To my dear husband Rumaizi and son Uwais
Combustion of gasified biomass:
Experimental investigation on laminar flame speed, lean blowoff limit and emission levels

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ABSTRACT

Biomass is among the primary alternative energy sources that supplements fossil fuels to meet today’s energy demand. Gasification is an efficient and environmentally friendly technology for converting the energy content in biomass into a combustible gas mixture, which can be used in various applications. The composition of this gas mixture varies greatly depending on the gasification agent, gasifier design and its operation parameters, and can be classified as low and medium LHV gasified biomass. The wide range of possible gas composition between each of these classes and even within each class itself can be a challenge in combustion for heat and/or power production. The difficulty is primarily associated with the range in the combustion properties that may affect the stability and the emission levels. Therefore, this thesis is intended to provide data of combustion properties for improving the operation and design of atmospheric combustion devices operated with such gas mixtures.

The first part of this thesis presents a series of experimental work on combustion of low LHV gasified biomass (a simulated gas mixture of CO/H₂/CH₄/CO₂/N₂) with variation in the content of H₂O and tar compound (simulated by C₆H₆). The laminar flame speed, lean blowoff limit and emission levels of low LHV gasified biomass based on the premixed combustion concept are reported in paper I and III. The results show that the presence of H₂O and C₆H₆ in gasified biomass can give positive effects on these combustion parameters (laminar flame speed, lean blowoff limit and emission levels), but also that there are limits for these effects. Addition of a low percentage of H₂O in the gasified biomass resulted in almost constant laminar flame speed and combustion temperature of the gas mixture, while its NOₓ emission and blowoff temperature were decreased. The opposite condition was found when H₂O content was further increased. The blowoff limit was shifted to richer fuel equivalence ratio as H₂O increased. A temperature limit was observed where CO emission could be maintained at low concentration. With C₆H₆ addition, the laminar flame speed first decreased, achieved a minimum value, and then increased with further addition of C₆H₆. The combustion temperature and NOₓ emission were increased, CO emission was reduced, and blowoff occurs at slightly higher equivalence ratio and temperature when C₆H₆ content is increased. The comparison with natural gas (simulated by CH₄) is also made as can be found in paper I and II. Lower laminar flame speed, combustion temperature, slightly higher CO emission, lower NOₓ emission and leaner blowoff limit were obtained for low LHV gas mixture in comparison to natural gas.
In the second part of the thesis, the focus is put on the combustion of a wide range of gasified biomass types, ranging from low to medium LHV gas mixture (paper IV). The correlation between laminar flame speed or lean blowoff limit and the composition of various gas mixtures was investigated (paper IV). It was found that H2 and content of diluents have higher influence on the laminar flame speed of the gas mixture compared to its CO and hydrocarbon contents. For lean blowoff limit, the diluents have the greatest impact followed by H2 and CO. The mathematical correlations derived from the study can be used to for models of these two combustion parameters for a wide range of gasified biomass fuel compositions.

**Keywords:** biomass gasification; gasified biomass; premixed combustor; laminar flame speed; blowoff; CO, UHC and NOx emissions
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PREFACE

This thesis is based on the following papers, which are referred to in the text by their Roman numerals and appended at the end of the thesis.


The contribution of the thesis author (first author of papers I - IV):

**Paper I:** First author was a main author; experimental work and analysis were done by the first author. Research ideas are from second and third authors. Second author also acted as the main mentors and reviewers. Fourth author acted as a reviewer.

**Paper II – III:** First author was a main author; experimental work and analysis were performed by the first author. Second author acted as the main mentor and reviewer. Third author acted as a reviewer.

**Paper IV:** First author was a main author; the data collection and analysis were performed by the first author. Second author acted as the main mentor and reviewer. Third author acted as a reviewer.
# TABLE OF CONTENTS

## ABSTRACT ......................................................................................................................... 1

## ACKNOWLEDGEMENTS ................................................................................................. 3

## PREFACE .......................................................................................................................... 4

## TABLE OF CONTENTS ....................................................................................................... 5

## INDEX OF TABLES ............................................................................................................. 7

## INDEX OF FIGURES ......................................................................................................... 7

## NOMENCLATURE ............................................................................................................... 7

### 1.0 INTRODUCTION .............................................................................................................. 8

#### 1.1 BIOMASS IN GENERAL .............................................................................................. 10

#### 1.2 BIOMASS GASIFICATION – GENERAL DESCRIPTION .................................................. 12

##### 1.2.1 Gasification process ............................................................................................ 12

##### 1.2.2 Gasifier designs ............................................................................................... 15

#### 1.3 GASIFIED BIOMASS GAS – INFLUENCING FACTORS .................................................. 18

##### 1.3.1 Biomass feedstock ............................................................................................ 18

##### 1.3.2 Gasification agent ............................................................................................ 21

##### 1.3.3 Gasifier operating conditions .......................................................................... 23

#### 1.4 COMBUSTION OF GASIFIED BIOMASS GAS ................................................................. 26

##### 1.4.1 Combustor concepts for gasified biomass .............................................................. 27

##### 1.4.2 Fundamental combustion properties of gasified biomass gas ................................. 31

##### 1.4.3 Combustor operability issues ............................................................................ 34

##### 1.4.4 Combustion emissions ..................................................................................... 37

#### 1.5 OBJECTIVES ............................................................................................................... 42

### 2.0 RESEARCH APPROACH ............................................................................................... 44

#### 2.1 INFLUENCE OF WATER VAPOUR AND TAR COMPOUND ON LAMINAR FLAME SPEED OF GASIFIED BIOMASS GAS (PAPER I) .............................................................................................................. 46

##### 2.1.1 Objectives of paper I ......................................................................................... 46

##### 2.1.2 Research approach of paper I ............................................................................. 46

#### 2.2 EMISSION CHARACTERISTICS AND LEAN BLOWOFF LIMIT OF SIMULATED GASIFIED BIOMASS GAS IN A PREMIXED COMBUSTOR .................................................................................................................. 48

##### 2.2.1 Objectives of paper II ......................................................................................... 48


2.2.2 Research approach of paper II ................................................................. 48

2.3 INFLUENCE OF WATER VAPOUR AND TAR COMPOUND ON COMBUSTION OF SIMULATED GASIFIED BIOMASS GAS ......................................................................................................................... 50
  2.3.1 Objectives of paper III ................................................................................. 50
  2.3.2 Research approach of paper III ....................................................................... 50

2.4 CORRELATIONS OF LAMINAR FLAME SPEED AND LEAN BLOWOFF LIMIT WITH THE FUEL COMPOSITION OF GASIFIED BIOMASS ........................................................................................................ 51
  2.4.1 Objectives of paper IV .................................................................................... 51
  2.4.2 Research approach of paper IV ........................................................................ 51

3.0 MAIN RESULTS .................................................................................................. 52
  3.1 INFLUENCE OF WATER VAPOUR AND TAR COMPOUND ON LAMINAR FLAME SPEED OF GASIFIED BIOMASS GAS (PAPER I) ........................................................................................................ 52
  3.2 EMISSION CHARACTERISTICS AND LEAN BLOWOFF LIMIT OF SIMULATED GASIFIED BIOMASS GAS IN A PREMIXED COMBUSTOR (PAPER II) ........................................................................................................ 54
  3.3 INFLUENCE OF WATER VAPOUR AND TAR COMPOUND ON COMBUSTION OF SIMULATED GASIFIED BIOMASS GAS (PAPER III) ........................................................................................................ 56
  3.4 CORRELATIONS OF LAMINAR FLAME SPEED AND LEAN BLOWOFF LIMIT WITH THE FUEL COMPOSITION OF GASIFIED BIOMASS (PAPER IV) ..................................................................................... 58

4.0 DISCUSSION ....................................................................................................... 60

5.0 CONCLUSIONS AND FUTURE WORK .............................................................. 63
  5.1 CONCLUSION ..................................................................................................... 63
  5.2 FUTURE WORK .................................................................................................. 65

6.0 REFERENCES ..................................................................................................... 66
INDEX OF TABLES

Table 1: Formation temperature and composition of tars .............................................. 13
Table 2: Gasified biomass composition for different gasifier agents used .................... 22

INDEX OF FIGURES

Figure 1: Gasifier designs .................................................................................................. 17
Figure 2: Illustration of simple non-premixed and premixed combustors .................. 27
Figure 3: Combustor design concept .................................................................................. 29

NOMENCLATURE

BFB  Bubbling fluidized bed
CFB  Circulating fluidized bed
ER  Equivalence ratio
ERblowoff  Blowoff equivalent ratio
FL  Flammability limit
GHG  Greenhouse gas
LFL  Lower flammability limit [% by volume]
LHV  Lower heating value [MJ/Nm³]
PAH  Polycyclic aromatic hydrocarbons
SBR  Steam-to-biomass ratio
UFL  Upper flammability limit [% by volume]
WGS  Water-gas-shift
λ  Air – to – biomass weight ratio (1/ER)
SL  Laminar flame speed
1.0 INTRODUCTION

Energy supply and global warming have become subjects of major international concern. Growth in the world’s population and economies are key drivers of the trend of increasing global energy demand. However, with uncertainties in both quantities and price of fossil fuels, i.e. the dominant energy resources, alternative fuel sources are urgently needed. Furthermore, the build-up of greenhouse gases (GHG) in the atmosphere associated with combustion of fossil fuels strengthens the reason for the shift towards a sustainable energy supply with minimal negative environmental impact. For such purposes, biomass for energy conversion seems suitable. Energy from biomass, when produced and utilized on a sustainable basis, can possibly reduce undesired emissions, especially CO₂, which is the most significant contributor to GHG emissions (Gao et al., 2012) (Li et al., 2004). Biomass is a versatile energy source which can be found across the globe and can be used for production of power, heat, liquid and gaseous fuels, and chemical feedstock through a wide range of conversion technologies.

In 2010, the total world primary energy supply was 535.2 EJ and biomass fuel accounted for about 9.8 % of the world energy market (Statistics, 2012). Most biomass fuel consumption, especially in developing countries, is utilized traditionally, i.e. burned directly and usually in inefficient devices for cooking, heating and lighting, and for industrial applications. Only a small portion of it is used in a modern way, for electricity production and as transport fuel (Kaygusuz, 2012). However, in the near-future, the target is to increase modern applications of biomass energy, for example in India, China and Brazil (Faaij, 2006). Power generation from biomass by advanced combustion technology, especially gasification-based integrated system, has received a great deal of attention. In addition to the potential for gasifying biomass feedstock of different physical nature and chemical composition, the gasification process also expands the possible end-use applications of biomass. For example, direct use in combustion engines, gas turbines or for high temperature combustion processes. When used for power generation, gasification technology integrated with a combined cycle offers higher efficiency than the conventional biomass combustion steam cycle (Jurado et al., 2003) (Bhattacharya et al., 2011) (Demirbas et al., 2009). In addition, compared to direct combustion of solid biomass, combustion of gasified biomass will lead to higher temperatures and lower NOₓ, CO and particulate emissions (Dornburg & Faaij, 2001), (Hernandez et al., 2012).

Gasified biomass is a mixture of combustible and non-combustible gases as well as other contaminants including tar. The exact composition of gasified biomass is highly variable depending on the chemical composition of the biomass feedstock, gasifier configuration, gasifying agent and operating conditions. Therefore, in order to
efficiently exploit gasified biomass in combustion systems, it is necessary to carry out studies about the combustion properties of gasified biomass to ensure smooth and reliable operation in combustion devices regardless of the resulting gas composition from the gasifier. Besides that, the combustion devices should also able to operate on backup fuels such as natural gas in case the gasifier operation needs to be stopped, for example for maintenance. The variety in composition of gasified biomass and the necessity to also operate on a backup fuel can significantly affect the combustion process and the combustion products (Lieuwen et al., 2006), (Lewis et al., 2012).

Unlike natural gas or other single fuels (such as H₂) and binary fuel mixtures (such as CO/H₂ and CH₄/H₂), there are fewer studies associated with the combustion of gasified biomass available in open literature. This applies especially to a fuel mixture with main components of gasified biomass such as a mixture of CO/H₂/CH₄/CO₂/N₂ (Monteiro et al., 2010). Motivated by this factor, the scope of this doctoral study is to technically investigate the combustion behaviour of gasified biomass gas and its performance in an atmospheric small-scale combustor.
1.1 Biomass in general

There are several definitions of biomass used by researchers. Biomass is defined in Unified Bioenergy Terminology as “a material of biological origin excluding material embedded in geological formations and transformed to fossil” (FAO, 2004). Biomass can be classified into several categories such as (Panwar et al., 2012), (Demirbas et al., 2009):

- Woody biomass, including plantation wood (e.g. willow, poplar, eucalyptus) and forestry by-products (e.g. wood blocks, wood chips from logging and thinning)
- Agricultural biomass, including herbaceous crops (e.g. miscanthus, reed canary grass, giant reed); oil, sugar, and starch energy crops (e.g. rape seed, sunflower); agricultural residues (e.g. wheat straw, rice straw) and livestock matter (e.g. animal manure)
- Municipal and industrial waste, including any organic fraction of municipal (waste from households) and from industry (e.g. waste from food-processing industry, waste from agricultural industries); biodegradable landfilled waste; landfill gas and sewage sludge

Biomass is composed primarily of glucose polymers such as cellulose, hemicelluloses, and lignin, along with minor amounts of extractives (Pereira et al., 2012). Biomass generally has high volatile matter content (about 70 – 80 wt%) but low fixed carbon content (15 – 25 wt%). It also typically has high moisture content (for example freshly cut wood chips can have moisture content in the range of 30 to 50 wt%) and high oxygen content (about 35 – 43 wt%), which result in a low heating value (Demirbas, 2005) (Blasi, 2008) (Demirbas et al., 2009), (Kaewluan & Pipatmanomai, 2011). The higher heating value (HHV, the total energy content released when the fuel is burned in air, including the latent heat contained in the water vapour) of biomass is approximately 20 MJ/kg (Telmo et al., 2010). Due to this low heating value, more fuel is required to obtain the same energy from biomass in comparison to fossil fuels. Moreover, biomass in its original form is bulky and dispersed and has low bulk densities (for example 64 – 96 kg/m³ for loose, uncompacted straw), which makes it less efficient in transportation, storage and handling process (Badger & Fransham, 2006). Therefore, often the raw biomass undergoes pre-treatment processes that transform it into other forms such as chips, briquettes, bales and pellets, which are homogenized in shape and size as well as improved in bulk densities (Richard et al., 2012), (Kaliyan & Morey, 2009).

Biomass can be considered the most significant renewable energy source, providing about 9.8 % of the global total primary energy supply in 2010; the fourth largest source after oil, coal and natural gas (Statistics, 2012). A potential deployment level of biomass for energy is expected to keep growing in order to meet the rapid rise of
global energy demand. Emerging interest in biomass energy is driven by the following facts (Vaezi et al., 2012) (Bapat et al., 1997) (Radmanesh et al., 2006) (Faaij, 2006):

- It is available abundantly across the globe in versatile resources
- It is renewable, as the formation of biomass takes place within a short-time period, thus promising a constant supply of energy and reducing dependence on fossil fuels
- It is carbon neutral, as biomass takes carbon out of the atmosphere while it is growing and returns it during the thermal conversion process
- It can improve the management of resources and wastes
- It can enhance agricultural production
- It can promote rural development by creating jobs and income

Despite these advantages, biomass energy also has negative aspects (Sciling & Esmundo, 2009; and Gokcol et al., 2009):

- Compared to fossil fuels, the energy density of biomass is lower and thus more biomass is needed to obtain the same energy capacity. Consequently, the cost for biomass production, harvesting, collection, transportation and storage is higher.
- A large area is needed to grow biomass for energy purposes and thus results in competition of land use for other purposes such as production of food crops, housing or resorts. Furthermore, more water, fertilizers, herbicides and insecticides are required for the production of biomass plant.
- Ineffective use of biomass as a fuel can contribute to more soot, particles and other pollutant emissions.
- Intensive harvesting of plant based biomass can have negative impacts on the environment such as deforestation or soil erosion.

Currently, most biomass energy is used inefficiently for cooking and heating purposes, especially in developing countries such as India, Sri Lanka and Nepal. For example, typical efficiency of traditional biomass-fired cooking stoves is in the range of 5 – 20 % (Battacharya & Abdul Salam, 2002). Cooking with these traditional cooking stoves leads to the production of incomplete combustion products such as CO, N₂O, and PAH as well as fine and ultra-fine particles, which not only contribute to global warming potential but also are very dangerous to human health (Battacharya et al., 2000), (Miah et al., 2009). According to Hassan et al. (2009), housewives who are exposed to indoor air pollutants when working in the kitchen can potentially develop respiratory tract infections and lung cancer. To better utilize biomass and to reduce both environmental effects and the negative social and
health impacts, it is necessary to shift towards more efficient applications with higher biomass conversion ratios and lower emission levels.

Generally, biomass can be converted to a number of secondary energy carriers such as liquid and gaseous fuels, heat, electricity or chemical feedstock. This diversity in end-use options results from a wide range of conversion technologies available to make optimum use of energy from various biomass raw materials. These conversion routes can be categorized as thermal, chemical and biochemical conversion routes. Among thermal conversion processes, the gasification technology is particularly attractive for converting biomass into useful energy due to its varied end-use applications and benefits as mentioned previously (Alauddin et al., 2010) (Kirkels & Verbong, 2011) (Kitzler et al., 2011).

1.2 Biomass gasification – General description

Biomass gasification is the process of converting biomass feedstock into a gas mixture. Gasified biomass consists primarily of carbon monoxide (CO), hydrogen (H₂), carbon dioxide (CO₂), methane (CH₄), nitrogen (N₂) (if air was used as an gasification agent) and water vapour (H₂O) (Balat et al., 2009), (Demirbas et al., 2009). There are several types of gasifiers used for biomass gasification. In addition to the design of the gasifier, there are other factors influencing the composition of gasified biomass which include biomass feedstock, gasification agent and gasification operating conditions.

1.2.1 Gasification process

The gasification process is carried out at elevated temperatures and atmospheric or elevated pressures. Through this thermochemical process, solid biomass will undergo several sub-processes in the course of producing gasified biomass in the presence of a gasifying agent. These sub-processes are drying, pyrolysis, combustion (if air or oxygen is used as gasifying agent) and char gasification (Puig-Arnavaet et al., 2010).

_Drying_ is the evaporation process of the water content in biomass in presence of heat and normally takes place in a temperature range of 100 to 200°C (Kaushal et al., 2010), (Puig-Arnavaet et al., 2010). The heat needed for evaporation originates from the combustion step. High moisture content in the biomass fuel will result in a less efficient gasification system since much of the heat supplied for the gasification process is spent in evaporation thus leaving less heat for the pyrolysis and char gasification processes (see below) (Pereira et al., 2012). On the other hand, some moisture is needed during the char gasification process (see below) so that H₂ gas can be produced (Erlich, 2009), (McKendry, 2002).
Pyrolysis or devolatilization is a decomposition process of biomass in absence of an oxidizer that normally occurs at a temperature above that of drying and up to 500°C (Bridgwater, 2003), (Kaushal et al., 2010). The heat required for the pyrolysis originates from the combustion step. The products from the pyrolysis process are volatile gases, tars and char. Volatile gases consist of condensable and non-condensable species including CO, CO₂, H₂, H₂O and light hydrocarbons. Char is the solid portion of the biomass that remains as a result of incomplete conversion. Char has high content of elemental carbon. The reactivity of the char produced during pyrolysis is determined by the heating rate supplied. A high heating rate of the biomass particles can produce high-reactivity char (Okumura et al., 2009). Tar is the condensable fraction of organic compounds contained in the pyrolysis gas and mainly consists of aromatic hydrocarbons, which range from low molecular weight components like benzene to heavy polycyclic aromatic hydrocarbons (PAHs) (Zhang et al., 2011). In the gasification process, tar is first produced during the pyrolysis stage due to the presence of oxygen in the biomass feedstock and is called primary tar (Pan et al., 1999), (Basu, 2010). The primary tar thermally decomposes to smaller, lighter non-condensable gases (such as CO₂, CO, H₂O) and a series of higher hydrocarbons called secondary tar at temperatures above 600°C and to PAH compounds known as tertiary tar at temperatures above 800°C (Basu, 2010). At a high temperature and/or a long residence time, secondary tar also decomposes to tertiary tar. The decomposition of tar in biomass gasification can be due to the thermal cracking, steam reforming and dry reforming reactions (Li & Suzuki, 2009). Examples of conditions that favour these reactions are the increase in gasification temperature, pressure and equivalence ratio (refers to section 1.3) (Gómez-Barea et al., 2013). Table 1 shows these categories of tar compounds depending on its formation temperature and composition (Milne & Evans, 1998). Tar in gasified biomass is an undesired compound as when it condenses (at temperatures below 350 – 400°C) it can lead to, for example, the detriment of pipe lines as well as damage in end-use applications such as engines and turbines (Nemanova et al., 2011).

<table>
<thead>
<tr>
<th>Category</th>
<th>Formation temperature</th>
<th>Constituents</th>
</tr>
</thead>
<tbody>
<tr>
<td>Primary</td>
<td>400 – 600°C</td>
<td>Mixed oxygenates, phenolic ethers</td>
</tr>
<tr>
<td>Secondary</td>
<td>600 – 800°C</td>
<td>Alkyl phenolics, heterocyclic ethers</td>
</tr>
<tr>
<td>Tertiary</td>
<td>800 – 1000°C</td>
<td>Polycyclic aromatic hydrocarbons (PAH)</td>
</tr>
</tbody>
</table>

Combustion or oxidation is an exothermic reaction that occurs in presence of oxygen. During the combustion process, parts of the pyrolysis products are reacting with the
oxygen supplied. The principal oxidation reactions that may occur are as follows (Erlich, 2009), (Hernández et al., 2012), (Gómez-Barea et al., 2013):

Gas combustion:
CO + $\frac{1}{2}$O$_2$ → CO$_2$                \hspace{1cm} (R1)
H$_2$ + $\frac{1}{2}$O$_2$ → H$_2$O                \hspace{1cm} (R2)
CH$_4$ + 2O$_2$ → 2H$_2$O + CO$_2$                \hspace{1cm} (R3)
C$_2$H$_4$ + 3O$_2$ → 2H$_2$O + 2CO$_2$                \hspace{1cm} (R4)

Solid combustion:
C + $\frac{1}{2}$O$_2$ → CO                \hspace{1cm} (R5)
C + O$_2$ → CO$_2$                \hspace{1cm} (R6)

According to Gómez-Barea, et al. (2013), tars may also undergo a partial oxidizing process and produce CO, H$_2$ and heat.
Tars + O$_2$ → CO, H$_2$                \hspace{1cm} (R7)

Another important exothermic reaction in gasification is the water-gas shift (WGS) reaction where CO and H$_2$O react to form H$_2$ and CO$_2$:
H$_2$O + CO ↔ H$_2$ + CO$_2$                \hspace{1cm} (R8)

This reaction strongly affects the final composition of the gasified biomass, as a higher gasification temperature (>800°C) shifts products towards more CO and less H$_2$ (Erlich and Fransson, 2011). The combustion zone has the highest temperature inherent to its exothermic nature of reactions. The heat released by these exothermic reactions is used for the endothermic reactions occurring in the drying, pyrolysis and reduction zones.

*Char gasification* or reduction is an endothermic process in which several reduction processes of the remaining char from pyrolysis occur through reactions with CO$_2$ and H$_2$O at high temperature in the absence of oxidizer, producing CO and H$_2$. Reduction reactions occurring during the char gasification process are as follows (Erlich, 2009), (Hernández et al., 2012), (Gómez-Barea et al., 2013):

Boudouard reaction:
C + CO$_2$ → 2CO                \hspace{1cm} (R9)

Char-steam reforming reactions:
C + H$_2$O → CO + H$_2$                \hspace{1cm} (R10)
C + 2H₂O → CO₂ + 2H₂  \hspace{1cm} (R11)

C + H₂O → \frac{1}{2}CH₄ + \frac{1}{2}CO₂ \hspace{1cm} (R12)

For gasification processes using steam as a gasifying agent (and an external heat source), the below reactions may also occur during the reduction process (Hernandez et al., 2012).

Gas-steam reforming reactions:

\[ \text{CH}_4 + \text{H}_2\text{O} \rightarrow 3\text{H}_2 + \text{CO} \hspace{1cm} (R13) \]

\[ \text{C}_2\text{H}_4 + 2\text{H}_2\text{O} \rightarrow 4\text{H}_2 + 2\text{CO} \hspace{1cm} (R14) \]

Tar-secondary cracking and reforming reactions:

\[ \text{Tars} + \text{H}_2\text{O} \rightarrow \text{CO}, \text{H}_2 \hspace{1cm} (R15) \]

\[ \text{Tars} + \text{CO}_2 \rightarrow \text{CO}, \text{H}_2 \hspace{1cm} (R16) \]

\[ \text{Tars} \rightarrow \text{Char} + \text{gases} (\text{CH}_4 + \text{H}_2 + \text{C}_n\text{H}_m \text{ (higher molecular hydrocarbons)}) \hspace{1cm} (R17) \]

1.2.2 Gasifier designs

In general, the types of gasifiers differ according to the movement of the biomass feedstock and the gasifying agent. Biomass gasification technologies can be classified by fixed beds and fluidized beds.

For fixed beds, the reaction zones of drying, pyrolysis, combustion and gasification can be distinguished and their locations depend on the relative movement of the biomass feedstock and the gasifying agent. The temperature in the combustion zone is the highest temperature of the reactor and can typically reach around 800 to 1400°C (Zhang et al., 2010). Two examples of fixed bed types are updraft and downdraft gasifiers.

For fluidized bed systems, the bed material (such as silica sand) is used as a heat transfer media, which is fluidized by the gasifying agent coming from the bottom of the gasifier. This bed material is initially heated and the biomass feedstock is introduced as soon as the bed temperature is high enough. The intense mixing between biomass feedstock, bed material and gasifying agent facilitates simultaneous reactions within the reactor, thus there are no distinct reaction zones as in fixed beds. The high fluidization velocity creates high turbulence and enables better gas-solid contact thus favouring the fuel conversion. Therefore, fluidized beds can achieve higher carbon conversion compared to fixed bed gasifiers. The operating
temperature of a fluidized bed is usually around 700 – 900°C, which is lower than the temperature of fixed bed gasifiers. Depending on the fluidizing velocity and gas path, fluidized beds gasifiers can be classified as bubbling fluidized bed (BFB) or circulating fluidized bed (CFB) (Zhang et al., 2010). Figure 1 shows the different gasification reactor designs as illustrated by Quaak, et al. (1999). The operation characteristics for these gasifiers are briefly described in the following sub-sections.

1.2.2.1 Updraft gasifier

In an updraft gasifier, the biomass feedstock is introduced at the top and the gasifying agent at the bottom of the gasifier via the grate. The reaction zones and motion of biomass feedstock and gas are illustrated in Figure 1(a). The gasifying agent travels downward to the combustion zone and reactions occur immediately above the grate where the temperature rises to its maximum value in this zone. The hot gas produced by the combustion process travels upward and transfers heat for the endothermic gasification reactions, leaving behind ash that will fall through the grate at the bottom. On the way up, the hot gas experiences temperature reduction and will meet the pyrolysis zone where the volatiles are released and the primary tar is formed. The volatile gases and tar have no opportunity for further conversion and leave the gasifier with the product gases at low temperature (200 – 400 °C) (Quaak et al., 1999). Therefore, the gasified biomass produced by an updraft gasifier is largely dependent on the presence of pyrolysis products and normally contains higher hydrocarbon concentrations and tar. Han & Kim (2008) reported that the tar content produced during updraft gasification is in the range of 10 – 150 g/Nm³, which is among the highest amount compared to other gasifier designs.

1.2.2.2 Downdraft gasifier

Figure 1(b) shows a closed constricted downdraft gasifier in which the biomass feedstock is fed from the top and the gasifying agent is fed from the side, above the constriction (throat) where the combustion zone is located. Compared to the updraft gasifier, the locations of combustion and gasification zones in the downdraft bed are interchanged. After being dried and pyrolyzed, the pyrolysis products go through the combustion zone first before proceeding to the gasification zone. Travelling downwards, these products encounter the high temperature region of combustion and gasification zones and tar have a great opportunity for further decomposition into smaller and lighter compounds such as CO, CO₂, H₂O, and CH₄. The gasified biomass leaves the gasifier from the bottom at higher temperature, around 700 °C (Quaak et al., 1999). For the closed constricted gasifier, the constriction allows mixing of gases in a high temperature region, which favours tar cracking as mentioned earlier. Therefore, the gasified biomass produced by a
downdraft gasifier has the lowest tar production, in comparison to other gasification techniques, and is in the range of $0.01 - 6 \text{ g/Nm}^3$ (Han & Kim, 2008).

**Figure 1: Gasifier designs**

1.2.2.3 Bubbling fluidized bed
In a bubbling fluidized bed (BFB), the bed material is agitated by the upward gasifying agent with a fluidization velocity in the range of 2 – 3 m/s, which is lower than the minimum fluidization point (Belgiorno et al., 2003). This fluidization velocity
of the gas creates the bubbles that have a continuous mixing behaviour. As the velocity is low, the bed material is only suspended in the lower part of the gasifier which is called “dense phase area” (Figure 1(c)). The fuel feedstock fed from the side of the gasifier will quickly mix with the hot bed material, resulting in rapid pyrolysis and a relatively large amount of pyrolysis gases. The remaining char will further react and gasify until it is small enough to be suspended together with the product gas in the freeboard (gas phase area) before being carried out from the upper part of the gasifier. The ash is removed from the bottom of the gasifier. BFB produces tar content in the range of 1 – 23 g/Nm³ (Han & Kim, 2008).

1.2.2.4 Circulating fluidized bed
The circulating fluidized bed (CFB) gasifier employs a system where the bed material circulates between the gasifier reactor and a secondary vessel (a cyclone) as illustrated in Figure 1(d). In this system, the fluidization velocity is in the range of 5 – 10 m/s, which is higher than the minimum fluidization velocity. Such high velocity continuously pushes and circulates the bed material and solid particles (biomass feedstock and chars) between the gasifier and a cyclone (Belgiorno et al., 2003). Therefore, there is no separation between dense phase and gas phase as in the BFB since the expended bed occupies the whole interior of the reactor. Through the circulation process, the char is separated from the product gas by the cyclone and returned back to the gasifier together with the bed material while ash is separated and removed from the bottom of the reactor. The gasified biomass leaves the system from the top of the cyclone. CFB produces tar content in the range of 1 – 30 g/Nm³ (Han & Kim, 2008).

1.3 Gasified biomass gas – Influencing factors
In order to obtain the desired composition of gasified biomass with minimal impurities and to increase energy conversion efficiency, the gasification operating conditions need to be optimized. Therefore, the purpose of the following sub-sections is to address the influence of some important parameters on the gasified biomass composition.

1.3.1 Biomass feedstock
There are a large number of different biomass feedstock types for use in a gasifier, each with different properties. The objective of this section is to briefly discuss examples of biomass properties that influence the composition of the gasified biomass.
1.3.1.1 Biomass Feedstock

As mentioned earlier, biomass consists of complex compounds that consist mainly of cellulose, hemicellulose and lignin, as well as a small amount of extractives (Pereira et al., 2012). The proportions of these constituents vary between different biomass feedstock and can significantly affect the gasified biomass composition.

For example, Gani and Naruse (2007) experimentally investigated the effect of cellulose and lignin content on pyrolysis of several types of biomass. They concluded that, for the biomass with higher cellulose content, the pyrolysis rate is faster, while higher lignin biomass gives a slower pyrolysis rate. Lv et al. (2010) explained that cellulose consists of a branching chain of polysaccharides and no aromatic compounds; thus easy to volatilize. However, the lignin consists of various —O– and C–C– containing functional groups and aromatic structural units, making it difficult to volatilize. Correspondingly, they found that cellulose produces more tar, gas and CO yield and lignin creates higher yield of H2 and CH4.

In another study, Hanaoka et al. (2005) experimentally investigated the gasification process of cellulose, xylan, lignin, Japanese oak (of which the main component is cellulose) and Japanese red pine bark (of which the main component is lignin). They found the carbon conversion of cellulose and xylan to be much higher than that of lignin. The results also suggest that the dominant gasification process of cellulose is pyrolysis while for xylan and lignin, the dominant processes would be both pyrolysis and partial oxidation. The H2/CO ratios produced by xylan, lignin and Japanese red pine bark are higher than those of cellulose and Japanese red pine. The opposite trend is found for the H2/CO2 ratio.

1.3.1.2 Moisture content

The influence of moisture content on the gasification system is briefly mentioned in section 1.2.1. The constraint of moisture content for gasifying biomass fuel is dependent on the gasifier design used. Particularly, the maximum moisture content acceptable for downdraft, updraft and fluidized gasifiers is 20 %, 50 % and 70 % respectively (Brammer & Bridgewater, 1999), (Faaij et al., 1997).

An increase in the moisture content of biomass implies that a higher mass of water has to be vaporized. This leads to a reduction in the maximum process temperature and the biomass consumption rate. The reduction in operating temperature can cause incomplete cracking of tars and hydrocarbon species from the pyrolysis zone. On the other hand, at low temperature (<800 °C), the forward reaction of WGS (R8) is more pronounced; in this reaction, moisture content reacts with CO to produce H2 and CO2 (McKendry, 2002), (Erlich, 2009). However, the consumption of CO and
production of CO₂ cannot be compensated by the H₂ formation, thus the heating value of the gasified biomass is reduced (Zainal et al., 2001), (Melger et al., 2007).

For high temperature operation (about >750 °C), moisture content can promote char-, gas- and tar-steam reforming reactions, (R10) to (R16) (González et al., 2008), (Pérez et al., 2012), (Narváez et al., 1996). The operating temperature can be maintained high either by increasing the heat supply externally or increasing the air/O₂ flow for combustion reaction enhancement. Further discussion on the influence of operating temperature on gasified biomass composition is discussed in section 1.3.3.1.

1.3.1.3 Particle size

The size and shape of the biomass particles are important for determining the movement, delivery and storage of the fuel, as well as the behaviour of the fuel inside the gasifier.

The biomass particle size affects the gasification reaction rate and the gas composition by influencing the heat and mass transfer during the conversion process (Yan et al., 2010). In general, an increase in particle size enhances the heat transfer resistance and hence the temperature inside the particle becomes lower than the temperature at the surface at a given time (Luo et al., 2009), (Hernández et al., 2010). Smaller biomass particle size will provide a more effective surface area, thus leading to greater heat transfer and better conversion during the gasification process. However, depending on the gasifier type, smaller particles create a large pressure drop, which in turn decreases the efficiency (Erlich, 2009)

Several researchers (Lv et al., 2004), (Hernández et al., 2010), (Pérez et al., 2012) found that the concentration of combustible gases (CO, H₂ and CH₄) in gasified biomass is higher for small biomass particles compared to large particles. For example, Hernández, et al. (2010) experimentally investigated combustion of biomass waste in an entrained flow gasifier and found the concentration of the combustible gases for 0.5 mm diameter particles higher than for a particle size of 8 mm. It was explained that, as smaller particles have larger surface area, the heating rate is faster and volatiles leave the particles more quickly during the pyrolysis process in comparison to larger particles. As the volatile release rate is high, the char and tar formed from pyrolysis is expected to be more porous and reactive and thus aids the endothermic gasification reactions progressively.

Lv et al., (2004) experimentally investigated biomass of different average sizes (0.25, 0.38, 0.53, 0.75 mm) in an air-steam fluidized gasifier. They suggested that the
pyrolysis process of small particles is mainly controlled by kinetic reactions (faster heating rate), and meanwhile the pyrolysis of larger particles is controlled by gas diffusion in which the volatiles inside the particle have greater difficulty in diffusing out. Correspondingly, the increase in particle size decreases the gas yield and increases the char and tar yields (Mohammed et al., 2011), (Pérez et al., 2012). Furthermore, larger particles produce more CO₂ and less CO and CH₄. In a fixed bed downdraft gasifier with self-regulated air intake, Pérez, et al. (2012) found that as the bed temperature is reduced, the increase in particle size also leads to reduction of the biomass consumption rate and increment in the air-fuel ratio to the gasifier. When the air-fuel ratio increases, the degree of combustion is enhanced and more CO₂ and N₂ are produced. More information regarding the influence of bed temperature and air-fuel ratio on biomass gasification will be discussed in a section on operating conditions.

1.3.2 Gasification agent

Various gasifying agents can be used during biomass gasification such as air, steam, O₂ or a mixture of these gases. If pure steam is used, an external heat source is needed. Gil et al., (1999) investigated three different gasification agents used in an atmospheric BFB gasifier of wood chips. The gasified biomass composition from this work is shown in Table 2.

Air gasification is known as an autothermal process since the heat is provided by partial combustion within the gasifier (Campoy et al., 2009). Air is the gasifying agent most used in both laboratory and commercial applications due to its availability and cheap price (Puig-Arnavat et al., 2010), (Hernández et al., 2012). However, as seen in Table 2, air gasification produces gasified biomass with low LHV, mainly due to its high N₂ content. The combustible gases are less than half of the total volumetric flow. Although the data for char content produced for air gasification is not available in Table 2, Hernandez, et al. (2012) reported that the carbon conversion in air gasification is higher than in steam gasification since the combustion reactions are faster than the gasification reactions. Correspondingly, a combination of high carbon conversion and high inert gas flow results in the highest gas yields in air gasification, as seen in Table 2. Furthermore, tar has a chance to oxidize in the high temperature region of the combustion zone, therefore air gasification produces lower tar content compared with steam based gasification agents (Table 2).

Biomass gasification with pure steam can be performed in fluidized beds and the external heat source could be either a heat exchanger or circulation of heat carrying bed material between the gasifier and an external combustor (Campoy et al., 2009), (Zhang, 2010), (Umeki et al., 2010). This means that steam gasification seldom
achieves the same high temperatures as in the case of autothermal gasification, which is observed in Table 2. The table also shows that steam gasification results in higher H₂ content in the gasified biomass and higher LHV than those of air gasification. The higher content of H₂ is a result of the WGS reaction, which becomes dominant in the forward direction due to the high availability of H₂O and moderate temperature. As the combustion step is skipped in steam gasification, the char-, gas- and tar-reforming reactions become important. As seen in Table 2, the tar content in the gasified biomass is significantly higher. The higher char and tar yields of steam gasification could be explained by: (i) lower reaction rate of steam-char reaction than oxygen-steam, (ii) thermal tar cracking is not achieved and (ii) tar oxidation has a larger effect on tar reduction than tar reforming since air is more active than steam (Luo et al., 2009), (Taba et al., 2012). Steam gasification is however attractive when producing liquid biofuels, since the H₂/CO is close to two (Table 2). For example, one methanol molecule (also the basis for DME - Dimethyl ether) consists of two molecules of H₂ and one molecule of CO.

Table 2: Gasified biomass composition for different gasifier agents used

<table>
<thead>
<tr>
<th>Gasifier agent</th>
<th>Air</th>
<th>Pure steam</th>
<th>Steam – O₂ mixture</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bed material</td>
<td>Atmospheric BFB</td>
<td>Silica sand</td>
<td></td>
</tr>
<tr>
<td>Biomass feedstock</td>
<td>small chips of pine (Pinus pinaster) wood</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Feedstock moisture</td>
<td>10 – 20 wt %</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Operating conditions</td>
<td>λ</td>
<td>0.18 – 0.45</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>SBR (kg/kg daf)</td>
<td>0.08 – 0.66</td>
<td>0.53 – 1.10</td>
</tr>
<tr>
<td></td>
<td>T (°C)</td>
<td>780 – 830</td>
<td>750 – 780</td>
</tr>
<tr>
<td>Gas composition</td>
<td>H₂ (Vol. %, dry basis)</td>
<td>5.0 – 16.3</td>
<td>38.0 – 56.0</td>
</tr>
<tr>
<td></td>
<td>CO (Vol. %, dry basis)</td>
<td>9.9 – 22.4</td>
<td>17.0 – 32.0</td>
</tr>
<tr>
<td></td>
<td>CO₂ (Vol. %, dry basis)</td>
<td>9.0 – 19.4</td>
<td>13.0 – 17.0</td>
</tr>
<tr>
<td></td>
<td>CH₄ (Vol. %, dry basis)</td>
<td>2.2 – 6.2</td>
<td>7.0 – 12.0</td>
</tr>
<tr>
<td></td>
<td>C₂Hₖ (Vol. %, dry basis)</td>
<td>0.2 – 3.3</td>
<td>2.1 – 2.3</td>
</tr>
<tr>
<td></td>
<td>N₂ (Vol. %, dry basis)</td>
<td>41.6 – 61.6</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>H₂O (Vol. %, wet basis)</td>
<td>11.0 – 34.0</td>
<td>52.0 – 60.0</td>
</tr>
<tr>
<td></td>
<td>Tars (g/Nm³)</td>
<td>2.0 – 20.0</td>
<td>30.0 – 80.0</td>
</tr>
<tr>
<td></td>
<td>Char (g/kg)</td>
<td>n.a</td>
<td>95.0 – 110.0</td>
</tr>
<tr>
<td></td>
<td>Gas (Nm³/kg)</td>
<td>1.25 – 2.45</td>
<td>1.3 – 1.6</td>
</tr>
<tr>
<td></td>
<td>LHV (MJ/Nm³)</td>
<td>3.7 – 8.4</td>
<td>12.2 – 13.8</td>
</tr>
</tbody>
</table>

n.a – not available
The LHV of the gasified biomass can be improved to the range of 12 – 28 MJ/Nm$^3$ if pure O$_2$ or O$_2$ enriched air is used as a gasifying agent (Göransson et al., 2011). The improvement of the gas quality of O$_2$ gasification can be attributed to the minimal (or absence of) N$_2$ dilution from air which results in more combustible gases (such as CO and H$_2$). The absence of N$_2$ dilution also causes a low flow rate to the gasifier and hence increases the residence time for better carbon conversion due to a high rate of carbon reactions (Zhou et al., 2009). According to Zhou et al. (2009), high content of O$_2$ in the gasifying agent strengthens the Boudouard reaction and more CO gas is produced. As a result, the H$_2$/CO ratio of O$_2$-gasification is lower than that of air gasification.

To improve the tar and char conversion while maintaining a higher LHV, a mixture of air-steam or O$_2$ -steam could also be used as a gasifying agent (Gil et al., 1999), (González et al., 2008), (Meng et al., 2011). The gasification agent blend will then provide the necessary heat for the gasification process and will thus eliminate the complexity of an external heat supplier. The lower char and tar yields depend on some of the char oxidizing, giving more porous and active char area for the forthcoming gasification reactions. At the same time the temperature will be higher in the gasifier compared to pure steam gasification (Table 2), enhancing tar cracking. Tars produced now also have the possibility to combust, providing some of the heat necessary for the endothermic char gasification reactions. However, the tar content remains higher than that of pure air-gasification as seen in Table 2. The absence of N$_2$ and presence of H$_2$O results in higher H$_2$ concentration and LHV compared to air gasification. It also can be seen that CO concentration is the highest for gasification with steam- O$_2$ –blend.

**1.3.3 Gasifier operating conditions**

Gasification operating temperature and pressure, equivalence ratio (ER), steam-to-biomass ratio (SBR) and possible use of catalyst material are among the operating parameters that affect the gasified biomass composition.

**1.3.3.1 Temperature**

The bed temperature is among the most important operating parameters in a biomass gasification process. The increase in bed temperature favours carbon conversion, which in turn increases the gas yield and decreases the char and tar yields (Lv et al., 2004). The production of combustible gases (H$_2$, CO, CH$_4$) is also increased when temperature is elevated thus resulting in higher heating value. This trend can be explained by a large release of volatiles from the fuel during the pyrolysis process leaving behind a more reactive char. At the same time, the endothermic gasification reactions (R10) to (R17) become faster at higher
temperature. For the WGS reaction (R8), the reverse direction is dominant at higher temperature, i.e. more CO is produced (Mayerhofer et al., 2012). This condition is consistent with the principle of Le Chatelier that states that any system at equilibrium if disturbed by a change in pressure, temperature or concentration, will shift its equilibrium position to counteract the change (Turns, 2006). Also, higher bed temperature favours cracking and reforming of tars and heavy hydrocarbons (Lv et al., 2004), (Wu et al., 2009).

Hernández et al. (2012) investigated the effect of temperature on air and air-steam biomass gasification. They found that the formation rate of CO and H₂ increases with a rise in temperature for air gasification due to the promotion of the Boudouard and steam-reforming reactions (R9) to (R16). While for air-steam gasification, the combination of the added steam in the gasification agent and high temperature favours the steam reforming reactions, (R10) to (R16), and the WGS reaction (R8) in the forward direction and thus producing more H₂ and CH₄.

1.3.3.2 Pressure
The operating pressure of a biomass gasification system is generally selected in accordance with the requirements of the downstream process or end-use equipment. For example, pressurized gasification can avoid a step of gas compression prior to the utilization of gasified biomass in a gas turbine (Valin et al., 2010).

Several researchers (Mayerhofer et al., 2012), (Kitzler et al., 2011), (Higman & van Der Burgt, 2003), (Valin et al., 2010), found that an increase in operating pressure in the biomass gasification process led to a reduction of the H₂ and CO formation and an increase in CH₄, CO₂ and tar production. They agreed on the fact that the equilibrium of the gasification reactions tends to shift to the side with the least number of gas molecules at high operation pressure, following the principle of Le Chatelier as mentioned in section 1.3.3.1.

1.3.3.3 Air to fuel ratio
For air or O₂ gasification, the air to fuel ratio is a crucial parameter. In this thesis, the air to fuel ratio required for biomass gasification is represented by the symbol λ and is defined as the actual air-to-biomass weight ratio divided by the stoichiometric air-to-biomass weight ratio (Narváez et al., 1996). In biomass gasification, the typical λ value normally varies between 0.20 and 0.40 (Narváez et al., 1996), (Gabra et al., 2001), (Zainal et al., 2002), (Pereira et al., 2012). The variation in λ affects the gasification temperature through the interaction between the exothermic and endothermic reactions. When the λ value is too small, the temperature is low thus
disfavouring the endothermic gasification reactions. On the other hand, when $\lambda$ is too high, the combustion reactions are progressively enhanced which produce more combustion products and increase the bed temperature but leave less char for the gasification process. Besides, at high $\lambda$, the higher amount of $N_2$ provided by the air (in the case that air is used as a gasification agent) dilutes the product gas and thus reduces its energy content.

Gai and Dong (2012) investigated experimentally the influence of $\lambda$ in the range of 0.18 to 0.41 for air gasification of corn straw. They showed that as $\lambda$ increased, the bed temperature and $N_2$ concentration in the gasified biomass constantly increased. The optimal $\lambda$ was found to be in the range of 0.28 to 0.32, which gave the highest LHV of the gasified biomass. Within this range, the concentration of combustible gases (H$_2$, CO and CH$_4$) was the highest and the concentration of CO$_2$ the lowest, mainly due to the enhancement of endothermic gasification reactions at higher temperature. Beyond this range, the opposite trend was observed for the concentration of H$_2$, CO, CH$_4$ and CO$_2$ indicating that progressive combustion became dominant. The tar content decreased with increased $\lambda$ since the elevated temperature results in the tar cracking and reforming reactions being enhanced.

The optimum $\lambda$ value of 0.23 is found by Lv et al. (2004) in experimental air-steam biomass gasification; here the optimum was defined as the operating condition at which maximum H$_2$ and other combustible components were produced. They observed that for $\lambda$ below this optimum value, both gas yield and LHV increased with $\lambda$. On the other hand, both parameters decreased when the increase of $\lambda$ exceeds this value. The improvement of oxidation reactions with higher $\lambda$ also decreased the concentration of CO, CH$_4$, C$_n$H$_m$ and increased CO$_2$.

1.3.3.4 Steam-to-biomass ratio
For a gasification agent containing steam, the steam-to-biomass ratio (SBR) is a significant important parameter which is defined as the mass flow rate of steam fed into the gasifier divided by the mass flow rate of the fed biomass (Campoy et al., 2009). According to Kumar et al. (2009), the increase in SBR increases the partial pressure of H$_2$O which consequently favours the steam reforming (R10) to (R16) and WGS reactions, (R8) (with help of high gasification temperature, i.e. above 750 - 800°C). High SBR also leads to improvement of the biomass conversion efficiency.

Mayerhofer et al. (2012) studied the influence of SBR (0.8 to 1.2) on tar content and gas composition for steam biomass gasification. At an operating temperature of 800°C, the minimum tar amount was found at the highest SBR and it is suggested that higher steam concentration favours the tar reforming which consequently
reduces the total tar content. However, at a gasification temperature of 750°C, only phenols and cresols are decreased with an increment of SBR and instead some light PAH compounds increased. An increment of SBR increases the concentration of H₂ and CO₂ and decreases the concentration of CO and CH₄ in the gasified biomass. It is suggested that the trend is due to the enhancement of the WGS reaction, (R8) when more steam is added. However, the decrease in CH₄ concentration is suggested to be due to the improvement of methane-steam reforming at higher steam addition. The production of gasified biomass with higher H₂ and CO₂ and lower CO and CH₄ concentrations at higher SBR was also observed by Xiao et al. (2011) and Lv et al. (2004).

1.4 Combustion of gasified biomass gas

As mentioned in the introductory section, the applications of gasified biomass are varied, including the chemical and liquid biofuel feedstock sector, or in various devices for power and heat production. For the power and heat sector, gasified biomass can be used as a fuel in combustion devices such as gas engines, gas turbines, boilers or industrial burners (Laurence & Ashenafi, 2012). Leung et al. (2004) reviewed the technological development of biomass gasification for a variety of applications in China including applications in gas engines and boilers for heating, domestic cooking and electric generation. Particularly, use of gasified biomass for electrical energy generation is foreseen as a very promising application, possessing great potential for research and development. López (2008) investigated a more suitable location for the project of biomass based electrical generation with three different technologies: gas engine, gas turbine and hybrid fuel cells-microturbines. Among the parameters studied: the location of the biomass distribution, the cost for the transportation between the biomass collection point and the plant, as well as the distance of the potential plant from the existing electrical distribution lines. Kirkels & Verbong (2011) presented a review of the future potential of biomass gasification based on a 30-year overview of the global developments of this technology. They indicated that high-end applications like Biomass Integrated Gasification Combined Cycle (BIGCC) are major interests in research and development. BIGCC technology has the potential to produce electricity at a higher efficiency through combustion of gasified biomass in gas and steam turbines. According to Franco and Giannini (2004), BIGCC of 30 – 50 MWe can achieve efficiencies of about 40 % for wood based biomass and atmospheric gasification. Currently, the BIGCC technology is still at the pre-commercial demonstration stage. Efforts towards both engineering improvements and cost reduction are ongoing. One potential area of improvement is the combustion system of the gasified biomass. One option could be to use externally fired gas turbines to remove the cost of clean-up systems and
compression of the gasified biomass (Kautz & Hansen, 2007), (Al-Attab & Zainal, 2010).

1.4.1 Combustor concepts for gasified biomass

The combustion of gasified biomass differs from that of natural gas particularly due to its different composition and heating value. Also, the product gases from gasification can have various mixtures and heating values, and thus the combustion process can also be different between them. The first focus of this section is to review combustion systems for low LHV gasified biomass since this kind of gas is cheapest to produce. Next there is a review of combustion systems that are capable of accepting a wide range of gasified biomasses.

There are two primary types of gas combustor designs; non-premixed (diffusion) and premixed combustion types. In the non-premixed combustor, fuel and combustion air are introduced separately to the combustor (Figure 2 (a)) and the flame is stabilized at the point where the fuel and air meet and mix. Therefore, for this configuration, the flame is considered diffusion limited since the combustion rate is dependent on how fast the reactants can diffuse towards the reaction zone where they oxidize and generate heat. This reaction zone is normally generated near the stoichiometric condition where the temperature is very high (Turns, 2006). This temperature is usually high enough to oxidize nitrogen in the combustion air thus producing an appreciable amount of NOx emissions (further information of NOx formation is discussed in section 1.4.4). In the premixed combustor type, fuel and combustion air (normally in excess) are premixed thoroughly before being fed into the combustor (Figure 2 (b)). The excess air serves to dilute the combustion and thereby keeps the flame temperature down, thus avoiding thermal NOx formation (Whitty et al., 2010). Considering the advantage of producing low emission levels, recent combustor concepts employ premixed type combustion and are frequently operated at fuel lean condition.

![Figure 2: Illustration of simple non-premixed and premixed combustors](image)

For low LHV gas combustion, there are two existing approaches; modification of an existing combustor for conventional fuels and development of a completely new combustor for specific fuel mixtures. Combustors designed for natural gas are
generally not suitable for low LHV gas due to stability problems (Adoune et al., 2002), (Hoppesteyn, 1999). Frequently the diffusion combustion concept is used for low LHV gas combustion since it allows stable combustion (Richards et al., 2010). However, modifications of an existing combustor are required in order to accommodate the higher mass flow compared to conventional fuel such as natural gas for the same thermal loads. The necessary alterations include the fuel delivery/injection system, fuel nozzles and fuel manifold and combustion (Neilson, 1998). For example, the diffusion type combustor of the Typhoon gas turbine (Alstom – Siemens Energy) as seen in Figure 3(a) that was originally designed for natural gas has been redesigned to suit the product gas from air-blown wood gasification in a pressurized circulating fluidized bed in the first BIGCC plant at Värnamo, Sweden (Ståhl & Neergaard, 1998).

There are several diffusion type combustors which have been tested with low LHV gasified biomass. An example is air staging; a non-premixed combustor design to maintain low flame temperature and hence prevent the formation of thermal NOx emissions (Richards et al., 2001). Adouane et al. (2010) performed an experimental study of gasified biomass with LHV of about 7.9 MJ/Nm³ in a new air staged combustor called Winnox-TUD (Figure 3(b)). The main characteristic of this combustor is its three air stages, which are mastered independently to enable good control of the equivalence ratio in each stage. Another air staging combustion concept tested with simulated low LHV gas from biomass gasification (4.2 - 5.9 MJ/Nm³) known as COSTAIR (continuous air staging with internal recirculation) was reported by Al-Halbouni et al. (2007) (Figure 3 (c)). The air-staging technique does help to maintain the combustion temperature below the temperature of thermal NOx formation, but on the other hand also complicates the combustion air control (Richards et al., 2001).

The use of catalytic premixed combustion of gasification biomass is a technique which can enhance the combustion stability in fuel-lean conditions and is well suited for low-LHV gas (Groppi et al., 1996), (Johansson et al., 2002) and (Andrea et al., 2005). An example of a catalytic combustion concept is given in Figure 3 (d). The research in this area focuses on the development of different catalysts to achieve better combustion, lower emission levels and lower cost. The catalytic premixed combustion of gasified biomass is still at the research/demonstration level due to the high cost associated with the technique (Carroni & Griffin, 2010), (Barbato et al., 2013).
Figure 3: Combustor design concept
Another important combustor design for gasified biomass is swirl stabilized combustion. Example of this design concept is the low-swirl burner (Littlejohn & Cheng, 2007) shown in Figure 3 (e). In this type of combustor, the swirling flow created allows high fluid mixing rates and long residence time for complete reaction to take place (De & Acharya, 2012). The swirl stabilization concept is used both in diffusion and premixed combustors. Lewis et al. (2012) experimentally investigated the combustion of product gas from air-blown wood gasification (LHV around 6 MJ/Nm³) in a swirl burner. They found that even though stable combustion can be achieved, a few blowoff events occurred due to the fluctuations in gas composition of the tested gasified biomass.

The above mentioned combustion systems are for operation of fuels within a low LHV limit. For medium LHV gasified biomass, higher content of CO and H₂ as well as heating value can lead to higher flame temperatures. Therefore, thermal NOₓ formation is the main issue when a diffusion type combustor is used. However, problems associated with high laminar flame speed such as auto-ignition, flashback and blowoff are major concerns when using a premixed type combustor (Richards et al., 2001). However, this combustor type may allow for a larger span of combustible gases to be used. In order to become commercialized, the research area of premixed combustion requires further study of combustion stability when using a fuel of different range of compositions and LHV.

Concerning the wide range of product gases which can be obtained from gasification of biomass, the subsequent combustion system should preferably not be limited by the specific composition or heating value range. Current commercially available combustion systems do not meet fuel flexibility allowing them to accept fuels that range widely in composition and heating values but also changes in gas composition over time. An effective fuel flexible combustor should not only meet these criteria but preferably also be able to use other types of fuel as backup while maintaining low emission levels. For example, the application of a fuel-flexible combustor could be in a gasifier-heat plant (e.g. for boilers, chemical heating, ceramic kilns), in which the use of gasified biomass can be switched to natural gas or similar during maintenance of the gasifier. By doing this, shut-down of the whole heat application during the maintenance process can be avoided without increasing the undesired emission levels.

In this thesis, the target combustion concept for gasified biomass is the premixed type since it may allow low emission combustion of a variety of fuels. The following section discusses the fundamental combustion properties and potential operating
issues associated with the premixed combustion system and how they are affected by the composition of the gasified biomass.

1.4.2 Fundamental combustion properties of gasified biomass gas

The combustion characteristics of gasified biomass are considerably dissimilar from those of natural gas due to its higher content of non-combustible components. The thermodynamic and transport properties of gasified biomass are a result of the combination of the properties of the individual gas species. In premixed combustion, it is essential to investigate the key combustion properties of gasified biomass within the combustor. Particularly in a premixed combustion system, the laminar flame speed is one of the essential parameters to characterize the combustion behaviour of gasified biomass as it contains information on reactivity, diffusivity and exothermicity (Munajat et al., 2012). In the following sub-sections, in addition to the laminar flame speed, the dependence of flammability limit and flame temperature on the composition of gasified biomass is also briefly discussed. This is because the flammability limit may serve as a first approximation of the condition in which the flame can propagate through the fuel and air mixture within certain limits of composition. At the same time, the flame temperature is important since it has a controlling effect on the chemical reaction rate.

1.4.2.1 Flammability limit

The flammability limit is defined as the range of fuel concentrations, normally in percentage volume, in which the fuel in air can be ignited and support the flame propagation (Turns, 2006). A lower flammability limit (LFL) describes the minimum fraction of fuel concentration in air below which there is not enough fuel to cause the flame propagation since the temperature cannot reach a level that sustains the combustion process. An upper flammability limit (UFL) describes the maximum fraction of fuel concentration in air above which there is an insufficient oxygen source for the reaction to cause flame propagation. In other words, the flame will only propagate if the ER of the particular fuel is within its flammability limit (Turns, 2006), (Lefebvre & Ballal, 2010).

For gasified biomass, the flammability limit is strongly dependent on its composition. The flammability limit of a gasified biomass fuel mixture can be estimated using the Le Chatelier’s law (Kondo et al., 2010),

\[
FL_{mix} = \sum \frac{C_i}{FL_i} \times 100
\]  

(Eq.1)

where \(FL_{mix}\) is a lower or upper flammability limit and \(C\) is the volumetric fraction of each combustible fuel component in the total fuel mixture. According to Natarajan
et al. (2010), gasified biomass can have a wider range of flammability limits than a conventional fuel such as natural gas, mainly due to the presence of H₂ and CO. H₂ as an individual fuel has an LFL slightly lower than CH₄ but a much higher UFL. Pure CO has a higher LFL than both H₂ and CH₄, but its UFL is almost similar to H₂ (Wierzba & Wang, 2006) and (Zlochower & Green, 2009). Wierzba and Wang (2006) experimentally found that the flammability limits for various H₂-CO-CH₄ mixtures are close to the predicted ones based on Le Chatelier’s law. Particularly, the mixture with higher H₂ content resulted in reduced values of LFL but higher UFL values. In contrast, the extension of the flammability limit of gasified biomass may be cut off due to its non-combustible contents. Usually, the combustion of diluted gasified biomass requires a higher ER (a leaner condition) or lower equivalence ratio (a richer condition) to ensure successful flame propagation throughout the gasified biomass mixture (Shrestha et al., 2009).

1.4.2.2 Flame temperature
Another important parameter in combustion analysis is the flame temperature, since it determines the amount of heat available in the flame and can significantly influence other combustion properties, including flame speed and extinction limit through the chemical reaction rate (Natarajan & Seitzman, 2010). Furthermore, in terms of emissions, the production of thermal NOₓ emission is sensitive to high temperature while the CO and unburned hydrocarbon (UHC) emissions are sensitive to low temperature (Natarajan & Seitzman, 2010), (Turns, 2006).

Measurement of the actual flame temperature is difficult. Frequently, the theoretical adiabatic flame temperature is used for combustion analysis. Adiabatic flame temperature is the temperature at which the reactant is oxidized adiabatically, i.e. complete combustion without any heat transfer from or to the surrounding (Turns, 2006). Considering the pure combustible gas components in gasified biomass, CO has the highest adiabatic flame temperature for a given equivalence ratio, closely followed by H₂, while CH₄ has the lowest. CH₄-air has a lower adiabatic flame temperature because it requires four times more air (on a molar basis) to achieve a stoichiometric mixture. Both CO and H₂ require the same amount of air for a similar equivalence ratio, but CO’s flame temperature is slightly higher than that of H₂ owing to its higher molar heating value. For a gasified biomass fuel mixture, the variation in fuel composition is among the main factors that defines its flame temperature. The adiabatic flame temperature increases with the increase of the volume fraction of the combustible constituents. The presence of non-combustible components thus tends to reduce the flame temperature due to limitation of reactive species and heat capacity of added diluents (Hui et al., 2007).
1.4.2.3 Laminar flame speed

The laminar flame speed is conventionally defined as the speed at which a flame front travels relative to the unburned gas in a direction normal to the flame surface (Lefebvre & Ballal, 2010), (Turns, 2006). Laminar flame speed is the rate at which the flame can propagate in a combustion mixture. If the flame speed is lower than the speed of the incoming unburned reactants, the flame may detach from the burner (known as blowoff) (Lieuwen et al., 2008). If the flame speed is higher than the flow rate of the unburned reactants, there is a possibility that the flame may travel back into the premixing section of the burner (known as flashback) (Lieuwen et al., 2008). Therefore, on a practical level, the laminar flame speed is used to predict flashback, blowout and flame stability. On a more fundamental level, it is an important target to validate chemical kinetics of the reacting mixture and is used in numerical studies of combustion.

There is an abundant amount of available measured and calculated laminar flame speed data for pure fuels at a variety of operating conditions. These studies show that the flame speed is strongly influenced by the air-fuel-ratio, pressure and preheating temperature. In descending order, H₂ has the highest flame speed (320 cm/s), followed by CO (55 cm/s) and CH₄ (40 cm/s). There is also a great amount of studies of laminar flame speed available for binary fuel mixtures such as CH₄/H₂ (Yu et al., 1986), (Halter et al., 2005), (Zhang et al., 2009), (Hu et al., 2009), CH₄/CO (Vagelopoulos & Egolfopoulos, 1994), (Coppens et al., 2007), (Wu et al., 2009) and CO/H₂ (Vagelopoulos & Egolfopoulos, 1994), (Dong et al., 2009), (Natarajan & Seitzman, 2010), (Bouvet et al., 2011), (Hassan et al., 1997). For fuel containing H₂ gas, it was found that the laminar flame speed increases as more H₂ is added to the fuel mixture. The increasing trend of laminar flame speed with increase in H₂ content can be explained as such: (i) the overall reactivity of the fuel mixture increases with the amount of H₂ and (ii) the low molecular weight of H₂ acts to increase the diffusivity of the reactant mixture. In the case of CO oxidation, the presence of hydrogen containing species either from H₂ or hydrocarbons can accelerate the reaction rate through the reaction (R18), which consequently increases the laminar flame speed.

\[
\text{CO} + \text{OH} \leftrightarrow \text{CO}_2 + \text{H}
\]  

(R18)

An increase in laminar flame speed by the acceleration of CO oxidation was shown by (Vagelopoulos & Egolfopoulos, 1994), (Zhang et al., 2009), (Wu et al., 2009), and (Brown et al., 1996). With regards to the hydrocarbon content, it is well known that CO and H₂ are produced during oxidation of hydrocarbons. Therefore, it is important to study the interaction of the intermediate CO and H₂ with those contained in the fresh fuel mixture of gasified biomass. Cheng et al. (2011) found that the flame
speed of a H2/CH4 fuel mixture is larger than that of a CO/CH4 mixture. This result is contradictory with its corresponding adiabatic flame temperature on which H2 has only a minor effect compared to CO. While in a mixture of H2/CO/CH4, higher H2 content resulted in higher laminar flame speed. They suggested that the trend of the laminar flame speed of this mixture is affected more by its chemical effect than its thermal effect. The other components included in gasified biomass not mentioned above, such as H2O, CO2 and N2, will also influence the flame speed. Also, when using raw gasified biomass directly from a gasifier without passing a clean-up process, the tar content may alter the reaction pathways thus affecting the laminar flame speed.

Several research projects have shown that the decrease in the laminar flame speed of a fuel mixture containing dilution components is caused by the increase in specific heat capacity and the decrease in heat release, flame temperature and thermal diffusivity. For example, the reduction of the laminar flame speed by N2 dilution in H2/CO is shown by Prathrap et al. (2008) and Burbano et al. (2010a). Tahtouh et al. (2009) found that N2 dilution non-linearly reduced the laminar flame speed of a H2/CH4 fuel mixture. Meanwhile, reduction of laminar flame speed by CO2 dilution in different proportions of H2/CO fuel mixtures is reported by Prathap et al (2011), Kishore et al. (2009), Natarajan et al. (2007) and (2008). There are several studies considering the influence of H2O on the laminar flame speed of a single component for example CH4 (Mazas et al., 2011) and H2 (Liu & MacFarlane, 1983), (Kuznetsov et al., 2011). However, there is little data available on laminar flame speed of fuel mixtures with H2O dilution. Das et al. (2011) found a different trend in the influence of H2O between low and high H2 content of H2/CO mixture. For the case of lower H2 content, the laminar flame speed of the fuel mixture first increases, reaches a maximum and then decreases with further increase of H2O addition. For the higher H2 content, however, the laminar flame speed always decreased with addition of H2O. The effect of diluents, such as tar and H2O, on laminar flame speed of gasified biomass has not been found in literature by the author.

### 1.4.3 Combustor operability issues

There are several important practical problems which need to be taken into account when dealing with gasified biomass as a fuel in a combustion device in addition to maintaining low emission levels. According to Lieuwen et al. (2008), the four most critical combustor operability issues are blowout, auto-ignition, flashback and combustion instability. These issues are primarily important in the premixed combustion type and are strongly affected by the fuel properties. Therefore, it is essential to investigate their interdependence on gasified biomass composition.
1.4.3.1 Auto-ignition

In premixed combustion, auto-ignition occurs when the fuel-air mixture spontaneously ignites and the flame is established in the premixing zone, which is not designed to sustain high temperature. For premixed combustion, longer mixing times are required in order to achieve uniform mixing for lowered emissions levels. However, this requirement is contradictory to the need to avoid auto-ignition in the premixer zone. Thus, the knowledge of ignition delay time and the residence time is important when designing the premixer in order to avoid auto-ignition (Lieuwen et al., 2008).

Comparing the combustible components in the gasified biomass fuel mixture, H₂ has the lowest ignition energy and shortest ignition delay time (Chacartegui et al., 2011), (Asai et al., 2010), (Lieuwen et al., 2008), (Delattin et al., 2010). Therefore, gasified biomass with high H₂ content may have high risk of auto-ignition. The CO content may affect the auto-ignition through the reactivity of the total fuel mixture since CO oxidation is sensitive to the presence of H₂. As mentioned above (R18), CO oxidation can be accelerated even with a trace amount of H₂, hydrocarbons or H₂O present in the gasified biomass fuel mixture. For CH₄ or other hydrocarbons, their low laminar flame speed and high auto-ignition temperature may suppress the reactivity tendency of gasified biomass to trigger the auto-ignition occurrence (Weinrotter et al., 2005), (Zhang et al., 2012). Also, the content of diluents weakens the reactivity of the total fuel mixture. Therefore, diluent appearance in the gasified biomass fuel mixture will result in longer ignition delay time and slower heat release rate. Both hydrocarbons and diluent gases reduce the risk of auto-ignition.

1.4.3.2 Flashback

The term flashback defines the propagation of the flame back to the premixer zone from where it has been stabilized in the combustion zone. Similar to auto-ignition, once the flashback sets in, the flame stabilized in the premixer zone results in rapid temperature increase, followed by material failure. There are four different mechanisms leading to flashback as discussed by Dam, Love and Choudhuri (2011), Kröner, Fritz and Sattelmayer (2003) and Lieuwen et al. (2008). These mechanisms are (i) flashback due to the turbulent flame propagation in the core flow, (ii) flashback in the boundary layer, (iii) flashback due to the combustion induced vortex breakdown (CIVB) and (iv) flashback due to the combustion instabilities. According to Wang et al. (2009) and Schäfer et al. (2003), the first two mechanisms occur if the flame speed is relatively higher compared to the velocity of the incoming unburned reactant mixture. However, the CIVB mechanism only occurs in swirl-combustors in which the interaction between turbulent and chemistry swirled flows causes the
flashback. For the last category, the flashback occurs due to velocity fluctuations in the combustion chamber associated with combustion instabilities.

For gasified biomass combustion, the tendency of flashback is increased compared to natural gas as H₂ content in gasified biomass has a high flame speed and low lean flammability limit (Delattin et al., 2010), (Bouvet et al., 2011), (Lee et al., 2001), (Dam et al., 2011). For example, Dam et al. (2011) investigated experimentally the combustion of H₂/CO containing fuel and five different fuel mixtures that resemble wood- and coal-gasified gas in a swirl-stabilized premixed combustor. They found that the flashback occurred at leaner conditions for fuel mixtures with higher H₂ content. While for fuel mixtures with diluents (CO₂ and N₂), CO₂ was shown to retard the flashback tendency. Fuel mixtures with higher CO₂ content experienced flashback at relatively rich condition.

1.4.3.3 Blowoff

Opposite to the flashback mechanism, flame blowoff in a combustion process occurs when the flame speed is much lower than the velocity of the incoming unburned reactant mixture (Delattin et al., 2010). As a consequence the flame detaches from its anchored location by the combustor outlet and is blown-out by the incoming gas. For premixed combustion the optimal lean conditions frequently occur close to blowoff condition.

For H₂-contained fuel mixtures, the equivalence ratio at which the blowoff occurs is decreased as the percentage of H₂ in the fuel increases, as found by Schefer (2003), Vagelopoulos and Egolfopoulos (1994) and Zhang et al. (2007) in CH₄/H₂, CO/H₂- and H₂/CH₄/CO-mixtures respectively. Alavandi and Agrawal (2008) investigated experimentally lean premixed combustion of simulated fuel mixtures containing H₂, CO and CH₄ in a porous burner. They found that the addition of H₂/CO in CH₄ resulted in lower flame temperature at the blowoff limit compared to pure CH₄, especially for higher contents of CO and H₂. Ding et al. (2011) investigated numerically the blowoff limit in terms of dilution level of N₂, CO₂ and H₂O for H₂/CO fuel mixture with and without the presence of CH₄ in premixed counter-flow flames. They found that the flame of H₂/CO mixture with CO₂ dilution blows off at the same level as with H₂O dilution, but at a lower temperature. The flame of H₂/CO/H₂O blows off at low level of N₂ dilution but still at a higher temperature compared to a pure H₂/CO mixture. The presence of CH₄ extends the dilution level at which the flame blowoff of H₂/CO occurs.
1.4.4 Combustion emissions

As with any fuel, the combustion of gasified biomass gas can produce emissions such as nitrogen oxides (NOx), carbon monoxide (CO) and unburned hydrocarbon (UHC). Besides the influence of combustor type and its operating temperature, fuel properties also play an appreciable role in the formation of emissions. Below, the dependence of emission formation on fuel composition is briefly described.

1.4.4.1 NOx emission

NOx is a collective term of nitric oxide (NO) and nitrogen dioxide (NO2) that are important in combustion because of their contribution to both environmental and health problems. NO is generally the major emitted component of the NOx species, however, once NO is released to the environment it easily oxidizes to NO2. Once in the atmosphere, NO2 can react with moisture and other compounds to form various acidic compounds, fine particles and tropospheric ozone (Turns, 2006). The formation of NO is strongly influenced by the temperature and the concentration of radical species in the flame. NO can be formed through several different mechanisms and these are thermal-, fuel NO-, prompt-, N2O-intermediate- and NNH mechanisms (Whitty et al., 2010).

Thermal mechanism:

Thermal NO is formed when air bound N2 is oxidized and this occurs normally in high-temperature combustion (>1370 °C) (Whitty et al., 2010). The three principal reaction steps associated with the formation of thermal NO are known collectively as the extended Zeldovich mechanism (Ding et al., 2011) and are given as follows:

\[ \text{O} + \text{N}_2 \leftrightarrow \text{NO} + \text{N} \quad \text{(R19)} \]
\[ \text{N} + \text{O}_2 \leftrightarrow \text{NO} + \text{O} \quad \text{(R20)} \]
\[ \text{N} + \text{OH} \leftrightarrow \text{NO} + \text{H} \quad \text{(R21)} \]

These reactions involve radical species such as O, N, H and OH which are initially formed through decomposition or abstraction reactions. However, compared to the influence of such radicals, the dependence of thermal NO formation on the combustion temperature is stronger since it requires high activation energy to decompose N2. Therefore, there is also a dependence of combustion residence time as longer residence time gives an opportunity for the temperature to develop in the combustion chamber (Göke et al., 2012), (Richards et al., 2010).

Considering the composition of gasified biomass, NO formation through the thermal route may be significant if the H2 and CO concentrations in the fuel mixture are high.
Both gases tend to cause high combustion temperature and thus triggering the formation of thermal NO (Whitty et al., 2010). Regarding hydrocarbon gases, the presence of CH₄ in the fuel mixture may decrease the flame temperature. However, other hydrocarbons with higher flame temperature than CH₄, have a tendency to contribute to an increase in the overall flame temperature (Richards et al., 2001). For such fuel mixtures, application of an air-staging combustor, a premixed combustor or operating the combustion at leaner condition is among the strategies to control the thermal NO. Another technology is to inject N₂ or H₂O in the fuel to limit the peak flame temperature through the dilution effect and hence inhibiting the emission of thermal NO (Whitty et al., 2010), (Ghenai, 2010).

High content of diluents in the gasified biomass may affect the thermal NOₓ formation in two ways. Firstly, their thermal dilution effect results in a flame temperature well below the critical temperature of thermal NO formation (Adoune et al., 2002). Consequently, the NO formation through the thermal route becomes less important. Secondly, the high diluent fraction means that more volumetric flow of total fuel mixture is needed to produce the same thermal power compared to natural gas. Therefore, lower thermal NO emission can be achieved since the overall residence time in the combustion chamber is lower for gasified biomass compared to natural gas (Popovic et al., 2010)

**Fuel-NO mechanism:**

The conversion of fuel-bound-nitrogen (FBN) to NO results in an emission called fuel-NO. Two primary FBN species are: (i) hydrogen cyanide (HCN) and (ii) ammonia (NH₃). In the case of gasified biomass, HCN and NH₃ are produced from the nitrogen content within the biomass feedstock during its gasification process. These species can be converted to NO in the presence of oxygen at combustion temperatures and at the same time can be competitively reduced to N₂ according to the overall reactions (Ahn et al., 2011);

\[
\text{Fuel N} \rightarrow \text{HCN} / \text{NH}_3 \quad (R22) \\
\text{HCN} / \text{NH}_3 + O_2 \rightarrow \text{NO} \quad (R23)
\]

According to Hasegawa et al., (2001) the rate of NH₃ to NO conversion is increased with an increase in the CO/H₂ ratio. They also found that presence of a small amount of CH₄ minimizes the influence of the CO/H₂ ratio on the conversion of NH₃ to NO. In another study, Adoune et al. (2002) reported that CH₄ in the gasified biomass fuel mixture resulted in higher NH₃-to-NO conversion compared to the fuel mixture in the absence of CH₄.
Prompt mechanism:
Prompt NO is formed when nitrogen in the combustion air reacts with a hydrocarbon radical, CH, to form intermediate species such as HCN, N and CN. The prompt mechanism can be described as follows (Turns, 2006):

\[ \text{CH} + \text{N}_2 \leftrightarrow \text{HCN} + \text{N} \quad (\text{R24}) \]

\[ \text{C}_2 + \text{N}_2 \leftrightarrow 2\text{CN} \quad (\text{R25}) \]

The intermediate species then react further in a similar way as in the fuel-NO mechanism described above, to finally form NO. This NO formation is dominant at lower temperature fuel-rich conditions of premixed combustion and diffusion combustion and occurs at short residence times. With regards to the small percentage of hydrocarbon in gasified biomass fuel mixtures, prompt NO contribution to the total NO emission may also be small.

The interaction of other components in the gasified biomass may have a significant influence on the production of CH species especially the content of diluents. Göke et al. (2012) experimentally investigated combustion of a fuel mixture containing H₂, CH₄ and H₂O in rich-lean-burn and premixed combustors. For both combustors they found that NOₓ formation through the prompt route can be increased when steam is present in methane-containing fuels. This is because the steam addition slightly increases the concentration of CH radicals in the flame, which are the critical species for the initiation reaction shown above. Giles et al. (2006) reported that presence of methane in their fuel mixture reduced the peak flame temperature and thus reduced thermal NO formation. However, the disadvantage is that methane both reduced the effectiveness of diluents for NO reduction and increased the NO formation through the prompt route. In another study, Lee et al. (2001) found in their numerical investigation of diffusion combustion of CH₄/CO₂ fuel mixture that CO₂ reduced not only thermal NO, but also prompt NO.

N₂O-intermediate:
NO formed from the N₂O-intermediate mechanism is important in fuel-lean combustion and at low temperature conditions. The radicals O and H are important in this NO formation route. The three steps of this mechanism are (Turns, 2006)

\[ \text{O} + \text{N}_2 + \text{M} \leftrightarrow \text{N}_2\text{O} + \text{M} \quad (\text{R26}) \]

\[ \text{H} + \text{N}_2\text{O} \leftrightarrow \text{NO} + \text{NH} \quad (\text{R27}) \]

\[ \text{O} + \text{N}_2\text{O} \leftrightarrow \text{NO} + \text{NO} \quad (\text{R28}) \]
where \( M \) in (R26) is a general third body. This mechanism is favourable at elevated pressure. Furthermore, the \( N_2O \) mechanism is most important under conditions when the total NO formation rate is relatively low (Turns, 2006).

**NNH mechanism:**

The NNH radical is produced when \( N_2 \) reacts with a hydrogen radical, H. Two essential reaction steps of NNH radical production are (Ding et al., 2011);

\[
N_2 + H \leftrightarrow \text{NNH} \quad \text{(R29)}
\]

\[
\text{NNH} + O \leftrightarrow N_2O + OH \quad \text{(R30)}
\]

NNH mechanism will become important in flames involving high concentrations of H and O radicals. Harrington et al. (1996) reported the evidence of production of NO by NNH mechanism in pure \( H_2 \) combustion. With a combustion temperature low enough to avoid thermal NO production they measured ppm levels of NO. For gasified biomass fuel mixture containing \( H_2 \), the NNH mechanism may also play a significant role in its NO formation.

1.4.3.2 CO emission

According to Whitty et al. (2010), the emission of carbon monoxide from the combustion of gasified biomass results from two primary sources. The first source is the unburned carbon monoxide of the fuel mixture itself due to the inefficient mixing that results in an equivalence ratio outside of the ignition range. The second is the incomplete combustion of hydrocarbon species in the gasified biomass.

In terms of fuel composition, the oxidation of CO to \( CO_2 \) can be accelerated by the presence of hydrogen, hydrocarbons and/or water components through reaction (R18) as mentioned previously. Therefore, as gasified biomass may contain all the contributors for hydrogen-containing radical species, low CO emissions in the combustion devices can be achieved. Alavandi and Agrawal (2008) experimentally investigated lean premixed combustion of a fuel mixture containing \( H_2 \), CO and \( CH_4 \). The results show that increasing \( H_2/CO \) ratio in the fuel mixture decreased the CO emissions. Schefer et al. (2002) and Tseng (2002) used swirl-stabilized combustors fuelled by \( CH_4 \) enriched with \( H_2 \). They showed low CO emission due to the presence of \( H_2 \) which created a high concentration of the radical pool and thus enhanced the CO oxidation.

In contrast to the function of \( H_2 \)-containing components in gasified biomass, the content of diluents may result in high CO emissions. It is well known that diluents in gasified biomass fuel mixture tend to decrease the flame temperature of the gasified
biomass and potentially suppress the oxidation of CO resulting in a high CO emission (Hui et al., 2007).

1.4.4.2 Unburned Hydrocarbons emission

Unburned hydrocarbons (UHC) include the hydrocarbon-containing fuel component itself that escapes from the combustor without being oxidized and the products of thermal degradation of these species that have lower molecular weight (Lefebvre & Ballal, 2010), (Turns, 2006).

In the case of gasified biomass, for example from air blown gasification, the major hydrocarbon species in the fuel mixture is CH₄ (1-5%) (Laurence & Ashenafi, 2012). There are also other slightly higher hydrocarbons in a gasified biomass fuel mixture such as acetylene (C₂H₂), ethylene (C₂H₄) and ethane (C₂H₆) (Gai & Dong, 2012), (Gil et al., 1999) and (Huynh & Kong, 2012). Normally in premixed combustion, the content of UHC in the combustion products is negligible (Turns, 2006).
1. 5 Objectives

As shown in the literature review of this thesis, there are many studies on the combustion of one to three fuel components, but there are fewer studies reporting data on real gasified biomass containing all the species CO/H₂/CO₂/CH₄/N₂. Besides CO₂ and N₂ (if air is used as gasifying agent), H₂O is also a non-combustible component in gasified biomass. In research studies of laminar flame speed, there are several studies that considered the individual effect of CO₂ and N₂ on the combustion of a certain gas mixture, as reported in the literature review but studies involving both CO₂ and N₂ are considerable rare. Considering the effect of H₂O on the laminar flame speed, this was found for binary fuel mixtures, but not for complete gasified biomass (see also paper IV as defined in the Preface). No study was found for a gasified biomass mixture including all three of these main non-combustible components simultaneously. Additionally, data on the combustion and emission characteristics of such a fuel mixture is very limited.

Another important component of gasified biomass that should be taken into consideration is tar. Tars are combustible components and contribute to increasing the heating value of the gasified biomass. As long as the tars stay in gas form, they may contribute positively to the combustion process. The advantage of using gasified biomass in a heat application and not for power generation is that it can be directly applied after the gasifier (after particulate removal) without the need for cooling. Tars will then be present in the gasified biomass when used. No combustion studies were found by the author for gasified biomass including tar as a part of the fuel mixture studying its effect on the emission characteristics as well as the laminar flame speed.

Considering the factors described above, the main objective of this study is to determine emission and flame data from premixed combustion of gasified biomass and compare this with that of natural gas (here simulated as methane) for the practical aspect of using a fuel flexible combustor for a heat application at atmospheric pressure. The data also would be a platform applied to combustion of gasified biomass using more complex fuel flexible combustors.

The specific objectives of this thesis are:

- To define the influence of H₂O and tar compound on fundamental combustion properties by experimentally measuring laminar flame speed of gasified biomass and compare to this to other combustible gases (paper I and paper IV).
• To find optimal operating conditions of a combustion system fuelled on gasified biomass containing CH₄ by experimentally investigating the combustion of these gases in an atmospheric premixed combustor (paper II).

• To define the influence of H₂O and tar compound on the combustion performance of gasified biomass by experimentally investigating the combustion of gasified biomass at lean-premixed conditions (paper III).

• To define the influence of different compositions of gases included in gasified biomass on the laminar flame speed and the lean blowoff limit and to develop empirical correlations (paper IV).
2.0 RESEARCH APPROACH

Biomass gasification is currently a very active research area since it enables biomass to be used in various applications due to its gaseous form. Among the different uses of gasified biomass, heat and/or electricity production based on gas combustion processes has been recognized as one of the main applications which at present exists in commercial biomass gasification plants. The ongoing research in the biomass gasification technology focuses not only on the improvement of the gasification itself and its cleaning system, but also on the subsequent combustion process.

The challenges associated with efficient combustion of gasified biomass include flame stability, varying LHV of the fuels as well as the contaminant content of the fuels such as tar. The common approach of combusting gasified biomass includes modification of an existing combustor for conventional fuel or development of a new combustor for a particular composition of the gasified biomass. The new research proposal herein is to develop an effective fuel flexible system which can use a wide range of combustible gases, from low to medium LHV gas, as well as backup fuels such as natural gas.

This thesis consists of a series of experimental studies for the development of a combustion system which is able to handle various mixtures of gasified biomass fuel. The first focus is given to the combustion of low LHV gas as this kind of gasified biomass is largely produced due to both the availability of air as a gasifying agent and to the economical aspect; air does not cost anything to produce in comparison to both pure oxygen and steam. The second focus is given to the combustion of different fuel mixtures of gasified biomass, covering its low and medium LHV, as well as a backup fuel simulating natural gas. There are two important technical areas addressed in this study as highlighted below:

1. **The varying composition in low LHV gasified biomass combustion**: In air blown biomass gasification, the variability in gasified biomass fuel composition is particularly caused by changes in biomass feedstock used as well as changes in the gasifier operating parameters. The combustion of low LHV gas itself can lead to problems associated with the flame stability and thus there is a need to investigate whether changes in the fuel composition mentioned earlier will enhance this problem. The focus is on the changes in the content of H₂O and tar compound of low LHV gasified biomass, which has not been studied previously. Also of importance is the influence of the varying fuel composition on the emission levels.
2. **The requirement of fuel flexible combustors**: A combustion system that can accommodate a wide range of gasified biomass compositions including backup fuel and that achieves better efficiency and lower emissions could lower the operating costs of the overall biomass based heat/power generation plant. Maintaining a stable combustion process and low emission levels are main challenges associated with the development such a universal fuel combustor. Therefore, besides the information on one specific gasified biomass fuel mixture and the backup fuel combustion, the development of fuel flexible combustion systems requires data compilation of combustion properties associated with flame stability to ensure a smooth and reliable operation in combustion devices. Particularly, laminar flame speed and blowoff limit are among the examples of important properties to ensure the combustion is in the stable region.

The research approach in this study is to address these two areas described above with a focus on premixed combustion applications. For the first part, a series of experimental work has been done. The laminar flame speed measurements are based on the burner stabilized flame method using a Schilieren photography system (paper I). For the purpose of combustion temperature, emission levels and lean blowoff limit measurements, a premixed combustion system has been used (papers II-III). The combustor consists of a combustion liner made of high temperature resistant fibrothal ceramic fibre, a premixed burner with a fuel injector, two tangential air inlets that are placed close to the fuel injector and a swirl air inlet that is close to the burner exit. The combustion facility used in this study is also equipped with gas and liquid flow measuring devices, heating system thermocouples, and gas sampling devices. For the second part of the study, a data compilation for laminar flame speed and lean blowoff limits for several fuel mixtures that resembles gasified biomass fuel mixture is used (Paper IV).

This complete study is composed based upon four separate publications as mentioned earlier. The detailed research approach for each publication is explained in the following sub-sections.
2.1 Influence of water vapour and tar compound on laminar flame speed of gasified biomass gas (Paper I)

Paper I focuses on the influence of gasified biomass composition on fundamental combustion properties such as laminar flame speed. As a comparison, CH₄ is also used. The gasified biomass used in this study is a simulated mixture and contains the major components of gasified biomass.

2.1.1 Objectives of paper I

Previously, no research has been performed on the laminar flame speed of a specific gasified biomass mixture with the inclusion of water and tar compound. Therefore, the main objective is to provide laminar flame speed data for gasified biomass in which the simulated gas mixture composition used is close to the actual gasified biomass composition which includes both water and tar. Particularly, the paper contains the following aspects:

- Specification of the influence of the gasified biomass composition on the laminar flame speed
- Specification of the influence of water or tar compound addition to the gasified biomass mixture on the laminar flame speed
- Specification of the influence of temperature and equivalence ratio on the laminar flame speed of gasified biomass containing tar, as compared to water
- Evaluation of thermodynamic and transport properties (diffusivity and Lewis number) related to changes in the laminar flame speed

2.1.2 Research approach of paper I

The burner stabilized flame approach was used and the laminar flame speed was measured on the basis of an area-weighted average of the laminar flame speed. The value of the laminar flame speed was calculated from the mass conservation balance, which is the ratio of the volumetric flow of the unburned air-fuel mixture and the flame surface area.

The simulated dry gasified biomass mixture of CO (25 % volume), H₂ (12.5 % volume), CH₄ (2.5 % volume), CO₂ (10 % volume) and N₂ (50 % volume) was used as a fuel in an atmospheric burner with a straight cylindrical nozzle. The composition of this mixture resembles the gasified biomass produced by an atmospheric, air downdraft gasification of wood pellet as published by Erlich and Fransson (2011). For water, the volume fraction was varied from 0 to 5 %, while the volume fraction for benzene (the model tar) was varied from 0 to 10 %. Benzene was used as a model tar compound because of its chemical stability and since it has a high proportion of the total tar content in actual gasified biomass fuel mixture (Basu, 2010), (Zhang et al., 2011).
The measured parameters in the laminar flame speed experiments were:

- Air, gasified biomass fuel mixture, water or benzene flows
- Temperature of the water/benzene injection line and temperature inside the premixing section of the burner
- Radius and height of the conical flame surface. (The flame surface area was captured by a Schlieren photography system)

The experimental setup was first validated by measuring the laminar flame speed of CH₄-air mixture and comparing with data from other researchers. Next, the measured laminar flame speeds of dry gasified biomass at different equivalence ratios were compared with data of other research studies on dry gasified biomass. The influence of the initial temperature of the air-fuel mixture, water vapour and benzene vapour were measured at different equivalence ratios.

The adiabatic flame temperature, thermal diffusivity and Lewis number were calculated at stoichiometric conditions to further investigate the influence of tar compound and water on transport phenomena in the flame.
2.2 Emission characteristics and lean blowoff limit of simulated gasified biomass gas in a premixed combustor

Paper II focuses on the combustion of gasified biomass and CH₄ in an atmospheric, premixed type combustion chamber. The gasified biomass used in this study has the same composition as used in the experimental work for paper I, but without added water or tar compound.

2.2.1 Objectives of paper II

As discussed in the introductory part of this thesis, the availability of combustion data for gasified biomass is relatively rare, especially for gas from air gasification. For the fuel mixture used in this study, no lean blowoff data has been found in the literature by the author. Therefore, the main objective is to determine the blowoff limit and emission levels for low LHV gasified biomass and CH₄ in one and the same burner at the same input power and equivalence ratios. This gas is characterized by relatively high content of diluents and thereby has a low heating value. Furthermore, the aim is to demonstrate the combustor operation with two different fuels, one with high LHV and the other with low LHV. Particularly, the paper contains the following aspects:

- Specification of the influence of equivalence ratios on CO, UHC and NOₓ emissions and the temperature distribution inside the combustor for both gasified biomass and CH₄
- Determination of the lean blowoff limit of gasified biomass and CH₄ combustion and their respective temperature behaviour during this event
- For gasified biomass, the above aspects are repeated for different input thermal loads
- Observation of auto-ignition and flashback occurrence

2.2.2 Research approach of paper II

The same dry gasified biomass composition as in Paper I was used as fuel for an atmospheric premixed combustor. The experiments were conducted from an equivalence ratio of one to fuel-lean condition. The input thermal load of the combustor was varied from 1.5 kW to 3.0 kW. The temperatures inside the premixing section were measured to observe any occurrence of auto-ignition or flashback. The lean blowoff event was determined once high UHC and CO emissions were measured and/or a sudden popping sound inside the combustor was produced since the spark plug reignited the incoming fuel-air mixture. CH₄ was tested under similar experiment conditions to compare a high LHV fuel with a low LHV fuel in the same combustor.
The measured parameters in the combustion experiment were:

- Air and fuel flows
- Temperature distribution inside the premixing section of the burner and in the combustion chamber
- Concentration of CO, NOx, UHC, CO₂ and O₂ in the combustion gas

The relation between the emission characteristics and blowoff limits with the combustion temperature was discussed. Thermal and chemical effects associated with the experimental results are suggested.
2.3 Influence of water vapour and tar compound on combustion of simulated gasified biomass gas

Paper III focuses mainly on the influence of water vapour and tar compound on the combustion of simulated gasified biomass. The gasified biomass used in this study is a simulated mixture containing the major composition of gasified biomass from air gasification with different levels of added water and tar.

2.3.1 Objectives of paper III

In addition to the relatively little data available for dry gasified biomass, no data has been found (by the author) for combustion of low heating value gasified biomass with addition of tar compound and water. Therefore, the main objective is to determine emission levels and blowoff limits for gasified biomass combustion when different levels of water vapour and tar compound are added to the gasified biomass. Particularly, the paper contains the following aspects:

- Specification of the influence of water vapour and tar compound addition in gasified biomass fuel mixture on its combustion temperature, concentration of CO, UHC and NOx and lean blowoff limits
- Observation of auto-ignition and flashback occurrence

2.3.2 Research approach of paper III

The same dry gasified biomass composition as in Paper I and II was used as fuel for an atmospheric premixed combustor. For water, the volume fraction was varied from 0 to 15 %, while the volume fraction for benzene (a model tar) was varied from 0 to 5 %. The combustion tests were performed at lean-fuel condition. The temperatures inside the premixing section were measured to observe any occurrence of auto-ignition or flashback. The lean blowoff event was determined once high UHC and CO emissions were measured and/or a sudden popping sound inside the combustor was produced since the spark plug reignited the incoming fuel-air mixture.

The measured parameters in the combustion experiments were:

- Air, gasified biomass fuel mixture, water and benzene flows
- Temperature distribution inside the premixing section of the burner and in the combustion chamber
- Concentration of CO, NOx, UHC, CO₂ and O₂ in the combustion gas

The relation between the emission characteristics and blowoff limits and the combustion temperature and equivalence ratio was discussed. Thermal and chemical effects associated with the experimental results are suggested. Analysis of the influence of the addition of water and tar compound is done.
2.4 Correlations of laminar flame speed and lean blowoff limit with the fuel composition of gasified biomass

Paper IV focuses on the potential of combusting gasified biomass with widely different gas compositions in the same combustion system by comparing a larger number of research studies in comprehensive diagrams. The fuel mixtures within this study cover a wide range resembling the variation in composition of gasified biomass.

2.4.1 Objectives of paper IV

There are a lack of data considering the combustion parameters of a wide range of gasified biomass. Therefore, the main objective is to empirically derive relations between combustion parameters and the composition of the gasified biomass based on collected open literature data. The combustion parameters studied are laminar flame speed and lean blowoff limit of various compositions of gasified biomass. Particularly, the paper contains the following aspects:

- Laminar flame speed as function of H₂ volumetric fraction, CO/H₂ ratio, hydrocarbons/H₂ ratio and diluents/H₂ ratio.
- Lean blowoff limit as function of H₂ volumetric fraction, CO/H₂ ratio, hydrocarbons/H₂ ratio and diluents/H₂ ratio.

2.4.2 Research approach of paper IV

The approach for this paper was to gather the available data on laminar flame speed and lean blowoff limit of gas mixtures resembling gasified biomass. By finding key parameters in common for the combustion studies performed, they could be merged into one and the same graph for analysis.

The data from the various gasified biomass fuel mixtures were separated into several groups in order to verify the influence of different components in gasified biomass on the parameters studied (refer to Table 1 and 2 in paper IV). The data were plotted against the volumetric percentage of H₂ in the gasified biomass as well as the ratios of CO/H₂, hydrocarbons/H₂ and diluents/H₂. The references associated with the trends in the graphs were suggested. Mathematical expressions were then derived from the graphs whenever a clear trend could be seen.
3.0 MAIN RESULTS

3.1 Influence of water vapour and tar compound on laminar flame speed of gasified biomass gas (Paper I)

The laminar flame speed is considered as a starting point for characterizing the combustion of gasified biomass. Knowledge of the laminar flame speed is essential to determine the behaviour of turbulent flames that is the actual flame condition in most combustion devices. Several other combustion parameters of importance when designing a combustor are developed from the laminar flame speed data. The composition of gasified biomass has an influence on its laminar flame speed. Specifically water is a non-combustible constituent which needs to be considered since it can react chemically, while tar is one of the combustible components that tend to increase the heating value of gasified biomass significantly.

For Paper I, the main results obtained were as follow:

- The dry gasified biomass-air mixture has a lower laminar flame speed when compared to the methane-air mixture. Among the main reasons is that a dry gasified biomass mixture contains a significant amount of diluents, i.e. CO₂ and N₂. The presence of diluents reduces the heating value of the gasified biomass (dilution effect), which also results in lower adiabatic flame temperature and hence the reaction rate (thermal chemical effect).

- The maximum laminar flame speed of dry gasified biomass was found at slightly fuel-rich condition. The laminar flame speed decreased as the mixture approached fuel-lean condition, while it was almost constant at richer conditions.

- From the comparison with data from other research studies, the difference in fuel mixture composition also influenced the position of a maximum laminar flame speed in which this maximum value was determined at richer conditions for the gasified biomass with higher CO, H₂, and thermal diffusivity.

- Increasing the initial temperature of the gasified biomass increases its laminar flame speed due to the influence of both diffusivity and thermal-chemical effects. The thermal diffusivity and the adiabatic flame temperature increased when the initial temperature of the fuel mixture was elevated.

- There was only a small decrease of the laminar flame speed with the addition of water vapour in the gasified biomass. The change is influenced by the thermal...
reactivity effects as the adiabatic temperature decreases with an increase of $\text{H}_2\text{O}$ in the gasified biomass fuel.

- The effect of benzene on laminar flame speed of gasified biomass exhibits a non-monotonic behaviour. It first decreases when more benzene vapour is injected in the gasified biomass mixture as the diffusivity effect is suggested to dominate. This is because addition of hydrocarbon can decrease the diffusivity of the mixture. With further increase in the amount of added benzene, the laminar flame speed reaches a minimum value, and thereafter increases again since now the thermal and chemical effects are suggested to dominate.

Overall, these results contribute to the increased understanding of combustion of low LHV gasified biomass with the inclusion of water and tar compound.
3.2 Emission characteristics and lean blowoff limit of simulated gasified biomass gas in a premixed combustor (Paper II)

As a continuation of the results obtained in Paper I, the study in this paper investigates the emission characteristics and lean blowoff limit of the low LHV gasified biomass in comparison to natural gas. The air-fuel ratio in a combustion process highly affects the temperature development and chemical reaction paths. If not adapted and optimized to the fuel used, unwanted factors can develop to a larger extent than necessary, for example blow-off, flashback, auto-ignition (gas combustion) and high levels of CO, NOx and UHC. Therefore, careful control of the air-fuel-ratio is needed not only to ensure a safe and stable combustion process, but also to manage the emission levels. The lean blowoff limit for gasified biomass in a particular combustor is important to determine since blowoff always happens near the air-fuel-ratio that gives the minimum CO emission. In this paper both emission levels and blow off limit for premixed combustion of gasified biomass are determined as well as for combustion of CH4 for comparison purpose. The effect of the input thermal power of the combustor is evaluated.

For Paper II, the main results obtained were as follow:

- The combustion temperature of gasified biomass is lower compared to that of CH4 at the same input thermal power. This is mainly due to the fact that the non-combustible constituent of the gasified biomass causes a larger volume to be heated. The high specific heat capacity of this non-combustible volume gives rise to a larger amount of the available heat being absorbed for heating purpose than for combustion of CH4 (thermal-dilution effect).

- In the specific combustor and at the specific dry gasified biomass composition used here, as low as about 15 ppm and about 5 ppm of CO and NOx emissions, respectively, could be achieved at an equivalence ratio of 0.5. UHC emission from gasified biomass combustion is negligible since there is only small fraction of hydrocarbon (CH4) included in the gasified biomass tested here. In comparison, both the lowest CO and NOx emissions for CH4 were measured as 8 ppm, at an equivalence ratio around 0.7 - 0.6. However, UHC emissions for CH4 increase abruptly from a negligible value once blowoff occurred.

- At lean condition, the combustion for both fuels is stable as long as the combustion temperature is well above 800°C. Below this limit, CO and UHC emissions start to rise from their stable and low concentration. The blowoff limit is determined when the CO emission exceeds 200 ppm. For gasified biomass, the lean blowoff limit is at an equivalence ratio of 0.50. This is at a leaner condition compared to CH4 of which the blowoff is at an equivalence ratio of 0.625.
Another observation is the occurrence of blowoff for gasified biomass is more abrupt than for CH₄.

- For similar operation conditions, the flame region of CH₄ is stabilized closer to the premixer zone exit compared to gasified biomass at the same thermal power input. It is suggested that this is due to the larger volumetric flow of gasified biomass compared to that of CH₄, which in turn causes a higher mean velocity of gas.

- For an increase in thermal power input of gasified biomass in the particular combustor, the CO emission is varied, while NOₓ emission is constantly reduced. The varying CO emission is suggested to have four limiting factors, with each one dominating at a certain operational condition: higher volumetric flow, lower temperature limit, availability of oxygen and residence time,

- The occurrence of auto-ignition or flashback can be detected by the temperature development in the premixer zone. However, neither one of these events are observed or recorded at the test conditions within this study.

These results have contributed to the understanding of combustion of low LHV gasified biomass in comparison to natural gas which has higher LHV in the same combustor.
3.3 Influence of water vapour and tar compound on combustion of simulated gasified biomass gas (Paper III)

Paper III complements the previous papers with a study on the emission characteristics and lean blowoff limit of gasified biomass with the addition of water and tar compound. As the water vapour and tar compound slightly change the laminar flame speed of gasified biomass, it will also affect other combustion parameters inside the combustion chamber. In this paper, the gasified biomass fuel mixture with water vapour or benzene vapour is combusted at two lean equivalence ratios: 0.7 and 0.6. These ratios were selected based on the results in Paper II; the combustion is stable and emissions are acceptable at these conditions. The effect of benzene as compared to that of water addition on the combustion and the emission levels can then be compared.

The combustion temperatures as well as the emissions of CO, NO\textsubscript{x}, and UHC are observed at different water and benzene vapour addition levels. The lean blowoff limit for each fuel mixture is determined. The occurrence of auto-ignition and flashback are observed.

For Paper III, the main results obtained were as follows:

- For water vapour addition up to 5%, the combustion temperature is nearly constant. By further increasing the addition of water, the combustion temperature is decreased.

- The NO\textsubscript{x} emission is decreased for water addition up to 5%, and levelled off when further water is added up to the level of 15%.

- The CO emissions are roughly constantly low with all levels of water vapour addition, except when the combustion temperature is approaching 800°C at an equivalence ratio of 0.6 and water addition of 10%. At this point, the CO emission begins to rise and more than 200 ppm is measured as the water addition level increased to 15%.

- The blowoff limit is nearly constant at ER=0.50 with the blowoff temperature remaining almost steady but lower compared to dry gasified biomass when the water addition is increased until 5%. Afterwards, further increase in the water vapour addition up to 15% increased the blowoff limit and the combustion temperature for this limit.
• Addition up to 5 % of benzene vapours into gasified biomass fuel increased the combustion temperature and the NOx emission; while the corresponding CO emission was decreased.

• The blowoff temperature is increased slowly, while the blowoff limit is increased for a benzene addition up to 2 %. With further addition up to 5%, the blowoff limit becomes nearly constant at ER=0.53 while the blowoff temperature continues to increase.

• For lean (ER=0.7 and 0.6) combustion of gasified biomass with addition of H2O and C6H6 vapours no UHC emissions were observed in the combustion gas.

• No auto-ignition or flashback is detected with water or benzene vapour addition in the combustion conditions tested here.

The results contributed to understanding of the emission level and blowoff condition of low LHV gasified biomass that contained water and tar compound, which have not been studied previously.
3.4 Correlations of laminar flame speed and lean blowoff limit with the fuel composition of gasified biomass (Paper IV)

Paper IV complements the previous papers with a study on the combustion parameters of a wide range of gasified biomass mixtures. A fuel-flexible combustion system must avoid combustion operating issues such as flashback, auto-ignition and blowoff, independently of fuel type or fuel impurities while producing low levels of undesired emissions. Therefore, the influence of two fundamental combustion parameters, namely laminar flame speed and lean blowoff limit (equivalence ratio), for various gasified biomass fuel mixtures are determined by merging data from literature into comprehensive graphs. The data are at stoichiometric, atmospheric pressure and room temperature. For fuels with * symbol, the initial temperature is higher than room temperature (see paper IV).

For Paper IV, the main results obtained were as follow:

- The laminar flame speed for most of the gas mixtures is increased with the increment in H₂ content, owing to the higher diffusivity and reactivity of H₂ compared to other fuel components. Laminar flame speed for diluted CO/H₂ (with CO₂ or N₂) is greater than for diluted CO/H₂/CH₄ (with CO₂, CO₂/N₂ or CO₂/N₂/H₂O) fuel mixtures for H₂ content between 20 – 40 %. This condition is suggested due to the content of hydrocarbon and diluent of latter fuel mixtures. Three empirical correlations are derived for fuel mixtures of CO/H₂/N₂, CO/H₂/CH₄/CO₂ and CO/H₂/CH₄/CO₂/N₂.

- The reduction of laminar flame speed for CO/H₂/CH₄/CO₂/N₂ with the enhancement of CO/H₂ ratio is clearly seen and an empirical correlation is established. The reduction may be caused by the lower diffusivity and reactivity of CO compared to H₂. Other gas mixtures showed less dependence on CO/H₂ ratio and suggested to be more dependent on other fuel components.

- There is a clear decrease trend for laminar flame speed of CO/H₂/CH₄/CO₂ when the hydrocarbons/H₂ ratio is increased and an empirical correlation is developed. CO/H₂/CH₄/CO₂/N₂ and CO/H₂/CH₄/CₙHₙ/CO₂/N₂ fuel mixtures also seem to follow a similar trend. The decrement can be attributed to the reduction in diffusivity and flame temperature by the content of hydrocarbon in the gas mixture.

- Laminar flame speed for CO/H₂, CO/H₂/CH₄/CO₂/N₂ and CO/H₂/CH₄/CₙHₙ/CO₂/N₂ gas mixtures is reduced with the enhancement of the diluents/H₂ ratio. The reduction is suggested due to the decrease in the heating value and the increase in specific heat of the gas mixtures which in turn reduce the flame temperature
and thus the reaction rate. The empirical relations for those gas mixtures are formulated.

- Five empirical correlations are obtained for the blowoff equivalence ratio of CO/H₂, CO/H₂/N₂, CO/H₂/CO₂/N₂, CO/H₂/CH₄/N₂ and CO/H₂/CH₄/CO₂/N₂ gas mixtures. The lean blowoff limits for these gas mixtures are decreased (approaching leaner fuel condition) as H₂ content is increased due to the increase in laminar flame speed and reaction rate as mentioned.

- The blowoff equivalence ratio for the fuel mixtures of CO/H₂, CO/H₂/N₂ and CO/H₂/CO₂/N₂ is enhanced, close to richer fuel condition when the CO/H₂ ratio is raised. This can be attributed to the reduction in laminar flame speed by the decrease in reaction rate as explained above, and the narrower flammability limit of CO than H₂. Empirical correlations are derived for these gas mixtures.

- No clear relation is observed between blowoff equivalence ratio and hydrocarbons/H₂ ratio.

- Almost all gas mixtures blowoff at higher equivalence ratio, near richer fuel condition when the ratio of diluents/H₂ content is increased. This is because, in addition to a decrease in the laminar flame speed by reducing the reaction rate, the content of diluent also can also narrow the range of flammability. Empirical correlations are formulated for CO/H₂/N₂, CO/H₂/CO₂/N₂, CO/H₂/CH₄/N₂, CO/H₂/CH₄/CO₂/N₂ and preheated CO/H₂/CH₄/CO₂/N₂/H₂O gas mixtures.

These results contribute to the understanding towards the development of a combustion system that is capable of accommodating a wide range of gasified biomass fuel mixtures.
4.0 DISCUSSION

Simulated gasified biomass gas of a mixture of CO/H₂/CH₄/CO₂/N₂ in such a proportion resembling the low LHV product gas from air-blown wood gasification (high in diluent content) was used in the experimental combustion studies performed within this thesis (paper I-III). Real gasified biomass fuel mixtures also contain both moisture and tars, which is why the addition of water vapour and benzene vapour (simulated tar compound) was studied. The comparison between the low LHV gasified biomass and natural gas (simulated by CH₄) was also made (paper I-II). In paper IV, the behaviour of two of the main combustion parameters for a varied range of gas mixtures approaching low and medium LHV gas mixtures was investigated.

The content of diluents is among the main issues when dealing with the combustion of low LHV gasified biomass. The content of diluents reduces the proportion of combustible components, the heating value, and the flammability range of the gas mixture. It is well known that the content of diluents can affect the combustion characteristics by influencing the adiabatic flame temperature, specific heat of the fuel mixture and chemical kinetic rates (Natarajan & Seitzman, 2010). Through a thermal-dilution effect, the low heating value and large specific heat of the gas mixture consequently reduce the flame temperature and hence the reaction rate. This reason is given to explain the lower laminar flame speed and combustion temperature of low LHV gasified biomass compared to natural gas (paper I-II). In addition to this thermal-dilution effect, the chemical kinetic effect is also suggested to influence the combustion of gasified biomass with H₂O content. H₂O in a fuel mixture can decompose into radicals such as O, OH and H, which enhances the chemical reaction rate or shifts the chemical reaction into different paths. For example, the CO oxidation can switch from a slow reaction (CO + O₂ → CO₂ + O) to a faster reaction (CO + OH = CO₂ + H) in the presence of hydrogen-containing radicals from H₂O (as well as H₂ and hydrocarbon contents). The balance between thermal-dilution and chemical kinetic effects may explain the almost constant laminar flame speed with addition of H₂O (up to 4 % vol.) before it decreases (paper I). The same reason is used to describe the nearly constant combustion temperature, as well as blowoff limit and temperature when a small fraction of H₂O is added to the gaseous fuel mixture (paper III). For a higher H₂O fraction, the dilution-thermal effect is suggested to dominate the combustion process, thus reducing the combustion temperature and increasing the CO emission. This is more pronounced for leaner fuel/air condition than richer condition. However, the increase in volumetric flow (at fixed thermal power) by H₂O addition reduces the time required to generate sufficient heat to ignite the incoming fresh reactant mixture and hence caused the
blowoff to happen at less lean condition and higher combustion temperature (paper III).

In terms of CO emission level, the main contributor for combustion of gasified biomass is suggested to be the unburned CO content in the gas mixture itself (Whitty et al., 2010). CO emission due to incomplete hydrocarbon oxidation is suggested to be a minor factor since there is only a small amount of hydrocarbon content in the gas mixture. In papers II and III, it is observed that for the CO oxidation there seems to exist a temperature limit of about 800°C, below which the CO emission starts to rise from its lower concentration level. The CO emission level is in accordance with the combustion temperature as discussed above when the volume fraction of H₂O is increased (paper III).

Within the temperature range measured for all experiments in papers II and III, the thermal formation of NO is unlikely. The NO emission is then suggested to be contributing by other NO formation mechanisms, i.e. prompt-, NNH-, and/or N₂O-routes. The lower combustion temperature for low LHV gasified biomass is possibly reducing the reaction rate of these formation routes and thereby causing lower NO emission compared to natural gas (paper II). In addition to this thermal-dilution effect by the content of diluents, the reaction for NNH- and N₂O is suggested to also be affected by the availability of O and H radicals since it needs to compete with the CO oxidation and OH+H+M=H₂O+M reaction. This reason possibly describes the reduction in NOₓ emission when a small fraction of H₂O is added to the gas mixture (paper III). For higher H₂O addition, the decrease in temperature may also increase CH and HCO radicals due to incomplete combustion of the content of hydrocarbons, thus an increase in prompt NO is expected. In agreement with the temperature as discussed above, this condition is probably more pronounced for leaner fuel conditions.

Tar compounds can exist in vapour form if the temperature of gasified biomass is maintained above its condensation point prior to the introduction into the combustion chamber, thus contributing positively to the heating value of the fuel mixture. However, the tar content can decrease the diffusivity of the gas mixture. The reduction in laminar flame speed of low LHV gasified biomass with small presence of tar compound (C₆H₆ addition up to 2.5 % vol.), probably due to the domination of diffusivity effect, is demonstrated in paper I. When the volume fraction of C₆H₆ is further increased (to about 10 % vol.), the combination of thermal and chemical effects is suggested to have more influence thus enhancing the laminar flame speed. From paper III, the increase of reactive species of gasified biomass with the addition of C₆H₆ up to 5 % vol. raises the combustion temperature and the NOₓ
emission but lowers the CO emission. It is suggested that the improvement of the overall reaction rate due to the increase in combustion temperature and higher availability of additional radical species by C₆H₆ decomposition results in the lower CO emission but higher NOₓ emission. Furthermore, the narrowed flammability limit of C₆H₆ is suggested to shift the lean blowoff limit of the gasified biomass to the fuel richer equivalence ratio.

In paper IV, it is demonstrated that the laminar flame speed of a wide range of gasified biomass is more influenced by the content of H₂ and diluents compared to the content of CO and hydrocarbons. For the lean blowoff limit, the content of diluents showed the greatest impact followed by H₂ and CO (smaller than that of H₂). The importance of H₂ content in the increase of the laminar flame speed and reduction of the blowoff equivalence ratio (to leaner fuel condition) owes to its higher reactivity and diffusivity compared to other gases (Natarajan & Seitzman, 2010). Both properties result in higher flame temperature and thus the reaction rate for the gas mixture with higher H₂ content. The leaner blowoff limit of low LHV gasified biomass in comparison to natural gas (paper II) is also suggested to be caused by these properties of the H₂ content. While, the content of diluents decreases the laminar flame speed and increases the blowoff equivalence ratio close to richer fuel condition due to thermal-dilution effect as explained previously. CO content has less impact since its oxidation is sensitive to the presence of hydrogen containing radicals (from other components) as discussed above. The reduced importance of hydrocarbon content in laminar flame speed (and no effect found on the lean blowoff limit) is probably due to its significantly low concentration in the gas mixtures studied.
5.0 CONCLUSIONS AND FUTURE WORK

5.1 Conclusion
This thesis serves as a first step towards the development of a combustion system that can handle the variation in the composition and heating value of gasified biomass due to changes during the biomass gasification process and also the wide range of fuel mixtures that cover low to medium LHV gases. This so called fuel flexible combustion system is also expected to accept other conventional fuels such as natural gas as a backup fuel. The first step in such a development is to characterize a number of fuel mixtures with regards to their combustion stability and emission levels.

The combustion studies on the specific composition of low LHV gasified biomass fuel mixtures used in this work, including the addition of water and tar compound, have not been found in open literature. Therefore, the results obtained from this study could be added to the existing data concerning the combustion of low LHV gasified biomass (papers I and III). The test rig used in this study allowed the combustion of low LHV gasified biomass and natural gas to give a same thermal power output without any modification (paper II). It is important to have a combustion system that is able to receive another fuel as a backup while maintaining the low emission levels, for example during maintenance of the gasifier. These issues are important for economic reasons for any industrial production process since every stop in the process is a net cost. The fuel flexible combustion system is supposed to combust a wide range of gasified biomass mixtures. Therefore, the results from paper IV that investigated the influence of fuel composition on the laminar flame speed and the lean blowoff limit is expected to serve as a basis to develop such a combustion system.

The main conclusions from the studies are:

- There is a possibility to use gasified biomass with high dilution content and pure methane in the same premixed combustor without compromising low emission levels. For gasified biomass, lower laminar flame speed, combustion temperature, lean blowoff limit and NOx emissions were measured (papers I – II). Gasified biomass combustion resulted however in somewhat higher CO emission.

- H2O content in a low fraction in the gasified biomass (up to 5%) can be an advantage in terms of NOx reduction and an increase of overall reaction rates through its chemical effect (paper III). On the other hand, H2O also exhibited thermal-dilution effect (papers I and III) and this effect dominated the decline in
laminar flame speed (paper I). At a large fraction of H$_2$O in the gasified biomass, the thermal-dilution effect dominated thus reducing the combustion temperature. This led to higher CO emission and a blowoff limit at richer condition compared to gasified biomass with small or no H$_2$O content (paper III).

- There is a lower combustion temperature limit of about 800°C, above which, low CO emission can be maintained (papers II and III). When the combustion temperature falls below this limit, the CO emission starts to increase rapidly.

- The effect of C$_6$H$_6$ vapour addition in the gasified biomass on laminar flame speed exhibits a non-monotonic trend (paper I). With small C$_6$H$_6$ addition (up to 2.5 % vol.), the laminar flame speed is decreased due to the dominating diffusion effect. When C$_6$H$_6$ is further increased (up to 10 % vol.), the laminar flame speed is increased due to the strong influence of thermal and chemical effects. In paper III, the C$_6$H$_6$ presence in gasified biomass displayed a trend of reduced CO emission and a tendency of increased NO\textsubscript{x} emission and lean blowoff equivalence ratio (richer condition). The increase in temperature due to the enhancement of the heating value of gasified biomass and addition of radicals from the breakdown of C$_6$H$_6$ are among the factors that have been suggested to explain these results.

- In paper IV, it was demonstrated that the content of H$_2$ and diluents content have greater impact on the laminar flame speed compared to the contents of CO and hydrocarbons for most gas mixtures studied that cover low to medium LHV gas mixtures. For the lean blowoff equivalent ratio, the influencing fuel composition in the descending order is the diluents, H$_2$ and CO content. The developed empirical correlations developed in this study can be used for models of the laminar flame speed and lean blowoff limit for varying gasified biomass fuel compositions.

The results of laminar flame speed in this thesis can be used for validation of chemical kinetic models as well as for a turbulent flame speed model, tools that are used for improving and designing the combustion system of gasified biomass as discussed in section 1.4.1. On a practical level, the data of laminar flame speed together with lean blowoff limits, combustion temperatures as well as CO and NO\textsubscript{x} emissions provide the information on the possible trend of these combustion characteristics for the design and operation of combustion devices intended to operate on gasified biomass that vary considerably in composition as well as a back-up fuel such as natural gas.
5.2 Future Work

The following recommendations discuss the various improvements to this study that could be performed:

- Use real gasified biomass from a gasifier.
  The definite gasified biomass composition from a gasifier is difficult to measure instantly and can vary continuously during operation. The variation of the gasified biomass composition may result in a difference in the combustion characteristics, which is why the fluctuations in the combustion data with time need further attention.

- Tests at elevated pressure and temperature.
  The current research is limited for the application of atmospheric combustion devices such as a boiler, industrial or external fired combustors. For example, the laminar flame speed of real gasified biomass at higher pressure than atmospheric is useful for the application of gasified biomass in a gas turbine.

- Measurement of other combustion properties.
  For example, the auto-ignition temperature and delay time are also useful for validation of chemical kinetic models that serve as an important tool in designing combustion devices for gasified biomass.
6.0 REFERENCES


