\mathrm{H}_2]^{\frac{1}{2}} [\mathrm{O}_2]^{\frac{9}{4}} [\mathrm{H}_2\mathrm{O}]^{-1}$
JL3 alt.	$H_2 + \frac{1}{2}O_2 \rightleftharpoons H_2O$	1	$0.68\cdot 10^{16}$	-1	$1.6744\cdot 10^8$	$[{\rm H}_2]^{\frac{1}{4}}  [{\rm O}_2]^{\frac{3}{2}}$
		2	$0.90\cdot 10^{16}$	-1	$1.6744\cdot 10^8$	$[{\rm H}_2]^{\frac{1}{4}}  [{\rm O}_2]^{\frac{3}{2}}$
		3	$0.85\cdot 10^{16}$	-1	$1.6744\cdot 10^8$	$[\mathrm{H}_2]^{rac{1}{4}} [\mathrm{O}_2]^{rac{3}{2}}$
		4	$0.75\cdot 10^{16}$	-1	$1.6744\cdot 10^8$	$[{\rm H}_2]^{\frac{1}{4}}  [{\rm O}_2]^{\frac{3}{2}}$
JL4	$\rm CO + H_2O \rightleftharpoons \rm CO_2 + H_2$	1-4	$0.275\cdot 10^{10}$	0	$8.372\cdot 10^7$	$[CO][H_2O]$

Table 4.5: The Jones-Lindstedt global reaction mechanism for  $C_nH_{2n+2}$  for  $n \leq 4$  given by Jones and Lindstedt [1988]. Quantities given in [kg], [m], [s], [kmol], [cal], [K].  $E_A$  is given in [cal/mol].

Reaction no.	Reaction	$A_r$	$\beta_r$	$E_A\left[\frac{\mathrm{cal}}{\mathrm{mol}}\right]$	Reaction orders
Mod. JL1	$CH_4 + \frac{1}{2}O_2 \rightarrow CO + 2H_2$	$7.82\cdot 10^{13}$	0	$30.0\cdot 10^3$	$[CH_4]^{0.5} [O_2]^{1.25}$
Mod. JL2	$\rm CH_4 + \bar{H}_2O \rightarrow \rm CO + 3H_2$	$3.00\cdot10^{11}$	0	$30.0\cdot10^3$	$[CH_4][H_2O]$
Mod. JL3	$H_2 + \frac{1}{2}O_2 \rightarrow H_2O$	$5.0\cdot10^{20}$	$^{-1}$	$30.0\cdot10^3$	$\left[\mathrm{H}_{2}\right]^{0.25} \left[\mathrm{O}_{2}\right]^{1.5}$
Mod. JL3 reverse	$H_2O \rightarrow H_2 + \frac{1}{2}O_2$	$2.93\cdot 10^{20}$	-0.877	$97.9\cdot 10^3$	$[\mathrm{H}_2]^{-0.75} [\mathrm{O}_2] [\mathrm{H}_2\mathrm{O}]$
Mod. JL4	$\mathrm{CO} + \mathrm{H}_2\mathrm{O} \rightleftharpoons \mathrm{CO}_2 + \mathrm{H}_2$	$2.75\cdot 10^{12}$	0	$20.0\cdot 10^3$	$[\rm CO][\rm H_2O]$

Table 4.6: Modified Jones-Lindstedt combustion mechanism for  $CH_4$  given by Andersen et al. [2009]. Quantities given in [cm], [s], [cal], [K] and [mol].

Reaction no.	Reaction	$A_r$	$\beta_r$	$E_A \left[ \frac{\mathrm{J}}{\mathrm{kmol}} \right]$	Reaction orders
Mod. JL1	$CH_4 + \frac{1}{2}O_2 \rightarrow CO + 2H_2$	$4.3975 \cdot 10^{11}$	0	$1.2558\cdot 10^8$	$[\mathrm{CH}_4]^{0.5}  [\mathrm{O}_2]^{1.25}$
Mod. JL2	$CH_4 + H_2O \rightarrow CO + 3H_2$	$3.00\cdot 10^8$	0	$1.2558\cdot 10^8$	$[CH_4][H_2O]$
Mod. JL3	$\mathrm{H}_{2} + \tfrac{1}{2}\mathrm{O}_{2} \rightarrow \mathrm{H}_{2}\mathrm{O}$	$2.8117 \cdot 10^{18}$	-1	$1.2558\cdot 10^8$	$[\mathrm{H}_2]^{0.25} [\mathrm{O}_2]^{1.5}$
Mod. JL3 reverse	$H_2O \rightarrow H_2 + \frac{1}{2}O_2$	$5.2104 \cdot 10^{19}$	-0.877	$4.0979\cdot 10^8$	$[\mathrm{H}_2]^{-0.75} [\mathrm{O}_2] [\mathrm{H}_2\mathrm{O}]$
Mod. JL4	$\rm CO + H_2O \rightleftharpoons CO_2 + H_2$	$2.75\cdot 10^9$	0	$8.3717\cdot 10^7$	$[CO] [H_2O]$

Table 4.7: The modified Jones-Lindstedt reaction mechanism from Andersen et al. [2009] for  $CH_4$  in SI units. Quantities given in [m], [s], [J], [K], [kmol].

#### 4.3 Modelling Natural Gas

The OXYFLAM-2 experiments were conducted with natural gas with a composition as given in Table 3.5 as reported by Lallemant et al. [1997]. The natural gas was primarily comprised of  $\rm CH_4$  at 86 [%Vol] and the hydrocarbons  $\rm C_2H_6$  up to  $\rm C_5H_{12}$ comprising a combined 7.99 [%Vol] of the natural gas. The WD mechanism only deals with  $CH_4$  and there are no reaction rate relations for larger hydrocarbons as there are in the JL mechanism for  $C_2H_6$  to  $C_4H_{10}$  given in Table 4.5. It is not possible to attain accurate simulation results using exclusively  $CH_4$  to model the hydrocarbon portion of the natural gas in order to use the WD mechanism. This is evident from the  $\rm CH_4~LHV$ of 50016  $\left\lceil \frac{kJ}{kg} \right\rceil$ , as given by Turns [2006], when compared to the natural gas hydrocarbon LHV of 44235  $\begin{bmatrix} kJ \\ kg \end{bmatrix}$  determined from the natural gas composition. The correct mass flow rates of C and H are also not matched by the use of  $CH_4$  to model natural gas. This is due to  $CH_4$  having a higher H/C ratio than  $C_2H_6$  to  $C_5H_{12}$ . This would cause an error in the predicted species mole fractions in the furnace. A numerical fuel is therefore created to model the natural gas hydrocarbon fraction. The WD mechanisms are then used with the numerical fuel by replacing  $CH_4$  in the reactions and reaction rate relations with the numerical fuel. The WD1 and mod WD1 reactions are balanced with respect to the numerical fuel. The following calculations are performed in determining the properties of the numerical fuel used to model the hydrocarbon fraction of the natural gas used during the OXYFLAM-2 experiments.

- 1. Mass fraction of  $CH_4$ ,  $C_2H_6$ ,  $C_3H_8$ ,  $C_4H_{10}$  and  $C_5H_{12}$ .
- 2. Mass flow rate of  $\rm CH_4,\, C_2H_6,\, C_3H_8,\, C_4H_{10}$  and  $\rm C_5H_{12}.$
- 3. Mole flow rate of  $CH_4$ ,  $C_2H_6$ ,  $C_3H_8$ ,  $C_4H_{10}$  and  $C_5H_{12}$ .
- 4. Total flow rate of C and H due to the hydrocarbons.
- 5. H/C ratio of the numerical fuel.
- 6. Total heat release from combustion of the hydrocarbons.
- 7. Enthalpy of formation of the numerical fuel.

The mass fraction, Y of each of the species in the natural gas is determined using Eq. 4.9.

$$Y_i = X_i \frac{M_i}{M} \tag{4.9}$$

where  $Y_i$  is the mass fraction of species i,  $X_i$  is the mole fraction of species i,  $M_i \lfloor \frac{\text{kg}}{\text{kmol}} \rfloor$  is the molecular weight of species i and  $M \lfloor \frac{\text{kg}}{\text{kmol}} \rfloor$  is the mixture molecular weight. For an ideal gas the mole fraction of species i is equal to the volume fraction. M can be determined from Eq. 4.10.

$$M = \sum_{i}^{N} X_i M_i \tag{4.10}$$

The mass flow rate  $\dot{m}_i \left[\frac{\text{kg}}{\text{s}}\right]$  and mole flow rate  $\dot{n}_i \left[\frac{\text{kmol}}{\text{s}}\right]$  of each species *i* in the natural gas are determined with Eq. 4.11 and 4.12 respectively.

$$\dot{m}_i = Y_i \dot{m}_{NG} \tag{4.11}$$

$$\dot{n}_i = \frac{\dot{m}_i}{M_i} \tag{4.12}$$

where  $\dot{m}_{NG} \left[\frac{\text{kg}}{\text{s}}\right]$  is the mass flow rate of natural gas given in Table 3.4.  $M_i$ , M,  $X_i$ ,  $Y_i$ ,  $m_i$  and  $n_i$  for each natural gas species is given in Table 4.9. The numerical fuel replaces the hydrocarbon mass fraction of the natural gas and must match the C and H mass flow rate from the hydrocarbons. The mass flow rate of C,  $\dot{m}_C \left[\frac{\text{kg}}{\text{s}}\right]$ , and H,  $\dot{m}_H \left[\frac{\text{kg}}{\text{s}}\right]$ , of each hydrocarbon is determined from Eq. 4.13 and 4.14 respectively.

$$\dot{m}_{\rm C,C_nH_{2n+2}} = \dot{m}_{\rm C_nH_{2n+2}} \frac{M_{\rm C}}{M_{\rm C_nH_{2n+2}}}$$
(4.13)

$$\dot{m}_{\mathrm{H,C_nH_{2n+2}}} = \dot{m}_{\mathrm{C_nH_{2n+2}}} \frac{M_{\mathrm{H}}}{M_{\mathrm{C_nH_{2n+2}}}}$$
(4.14)

where  $\dot{m}_{\rm C,C_nH_{2n+2}} \left[\frac{\rm kg}{\rm s}\right]$  and  $\dot{m}_{\rm H,C_nH_{2n+2}} \left[\frac{\rm kg}{\rm s}\right]$  are the C and H mass flow rate from the  $\rm C_nH_{2n+2}$  respectively and  $\dot{m}_{\rm C_nH_{2n+2}} \left[\frac{\rm kg}{\rm s}\right]$  is the mass flow rate of  $\rm C_nH_{2n+2}$  given in Table 4.9.  $\dot{m}_{\rm C,C_nH_{2n+2}}$  and  $\dot{m}_{\rm H,C_nH_{2n+2}}$  determined are given in Table 4.9. The mole H/C ratio of the numerical fuel is determined from the total mass flow rates of C and H given in Table 4.9 with Eq. 4.15.

$$\frac{\dot{n}_{\rm H}}{\dot{n}_{\rm C}} = \frac{\frac{m_{\rm H}}{M_{\rm H}}}{\frac{\dot{m}_{\rm C}}{M_{\rm C}}} = 3.783 \tag{4.15}$$

From the result in Eq. 4.15 the numerical fuel chemical formula is specified as  $CH_{3.783}$ . From the chemical formula the  $CH_{3.783}$  molecular weight 15.783  $\left[\frac{kg}{kmol}\right]$  follows. The H/C ratio of the numerical fuel is lower compared to  $CH_4$ . This is consistent with the lower H/C ratio of the other hydrocarbons present in the natural gas. The other hydrocarbons lower the H/C ratio only slightly due to their low mass fraction. The numerical fuel must match the heat release rate  $\dot{Q} \left[\frac{kJ}{s}\right]$  of the natural gas hydrocarbons. The heat release rate of the natural gas hydrocarbons is determined by summing the heat release rate of each hydrocarbon. As the furnace operated at  $\Phi = 0.9871$  there is enough reaction for the stoichiometric reactions to occur. For the case of CH<sub>4</sub> the stoichiometric oxy-CH<sub>4</sub> reaction is repeated in Eq. 4.16.

$$\mathrm{CH}_4 + 2\mathrm{O}_2 \to \mathrm{CO}_2 + 2\mathrm{H}_2\mathrm{O} \tag{4.16}$$

The heat release rate from  $CH_4$ ,  $\dot{Q}_{CH_4}$   $\left[\frac{kJ}{kg\cdot s}\right]$ , is determined by the energy balance given in Eq. 4.17.

$$\dot{Q}_{\mathrm{CH}_{4}} = \dot{n}_{\mathrm{CH}_{4}} \bar{h}_{f,\mathrm{CH}_{4}}^{0} + n_{\mathrm{O}_{2}} \bar{h}_{f,\mathrm{O}_{2}}^{0} - \dot{n}_{\mathrm{CO}_{2}} \bar{h}_{f,\mathrm{CO}_{2}}^{0} - n_{\mathrm{H}_{2}\mathrm{O}} \bar{h}_{f,\mathrm{H}_{2}\mathrm{O}}^{0} = \dot{n}_{\mathrm{CH}_{4}} \bar{h}_{f,\mathrm{CH}_{4}}^{0} - \dot{n}_{\mathrm{CH}_{4}} \bar{h}_{f,\mathrm{CO}_{2}}^{0} - 2\dot{n}_{\mathrm{CH}_{4}} \bar{h}_{f,\mathrm{H}_{2}\mathrm{O}}^{0}$$

$$(4.17)$$

where  $\bar{h}_{f}^{0} \left[\frac{\text{kJ}}{\text{kmol}}\right]$  is the enthalpy of formation at standard reference state. 298.15 [K] and 101.325 [kPa] is used as the standard reference state. The mole flow rates  $\dot{n}_{O_2}$ ,  $\dot{n}_{CO_2}$  and  $\dot{n}_{H_2O}$  are determined from Eq. 4.16 as function of  $\dot{n}_{CH_4}$ . The heat release rates from C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, C<sub>4</sub>H<sub>10</sub> and C<sub>5</sub>H<sub>12</sub> are determined in a similar manner from Eq. 4.18 to 4.21. The heat release rates are given Table 4.9.

$$\dot{Q}_{C_2H_6} = \dot{n}_{C_2H_6}\bar{h}^0_{f,C_2H_6} + 3.5\dot{n}_{C_2H_6}\bar{h}^0_{f,O_2} - 2\dot{n}_{C_2H_6}\bar{h}^0_{f,CO_2} - 3\dot{n}_{C_2H_6}\bar{h}^0_{f,H_2O}$$
(4.18)

$$\dot{Q}_{C_3H_8} = \dot{n}_{C_3H_8}\bar{h}^0_{f,C_3H_8} + 5\dot{n}_{C_3H_8}\bar{h}^0_{f,O_2} - 3\dot{n}_{C_3H_8}\bar{h}^0_{f,CO_2} - 4\dot{n}_{C_3H_8}\bar{h}^0_{f,H_2O}$$
(4.19)

$$\dot{Q}_{C_4H_{10}} = \dot{n}_{C_4H_{10}}\bar{h}^0_{f,C_4H_{10}} + 6.5\dot{n}_{C_4H_{10}}\bar{h}^0_{f,O_2} - 4\dot{n}_{C_4H_{10}}\bar{h}^0_{f,CO_2} - 5\dot{n}_{C_4H_{10}}\bar{h}^0_{f,H_2O} \quad (4.20)$$

$$\dot{Q}_{C_5H_{12}} = \dot{n}_{C_5H_{12}}\bar{h}_{f,C_5H_{12}}^0 + 8\dot{n}_{C_5H_{12}}\bar{h}_{f,O_2}^0 - 5\dot{n}_{C_5H_{12}}\bar{h}_{f,CO_2}^0 - 6\dot{n}_{C_5H_{12}}\bar{h}_{f,H_2O}^0$$
(4.21)

The total heat release rate of the natural gas,  $\dot{Q}_{\rm NG}$ , is determined with Eq. 4.22.

$$\dot{Q}_{\rm NG} = \dot{Q}_{\rm CH_4} + \dot{Q}_{\rm C_2H_6} + \dot{Q}_{\rm C_3H_8} + \dot{Q}_{\rm C_4H_{10}} + \dot{Q}_{\rm C_5H_{12}} \tag{4.22}$$

The  $\text{CH}_{3.783}$  mass flow rate is set equal to the total mass flow rate of the natural gas hydrocarbons given in Table 4.9. From  $\dot{m}_{\text{CH}_{3.783}}$  and the determined molecular weight the mole flow rate  $\dot{n}_{\text{CH}_{3.783}} = 9.913 \cdot 10^{-4}$  follows. The numerical fuel must equal the heat release rate of the natural gas hydrocarbons. The  $\text{CH}_{3.783}$  stoichiometric reaction is given in Eq. 4.23.

$$CH_{3.783} + 1.9457O_2 \rightarrow CO_2 + 1.8915H_2O$$
 (4.23)

The numerical fuel enthalpy of formation is determined from Eq. 4.24, an energy balance for the stoichiometric reaction in Eq. 4.23.

$$\bar{h}_{f,\mathrm{CH}_{3.783}}^{0} = \frac{\dot{n}_{\mathrm{CH}_{3.783}}\bar{h}_{f,\mathrm{CO}_{2}}^{0} + \left(\frac{3.783}{2}\right)\dot{n}_{\mathrm{CH}_{3.783}}\bar{h}_{f,\mathrm{H}_{2}\mathrm{O}}^{0} + \dot{Q}_{NG}}{\dot{n}_{\mathrm{CH}_{3.783}}} = 69958.3099 \left[\frac{\mathrm{kJ}}{\mathrm{kmol}}\right]$$
(4.24)

The calculated properties of the numerical fuel  $CH_{3.783}$  used to model the hydrocarbon mass fraction of the OXYFLAM-2 natural gas are summarized in Table 4.8.

Chemical formula	CH <sub>3.783</sub>
$M_{\mathrm{CH}_{3.783}}$	$15.783 \left  \frac{\text{kg}}{\text{kmol}} \right $
$\bar{h}^{0}_{f, \mathrm{CH}_{3.783}}$	$69958.3099 \left[\frac{\mathrm{kJ}}{\mathrm{kmol}}\right]$

Table 4.8: Properties of the numerical fuel  $CH_{3.783}$ .

Species	$M_i\left[\frac{\mathrm{kg}}{\mathrm{kmol}}\right]$	$X_i$	$Y_i$	$\dot{m}_i \left[\frac{\mathrm{kg}}{\mathrm{s}}\right]$	$\dot{n}_i \left[\frac{\mathrm{kmol}}{\mathrm{s}}\right]$	$\dot{m}_{\rm C} \left[\frac{\rm kg}{\rm s}\right]$	$\dot{m}_{\rm H} \left[\frac{\rm kg}{\rm s}\right]$	$\dot{Q}_i \left[\frac{\mathrm{kJ}}{\mathrm{s}}\right]$
$CH_4$	16.04	0.86	0.7393	0.01304	$8.131\cdot 10^{-4}$	$9.783\cdot10^{-3}$	$3.261\cdot 10^{-3}$	647.2
$C_2H_6$	30.07	0.054	0.08701	$1.535 \cdot 10^{-3}$	$5.105 \cdot 10^{-5}$	$1.228 \cdot 10^{-3}$	$3.07\cdot 10^{-4}$	72.34
$C_3H_8$	44.1	0.0187	0.04419	$7.796 \cdot 10^{-4}$	$1.768 \cdot 10^{-5}$	$6.379\cdot10^{-4}$	$1.417\cdot 10^{-4}$	35.83
$C_4H_{10}$	58.12	0.0058	0.01807	$3.187\cdot10^{-4}$	$5.483 \cdot 10^{-6}$	$2.638\cdot10^{-4}$	$5.495 \cdot 10^{-5}$	14.45
$C_5H_{12}$	72.15	0.0014	0.005413	$9.55\cdot10^{-5}$	$1.324 \cdot 10^{-6}$	$7.958 \cdot 10^{-5}$	$1.592 \cdot 10^{-5}$	4.3
$\sum_{i=1}^{n}$	16.68	0.9399	0.894	0.01577	$8.886\cdot10^{-4}$	0.01199	$3.781 \cdot 10^{-3}$	774.1
$\rm CO_2$	44.01	0.0179	0.04222	$7.448 \cdot 10^{-4}$	$1.692 \cdot 10^{-5}$			
$N_2$	28.01	0.0401	0.0602	$1.062 \cdot 10^{-3}$	$3.791 \cdot 10^{-5}$			
$O_2$	32	0.0021	0.003601	$6.353 \cdot 10^{-5}$	$1.985 \cdot 10^{-6}$			
$\sum$	18.66	1	1	0.01764	$9.454 \cdot 10^{-4}$			

Table 4.9: Mole fraction, mass fraction, mass flow rate and mole flow rate of natural gas components and the mass flow rate of C and H due to each hydrocarbon.

### Chapter 5

# Radiation

In this chapter the modelling of the radiative properties of the combustion product gases is discussed. Thermal radiation and the thermal radiative properties of combustion product gases are discussed. The weighted sum of grey gases model (WSGGM) used in simulations to determine the emissivity of mixtures of oxy-fuel combustion product gases is discussed. The determination of appropriate WSGGM coefficients for oxy-fuel combustion products using the exponential wide band model (EWBM) is discussed and the coefficients are presented.

#### 5.1 Gas Radiation

The thermal radiative heat flux  $q \left[\frac{W}{m^2}\right]$  from a blackbody to isothermal surroundings is given by Eq. 5.1.

$$q = \sigma (T^4 - T_{surr}^4) \tag{5.1}$$

where  $\sigma \left[\frac{W}{m^2 \cdot K^4}\right]$  is the Stefan-Boltzmann constant and  $T_{surr}$  [K] is the temperature of the surroundings. From Eq. 5.1 it is apparent that the radiative flux is proportional to  $T^4$ . Thermal radiative heat transfer therefore usually becomes significant relative to convective heat transfer as the temperature of the system increases. In combustion systems temperatures of several thousand degrees Kelvin usually occur and from the preceding discussion it follows that radiative heat transfer becomes significant. Radiative heat transfer must therefore be modelled accurately for accurate simulations of combustion systems. Thermal radiation is concentrated in the infrared spectrum. The diatomic gases N<sub>2</sub> and O<sub>2</sub> have no significant absorption bands in the infrared spectrum. This is in contrast to H<sub>2</sub>O and CO2 which absorb and emit a significant amount of radiation in the thermal spectrum. It is therefore apparent due to the large concentration of CO<sub>2</sub> and H<sub>2</sub>O that . Johansson et al. [2010] evaluated a number models for gas emissivity and found that the WSGGM model provides a good compromise between accuracy and computation time.

#### 5.2 Weighted Sum of Grey Gases Model

The weighted sum of grey gases model assumes that the emissivity of a gas mixture can be approximated as a weighted sum of I grey gases each with a discrinct absorption coefficient. The emissivity is determined with the WSGGM from Eq. 5.2.

$$\varepsilon = \sum_{i=0}^{I} a_{\varepsilon,i} \left(T\right) \left[1 - e^{-\kappa_i PS}\right]$$
(5.2)

where T [K] is the gas mixture temperature, P [bar] is the sum of the partial pressures of the absorbing species in the gas mixture and  $\kappa_i$  [bar<sup>-1</sup> · m<sup>-1</sup>] is the absorption coefficient of the grey gas i. The weighting of each grey gas is temperature dependent and a polynomial function of temperature is used. The weighting factor function for the grey gas i,  $a_{\varepsilon,i}$  is given in Eq. 5.3.

$$a_{\varepsilon,i} = \sum_{j=0}^{J} b_{\varepsilon,i,j} \left(\frac{T}{T_{ref}}\right)^{j}$$
(5.3)

where  $b_{\varepsilon,i,j}$  is the polynomial coefficient j of the weighting factor function for the grey gas i and  $T_{ref}$  [K] is a reference temperature. The weighting factor  $a_{\varepsilon,i}$  can be interpreted as the fraction of black body radiation energy in the spectral interval where the grey gas i has an absorption band with value  $\kappa_i$  as discussed by Smith et al. [1982]. Spectral intervals between the absorption bands modelled by the I grey gases are accounted for by one clear gas. The clear gas is assigned an aborption  $\kappa_i = 0$  and the weighting factor for the clear gas  $a_{\varepsilon,0}$  is determined using Eq. 5.4.

$$a_{\varepsilon,0} = 1 - \sum_{i=1}^{I} a_{\varepsilon,i} \tag{5.4}$$

The weighting factor function presented by Smith et al. [1982] does not employ the reference temperature present in Eq. 5.3. The weighting with reference temperature is employed by Johansson et al. [2010] to normalize the temperature and thereby reduce the magnitude of the term  $\left(\frac{T}{T_{ref}}\right)^j$  which would become large due to the increasing exponent and power j in the last terms of  $a_{\varepsilon,i}$ . This then reduces the magnitude of the coefficients  $b_{\varepsilon,i,j}$  which are on the order of  $10^4$  and increase to  $10^{11}$  for the coefficients presented by Smith et al. [1982] for a weighting function with a polynomial degree of 4. For coefficients of such magnitudes more significant figures are necessary in order to adjust the large temperature terms accurately. The normalization procedure can be employed as the temperatures are limited to a finite interval of between approximately  $300 \ [K]$  and  $3000 \ [K]$  in furnaces. It is therefore possible to normalize the temperature to approximately one order of magnitude difference between the two extremes of the temperature range.

Each mixture of gases requires a distinct set of coefficients and the WSGGM is therefore not immediately applicable to different mixtures. For each gas mixture a set of coefficients is determined on the basis of emissivity data for the given mixture obtained from another source. The WSGGM is fitted to emissivity data by choosing appropriate coefficients in order for the WSGGM to yield correct emissivities. In the original presentation of the WSGGM the coefficients were determined using a graphical method to fit the WSGGM to the emissivity data. Another procedure is to use optimization methods to minimize the square of the relative error in emissivity between the data source and the emissivity obtained from the WSGGM as first presented by Smith et al. [1982]. Smith et al. [1982] developed WSGGM coefficients for air combustion conditions using this method. This method has also been used by Yin et al. [2010], Johansson et al. [2010] and Johansen et al. [2009] to determine coefficients for the WSGGM applicable to conditions encountered in oxy-fuel combustion. Yin et al. [2010] used the exponential wide band model to provide the required emissivity data for the fitting of the WSGGM. The WSGGM coefficients were then obtained by fitting to the EWBM emissivity data. This has been done for various partial pressure ratios of  $H_2O$  and  $\rm CO_2$  cases encountered in an oxy-fuel furnace and a total pressure of 1 [atm]. The WSGGM coefficients determined by Yin et al. [2010] are given in Figure 5.1. Details of the EWBM emissivity calculations and the determination of WSGGM coefficients are given by Yin et al. [2010] and Johansen et al. [2009].

A user defined function (UDF) to evaluate the absorption coefficient required for the radiative transfer equation from the emissivity obtained from the new WSGGM for oxy-fuel conditions has been developed by Yin et al. [2010]. Eq. 5.5 relates the absorption coefficient  $a \, [m^{-1}]$  to the emissivity as shown by Siegel and Howell [2002] and ANSYS Inc. [2009].

$$a = -\frac{\ln\left(1-\epsilon\right)}{S} \tag{5.5}$$

The new WSGGM model has been used in CFD simulations to model the radiative properties of the gas mixture in the furnace. The peak temperature in the furnace is 4500 [K]. This is greater than the temperatures for which the WSGGM coefficients in Figure 5.1 and the default WSGGM coefficients for air combustion developed by Smith et al. [1982]. An error is therefore introduced the absorption coefficient determined at temperatures above 3000 [K]. Two methods are used for determination of the beam length S in the WSGGM, the cell based and domain based beam length. Both beam lengths are determined from Eq. 5.6.

$$S = 3.6 \frac{V}{A} \tag{5.6}$$

where  $V \text{ [m^3]}$  is the volume of the cell or domain and  $A \text{ [m^2]}$  is the surface area of the volume of the cell or domain. A comparison of the emissivities obtained from the air WSGGM, the oxy WSGGM and the EWBM as a function of beam length at various partial pressure ratios is shown in Figure 5.2. A comparison of emissivity data as a function of temperature is shown Figure 5.3 for the cell based beam length and domain based beam length. From Figure 5.2 it can be concluded that at beam lengths below 10 [m] the emissivity determined with the air WSGGM and oxy WSGGM are essentially equal. As the beam length in the CFD simulations is 0.79 [m] no significant difference is likely to be observed between results obtained using the oxy WSGGM and air WSGGM. The domain based beam length is used in all CFD simulations performed.

i	k <sub>i</sub>	$b_{\varepsilon,i,1}$	$b_{\varepsilon,i,2}$	$b_{\varepsilon,i,3}$	$b_{\varepsilon,i,4}$
$P_w \rightarrow 0$ atm, $F$	$P_c \rightarrow 0 \text{ atm}$				
1	0.009422	0.778969	-1.342848	0.964858	-0.195747
2	0.415646	-0.011449	0.343754	-0.234886	0.044008
3	11.617018	-0.007627	0.242233	-0.173738	0.033868
4	319.911168	0.080082	-0.049280	0.001861	0.002232
$P_{w} = 0.1 \text{ atm}, 1$	$P_c = 0.1  \mathrm{atm}$				
1	0.256738	0.492304	-0.433789	0.279329	-0.057770
2	3.108033	0.082686	0.486294	-0.369752	0.070509
3	52.585782	0.144385	-0.083662	0.002003	0.003902
4	440.845718	0.079515	-0.110361	0.051379	-0.007983
$P_w = 0.3 \text{ atm}, 1$	$P_c = 0.1  \mathrm{atm}$				
1	0.132242	0.478371	-0.608643	0.475098	-0.109044
2	14.660767	0.101065	0.204118	-0.202202	0.042771
3	1.750654	0.185155	0.299794	-0.240346	0.046968
4	165.763926	0.191665	-0.277448	0.133514	-0.021280
$P_w / P_c = 1/8$ ,	$P_w + P_c = 1 \text{ atm} (\text{corresp})$	onding to dry flue ga	is recycling, FGR)		
1	0.051237	0.515415	-0.618162	0.430921	-0.092082
2	0.688383	0.199807	0.298581	-0.265758	0.052910
3	13.763205	0.138767	-0.001851	-0.049353	0.013012
4	289.841885	0.08/511	-0.06/295	0.013489	-5.54E-06
$P_{W}/P_{c} = 1/4$ ,	$P_w + P_c = 1 \text{ atm}$				
1	0.052694	0.486247	-0.644137	0.485654	-0.107808
2	0.752776	0.213959	0.306543	-0.264417	0.051889
3	11.543306	0.181991	-0.020460	-0.053791	0.015058
$\frac{4}{D/D} = 1/2$	252.958841	0.106180	-0.096088	0.028114	-0.002443
$\Gamma_W / \Gamma_C = 1/2 ,$	$r_w + r_c = 1 \text{ aun}$	0.000005	0.510025	0.4400.01	0.10(000
1	0.052378	0.383225	-0.510937	0.442201	-0.106398
2	0.712283	0.251481	0.161562	-0.150405	0.028982
3	105 802573	0.208259	0.156339	-0.133008	0.032090
P/P - 3/4	P + P - 1 atm	0.147235	-0.150555	0.057098	-0.007200
1 1	$1_W + 1_c = 1$ addi	0.255052	0.276222	0 211285	0.084003
1 2	0.031039	0.233933	-0.276222	0.0511285	-0.084903
2	6.051770	0.160253	0 289548	-0 284144	0.060344
4	150.875915	0.201452	-0.233937	0.095159	-0.013302
$P_{\rm w}/P_{\rm c} = 1/1$ ,	$P_{\rm su} + P_{\rm c} = 1  {\rm atm}  ({\rm corresp})$	onding to wet FGR)			
1	0.051487	0 164048	-0.087793	0 195253	-0.063573
2	0.571797	0.412652	-0.339810	0.197886	-0.038963
3	5.398936	0.112364	0.450929	-0.388486	0.079862
4	130.622859	0.238339	-0.288619	0.121962	-0.017651
$P_w / P_c = 2/1$ ,	$P_w + P_c = 1 \text{ atm (corresp})$	onding to, e.g., oxy-	fuel combustion of n	atural gas, without F	GR)
1	0.054480	-0.002188	0.286129	-0.048594	-0.016243
2	0.555304	0.546857	-0.714799	0.452812	-0.088841
3	5.040174	-0.001911	0.764177	-0.581819	0.115069
4	100.372663	0.317219	-0.415470	0.186570	-0.028335
$P_w / P_c = 4 / 1$ ,	$P_w + P_c = 1 \text{ atm}$				
1	0.060800	-0.053999	0.434975	-0.152413	0.005094
2	5.608831	-0.094953	0.952010	-0.696161	0.136316
3	0.676040	0.606525	-0.853216	0.545562	-0.107328
4	84.540632	0.369661	-0.517493	0.244011	-0.038451

Figure 5.1: WSGGM coefficients developed by Yin et al. [2010] for oxy-fuel conditions. Coefficients are given for various partial pressures and partial pressure ratios of  $\rm H_2O$  and  $\rm CO_2$ . From Yin et al. [2010].



Figure 5.2: Comparison of emissivities obtained from EWBM, Smith WSGGM, Johansson WSGGM and new WSGGM at various beam lengths and gas compositions from Yin et al. [2010].



Figure 5.3: Comparison of emissivities obtained from EWBM, Smith WSGGM, Johansson WSGGM and new WSGGM at gas temperatures and gas compositions from Yin et al. [2010]. The cell based approach is shown to the left and the domain based to the right.

### Chapter 6

## **RANS** Simulations

In this chapter the RANS models and simulations performed are discussed. The RANS equations and solution method are presented along with the models employed for turbulence, chemistry-turbulence interaction and radiation transfer. The parameters used for each model are presented. The mesh independence study and the cases simulated are discussed.

#### 6.1 Reynolds Averaged Navies Stokes Equations

The RANS equations constitute one approach to account for the influence of turbulence on fluid flows. Turbulence has an effect on all transported properties in a flow and the importance of turbulence generally increases with the Reynolds number. Turbulent flows have the common properties of velocity fields that fluctuate randomly in time, that are disordered and are chaotic as discussed by Davidson [2004]. There are three approaches to simulating turbulent flows. The Reynolds Averaged Navier Stokes (RANS) equations model all scales of turbulence, the large eddy simulation (LES) equations simulate the larger turbulence scales and model the smaller scales and direct numerical simulation (DNS) simulates all scales of turbulence. The approaches are in order of increasing computation time. In this investigation the RANS approach has been used. For a turbulent flow an instantaneous scalar variable  $\varphi$  can be decomposed to a mean component with respect to time,  $\overline{\varphi}$ , and a fluctuating component  $\varphi'$  as shown in Eq. 6.1.

$$\varphi = \overline{\varphi} + \varphi' \tag{6.1}$$

The RANS equations for incompressible flows are obtained by decomposing each variable in the Navier Stokes equations in the form of Eq. 6.1 and time averaging the equations. The RANS equations include terms involving fluctuating properties and these require modelling as discussed by Versteeg and Malalasekra [2007]. For flows with density fluctuations, such as flows involving combustion, the decomposition given in Eq. 6.1 leads to RANS equations with many more terms containing fluctuating components compared to the RANS equations for incompressible flow. These additional terms also require modelling. To reduce the number of terms containing fluctuating variables, a density weighted averaging procedure is used instead of Eq. 6.1 as discussed

by Versteeg and Malalasekra [2007]. This is known as Favre averaging. As shown by Versteeg and Malalasekra [2007], a density weighted mean variable is defined as given in Eq. 6.2.

$$\widetilde{\varphi} = \frac{\overline{\rho \varphi}}{\overline{\rho}} \tag{6.2}$$

The instantaneous variable  $\varphi$  is then decomposed according to Eq. 6.3.

$$\varphi = \widetilde{\varphi} + \varphi'' \tag{6.3}$$

where  $\varphi''$  is a fluctuating component which includes the effect of density fluctuations. If the flow is incompressible  $\tilde{\varphi}$  and  $\varphi''$  reduce to  $\overline{\varphi}$  and  $\varphi'$  respectively. Decomposing each scalar variable in the Navier Stokes equations according to Eq. 6.3 and time averaging yields the Favre averaged RANS equations given in Eq. 6.4, Eq. 6.5, Eq. 6.6 and Eq. 6.10 as presented by Versteeg and Malalasekra [2007].

$$\frac{\partial \overline{\rho}}{\partial t} + \frac{\partial}{\partial x_i} \left( \overline{\rho} \widetilde{u}_i \right) = 0 \tag{6.4}$$

where  $\rho \left[\frac{\text{kg}}{\text{m}^3}\right]$  is the density,  $x_i$  is the coordinate i, t [s] is time and  $u_i$   $\left[\frac{\text{m}}{\text{s}}\right]$  is the component of velocity in the i coordinate direction.

$$\frac{\partial \overline{\rho} \widetilde{u}_i}{\partial t} + \frac{\partial}{\partial x_j} \left( \overline{\rho} \widetilde{u}_i \widetilde{u}_j \right) = -\frac{\partial \overline{p}}{\partial x_i} + \frac{\partial}{\partial x_j} \left( \overline{\tau}_{ij} - \overline{\rho u_i'' u_j''} \right)$$
(6.5)

where  $x_j$  is the coordinate j, p [Pa] is the pressure and  $\tau \left[\frac{N}{m^2}\right]$  is the viscous stress on a surface normal to the *i* coordinate direction and acting in the *j* coordinate direction.

$$\frac{\partial \overline{\rho} \widetilde{h}}{\partial t} + \frac{\partial}{\partial x_j} \left( \overline{\rho} \widetilde{h} \widetilde{u}_j \right) = \frac{\partial}{\partial x_j} \left( \Gamma_h \frac{\partial \widetilde{h}}{\partial x_j} \right) + \overline{S}_h \tag{6.6}$$

where  $h \begin{bmatrix} \frac{kJ}{kg} \end{bmatrix}$  is the total enthalpy,  $\Gamma_h \begin{bmatrix} \frac{kg}{m \cdot s} \end{bmatrix}$  is determined from Eq. 6.7 and  $\overline{S}_h \begin{bmatrix} \frac{kJ}{m^3 \cdot s} \end{bmatrix}$  is an energy source term.

$$\Gamma_h = \frac{\mu}{Pr} + \frac{\mu_t}{Pr_t} \tag{6.7}$$

where  $\mu \begin{bmatrix} \frac{\text{kg}}{\text{m}\cdot\text{s}} \end{bmatrix}$  and  $\mu_t \begin{bmatrix} \frac{\text{kg}}{\text{m}\cdot\text{s}} \end{bmatrix}$  are the laminar and turbulent viscosity respectively and Pr and  $Pr_t$  are the laminar and turbulent Prandtl numbers respectively determined from Eq. 6.8 and 6.9.

$$Pr = \frac{c_p \mu}{k} \tag{6.8}$$

$$Pr_t = \frac{\mu_t}{\rho D_t} \tag{6.9}$$

where  $k \begin{bmatrix} W \\ m \cdot K \end{bmatrix}$  and  $k_t \begin{bmatrix} W \\ m \cdot K \end{bmatrix}$  are the laminar and turbulent thermal conductivity and  $D \begin{bmatrix} \frac{m^2}{S} \end{bmatrix}$  and  $D_t \begin{bmatrix} \frac{m^2}{s} \end{bmatrix}$  are the laminar and turbulent diffusion coefficients.

$$\frac{\partial \overline{\rho} \widetilde{Y}_k}{\partial t} + \frac{\partial}{\partial x_j} \left( \overline{\rho} \widetilde{Y}_k \widetilde{u}_j \right) = \frac{\partial}{\partial x_j} \left( \Gamma_k \frac{\partial \widetilde{Y}_k}{\partial x_j} \right) + \frac{\partial}{\partial x_j} \left( \overline{\rho} D_k \frac{\partial \widetilde{Y}''_k}{\partial x_j} \right) + R_i$$
(6.10)

where  $Y_k$  is the mass fraction of species k,  $D_k \left[\frac{\mathrm{m}^2}{\mathrm{s}}\right]$  is the binary diffusion coefficient of species k,  $R_i \left[\frac{\mathrm{kg}}{\mathrm{m}^3 \cdot \mathrm{s}}\right]$  is the net rate of reaction of species i and  $\Gamma_k \left[\frac{\mathrm{kg}}{\mathrm{m} \cdot \mathrm{s}}\right]$  is determined from Eq. 6.11.

$$\Gamma_h = \frac{\mu}{Sc} + \frac{\mu_t}{Sc_t} \tag{6.11}$$

where Sc and  $Sc_t$  are the laminar and turbulent Schmidt numbers respectively which are determined from Eq. 6.12 and 6.13.

$$Sc = \frac{\mu}{\rho D} \tag{6.12}$$

$$Sc_t = \frac{\mu_t}{\rho D_t} \tag{6.13}$$

Eq. 6.4 is the continuity equation, Eq. 6.5 is the momentum conservation equation, Eq. 6.6 is the enthalpy conservation equation and Eq. 6.10 is the species conservation equation. The ideal gas equation given in Eq. 6.14 and the definition of total enthalpy given in Eq. 6.15 provide closure along with a model for the Reynolds stress term, discussed in Section 6.2.

$$p = \rho RT \tag{6.14}$$

$$h = h_f^0 + \int_{T0}^T c_p dT$$
 (6.15)

#### 6.2 Turbulence Modelling

The additional unknown Reynolds stress term  $-\overline{\rho u'_i u'_j} \begin{bmatrix} N \\ m^2 \end{bmatrix}$  is introduced in Eq. (6.5) by the Favre averaging of the Navier Stokes equations. In order to solve the RANS equations a model for the Reynolds stress term is required in order to achieve closure. A common model for the Reynolds stress term is the Boussinesq hypothesis. The Boussinesq hypothesis assumes that the Reynolds stresses are proportional to the time averaged velocity gradients . The Boussinesq equation is given in Eq. 6.16.

$$-\rho \overline{u'_{i}u'_{j}} = \mu_{t} \left(\frac{\partial u_{i}}{\partial x_{j}} + \frac{\partial u_{j}}{\partial x_{i}}\right) - \frac{2}{3} \left(\rho k + \mu_{t} \frac{\partial u_{k}}{\partial x_{k}}\right) \delta_{ij}$$
(6.16)

where  $\delta_{ij}$  is the Kronecker delta function.  $\delta_{ij} = 1$  for i = j and  $\delta_{ij} = 0$  for  $i \neq j$ . The Boussinesq hypothesis is used in the CFD simulations along with the  $k - \epsilon$  turbulence model. The  $k - \epsilon$  turbulence model solves transport equations for the turbulent kinetic energy  $k \left[\frac{\mathrm{m}^2}{\mathrm{s}^2}\right]$  and turbulent energy dissipation rate  $\epsilon \left[\frac{\mathrm{m}^2}{\mathrm{s}^3}\right]$ . The  $k - \epsilon$  turbulence

model transport equations for k and  $\epsilon$  are given in Eq. 6.17 and 6.18 as given by ANSYS Inc. [2009]. The variables should be viewed as density weighted mean values consistent with the Favre averaged approach.

$$\frac{\partial}{\partial t}\left(\rho k\right) + \frac{\partial}{\partial x_{i}}\left(\rho k u_{i}\right) = \frac{\partial}{\partial x_{j}}\left(\left(\mu + \frac{\mu_{t}}{Pr_{k}}\right)\frac{\partial k}{\partial x_{j}}\right) + G_{k} + G_{b} - \rho\epsilon - Y_{m}$$
(6.17)

$$\frac{\partial}{\partial t}\left(\rho\epsilon\right) + \frac{\partial}{\partial x_{i}}\left(\rho\epsilon u_{i}\right) = \frac{\partial}{\partial x_{j}}\left(\left(\mu + \frac{\mu_{t}}{Pr_{\epsilon}}\right)\frac{\partial\epsilon}{\partial x_{j}}\right) + C_{1\epsilon}\frac{\epsilon}{k}\left(G_{k} + C_{3\epsilon}G_{b}\right) - C_{2\epsilon}\rho\frac{\epsilon^{2}}{k}$$
(6.18)

where  $Pr_k$  and  $Pr_{\epsilon}$  are the turbulent Prandtl numbers for k and  $\epsilon$  respectively,  $G_k$  represents the generation of turbulent kinetic energy due to the mean velocity gradients,  $Y_M$  is the dissipation rate due to fluctuating dilation in compressible flow, and  $C_{1\epsilon}$ ,  $C_{2\epsilon}$ ,  $C_{3\epsilon}$  are model constants. The default values given by ANSYS Inc. [2009] have been used for the model constants. Eq. 6.19 is used to evaluate the turbulent viscosity  $\mu_t$ .

$$\mu_t = \rho C_\mu \frac{k^2}{\epsilon} \tag{6.19}$$

where  $C_{\mu}$  is model constant. The  $k - \epsilon$  model exhibits excellent performance for many industrially relevant flows and is the most widely validated turbulence model according to Versteeg and Malalasekra [2007]. The model is known to perform poorly in weak shear layers such as far mixing layers and the rate of spreading for an axis symmetric jet is over predicted as discussed by Versteeg and Malalasekra [2007]. Casey et al. [2000] states that turbulence driven secondary flows in straight ducts with a non circular cross section are not predicted.

The realizable  $k - \epsilon$  model model is an improvement on the standard  $k - \epsilon$  model with respect to prediction of jet spreading rate according to ANSYS Inc. [2009]. The realizable  $k - \epsilon$  uses different expressions for the turbulent viscosity and the transport equation for  $\varepsilon$ . Simulations have been performed to evaluate the effect of using the realizable  $k - \epsilon$  model. The equations for the realizable  $k - \epsilon$  model are given by ANSYS Inc. [2009].

#### 6.3 Radiative Heat Transfer Modelling

Radiation heat transfer is accounted for by solving the radiative transfer equation (RTE) given in Eq. 6.20 as shown by ANSYS Inc. [2009].

$$\nabla \cdot \left( I\left(\mathbf{r},\mathbf{s}\right)\mathbf{s}\right) + \left(a + \sigma_{s}\right)I\left(\mathbf{r},\mathbf{s}\right) = an^{2}\frac{\sigma T^{4}}{\pi} + \frac{\sigma_{s}}{4\pi}\int_{0}^{4\pi}I\left(\mathbf{r},\mathbf{s}'\right)\Phi\left(\mathbf{s}\cdot\mathbf{s}'\right)d\Omega' \qquad (6.20)$$

where  $I\left[\frac{W}{m^2 \cdot sr}\right]$  is the total radiation intensity,  $a\left[m^{-1}\right]$  is the absorption coefficient,  $\sigma_s\left[m^{-1}\right]$  is the scattering coefficient, n is the refractive index,  $\Omega$  [sr] is the solid angle, **s** and **s'** are the incoming and outgoing radiation direction vectors and **r** is the position vector. The discrete ordinates (DO) method solves the RTE in a finite number of solid angles.  $8N_{\theta}N_{\phi}$  equations are solved where  $N_{\theta}$  and  $N_{\phi}$  are the number subdivisions of

1/8 of a sphere in the  $\phi$  and  $\theta$  spherical coordinates. The DO method is used to solve Eq. 6.20 in all CFD simulations performed.  $N_{\theta} = N_{\phi} = 2$  subdivisions have been used for all simulations. One simulation with  $N_{\theta} = N_{\phi} = 3$  has been performed in order to determine whether  $N_{\theta} = N_{\phi} = 2$  subdivisions are sufficient. The simulation exhibits no difference and  $N_{\theta} = N_{\phi} = 2$  is therefore concluded to be sufficient.

#### 6.4 Chemistry Modelling

The WD, modified WD and modified WD B mechanisms in Section 4.2 have been used in the CFD simulations performed. The WD scheme was used for the mesh independence study and the modified WD B scheme has been used for one simulation. The remaining simulations have used the modified WD mechanism. As the simulated flow is fully turbulent the chemistry-turbulence interaction must be accounted for. Two models for this interaction and the reactions are used. The eddy dissipation (ED) model, given in Eq. 6.21 and 6.22 as presented by ANSYS Inc. [2009] has been used initially in the mesh independence simulations before changing to the eddy dissipation concept (EDC) model for all subsequent iterations and simulations. This has been done stability during the simulations.

$$R_{i,r} = \nu'_{i,r} M_{w,i} A \rho \frac{\epsilon}{k} \min_{\mathcal{R}} \left( \frac{Y_{\mathcal{R}}}{\nu'_{\mathcal{R},r} M_{\mathcal{R}}} \right)$$
(6.21)

where  $\mathcal{R}$  is a reaction reactant and A = 0.4 is an empirical model constant.

$$R_{i,r} = \nu'_{i,r} M_{w,i} A B \rho \frac{\epsilon}{k} \frac{\sum^{\mathcal{P}} Y_{\mathcal{P}}}{\sum_{j}^{N} \nu''_{j,r} M_{j}}$$
(6.22)

where  $\mathcal{P}$  is a reaction product and B = 0.5 is an empirical model constant. In the eddy dissipation model the reaction rate is proportional to the large eddy mixing time scale given by  $k/\epsilon$ . The reaction rate  $R_{i,r} \left[\frac{\text{kg}}{\text{m}^3 \cdot \text{s}}\right]$  is taken as the smaller of the values determined with the equations. The ED model only models turbulent reaction rates and does not include kinetic reaction rates. The reaction mechanisms used employ the Arrhenius expression to model reaction rates. The ED model can therefore not predict intermediate species and is ill suited for the WD and JL multi step mechanisms.

The EDC model can be used with Arrhenius rate expressions and can therefore be used with multi step reaction mechanisms. The EDC model assumes that reactions take place in the smallest turbulent scales, referred to as fine scales, where viscous dissipation occurs and mixing is completed as discussed by Magnussen [2005], Versteeg and Malalasekra [2007] and ANSYS Inc. [2009]. The EDC model equations are given in Eq. 6.23, 6.24 and 6.25.

$$\zeta^* = C_{\zeta} \left(\frac{\mu\epsilon}{\rho k^2}\right)^{\frac{1}{4}} \tag{6.23}$$

where  $\zeta^*$  is the length fraction of the fine scales and  $C_{\zeta} = 2.1377$  is the volume fraction constant. The volume fraction of the fine structure is determined as  $(\zeta^*)^3$ .

Parameter	Value
Integration method	ISAT
ODE abs. error tol.	$10^{-8}$
ODE rel. error tol.	$10^{-9}$
ISAT error tol.	0.001
Number of trees	1

Table 6.1: Chemistry integration parameters.

$$\tau^* = C_\tau \left(\frac{\nu}{\epsilon}\right)^{\frac{1}{2}} \tag{6.24}$$

where  $C_{\tau} = 0.4082$  is a time scale constant. The fine scale volume fraction is treated as a constant pressure reactor (CPR) and the system of differential equations that model the CPR is then integrated over the time scale  $\tau^*$ . The source term in the conservation equation is determined from Eq. 6.25.

$$R_{i} = \frac{\rho(\zeta^{*})^{2}}{\tau^{*} \left(1 - (\zeta^{*})^{3}\right)} \left(Y_{i}^{*} - Y_{i}\right)$$
(6.25)

where  $Y_i^*$  is the fine scale mass fraction of species *i* after the constant pressure reactor system is solved and  $Y_i$  is the mass fraction in the fluid surrounding the fine scale volume. The parameter values used for the ISAT integrator used to solve the are given in Table 6.1. The values used are the default values. ANSYS Inc. [2009] states that the default ISAT error tolerance is relatively large. The ISAT error tolerance should therefore be sequentially reduced until species concentrations remain constant. A simulation with the modified WD mechanism has performed with the ISAT error tolerance first reduced to 0.0005 and subsequently 0.0001 to investigate the effect of ISAT error tolerance. No change in the flow variables occurred for the converged solution with either of the lower ISAT error tolerances. The default ISAT error tolerance has therefore been assumed to be sufficient for all other simulations performed.

#### 6.5 RANS Solution

The RANS equations and models employed have been solved using the FLUENT 12.0.16 CFD software. FLUENT employs the finite volume method to solve the RANS partial differential equations. The SIMPLE pressure velocity coupling has been used. For all but one case, steady state simulations have been performed. In all cases the Green-Gauss node based method has been chosen to evaluate gradients and derivatives. This is due to the increased accuracy of the method relative to the less computationally expensive Green-Gauss cell based method as discussed by ANSYS Inc. [2009]. One transient simulations has been performed. Second order temporal discretization has been used for this simulation. The time step used is 0.0003 [s]. This yields a maximum cell Courant number of 30. This is between 20 and 40 as recommended by ANSYS Inc. [2009]. Six cycles of the furnace based on the residence time of gas on the centerline

Equation	Initial under relax.	Final under relax.
Pressure	0.3	0.3
Density	0.7	0.9
Momentum	0.6	0.6
k	0.75	0.75
$\epsilon$	0.75	0.75
$\mu_t$	0.8	0.8
Species	0.8	0.9
Energy	0.9	0.95
DO	0.9	0.95

Table 6.2: Under relaxation factors employed.

have been simulated. Sampling with a frequency of 1/10 of the residence time in the furnace has then been performed for the simulation of one furnace cycle.

The first order upwind discretization scheme has been used in all the mesh independence study simulations for the first 1000 iterations. The second order upwind discretization is then used for the remaining iterations. All other simulations have also used the second order upwind discretization scheme. Second order upwind discretization is better able to resolve gradients than the first order upwind discretization scheme. Numerical diffusion is therefore reduced compared to the first order scheme as illustrated by Versteeg and Malalasekra [2007]. Furthermore, the discretization scheme truncation error decreases twice as fast with increasing mesh cell number for a second order scheme compared to a first order scheme. A mesh independent solution will therefore be reached for a mesh with fewer cells compared to the mesh identified using first order discretization.

All simulations have used the converged mesh independent solution obtained in the mesh independence study for the initial conditions. This has been done in order to speed convergence as the mesh independent solution is assumed to be a good approximation to the solution of all other simulations performed.

The third order QUICK discretization scheme has also been used in one simulation. This has been done to determine the effect of higher order scheme with a further improved ability to resolve large gradients compared to the second order scheme. It has been found that the QUICK scheme does not produce any significant difference in the solution obtained compared to the second order scheme. The under relaxation factors given in Table 6.2 have been used for the initial and subsequent iterations respectively.

#### 6.6 Material Properties

The material properties given in Table 6.3 have been used. The constant pressure specific heat capacity  $c_p$  for the gas mixture is determined from Eq. 6.26 as the sum of the mass fraction weighted  $c_p$  of each species.  $c_{p,i}$  is determined from the polynomial functions of temperature. The default polynomials provided in FLUENT are used. The  $c_p$  polynomial provided for  $CH_4$  is used for the numerical fuel. As the natural gas is primarily comprised of  $CH_4$  the error introduced by using the  $c_p$  of  $CH_4$  for the entire hydrocarbon fraction of the natural gas is assumed to be minimal. The

Property	Value
Thermal conductivity $k \left[ \frac{W}{m \cdot K} \right]$	0.0454
Viscosity $\mu \left[\frac{\text{kg}}{\text{m}\cdot\text{s}}\right]$	$1.72\cdot 10^{-5}$
Mass diffusion coefficient $D\left[\frac{\mathrm{m}^2}{\mathrm{s}}\right]$	$2.88\cdot 10^{-5}$
Scattering coefficient $\sigma_s \left[ m^{-1} \right]$	0
Refractive index $n$	1

Table 6.3: Material properties used in all CFD simulations.

laminar thermal conductivity  $k \left[\frac{W}{m \cdot K}\right]$ , mass diffusion coefficient  $D \left[\frac{m^2}{s}\right]$  and viscosity  $\mu \left[\frac{kg}{m \cdot s}\right]$  are the default given by FLUENT for the WD mechanism gas mixture. As the flow is turbulent the turbulent properties corresponding to the laminar properties provided here are dominant and detailed modelling of the laminar properties is assumed to be unnecessary. The absorption coefficient is determined using the new WSGGM as described in Section 5.2.

$$c_p = \sum_{i=1}^{N} Y_i c_{p,i} \tag{6.26}$$

where  $c_{p,i}$  is the constant pressure specific heat capacity of species i.

#### 6.7 Boundary Conditions

The boundary conditions given in Table 6.4, 6.5 and 6.6 have been used in all simulations unless otherwise sepecified. The symmetry boundary type has been used for the symmetry planes. The wall emissivities are those used by Bollettini et al. [1997]. The wall roughness constants and roughness height are the default values provided by FLUENT. The mass flow rates and mass fractions at the inlets given in Table 6.4 are determined from Table 3.4 and 4.9. The mass flow rates are a quarter of the values given in the aforementioned tables due to the mesh being one quarter of the furnace. The inlet turbulence intensity  $I_{turb}$  in specified in Table 6.4 is based upon the values used by Bollettini et al. [1997]. The inlet turbulent kinetic energy is then determined from Eq. 6.27 given by ANSYS Inc. [2009].

$$k = \frac{3}{2} \left( u_{avg} I_{turb} \right)^2 \tag{6.27}$$

where  $u_{avg} \left[\frac{\text{m}}{\text{s}}\right]$  is the mean flow velocity. The turbulent kinetic energy dissipation rate is determined using Eq. 6.28 given by ANSYS Inc. [2009].

$$\epsilon = C_{\mu}^{\frac{3}{4}} \frac{k^{\frac{3}{2}}}{l} \tag{6.28}$$

where  $C_{\mu} = 0.09$  is an empirical constant and l [m] is the turbulence length scale estimated from Eq. 6.29 as given by ANSYS Inc. [2009].

Inlet	Gas	Oxygen
Boundary type	Mass flow inlet	
Mass flow rate $\left[\frac{\text{kg}}{\text{s}}\right]$	$4.4106\cdot10^{-3}$	0.01574
Flow direction	Normal to inlet	
$Y_{\rm CH_{3.783}}$	0.894	0
$Y_{O_2}$	0.003601	1
$Y_{N_2}$	0.0602	0
Turbulence intensity	20%	20%
Hydraulic diameter [m]	0.016	0.008
Total temperature [K]	300	300
Ext. black body temp. method	Boundary temp.	
Internal emissivity	1	1

Table 6.4: Inlet boundary conditions.

Boundary type	Pressure outlet
$Y_{ m N_2}$	0.02
$Y_{\rm H_2O}$	0.45
Backflow turbulent kinetic energy $\left[\frac{m^2}{s^2}\right]$	1
Backflow turbulent dissipation rate $\left[\frac{m^2}{s^3}\right]$	1
Backflow total temperature [K]	1500
Ext. black body temp. method	Boundary temp.
Internal emissivity	1

Table 6.5: Outlet boundary conditions.

$$l = 0.07 D_H$$
 (6.29)

where  $D_H$  [m] is the hydraulic diameter of the inlet given in Table 6.4. This approach to estimation of k and  $\epsilon$  at the inlet assumes fully developed flow. In order to investigate the effect of the inlet turbulence intensity on the simulation results obtained, simulations have been performed with a turbulence intensity of 5%. This turbulence intensity is based upon an estimation of the turbulent intensity at the core of fully developed pipe flow obtained with the empirical correlation given in Eq. 6.30 from ANSYS Inc. [2009].

$$I_{turb} = \frac{U'_{\rm rms}}{\overline{U}} = \frac{0.16}{(Re_H)^{\frac{1}{8}}}$$
(6.30)

where  $Re_{D_H}$  is the Reynolds number based on hydraulic diameter. Using the Reynolds numbers given in Table 3.4 for the gas and oxygen flows an estimate of 3.7% and 4% respectively has been found. An intensity of 5% has been chosen as it is exactly 1/4 of the turbulence intensity otherwise employed and is approximately equal to the two estimated values.

During the mesh independence study initially a heat flux of  $-35 \left[\frac{kW}{m^2}\right]$  was specified for

Horiz. wall	Vert. wall	Bottom wall	Top wall	Chimney	Nozzle
0	0	0	0	0	0
0.5	0.5	0.5	0.5	0.5	0.5
Eq. 6.31	Eq. 6.31	-	-	-	-
-	-	0	0	0	0
0.6	0.6	0.6	0.6	0.4	0.6
	Horiz. wall 0 0.5 Eq. 6.31 - 0.6	Horiz. wall         Vert. wall           0         0           0.5         0.5           Eq. 6.31         Eq. 6.31           -         -           0.6         0.6	Horiz. wall         Vert. wall         Bottom wall           0         0         0           0.5         0.5         0.5           Eq. 6.31         Eq. 6.31         -           -         0         0           0.6         0.6         0.6	Horiz. wall         Vert. wall         Bottom wall         Top wall           0         0         0         0           0.5         0.5         0.5         0.5           Eq. 6.31         Eq. 6.31         -         -           -         0         0         0           0.6         0.6         0.6         0.6	Horiz. wall         Vert. wall         Bottom wall         Top wall         Chimney           0         0         0         0         0           0.5         0.5         0.5         0.5         0.5           Eq. 6.31         Eq. 6.31         -         -         -           -         0         0         0         0           0.6         0.6         0.6         0.4         0.4

Table 6.6: Wall boundary conditions. A temperature is specified for the horizontal and vertical walls and for the bottom wall, top wall, chimney and nozzle a heat flux is specified.

the horizontal and vertical walls shown in Figure 3.4. This value is based on the average heat flux determined from the total furnace heat transfer measured by Lallemant et al. [1997]. Simulations converged slowly and the boundary condition was changed to the temperature boundary condition to determine whether this would increase the rate of convergence. Simulations converged significantly faster and a temperature boundary condition has been employed for all simulations as a result. The wall temperature boundary condition determines the heat flux from the flow conditions calculated and the specified wall temperature. Contours of the vertical wall heat flux for the mesh independent solution, determined from the mesh independence study described in Section 6.8, are shown in Figure 6.1. It is apparent from Figure 6.1 that the wall heat flux is not uniform with the largest heat flux near the flame and the lowest heat flux at near the chimney. The total furnace wall heat transfer for the mesh independent solutions is 540 kW. The total heat transfer for the specified heat flux is 505 kW. The 1% difference between the two boundary conditions is considered to be neglible. The heat flux boundary condition results in a lower heat flux near the flame and a higher heat flux near the chimney compared to the heat flux resulting from the temperature boundary condition. This is likely the cause for the slow simulation convergence. The wall temperature is determined from the polynomial fit by Bollettini et al. [1997] of measured wall temperatures by Lallemant et al. [1997] given in Eq. 6.31. A user defined function (UDF) is used to specify the wall temperature in the CFD simulations based on Eq. 6.31. The UDF

$$T_{\text{wall}} = 1.700598 \cdot 10^3 + 2.125872 \cdot 10^2 \cdot z - 4.666929 \cdot 10^1 \cdot z^2 \tag{6.31}$$

where z; [m] is the distance along the wall measure from the furnace bottom.

#### 6.8 Mesh Independence

As a mesh cell number increases a CFD solution will in general become more accurate as stated by Casey et al. [2000]. As the mesh cell number increases the solution should at some mesh cell number converge and remain constant for larger mesh cell numbers. This is the less stringent definition of a mesh independent solution as discussed by AIAA [1998]. A mesh independence study has been performed in order to identify which of the meshes presented in Section 3.2 yields a mesh independent solution.

The WD mechanism has been used for the simulations with the EDC and  $k - \epsilon$  turbulence and new WSGGM models. The boundary conditions given in Table 6.4, 6.5



Figure 6.1: Contours of heat flux for the vertical wall from the mesh independent simulation from the mesh independence study.

and 6.6 have been used along with the material properties in Table 6.3. Ideally a mesh independence study for each case should be performed. This is however not possible due to the time required. It is therefore assumed that the mesh found to yield a mesh independent solution for the current case will also yield a mesh independent solution for the current case will also yield a mesh independent solution for the current case will also yield a mesh independent solution for the other simulations performed. Convergence of the solution has been monitored with the temperature at 0.22, 0.82, 1.42 and 2.21 [m] downstream of the burner on the centerline and the average temperature on the vertical symmetry plane. Temperature has been chosen because it is dependent on the energy equation and the heat release from the combustion reactions. Ideally all variables of interest should be monitored. However, this leads to a large amount of data handling. The mass conservation and total heat conservation values are also inspected when the simulation is believed to have converged. The same variables and points are monitored for convergence in subsequent simulations. Plots of the temperature at the monitored points for each mesh are shown in Figure 6.2, 6.3, 6.4, 6.5 and 6.6.



Figure 6.2: Temperature on at z = 0.22 [m] on the centerline as a function of iterations.

The species mass flow rates and temperature at the furnace outlet solution obtained for each mesh are given in Table 6.7. From the converged values shown in Figure 6.2 to 6.6, the values given in Table 6.7 and comparison of radial plots of velocities, temperatures and species at the at the monitored points mesh q2 is determined to yield a mesh independent solution and this mesh has been used for all subsequent simulations.

#### 6.9 Simulation Cases

A summary of the cases simulated is given in Table 6.8. The case names are referenced in Chapter 7. The default simulation models and settings discussed in the preceding sections are used where nothing else is specified. The default models are the  $k - \epsilon$ turbulence model, the EDC model and the modified WD mechanism. In addition to cases summarized in Table 6.8 other simulations have been performed. Simulations performed with the modified WD mechanism, modified WD B mechanisms, QUICK discretization, DO angular discretization increased and lowered ISAT error tolerance



Figure 6.3: Temperature on at z = 0.82 [m] on the centerline as a function of iterations.



Figure 6.4: Temperature on at z = 1.42 [m] on the centerline as a function of iterations.



Figure 6.5: Temperature on at z = 2.21 [m] on the centerline as a function of iterations.



Figure 6.6: Average temperature on the vertical symmetry plane as a function of iterations.

Mesh	q	q1	q2	q3	q4
Avg. $T$ [K]	1941	1941	1941	1941	1941
$\dot{m}_{\rm CH_{3.783}} \left[ \frac{\rm kg}{\rm s} \right]$	0	0	0	0	
$\dot{m}_{\rm O_2} \left[\frac{\rm kg}{\rm s}\right]$	$3.208\cdot 10^{-4}$	$3.183\cdot10^{-4}$	$3.108\cdot 10^{-4}$	$3.129\cdot 10^{-4}$	$3.109\cdot10^{-4}$
$\dot{m}_{\rm CO}\left[\frac{\rm kg}{\rm s}\right]$	$2.498\cdot 10^{-4}$	$2.495\cdot10^{-4}$	$2.511\cdot 10^{-4}$	$2.506\cdot 10^{-4}$	$2.501\cdot 10^{-4}$
$\dot{m}_{N_2} \left[\frac{kg}{s}\right]$	$2.647\cdot 10^{-4}$	$2.656\cdot 10^{-4}$	$2.655\cdot 10^{-4}$	$2.656\cdot 10^{-4}$	$2.666\cdot 10^{-4}$
$\dot{m}_{\rm H_2O} \left[\frac{\rm kg}{\rm s}\right]$	$8.477\cdot 10^{-3}$	$8.503\cdot10^{-3}$	$8.505\cdot10^{-3}$	$8.507\cdot 10^{-3}$	$8.539\cdot 10^{-3}$
$\dot{m}_{\rm CO_2} \left[ \frac{\rm kg}{\rm s} \right]$	$1.065\cdot 10^{-2}$	$1.062\cdot 10^{-2}$	$1.063\cdot 10^{-2}$	$1.063\cdot 10^{-2}$	$1.060\cdot10^{-2}$
$\dot{m} \left[\frac{\text{kg}}{\text{s}}\right]$	$2\cdot 10^{-2}$	$2 \cdot 10^{-2}$	$2 \cdot 10^{-2}$	$2 \cdot 10^{-2}$	$2 \cdot 10^{-2}$

Table 6.7: Mesh independence study outlet flow properties.

Case name	Settings
WD	WD mechanism (mesh independent solution obtained)
I=5%	Mod. WD, $I_{turb} = 5\%$
Rea	Mod. WD, realizable turbulence model
Rea, $I=5\%$	Mod. WD, realizable turbulence model, $I_{turb} = 5\%$
Def. WSGGM	Mod. WD, Default WSGGM
Transient	Transient, mod. WD, $I_{turb} = 5\%$
ED	Eddy dissipation, mod. WD, $I_{turb} = 5\%$

Table 6.8: Simulation cases.

respectively have been found to yield the same results as case WD or yielded results with no significant difference.

### Chapter 7

### Results

In this chapter the results of the CFD simulations performed for each case discussed in Section 6.9 are presented. The velocity, velocity fluctuations, temperature and mole fractions of  $CH_4$ ,  $O_2$ , CO,  $N_2$ ,  $CO_2$  obtained from the CFD simulations are compared with measurements from Lallemant et al. [1997] and discussed.

As mentioned in Section 6.9, results from simulations with the modified WD mechanism, modified WD B mechanism, QUICK discretization, DO with increased angular subdivisions and a lowered ISAT error tolerance respectively show no significant difference when compared to the WD case. The results have been compared with respect to the same variables discussed in the following sections. Due to the number of cases, the simulation results which exhibit no difference with respect to the WD case have been omitted in the following graphs comparing results. The def. WSGGM case results have been included in the temperature graphs only. For all other variables the results from this case show no difference to the WD case.

From the results of CFD simulations by Andersen et al. [2009] the mod. WD is expected to yield both more accurate temperature and  $X_{\rm CO}$  results. The absence of these results seem to indicate that changes in the reaction kinetics in the current test case have are unimportant for the reaction set used in the WD mechanisms. ISAT error tolerance is judged to be irrelevant to the lack of change between the WD, modified WD and modified WD B mechanism predicted species concentrations due to the simulation with lowered ISAT error tolerance having no effect on concentrations. It is therefore assumed that lowered ISAT error tolerances would not have any effect on simulations with the other mechanisms.

#### 7.1 Axial Velocity

Figure 7.1 to 7.3 show the axial velocity  $w\left[\frac{m}{s}\right]$  at 0.22, 0.82 and 1.42 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997]. At 0.22 [m] the best agreement is obtained for the Rea  $I_{turb} = 5\%$  case. Both the spreading rate of the jet and core velocity is matched. The ED case is also in good agreement, matching the spreading rate and slightly overpredicting the core jet velocity while the  $I_{turb} = 5\%$  and transient simulations slightly under predict the core velocity. The WD and Rea simulations match the core velocity but over predict the spreading rate. At 0.82 and 1.42 [m] the primary difference in results is at the jet core. At 0.82 all cases are in good agreement with measurements for x > 0.05. For x < 0.05 all simulations over predict the centerline velocity with the WD simulation in closest agreement with measurements. At 1.42 [m] all cases under predict the velocity for x > 0.05 and are spread above and below at the centerline. These results indicate that turbulence intensity has as great an effect on predicting spreading rate as using the realizable  $k - \epsilon$  model. Neither parameter can model the core flow precisely although good agreement is obtained. All simulations yield accurate results as the distance from the centerline increasess.



Figure 7.1: Comparison of w at z = 0.22 [m].

#### 7.2 Fluctuating Velocity

Figure 7.4 to 7.6 show the rms fluctuating velocity  $U'_{\rm rms} \left[\frac{\rm m}{\rm s}\right]$  at 0.22, 0.82 and 1.42 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997].  $U'_{\rm rms}$  is determined from equation Eq. 7.1.  $U'_{\rm rms}$  results are an indicator of turbulence prediction. At 0.22 [m] all simulations exhibit the approximate trend in the measurements with the WD simulation in closes followed by the transient simulation. All simulations under predict  $U'_{\rm rms}$  at x > 0.1 and the same is true for most simulations in the peak region. At 0.82 and 1.42 [m] all simulations approximate the trend in exhibited by the measurements but under predict the turbulence levels. The discrepancy is greates far form the centerline. It can be concluded no simulation is clearly more accurate in predicting the fluctuating velocity.

$$U_{\rm rms}' = \sqrt{\frac{2k}{3}} \tag{7.1}$$



Figure 7.2: Comparison of w at z = 0.82 [m].



Figure 7.3: Comparison of w at z = 1.42 [m].



Figure 7.4: Comparison of  $V'_{\rm rms}$  at  $z = 0.22 \ [m]$ .



Figure 7.5: Comparison of  $V'_{\rm rms}$  at  $z = 0.82 \ [m]$ .



Figure 7.6: Comparison of  $V'_{\rm rms}$  at  $z = 1.42 \ [m]$ .

#### 7.3 Temperature

Figure 7.7 to 7.9 show the temperature T at 0.22, 0.82 and 1.42 [m] downstream of the burner as a function of radial distance from the centerline compared with data from Bollettini et al. [1997]. At 0.22 [m] all simulations except ED significantly over predict the peak temperature. Peak temperatures above the adiabatic flame temperature with only WD species included are reached. The Rea, def. WSGGM and WD simulations predict a peak temperature of 4500 [K], approximately 750 [K] higher than the adiabatic flame temperature.  $I_{turb} = 5\%$ , Rea  $I_{turb} = 5\%$  and the transient simulations yield lower peak temperature predictions. The ED under predicts the peak temperature but is in closest agreement. All simulations exhibit the correct temperature trend and  $I_{turb} = 5\%$ , Rea  $I_{turb} = 5\%$ , ED and the transient predict the temperature accurately at the centerline. At x > 1 all simulation yield the correct temperature. At 0.82 and 1.42 [m] all simulations again show the same trend as the measurements but over predict temperatures significantly. Near the burner it can be concluded that the ED, transient and  $I_{turb} = 5\%$  simulations provide the most accurate results but fail on an equal footing with the other simulations at all other downstream distances. The default WSGGM results in a peak temperature at 0.22 [m] 100 [K] higher compared to the oxy WSGGM and at 0.82 [m] 50 [K] higher. Thus the minimal effect predicted due to the relatively short mean beam length holds.



Figure 7.7: Comparison of T at z = 0.22 [m].



Figure 7.8: Comparison of T at z = 0.82 [m].


Figure 7.9: Comparison of T at z = 1.42 [m].

### **7.4** $X_{CH_4}$

Figure 7.10 to 7.13 show the dry mole fraction of  $CH_4$  at 0.22, 0.82, 1.42 and 2.21 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997]. At 0.22 [m] all simulations under predict  $X_{CH_4}$ . The transient,  $I_{turb} = 5\%$  and ED are the most accurate simulations. At 0.82 the ED simulation predicts a higher  $X_{CH_4}$  than measured while all other simulations yield a lower  $X_{CH_4}$ . The transient and  $I_{turb} = 5\%$  simulations are the most accurate. At 1.42 [m] the ED simulations still predicts  $CH_4$  to be present where the is none. The rate of reaction of  $CH_4$  therefore seems to most accurately modelled by the  $I_{turb} = 5\%$  and transient simulations.

#### **7.5** $X_{O_2}$

Figure 7.14 to 7.17 show the dry mole fraction of  $O_2$  at 0.22, 0.82, 1.42 and 2.21 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997]. At 0.22 [m] the Rea and WD simulations show incorrect trends in  $X_{O_2}$ . The remaining simulations better predict the trend but under predict the concentration. At 0.82 [m] all simulations predict a significantly lower  $O_2$  concentration and the same is the case at 1.42 [m]. At 2.21 [m] the predictions of all but the ED simulation are accurate.



Figure 7.10: Comparison of  $X_{CH_4}$  dry at  $z = 0.22 \ [m]$ .



Figure 7.11: Comparison of  $X_{\text{CH}_4}$  dry at  $z = 0.82 \ [m]$ .



Figure 7.12: Comparison of  $X_{CH_4}$  dry at  $z = 1.42 \ [m]$ .



Figure 7.13: Comparison of  $X_{CH_4}$  dry at  $z = 2.21 \ [m]$ .



Figure 7.14: Comparison of  $X_{O_2}$  dry at z = 0.22 [m].



Figure 7.15: Comparison of  $X_{\mathrm{O}_2}$  dry at  $z=0.82\;[m].$ 



Figure 7.16: Comparison of  $X_{O_2}$  dry at  $z = 1.42 \ [m]$ .



Figure 7.17: Comparison of  $X_{O_2}$  dry at  $z = 2.21 \ [m]$ .

#### **7.6** *X*<sub>CO</sub>

Figure 7.18 to 7.21 show the dry mole fraction of CO at 0.22, 0.82, 1.42 and 2.21 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997]. At 0.22 [m] the Rea and WD cases predict a far too high concentration of CO indicating that combustion is concentrated in this region for the two simulations. The remaining under predict the peak concentration. At 0.82 [m] all but Rea and ED simulations provide reasonable estimates of CO. At 1.42 [m] the predicted CO level is reasonable but the distribution is too uniform with respect to radial position. At 2.21 [m] the concentration is well predicted. The equilibrium concentration of CO for the temperature at 1.42 [m] and 2.21 [m] is in reasonable agreement with the actual concentration measured.



Figure 7.18: Comparison of  $X_{\rm CO}$  dry at  $z = 0.22 \ [m]$ .

#### **7.7** $X_{N_2}$

Figure 7.22 to 7.25 show the dry mole fraction of  $N_2$  at 0.22, 0.82, 1.42 and 2.21 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997]. As  $N_2$  does not react it provides an evaluation of the turbulent species transport. The Rea simulation and WD simulation do not predict the correct trend at 0.22 [m] while the remaining simulations approach the correct trend while over predicting the concentration close to the centerline. At 0.82 [m] the  $N_2$  concentrations are more uniform than those measured and are over predicted. As distance downstream increases the correct distribution and concentration of  $N_2$ .



Figure 7.19: Comparison of  $X_{\rm CO}$  dry at  $z = 0.82 \ [m]$ .



Figure 7.20: Comparison of  $X_{\rm CO}$  dry at  $z = 1.42 \ [m]$ .



Figure 7.21: Comparison of  $X_{\rm CO}$  dry at  $z = 2.21 \ [m]$ .



Figure 7.22: Comparison of  $X_{N_2}$  dry at  $z = 0.22 \ [m]$ .



Figure 7.23: Comparison of  $X_{N_2}$  dry at  $z = 0.82 \ [m]$ .



Figure 7.24: Comparison of  $X_{\mathrm{N}_2}$  dry at  $z=1.42\;[m].$ 



Figure 7.25: Comparison of  $X_{N_2}$  dry at z = 2.21 [m].

### **7.8** $X_{\rm CO_2}$

Figure 7.26 to 7.29 show the dry mole fraction of  $N_2$  at 0.22, 0.82, 1.42 and 2.21 [m] downstream of the burner as a function of radial distance from the centerline compared with measurements from Lallemant et al. [1997]. All simulations yield the correct trend of  $CO_2$  but over predict the concentrations at all cnterline distances except at 0.22 [m] where good agreement is achieved for all simulations.

#### 7.9 Remarks

The velocities and fluctuating velocities are in general predicted well by all simulations. The realizable  $k - \epsilon$  model and a turbulence intensity of 5% most accurately predict the spreading rate of the jet. They provide no improvement in predicting the fluctuating velocities. The temperatures are in general poorly predicted by all simulations. The peak temperatures are much greater than those measured. Near the burner the transient and 5% intensity simulation are more accurate and predict the trend well. Further downstream neither simulation performs better than the other simulations. CH<sub>4</sub> is predicted to burn to rapidly. The transient and 5% simulations provide the slowest decrese in CH<sub>4</sub>. The rapid consumption of CH<sub>4</sub> is confirmed by the low O<sub>2</sub> predictions at all points. The ED simulation poorly predicts CO as expected. Turbulent mixing is not modelled accurately based on the concentration distribution of inert N<sub>2</sub>. The transient simulation is found to yield a different result from the corresponding steady state simulation with a 5% inlet turbulence intensity. However the simulation does not provide more accurate predictions and does not yield significant differences.



Figure 7.26: Comparison of  $X_{\text{CO}_2}$  dry at  $z = 0.22 \ [m]$ .



Figure 7.27: Comparison of  $X_{{\rm CO}_2}$  dry at z=0.82~[m].



Figure 7.28: Comparison of  $X_{\text{CO}_2}$  dry at  $z = 1.42 \ [m]$ .



Figure 7.29: Comparison of  $X_{\text{CO}_2}$  dry at  $z = 2.21 \ [m]$ .

Further transient simulations should be performed with smaller time steps to determine the effect of the time step on accuracy. The CO concentrations downstream are in good agreement with equilibrium concentrations at the temperatures found at these locations. This can be taken to indicate that equilibrium calculations provide a reasonable estimate of the dissociation species. Furthermore, the peak temperature measured is approximately 2750 [K]. The adiabatic flame temperature is approxiantely 3000 [K] indicating that the flame temperature in the furnace is predicted well by equilibrium calculations. It can be concluded that CFD calculations using the mixture fraction approach with an equilibrium chemistry model seem likely to predict temperature and dissociation species well in the furnace and should be investigated.

# Chapter 8

## Conclusion

The investigation documented in this report has sought to determine the accuracy of CFD simulations using the RANS approach. In particular, the effect of different models used in the RANS CFD simulations, on the accuracy of results obtained has been investigated. The IFRF OXYFLAM-2 furnace experiments with oxy-natural gas combustion have been chosen as the test case for the CFD simulations. The OXYFLAM-2 experiments have been chose due to the in furnace data available for comparison. Based upon the geometry of the furnace and measurements reported by Lallemant et al. [1997], vertical and horizontal symmetry has been assumed to reduce the size of the meshes created for the CFD simulations. The validity of the symmetry assumption has not been verified. Meshes of the top left quarter of the furnace have been constructed.

The combustion chemistry of oxy-natural gas has been investigated with chemical equilibrium calculations using the CEA software. The adiabatic flame temperature for airnatural gas combustion with all species included has been determined. The oxy-natural gas combustion adiabatic flame temperature, with the species included varied between all species and those present in the Westbrook-Dryer and Jones-Lindstedt mechanisms, has been determined. Oxy-natural gas has been found to have a significantly higher adiabatic flame temperature than air-natural gas combustion. The adiabatic flame temperature for oxy-natural gas combustion is significantly higher when species are limited to those present in the WD and JL mechanisms. Limiting of species to those present in the WD mechanism yields the highest adiabatic flame temperature. The equilibrium composition of oxy-natural gas combustion as a function of temperature has been investigated. The importance of dissociation has been found to increase with temperature and the mole fraction of dissociation species CO,  $O_2$ ,  $H_2$  and OH increases significantly. The JL mechanism is therefore expected to yield the most accurate CFD calculations. A numerical fuel model of the natural gas used in the OXYFLAM-2 experiments has been calculated in order to use the WD mechanism.

The weighted sum of grey gases model is used to model the radiative transfer properties of the furnace gas mixture. A WSGGM applicable to oxy-fuel conditions is presented. The new WSGGM is found unlikely to produce different results from the WSGGM for air combustion conditions. This is due to the relatively short mean beam length in the chosen test case.

The RANS approach has been chosen for the CFD simulations of the test case. The  $k - \epsilon$  and realizable  $k - \epsilon$  turbulence models have been used. The discrete ordinates

method is used to solve the radiative transfer equation. The eddy dissipation and eddy dissipation concept models have been used to model reactions and the chemistryturbulence interaction. A mesh independence study has been performed in order to identify which of the meshes constructed lead to a mesh independent solution. The mesh q2 was found to fulfill this requirement and has been used for all subsequent simulations.

Simulations employing different models and settings for turbulence, reaction mechanism, chemistry-turbulence interaction, radiation properties, discretization, chemistry integration and radiative transfer have been performed. The reaction kinetics of the WD mechanisms employed have been found to have no effect on the results indicating that mixing is likely the limiting factor. It has been found that velocities and velocity fluctuations are predicted as accurately with a lower inlet turbulence intensity and the  $k - \epsilon$  model as with the realizable  $k - \epsilon$ . For the species concentrations, best agreement has been found for a lower inlet turbulence intensity and for the transient calculations. The higher turbulence intensity simulations predicts a more rapid  $CH_4$  consumption than that measured at the burner. Downstream the lower turbulence simulations also over predict the reaction of  $CH_4$ . The rapid reaction of  $CH_4$  causes  $O_2$  to be under predicted. Turbulent mixing is evaluated from N<sub>2</sub> concentrations be poorly predicted for all simulations. A more uniform turbulent species transport is predicted compared to measurements  $N_2$ . CO is over predicted by for the high turbulence inlet intensity and under predicted for the low inlet turbulence intensity. In general it is concluded that inlet turbulence intensity affects the accuracy of simulations more than any other parameter investigated. Accurate modelling of inlet turbulence is therefore vital to accurate simulations.

The JL mechanism failed due to a program error and has therefore not been evaluated. The JL mechanism is expected to provide more accurate temperature and dissociation species concentration predictions due to the inclusion of  $H_2$ . This is based upon chemical equilibrium calculations. Transient calculations with smaller time steps are needed to evaluate the effect of the time step on accuracy. The measured concentrations of CO and flame temperature have been found to be in good agreement with equilibrium calculations. In light of the success of the equilibrium approach in the literature and the findings in this report, CFD simulations using the mixture fraction approach with equilibrium chemistry calculations are found likely to provide more accurate results than those obtained in the present investigation.

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