Development of the MILENA gasification technology for the production of Bio-SNG

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Development of
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Contents

Summary

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Development of the MILENA gasification technology for the production of Bio-SNG

The production of Substitute Natural Gas from biomass (Bio-SNG) is an attractive option to reduce CO_2 emissions and replace declining fossil natural gas reserves. The Energy research Center of the Netherlands (ECN) is working on the development of the MILENA gasification technology that is ideally suited to convert a wide range of biomass fuels into a gas that can be upgraded into Bio-SNG.

Production of a synthetic natural gas that can be readily injected into the existing natural gas infrastructure is a major challenge to make a big step into bringing renewable energy to the public. To achieve such a goal it is necessary to produce an SNG with similar properties as natural gas and also at a price that makes it competitive with current and future prices.

This goal is translated into some major scientific and technological challenges. The process, in which the gasification step is a major one, should have the highest possible thermodynamic efficiency, meaning that most of the calorific value of the input biomass is retained in the product gas. Next to this the quality of the gas should be such that it can be effectively cleaned to allow for a long lasting high efficiency SNG synthesis. This requirement is translated into the goal of making a product gas with minimum non convertible components like nitrogen and H_2O and CO_2. The inherent production of tar like components should be such that these can be beneficially re-used in the process or be converted to components adding to the amount and quality of the SNG.

On top of this the major technical challenge is that the design of the process should be such that it can be up scaled into a process with capacities of well over several hundreds of Megawatts input.

The gasification process fulfilling these technical and scientific challenges is designed to produce a medium calorific value gas (approximately 16 MJ Nm^{-3} on dry basis) with a high content of hydrocarbons like methane and ethylene.

The available knowledge from an existing 500 kWth biomass gasifier was used to make the first design of the MILENA gasifier. On the basis of this the final MILENA gasification process has been established which is best described as an Indirect or Allothermal fluidized bed gasifier. One of the major advantages of Indirect gasifiers is the near 100% conversion of the fuel into a combustible gas and latent heat. The residual ash is virtually carbon free (< 1 wt.% C), which means that the loss in heating value of the remains including the ash is close to zero. The overall efficiency of the MILENA gasifier is relatively high, compared to the alternatives, because of the complete fuel conversion and the relatively low amount of steam required in the process.

The objective of the development described in this thesis was to design an up-scalable biomass gasification process with a high cold gas efficiency (> 80% for dry wood) producing a gas which is suitable to be converted into Bio-SNG with a higher overall efficiency than the alternative biomass gasification processes. The nitrogen content of the producer gas should be below 3 vol.%, to prevent dilution of the Bio-SNG.

Verified relations to calculate the gas composition, compound and energy balances are required for the design of a commercial scale demonstration plant which is scheduled for construction in 2011. Reliable relations for carbon conversion and hydrocarbon yields in an indirectly heated riser gasifier as function of temperature were not available from literature. Data from an extensive test program was used to produce and verify the required relations. The models to describe the process were designed by the author of this thesis. The relations for hydrocarbon yield can also be used for comparable biomass gasification processes (e.g. BFB and CFB gasification), but experimental verification is recommended.

An introduction into Bio-SNG is given and the MILENA development for Bio-SNG production is given in Chapter 1.

In Chapter 2 the biomass gasification process in fluidized bed reactors and the typical problems related to biomass fluidized bed gasification are described.

In Chapter 3 background information is given on the production and usage of Bio-SNG.

Chapter 4 presents an analysis of the obtainable Bio-SNG process efficiency using three different, more or less suitable, gasification technologies and associated gas cleaning and methanation equipment. These technologies are: 1) Entrained Flow, 2) Circulating Fluidized Bed (CFB) and 3) Indirect gasification. Overall efficiency to SNG is highest for Indirect gasification. The net overall efficiencies on an LHV basis, including electricity consumption and pretreatment, but excluding transport of biomass, are 54% for Entrained Flow, 58% for CFB and 67% for Indirect gasification. Because of the significant differences in overall efficiencies to SNG for the different gasifiers, ECN has selected the Indirect gasification as the preferred technology for the production of SNG.

A pseudo-equilibrium model is made to describe the MILENA gasification process. This MILENA model was used to design the lab-scale and pilot-scale installations. The model is described in Chapter 5.

In 2004 the 30 kW_{th} lab-scale MILENA gasifier was built. After successful operation of the MILENA lab-scale gasifier for some years it was, at the end of 2006, decided to start the realization of a pilot-scale gasifier. Construction started in 2007 and the 800 kW_{th} pilot plant was taken into operation in 2008. First tests with the complete system (gasifier and gas cleaning) were done in 2009. The MILENA process and the lab-scale and pilot-scale installation are described in Chapter 6.

An extensive test program was done in the lab-scale and pilot-scale MILENA installations. Different fuels, such as clean wood, demolition wood, sewage sludge and lignite were tested. Test results were used to verify the MILENA model. Tests with demolition wood were done to produce data for the engineering of a MILENA demonstration plant. Results of the lab-scale and pilot-scale tests using different fuels are described in Chapter 7.

Chapter 1

Introduction

1.1 Sustainable Energy

Energy is one of the essential ingredients of modern society. Nowadays energy comes for the greater part from fossil fuels like oil, natural gas and coal. The proven fossil oil and natural gas reserves are declining in North America and Europe [1]. According a study of the Energy Research Centre of the Netherlands (ECN) the global production of oil might decline within 30 years [2]. According to the International Energy Agency (IEA) the consumption of primary energy is expected to increase by 1.6% per year. By 2030 consumption is expected to have risen by just over 45% compared to 2006 [3].

On top of the problem of securing the supply, the combustion of fossil fuels produces CO_2, which contributes to global warming. CO_2 emissions from fossil fuels can, to some extent, be countered by sequestration of CO_2. This CO_2 sequestration, however, lowers overall efficiency significantly, resulting in a higher consumption of fossil fuels per unit of energy delivered and consequently a faster decline of fossil fuels reserves.

Sustainable alternatives like wind, solar or biomass energy are required to replace the declining production of fossil fuels without increasing the amount of CO_2 in the atmosphere.

1.2 Biomass Energy

Biomass energy is expected to make a major contribution to the replacement of fossil fuels. The future world-wide available amount of biomass for energy is estimated to be 200 to 500 EJ per year, based on an evaluation of availability studies [4]. Word wide oil consumption was 161 EJ (82.5 million barrels of oil per day) in 2005 [1].

Biomass is considered a CO_2 neutral fuel, as the amount of CO_2 released on burning biomass equals the amount taken from the atmosphere during growth of the biomass. Fuels like hydrogen, methane, Fischer Tropsch (FT) diesel and methanol produced from biomass have the potential to become a CO_2 negative fuel, because part of the biomass carbon is separated as CO_2 during the production process and can be sequestrated. This might be an attractive option for reducing the level of greenhouse gases in the atmosphere.

Biomass for the production of energy is controversial for several reasons. Corn is used on a large scale to produce ethanol to replace fossil gasoline. Palm oil is used to produce biodiesel. This resulted in the fuel versus food discussion. Large areas of rainforest have been cut down in Malaysia to create space for palm oil production. On top of this, some production processes for Bio-fuels require a large (fossil) energy input for logistic reasons and to upgrade the fuel to an acceptable quality. A well know example is the distillation of the water ethanol mixture to produce fuel quality ethanol. Some fast growing biomasses require nitrogen fertilizers, which are normally produced from natural gas. This has a negative effect on the overall CO_2 balance of the Bio-fuel. To deal with these issues Sustainability Criteria were introduced. These criteria include issues like the greenhouse gas balance, competition with food, biodiversity and local environmental issues. Woody biomass performs very well on these criteria, especially when the wood is converted into a low carbon fuel like methane.

1.3 Biomass gasification

The term gasification applies to processes which convert solid or liquid fuels into a combustible gas at high temperature. The heat required for the heating of the fuel and the endothermic gasification is supplied by the combustion of part of the fuel (Direct gasification) or comes from an external source (Indirect or Allothermal gasification). The MILENA gasifier described in this thesis belongs to this latter category. Background information on biomass gasification is given in Chapter 2.

1.4 Bio-SNG as renewable fuel

Natural gas plays an important role as an energy source world wide. According to the Energy Information Administration of the U.S. Government natural gas consumption in 2003 was one-quarter of the world primary energy consumption and is expected to rise by 2.4 percent per year [5].

Natural gas is a relatively clean primary energy carrier and is therefore often the fuel of choice in many regions of the world. The share of natural gas in the world energy consumption is expected to rise.

In the Netherlands natural gas contributes nearly 50% of the primary energy supply as it is by far the most popular fuel for heating of buildings.

Replacing part of natural gas by a Substitute Natural Gas (SNG) or Synthetic Natural Gas, produced out of a sustainable primary energy source, with the same properties as natural gas makes the implementation of sustainable energy easy as natural gas grids are widely spread in the Netherlands and in many other countries.

Sustainable electricity has become popular in recent years. In 2005 7 percent of the consumers in the Netherlands used sustainable electricity. The number reduced to 5% in 2007. This was a consequence of the fact that the subsidies for production of electricity out of biomass became less available. Presently the majority of the growth in renewable electricity generation is due to wind power. The majority of the electricity out of biomass is from co-combustion in coal fired plants.

Replacing heat produced from fossil fuel with sustainable heat on a household scale is more challenging than replacing fossil electricity with sustainable electricity. Direct local combustion of biomass can be attractive from an efficiency point of view but local biomass boilers (on household scale) would require extensive and expensive flue gas treatment equipment to keep emissions at an acceptable level (compared to emissions from large power plants or the presently used local natural gas fired boilers). Decentralized biomass fired boilers or combined heat and power plants require a district heating network. In most cities district heating network is not present and the installation of such a network is expensive.

A Substitute Natural Gas can be produced from biomass with a high efficiency and with low emissions from the SNG plant (comparable with modern power plants). Biomass transport can be limited to the central SNG plants, which would be located next to harbors.

Bio-SNG is the obvious choice for sustainable heating of houses in the Netherlands and in many other countries. It is likely that in the near future conventional house heating boilers will be replaced by natural gas fired Micro Combined Heat and Power plants (Micro-CHP), which increases overall energetic efficiency compared to local heat production and decentralized electricity production. The production process of Bio-SNG via gasification is described in chapter 3.

1.5 CO_2 balance of Bio-SNG

Biomass (wood) with 20 wt.% of moisture has, by approximation, the composition of $C_6H_{12.5}O_{5.75}$. In theory biomass can be converted directly into a mixture of CH_4 and CO_2 by the following exothermic reaction:

$$C_6H_{12.5}O_{5.75} \rightarrow 3.12\ CH_4 + 2.88\ CO_2 \tag{1.1}$$

After removal of CO_2 the Bio-SNG can be injected into the gas grid. This means that almost half of the carbon (on mol basis) in the fuel is separated as pure CO_2 and is available for CO_2 sequestration. This makes the production of SNG carbon negative. Figure 1-1 shows an indicative overall CO_2 balance, including emissions from harvesting and transport, for a Bio-SNG production facility based on the MILENA indirect gasifier as described is this thesis.

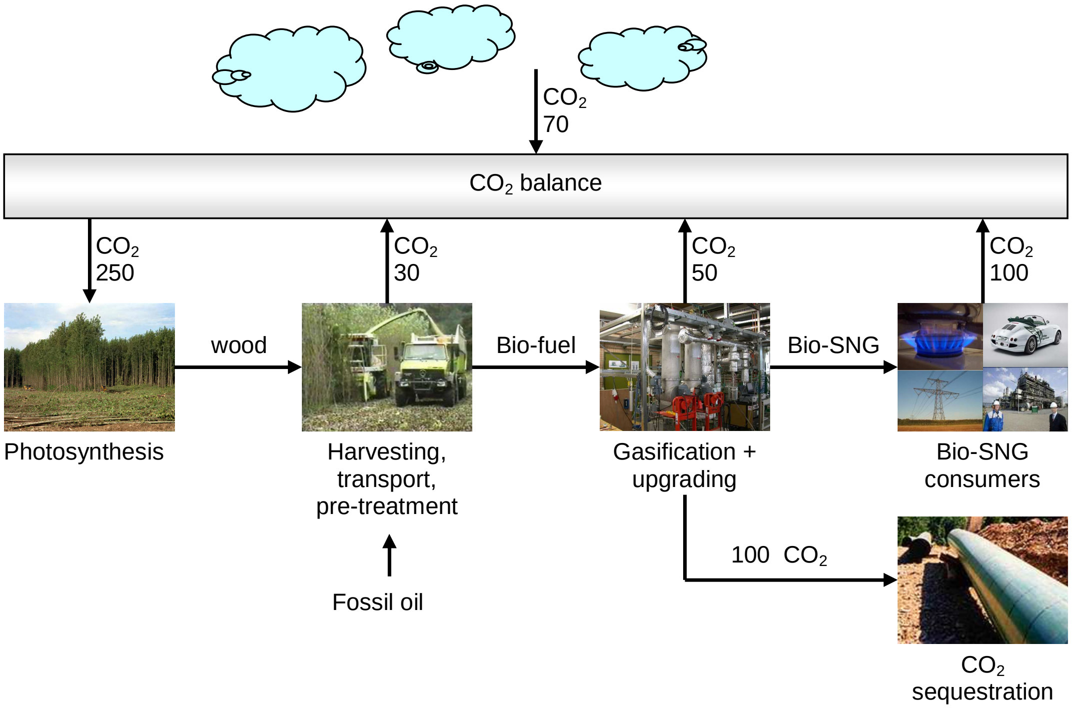

Figure 1-1: Indicative CO_2 balance for Bio-SNG system based on MILENA gasification.

1.6 Bio-SNG development at ECN

The Energy research Centre of the Netherlands (ECN) became interested in the production of Bio-SNG out of biomass already in the nineties. The original interest in SNG was based on the possibility to use biomass to store sustainable hydrogen coupled to sustainable carbon from biomass. This was achieved by the hydrogasification process. Several studies and some experimental work were done on the hydrogasification process [6]. The hydrogasification process uses hydrogen as gasification agent in a pressurized reactor operating around 30 bara and 800°C, where no external heat or oxygen supply is required. Hydrogen reacts exothermally with the carbon in the biomass to form methane. The hydrogasification development was not continued, because it became clear that the availability of a surplus of sustainable hydrogen was not likely for the near future. As a second option the production of SNG using more conventional gasification processes for the production of SNG gained interest.

ECN did the first SNG production tests using a steam-oxygen blown lab-scale gasifier in 2003. These first results were promising and led to the continuation of the research program. Indirect gasification was identified as most suitable gasification technology for the production of SNG [7]. Indirect gasification technology was by that time already in development. Further research concentrated on the production of SNG from gas coming from the ECN MILENA gasifier.

1.7 Objective of the MILENA development

The objective of the MILENA SNG development at ECN is to develop an economically viable and up scalable process for the production of Bio-SNG from cellulosic biomass and to bring this development to the market. The design of the gasifier is made such that it should be up scalable to at least 100 MW_{th}.

The MILENA producer gas should contain a high concentration of CH_4 (>12 vol% on dry basis), because this has a positive effect on overall efficiency to SNG. The N_2 content in the producer gas should be below 3 vol% (dry basis), because N_2 dilutes the final Bio-SNG.

The technology will focus on woody biomass to start with, because experiences at ECN and elsewhere (the FICFB gasifier in Güssing) have shown that woody biomass is an ideal fuel for an indirect gasifier. Figure 1-2 shows the foreseen scale-up steps and demonstration trajectory.

The demonstration of the technology is done with commercial partners, as they are essential for the implementation after a successful demonstration. One of the demonstration steps is a 10 MW_{th} MILENA gasifier in combination with

OLGA gas cleaning and a gas engine. The 10 MW_{th} Combined Heat and Power (CHP) demo is considered to be a crucial intermediate step towards commercial Bio-Methane plants. The CHP demo, however, is also considered to be a demonstration of a commercial size CHP unit and therefore serves two goals. After a successful CHP demonstration, further scale-up to a 50 MW_{th} Methane demonstration unit is foreseen.

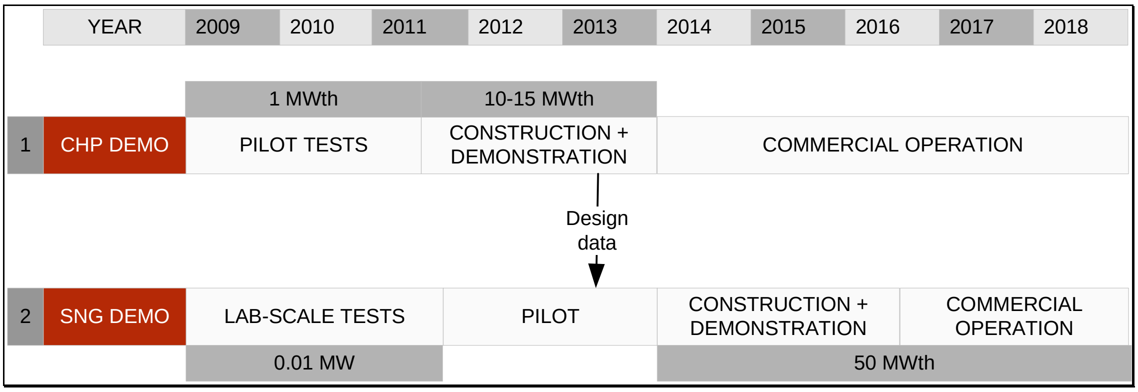

Figure 1-2: Foreseen scale up and demonstration trajectory for MILENA technology.

The development focuses on high overall energetic efficiency, because biomass is seen as a valuable primary energy source. The aim is to have a net overall energetic efficiency from fresh woody biomass to SNG of more than 70% (LHV basis).

1.8 Objective of this thesis

This thesis has two main objectives:

1) Quantify the differences in overall efficiency from wood to Bio-SNG with other biomass gasification processes to prove that the overall efficiency from wood to Bio-SNG is significantly higher than for other biomass gasification processes that will be commercially available within ten years.
2) Generate verified relations that are required to calculate the gas composition, the mass balance and the energy balance for an indirectly heated biomass riser gasifier like the MILENA.

The comparison on overall efficiency basis of indirect gasifiers with alternatives like Entrained Flow and Circulating Fluidized bed gasifiers is described in chapter 4.

The empirical relations used in the model to predict hydrocarbon yields and carbon conversion are based on a wide range of experiments described in chapter 7.

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Chapter 2

Background on Biomass gasification

Abstract

This chapter describes the biomass gasification process in fluidized bed reactors and the typical problems (tars and agglomeration of bed material) related to biomass fluidized bed gasification. Indirect gasifiers and the MILENA process are described in more detail.

A clear definition of Cold Gas Efficiency (CGE) is given, because the recycle of char and tar flows creates several options to define this efficiency.

2.1 Biomass gasification technologies

Gasification processes have been in use since the 1800s. The first application was the production of town gas from coal. From the 1920s gasification was used to produce synthetic chemicals. Most well known is the production of Fischer Tropsch oil out of synthesis gas in Germany to run the military machinery during the Second World War and, more recently in South Africa.

Nowadays, commercial coal gasifiers are in operation on a scale over 1 GW_{th} [1]. The number of gasifiers based on biomass as a fuel is still limited. The technology of gasification to liquid and gaseous fuels on the basis of biomass as feedstock will get a new boost as it opens the road to produce a green alternative to fossil fuel based energy carriers

The term gasification is applied to processes which convert solid or liquid fuels into a combustible gas at high temperature. The heat required for the heating of the fuel and to energize the endothermic gasification reactions is supplied by the combustion of part of the fuel (Direct gasification) or is supplied from an external source (Indirect or Allothermal gasification).

Gasifiers can be divided into high temperature gasifiers (typical 1300 - 1500°C) which produce a syngas and medium temperature gasifiers (typical 850°C) which produce a producer gas. Syngas contains almost no hydrocarbons like methane. Gas coming from medium temperature gasifiers contains on energy basis up to 50% of hydrocarbons (mainly CH_4, C_2H_4 and C_6H_6). The producer gas from medium temperature gasifiers also contains some tars. Tars are heavy hydrocarbons, which can cause fouling problems when the gas is cooled. Producer gas also contains several other pollutants like H_2S, COS, thiophenes, NH_3, HCl, HCN and dust which need to be removed before application of the gas.

For processes like the synthesis of Fischer Tropsch Diesel or methanol the presence of large quantities of hydrocarbons is unwanted, because only CO and H_2 (and probably C_2H_4 in the case of Fischer Tropsch synthesis) are converted into the desired product. Next to the fouling due to heavy hydrocarbons, the other hydrocarbons have negative effects on the downstream catalytic process due to the risk of deactivation. For the production of SNG the presence of hydrocarbons is an advantage, because most of the hydrocarbons are present as CH_4 and the other hydrocarbons can be converted into methane with a higher efficiency than the conversion of syngas into CH_4.

The most common type of gasifier for the production of syngas is the Entrained Flow (EF) gasifier. EF gasifiers operating on coal are commercially available from large companies like Shell, General Electric and Siemens. The typical scale is over 500 MW_{th}. The gasifiers are always operated at high pressure (typically 30 bar), because the syngas is needed at high pressure while the compression of a solid requires less energy than the compression of a gas. Feeding pulverized coal at elevated pressures using lock hopper systems is proven technology, which is not the case for biomass. The solid fuel (mostly coal) is pulverized and pneumatically fed into the Entrained Flow gasifiers. N_2 is normally used as feeding gas. O_2 (diluted with steam) is mostly used as gasification agent. The gasifier is always operated above the ash melting temperature to keep the ash in the liquid phase in the gasifier. The syngas produced by the gasifier is quenched with cooled syngas to solidify the ash.

A well known example of an Entrained Flow gasifier is the Shell gasifier in Buggenum, where the produced syngas is fired in a combined cycle to produce 253 MW of electricity. The fuel for this plant is normally coal, but co-gasification with up to 30 mass% of biomass has been demonstrated [2].

Entrained Flow gasifiers can in principle be used to gasify biomass if the particles are milled. However, milling of biomass particles is energy intensive and the pneumatic feeding of those particles can be problematic. Torrefaction is a biomass pretreatment step under development to reduce the required milling energy and to increase the energy density of the biomass to make a.o. the transport more economic [3]. Another pretreatment option is the production of bio-oil by the flash pyrolysis process. Bio-oil as a liquid can easily be fed into a gasifier using a high pressure pump [4]. The disadvantage of both pretreatment steps is the energy loss caused by the thermal conversion of the fresh biomass into a more manageable fuel. The overall efficiency (including electricity consumption) of both pretreatment methods is still not known, as both processes are still under development and data from commercial scale demonstration units are not yet available. There are no known demonstrations of EF gasifiers using only biomass or pre-treated biomass as a fuel. There is a strong interest from industry in using this coal derived gasification technology for biomass.

The medium temperature gasifiers can be divided in fixed bed gasifiers and fluidized bed gasifiers. The fixed bed gasifiers can be separated in Downdraft and

Updraft gasifiers. Both are in use for biomass gasification as well. Figure 2-1 depicts the basic operating principles of typical Updraft and Downdraft gasifiers.

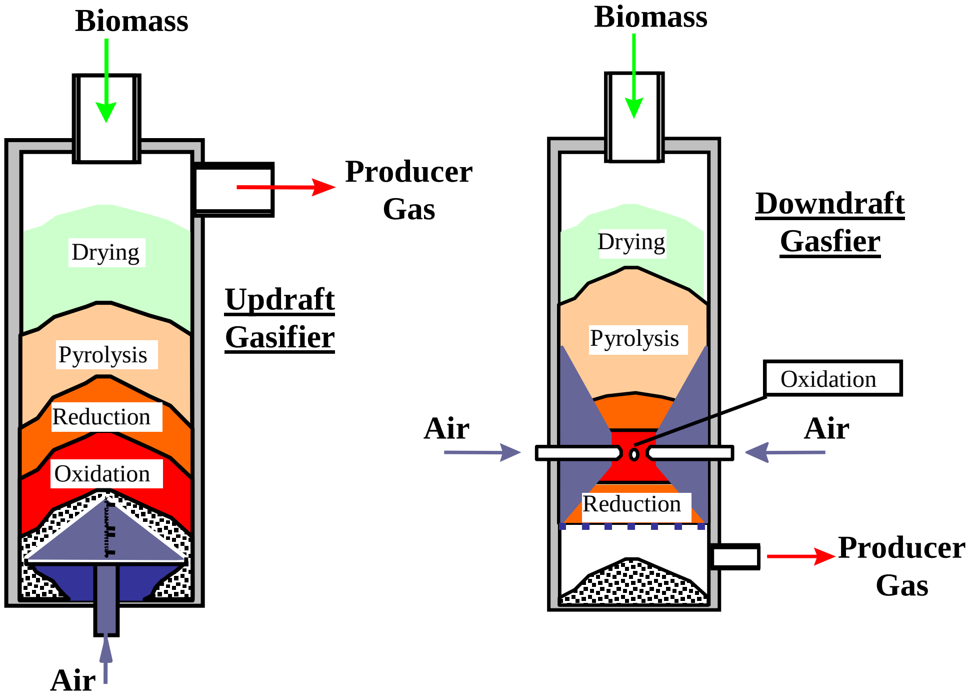

Figure 2-1: Schematic comparison of Updraft and Downdraft Gasification

Downdraft gasifiers are widely in use for small scale CHP generation. The typical size of a gasifier is between 100 and 1000 kW_{th} input. The fuel is normally dry wood. The gas is mostly used to fuel a gas engine. The advantage of this technology is that the produced gas is rather clean (low tar and dust content) and the technology is relatively simple. The gasifiers require a well defined dry fuel for continuous and reliable operation. The carbon content of the ash coming from the bottom of the gasifiers normally is relatively high (>10 wt.%), because the fuel conversion in the gasification section is not complete. The Downdraft technology is not well suited for scaling up. One of the reasons is that scale up requires an increase in bed diameter. A large bed diameter increases the risk of channeling in the bed and leads to inhomogeneous supply of oxygen to the gasification zone. If a channel in the bed occurs, a larger than average part of the gas goes through such a channel and not through the char bed. The char bed should help to reduce the tar concentration in the gas, but if the gas ‘escapes’ through the hole the tar concentration increases and downstream equipment gets fouled.

Updraft gasifiers are better suited for scale up and less sensitive regarding moisture content and geometry of the fuel, but produce a lot of tar. If tar removal technology is applied the gas can be fired in a gas engine. Tar is normally removed in combination with water. This water stream requires extensive cleaning before it can be disposed in a sewer system. The overall efficiency of the Updraft process can be high because of the complete conversion of the fuel and the low outlet temperature of the gasifier. The tar removal and water clean up make the process complex and too expensive for small scale (< 1 MW_{th}). A successful example of an Updraft gasifier is the Harboøre plant in Denmark [5].

Updraft and Downdraft Fixed Bed biomass gasifiers are operated in ‘dry’ mode. This means that the ash in the gasifiers is not a molten state. This is achieved by keeping the operating temperature below the melting temperature of the ash. Both types of gasifiers use air as gasification agent. For the production of SNG the air needs to be replaced by oxygen, as the nitrogen in the gas dilutes the final product. Replacing air by oxygen, however, is not an option, because this would result in a local increase in temperature, which increases the risk of ash melting. Fixed Bed gasifiers are not seen as an option for Bio-SNG production because of the inability of this type of biomass gasifier to produce a nitrogen free gas.

Fluidized Bed gasifiers, as described in the next paragraph, can be operated in such way that they produce a nitrogen free or nitrogen poor gas and the technology is suitable to be scaled up to several hundreds of MW.

2.2 Fluidized bed gasification

Fluidized Bed gasifiers can be divided into three main categories: Bubbling Fluidized Bed (BFB), Circulating Fluidized Bed (CFB) and Indirect or Allothermal twin bed concepts. All Fluidized Bed gasifiers use a bed material. That can be ordinary sand, the ash from the fuel or a catalytically active bed material like dolomite or olivine. The purpose of the bed material is to distribute and transport the heat in the gasifier which prevents local hot spots, mix the fuel with the gasification gas and the produced gases and, in the case of a catalytically active material, reduce the concentration of tars. Figure 2-2 shows the basic principles and differences of three types of Fluidized Bed gasifiers.

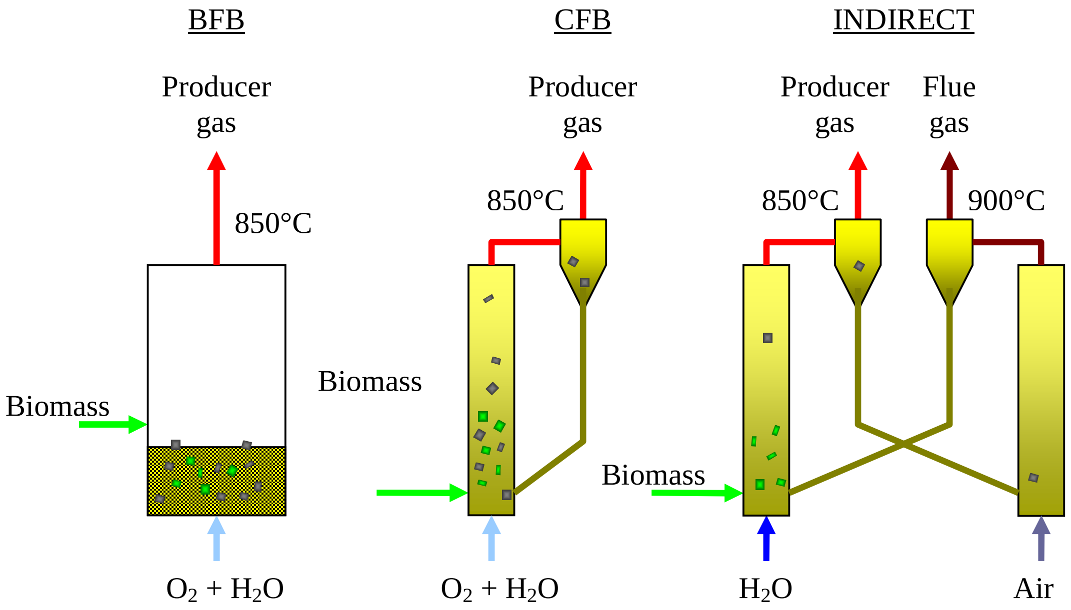

Figure 2-2: Schematic comparison of BFB, CFB and Indirect gasification

In a BFB gasifier the fuel is normally fed in or above the fluidized bed. The bed material is fluidized by a gas (air or an oxygen steam mixture) entering the gasifier through nozzles distributed over the bottom of the reactor. The air is used in the bed to combust part of the gas and/or the char to produce the heat required for heating the biomass and the endothermic gasification processes. The typical gas velocity in this gasifier is 1 m s^{-1}. BFB gasifiers are normally applied at a scale below 10 MW_{th}. The reason for this scale limitation is probably the requirement of a good fuel distribution over the bed, which becomes more difficult when the diameter of the reactor increases.

At higher gas velocities, the bed material gets entrained and a circulation of the bed material is required. This type of gasifiers is called Circulating Fluidized

Bed (CFB) gasifiers. Typical velocity in the gasifier is normally between 3 and 10 m s^{-1}. The entrained bed material and the, not completely, converted fuel particles (char) are removed from the produced gas by a cyclone or another separation device. The particles are normally returned to the bottom of the gasifier via a non-mechanical valve. This ‘non-mechanical’ valve can be a stand pipe which also serves the function of preventing gas leakage from the bottom of the riser into the solids outlet of the cyclone. Foster Wheeler has successfully demonstrated this type of gasifier on a commercial scale in Lahti in Finland and Ruien in Belgium.

Separating the gasification of the biomass and the combustion of the remaining char leads to the Indirect or Allothermal gasification process as shown in the right part of figure 2-2. The biomass fed to the ‘gasifier’ is converted into a gas and char (pyrolysis). The heat required for the heating of the biomass comes from the combustion reactor. This heat is transported by the circulating bed material. Char and bed material are separated from the gas by a solid gas separation device (e.g. a cyclone). The produced gas exits the gasifier to the gas cleaning. The char and bed material are fed to the combustion reactor. The char is combusted to produce the required heat for the gasification reactor. The heated bed material is returned to the gasifier reactor again. Indirect gasification will be explained in more detail in paragraph 2.5.

2.3 Tar

All biomass gasifiers which produce a gas containing methane (e.g. Fluidized Bed gasifiers) produce tar as well [6]. The syngas from gasifiers operating above ≈1200°C, like Entrained Flow gasifiers, contains almost no methane and tar.

Tar is a complex mixture of (poly-aromatic) hydrocarbons which varies in amount and composition. Tar consists largely of aromatic compounds [7]. The general definition is "hydrocarbons with molecular weight higher than benzene".

The tar properties are influenced by gasifier operating conditions as temperature, residence time, etc. and the presence of a catalyst like olivine or dolomite. Tars can cause fouling of downstream equipment and produce polluted condensation water. Therefore, the type and concentration of tars in producer gas are major issues in operating biomass gasification plants. Figure 2-3 shows an example of tar related problems in downstream equipment. The picture to the left shows a demister behind a scrubber fouled with tar and dust. The right one shows water from a wet scrubber fouled with tar and dust.

Figure 2-3: Examples of tar fouling problems

Several classification systems for tars are in use. Evans and Milne defined tars based on the formation conditions [7]. Primary tars are formed by decomposition of the building blocks of biomass and contain oxygen in significant amounts. Secondary and tertiary tars are formed by destruction of primary tar compounds and recombination of fragments.

The amount or concentration of tars is often not the most important parameter in running a biomass gasification plant. The type of tars in combination with the concentration is of higher relevance. ECN has set up a tar classification system based on the physical tar properties like water solubility and dew point of the tar components. Table 2.1 gives a description of the five tar classes in the classification system with the focus on the tar properties.

Table 2.1: Description of the tar classes

Class	Description
1	GC undetectable tars. This class includes the heaviest tars that condense at high temperature even at very low concentrations.
2	Heterocyclic components (like phenol, pyridine, cresol). These are components that generally exhibit high water solubility, due to their polarity.
3	Aromatic components. Light hydrocarbons that are not important in condensation and water solubility issues.
4	Light poly-aromatic hydrocarbons (2-3 rings PAH's). These components condense at relatively high concentrations and intermediate temperatures.
5	Heavy poly-aromatic hydrocarbons (4-5 rings PAH's). These components condense at relatively high temperature at low concentrations.

Class 1 and class 5 tars can condense in the producer gas cooler that normally is located directly after the gasifier. Condensation of heavy tars can be prevented by keeping the temperature of the cooling wall high, but this limits the final cooling temperature. Further cooling can be accomplished by systems that can handle condensed tars, like the OLGA gas tar removal system [8]. If some condensation of tars occurs, the walls and ducts can be cleaned by using the larger entrained particles (>20 μm) to sand blast the wall [9]. These principles were used for the design of the MILENA pilot plant gas cooler. An optimized cooler applying these principles is under development at ECN [10], with the goal of making the producer gas cooler less sensible to fouling by tar.

The concentrations of class 1 and 5 tars can be reduced strongly in a Fluidized Bed gasifier by using a catalytically active bed material like olivine or dolomite. The concentration of class 2 tars is of importance if the water from the producer gas is removed by condensation. In that case the waste water will contain most of the class 2 tars and has to be cleaned. The concentration of class 2 tars increases with decreasing gasification temperature. Updraft gasifiers produce relatively high amounts of class 2 tars, whereas the concentration of class 2 tars in the gas from a Fluidized Bed gasifier is lower but strongly temperature dependent. In the case of Fluidized Bed gasifiers the concentration of class 2 tars can be significantly reduced by using a catalytically active bed material.

Reported measured tar concentrations are hard to compare for different installations because the tar measurement method is often not clear. To solve this problem a standard tar measurement method (the Tar Protocol) has been developed. A draft version of this tar measurement standard and background information can be found at the website www.tarweb.net. ECN uses the Solid Phase Adsorption (SPA) method to measure the tar concentration behind Fluidized Bed gasifiers like in the case of the MILENA gasifier. The results using this method agree with the Tar Protocol for compounds from phenol to pyrene [11]. The concentrations of heavier tar molecules are relatively low if the gasification temperature in a Fluidized Bed gasifier is above 800°C [12]. The SPA method was selected because the sampling of the tars is relatively simple.

The tar dew point is more relevant than the tar concentration. The tar dew point is the highest temperature of the gas at which condensation of tar components occurs. The tar dew point can be calculated from the gas composition or directly be measured. The ECN website www.thersites.nl provides a useful tool to calculate the tar dew point. Direct measurement of the tar dew point is possible with devices like the tar dew point analyzer [13].

The tar concentration and the tar dew point can be reduced in a Fluidized Bed gasifier by using a catalytically active bed material like olivine and dolomite as the most common catalytic bed materials. Especially olivine has become of interest because of the success of the FICFB gasifier in Güssing where it is used as the standard bed material. The reduction of the tar concentration in the gasifier is in literature described as a “primary measure”. A “secondary measure” is defined as a measure taken downstream the gasifier like thermal cracking, catalytic cracking or scrubbing.

Thermal cracking reduces the cold gas efficiency because the gas needs to be heated up to above 1200°C. Normally air or oxygen is added to the gas to increase the temperature by combustion of part of the produced gas. Under these conditions also methane is broken down, which has a negative impact on the calorific value of the gas. Thermal tar cracking is often applied in combination with a chemical quench. In this case the latent heat in the gas is used to gasify the char which remains after pyrolysis. A classical Downdraft gasifier is a good example of the combination of thermal tar cracking and a chemical quench using char. Another example is the Carbo-V process [14].

Much research is done on catalytic tar cracking directly downstream the gasifier. Catalytic tar cracking has the advantage that the temperature of the gas does not need to be increased too much which has a positive influence on the Cold Gas Efficiency (CGE). The catalysts that are used, mostly nickel based, are sensitive to the pollutants in producer gas (e.g. sulphur and dust). Several projects were done to demonstrate that it is possible to operate a catalytic tar cracker on raw producer gas [15], but so far catalytic tar crackers were not successful in commercial operation. Deactivation of the (expensive) catalyst is still a major problem.

Catalytic tar cracking can be an interesting option if a cheap catalyst can be applied, because replacement of the deactivated catalyst becomes less expensive. Char from biomass is such a catalyst. Several tests at ECN and other institutes showed that under the right process conditions char can reduce the tar concentration to some extent. Experiments at ECN showed that a concentration of fine char particles of 1500 mg Nm^{-3}, a gas residence time of 1.5 to 3.5 s and a temperature above 800°C were sufficient to reduce the concentration of heavy tars by 80 - 90% [16]. The settling chamber in the MILENA gasifier, at that time called the STAR gasifier, was originally intended to create a zone with a high char concentration. Hydrodynamic testing showed that it was not possible to reach the required char concentration in the MILENA settling chamber. A new design of a stand alone reactor was made to achieve the required high char concentration and sufficient contact time. This reactor was called the TREC (Tar REduction by Char) reactor [17] and was constructed and tested in the EU project “Green Fuel Cell”

[18]. The TREC reactor is a kind of granular bed type. The TREC reactor removes fly ash and char from producer gas which flows in radial direction through the bed. The char that is collected between granules acts as catalyst for tar cracking. The effectiveness of TREC can be enhanced by a catalytically active loading. The TREC reactor can reduce the tar dew point from 350°C down to 170°C, but this is not sufficient for most applications (e.g. gas engines). The TREC reactor was also tested with a (inexpensive) catalytically active bed material (olivine), that resulted in a tar dew point below 100°. The TREC reactor is possibly an attractive option for tar removal in the MILENA SNG system. The TREC development will probably be continued in the future.

ECN has selected tar removal technology as a secondary measure for further tar reduction. Several wet tar removal systems were developed and tested [6, 19]. The Updraft gasifier in Harboøre successfully applies a wet electrostatic precipitator (ESP). Because of the positive experiences in Harboøre, tests have been done at ECN to check whether such a system was also applicable in combination with Fluidized Bed gasifiers. The system using a wet ESP was able to reduce the tar dew point to a sufficiently low level such that the gas can be combusted in a gas engine. The tar (and dust) ended up in the water system and the cleaning of the water appeared to be problematic and expensive [6]. ECN decided to switch to an oil based tar removal system named OLGA. The OLGA tar removal system is operated at a temperature above the water dew point to prevent the mixing of water and tar. The tar can be removed from the producer gas down to a tar dew point temperature below 0°C. Water soluble components (class 2 tars) are almost completely removed. Water is condensed out of the gas downstream OLGA. Water is condensed from the gas at a temperature above the tar dew point, such that condensation of tars in the water is prevented. A small amount of benzene in the gas will dissolve in the condensed water. The benzene can be removed from the water by active carbon before disposure.

The tar concentration and the tar dew point in the gas produced in the MILENA gasifier are reduced by a combination of primary and secondary measures. Olivine is often used for the first reduction of the tar dew point. This already simplifies the gas cooling. The tar dew point is further reduced by removal of the tars in the OLGA gas cleaning system. The removed tars can be recycled to the gasifier, preferably to the combustion zone.

2.4 Agglomeration

One of the major operational problems in Fluidized Bed combustors and gasifiers is agglomeration of the bed material. Bed agglomeration can result in de-fluidization of the bed which normally leads to local temperature deviations. This can result in local melting and will finally lead to a complete shutdown of the installation. Agglomeration is caused by melting of the inorganic components in the fuel. Especially biomass fuels contain inorganic components which can cause bed agglomeration. The most well known inorganic component to cause agglomeration is potassium (K). Potassium and silicon can form a low melting potassium silicate eutectic. Silicon is normally present in large quantities from the (silica) sand often used as bed material and/or sand present in the biomass.

Two different types of agglomeration were identified during gasification and combustion tests in the ECN Fluidized Bed facilities [20]. Figure 2-4 shows the basic difference between type I and type II agglomeration.

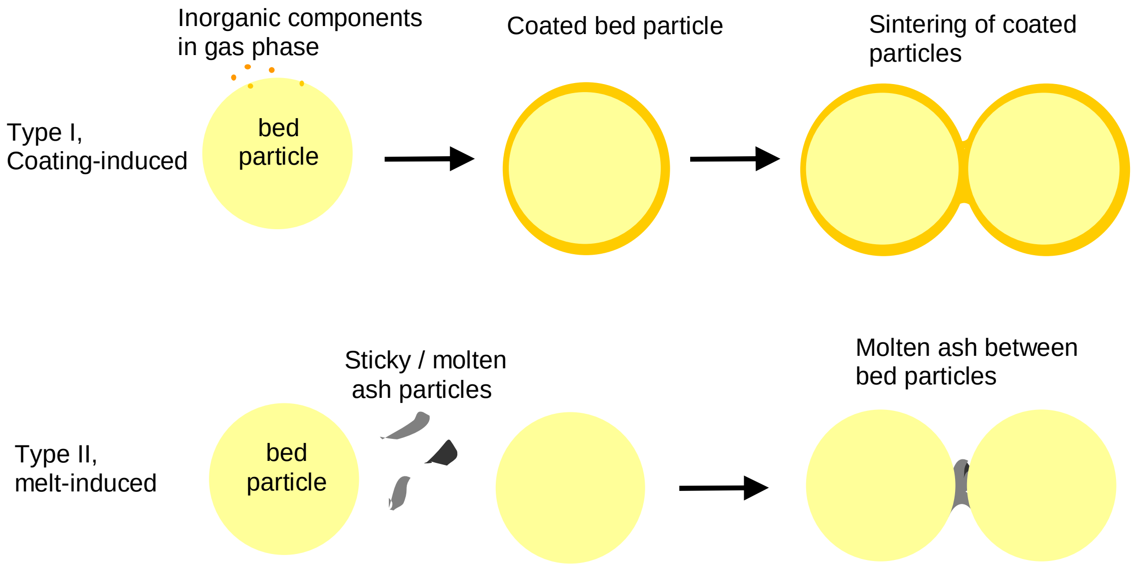

Figure 2-4: Basic principles for type I and type II bed agglomeration

Coating induced agglomeration is a result of the interaction between inorganic components in the gas phase (e.g. potassium) and the bed material (e.g. silicon). The coating formed can be sticky and cause agglomeration of the particles. The typical coating layer thickness is between 2 and 20 µm. The tendency to agglomerate increases with increased coating layer thickness. Type I agglomeration can be suppressed by preventing the formation of a ‘thick’ coating. This can be realized by replacing part of the bed material during operation. Replacing the bed material with a more ‘inert‘ bed material is not always an option, because many biomass (waste) streams contain large quantities of silica sand.

Melt induced agglomeration is observed when a fuel is gasified with a low ash melt temperature, like grass, or when unstable operation of the gasifier has led to temperature excursions. Preventing melt induced agglomeration is possible by keeping the operating temperature of the fluidized bed well below the ash melting point and by preventing local hot spots. Local hot spots are best prevented by stable operation and a high bed material to char/fuel ratio.

2.5 Indirect gasification

The MILENA gasifier is an Indirect or Allothermal gasifier. The conversion of the fuel is being done in two separate reactors. For this reason this type of reactor is sometimes called a Twin Bed gasifier. The first reactor is called the gasifier (left reactor in Figure 2-2) and the second reactor is called the combustor. The processes in the gasifier are endothermic and the processes in the combustor are exothermic. In the ‘gasifier’ reactor the biomass is pyrolysed or degasified by hot bed material coming from the second reactor. The typical gasification temperature is 850°C. The gasifier is normally fluidized by steam and the gas produced by the gasification process. The producer gas and the solids are separated, after which the producer gas is led to a gas cooler. The solids (bed material and char) are returned to the combustor reactor. The char is combusted to heat up the bed material up to a temperature of typically 930°C. The heated bed material is sent back to the gasifier.

The main advantages of Indirect gasification over Direct gasification processes like BFB and CFB gasification is the higher heating value of the produced gas and the complete conversion of the fuel. The heating value of the gas produced in an Indirect gasifier is higher than the heating value of the gas produced from an air blown Direct gasifier, because the air used in a Direct gasifier dilutes the producer gas with N_2 and CO_2. Indirect gasification is a high temperature pyrolysis process, so no air or oxygen is required. The nitrogen content in the producer gas from an Indirect gasifier can be kept below 5 vol.%. The small amount of nitrogen in the gas originates from the nitrogen purge for the feeding screw and a small amount of air/flue gas in-leak from the combustor into the gasifier. The nitrogen content can be lowered by using CO_2 as a purge gas or by minimizing the leakage between combustor and gasifier.

The conversion of biomass in a Direct gasifier like a BFB or CFB gasifier is not complete, because the gasification of the char that remains after the devolatization of the biomass is a slow process at the typical operating temperature.

The residence time in a BFB or CFB is far from sufficient to gasify all the char [21]. An acceptable conversion can only be achieved by subsequent combustion of the char as it is done in an Indirect gasifier so that the total process conversion is complete.

Well known examples of Indirect gasifiers are the FICFB gasifier developed by the Vienna University of Technology [22] and the SilvaGas process developed at Battelle’s Columbus Laboratories [23]. The FICFB process was successfully demonstrated in Güssing (Austria) on an 8 MW_{th} scale [24]. A second commercial FICFB gasifier was built in Oberwart (Austria) and several others are under construction. The FICFB gasification technology was also selected for the Bio-SNG demonstration project in Gothenburg called GoBiGas. The SilvaGas process was demonstrated in Vermont (US) [23], but this demonstration was cancelled after a relatively short period. Unfortunately the process data are insufficiently documented. The SilvaGas process is continued by Biomass Gas & Electric. Several large commercial projects are under construction in the U.S. The ECN OLGA tar removal technology, delivered by Dahlman will be used for gas cleaning in these initiatives.

2.6 MILENA gasification process

The Energy research Center of the Netherlands developed CFB gasification technology for approximately 12 years [21]. The experience gained with modifying and operating a 500 kW_{th} pilot plant led to the development of the Indirect MILENA gasifier. The gasifier contains separate sections for gasification and combustion. The gasification section consists of three parts: gasifier riser, settling chamber and downcomer. The combustion section consists of only one part. The red arrows in Figure 2-5 represent the circulating bed material. The processes in the gasification section will be explained first.

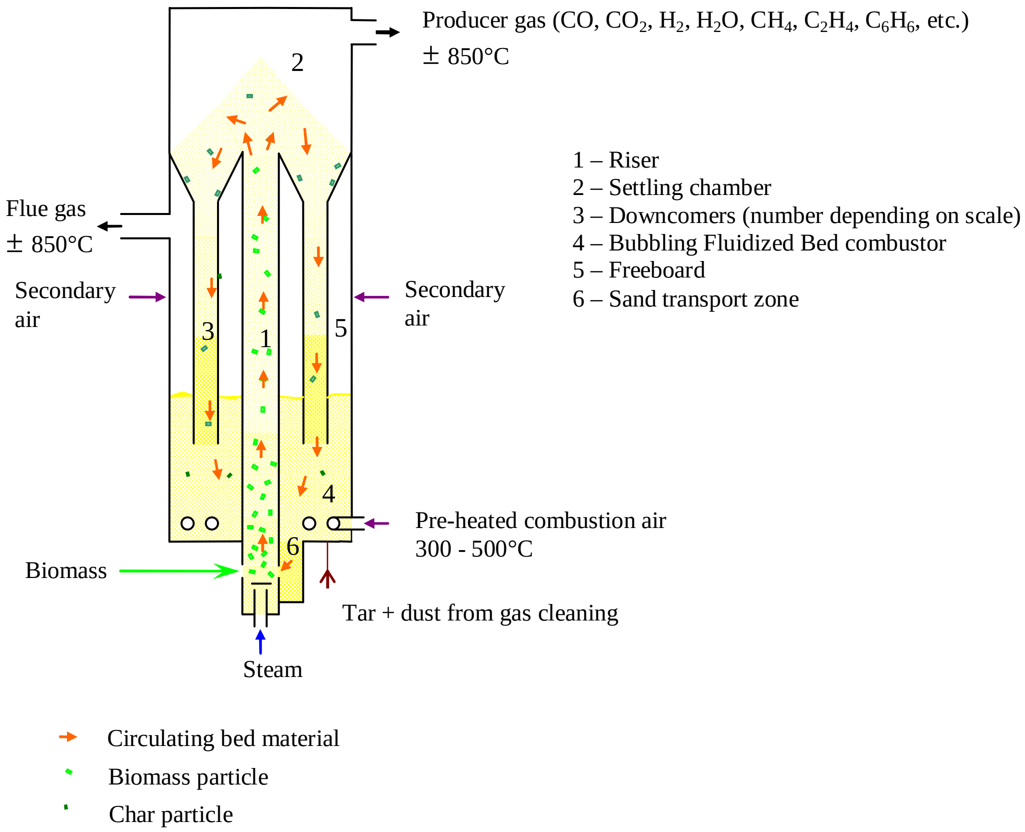

Figure 2-5: Simplified scheme of MILENA gasification process.

Biomass (e.g. wood) is fed into the gasifier riser. A small amount of superheated steam is added from below to create a linear gas velocity of approximately 0.5 m s^{-1} in the bottom part of the riser gasifier. Steam can be replaced by air if nitrogen dilution of the producer gas is not a problem (e.g. if the gas is fired in a gas engine). Hot bed material (typically 925°C sand or olivine of 0.2 – 0.3 mm) enters the gasifier riser from the combustor through a hole in the gasifier riser (opposite of the biomass feeding point). The typical bed material circulation rate on a mass basis is 40 times the amount of biomass fed to the gasifier riser. The bed material heats the biomass to 850°C in the gasification section. The heated biomass particles degasify and are partially converted into gas. The volume created by the gas from the biomass results in a vertical velocity increase over the length of the gasifier riser to approximately 6 m s^{-1}. It will ultimately create a “turbulent fluidization” regime in the gasifier riser and carry-over of the bed material together with the degasified biomass particles (char). The vertical velocity of the gas is reduced in the settling chamber, causing the larger solids (bed material and char) to separate from the gas and fall down into the downcomer. The producer gas leaves the reactor from the top and is sent to the cooling and gas cleaning section. The typical residence time of the gas is several seconds.

The combustor operates as a Bubbling Fluidized Bed (BFB). The downcomers transport bed material and char from the gasification section into the combustor. Tar and dust, separated from the producer gas, can also be fed to the combustor. Char, tar and dust are burned with air to heat the bed material to approximately 925°C. Secondary air is added in the freeboard to reduce CO and C_xH_y emissions. Flue gas leaves the reactor to be cooled, de-dusted and emitted. The heated bed material leaves the bottom of the combustor through a hole into the gasifier riser. No additional heat input is required since all heat for the gasification process is produced by the combustion of the char, tar and dust in the combustor.

The mechanical design of the MILENA reactor is patented [25]. The reactor vessel is a conventional carbon steel vessel with a refractory wall lining to reduce heat loss and keep the temperature of the carbon steel wall low. The insert (gasifier riser, downcomers and settling chamber) is made of high temperature steel like 310 Stainless Steel.

The main difference between the MILENA and the FICFB (Güssing) are the reversed roles of the BFB and the riser. The FICFB process applies a BFB as the gasifier and a riser as the combustor. The MILENA process applies a riser as gasifier and a BFB as combustor. The advantage of using a riser is that the area that needs to be fluidized is smaller. Therefore the amount of fluidization gas (steam) is smaller. All the fluidization gas needs to be heated to the gasification temperature, which has a negative effect on the Cold Gas Efficiency (see paragraph 2.7 for a detailed explanation). On the other hand process conditions in the steam blown BFB gasifier are optimum for primary tar reduction, because an excess of steam is available for tar reforming and the contact with (catalytic) bed material is better than in a riser. Tests at ECN have shown that the tar dew point can be around 100°C when biomass is gasified in a steam blown BFB using (Austrian) Olivine as a bed material. This agrees with reported results from the Güssing gasifier.

The SilvaGas or Battelle gasifier is more similar to the MILENA process. Cold Gas efficiency and gas compositions are similar when both processes are operated under similar process conditions. One major difference is the use of a settling chamber in the MILENA process instead of a cyclone to separate the char and bed material from the producer gas. The settling chamber was selected to create a zone with a high gas residence time in an environment with a lot of dust (char/fine bed material), because these conditions are advantageous for tar reduction. The settling chamber makes an integrated design of gasifier riser, solids removal (the settling chamber) and combustor possible and more logical. By placing all the key components in one vessel, pressurized operation becomes easier. The SilvaGas applies two riser reactors, one for gasification of the biomass and one for combustion of the char. The MILENA process uses a BFB for the char combustion. The BFB was selected because the bed material/char ratio is higher in a BFB than in a riser. Char particles are surrounded by more sand particles during the combustion process. The bed material acts a heat carrier, and cools the burning char particle which prevents local hot spots. Local hot spots are a cause for agglomeration (type II, melt-induced). The relatively high bed material/char ratio is expected to help preventing agglomeration problems.

2.7 Gasifier Efficiency

The efficiency of a gasifier is generally expressed as Cold Gas Efficiency (CGE). CGE is defined as the chemical energy of the gas at room temperature divided by the chemical energy of the biomass input. This definition leaves room for a several different interpretations. The calorific value of biomass and the produced gas can be defined on Lower Heating Value (LHV) basis or on Higher Heating Value (HHV) basis. The LHV of a fuel excludes the condensation heat of the water in the flue gas after combustion, the HHV includes the heat of condensation. When calculating the CGE both the heating value of the biomass and the gas should be calculated on the same basis. The chemical energy in the cleaned gas is normally lower than the chemical energy in the raw gas leaving the gasifier, due to tars and NH_3 in the untreated gas. Both can contribute significantly to the heating value of the gas. When comparing CGE’s it should be made clear whether the heating value of the cleaned gas or the raw gas is used. The CGE reported in this thesis is always defined using the heating value of the cleaned gas. Both LHV and HHV are given, to make the given efficiencies comparable with other publications.

The calculation of efficiencies based on LHV can give remarkable results, because the heating value of a solid fuel is corrected for the heat required to evaporate the water from the fuel (Equation 2.1).

$$LHV_{a.r.} = LHV_{dry} \cdot (1-w/100) - 2.442 \cdot w/100 \qquad (2.1)$$

Where: w = mass% of moisture in the biomass on as received (a.r.) basis and the heating value is expressed in MJ kg^{-1}.

This definition results in negative Lower Heating Values (LHV) if the biomass is wet enough. Figure 2-6 shows the calculated heating values of dry wood with different moisture contents and the calculated CGE’s on LHV and HHV basis for a MILENA type gasifier with integrated dryer. The biomass is dried to 15 mass% of moisture. The drying is not limited by the availability of heat produced by the gasifier system As can be seen from the figure the lower heating value of wood becomes negative above 88 mass% of moisture.

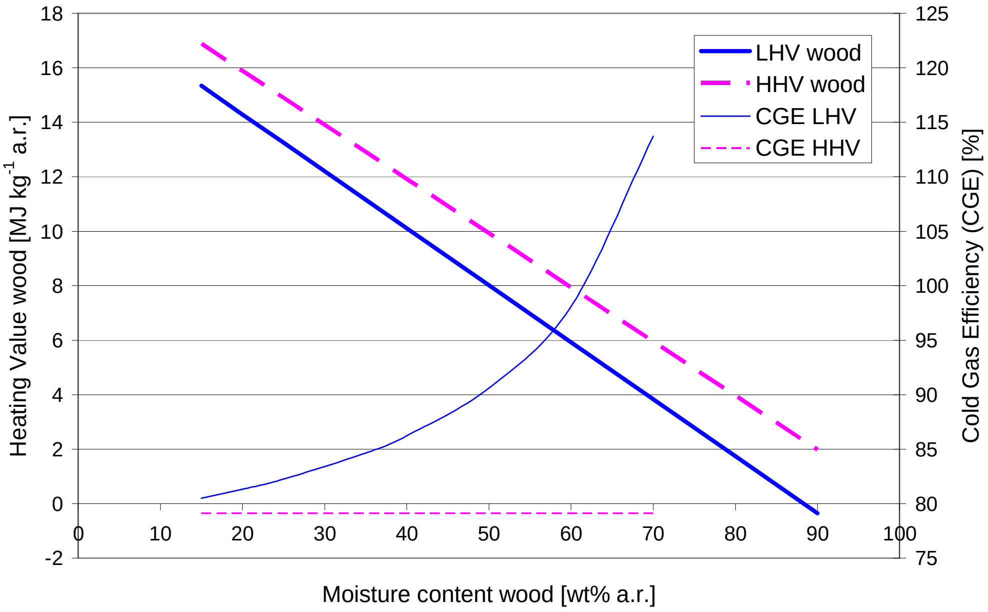

Figure 2-6: Heating value wood and calculated CGE as function of moisture content fuel.

When relatively wet biomass is gasified and waste heat or non evaporative drying is applied to dry the biomass, the CGE (on LHV basis) can be increased significantly to values eventually higher than 100%. Obviously, when comparing efficiencies for biomass installations on LHV basis the moisture content of the fuel is an important factor.

The CGE of a gasification system is determined by the losses. These losses are latent heat of the produced gases minus the heat of the fluidization gas, heat loss, tar loss and char loss. The heat loss of commercially sized gasifiers (>10 MW_{th} input) is normally below 1% if no active wall cooling is applied. Most Fluidized Bed gasifiers do not use reactor wall cooling, but cooling is common for high temperature Entrained Flow (EF) gasifiers. Tar is normally recycled to the gasifier, which reduces the loss. The latent heat of the producer gas is influenced by the amount of gasification or fluidization gas fed to the gasifier. The char loss is determined by the fuel or carbon conversion.

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Chapter 3

Bio-SNG

Abstract

The application and production options of Bio-SNG are described in this chapter. Bio-SNG can directly replace fossil natural gas, because the composition is similar. The application of natural gas as transport fuel has been increasing rapidly in recent years. This creates the possibility to apply Bio-SNG in the transport fuel market as well. It is argued in this chapter that Bio-SNG is as more attractive transport fuel than alternatives (like FT diesel) that are under development at the moment.

3.1 Application of (Bio)-SNG

Natural gas is a convenient and environmentally friendly fuel used all over the world for a wide range of applications. The most relevant applications are:

- Large scale Electricity production in Combined Cycles.
- Decentralized Combined Heat and Power production (CHP).
- Transportation, 10 million cars use Compressed Natural Gas (CNG).
- Chemical industry, as a feedstock for many chemicals.

SNG can directly replace fossil natural gas in al these applications if the composition of the produced SNG complies with the specifications put forward. Limits are normally set for Heating Value, Wobbe index, concentrations of impurities and condensables. Exact specifications and certification for gas grid injection of Bio-SNG are still topic of discussion. For some countries specifications are available for the injection of upgraded biogas [1].

SNG produced via gasification mainly consists of CH_4, together with some CO_2, N_2 and H_2. The H_2 concentration has a strong influence on the calculated Wobbe index. CO_2 and N_2 lower the heating value and Wobbe index whereas $CO_{2,}$ together with water can also condense in the high pressure gas grid.

The composition of natural gas varies. The main component of natural gas is always CH_4. Some alkanes (mostly C_2H_6 and C_3H_8) are also present in the gas. Because of the presence of the alkanes the heating value of the gas mixture can be higher than the heating value of pure CH_4. SNG does not contain alkanes and therefore it can be difficult to meet the specifications for Wobbe index and Heating Value. A possible solution is mixing in Liquid Propane Gas (LPG). LPG is a relatively cheap and widely available fuel.

Table 7.11 (Chapter 7) gives the expected gas Bio-SNG for the MILENA SNG system.

3.2 Bio-SNG Production routes

There are two main options to produce SNG from biomass:

1. Anaerobic digestion (biological conversion at low temperature).
2. Gasification (thermo chemical conversion at high temperature).

Anaerobic digestion is a process carried out by bacteria. The bacteria grow by converting organic matter into biogas (mainly CH_4 and CO_2). Biogas production is a proven technology. In 2007 more than 3500 anaerobic digesters were in operation in Germany [2].

Most of the present biogas production comes from landfills and waste water treatment plants. The biogas production from landfills is in decline, due to the ban on depositing organic material, whereas the number of dedicated co-digestion plants using manure and food wastes is increasing. Biogas can be used in a gas engine after relatively simple cleaning of the gas. After CO_2 removal, gas cleaning and compression the gas can be injected in the grid. The relatively small scale of anaerobic digestion facilities makes upgrading of the gas expensive. In 2007 three anaerobic digestion plants were in operation in Sweden which injected the produced SNG in the gas grid [3].

Gasification of biomass is less limited by biomass supply as in the case of digestion, because a wider range of biomass fuels are suitable to be used as feedstock and the amounts available of these feedstocks are larger.

The estimated potential for biogas in the Netherlands is estimated to be 50-60 PJ or 2 to 3% replacement of fossil natural gas [4]. Additional production of Bio-SNG via gasification is required to replace a significant amount of present day use of natural gas. Anaerobic digestion can speed-up the introduction of Bio-SNG. Figure 3-1 depicts the Bio-SNG implementation in time. Both contributions of anaerobic digestion and gasification are foreseen [5].

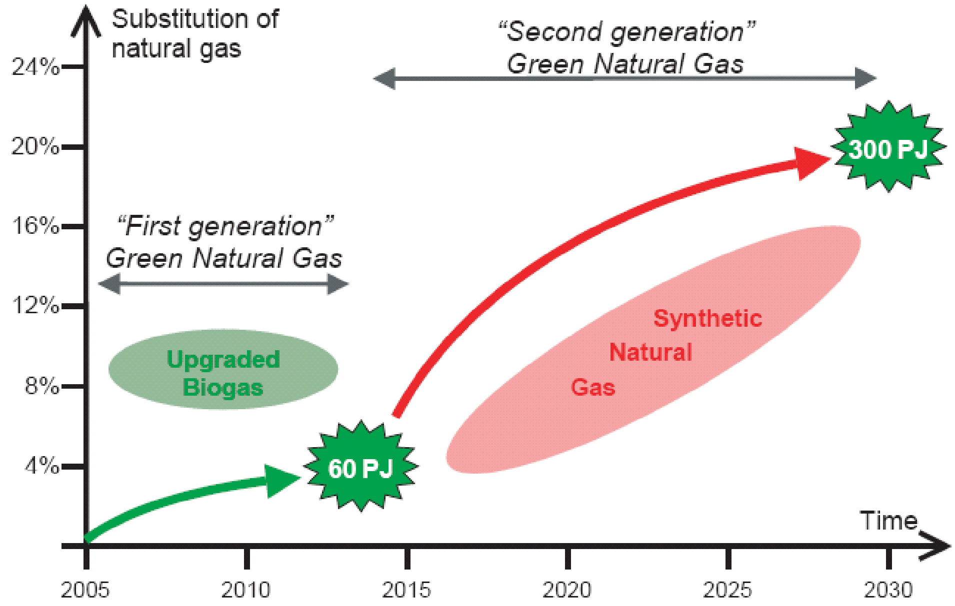

Figure 3-1: Foreseen implementation trajectory of Bio-SNG.

3.3 Historical Background

Coal gas can be seen as the predecessor of SNG. Gas produced from coal was produced in London for public street lighting in the beginning of the 19th century. Main combustible components were CO, H_2 and hydrocarbons. The main problems with coal gas were: its toxicity (CO) and the cleaning of the gas (tar). Figure 3-2 shows a picture of a coal gas production and storage facility.

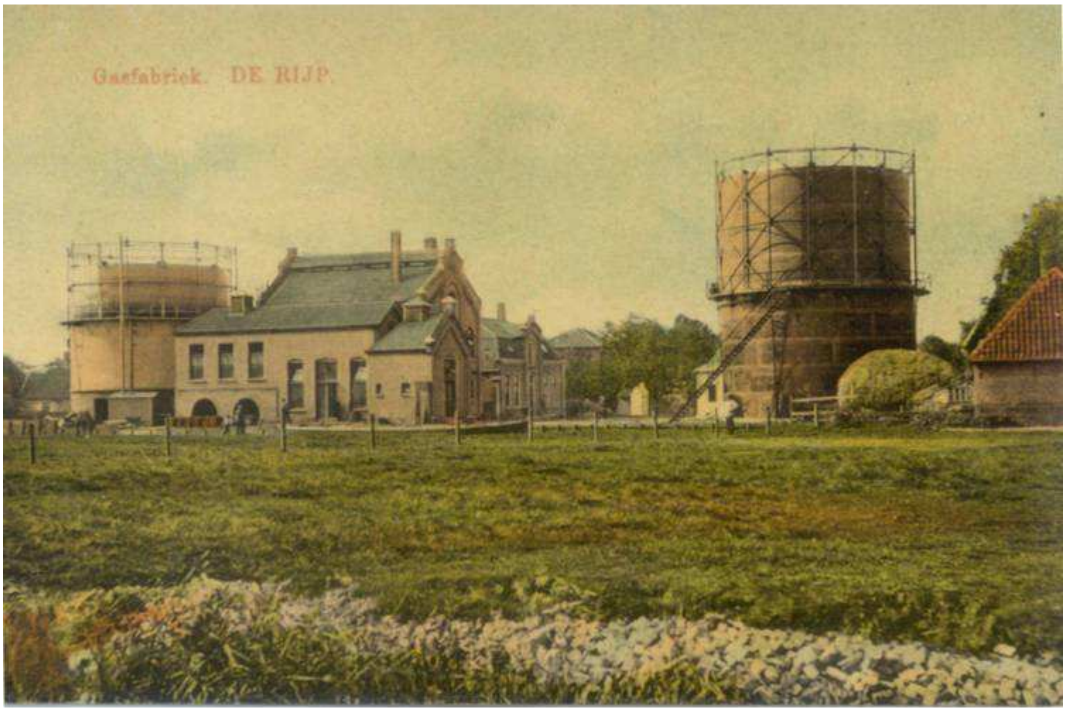

Figure 3-2: Coal gas production facility in the Netherlands in 1911.

The use of coal came to an end when relatively cheap and safe natural gas became available.

Due to the oil crisis of the seventies, governments started to realize that dependence of (foreign) oil and in a lesser extent natural gas was an unwanted situation. Projects were started to develop alternatives. One of those alternatives was the production of SNG from lignite coal. In the United States several large scale demonstrations were realized. Most of these demonstrations were not successful. One important reason for the failure of many projects was probably the drop in oil prices, which made these facilities uneconomic. The Great Plains

Synfuels plant in North Dakota survived the period of low oil prices and is still producing SNG from lignite coal [6]. The plant has a scale of 3 GW_{th} (lignite) input and produces, besides SNG, electricity and many valuable chemicals like ammonium sulfate, phenol, benzene and toluene. The CO_2 that is removed during the SNG production process is used for Enhanced Oil Recovery (EOR). This decreases the CO_2 emissions from the plant and gives financial benefits. Figure 3-3 shows the Great Plains Synfuels plant in North Dakota.

Figure 3-3: Pictures of Great Plains Synfuels plant.

The Great Plains Synfuels plant operates a Lurgi dry bottom fixed bed coal gasification technology with an oxygen/steam mixture as gasification agent. In total 14 of such gasifiers are in use. The tar loaded hot gas is cooled and quenched with water which removes part of the tar. A Rectisol unit is used for further purification of the gas. The tar is separated from the liquid streams and upgraded into saleable products.

The rise in oil and natural gas prices has resulted in a renewed interest in SNG from lignite and resulted into several new projects where lignite again is the feedstock.

Biogas was already produced at the end of the 19th century to supply gas for street lightning. Nowadays biogas is mostly used in gas engines to produce heat and electricity. In many cases heat is not required at the location of the digestion facility and injection of the gas in the (local) gas grid would be more beneficial. Because the gas grid is normally used to transport natural gas the biogas should converted into a similar composition and quality. This can be done by CO_2 and sulphur removal.

In 1994 a demonstration was started in Lille. Biogas produced by the digestion of sewage sludge was upgraded into SNG and used to fuel local buses. In 2007 127 Bio-SNG fueled buses were in use. The “Biogasmax” project is linked to this demonstration and demonstration in other locations. Useful information can be found on the project website (www.biogasmax.eu).

The production of SNG from biomass by gasification has only recently started. In Güssing (Austria) a 1 MW SNG demonstration coupled to the 8 MW_{th} FICFB gasifier was taken into operation at the end of 2008.

3.4 Centralized versus Decentralized production

The production of SNG by gasification is a complex process and therefore expensive. The costs per unit of SNG produced reduce drastically with increasing scale. Figure 3-4 shows the estimated production costs for SNG at different scales and biomass prices [5].

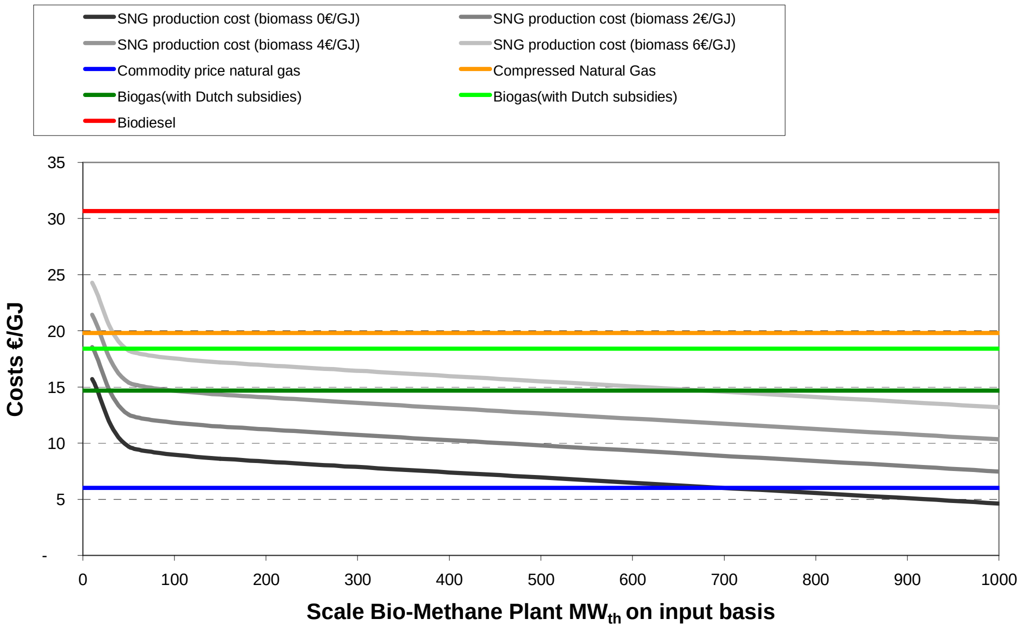

Figure 3-4: Estimated production costs for Bio-SNG.

The available amount of biomass is normally limited in a certain region. To limit transport (thus costs) of biomass the size of a Bio-SNG plant should be restricted. A good size indication is probably set by a paper mill. Paper mills have developed over time to a size that equals several hundreds of MW_{th} of biomass input. A scale between 50 and 500 MW_{th} is probably realistic for local biomass as feedstock. If biomass is imported from overseas a larger scale is beneficial.

In many areas where natural gas is consumed as a primary energy source the amount of available biomass is insufficient to fully replace fossil natural gas by Bio-SNG. This makes import of biomass or Bio-SNG necessary. Transport of the raw material is inefficient as biomass has a low energy density. Several densification processes are available (pelletisation or pyrolysis) or under development (torrefaction in combination with pelletisation) to reduce the transport cost. The disadvantage of these pretreatment processes is the energy consumption required for the densification in addition to investments in local pretreatment plants.

Bio-SNG can be produced where the biomass is available in sufficiently large quantities and can be transported using existing gas grids or as Liquid Natural Gas (LNG) if the production facility is close to a harbor. Figure 3-5 depicts an overview of the existing European gas grid for gas transport. As can be seen from the figure the gas grid is widely spread throughout Europe, with the exception of large parts of Scandinavia.

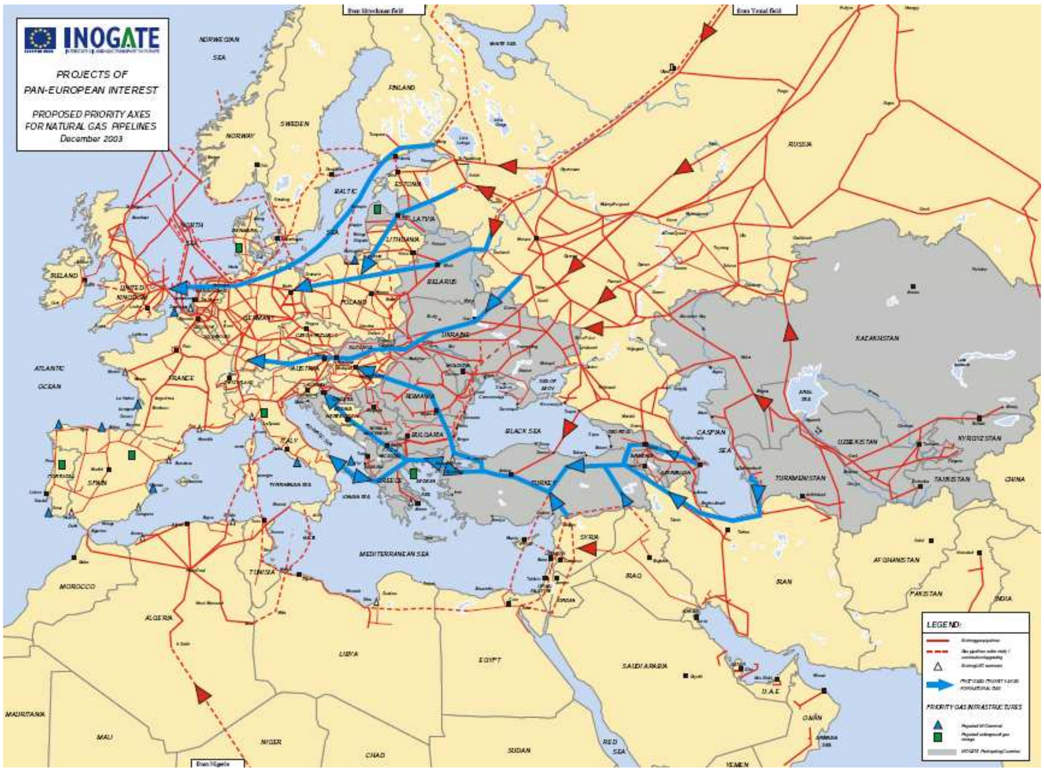

Figure 3-5: Overview of European gas grid.

3.5 Bio-SNG as transport fuel

Compressed Natural Gas (CNG) is becoming more and more important as a transport fuel. In 2010 more than 10 million cars were fueled by CNG according to the International Association for Natural Gas Vehicles (www.iangv.org). This number is rapidly increasing. All major European car suppliers have one or more CNG models commercially available. Cities like Haarlem, Grenoble, Lille and many others use CNG as a fuel for their public transportation. Municipal refuse collection trucks use CNG or LNG in Madrid, Los Angeles and many other cities around the world.

CNG is a relatively clean transport fuel. The emissions of CO_2, particles and sulphur are significantly lower than for other conventional fossil fuels. Noise production can be significantly lower, which is of special importance in urban areas.

Compressing Bio-SNG to 250 bar makes the fuel suitable to be used as Bio-CNG, which is primarily used in passenger cars. Liquefied Natural Gas is often used in heavy trucks and buses, but they can also use CNG. Bio-LNG can be produced from Bio-SNG by cooling it to approximately −163 °C.

Bio-CNG produced by digestion of digestible feedstocks, landfill gas and sewage gas is already demonstrated at several locations. A good example is the Biogasmax project (www.biogasmax.eu). Bio-CNG produced by gasification of non-digestible feedstocks like wood is being demonstrated in Güssing, Austria.

The disadvantage of any gaseous transport fuel is the energy needed for compressing the gas to the required pressure for storage in gas cylinders (approximately 250 bar). The compression of 30 bar SNG to 300 bar CNG consumes approximately 0.014 kW_e kW_{CH4}^{-1}. The overall efficiency of the conversion of biomass to Bio-CNG at 300 bar is 2%-point lower than overall efficiency from biomass to Bio-SNG at 30 bar. The estimated overall efficiency from woody biomass to Bio-SNG at 30 bar is 70% (LHV bases), so overall efficiency to Bio-CNG would be 68%. A popular alternative Bio-Fuel is Fischer Tropsch (FT) Diesel produced out of syngas made by gasification of biomass in high temperature oxygen blown Entrained Flow (EF) gasifiers. An average overall net efficiency from biomass to Fischer Tropsch Diesel is less than 50% [7]. The difference in efficiency for producing Bio-FT Diesel and Bio-CNG is at least 18 %-point. The lower efficiency for Bio-FT is a.o. due to inherent energy losses in the conversion chain of biomass to FT products.

In a recent European study [8] different fuels were compared regarding well-to-wheels energy efficiencies. Part of this comparison is the tank-to-wheels energy use which is relevant for comparing Bio-CNG with Bio-FT diesel. Conclusion of this study was that technical improvements will bring the energetic performance of CNG close to diesel in the near future (2010+) and hybridization is particularly favorable for CNG. Overall “wood”-to-wheels efficiency will be higher for Bio-CNG than for Bio-FT.

An important driver for the development of Biofuels in the European Union is the Bio-Fuel directive 2003/30/EC. In 2010 5.75% of the road transport fuels have to be replaced by Bio-Fuels. Both diesel and gasoline have their own replacement obligation. CNG is taken to the diesel category, so Bio-CNG counts as Bio-fuel for the replacement of CNG and Diesel.

Fossil based gasoline is mostly replaced by ethanol produced from corn or wheat and fossil diesel is mostly replaced by Bio-Diesel produced from rape seed. Both Biofuels are produced from glucose based biomass and are seen as “first generation” Biofuels. Biofuels produced from non-food cellulosic material like wood are seen as “second generation” Biofuels. To promote the development of “second generation” Biofuels the European Commission has proposed to make “second generation” Biofuels accountable for twice the amount of fossil fuel they replace to achieve the European targets [9]. This double counting would give Bio-SNG a significant advantage over “first generation” Biofuels.

3.6 Concluding remarks

The production of gas/SNG from coal and biomass is not new. The availability of cheap fossil natural gas during most of the 20th century prevented the breakthrough of Bio-SNG technology. The urgent need to reduce fossil CO_2 emissions and to replace declining fossil fuels reserves has renewed interest in Bio-SNG technology.

Production of Bio-SNG via anaerobic digestion is an attractive option to convert digestible feedstocks like manure and food wastes, but additional production of Bio-SNG via gasification is required to replace a significant amount of present day use of natural gas.

The production costs for Bio-SNG are strongly influenced by scale. A scale between 50 and 500 MW_{th} is probably a realistic scale if local biomass is used. If biomass is imported an even larger scale is beneficial.

For some countries the, like the Netherlands, the amount of biomass available is insufficient to replace the present natural gas consumption, so that import is required. In Europe an extensive natural gas transport grid is in place. The areas with a high potential for energy crops production are mostly near a gas grid, so import of Bio-SNG instead of (pelletized) wood is an option.

Bio-SNG (Bio-CNG) is an ideal transport fuel. The overall efficiency from wood to fuel is significantly higher than for the alternative Fischer Tropsch Diesel. The number of cars using CNG is growing rapidly. All these cars can be fuelled with Bio-SNG as it becomes available.

3.7 References

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Chapter 4

Selection of optimal gasification route for SNG production

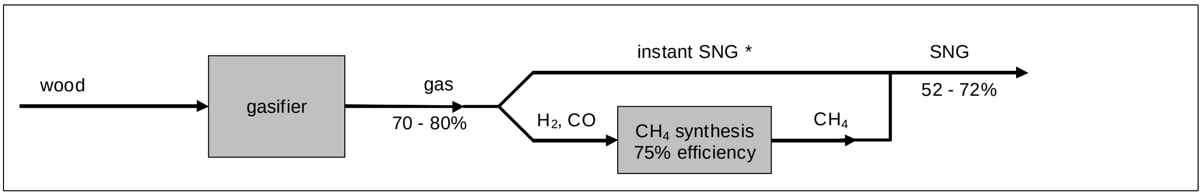
Abstract

The present chapter contains an analysis of the Bio-SNG process efficiency that can be obtained using three different gasification technologies and associated gas cleaning and methanation equipment. These technologies are: 1) Entrained Flow, 2) Circulating Fluidized Bed and 3) Allothermal or Indirect gasification. It was concluded that Indirect gasification results in the highest overall efficiency from wood to Bio-SNG.

4.1 Introduction

Various biomass gasification technologies are suitable to produce a gas which can be upgraded into Bio-SNG. A comparison of these technologies, using similar boundary conditions is required to quantify the assumed higher overall efficiency of Indirect gasification over the alternative biomass gasification processes. This comparison was not available in public literature. This comparison is essential in convincing industrial partners that it is worthwhile to select indirect gasification for Bio-SNG production instead of more conventional gasification processes.

The overall efficiency from wood to Bio-SNG was modeled using three different gasification technologies and associated gas cleaning and methanation equipment. These technologies are: 1) Entrained Flow, 2) Circulating Fluidized Bed and 3) Allothermal or Indirect gasification. The aim of this work was to identify the gasification route with the highest process efficiency from biomass to SNG and to quantify the differences in overall efficiency. This study does not focus on the MILENA gasifier as indirect gasifier, but an "average" Indirect gasifier is assumed. Aspen Plus® was used as modeling tool. The heat and mass balances are based on experimental data from literature and ECN experience. The three gasifier types under consideration all deliver gas suitable for upgrading to SNG, but the high temperature Entrained Flow gasifier (± 1300°C) produces a syngas containing mostly CO, H_2, CO_2 and H_2O while the medium temperature Indirect and CFB gasifiers (±850°C) give a producer gas which, in addition to CO, H_2, CO_2 and H_2O, contains CH_4, unsaturated and aromatic hydrocarbons like C_2H_4 and C_6H_6 and tar. Pollutants like dust, sulphur and chloride need to be removed from the gas in all cases. The medium temperature gasifiers also require tar removal. Figure 4-1 shows the general process efficiencies for wood to SNG. A high amount of 'instant SNG' gives the highest efficiency to SNG, because the loss caused by the exothermic synthesis of CO and H_2 is minimized.



* *mainly CH_4, but also olefins and aromatics*

Figure 4-1: General process efficiencies for wood to SNG.

Syngas, as produced conventionally by oil or coal gasifiers, is seen as a more common feedstock gas than producer gas because gas cleaning processes for syngas applications are already commercially available. The gas cleaning processes for producer gas are still under development.

Both types of cleaned gases can be converted into a gas containing only CH_4, CO_2, H_2O and a small amount of Ar and N_2. CO_2 and H_2O are easily removed from the gas, leaving an SNG that can contain small amounts of N_2 and Ar. The maximum allowable concentration of non-combustible gases is site-specific.

In this study a model is made to calculate the overall efficiencies to SNG for large scale systems of 1 GW_{th} input on HHV basis, based on three different types of gasifiers. The relatively large scale was selected because it matches the commercial scale of Entrained Flow coal gasifiers and the production capacity of a small natural gas field. Oxygen blown Entrained Flow gasification is compared to both Indirect (or Allothermal) gasification and oxygen-blown Circulating Fluidized Bed gasification at the same scale. The comparison is solely based on energetic efficiency numbers. Economics and transport of biomass are not taken into account. The goal of this comparison is to quantify the differences in overall process efficiencies for the three different gasifiers. The results were used by ECN to select to most promising gasification technology for SNG production.

Dry wood (moisture content = 15 wt.%) was selected as the fuel for this study, as it can be readily used for all three different types of gasifiers. Wood mixed with coal was gasified in the Shell Entrained Flow gasifier in Buggenum in the Netherlands and wood is gasified in several Fluidized Bed gasifiers.

The next paragraph describes the system lay-out for each gasification technology. Sub systems are described in detail in separate paragraphs. The final paragraph presents results of the analysis.

4.2 Process configurations

In all cases the gas cleaning and gas upgrading steps are kept similar as much as possible to allow a fair comparison of the overall efficiencies due to various types of gasifiers. Operating pressures were selected on the basis of what is thought to be commercially available in the near future. Figure 4-2 shows the process scheme for the Entrained Flow SNG system.

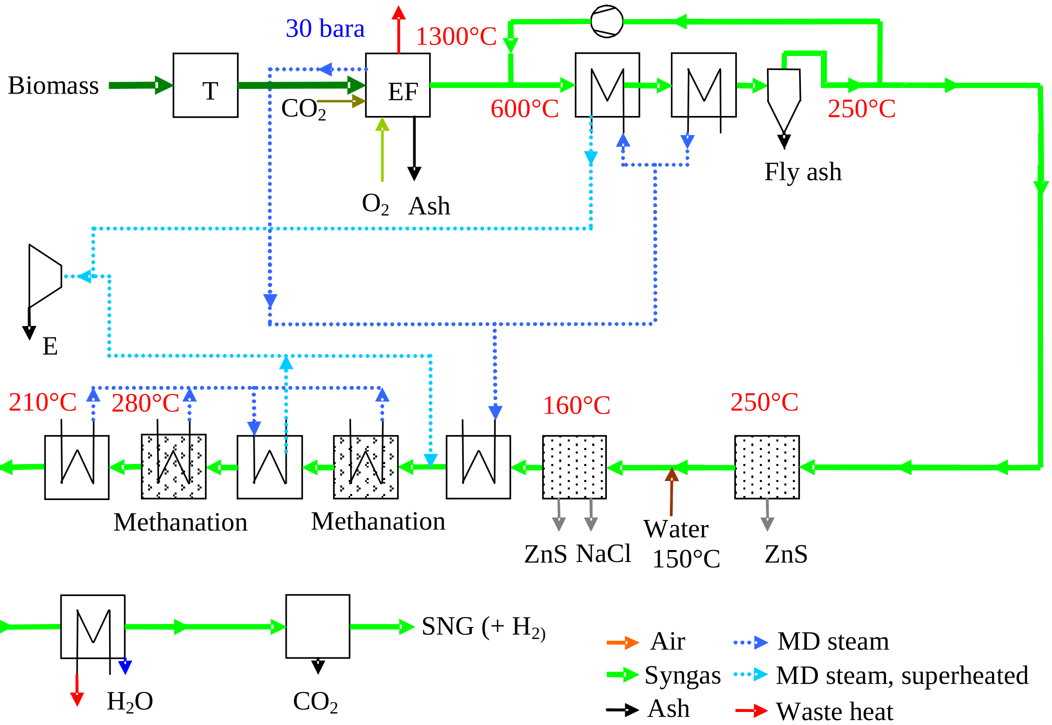


Figure 4-2: Scheme of EF SNG system.

Entrained Flow gasification requires an energy intensive pretreatment to produce fine powder-like feedstock. Torrefaction [1] is selected, followed by milling, because of the relatively low energy requirement. Dried biomass is fed into the torrefaction reactor (T in Figure 4-2). The torrefied biomass is milled, pressurized with CO_2 and pneumatically fed into the Entrained Flow gasifier (EF in Figure 4-2) which operates at 30 bar and 1300°C. Syngas leaving the gasifier is first cooled by a gas quench down to 600°C and subsequently cooled in a heat exchanger producing steam. Fly ashes are removed from the cooled gas by a filter. Part of the gas is recycled to act as quench gas. Sulphur and chloride are removed from the gas by adsorbents. The gas is moisturized and preheated prior to conversion into methane. The heat produced in the methanation section is used for steam generation. Water and carbon dioxide are removed from the gas prior to injection into the natural gas grid.

Figure 4-3 shows the CFB configuration. Wood chips are pressurized by a lock hopper system and fed into the gasifier which operates at 10 bar and 850°C.

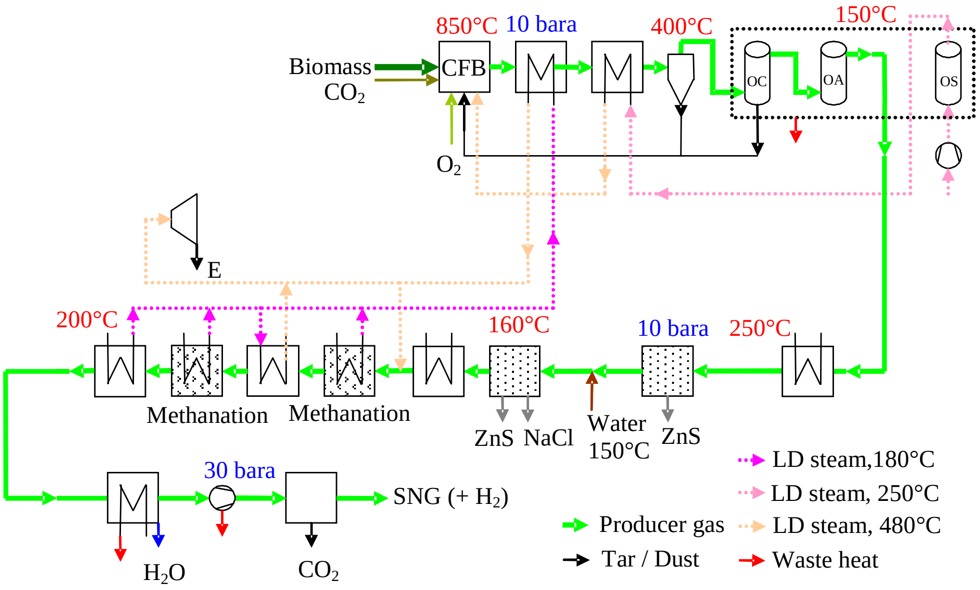


Figure 4-3: Scheme of CFB SNG system.

Producer gas leaving the gasifier is cooled down to 400°C. Most of the dust (carbon containing ash) is removed from the producer gas by a cyclone. The remaining dust and tar are dissolved in oil in the OLGA gas cleaning system. Heavy tars are removed in the collector (OC in Figure 4-3) and light tars are removed in the absorber (OA in Figure 4-3). Heavy tars and a small amount of ash are recycled back to the gasifier. Light tars are removed from the oil with steam in a stripper (OS in Figure 4-3). The steam/light tar mixture is preheated and sent back to the gasifier. Prior to the sulphur and chloride removal by adsorbents the producer gas is preheated to 250°C. The cleaned gas is then preheated, moisturized and converted into methane in several methanation reactors. The heat produced is used for steam generation. Water and CO_2 are removed from the gas before the SNG is compressed to 30 bar and injected into the natural gas grid.

Figure 4-4 shows the system based on the Indirect gasifier. Wood chips are fed into the gasifier (MG in Figure 4-4) which operates at atmospheric pressure and 850°C.

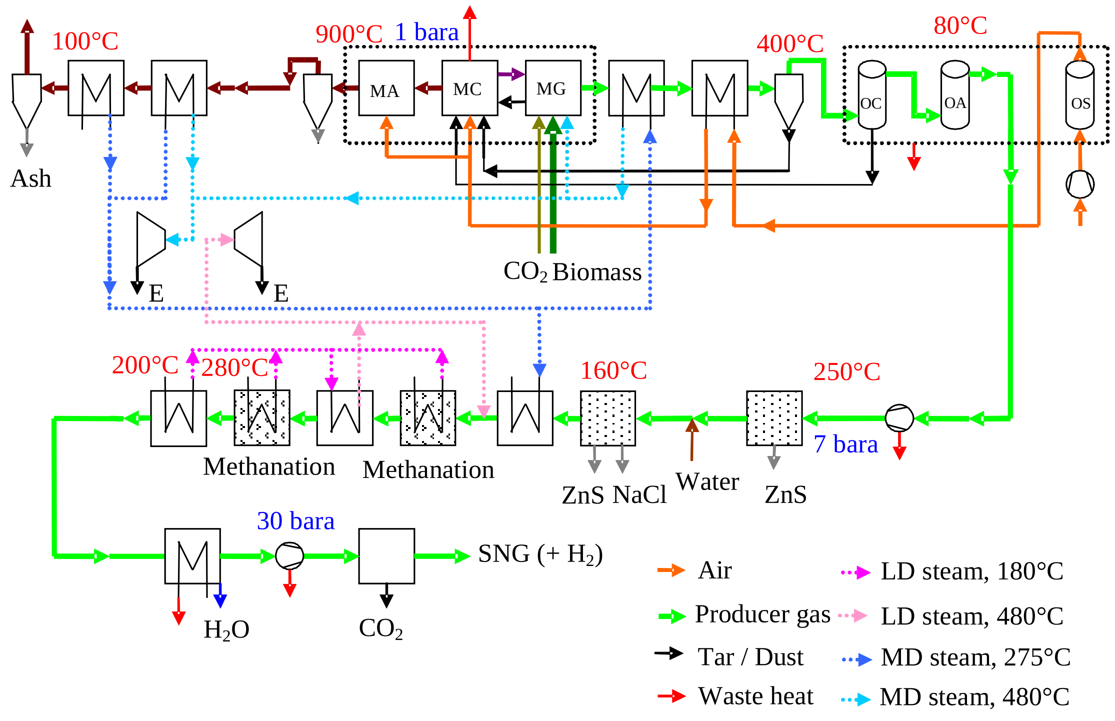


Figure 4-4: Scheme of Indirect gasifier SNG system.

Producer gas leaving the gasifier is cooled down to 400°C. Most of the dust (carbon containing ash) is removed from the producer gas by a cyclone. The remaining dust and tar are dissolved in oil in the OLGA gas cleaning system (OC and OA in Figure 4-4). Heavy tars and a small amount of ash are recycled back to the combustion section of the Indirect gasifier. Light tars are removed from the oil with air in a stripper (OS in Figure 4-4). The air/light tar mixture is preheated and used as fuel enriched combustion air in the combustion section (MC in Figure 4-4) of the Indirect gasifier. For the rest the SNG production steps are the same as in the previous case. Air is added to the flue gas leaving the fluidized bed combustor (MC in Figure 4-4) in the freeboard or afterburner section (MA in Figure 4-4) to reduce CO and C_xH_y emissions. The flue gas is cooled and dedusted by a bag house filter before it is sent to the stack. Heat is used for steam production.

4.2.1 Fuel pretreatment

In case of Entrained Flow gasification a pretreatment step is required to be able to feed the biomass into the gasifier. Torrefaction is seen as the most logical pretreatment step for Entrained Flow gasification [1]. The energy required for milling the fuel is significantly reduced and the fuel contains less H_2O, which improves the Cold Gas Efficiency. Torrefaction also reduces transportation and storage costs for biomass [2]. For the Fluidized Bed gasifiers no such pretreatment is required. Because the torrefaction process requires a relatively dry fuel to operate without additional heat input, wood with 15% moisture was selected as the input fuel for all three systems in this comparison. Integrating a dryer using low temperature waste heat will increase efficiencies (on LHV basis) for all systems, but drying is not considered in this study.

The energetic efficiency of the torrefaction process as estimated from data published by ECN for dry wood [1, 3], was assumed to be 90% on HHV basis and 93% on LHV basis. The electricity consumption of the torrefaction process is assumed to be 1% of the biomass input (on LHV basis). At the moment of writing the ECN torrefaction pilot plant is under construction. More reliable data for overall efficiency and electricity consumption of the process will be available in the future. The moisture content of torrefied biomass varies between 1 and 6% depending on the torrefaction conditions and the post-treatment [3]. For this study a moisture content of 3.5% was assumed. The torrefied biomass particles fed into the gasifier have to be relatively small to be completely converted into syngas. An average particle size of 0.1 mm is assumed. The required milling electricity is 0.017 $MW_e\ MW_{th}^{-1}$ input (LHV) [1].

Table 4.1 gives the composition of clean wood and torrefied wood used in this study. Data have been taken from the Phyllis biomass database (www.ecn.nl/phyllis/). The ash content of wood is estimated at 1%. The composition of the torrefied wood is derived from ECN measurements.

Table 4.1: Composition of wood used in analysis.

	Clean wood	Torrified wood
Moisture [wt.% a.r.]	15.0	3.5
Ash [wt.% d.b.]	1.0	1.1
C [wt.% d.a.f.]	50.7	56.1
H [wt.% d.a.f.]	6.1	5.7
O [wt.% d.a.f.]	42.8	37.9
N [wt.% d.a.f.]	0.3	0.3
S [wt.% d.a.f.]	0.06	0.06
Cl [wt.% d.a.f.]	0.05	0.05
LHV [MJ kg^{-1} d.a.f.]	18.8	20.8
HHV [MJ kg^{-1} d.a.f.]	20.1	22.0

The higher heating value (HHV) of biomass includes the condensation heat of water from the moisture in the biomass and the water formed from hydrogen in the biomass. The lower heating value (LHV) of biomass excludes the condensation heat of the water formed from the hydrogen in the biomass and the moisture. It should be noted that the heating values of wood can vary substantially [4] and have a strong effect on calculated Cold Gas Efficiencies of the gasifiers. The heating values and compositions for clean wood given in Table 4.1 are an average of approximately 200 samples of different types of wood, so the inaccuracy in the heating value is negligible. The number of representative samples for torrefied wood is still low, so the heating value can be inaccurate.

4.2.2 Gasifiers

The Shell type high temperature slagging Entrained Flow gasifier was selected because this is the only commercial high temperature gasifier where solid biomass has been gasified so far (co-fired with coal) [5]. It uses oxygen as gasification agent, containing only 1% of nitrogen to minimize the amount of nitrogen in the syngas. The inert gas (CO_2) requirement for pneumatic feeding was calculated to be 0.006 m^3 kg^{-1} biomass. This is based on a void fraction of 0.75 required for dense phase transport and a particle density of 500 kg m^{-3}. It is assumed that the amount of water present in the fuel is sufficient to prevent the formation of soot in the gasifier so no external steam has to be added to the gasifier. The slag flowing down the reactor wall is kept liquid by controlling the mineral composition [6]. To maintain the right composition and the right amount of slag to cover the membrane wall of the reactor, a flux material is added. The amount of flux added is assumed to be 8 wt.% of the biomass. The typical operating temperature of an Entrained Flow gasifier is between 1300 and 1500°C. At lower temperatures the fuel is not converted completely and the viscosity of the ash can become too high. Higher temperatures decrease the Cold Gas Efficiency (CGE). The temperature selected is a trade-off between fuel conversion and CGE. As the biomass is more reactive than coal, a gasification temperature of 1300°C was selected [6]. The assumed carbon conversion at this temperature is 99.5%. Heat loss through the membrane wall of the gasifier vessel is assumed to be 2% of the thermal input (LHV basis) [7]. 1% of the heat is used to produce medium pressure steam; the remaining 1% is seen as an overall heat loss.

Conventional Circulating Fluidized Bed biomass gasifiers use air as the gasification medium. This dilutes the producer gas with nitrogen, which will end up in the SNG and cannot be easily removed. To prevent this nitrogen dilution oxygen has to be used as gasification medium. Replacing air by oxygen will increase the chance of local hot spots in the fluidized bed resulting in an increased risk of agglomeration of the bed material which is a major problem in operating Fluidized Bed gasifiers [8]. To reduce the risk of agglomeration the oxygen is diluted with steam. The selected steam to oxygen ratio is 1 (kg kg^{-1}) [9]. It should be noticed, however, that this assumption has a major impact on overall efficiency. VTT in Finland has built a pressurized steam/oxygen blown CFB biomass gasifier, but at this moment no experimental results are available in the public domain. ECN has previously done some experiments with atmospheric steam/oxygen blown CFB gasification where the steam to oxygen ratio was taken similarly to the ratio assumed in this study.

A heat loss of 1% from the reactor was assumed as for the EF gasifier. For this study a carbon conversion of 90% is assumed. This assumption is based on published data [10] and practical experience from the ECN CFB gasifier BIVKIN [11].

The system for the Indirect gasifier is based on the ECN MILENA technology [12, 13]. The basic working principles of the MILENA gasifier are similar to the SilvaGas [14] Indirect gasifier. Biomass is fed into a riser where a small amount of superheated steam is added (5wt.% of the biomass input). Hot bed material (typically sand) enters the riser from the combustor through a hole in the riser, which is located under the biomass feeding point. The bed material heats the biomass to typically 850°C. The heated fuel particles degasify and create a vertical linear velocity of approximately 6 m s^{-1}, leading to a “turbulent fluidization” regime in the riser and a carry-over of the bed material together with the degasified biomass particles (char). The vertical velocity of the gas is reduced in the settling chamber, causing the larger solids (bed material and char) to separate from the gas and to fall down into the downcomer. The producer gas leaves the reactor from the top and is sent to the cooling and gas cleaning section. The char is burned in the Bubbling Fluidized Bed, were it heats the bed material to approximately 925°C. The heated bed material leaves the Bubbling Fluidized Bed from the bottom and is sent to the riser again. Measurement in the lab-scale Indirect gasifier, MILENA, have shown that the nitrogen content in the producer gas can be as low as 1 vol.% (in dry gas) by purging the feeding system with CO_2 and minimizing the flue gas leaking from the combustor to the gasifier. A leakage of flue gas from combustor to gasifier of 1% is calculated from the measured nitrogen concentration in the producer gas.

The carbon conversion (CC) in the Indirect gasifier was calculated using the following relation:

$$CC(\%) = 65 + 0.15 * (T-760) \qquad T \text{ in } ^\circ C \qquad (4.1)$$

This relation is an estimate based on experimental results from the ECN MILENA lab-scale gasifier using wood particles between 0.3 and 3 mm and conversion data published for the Battelle/Silvagas Indirect gasifier [14]. Again a gasifier heat loss of 1% was assumed.

The gasifier exit gas compositions for all cases were calculated using empirical relations and assuming an offset temperature for water gas shift equilibrium. Table 4.2 shows the relations used for the two Fluidized Bed gasifiers and the Entrained Flow gasifier.

Table 4.2: Relations used to calculate gasifier outlet gas composition.

Component	Fluidized Bed	Entrained Flow
CH_4 [kg kg^{-1} fuel d.a.f.]	0.0873-0.082•ER	0
C_2H_2 [mol mol^{-1} CH_4]	0.02	0
C_2H_4 [mol mol^{-1} CH_4]	0.32	0
C_2H_6 [mol mol^{-1} CH_4]	0.02	0
C_6H_6 [mol mol^{-1} CH_4]	0.08	0
C_7H_8 [mol mol^{-1} CH_4]	0.01	0
NH_3 [mol]	0.5 • N in fuel	0
H_2S [mol]	0.9 • S in fuel	0.9 • S in fuel
COS [mol]	0.1 • S in fuel	0.1 • S in fuel

The ER (equivalence ratio) is defined as the amount of oxygen fed into the gasifier divided by the amount of oxygen required for stoichiometric combustion of the biomass. The relation given in the table and used in the model for the yield of methane as function of ER was published by Maniatis [15]. These relations correspond well with ECN measurements from Bubbling Fluidized Bed, Circulating Fluidized Bed and Indirect gasification between 800 and 900°C at atmospheric pressure and wood as fuel. For Indirect gasification an ER of 0 was used in the calculation. The relations for the higher hydrocarbons are determined from experiments in the ECN lab-scale MILENA gasifier [13]. The influence of pressure on the formation of hydrocarbons in Fluidized Bed biomass gasifiers is not known yet, therefore the same relations as for atmospheric operation were used. The tar concentration in the raw producer gas is 30 g Nm^{-3} dry for both Fluidized Bed gasifiers. This figure is relatively high because no catalytically active bed material, like olivine, is used in the gasifier to reduce the tar concentration and tar from the gas cleaning section is returned to the gasifier in the case of CFB gasification. The water gas shift equilibrium is set at 1200°C for the fluidized bed gasifiers and at operating temperature for the EF gasifier.

Chloride (as HCl) and NH_3 leaving the gasifiers have minor influence on the overall energy balance and are therefore neglected.

4.2.3 Gas cooling and gas cleaning

Syngas from the Entrained Flow gasifier is cooled down to 600°C by quenching with its own cooled gas of 250°C. A gas quench is chosen to prevent salt condensation and deposition on the downstream heat exchangers. The gas is cooled further by a conventional heat exchanger. The heat is used to produce steam.

Producer gas leaving the CFB or Indirect gasifier is cooled down to 400°C. This temperature is selected to stay above the dew point of the tar in the gas. The heat is used to preheat the air to the Indirect gasifier and/or to superheat steam. In the Indirect gasifier the fly ashes removed by a cyclone from the producer gas are sent back to the combustor, where the carbon in the ash is converted into flue gas. The completely converted ashes are blown out of the combustor and are collected in a bag-house filter. The carbon containing ashes from the CFB gasifier are disposed. Tar and remaining dust are removed by the OLGA gas cleaning technology [16]. The OLGA gas cleaning system consists of three stages. In the first stage, the collector (OC in Figure 4-3 and Figure 4-4), heavy tars are condensed from the gas by contacting the gas with cooled scrubbing oil. Most of the fine dust that passes through the upstream cyclone is also removed with the scrubbing liquid. Dust and heavy tar are separated from the oil and returned to the gasifier. In the Indirect gasifier the heavy tar is used as fuel for the combustor. In the second stage, the absorber (OA in Figure 4-3 and Figure 4-4), light tar components are dissolved in the oil. The absorber is operated above the water dew point to prevent condensation of water. Producer gas exiting the OLGA absorber is free of condensable tars. The oil from the absorber is sent to the third stage of the OLGA system, the stripper (OS in Figure 4-3 and Figure 4-4). The stripper is either operated with air at 180°C (Indirect gasifier) or with steam at 210°C (CFB gasifier). The air or steam leaving the stripper contains some oil, that can be partly regained in a condenser if air is used to operate the stripper. The oil consumption of the OLGA system is 0.1 g Nm^{-3} for the stripper operated on air (Indirect gasifier) and 1 g Nm^{-3} for the stripper operated on steam (CFB). The oil consumption is only a fraction of the thermal input of the plant and is therefore neglected in the overall heat balance. More information on the OLGA can be found in paragraph 6.6.4.

4.2.4 Cl and Sulphur removal

Clean biomass like wood contains low amounts of sulphur and chloride. A 1 GW_{th} gasifier fed with clean wood produces 2500 kg day^{-1} of sulphur. Typically 90% of the sulphur converts into H_2S and 10% into COS. Other organic sulphur compounds are neglected in this study. Measured HCl concentrations in raw producer gas are low (<5 ppm). The Cl concentration in wood is low already and most of the chloride is removed from the gas together with the ash. The maximum allowable sulphur inlet concentration for the different methanation catalysts is not known yet. An acceptable inlet concentration of 100 ppb is assumed for this study. A ZnO sorbent is selected for the bulk sulphur removal. This sorbent can remove H_2S and COS. Non regenerative processes are commercially available and regenerative processes are under development. The removal principle is based on the following equilibrium:

$$ZnO + H_2S \leftrightarrow ZnS + H_2O \qquad (4.2)$$

In this case the majority of H_2S is removed at 250°C. A guard bed removes the remaining H_2S at a slightly lower temperature (160°C). The same guard bed reactor is used to remove HCl. In the case of Fluidized Bed gasification other organic sulphur compounds are present in the producer gas as well. It is assumed that these components are converted to H_2S and removed by the ZnO sorbent. ECN is testing different sorbents/catalysts for this goal. Ammonia removal is not included in the analysis.

4.2.5 Methanation

CO and H_2 in the cleaned gas are converted into CH_4 by the following strongly exothermic reactions:

$$CO + 3\,H_2 \leftrightarrow CH_4 + H_2O \qquad \Delta H^\circ_{298} = -206 \text{ kJ mol}^{-1} \qquad (4.3)$$

$$CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O \qquad \Delta H^\circ_{298} = -165 \text{ kJ mol}^{-1} \qquad (4.4)$$

The generated heat is used to produce steam. In the case of Fluidized Bed gasification a major problem is the formation of soot in the methanation reactors which deactivates the catalyst and blocks the system. The presence of higher hydrocarbons in the producer gas seems to enhance the formation of soot on the methanation catalysts [17]. The required boundary conditions to prevent the formation of soot on the methanation catalyst are not well known yet, but, in theory, it can be suppressed by adding steam prior to the methanation. Table 4.3 shows the resulting steam to dry gas ratios for the three different configurations on the basis of Aspen Plus® calculations.

Table 4.3: Required steam to dry gas ratios to prevent soot formation

Gasifier	Operating pressure methanation unit [bar]	Final methanation temperature [°C]	Steam to dry gas ratio [mol mol^{-1}]
EF	30	280	1.4
CFB O_2/Steam	10	280	1.4
Indirect	7	280	1.5

The steam gas mixture is preheated to 350°C before it enters the first methanation reactor.

4.2.6 SNG upgrading

The SNG upgrading step is similar for the different systems, therefore this step is not modeled in detail. Gas leaving the last methanation reactor is cooled and water is condensed out of the gas which is then compressed to 30 bar (if required).

Different CO_2 removal technologies are commercially available: PSA (Pressure Swing Adsorption), Physical absorption (e.g. Selexol process) or membrane gas separation (UOP Separex membrane). In this study CO_2 is assumed to be removed for 98%. The energy consumption for CO_2 removal is calculated from the required pumping energy for the solvent in a Selexol unit [18]. The CH_4 loss is assumed to be 1%. The CO_2 rich gas is sequestrated in empty gas fields, which means that the overall process becomes CO_2 negative. The required compression energy is not included in the calculated overall efficiency.

The Swiss national standard for unlimited gas injection limits the hydrogen concentration in SNG to 5 vol.% [19]. This value was used in this study as the maximum allowable hydrogen concentration in the final SNG. The H_2 concentration can be kept below this concentration by choosing the appropriate operating conditions (temperature and pressure) of the last methanation reactor. Although additional H_2 removal is not required, selective oxidation could be applied if necessary.

4.2.7 Steam system

All heat above 200°C is used within the system to generate steam or preheat combustion air. The remaining low temperature heat is sufficient for heating all feed water to 150°C. Most of the heat is used to produce low pressure (10 bar) and medium pressure (60 bar) steam of which a significant part is mixed into the producer gas before the methanation unit to prevent soot formation. The remaining steam is sent to medium and low pressure steam turbines. An isentropic efficiency of 80% and a mechanical efficiency of 98% are assumed for the steam turbines.

4.2.8 Electricity consumption

An overall electricity consumption of 1.5% of biomass input to the gasifier (HHV basis) is assumed. Major electricity consumers like the air separation unit and the different gas compressors, are not included in this figure and calculated separately. An isentropic efficiency of 80% and a mechanical efficiency of 98% are assumed for the gas compressors. The cryogenic air separation unit consumes 0.4 kWhe Nm^{-3} oxygen [20].

4.3 Results

The performance of a gasifier is given by its Cold Gas Efficiency (CGE), which is defined here by the heating value of the dry, cold producer gas after gas cleaning divided by the heating value of the biomass or torrefied biomass to the gasifier. The CGE on HHV basis is defined as the higher heating value of the dry and cleaned gas divided by the higher heating value of the biomass. The CGE on LHV (lower heating value) basis is defined as the lower heating value of the dry and cleaned gas (thus excluding condensation heat of water in the gas produced when the gas is combusted) divided by the lower heating value of the biomass. Figure 4-5 shows the calculated Cold Gas Efficiencies for the different gasifiers.

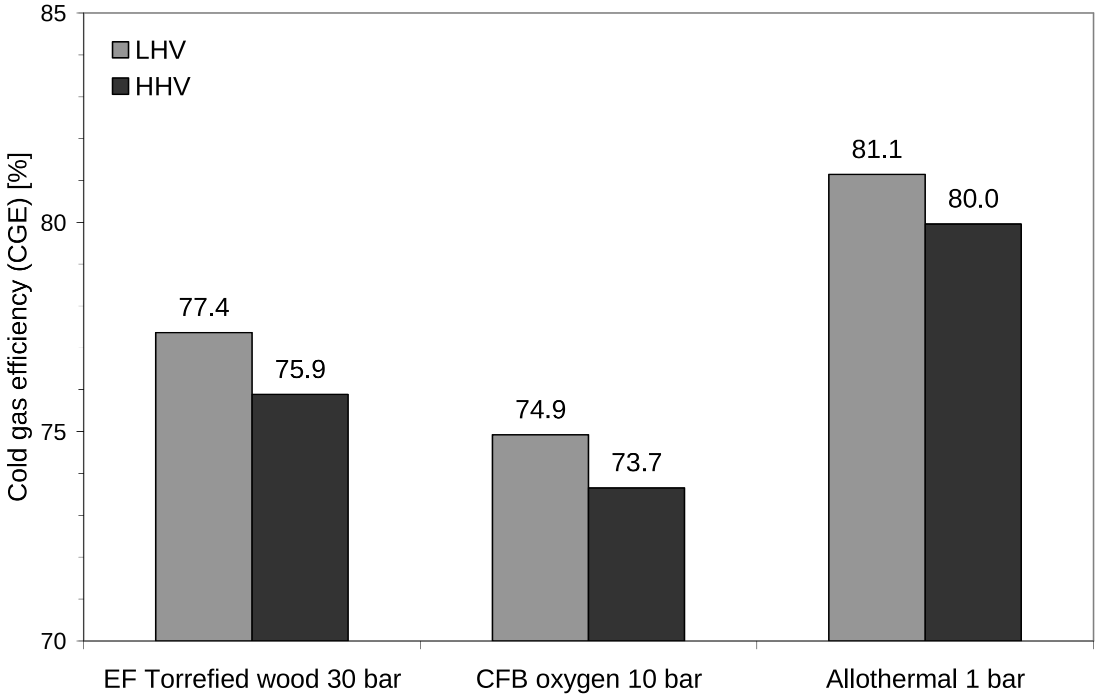


Figure 4-5: Cold Gas Efficiencies for the three different biomass gasifiers.

It should be noted that the cold gas efficiency presented for Entrained Flow gasification (EF) does not include the efficiency from wood to torrefied wood. Torrefaction gives a loss of approximately 10% on HHV basis and 7% on LHV basis. The torrefaction process is still under development. The efficiencies assumed for torrefaction are based on preliminary data. Figure 4-6 visualizes the different losses on Cold Gas Efficiency for the three gasifiers.

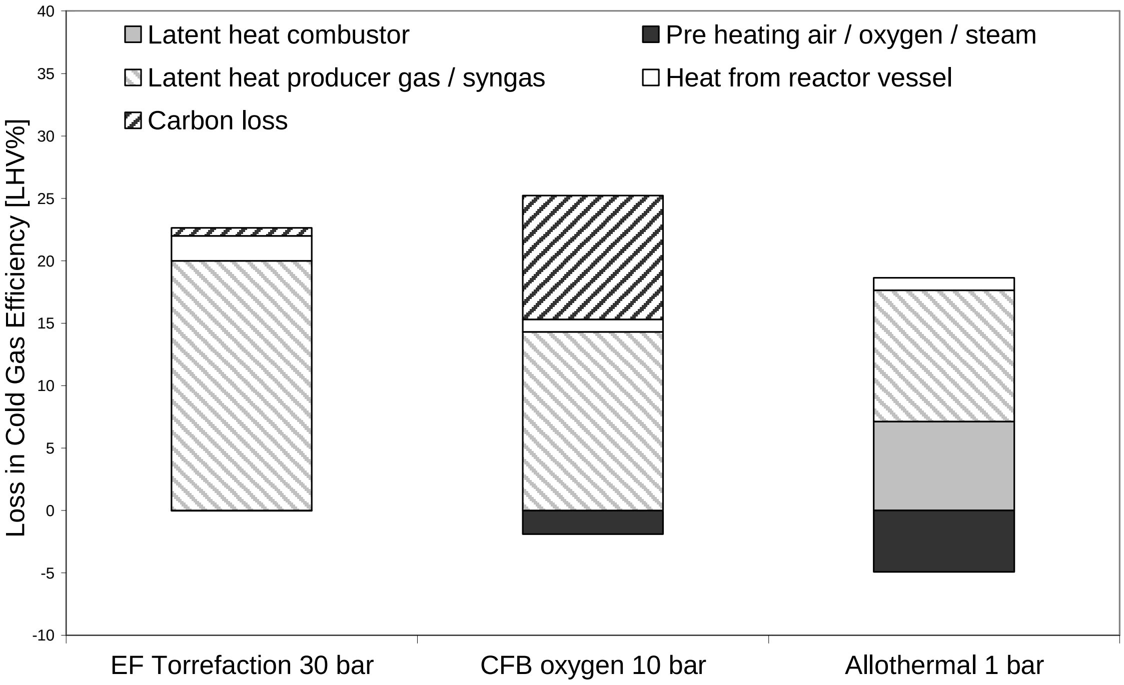


Figure 4-6: Visualization of losses in Cold Gas efficiency (CGE).

The main loss in cold gas efficiency for the Entrained Flow gasifier is due to the latent heat in the syngas. The gasifier exit temperature of the Entrained Flow gasifier is ±450°C higher than the exit temperatures of the two Fluidized Bed gasifiers. The main losses for the CFB gasifier are due to the unconverted carbon in the ash and the latent heat in the producer gas. The Indirect gasifier has the highest cold gas efficiency, because the losses are reduced to a minimum. The combined latent heat in flue gas and producer gas is the highest, but is compensated by preheating the combustion air. Table 4.4 gives the gas compositions for the three gasifiers after tar removal.

Table 4.4: Gasifier wet gas compositions (after tar removal, if applicable).

	EF Torrefied wood 30 bar	CFB oxygen 10 bar	Indirect 1 bar
CO [mol%]	49.8	18.7	25.9
H_2 [mol%]	18.6	14.4	21.3

CO_2 [mol%]	14.7	19.8	12.0
O_2 [mol%]	0.0	0.0	0.0
H_2O [mol%]	16.5	39.2	24.8
CH_4 [mol%]	0.0	5.1	10.3
N_2 + Ar [mol%]	0.3	0.4	0.9
C_2H_2 [mol%]	0.0	0.1	0.3
C_2H_4 [mol%]	0.0	1.7	3.4
C_2H_6 [mol%]	0.0	0.1	0.2
C_6H_6 [mol%]	0.0	0.3	0.6
C_7H_8 [mol%]	0.0	0.0	0.1
H_2S [ppm]	195	200	314
COS [ppm]	22	22	35
NH_3 [ppm]	0	1471	2308

The concentrations of methane and other hydrocarbons in the gas from the Indirect gasifier are higher than those in the gas from the CFB gasifier, because the water concentration is lower and no methane is burned. As can be seen in Table 4.2 the yield of hydrocarbons is a function of the air to fuel ratio (ER). A typical ER for an oxygen – steam blown CFB is 0.25. The air to fuel ratio in an Indirect gasifier is near zero, giving the maximum yield in hydrocarbons. A high initial concentration of methane and other hydrocarbons has a positive influence on overall efficiency to SNG.

The overall efficiencies to SNG are calculated on a net and gross basis. Gross efficiency does not take into account the electricity production or consumption of the system. To calculate the net efficiency the electricity consumed (or produced) is assumed to be produced by converting SNG into electricity with an efficiency of 60% for electricity generation. Figure 4-7 shows the calculated overall efficiencies of biomass to SNG at 30 bar. As can be seen from the figure the gross efficiencies to SNG are relatively low for Entrained Flow gasification. The net efficiency is higher, because the system produces electricity. The other two systems consume electricity. The reason for the net electricity production in the EF based system is the significantly higher production of steam in both the syngas cooler and the methanation unit. The overall efficiencies are higher on HHV basis than on LHV basis. It is caused by the conversion of CO into CH_4 in the methanation reactors. The lower and higher heating values of CO are equal, while the higher heating value of CH_4 is higher than the lower heating value.

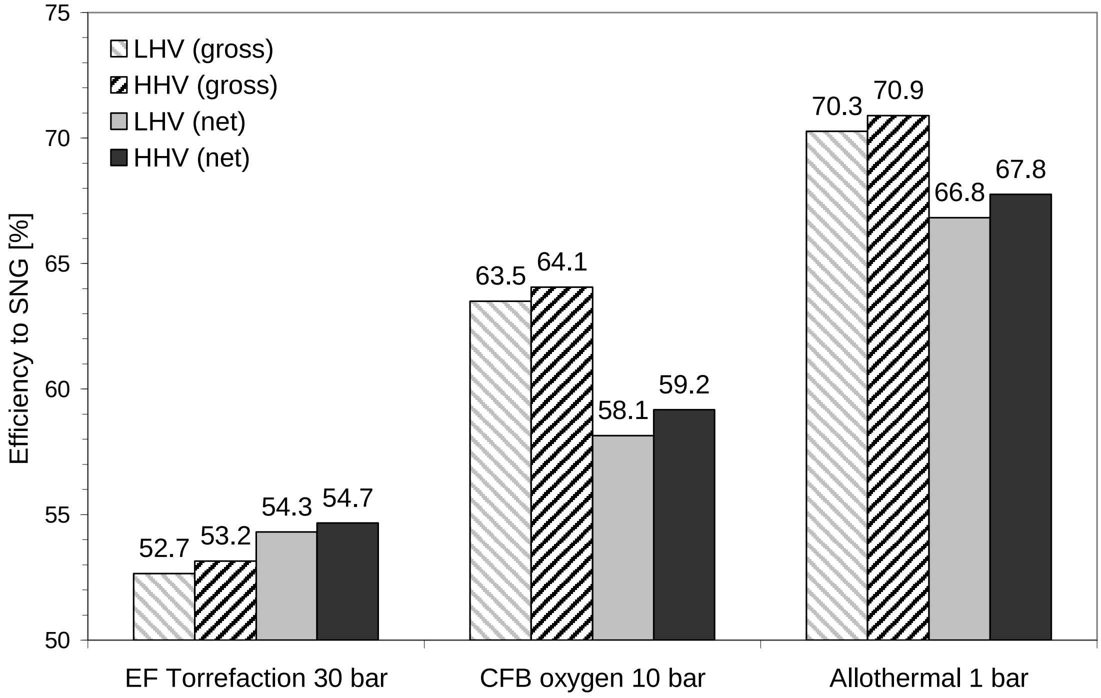


Figure 4-7: Gross efficiency to SNG and net efficiency to SNG and electricity.

Gross and net overall efficiencies on LHV and HHV basis are highest for the Indirect atmospheric gasifier. These high values are due to the low losses, both heat and unconverted carbon, and the fact that the gas from the gasifier already contains a significant amount of hydrocarbons. These hydrocarbons, as indicated in Table 4.4, amount to a 55% contribution to the calorific value of the producer gas. Hydrocarbons are converted into methane with a relatively high efficiency compared to the conversion of syngas into methane. The gas after the methanation reactors in all three systems contains approximately 60 mol% of water. All the condensation heat of this water (approximately 20% of the thermal input) is not used in the defined systems. If a biomass dryer is integrated in the system this low temperature heat can be used to dry the biomass. Integrating the drying step will increase overall efficiency for all defined systems. Another possible improvement of the systems efficiency is reduction of the amount of steam required in the different methanation steps to prevent formation of soot. The amount of steam required is now based on thermodynamic equilibrium calculations.

The Indirect gasifier is operated at atmospheric pressure, because pressurized operation of an Indirect gasifier is not yet demonstrated. In principle it is possible to operate an Indirect gasifier at elevated pressures which will further increase the overall efficiency. The methane yield is possibly increased by pressure. This will have a positive effect on overall efficiency to SNG.

Table 4.5 shows the calculated concentrations of the major components in the produced SNG after water and CO_2 removal. As can be seen the calculated heating values are similar. The amount of H_2 is lower in the case of EF gasification, because the methanation pressure is higher. The concentration of CO is below 0.1 mol% in all cases. The gas composition can be influenced by changing the level of CO_2 removal.

Table 4.5: SNG gas compositions.

	EF Torrefied wood 30 bar	CFB oxygen 10 bar	Indirect 1 bar
CH_4 [mol%]	90.5	89.9	90.7
H_2 [mol%]	2.5	3.9	4.1
CO_2 [mol%]	5.1	3.3	1.8
N_2 + Ar [mol%]	1.7	2.4	3.0
LHV [MJ m^{-3}]	33.5	32.8	33.3
HHV [MJ m^{-3}]	37.2	36.4	37.0

From a technical point of view the gas should be suitable to replace fossil natural gas, but exact specifications for injection in the natural gas grid are not yet clear and may vary per country.

4.4 Concluding remarks

Overall efficiency to SNG is highest for Indirect gasification. The net overall efficiencies on LHV basis, including electricity consumption and pretreatment but excluding transport of biomass are 54% for Entrained Flow, 58% for CFB and 67% for Indirect gasification.

Because of the significant differences in overall efficiencies to SNG for the different gasifiers, ECN decided to select the Indirect gasification as the preferred technology for the production of SNG.

In a recent study a comparison was made between a MILENA based SNG system and a SNG system based on the FICFB gasification technology combined with the PSI methanation technology [21]. The differences between the two concepts have been quantified and estimated to be 6 $\%_{abs}$ on overall efficiency from wood to Bio-SNG in favor of the MILENA concept. The main reason for this difference is the relatively large amount of steam that is required for the FICFB process.

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Chapter 5

MILENA model

Abstract

A pseudo-equilibrium model was made to calculate the gas composition and the mass and energy balance. The essential empirical relations were obtained from literature and experiments in CFB and BFB gasifiers. These relations were updated using data from the lab-scale and pilot scale gasifiers. The MILENA model is described in this chapter. The relations obtained from experimental work in the MILENA are described in chapter 7.

5.1 Fluidized Bed biomass gasification models

The objective of a biomass gasification model is to predict gas composition, gas flow and reactor temperature at specified process conditions. These data are required to dimension the gasification reactor and the connected gas cleaning equipment. Several models for Fluidized Bed gasification are described in literature [1, 2]. Many of these models are based on theories about fluidization hydrodynamics, coupled with kinetic schemes for the heterogeneous and homogeneous processes occurring inside the gasifier such as drying, pyrolysis, tar cracking and char gasification. These models are in general very complex and only include the main gas compounds. This makes practical application difficult.

More simplistic models (excluding hydrodynamics and kinetic schemes) are often used to describe an integrated gasification system. These models are mostly based on chemical equilibrium. Aspen Plus® is generally used as modeling tool. The assumption of chemical equilibrium works very well if high temperature gasification, like Entrained Flow gasification, is to be described. For Fluidized Bed gasification this method is not applicable, because a major part of the combustible compounds is present in the gas as hydrocarbons. At chemical equilibrium the presence of hydrocarbons is negligible at typical operating conditions of Fluidized Bed gasifiers.

A commonly used method to apply the equilibrium calculation is to define a negative offset temperature for the equilibrium temperature of the methanation reaction. By doing this it is possible to fit the main gas composition (CO, CO_2, H_2, H_2O and CH_4) to measured values. The higher hydrocarbons are not included, but their impact is not large as CH_4 represents the major part of the hydrocarbons. This method is only applicable at the temperature and pressure at which the offset temperature could be fitted to the experimental data.

The method selected by ECN for modeling the MILENA process and other Fluidized Bed processes is a combination of chemical equilibrium for the CO shift reaction and empirical relations for the yield of hydrocarbons. This model is best described as a pseudo-equilibrium model. VTT uses a similar approach [3, 4].

CFD analysis was used to predict gas flow patterns in the MILENA settling chamber. A more detailed model of the gasifier riser was made to predict the char conversion of different biomass particles in the gasifier riser [5]. A summary of the results is given in paragraph 6.2.

5.2 MILENA pseudo-equilibrium model

The first step in the design process of the gasifier is the calculation of the mass and energy balances. Aspen Plus® has been used in the past to model Fluidized Bed gasification, but because of the many empirical relations that are required the use of

Excel is more convenient. The Excel model made for this purpose solves the component- and energy balances. The model is an adaptation of a model made for the CFB gasifier BIVKIN, which is extensively verified using data from lab-scale, pilot-scale and commercial scale Fluidized Bed gasifiers.

5.2.1 Model layout

Figure 5-1 shows the basic layout of the Excel model. The model includes the conversion of the producer gas into Bio-SNG. This makes it possible to see the effect of gasifier process parameter variations on overall efficiency to SNG. The model is divided into different process blocks (e.g. drying, gasification, char combustion, etc.). The energy and component balances are solved for each process step where the individual process blocks produce outputs which act as inputs for the next step. For example: the gasifier produces char. The amount and composition of the char is an input variable for the combustion step. The temperature of the exiting gases and solids results from the energy balance over the block describing the combustion.

The Gasifier and Combustor block are described in more detail, because these are the essential blocks to describe an Indirect Gasifier.

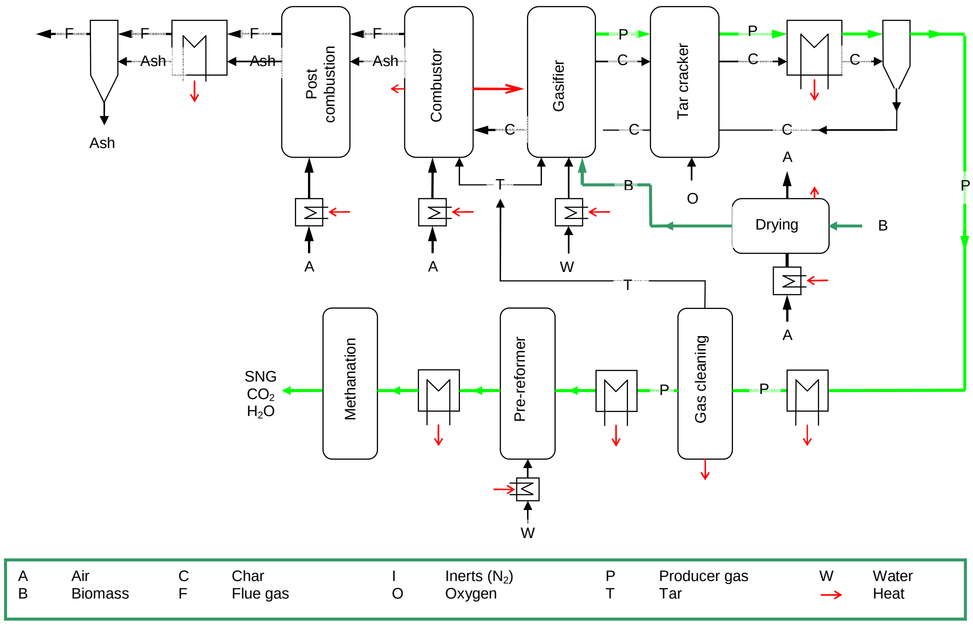

Figure 5-1: Basic layout of the integrated MILENA thermodynamic model

The Drying block simulates a simple single pass rotary air drum dryer in which heated air is used to dry the biomass. The outlet air temperature is set at a fixed value (typically 20°C) above the water dew point of the air exiting the dryer. The block is used to calculate the influence of integrating a dryer when relatively wet biomass is foreseen as fuel. The integration of a dryer can increase the overall efficiency on LHV basis significantly.

The Gasifier block converts the solid fuel into a combustible gas containing also pollutants like fly-ash, tar, chloride and sulphur.

The Tar cracker block can be used to describe a thermal or a catalytic tar cracker/reformer. Oxygen is normally added to keep the process at the required operating temperature.

A simple Gas cleaning block is integrated into the overall model to calculate the amount of light and heavy tars that are removed and become available as fuel for the combustor. The NH_3 concentration after the gas cleaning system can be entered. Ammonia has a relatively high heating value, so the removal rate has a significant influence on the heat balance. Ammonia is not returned to the gasifier. The water removal rate is calculated from the specified water dew point. The Cold

Gas Efficiency (CGE) is calculated in the Gas cleaning block, because the removal or recycle of tars, BTX and NH_3 influences the CGE. The raw gas from the MILENA is normally not directly usable (with the exception of direct firing in a boiler), so defining the CGE after cleaning the gas is more logical.

Upgrading of the gas is described by the Pre-reformer and Methanation block. Both reactors are normally at chemical equilibrium. Steam can be added to prevent soot formation. The outlet of the Methanation block is used to calculate the overall efficiency from wood to Bio-SNG.

A separate calculation block is used in the model to check the hydrodynamic behavior of the bed material and char particles.

5.2.2 MILENA gasifier

The MILENA gasifier is described by the combination of the Gasifier block and the Combustor block. Figure 5-2 shows the two blocks and their connections in more detail. Biomass (B) is fed into the gasifier with some inert gas (I) to purge the feeding screw. Air (A) or steam is added to the bottom of the gasifier as fluidization agent. The biomass is converted in producer gas (P) and char (C). The tars are included in the producer gas (P). Most of the char is separated from the producer gas and falls down in the combustor. The remaining char leaves the gasifier as fine dust.

Some of the producer gas (P) leaks into the combustor. The red arrows represent the heat that is transferred from the combustor to the gasifier by the circulating bed material. Some air (A) or Flue gas (F) can leak from the combustor into the gasifier. Oxygen (O) can be transported from the combustor to the gasifier by the circulating bed material.

Air (A) is added to the combustor to convert the char in flue gas. Light tars (T2) and heavy tars (T1) from the OLGA gas cleaning are combusted as well. The flue gas (F) and the remaining ash leave the combustor bed. The small red arrow represents the heat loss from the reactor.

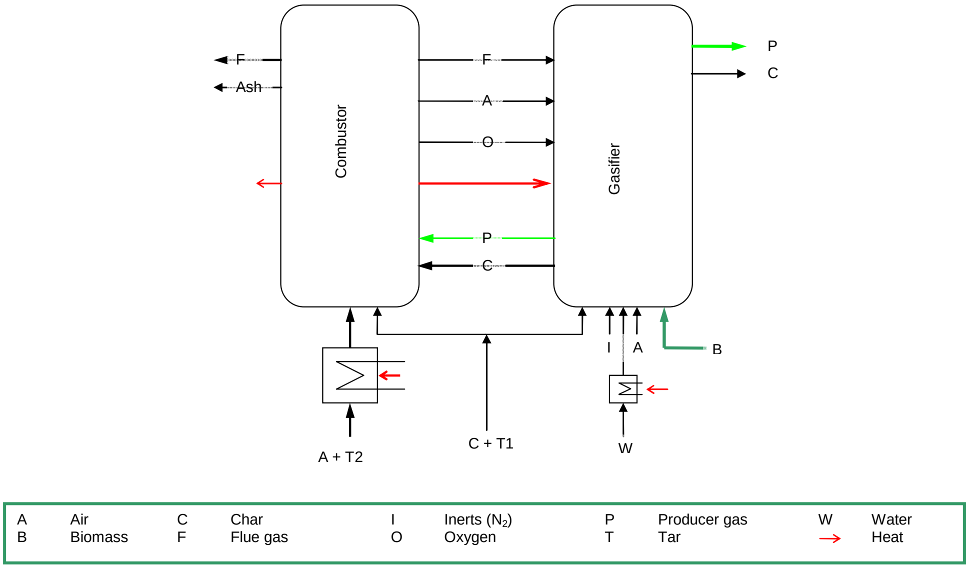

Figure 5-2: MILENA gasifier and combustor blocks.

The Gasifier block calculates the gas composition using empirical relations for the hydrocarbons. The concentrations of CO, CO_2, H_2 and H_2O are results of the component balances and the water gas shift equilibrium. The method of calculating the gas composition and the carbon conversion is explained in more detail below.

The net heat input into the gasifier is calculated from the temperature difference between combustor and gasifier and the circulation rate of the bed material. This circulation rate of the bed material is used as an input parameter. The higher the circulation rate, the lower the temperature difference between combustor and gasifier. The circulation rate is normally set at 40 times the amount of biomass fed to the gasifier, which results in a typical temperature difference between combustor and gasifier of 70°C when an inert bed material like sand is used.

The main purpose of the Combustor block is to calculate the amount of heat that is available to the Gasifier block. The majority of the fuel for the Combustor block is the char which is produced in the Gasifier. Separation efficiency for char in the settling chamber must be specified. The typical value is 90%, leaving 10% of the char to be entrained with the producer gas leaving the gasifier. This residual char is collected after the producer gas cooler and recycled to the Combustor. The Combustor is operated at a low air to fuel ratio (typical lambda value of 1.1). The lambda value can be entered as a set point value so that the combustion air flow can be calculated. The combustion air temperature is an input value. Preheating has a strong positive effect on CGE, so, in theory, this temperature should be as high as possible, but for reasons of practical integration, normally the pre-heat temperature is limited to 320 – 400°C. The char composition influences the concentration of pollutants like HCl and SO_x in the flue gas.

The assumed char composition and distribution of pollutants over the producer gas and char are explained in more detail below. The NO_x concentration in the flue gas is not calculated, but taken as an input value. Part of the fuel (char) bound nitrogen is converted into NO_x, but no (empirical) relations are known for their concentrations. The concentration of CO and unburned hydrocarbons in the flue gas is taken to be zero in the model. In practice these concentrations are very low (<< 0.1 vol.%) and have a negligible effect on the overall balance.

The flue gases from the BFB section of the combustor are post combusted in the freeboard of the MILENA to reduce emissions of CO and C_xH_y. The typical O_2 concentration in the flue gas of the MILENA combustor outlet is between 4 and 6 vol.%. The Post combustion block is used to simulate this section (see Figure 5-1). In the model the concentrations of CO and C_xH_y are already zero in the gas exit of the Combustion block, so the Post combustion block calculates only the drop in flue gas temperature due to dilution of the flue gas with air.

Optionally, gas exchange between the Combustor block and Gasifier block can be integrated into the model. Flue gas can leak from the Combustor into the gasifier. The typical leakage is set to 0.8% of the total flue gas flow in the Combustor block. This value was obtained from measurements. Producer gas can leak via the downcomers into the combustor, where it acts as additional fuel. During normal operation this leakage is very small and therefore neglected. It was observed that some bed materials (e.g. olivine) can transport oxygen from the combustor to the gasifier. This is caused by oxidation and reduction of, for example, iron. In the model this is simulated by an oxygen flow from the Combustor block into the Gasifier block.

5.2.3 Gas composition

The gas composition of a gas produced in a Fluidized Bed gasifier at 850°C is not at chemical equilibrium, except for the water gas shift reaction if a catalytic bed material is used. The concentrations of hydrocarbons are significantly higher than calculated using the equilibrium approach. Empirical relations are used to estimate the yield of hydrocarbons. A similar approach was selected by VTT [3]. The equations used by ECN are given table 4.2. The relation to predict the CH_4 yield as function of the Equivalence Ratio (ER) is obtained from literature [6]. This relation is verified with data from various ECN Fluidized Bed gasifiers and appears to be reliable. ER is defined as the amount of oxygen fed into the gasifier divided by the amount of oxygen required for stoichiometric combustion of the biomass.

The yield of higher hydrocarbons is a fraction of the methane yield. The fractions are obtained from measurements originally done in the CFB gasifier BIVKIN, but later are adapted using data from the lab-scale and pilot-scale MILENA gasifiers. The work described in this thesis resulted in updated relations for the hydrocarbon yield. The updated relations can be found in chapter 7.

It must be noticed that the relation used for CH_4 does not include temperature. Measurements have shown that the influence of temperature on methane yield is small at the typical operating window of Fluidized Bed gasifiers (770 – 880°C).

5.2.4 Carbon Conversion

Equation 5.1 defines the carbon conversion. The carbon in the tar is included in $C_{producer\_gas}$.

$$\varsigma_{c,gas} = \frac{C_{producer\_gas} - C_{feed\_gas} - C_{additives}}{C_{fuel}} \qquad (5.1)$$

C is defined as mass flow of carbon in the different streams. $C_{additives}$ is introduced in the relation to compensate for the carbon released by calcination reactions. Dolomite, for example, undergoes calcination at typical gasifier conditions releasing CO_2. The calcination reactions are endothermic which has to be taken into account in the heat balance.

Equation 5.2 shows the relation used to calculate the carbon conversion. The residence time of a biomass/char particle is relatively short, so gasification reactions of the char with steam or CO_2 can be neglected.

$$\varsigma_{c,gas} = X + Y*(T-760);\ T \text{ in °C.} \qquad (5.2)$$

The original carbon conversion relation was based on data published by Battelle [7] for the gasification of different wood particles/chips. The obtained values for X and Y were: X = 55 and Y =0.11/°C, but it is not clear whether the carbon conversion definition used for the conversion by Battelle also includes tar in the producer gas. The carbon conversion definition used in the MILENA model includes tar, because tar is in the gas phase at the operating temperature of the gasifier. Because the residence time in the MILENA riser plus settling chamber is longer than in the Battelle riser, a higher carbon conversion is assumed. The following values are estimated and used for the basic design of the MILENA reactor: X=65 and Y = 0.15/°C.

More recent data found in literature for the Battelle / SilvaGas gasifier result in the values: X = 56 and Y = 0.126/°C [8] and X = 58 and Y = 0.16/°C [9]. Especially the latter values are close to the assumptions originally made for the MILENA design. The assumed relations are shown and compared to measured carbon conversions for the lab-scale installation and pilot plant in chapter 7 (Figure

7-6). The relation is modified based on experimental data from the MILENA pilot plant (see Chapter 7).

VTT has published a relation for carbon conversion in Fluidized Bed biomass gasifiers [3, 4], but this relation is fitted to experimental data generated with an air or steam/oxygen blown gasifier and is therefore not applicable to the MILENA gasifier.

5.2.5 Char and tar composition

In earlier versions of the MILENA model it was assumed that the char leaving the gasifier was pure carbon and (inert) ash. The content of other elements was neglected. From measurements and literature [10] it was found that, especially, the oxygen content can be significant. Analyses of char produced in the ECN CFB gasifier are used to set a standard composition of char. Table 5.1 shows the measured compositions (wood was used as fuel). The average composition is used in the MILENA model.

Table 5.1: Char composition used in MILENA model

		BIVKIN	BIVKIN	Average
Date		20-11-1997	17-12-2002	-
C	[wt.% daf]	90	95	92
H	[wt.% daf]	1	1	1
O	[wt.% daf]	9	3	6

Experimental data from the pilot plant are used to update the composition. The updated composition values can be found in Chapter 7.

The sulphur content in the gas is calculated by assuming that all sulphur in the fuel that is not converted into H_2S or COS remains in the char. The same is done for chloride. The nitrogen content in the char is assumed to be $^1/_3$ of the nitrogen in the fuel.

The composition of the tar leaving the MILENA lab-scale gasifier, operated at a gasification temperature of approximately 800°C and fed with beech wood (Rettenmaier Rauchergold, type HBK 750/2000) was measured during the December 2006 test campaign. During these tests the bed material in the gasifier was olivine. Six samples were taken and analyzed used the SPA method [11]. The variations in composition of the different samples are small. The average tar concentration was 28 g Nm^{-3} dry. Part of the tar components (22 wt.%) cannot be identified. Table 5.2 shows the tar composition used in the model.

Table 5.2: Tar composition used in MILENA model

C	[wt.%]	94.2
H	[wt.%]	5.8
O	[wt.%]	-

5.3 Use of the model

The MILENA model is used to dimension the lab-scale, pilot-scale and demonstration-scale MILENA reactors. The model is used to specify the required operating conditions for the different tests. The model is also used to calculate deviation from CO shift equilibrium, the carbon conversion, and solids circulation rate from test data.

Figure 7-19 shows the first page of the MILENA Excel model. The fuel composition is the first required input. It is also the input parameter that is responsible for most of the uncertainty in the results. In practice the specified fuel/biomass composition and heating values are often uncertain. This is checked by using empirical relations to calculate the heating value from the composition [12]. Normally the calculated value should be within 3% of the measured heating value.

The deviation from CO shift equilibrium is defined as the difference between the calculated CO shift equilibrium temperature and the actual temperature at the gasifier riser outlet. When a catalytically active bed material like olivine is used the CO shift reactions are normally close to equilibrium. The typical deviation is normally between 0 and +400°C. Deviations outside this range are indications for errors or a catalytically inactive bed material.

Carbon conversion is estimated from the compound balance by fitting the calculated producer gas CO, producer gas CO_2, flue gas O_2 and flue gas CO_2 concentrations to measured values. The sum of the square of the differences between calculated and measured gas concentrations is minimized to get the most reliable results. All other measured values (e.g. flue flow, fuel composition, gas flows, CH_4 concentration, etc.) are put in the model as constants.

The bottom part of the Gasifier block in the model shows the heat balance over the Gasifier block. The heating value on HHV basis is used to solve the balance. The heat balance on LHV balance is only used to check for deviations. The solids circulation rate is calculated from the combined heat balance of the Gasifier and Combustor block.

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Chapter 6

MILENA gasification technology

Abstract

In this chapter the MILENA gasification technology and the MILENA lab-scale and pilot scale plants are described in more detail. A historical overview of the development is given in the first paragraph. The different design considerations are described to explain the benefits of the MILENA concept against the CFB concept which was originally used by ECN for biomass gasification. This chapter ends with a comparison of the MILENA concept with other Indirect biomass gasification processes (SilvaGas and FICFB).

6.1 Development

ECN started the development of Fluidized Bed gasification in 1996 with the realization of the air blown BIVKIN CFB gasifier [1]. Tests done during the first years of operation showed some limitations of the CFB concept. The carbon conversion was limited to approximately 90% for dry wood [2], resulting in a reduced overall efficiency. The residual fly ash therefore contained a high amount of carbon (typical 50%), making it difficult to dispose this as a waste stream. The BIVKIN gasifier is of the air blown type, so the producer gas is diluted with nitrogen up to a concentration of 50 vol.% on dry basis, resulting into relatively low heating values of less than 7 MJ Nm^{-3} of the dry gas on LHV basis. When wet fuels were gasified the heating value was even lower. The relatively low heating value made it hard to achieve stable combustion of the gas and made it also unfeasible to upgrade the gas into Bio-SNG. A limited test program was done with an oxygen steam mixture as gasification agent to prevent the nitrogen dilution and increase the heating value of the produced gas. The results were somewhat disappointing, the heating value of the gas increased, but the carbon conversion decreased and problems with bed agglomeration occurred. ECN did not continue steam/oxygen blown CFB gasification, but others like VTT were more successful and continue the development of steam/oxygen blown CFB gasification [3].

Battelle’s Columbus Laboratories had been working on the development of Indirect gasification since the late 1970's and reported positive results generated with a 10 ton/day pilot plant using a variety of biomass fuels [4]. The results reported by Battelle and the experience with the ECN CFB gasifier resulted in the first design of an ECN indirect gasifier. The concept was originally called STAR.

The STAR cold flow test rig was adapted and used for the further development of the MILENA process. The settling chamber is still part of the concept, because even a limited tar reduction can be beneficial, especially in preventing tar fouling in the producer gas cooler. The successful development of the OLGA tar removal technology at ECN made it possible to allow the relatively high tar content in the gas.

In 2003 it was decided to build a second lab-scale Fluidized Bed gasifier at ECN, because the existing lab-scale gasifier was frequently overbooked. At that moment Indirect gasification was identified as a promising option for the production of Bio-SNG [5]. Therefore a lab-scale concept allowing both Indirect and Direct gasification was adopted. The gasifier was named the “Multipurpose Integrated Lab-unit for Explorative and iNnovative Achievements in biomass gasification” or MILENA. By means of a metal insert the gasifier could be transferred from a Bubbling Fluidized Bed (BFB) gasifier into an Indirect gasifier.

After successful operation of the MILENA lab-scale gasifier for some years it was decided at the end of 2006 to start the realization of a pilot-scale gasifier. Construction started in 2007 and the pilot plant was taken into operation in 2008. First tests with the complete system (gasifier and gas cleaning) were done in 2009. Figure 6-1 shows the MILENA pilot plant gasifier and the installation of the OLGA gas cleaning pilot plant.

The basic working principles of the MILENA process are explained in paragraph 2.6. In this chapter a more detailed explanation of the MILENA process and reactor design are given.

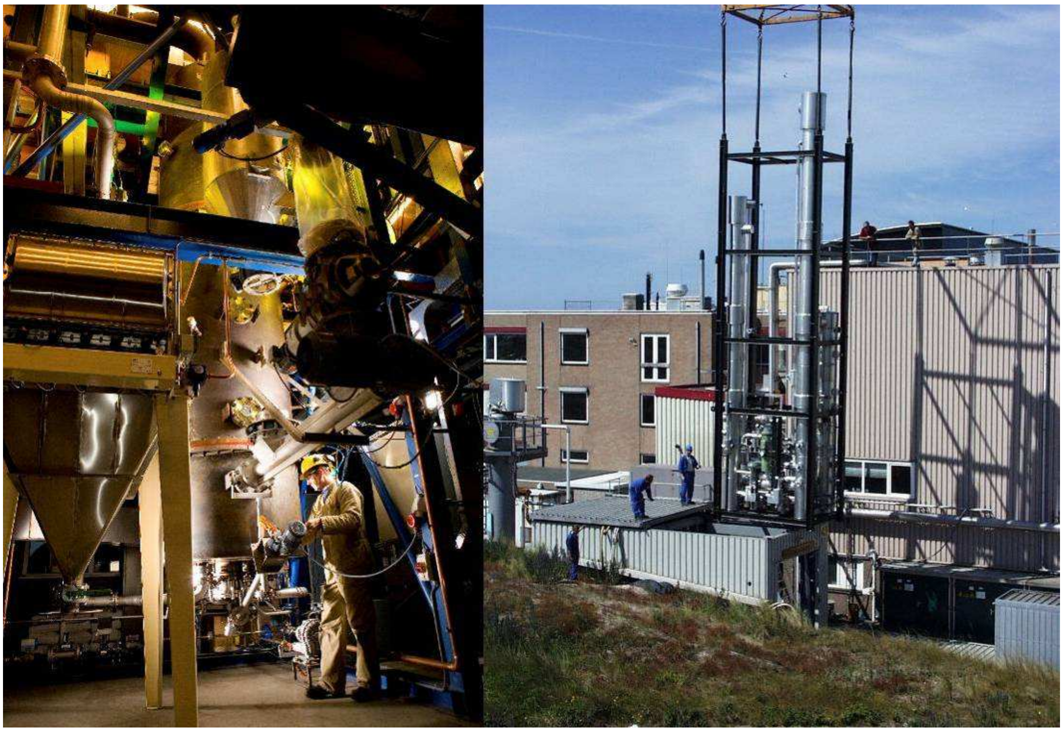

Figure 6-1: pilot-scale MILENA gasifier (left) and installation of the OLGA pilot-scale gas cleaning (right) at ECN.

6.2 Design considerations

The original MILENA design was made after the first years of experience with operating the CFB gasifier BIVKIN, and therefore many of the design considerations were influenced by these experiences. The preference for Indirect gasification over Direct gasification was primarily based on the increase in heating value of the gas and the increase in overall carbon conversion. The following design considerations resulted in the present MILENA design.

Integrated reactor:

The process was designed to be accommodated in a single reactor vessel. The original main reasons for doing this were a lack of space and the requirement to limit the heat loss from the gasifier. The desire to limit horizontal transport of solids and problems with expansion joints in the ECN CFB gasifier made the design of an integrated concept more logical. An integrated reactor design is more suitable for pressurized operation. In the ECN CFB gasifier (BIVKIN) the circulating solids were transported from the circulation cyclone via the sealpot (a fluidized bed acting as a non mechanical valve) and a pipe for further downward flow into the riser. Because of the horizontal distance between the sealpot and the riser the transport pipe was relatively long which gave some operational problems. The effective flow diameter of the pipe sometimes decreased because of agglomerated bed particles sticking to the wall of the pipe. The designs for the commercial plants that were based on the BIVKIN design were made with an increased angle for this pipe. The MILENA design was made such that horizontal transport of the circulating bed material would not be an issue at all. The horizontal transport (outside the BFB) is limited to the wall thickness of the riser pipe.

When two high temperature reactors are connected to each other, differences in thermal expansion need to be compensated. This was mostly done by metal expansion joints which allow differential expansion. In the ECN CFB gasifier this was a major problem during the first years of testing. The expansion joints broke frequently and replacement was expensive. After modification the remaining thermal stresses were still relatively high and resulted in deformation and small leakages in the sand recirculation system. The MILENA design has no expansion joints and stresses due to thermal expansion are minimized by an integrated design. This design allowed the metal insert (the riser, settling chamber and downcomers) to expand more than the refractory lined reactor. The insert is only connected at the top of the reactor and can freely move at the bottom part of the reactor. This design feature is patented [6].

An important characteristic of the lab-scale MILENA design is the ability to remove the insert (the riser, downcomer and settling chamber). Without the insert the installation can be used as conventional BFB gasifier or combustor. This made the MILENA reactor multifunctional, which is important for research purposes.

An integrated reactor design is more suitable for pressurized operation, because all the parts are placed in a single pressurized vessel. The insert is not exposed to additional mechanical stresses at pressurized operation. It is expected that pressurized operation (e.g. 3- 7 bar) of the MILENA will become important for large scale Bio-SNG production.

Steel insert:

The selection of a steel insert was logical for the lab-scale and pilot plant installation because alternatives (refractory) are not practical at these scales. The ECN Engineering and Service department made a short survey and selected the steel grade 253MA for the lab-scale installation. This same grade was also used in the pilot plant. Extensive thermal stress analysis using the finite element method showed that a metal insert was a good solution for the larger scale installations as well. The steel insert is seen as a part that needs replacement after a certain period of operation time and is therefore made in such a way that it can easily be removed from the reactor.

A re-evaluation after some years of operational experience with the lab-scale and pilot-scale installation revealed that better alloys (higher creep strength at elevated temperatures and better corrosion resistance) are available at acceptable prices. These alloys will be used for future testing with the aim to increase the lifespan of the insert. The goal is to create an insert with a lifespan of several years.

BFB combustor & Riser gasifier:

The primary reason for selecting a BFB combustor was the idea that the temperature of a char particle during combustion needs to be limited as much as possible to reduce the risk of bed agglomeration. A BFB is better suited for this purpose than a riser because the bed density is higher, so more bed particles can absorb the heat from the burning char particles. The selection of a BFB as combustor automatically results in the selection of a riser for the gasifier, because vertical transport of the bed material is required.

The selection of a riser reactor for the gasification process has a positive effect on Cold Gas Efficiency compared to a BFB, because less dilution gas is introduced into the gasifier. Fluidization gas is required to fluidize the bottom part of the riser, not to create the velocity required for vertical transport of the bed material. The amount of required fluidization gas is mainly influenced by reactor area and this is much smaller for a riser than a BFB reactor. The velocity in the riser required for vertical transport of the bed material originates from the fact that the gas produced during the devolatilization of the biomass adds to the transport gas flow in the riser.

Number of biomass feeding points:

The number of biomass feeding points is set by the area of the riser were the biomass is introduced. An important characteristic of risers is that they require less feeding points than Bubbling Fluidized Beds, as the relatively smaller area and the increased height of the reactor give a homogeneous radial mixing. The design of the MILENA gasification concept is such that feeding from two opposite feeding points is both possible and practical.

An overview of atmospheric CFB combustion boilers given in [7] gives a largest bed area of 22.5 m^2 per feeding point with an average bed area of 5.3 m^2 per feeding point. CFB combustion processes are more sensitive to fuel distribution than gasification processes, because a local variation in air to fuel ratio can result in a high concentration of CO and unburned hydrocarbons. Assuming an area/feeding point ratio of 20 m^2 per feeding point, the MILENA technology can be scaled up to

900 MW_{th} under atmospheric conditions using only one feeding point. From a practical point of view two fuel feeding points are more logical, because biomass feeding systems can be relatively unreliable. By using two feedings systems at a design capacity of 70% of the total biomass flow the availability of the plant will increase drastically, because the operation of the gasifier can continue if one feeding line fails. The pilot plant is equipped with one feeding system, due to physical space limitations. The foreseen 10 MW_{th} demonstration plant will be equipped with two feeding systems.

Bed material:

Sand was selected as the design bed material for both the lab-scale and the pilot plant. This type of bed material was already in use by ECN in a lab-scale Fluidized Bed gasifier. This type of bed material shows no catalytic activity towards tar reduction or the water gas shift reaction. The gas cleaning technology (OLGA) foreseen can handle the high tar load.

The bed material properties influence the dimensioning of the reactor. Table 6.1 shows the main bed material properties and calculated values for minimum fluidization velocity and terminal velocity [8] for typical MILENA process conditions.

Table 6.1: Hydrodynamic bed material properties at atmospheric operating conditions.

Bed material	[-]	Sand	Olivine
Average particle diameter	[mm]	0.3	0.3
Particle density	[kg m^{-3}]	2600	3200
Bulk density	[kg m^{-3}]	1500	1700
Particle sphericity (ψ_p)	[-]	0.8	0.7
Gas pressure	[bar]	1	1
Min. fluidization velocity (u_{mf}) in flue gas at 900°C	[m s^{-1}]	0.05	0.05
Linear gas velocity BFB combustor bed/u_{mf}	[-]	10	10

Min. fluidization velocity (u_{mf}) in producer gas at 850°C	[m s^{-1}]	0.06	0.06
Terminal velocity (u_t) in producer gas at 850°C	[m s^{-1}]	2.2	2.4
Linear gas velocity riser - Terminal velocity particle (u_g - u_t)	[m s^{-1}]	3.8	3.6

Later tests in the lab-scale installation were done using olivine as bed material. The hydrodynamic properties are given in the right hand column. The hydrodynamic properties are close to the properties of the design bed material (sand) if the same size bed material is used. Olivine is catalytically active. It reduces the tar content of the producer gas and promotes the water gas shift reaction. A reduction in tar concentration in the producer gas was required to prevent clogging in the piping between the MILENA and OLGA at lab-scale. The pilot plant was also operated with olivine to reduce the risk of fouling problems in the producer gas cooler. The aim is to replace the olivine again by sand because this type of bed material is less expensive and the tests done so far have shown that the loss of bed material is lower when sand is used instead of olivine. The aim to reduce bed material loss is to some extent conflicting, because some dust in the producer gas is desired to prevent fouling problems in the gas cooler. The bed material particles that are blown out of the gasifier are relatively large (> 20 μm) and dense (typical 2600 kg m^{-3}) compared to the biomass char/ash particles which are entrained. These large particles sand blast the wall of the producer gas cooler and thereby prevent fouling.

Biomass residence time:

The MILENA concept is based on (almost) complete devolatilization of the particles before the remaining char is used in the combustor to generate the heat of the process. A simplified model was made to estimate the conversion and residence time of different fuel particles [9]. The required biomass residence time influences the dimensions of the riser.

The residence time of a biomass particle needs to be sufficient for almost complete devolatilization. The required residence time strongly depends on the geometry and size of the particle. Equation 6.1 gives an indication of the required residence time for spherical woody biomass as function of temperature (to be entered in Kelvin) and particle diameter (to be entered in meter). The formula was obtained from experiments in a BFB operated at similar temperatures as the MILENA riser using spherical wood particles [10].

$$\tau = 2.673(1000 d_p)^n \quad [\mathrm{s}], \qquad \text{where} \quad n = 1.02 e^{233/T} \tag{6.1}$$

Figure 6-2 depicts the relation for typical MILENA operating temperatures. As can be seen from the figure the operating temperature has only a small influence on the required residence time.

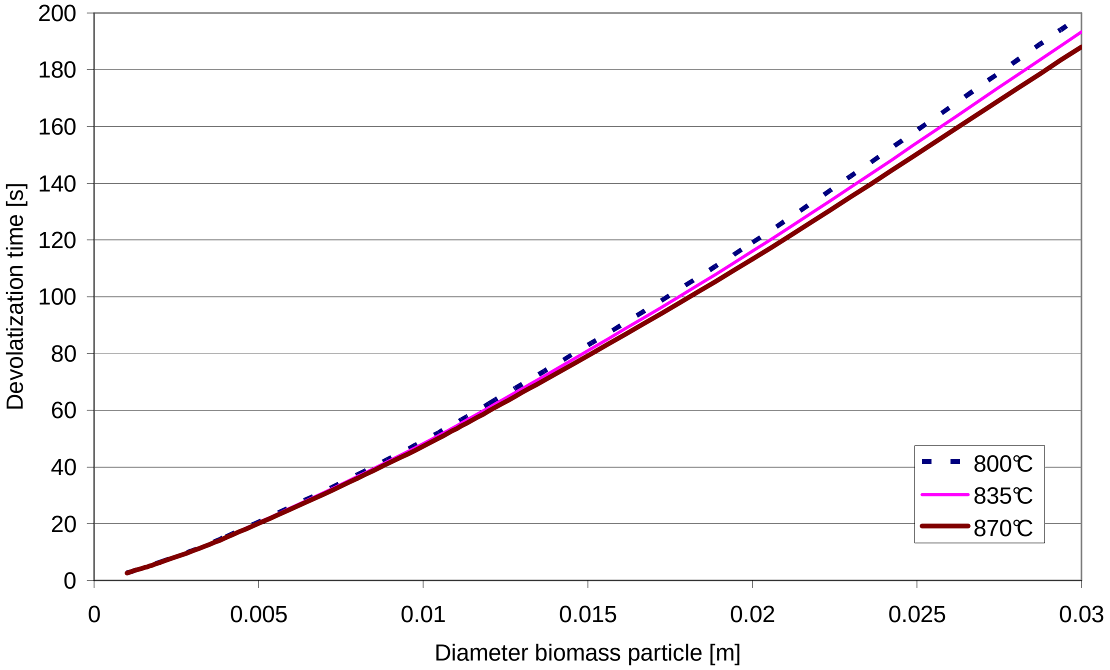

Figure 6-2: Calculated devolatization times assuming spherical particles.

Typical commercial size biomass chips have a smallest diameter or width of 0.01 m. These particles can be compared with spherical particles of the same diameter. Figure 6-2 shows that the required devolatilization time in the MILENA gasifier is approximately 50 seconds.

The devolatilization of the biomass particles takes place in the riser and the settling chamber. The riser is divided in two zones: a dense zone and a lean zone. A biomass particle starts its devolatilization in the dense zone of the riser. In this zone the conditions are similar to a highly expanded stationary Bubbling Fluidized Bed [11]. In the dense zone the biomass will rise with the same velocity as the bubbles in the Fluidized Bed [12]. In the lean zone the biomass particles will rise with the difference in linear gas velocity (u_g) and terminal velocity of the biomass particle (u_t).

The terminal velocity of a particle is the result from the equilibrium between gravity, buoyancy and drag forces. The gravity forces on a particle will reduce in time because of loss of mass. The buoyancy forces on a particle are mainly determined by the location in the riser. The density of the surrounding gas-bed material mixture reduces with height, thereby decreasing the buoyancy force. Therefore the terminal velocity of a biomass particle will decrease over time and will decrease when the particle is in a higher location in the riser. When the terminal velocity of the char particle has decreased below the linear gas velocity the particle will be blown out of the riser.

Figure 6-3 shows an example of results from modeling work done by Martin Horstink for the MILENA pilot plant using wood pellets as fuel [9]. The results show that the particle is close to complete devolatilization when it leaves the riser.

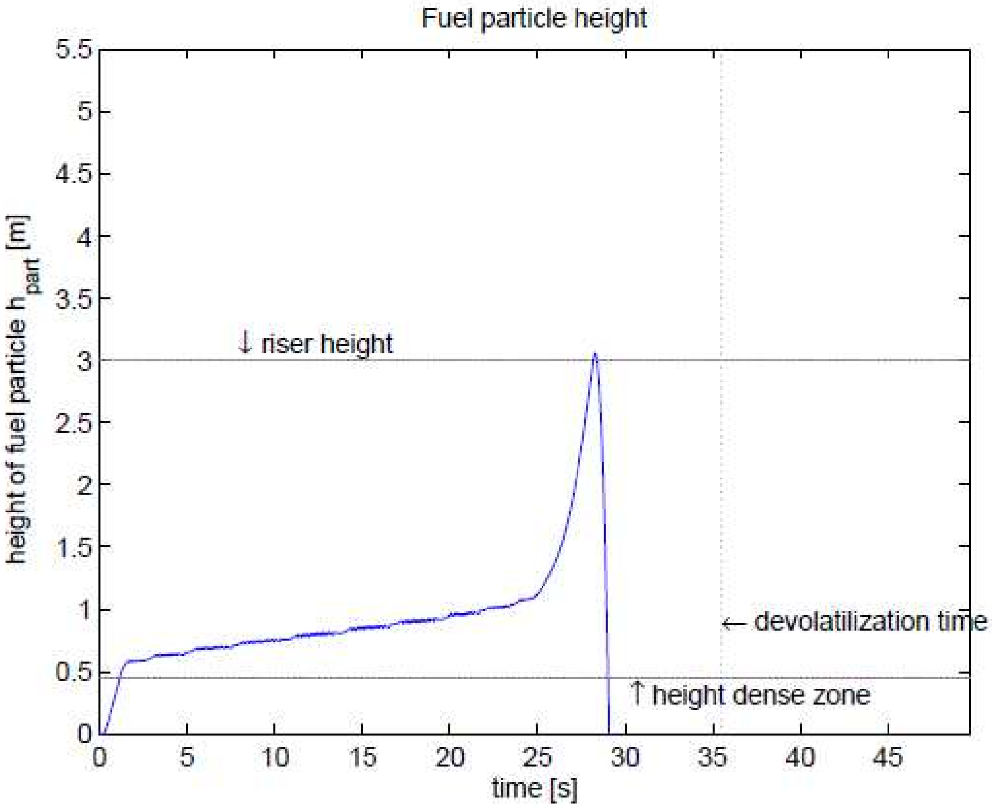

Figure 6-3: Example of fuel particle height profile in the pilot-scale.

Riser dimensions:

The riser diameter is determined by the terminal velocity (u_t) of the bed material and the calculated volume flow of the producer gas. The design gas velocity is the calculated terminal velocity plus a chosen off-set velocity. Table 6.1 shows the design values.

The required height of the riser is set by the required time for devolatilization of the biomass particles. The combined residence time of the biomass particles in the riser and the settling chamber needs to be sufficient to convert the biomass into gas and char. Background information on the biomass residence time is given in the previous section.

Settling chamber:

A settling chamber was selected instead of a cyclone for gas solid separation because a settling chamber increases the residence time of the producer gas and the contact time of the gas and the solids (char + bed material). An increased residence time has a positive effect on fuel conversion and possibly on tar conversion.

The disadvantage of a settling chamber over a cyclone is the lower collection efficiency of small particles (<0.1 mm). Small particles will be blown out of the gasifier and are collected by the cyclone that is normally placed before the gas cleaning. They are recycled to the combustor. The BFB combustor is designed such that the small particles will leave the system with the flue gas. A positive side effect of the low removal efficiency of a settling chamber for small particles is the increased dust load of the gas going through the producer gas cooler. The coarser particles in the gas (> 20 µm) are used to sand blast the cooler, thereby preventing fouling problems.

Downcomers:

In general a downcomer (or dipleg or standpipe) is used to transport solids against a pressure gradient, e.g. in a CFB where solids are transported from a low-pressure region (outlet cyclone) to a high-pressure region (bottom of the riser). In the MILENA gasifier the downcomer is used to transport solids between the settling chamber and the BFB combustor. To minimize gas leakages between combustor and settling chamber the downcomer is sealed in the fluidized bed. To prevent (large) bubbles from the fluidized bed entering the downcomer, the end of the downcomer is protected by a plate. From observations during cold flow tests we learned that the solids normally move down in moving bed or fluidized bed flow. Both flow regimes are acceptable for stable operation of the gasifier. The gas pressure at the outlet of the downcomer is during normal operation approximately 10 mbar higher than the inlet pressure (the pressure in the settling chamber). This results in an upwards flow of flue gas from the combustor relative to the downwards moving bed of bed material.

The diameter of the downcomer is set by the velocity of the bed material flowing down through the downcomer. A moving bed kind of flow was assumed. The design velocity is 0.1 m s^{-1} and the design solid circulation rate is 6000 kg h^{-1}

for the pilot plant. The downward velocity of the bed material is, in general, higher then the upward flow (relative to the bed material) of flue gas, resulting in a net downwards moving gas flow. A relatively small amount of producer gas leaks into the combustor bed at normal operation. The amount is influenced by the pressure difference over the downcomer.

The number of downcomers is set by the required fuel (char) distribution in the bed and the mechanical design of the ‘insert’. A symmetrical design reduces mechanical stresses. A local increase in fuel (char) concentration in the bubbling fluidized bed can result in emissions of CO and unburned hydrocarbons. Large scale MILENA gasifiers will benefit from multiple downcomers. The lab-scale installation is equipped with one downcomer. The pilot plant was equipped with two downcomers, but this was later reduced to one, because the concept of ‘multiple’ downcomers was proven and is not required at the relatively small scale of the pilot plant (or 10 MW_{th} demo plant.)

Combustor:

The combustor diameter is determined by the minimum fluidization velocity of the bed particles and the calculated volume flow of flue gas. The design velocity is 10 u_{mf} for the pilot plant. A higher ratio would result in a smaller reactor, but it would increase the loss of bed material.

The downcomer pipe must be submerged in the fluidized bed. Otherwise flue gas will leak into the settling chamber. A depth of 0.3 m was selected for the pilot plant to have an operational margin. The distance between the outlet of the downcomer and the top of the fluidization nozzles is 0.6 m. The total bed height from the top of the air nozzles to the top of the fluidized bed is 0.9 m for the pilot plant. The typical pressure drop gradient over a fluidized bed of sand is 10 kPa m^{-1}. The typical pressure drop over the fluidized bed of the pilot plant is approximately 9 kPa.

As a rule of thumb the pressure drop over the air nozzles must be approximately 1/3 of the pressure drop over the bed, to guarantee a good distribution of the air [8]. This results in typical exit velocities of approximately 40 – 60 m s^{-1}.

For the lab-scale installation a plate with small holes was used for air distribution. This is a typical solution for lab-scale fluidized beds, but is not applicable for commercial size installations because the required discharge of solids from the bed is not possible if a plate is used. Moreover, the holes are sensitive to plugging. The design of the air nozzles for the pilot-scale installation was copied from the BIVKIN gasifier. Several pipes with small holes were distributed over the fluidization area.

Height of Combustor Freeboard :

The height of the freeboard is defined as the height difference between the top of the fluidized bed and the flue gas exit. The freeboard above the fluidized bed serves two goals:

1. Post combustion chamber to reduce emissions of CO and C_xH_y. Secondary air is injected for this purpose.
2. Transport disengaging zone for the entrained solids (bed material) to minimize the loss of bed material.

The gas residence time in the freeboard is several seconds, which is more than sufficient for complete combustion of the gases at the typical freeboard conditions. The gas phase residence time requirements therefore do not set the limit for the required freeboard height. The freeboard height is defined by the required transport disengaging height (TDH). Solids are thrown from the bed by bursting bubbles rising from the bottom to the top of the bed. The solids thrown up into the freeboard contain the whole spectrum of particle sizes present in the bed. The larger particles (bed material) should fall back in the bed and the smaller particles (fly ash) are allowed to leave the bed. The TDH is defined as the height at which entrainment does not change appreciably. Several empirical relations are available to estimate the required transport disengaging height. The required height is mainly influenced by the minimum fluidization velocity of the smallest particles which are required to fall back in the bed and the linear gas velocity in the fluidized bed and freeboard. The size of the bubbles in the bed also influences the required TDH. The bubble size is influenced by the combustion air distribution design. For the design of the MILENA reactor the relations found by Amitin and Horio [13] were used to estimate the height. The estimated TDH for the MILENA pilot is ≈ 2.3 m, but this height could not be realized in practice due to height limitations in the building.

Bed material circulation rate:

The gasification/pyrolysis reactions in the riser are endothermic. The required heat is produced in the combustion reactor. The circulating bed material transfers the heat from the combustor to the riser. The required circulation rate of the bed material follows from the energy balance. The holes in the riser set the circulation rate. The circulation rate is influenced by the pressure difference over these holes. The pressure difference is normally controlled at a constant value. The diameter of the sand circulation holes is the main design parameter in setting the solids circulation rate (G_s). The diameter is empirically determined from cold flow tests and CFB gasifier tests. The solids circulation through the hole(s) is approximately 300 kg m^{-2} s^{-1}.

Table 6.2 summarizes the basic design data of the pilot plant and the lab-scale installation.

Table 6.2: Basic design data MILENA lab-scale & pilot plant.

		Lab-scale	Pilot plant
Thermal input (HHV basis)	[kW]	30	800
Biomass mass flow	[kg h^{-1}]	5 - 6	160
Steam to gasifier	[kg h^{-1}]	0.1 - 2	19
Riser diameter	[m]	0.036	0.2
Combustor (fluidized bed) diameter	[m]	0.25	0.8
Fluidized bed height	[m]	0.4	0.6

Overall reactor height	[m]	2	8
Number of downcomers	[-]	1	2
Combustor temperature	[°C]	925	925
Operating pressure	[bara]	1.1	1.1
Heat loss	[kW]	0	~30
Estimated circulation rate bed material	[kg h^{-1}]	150	6300
Producer gas volume flow wet	[Nm^3 h^{-1}]	6	174
Tar and BTX to combustor	[kW]	0	55
HHV gas wet basis excl. tar	[MJ Nm^{-3}]	13	13
HHV gas dry basis excl. tar	[MJ Nm^{-3}]	18	18

6.3 Description of cold-flow setup

The main goal of the cold flow model was to establish insight into the hydrodynamics (circulation of bed material) of the MILENA reactor. The cold flow model was basically used to visualize the hydrodynamics, but also tests were done to determine the solids recirculation rate. Figure 6-4 depicts the hydrodynamic principles of the MILENA process.

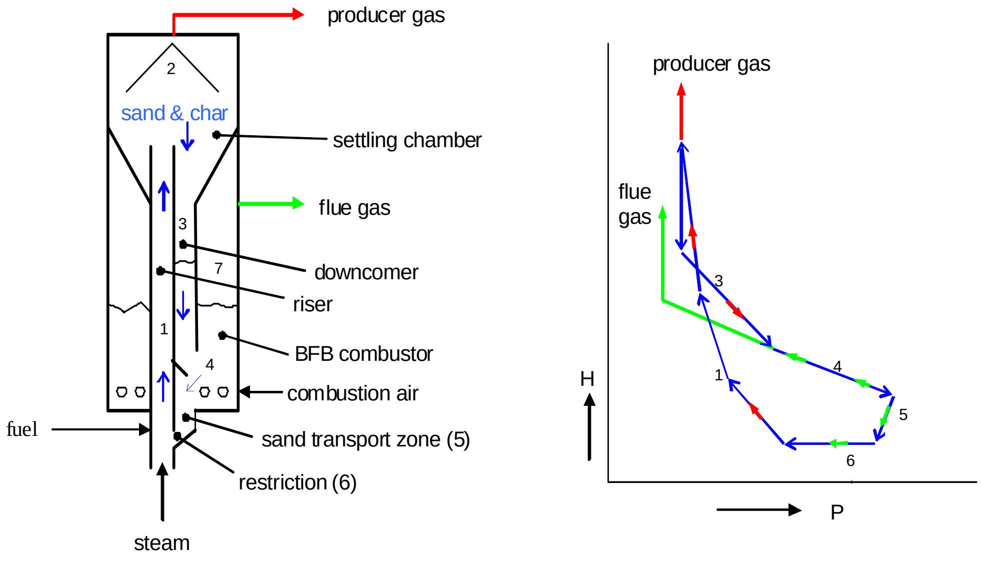

Figure 6-4: Schematic representation of solids circulation in MILENA gasifier.

The fluidization behavior of the system is completely determined by the bed particles, because the amount of char in the system is relatively low. The typical amount of char in the system is less than 1 mass%. This is a fraction of the amount of char that can build up in a CFB or BFB gasifier, because the circulating char is completely combusted in the combustor. The low char build up is beneficial from a hydrodynamic point of view.

The selected bed particles used in the MILENA gasifier can be classified as group B particles according the Geldart classification system [8].

Table 6.3 gives a brief overview of the functions, hydrodynamic behavior and boundary conditions for the different parts of the MILENA gasifier. A more detailed explanation is given below.

The hydrodynamic principles of the MILENA gasifier are explained by starting at the producer gas outlet of the system. The pressure in the settling chamber (2) is determined by the pressure drop of the downstream equipment (gas cooler, gas cleaning, etc.). In the MILENA lab-scale and pilot plant the typical overpressure in the settling chamber is 50 mbar. The solids in the settling chamber drop down into the downcomer (3). The solids flow into the Bubbling Fluidized Bed combustor (4) where the pressure is higher. The pressure increase is overcome by gravity. The flue gas produced in the BFB combustor flows into the freeboard (7) of the BFB and exits the installation. The pressure in the freeboard is controlled by varying the resistance of the flue gas exit. This is done by a (manually operated) valve. Normally the set point for the freeboard pressure is set to the pressure of the settling chamber +5 to +15 mbar. The pressure difference is kept small to minimize the leakage of gas through the downcomer.

Table 6.3: Overview of functions, preferred modes of operations and boundary conditions.

Part	Function	Preferred mode of hydrodynamic operation	Boundary conditions
Riser (1)	Solids transport. Mixing gas and solids. Heating of biomass (drying and pyrolysis)	Fast fluidized bed	Solids transport not restricted by riser throughput. Velocity > u_t
Settling chamber (2)	Separating char and bed material from producer gas. Further degasification of biomass.	-	Velocities far below terminal velocities of bed particles.
Downcomer (3)	Solids transport. Gas barrier between combustor and settling chamber.	Moving bed Bubbling fluidized bed	Solids transport capacity not limiting solids circulation. No/limited gas transport from combustor to settling chamber → downward velocity of solids higher than upward velocity of gas.
Combustor (4)	Combustion of char Heating of circulating sand Separating ash	Bubbling fluidized bed	Stoichiometric air-fuel ratio approx. 1. Superficial gas velocities > 3 u_{mf} and $<u_t$
Transport - zone (5)	Transporting sand from combustor to opening in riser (restriction).	Moving bed	
Restriction (6)	Restricting circulation rate.	Moving bed	Solids circulation capacity between 30 - 80 times the biomass throughput.
Freeboard (7)	Disengagement of bed material	-	Superficial gas velocity < U_t Height > TDH

Under normal operating conditions the gas pressure at the bottom end of the downcomer is slightly higher than the pressure in the settling chamber, resulting in an upwards gas flow relative to the solids. The solid particles in the downcomer move down with a vertical velocity of approximately 0.1 m s^{-1}. The net gas flow through the downcomer is minimized by controlling the pressure in the freeboard. A small downward gas flow from the settling chamber into the combustor is preferred, to prevent dilution of the producer with nitrogen from the flue gas.

The solids in the BFB (4) move downward into the sand transport-zone (5) by gravity. The gas pressure is highest just above the nozzles where the combustion air is injected. The gas pressure decreases again in the direction of the restriction (6). Sand moves from the transport zone through a restriction/hole (6) into the riser. The opening controls the sand circulation flow. Tests were done in the cold-flow to determine the required diameter of the opening to obtain a sand circulation rate of approximately 40 times the biomass input on mass basis. The mass ratio of 40 is required to have a relatively low temperature difference of approximately 60°C between combustor and gasifier. A lower circulation rate increases the temperature difference between combustor and riser which has a negative influence on the Cold Gas Efficiency. The pilot plant was used to verify the selected restrictions.

The riser is operated in the fast fluidization regime. This type of operation results in a typical low solids density compared to a fluidized bed, leading to a relatively low pressure drop. The difference in pressure between the riser and the BFB combustor is the driving force for the solids circulation. The superficial velocity in the riser (U_r) is defined as the volume flow of gas created by the gasification of biomass plus the amount of steam added divided by the area of the riser. The bulk of the gas volume is due to the degasification of the biomass.

The cold flow model is made of glass to be able to visualize the hydrodynamic process. Figure 6-5 depicts the cold-flow setup and shows the main dimensions. A pre-design of a lab-scale MILENA reactor was made before the cold flow model was designed. The size of the cold flow setup is similar to the lab-scale setup which was constructed several years later.

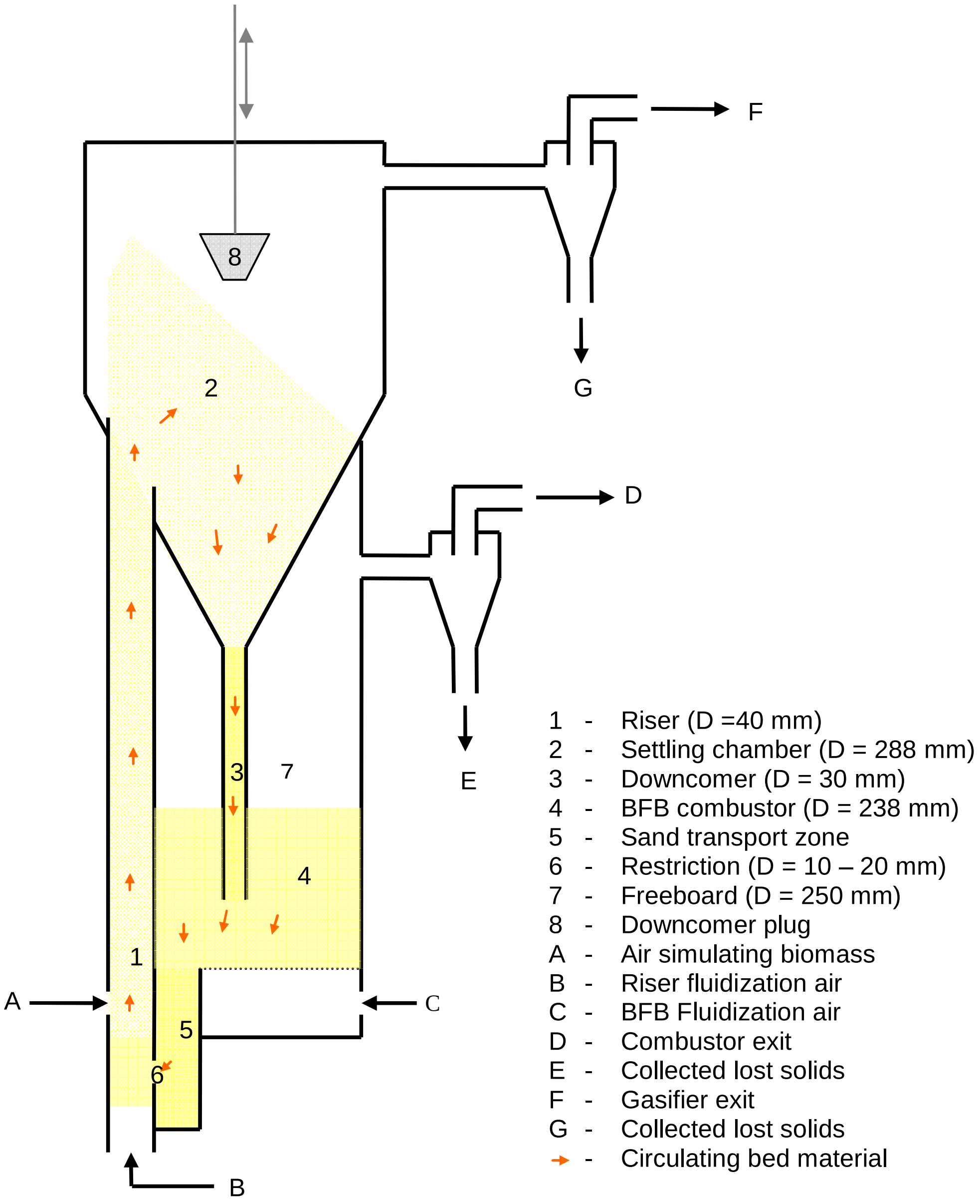

Figure 6-5: MILENA Cold Flow Set-up.

The solids circulation rate is influenced by the diameter of the hole in the riser (the restriction). Several tests were done to establish an applicable relation for the design of the lab-scale MILENA and further scale up. The bottom part of the riser was made exchangeable to vary the dimensions of the hole. Three different sizes were tested to see how this influences the solids circulation rate. The solids circulation rate was measured by closing the inlet of the downcomer and measuring the increase in height of the solids in the funnel.

Most Cold Flow experiments were done using quartz sand and olivine. The main properties of these bed materials are given in Table 6.1. The size distribution of olivine varied to some extent. This material was ordered from different suppliers, but also batches from the same supplier varied sometimes. The diameter of the fluidized bed was chosen such that operation at approximately 5 - 8 * u_{mf} is possible. The riser was dimensioned to operate approximately 3 m s^{-1} above the terminal velocity of the average bed material particles. The minimum vertical velocity in the settling chamber was 0.1 m s^{-1}. This is low enough to separate char particles with a diameter of more than 0.2 mm from the gas.

6.4 Lab-scale installation

The design of the lab-scale MILENA configuration was based on a preliminary design made for the pilot plant and the cold flow model. The cold flow was used to measure the bed material circulation rate for different configurations and dimensions. The original design capacity of the installation was 5 kg h^{-1} of wood, because this was the limit for lab-scale installations at that time. Installations with capacities above this scale required elaborate safety measures, which would increase the costs. A smaller scale was seen as less realistic from hydrodynamics point of view.

The main dimensions of the lab-scale installation compared to the pilot plant are given in Table 6.2.

The lab-scale gasifier is coupled to a lab-scale gas cleaning installation and a methanation unit. The entire system operates at atmospheric pressure. Figure 6-6 shows a simplified scheme of the integrated lab-scale setup (including gas cleaning and methanation units).

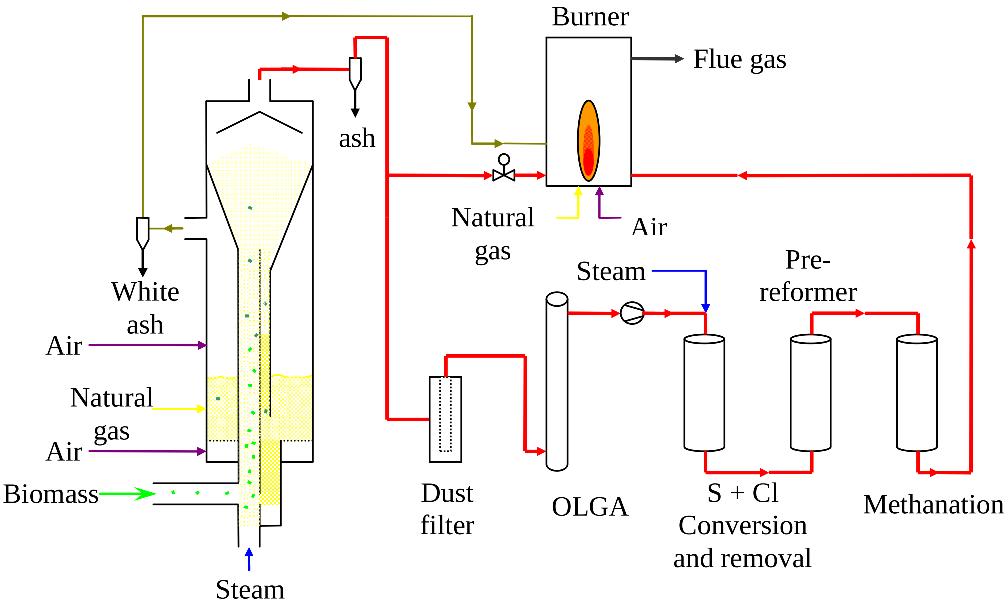

Figure 6-6: Basic layout of MILENA lab-scale installation.

Most of the gas cleaning equipment (hot gas filters and OLGA tar removal) was originally designed to be used with the lab-scale Fluidized Bed gasifier WOB [14]. The typical producer gas flow from this gasifier was 1 – 2 $Nm^3 h^{-1}$. The producer gas flow from the MILENA is 6 – 7 $Nm^3 h^{-1}$, only part of the gas produced can be used in the gas cleaning system. Most of the gas is directly combusted in a burner. A high temperature valve in front of the burner regulates the gas split between the burner and the gas cleaning system. Because of the small scale, the heat loss from the pipes is relatively high and electrical heating is required to keep the temperature of the gas above the tar dew point. The dust is removed from the gas by high temperature porous candle filters. Typical operating temperature is between 400 and 450°C**.** Metal and ceramic filters were both applied successfully. The filters were cleaned off-line. The removal of dust using high temperature gas filters is seen as a lab-scale solution. In the pilot plant the coarse dust particles are removed by a cyclone and the fine dust particles are removed in the OLGA unit. This solution is more economic, but less practical at lab-scale. The dust-free gas is sent to the OLGA unit for tar removal. The OLGA technology is based on scrubbing the producer gas with oil [15].

The cleaned gas is sent to the chloride and sulphur conversion and removal unit. Higher hydrocarbons like C_6H_6 are converted into CO, CO_2, H_2, H_2O and CH_4 in a catalytic reactor (the pre-reformer in Figure 6-6). After this process step the gas is ready to be compressed and converted into Bio-SNG in commercial processes. In the lab-scale installation commercial catalysts are used at atmospheric pressure to convert the gas into CH_4, CO_2 and H_2O. The gas flow through the reactors is regulated by a small booster. A steam generator is installed to increase the moisture content of the gas before the catalytic processes. Steam is required to prevent soot formation. The different process steps will be explained in more detail in the next paragraphs.

Figure 6-7 shows the MILENA lab-scale gasifier (left) and the OLGA tar removal system (right). The two bunkers on the left are used to feed the fuel (small wood particles) to the gasifier.

Figure 6-7: Photos of lab-scale MILENA (left) and OLGA (right) installation.

6.4.1 Gasifier

Figure 6-8 shows a scheme of the lab-scale gasifier. The MILENA lab-scale gasifier installation has been used for more than 2000 hours.

The feeding system is the most sensitive part of the complete installation. Most problems with the gasifier were a direct or indirect result of problems with the feeding system. The biomass is fed into the gasifier from one of the two fuel bunkers. The bunkers are also used to control the feed rate by changing the rotation frequency of the discharge screw. Several discharge screws are available for different types of fuels. The feeding bunkers are calibrated before a test by measuring the weight loss of the bunkers over a certain time at a set rotation frequency. After a test the weight loss of the bunkers is measured again to check the correct fuel flow. N_2 was used as a purge gas during the first experiments. This was later changed into CO_2 to minimize the nitrogen dilution of the producer gas.

The fuel bunkers are directly coupled to the gasifier and are operated at the same pressure as the riser. A small N_2 purge flow is used to keep producer gas out of the feeding bunkers. For refueling the feeding bunker is stopped while the other feeding bunker is started. The valve at the bunker outlet is closed. The lid is opened and fresh biomass is added. The flanges of the lid are carefully cleaned and closed. The pressure in the bunker is increased to operating pressure by adding nitrogen after which the feeding bunker is ready to be taken into operation again. Switching of the feeding bunkers is done automatically after a certain running time. The filling of the bunkers is done manually. This was also automated, but the feeding system appeared not reliable enough for duration tests. The two bunkers can operate for approximately 10 hours before an operator refills both bunkers. This procedure was developed over time and proved to be reliable enough for running duration tests of typically 100 – 200 hours.

Because of the small size of the feeding screw the size of the biomass particles is limited to several millimeters. The most left sample shown in Figure 7.1 (Chapter 7) gives an impression of the physical dimensions of the biomass particles used for tests in the lab-scale installation.

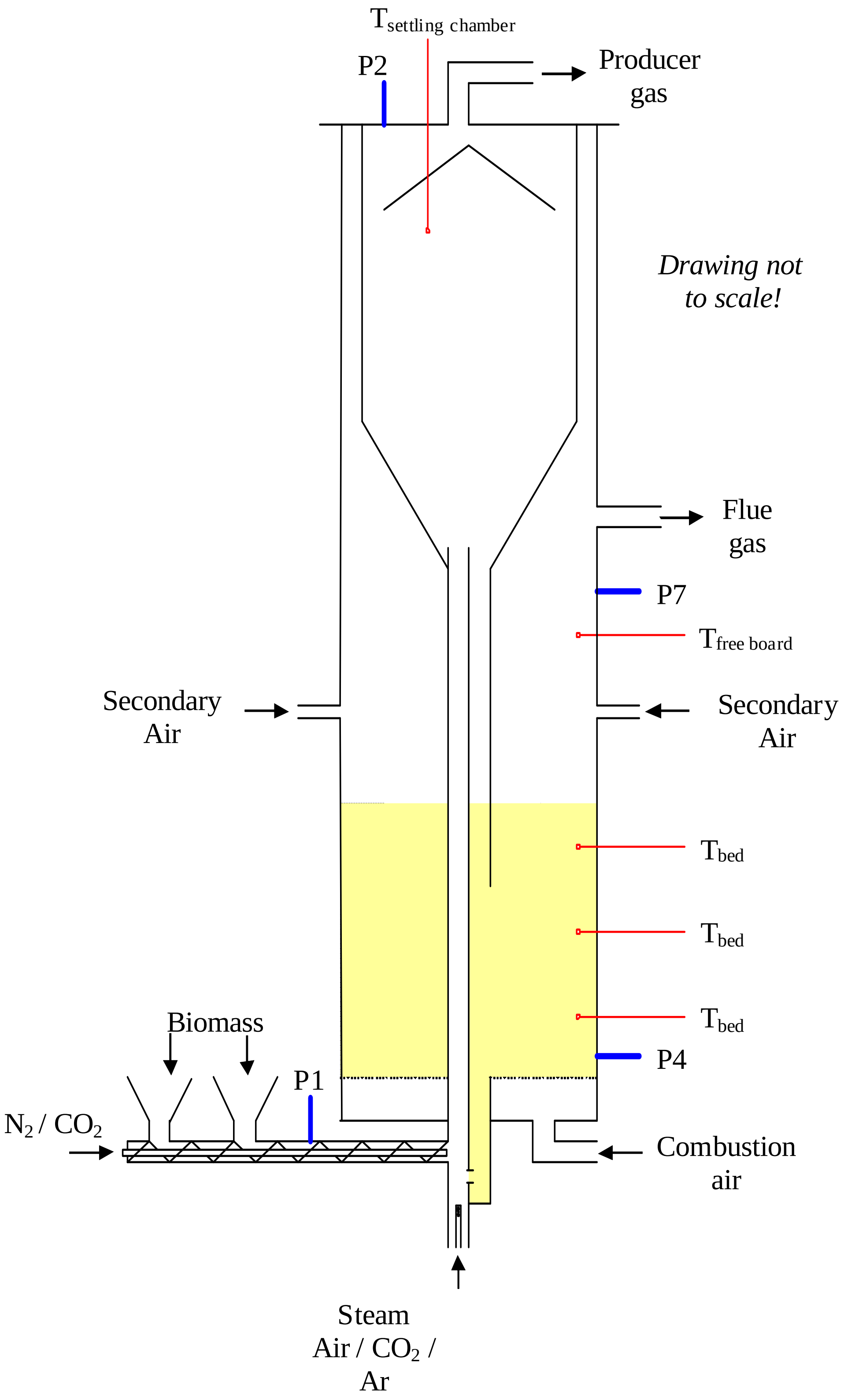

Figure 6-8: MILENA lab-scale gasifier.

The feeding screw transports the fuel into the riser. The feeding screw is not cooled, some drying and pyrolysis may occur in the feeding screw, which so far did not lead to operational problems. A thermocouple on the outside of the screw is used to monitor gas leakage from the gasifier into the feeding system. A gas leakage through the feeding screw results in an increase of the screw temperature. A small leakage in the feeding system is a very common problem, which needs to be solved directly to prevent wetting of the fuel due to condensation of water coming from the producer gas. The moisture on the fuel can make the fuel sticky, which gives feeding problems.

The lab-scale MILENA gasifier was built in 2004. All engineering and construction were done by the Engineering & Service department of ECN. The lab-scale reactor vessel and insert are made of stainless steel (grade 253MA). This type of steel can withstand temperatures of up to 1100°C and is available in the required pipe sizes. Heat loss from the process is compensated for by high temperature electrical trace heating. The heat loss is reduced by external insulation.

The operating pressure is limited to 0.4 bar overpressure, to stay below the 0.5 bar overpressure limit set in the Pressure Equipment Directive (PED 97/23/EC). A higher pressure would significantly increase the costs, because of the required safety measures.

The riser is fluidized with steam. The amount of fluidization steam can be varied between 0.1 and 2 kg h^{-1}. The amount of steam required to fluidize the riser is low (0.1 kg h^{-1}), but additional steam is used to increase the water content of the producer gas because the biomass used for lab-scale experiments is relatively dry (10 wt.% moisture), whereas the fuel foreseen for commercial applications contains more moisture (25 wt.%).

The temperature of the riser and the reactor will differ, especially during heating. The riser can move freely through the bottom of the reactor while gas leakage is minimized by a seal. This construction minimizes mechanical stresses due to differences in expansion. This solution makes it possible to use a steel construction at temperatures at which the strength of the material is relatively low.

6.4.2 Flue gas cooling and dust removal

Flue gas from the combustion section is partially cooled by heat loss. Approximately 90% of the dust/fly ash is removed by a cyclone. Most of the smaller particles (e.g. aerosols of salts) will probably stay in the gas. The temperature of the cyclone is not controlled or measured. Because of the simple layout of the flue gas treatment line the collected ash samples are not representative for commercial scale installations. Pilot scale tests are required to produce ash samples under more realistic conditions. The flue gas composition is measured before the gas is sent to the stack.

6.4.3 Producer gas cooling and dust removal

A cyclone located directly after the gasifier is used to remove approximately 90% of the carbon containing dust from the producer gas. In order to be able to make the carbon balance over the gasifier, the carbon content of the collected ash is measured. The cyclone was not always present in the system layout, because the cyclone caused operational problems. The relatively large flanges of the cyclone caused local cold spots, resulting in fouling problems (tar condensation). Most of the duration tests were done without cyclone to prevent fouling problems in the gas tubes. The amount of lost carbon had to be estimated from previous tests under similar operating conditions.

Cooling of gas is achieved by heat loss. The relatively small flow and large wall area of the piping result in a relatively high heat loss, this makes a separate gas cooler unnecessary. The wall temperature of the tubes is temperature controlled by electrical trace-heating. The typical wall temperature of the tubes with the tar loaded gas is kept at 450°C. Keeping all the parts of the piping at this temperature proved to be very important. Defective trace-heating and mistakes in the layout of the piping resulted in blockages by tar and dust leading to many shutdowns of the system.

Approximately $^2/_3$ of the producer gas is sent directly to the flare. The remaining gas is sent to the gas cleaning test rig. The gas cleaning test rig was designed for a smaller capacity gasifier.

6.4.4 Producer gas cleaning

In the producer gas cleaning section the main impurities in the gas are removed and converted to meet the stringent demands of the methanation catalysts. Tars are removed by the OLGA tar removal unit. Chloride and sulphur are removed by adsorbents.

The OLGA tar removal technology is based on scrubbing the producer gas with oil [15]. The tars are removed in two stages (only one shown in Figure 6-6). In the first stage (collector column) the producer gas is cooled and the condensed tars are collected. A bleed stream of the oil–tar mixture is available for use as fuel in the combustor of the MILENA gasifier. In the lab-scale installation this tar-fuel is simulated by methane, because feeding oil/tar in low quantities (typically 100 – 200 grams h^{-1}) proved to be very cumbersome. In the second stage (absorber column) the light tars are removed from the producer gas. The scrubbing oil from the second stage is regenerated in a stripper. The stripper uses air to strip the light tars from the oil. The air containing light tars can be used as combustion air in the MILENA combustor. In the present system layout, the air is used as combustion air in the flare for combusting the cleaned gas. The temperature in the OLGA gas cleaning is kept above the dew point of the water in the gas (typically 74°C) to prevent condensation of water.

The typical concentration of sulphur compounds in the gas is between 100 and 200 ppm for clean wood as a fuel. Most of the sulphur is present in the gas as

H_2S, but also as COS and thiophenes. Sulphur needs to be removed from the gas to a concentration below 1 ppm.

A fixed bed reactor filled with hydrodesulphurization (HDS) catalysts converting the thiophenes in H_2S. Hydrodesulphurization is a well known process in the oil industry, but the producer gas conditions are very different. The conversion of thiophenes in the producer gas was major research topic during several years, but is not a topic in this thesis. Results from this research are not publicly available. It was decided to keep this part of the system confidential to protect the IP position of ECN. H_2S, COS and HCl are removed from the gas by commercially available sorbents (e.g. ZnO).

The producer gas pressure is slightly increased by a blower in order to compensate for the pressure losses over the downstream reactors. At the moment the pressure is kept below 400 mbar for safety reasons. This pressure will probably be increased in the future.

Conventional methanation processes were not developed to handle hydrocarbons like C_2H_2, C_2H_4 and C_6H_6. Therefore these hydrocarbons are converted into a mixture of CO, CO_2, H_2, H_2O and CH_4 in an adiabatic catalytic reactor operating at approximately 550°C (the pre-reformer). Steam is added to prevent soot formation on the catalyst surface.

6.4.5 Methanation

The methanation test rig consists of three fixed bed reactors placed in series. The reactors are filled with commercial catalysts and operated at atmospheric pressure. The methanation reactions are exothermic. The reactors are operated adiabatically. The gas is cooled between each reactor to lower the methanation temperature. The last reactor is operated at a typical temperature of 270°C.

The produced gas consists of CH_4, CO_2, H_2O and a low concentration of H_2. To bring the gas on specification the H_2O and CO_2 need to be removed as it is being done in conventional processes.

The methanation of the clean producer gas is not seen as a new development for this purpose. The test rig is used to see whether the gas is clean enough and to demonstrate the concept. On a commercial scale the methanation will be done at increased pressure (e.g. 20 bar), because pressure favors the equilibrium toward CH_4 and the end product is mostly required at elevated pressures.

6.5 Pilot plant

The MILENA plant replaced the old ECN CFB gasifier BIVKIN. The existing infrastructure, feeding system, producer gas cooler and gas cleaning were re-used for the MILENA pilot plant. Figure 6-9 shows the basic layout of the pilot plant.

Three different fuel bunkers are available for feeding the different biomass feedstocks. This makes it possible to use fuel mixtures (e.g. sewage sludge with wood) or to switch from one fuel to another. Nitrogen is used as purge gas.

The gasifier has a flue gas outlet and a producer gas outlet. The flue gas is cooled down to approximately 200°C. Part of the heat is used to pre-heat the combustion air up to a value of typically 340°C. Dust is removed from the flue gas by a conventional bag-house filter. An isokinetic sampling point is installed after the bag-house filter for emission measurements. Samples from the bag-house filter solids outlet are taken on a regular basis to monitor the fly-ash quality. The carbon content of these ashes is always low (<1 wt.%), this indicates that the overall fuel conversion is complete.

The producer gas is cooled in a double tube cooler. The heat is used to pre-heat air. The preheated air is used as combustion air in the boiler. The cooled producer gas is sent to the gas cleaning test rig which is located outside the building. A cyclone removes most of the dust (ash, carbon and lost bed material) from the gas.

The producer gas at a temperature between 350 and 400°C, containing some small dust particles (typical 1 – 2 g Nm^{-3}), is cooled and cleaned in the OLGA gas cleaning test rig. Tar is removed from the gas. The heavy tars, containing some small dust particles are returned to the combustor section of the MILENA. During the first tests the tar recycle system was not operational and the recycle of heavy tars was simulated by adding natural gas to the bottom of the fluidized bed combustor. The light tars are sent to the boiler.

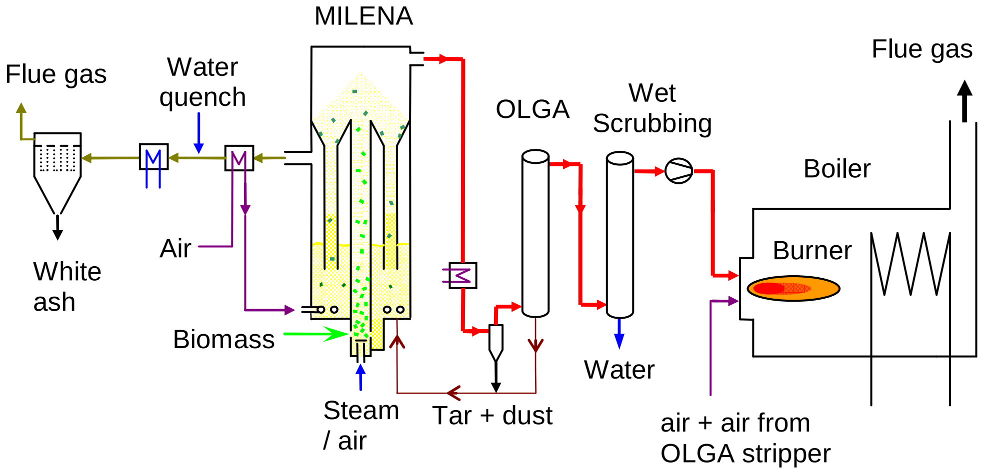

Figure 6-9: Basic layout of pilot plant

The producer gas exits the OLGA at a temperature of 80 – 90°C. The gas is further cooled in the wet scrubbing system down to approximately 35°C (strongly depending on ambient conditions). Most of the water in the gas is condensed out.

The pressure of the gas is increased by a booster to approximately 75 mbar. This pressure was required in the past to operate the gas engine on the gas from the previous BIVKIN gasifier. The gas engine is removed because this is not a topic of research anymore. Gas engines can be operated on producer gas if the calorific value is high enough and the tar dew point is low enough [16]. During the test with the previous gasifier the gas was also used in a gas turbine to prove that the gas is clean enough for this application as well [17].

The clean and nearly dry prodcuer gas is fired in a boiler. The heat is used to heat up water. At the exit of the boiler an isokinetic sampling point is installed for emission measurements.

6.5.1 Biomass feeding system

Biomass feeding problems are probably the main reasons for unplanned shut downs of biomass gasification plants. The tests described in this thesis were no exception. Problems can be minimized by setting stringent limitations on allowable particle dimensions. For small batches such stringent limitations are achievable, but for the MILENA pilot plant the typical fuel batch for tests was 15 tons. The fuel batches were pre-treated with the same equipments which is being used for commercial size biomass boilers. These batches also contained large particles exceeding the specifications, which made feeding challenging.

The MILENA gasifier requires a relatively stable biomass feeding flow, because the velocity in the riser depends directly on the fuel flow. A sudden drop in feeding rate results in a drop in gas velocity. If the velocity in the riser drops below the terminal velocity of the bed material the circulation of the bed material will stop. This will stop the heat transfer from the combustor into the riser.

The gasifier is directly coupled to the gas cleaning system, so the variations in fuel flow will result in pressure fluctuations in the entire system. For the tests done in the past with the integrated BIVKIN OLGA system wood pellets were used as the standard fuel [16]. Wood pellets are easy to feed into a gasifier and the feed flow is relatively stable, but wood pellets are not an option for a commercial gasification system because they are far too expensive. The development of the MILENA gasifier focuses on less expensive fuels. For the first 10 MW_{th} MILENA demonstration plant, demolition wood was selected. Demolition wood is irregularly shaped, so a lot of effort was spent to obtain stable biomass feed flow, to prevent solid circulation problems, and to minimize pressure fluctuation in the gas cleaning. Figure 7.1 in Chapter 7 shows the demolition wood fed to the MILENA pilot plant and the wood pellets used in the past and the first operational tests of the MILENA – OLGA pilot system.

For the tests described in this thesis open feeding systems were used. This means they can be re-filled during operation. The feeding bunkers are separated from the gasifier by a rotary valve. Nitrogen is used to purge the feeding screw and the rotary valve. Some of the nitrogen will end up in the gasifier and some will be lost in the rotary valve. The weight of the feeding bunkers is registered by load cells, which makes it possible to determine the exact fuel flow during the tests.

The gas leakage from the feeding screw is determined by the pressure in the feeding screw and the wear of the rotary valve. Leakage of producer gas is minimized by using purge gas (nitrogen). The gas leakage is monitored by temperature measurement. If the temperature of the feeding screw increases, the leakage from the gasifier into the rotary valve is increased and measures need to be taken to solve or reduce the leakage. Gas leakage in the feeding system is a major issue. The rotary valve requires a lot of maintenance to keep the leaking rate at an acceptable level.

6.5.2 Gasifier

The design for the MILENA gasification technology was originally done at a scale of 10 MW_{th}. The original foreseen application of the technology was producing gas for a gas engine. 10 MW_{th} biomass input is seen as attractive commercial scale.
The pilot plant design is a downscaling of the 10 MW_{th} design.

The MILENA pilot plant replaced the 500 kW_{th} CFB gasifier BIVKIN [16]. The BIVKIN gasifier was connected to a producer gas cooler, OLGA tar removal system, wet scrubbers for NH_3, HCl and water removal, a gas engine and a boiler. The gas cleaning system and the producer gas cooler are dimensioned on volume flow basis. Because the producer gas cooler and the gas cleaning system had to be used for the MILENA gasifier as well, the volume flow was fixed. The volume flow from the BIVKIN gasifier was approximately 190 $Nm^3\ h^{-1}$. The design volume flow for the MILENA was chosen to be 10% lower, to reduce the required biomass input and heat output from the boiler. Because of the increase in heating value of the gas from the MILENA gasifier compared to the BIVKIN gasifier, the thermal input increased from 500 kW_{th} to 800 kW_{th}, which corresponds with a fuel flow of 158 kg h^{-1} in case of dry wood.

The design pressure of the reactor vessel is 0.4 bar overpressure. The design pressure was chosen to be below the 0.5 bar overpressure limit set in the Pressure Equipment Directive (PED 97/23/EC) to prevent additional costs for safety checks. The reactor concept is in principle suited for pressurized operation of up to approximately 7 bar, but this would increase cost because of the additional safety measures required for pressurized vessels filled with a high temperature toxic and inflammable gas.

Figure 6-10 shows the basic concept of the MILENA pilot reactor. The drawing is not to scale, but Table 6.2 gives the basic dimensions of the reactor.

The inside of the outer reactor wall is refractory lined to reduce heat loss and keep the reactor wall at an acceptably low temperature. The inner layer is made of an attrition resistant material, which is suitable for typical fluidized bed conditions. The outer layer has good insulation properties. Refractory lining is very common for commercial scale gasifiers and is also foreseen to be used for further scale up. The reactor wall is externally insulated with a thin layer of insulation wool to further reduce the heat loss.

Start-up procedure:

The refractory lining requires a gradual heating of the reactor vessel (50 °C h^{-1}). This is accomplished by natural gas burners that are placed in the secondary air ports above the fluidized bed combustor. The burners are positioned such that the flames point downwards to the top of the fluidized bed. The flue gas exiting the combustion section passes through a heat-exchanger which preheats the fluidization/combustion air. When the bed temperature is high enough (T>650°C), natural gas is fed to the bottom of the fluidized bed to realize a further increase of the bed temperature. The heat transfer to the metal insert is relatively high, so no special measures are required to heat up the insert.

The installation is normally kept in natural gas combustion mode over night. The typical operating temperature is between 800 and 900°C, such that the refractory stays at a constant temperature. The installation is further heated in biomass combustion mode. A large air flow (typically 140 $Nm^3\ h^{-1}$) and a small biomass flow are added to the riser (typically 12 $kg\ h^{-1}$) to start circulation of bed material and produce hot flue gas for heating the producer gas cooler and the piping. After one or two hours of biomass combustion the installation is switched to gasification mode by decreasing the amount of air to the riser and increasing the fuel flow. The produced gas is ignited in the boiler.

Temperature measurement:

Type K thermocouples are used for all temperature measurements. The most relevant temperature is the gasification temperature. In most gasifiers this temperature is measured at several locations in the riser. Due to the mechanical construction of the MILENA reactor, it is not possible to place thermocouples in the riser. Therefore, the gasification temperature is measured at the outlet of the riser in the settling chamber ($T_{\text{settling chamber}}$). The process temperature normally decreases with height in a riser reactor that is used for gasification processes, because the process is endothermic and the heat is supplied to the process at the bottom of the reactor. The average gasification temperature is between the combustion bed temperature and the settling chamber temperature.

The temperature in the fluidized bed combustor is measured by several thermocouples, evenly distributed over the height of the bed. During normal operation the measured differences in temperature are small (<20°C). Large temperature deviations are an indication of fluidization problems, which are normally caused by bed agglomeration. The upper thermocouple is used as an indication of the bed height. Fast variations in temperature indicate that the bed height has dropped below the location of the specific thermocouple.

Pressure measurement:

Pressure measurement points are numbered as shown in Figure 6-10. The pressure difference between the freeboard (P7) and the settling chamber (P2) is the most relevant pressure control parameter. During normal operation this pressure difference is kept close to -15 and +15 mbar by variation of the opening position of a valve in the flue gas line (see paragraph 6.3 for more information about this control parameter).

Flow measurement:

The amount of combustion air is measured with relatively accurate devices, because the combustion air flow is one of the main parameters in the overall balance over the gasifier. In the lab-scale installation a mass flow meter is used with a typical accuracy of 1% at the typical operating range. In the pilot plant a mass flow meter with a typical accuracy of 2% at the typical operating range is used.

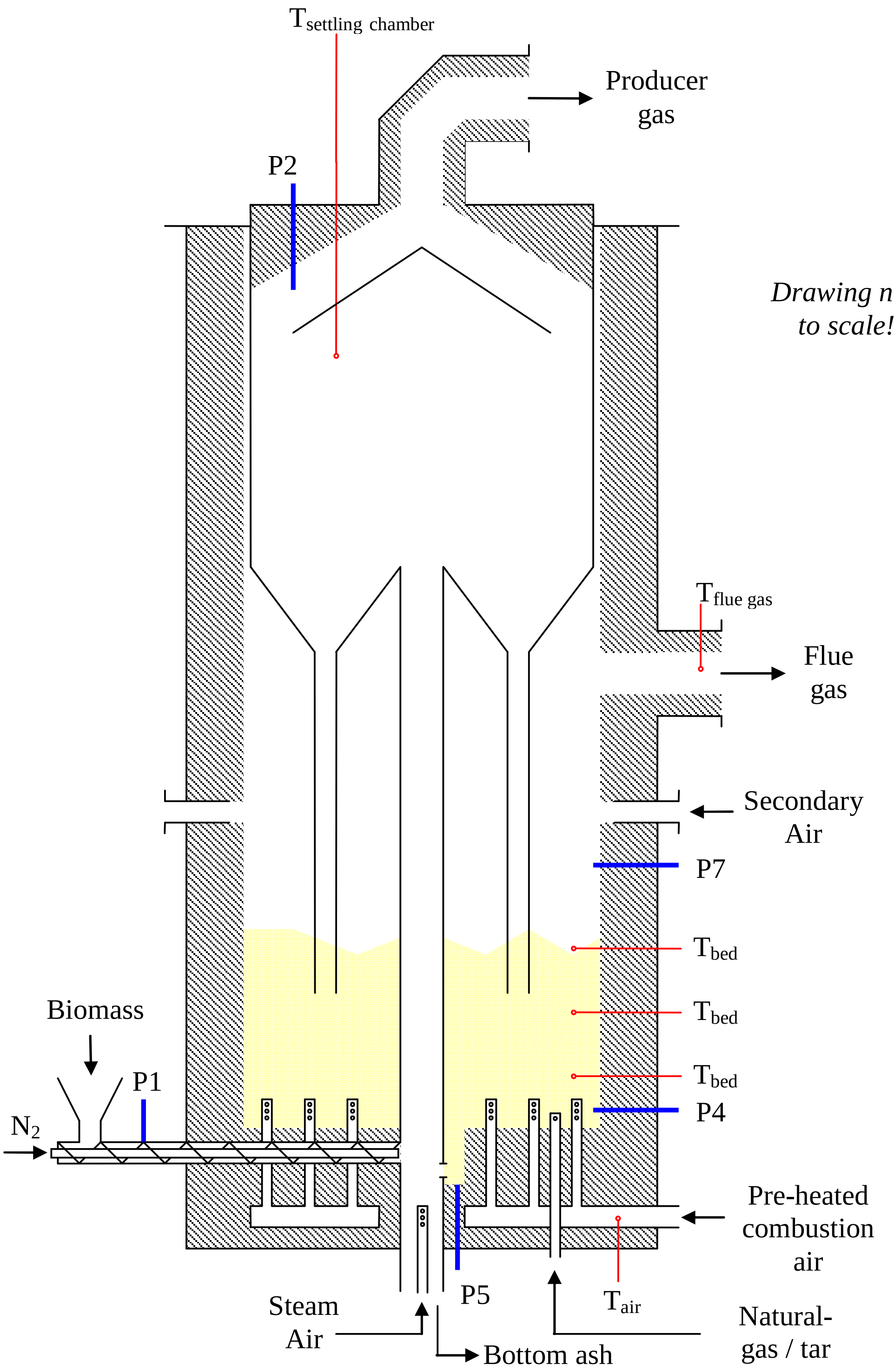

Figure 6-10: MILENA pilot-scale reactor

6.5.3 Flue gas cooling and dust removal

The flue gas is cooled to approximately 500°C in the first cooler (single tube cooler). The heat is used to pre-heat the combustion air for the combustor. A second air cooled heat exchanger was installed to reduce the flue gas temperature from approximately 500°C down to the bag-house filter inlet temperature, but the cooling capacity of this cooler was insufficient. A water quench was installed between the first and second cooler to control the bag-house filter inlet temperature between 160 and 200°C. This water quench cooler was the source of many operational problems, because the water droplets were not completely evaporated when they hit the wall of the cooler. The liquid water on the wall mixed with the dust, resulting in serious fouling of the piping. The pressure drop over the cooler increased to values of some 100 mbar when demolition wood was used as fuel. The water quench cooler is not a component planned for commercial plants, so it was decided to replace the water quench and the second cooler by a conventional cooler (fire tube) in the second quarter of 2010. This new cooler worked without problems and the problems with a high pressure drop were solved. The operating temperature of the bag-house filter was lowered to 120 – 150°C, because of the higher cooling capacity of the new fire tube cooler.

The pressure difference between the settling chamber and the freeboard in the combustor is controlled by a valve that is located upstream of the bag-house filter (not shown in Figure 6-9). The normal control set point is between -15 and +15 mbar, a value obtained by manually closing the valve for about 50%. In case of fouling problems in the flue gas duct the valve is completely opened and the pressure difference can not be regulated anymore. The pressure difference between the settling chamber and the freeboard in the combustor can drop to approximately -60 mbar. This influences the solid circulation rate and the flue gas leakage rate between combustor and riser.

6.5.4 Producer gas cooling and dust removal

The producer gas is cooled in a double pipe cooler that was used in the past to cool the producer gas from the BIVKIN CFB gasifier [18]. The producer gas flows through the center tube, the cooling medium (air) flows through the outer tube in parallel flow. Parallel flow was selected to keep the wall temperature (of the cooler) high enough (>350°C) to prevent problematic condensation of tars on the wall of the cooler. The relatively large dust particles (>20 µm) effectively remove some of the condensed heavy tars from the wall of the cooler. The producer gas outlet temperature from the cooler is approximately 400°C.

The preheated air is used as combustion air in the boiler. The cooled producer gas is sent to the gas cleaning test rig which is located outside the building. The heat loss in the relatively long pipe between the gasifier building and the gas cleaning building is high. It is partially compensated by electrical trace-heating of the last part of the pipes. A cyclone is used to remove most of the dust (ash, carbon and lost bed material) from the gas. The typical operating temperature of the cyclone is 350 – 400°C. A cyclone removes only part of the dust, so the remaining dust removal has to be done in the OLGA gas cleaning. This is a major difference with the lab-scale installation where all the dust is removed before the OLGA.

6.5.5 Producer gas cleaning

The pilot-scale producer gas cleaning test rig consists of the OLGA tar removal system and a wet scrubber. The ECN gas cleaning installation was developed after several less successful tests with different kind of tar and dust removal technologies [19].

The OLGA consists of two washing/scrubbing columns (collector + absorber) and an oil regenerating column (stripper). The first scrubbing column cools the producer gas with oil from approximately 400°C to 80 – 90°C. Heavy tars and dust are captured in the oil. A wet Electro Static Precipitator is installed downstream the first column to collect the aerosols that were not removed in the first column. The mixture of tar and dust can be used as a fuel in the MILENA combustor or gasifier. In the first tests this was simulated by firing natural gas in the combustor. In the second OLGA column the light tars (e.g. naphthalene) are absorbed. The absorption column is operated above the water dew point temperature of the gas, to prevent condensation of water in the oil. The typical water dew point temperature of the MILENA gas is 65 - 75°C at atmospheric pressure. The light tars are removed from the scrubbing oil in a stripper column using preheated air. The air containing light tars is used as combustion air in the boiler.

The gas is further cooled with water in the wet scrubbing system to approximately 35°C (partly depending on ambient conditions). Most of the water in the gas is condensed out. The water scrubber also removes most of the HCl in the producer gas and part of the NH_3. The typical water content of the producer gas after the wet scrubbing system is 7 vol.%.

6.5.6 Boiler

The producer gas is combusted in a boiler during start-up and normal operation. During start-up the gas cleaning is bypassed. The tar loaded gas is sent directly to the burner of the boiler. The burner was originally designed to combust the producer gas from the ECN CFB gasifier BIVKIN [20]. The burner design was modified to handle the higher calorific gas with higher tar content from MILENA. Although the burner in the boiler was especially designed to handle the gas with tar, it is still sensitive to fouling if operated too long on gas with high tar content. Fouling of the burner is prevented as much as possible by taking the gas cleaning in operation as fast as possible.

6.5.7 Construction and commissioning

The engineering of the MILENA pilot plant started in 2005. Financing was approved at the end of 2006. The detailed engineering was done by the Engineering and Services department of ECN in the beginning of 2007. The construction of the reactor vessel was done by HoSt BV, together with Klaas Zijlstra Metaalbewerking BV. The reactor vessel was delivered to ECN by the end of November 2007. Figure 6-11 shows the different reactor segments when they arrived at ECN.

Figure 6-11: Arrival of the MILENA reactor segments at ECN.

The construction and connection of the installation to the existing infrastructure was finished by the end of April 2008. Pressure testing and the required fixing of leakages caused some delays.

The commissioning of a biomass gasification pilot plant is almost always problematic. The commissioning of MILENA was no exception. Commissioning of the pilot plant started in the summer of 2008. The first tests showed numerous problems. Measures taken to compensate for thermal expansion were not incorporated in the design correctly. This resulted in mechanical failure of the metal insert. It took almost a year to solve most of the problems and to adapt the existing gas cleaning for the MILENA producer gas. The integrated system was taken into operation in the summer of 2009 using wood pellets as a fuel.

6.6 The alternatives

Alternative Indirect gasification concepts under development or demonstration are the SilvaGas process and the FICFB process. Both processes are described in chapter 2. In spite of the fact that Indirect gasification was not new ECN decided to start/continue the development of the MILENA gasifier, because of the advantages over the alternatives.

The FICFB process was not known at the start of the MILENA development, but became very well known in the beginning of the millennium because of the very successful demonstration of this technology in the city of Güssing in Austria [21]. This successful demonstration made it possible to continue the development of the MILENA process, because the Güssing plant showed that successful operation of an Indirect gasifier was possible. This reduced the many doubts that Indirect gasification would not be a viable concept.

The FICFB gasifier requires a relatively large amount of steam to gasify the biomass. This results into a low tar content, but creates an efficiency loss, because the steam is heated up to process temperature. Typical steam to biomass ratios for the FICFB gasifier are in the range between 0.7 and 1.1 [22] compared to approximately 0.1 for the MILENA gasifier. The increased efficiency of the MILENA process over the FICFB process is the main driver for the development of the MILENA technology [23].

The SilvaGas process is very similar to the MILENA process. The main difference is the mechanical construction of the reactor and the integration of the process in one vessel. The SilvaGas process was demonstrated in the United Stated [4], but this demonstration was not seen as very successful. The produced gas was directly fired in a wood fired boiler, where it replaced wood. So operating the gasifier was not beneficial. The demonstration plant was shutdown after a few years. The unsuccessful demonstration of the SilvaGas process was used as an argument in the past not to develop an alternative Indirect gasification process, it was seen as too complex. This changed after the successful demonstration of the FICFB process. Because the SilvaGas process never reached the stage of a successful demonstration or commercial plant it was never seen as a reason to stop the development of the MILENA process. Nowadays several initiatives are underway to demonstrate the SilvaGas Technology in combination with the OLGA gas cleaning technology. The producer gas will be used in a gas turbine. The experience of operating the MILENA pilot plant in combination with the OLGA gas cleaning is very advantageous for the large scale commercial projects based on the SilvaGas technology, because the raw gas composition is similar.

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Chapter 7

Experiments

Abstract

The test results from lab-scale and pilot scale experiments in the MILENA gasifiers are presented in this chapter. Different types of woody fuels were used in the tests. The test data was used to produce and verify the relations that are required to describe an indirectly heated biomass riser gasifier like the MILENA. The relations were used to calculate the Cold Gas Efficiency and the overall efficiency to Bio-SNG of a commercial size MILENA gasifier.

In the last paragraphs the more practical issues like agglomeration and fuel flexibility, are described.

7.1 Introduction

The main goal of the experimental work done in the cold-flow, lab-scale and pilot-scale installations described in the previous chapter was to generate and verify the required relations for the MILENA Excel model as described in chapter 5, since they were not available in literature. he second goal was to show that the technology can operate for a prolonged time using a "commercial" fuel without operational problems. Clean wood chips were originally seen as a "commercial" fuel, but this was later changed to demolition wood.

Many of the lab-scale tests were done in combination with the methanation test rig. The standard fuel was beech wood. The main aim of these tests was to test the lifespan of the different catalysts in the gas pretreatment test rig. The performance data of the gas pretreatment catalysts are still confidential and are not part of this thesis.

The most relevant relations for the MILENA model are the carbon conversion as a function of temperature for different particle sizes of woody biomass and the yield of hydrocarbons. Data were generated at different process conditions, different bed materials (olivine and sand) and using different particle sizes of woody biomass. Some experiments were done with alternative fuels like sewage sludge and lignite coal. Data from the experiment with the non woody fuels were excluded from the data set used to generate the relations.

The test program executed in the pilot plant was aimed at generating engineering data for the 10 MW_{th} MILENA Bio-CHP demonstration plant to be built in Alkmaar. The selected fuel is demolition wood (B-wood), so most tests were done using demolition wood.

The MILENA gasifier was designed to use steam as a fluidization agent for the riser, because the N_2 in air dilutes the producer gas. This is not acceptable if the gas is going to be upgraded into Bio-SNG, for which the N_2 concentration is critical. For gas engine application dilution of the gas with some N_2 is allowed. In a commercial scale plant air is cheaper than steam, so many of the tests done to generate data for the foreseen Bio-CHP demonstration plant were done with air instead of steam.

The MILENA gasification tests described in this chapter were done at atmospheric pressure. Pressurized gasification would be beneficial if the desired end-product is Bio-SNG, which must be injected in the gas grid at elevated pressures and the methanation reactions are favored by an increased pressure. Future development of the MILENA technology will also include pressurized operation.

In the first paragraphs of this chapter the results of the experimental work relevant for the MILENA model are described. In the last paragraphs the more practical issues, like agglomeration and fuel flexibility, are described.

7.2 Fuels and bed materials used in MILENA tests

Most of the tests done in the lab-scale and pilot-scale MILENA were done with woody fuels. Figure 7-1 depicts the fuels used. The diameter of the wood pellets is 6 mm.

Figure 7-1: Beech wood, wood pellets and demolition wood as tested in MILENA gasifiers.

Table 7.1 shows the measured compositions of the main fuels used in the MILENA lab-scale and pilot-scale gasifiers. The demolition wood used was of the so called ‘B’ quality. This means that it includes painted waste wood and particle board. It must be noted that the composition of the demolition wood varied strongly during the tests, some batches contained large amounts of particle board material and others contained significantly more gypsum board material than average. Some batches contained very large particles (length > 10 cm), which gave rise to feeding problems. The MILENA pilot-scale feeding system can handle particles with a maximum length of approximately 25 mm. The additional size reduction resulted in a high dust content of the fuel, especially when a large fraction of particle board material was present in the fuel. Pretreatment of the feed is one of the most problematic issues in lab-scale and pilot-scale biomass gasification research. Luckily this gets less important at commercial scale.

Because of the unstable feeding and the inhomogeneous composition of the demolition wood, many of the test results were too unreliable to be included in this chapter.

A limited number of tests were done with alternative fuels like sewage sludge, grass and lignite coal. Results from these tests are described in paragraph 7.14.

Table 7.1: Average fuel compositions

	Beech wood	Wood pellets	Demolition wood B
Moisture [wt.% a.r.]	10.1	8.3	19.0
Ash [wt.% d.b.]	1.0	0.3	2.7
C [wt.% d.a.f.]	49.2	48.2	50.2
H [wt.% d.a.f.]	6.1	6.4	6.1
O [wt.% d.a.f.]	44.5	45.2	41.6
N [wt.% d.a.f.]	0.2	0.1	1.9
S [wt.% d.a.f.]	0.017	0.009	0.10
Cl [wt.% d.a.f.]	0.005	0.012	0.12
LHV [MJ kg^{-1} d.a.f.]	18.2	18.5	18.9
HHV [MJ kg^{-1} d.a.f.]	19.5	19.9	20.2

All duration tests in the lab-scale installation were done using beech wood as fuel. The total number of lab-scale operating hours with beech woods exceeds 1500 hours.

Quartz sand (sieve fraction 0.1 – 0.4 mm) was used as a standard bed material in the lab-scale MILENA gasifier during the first years of operation and for the first tests of the MILENA pilot-scale plant. Quartz sand shows no or no significant catalytic activity in tar cracking, but it was selected as bed material because of its wide availability and its high resistance to attrition. Some wood fractions can contain a lot of sand. Tests in the ECN CFB pilot plant (BIVKIN installation) showed that no external addition of bed material is required if these fractions are used. When these fractions are used the bed inventory will be replaced by the sand in the fuel automatically.

Olivine is a well known catalytic bed material for reducing the tar concentration in a fluidized bed gasifier [1, 2]. It was observed that the origin of the olivine catalyst has an influence on its catalytic activity. The first tests with olivine as bed material in the lab-scale MILENA gasifier were done with olivine from Norway. Later on this bed material was replaced by olivine from Austria ordered from Magnolithe GmbH, because Austrian olivine showed more activity for tar cracking. This was changed to Norwegian olivine for the pilot plant in 2010 again, because of the better quality (less dust) and better availability.

7.3 Hydrodynamics

The hydrodynamical behavior of the MILENA reactor determines the temperature difference between combustor and gasifier riser, the gas exchange between the combustor and gasifier riser and the loss of char from the system.

Solids circulation rate:

The gasifier riser is preferably operated at a high temperature, because the carbon conversion and the conversion of tar into producer gas increases with increasing temperature. This has a positive effect on the overall efficiency. The combustor is preferably operated at low temperature, because this reduces the risk of bed agglomeration. The heat produced in the combustor is transported by the circulating bed material. To transport the heat a positive temperature difference between combustor and gasifier riser is required, although a small temperature difference gives the highest gasifier riser temperature and the lowest combustor temperature. The solids circulation rate determines the temperature difference between combustor and riser. The solids circulation rate is “controlled” by the size of the sand circulation holes in the riser (see chapter 6.2) and the pressure difference over these holes. Figure 7-2 shows the calculated temperature difference for a commercial size MILENA gasifier using wood with 25 wt.% moisture for different solid circulation rates. The MILENA model that is described in chapter 5 was used to simulate the different conditions. The ‘old’ relations for hydrocarbons yield and carbon conversion were used. The right hand y-axis shows the calculated Cold Gas Efficiency (CGE).

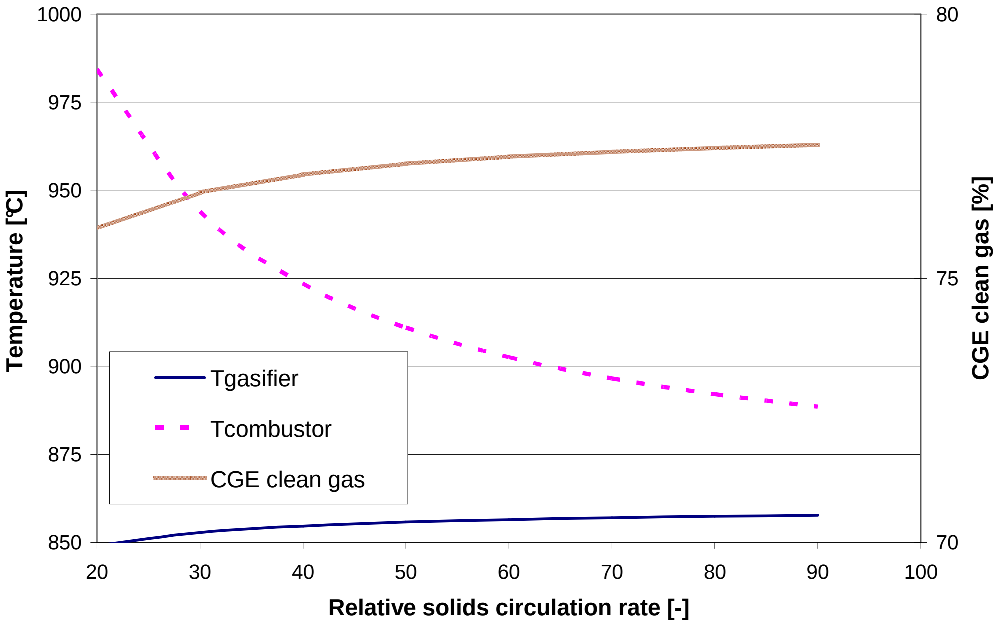


Figure 7-2: Effect of relative solids circulation rate (defined as mass flow solids per mass flow biomass on a.r. basis) on reactor temperatures and CGE.

As can be seen from the figure the solids circulation rate has an influence on the temperature in the fluidized bed combustor and CGE. The MILENA gasifier was originally designed for a solids circulation rate of 40. From the figure it can be concluded that a relative circulation rate above 50 is recommended to limit the combustor temperature and to increase the CGE.

Figure 7-3 shows the measured temperature differences between the riser outlet and the fluidized bed of the combustor. Data from the lab-scale installation have not been used, because the heat balance over the lab-scale installation is unreliable due to the electrical heating of the installation and a relatively high heat transfer from the combustor to the riser through the metal wall of the riser. As can be seen from the figure the temperature difference varies. This is partially caused by variation in pressure between the sand transport zone and the riser. Pressure control appeared to be difficult when demolition wood was used as fuel and the MILENA outlet was connected to the gas cleaning test rig. Automation of the pressure control was only implemented in the second half of 2010. Another reason for the variation in temperature difference was the observed mechanical deformation of the riser. This resulted in an additional opening for the sand to flow from the combustor into the riser. This is probably the reason for the small temperature difference during the demolition wood tests.

The temperature difference decreases with an increase in Equivalence Ratio (ER). ER is defined as the amount of oxygen fed into the gasifier divided by the amount of oxygen required for stoichiometric combustion of the biomass. ER varies because of oxygen transport from the combustor to the gasifier riser by circulating bed material (see paragraph 7.12) and switching between air and steam as gasification agent for the gasifier riser. The temperature difference decreases because the riser gasifier temperature is increased by the partial combustion of producer gas when air or oxygen is added.

From the measurements it can be concluded that the circulation rate is in the correct range. Further testing is required to produce more accurate and reliable empirical relations for the circulation rate as function of pressure difference between the sand transport zone and the riser inlet.

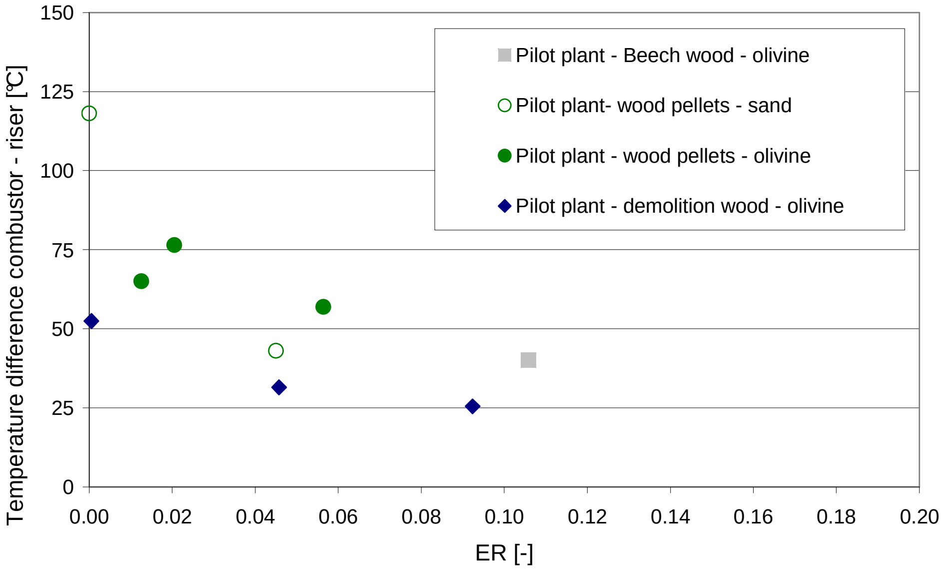


Figure 7-3: Measured temperature differences between combustor and riser as function of Equivalence Ratio (ER)

Gas exchange:

The free space between the circulating bed particles, filled with gas, will move with the gas from the gasifier riser to the combustor and from the combustor to the gasifier riser. This will lead to some gas exchange between the two reactors. If the producer gas is going to be upgraded into Bio-SNG the leakage of flue gas into the producer gas should be limited, because the N_2 in the flue gas will end up in the Bio-SNG. If the N_2 concentration in the Bio-SNG is too high, the specifications for heating value can-not be met.

The producer gas from the gasifier riser moves with the bed material from the settling chamber through the downcomers into the combustor. The pressure in the settling chamber is at normal operating conditions, approximately 10 mbar higher than the pressure in the freeboard of the combustor. The pressure at the outlet of the downcomer is under these conditions higher than the pressure in the settling chamber, because the depth of the exit of the downcomers in the fluidized bed is approximately 30 cm. Every cm of bed height gives a pressure drop of approximately 1 mbar. This results in a positive pressure difference of approximately 20 mbar between downcomer exit and settling chamber. This pressure difference results in an upwards gas flow relative to the downwards moving solids. This velocity of the upwards gas flow is lower than the velocity of the downwards moving solids and results into a net gas flow through the downcomer from the settling chamber into the combustor. See also chapter 6.3 for a more detailed explanation. The net gas exchange through the downcomer is small and is calculated to be approximately 0.8% of the producer gas production. The effect on the overall balance is small. Experiments have shown that the amount of producer gas leaking from the settling chamber into the combustor can be increased by increasing the pressure in the settling chamber or reducing the pressure in the combustor. The impact of the gas leakage under normal operating conditions is small.

More relevant is the leakage of flue gas from combustor into the bottom of the riser, mainly because of the resulting N_2 dilution of the producer gas. Several experiments were done to determine the leakage of flue gas into the riser. In the past N_2 was used as an inert purge gas, so part of the N_2 in the gas came from the purge gas. More recently CO_2 was used as purge gas to minimize the N_2 dilution and to determine how much N_2 came from the flue gas. The experiments showed that the N_2 in the producer gas can be limited to 1 – 3 vol.% if clean wood is used as fuel. This is seen as low enough for upgrading the gas into Bio-SNG. The typical value for flue gas leakage from combustor to gasifier riser is 1% of the amount of flue gas produced in the combustor bed. This value is used in the up-dated MILENA model.

Collection efficiency settling chamber:

The main purpose of the settling chamber is to separate the solids from the gas. The solids entering the settling chamber can be divided into char and bed material. The bed material particles have a relatively high density of approximately 3000 kg m^{-3} and therefore are easily separated from the gas. The char particles can be very fine and have a relatively low density. The average measured bulk density of the particles blown out of the settling chamber is 250 kg m^{-3}. Because of the lower density the separation efficiency for the char particles will be lower than for the circulating bed material. The collection efficiency for the char particles is relevant for the mass and heat balance over the MILENA gasifier.

The collection efficiency for char particles is of course strongly influenced by the particle size of the char, and therewith the size of the biomass particles being used as fuel. The tests with demolition wood as a fuel were seen as the most realistic for further scaling up. These tests therefore were used to calculate the collection efficiency.

The collection efficiency of the settling chamber in the pilot plant for char particles is calculated from the amount of char in the cyclone ashes collected by the cyclone in front of the OLGA gas cleaning. A collection efficiency of 90% for the cyclone was assumed. Approximately 10% of the char particles that are blown out of the MILENA settling chamber end up in the OLGA gas cleaning. These particles will be returned with the tar in commercial scale installations. The collecting efficiency of the MILENA settling chamber varied between 75 and 90%. The strong variation in the collection efficiency was probably caused by the inhomogeneous particle size distribution of the fuel and the fact that some batches contained a lot of dust. The remaining char that is collected by the cyclone will in the future be returned to the combustor.

7.4 Carbon conversion in gasifier riser

Carbon conversion is an important parameter in the Excel model made for the MILENA gasifier. Because Carbon conversion is mainly influenced by the gasifier riser temperature, a temperature dependent relation is used in the model (Equation 7.1). Carbon conversion increases at higher temperatures, resulting in a lower char yield. A lower char yield results in a lower heat production in the combustion section giving a lower temperature in the combustor and the gasifier riser. The temperature dependency of the carbon conversion results into an equilibrium temperature at which the amount of char produced equals the amount of char required to sustain the process temperature. The equilibrium temperature should be between limits posed at the higher end by ash melting and at the lower end by tar composition and tar concentration requirements. The tar content and composition can change significantly at relatively low gasification temperatures (<750°C). This can lead to problems in the equipment downstream which is normally designed for a specific type of tar. Equation 5.1 in chapter 5 shows the definition of Carbon conversion used in this thesis.

<u>Assumed carbon conversion relation:</u>

SilvaGas did several gasification tests in a pilot plant (PRU in Columbus) and a commercial plant (Vermont) to determine the carbon conversion at different process conditions [3]. Figure 7-4 shows the reported carbon conversion as a function of temperature. The presented data indicates that the Vermont gasifier was mainly operated at a relatively low temperature of 677°C (1250°F). It is not clear how the carbon conversion was defined (with or without tar).

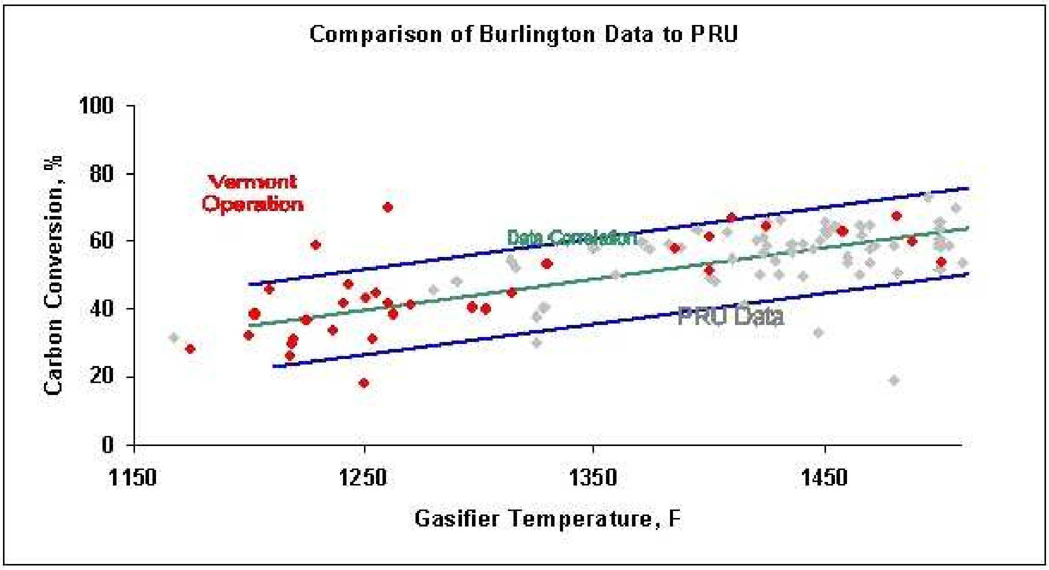


Figure 7-4: Original Carbon Conversion to Gas Results for SilvaGas Process.

Several authors have published relevant carbon conversion data as well. These data are shown in Figure 7-5 for reasons of comparison. Relations obtained from SilvaGas publications [4, 5] are shown as SilvaGas 1 and SilvaGas 2. As mentioned before it is not clear if tar is included in the definition.

Data from the FICFB gasifier cannot be used because the biomass is gasified in a BFB using a much higher steam to biomass ratio than in the MILENA or SilvaGas process.

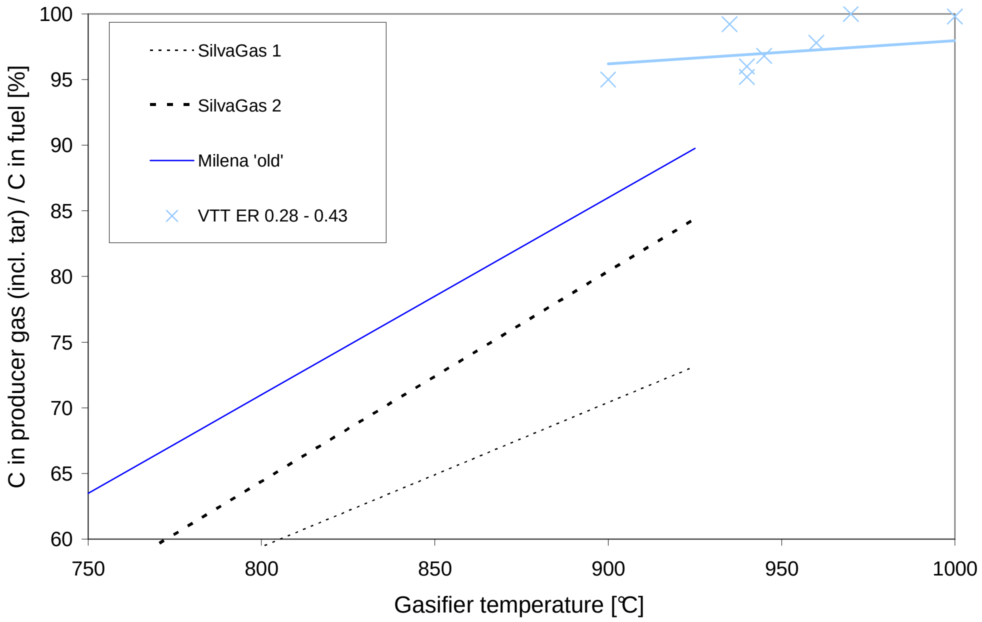


Figure 7-5: Carbon Conversion data from literature

The data published by VTT were for air/oxygen blown gasification [6] and are therefore less relevant than the other data. The reported carbon conversion is higher than for the indirect gasifiers, which is logical because part of the available oxygen will be used to directly combust the carbon in the fluidized bed gasifier. The measurements show a small increase in conversion by increased gasification temperature.

The MILENA ‘old’ relation showed in Figure 7-5 was estimated from the SilvaGas data and from tests with the ECN CFB BIVKIN gasifier. The MILENA ‘old’ relation was used to design the lab-scale and pilot-scale gasifiers.

Carbon conversion measured in MILENA reactor:

The carbon conversion in the riser and the settling chamber cannot be measured directly. The char leaving the gasifier riser is separated from the producer gas in the settling chamber. Approximately 80 - 90% is sent directly to the combustor. The remaining 10 - 20% leaves the MILENA gasifier riser with the producer gas and for approximately 90% it is collected by a cyclone. It will be returned to the combustor in commercial scale installations. The amount of char sent to the combustor is calculated indirectly from the C and O balance over the combustor. The amount of C and O leaving the combustor as flue gas is calculated from the measured dry flue gas composition (concentration of CO_2 and O_2) and the amount of flue gas. This amount of flue gas is calculated from the nitrogen balance over the combustor, assuming that all nitrogen entering the combustor comes from the combustion air. The amount of combustion air is measured.

Figure 7-6 shows the calculated carbon conversion for a wide range of experiments in the lab-scale and pilot-scale installation using different woody biomass fuels. The carbon conversion determined for pilot-scale experiments is lower than for lab-scale experiments. An experiment was done with the same size fuel (beech wood) as used in the lab-scale MILENA. Also this test resulted in a lower conversion for the pilot plant. A higher carbon conversion in the pilot plant was expected, because residence time of the biomass/char in the riser and the settling chamber is longer. The lower carbon conversion is probably due to leakage of fuel from the feeding system into the combustor. This problem will be solved in future tests.

The graph also shows the relation used to design the MILENA process and the ‘new’ relation based on the experiments described in this thesis. It must be noted that the carbon conversions calculated from measurements in the pilot-scale installation can be too low due to leakage of biomass from the feeding system in the combustor.

The data for carbon conversion measured for the MILENA gasifier are significantly higher than what was presented by SilvaGas. This difference is probably due to the higher residence time of the biomass/char particles in the MILENA settling chamber.

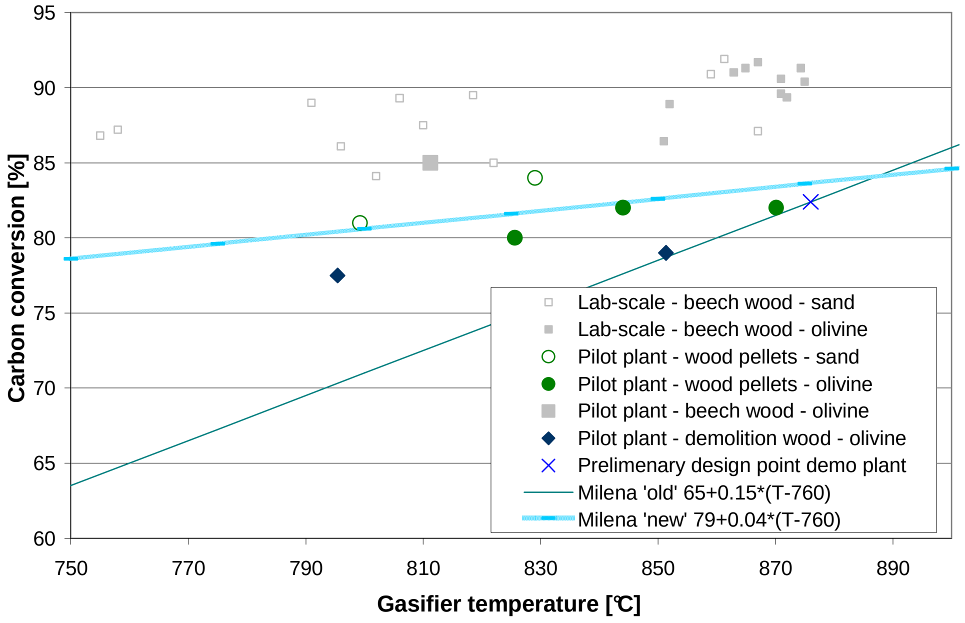


Figure 7-6: Measured carbon conversions in MILENA lab-scale and pilot plant.

Equation 7.1 shows the relation for carbon conversion that is going to be used in the updated MILENA model. At the typical MILENA process conditions the difference with the previous relation are small.

$$\varsigma_{c,gas} = 79 + 0.04*(T-760);\ T \text{ in } ^\circ C. \qquad (7.1)$$

The MILENA process temperature is self controlled. If the gasification temperature drops, the amount of char/fuel to the combustor increases, leading to an increase in temperature. The equilibrium temperature is between 800 and 880°C, depending on the moisture content of the fuel. The fact that the carbon conversion is less temperature dependent than assumed in the past might make additional process control necessary. The idea is to control the gasification and combustion temperature by splitting the recycle of tar between gasifier riser and combustor. A recycle of tar to the gasifier riser will decrease the gasification and combustion temperatures. This has a positive effect on Cold Gas Efficiency, but the concentration of tar in the producer gas will increase. Tests are planned to measure the effect of tar recycling to the gasifier riser on tar composition and concentration.

Char composition:

The char entering the combustor is mainly carbon, but the oxygen and hydrogen content cannot be neglected. The left column of Table 5.1 shows the char composition that was found in literature. These data were obtained from drop tube measurements at temperatures between 750 and 900°C [7].

The second column shows the char composition which was previously used in the MILENA model. The composition was an average of the literature data and the measured composition of the collected cyclone ashes from the ECN CFB gasifier BIVKIN.

Experiments were done in the MILENA pilot reactor to generate more reliable data. Demolition wood was used as fuel. It is not practical to collect the char particles that fall down in the settling chamber and fuel the combustor. The smaller particles that are blown out of the settling chamber are collected by a cyclone. These particles were sampled and analyzed. It is assumed that the composition of the fine char particles is similar to that of the larger particles. The third column in Table 5.1 shows the measured char composition.

The right hand column shows the average values of the previously used data and the data obtained from MILENA pilot measurements. These values are used in the updated MILENA model. Nitrogen is not included in the standard char composition, because the concentration is strongly fuel dependent. The nitrogen content in the char is assumed to be 30% of the nitrogen in the fuel.

Table 7.2: Char composition used in MILENA model

	Literature	Previously used in model	MILENA pilot measurements	Updated Model
Fuel	Hardwood	-	Demolition	-

				Wood	
C	[wt.% daf]	85.2	92.6	89.7	91.1
H	[wt.% daf]	2.5	1.0	1.1	1.1
O	[wt.% daf]	12.3	6.4	9.2	7.8

7.5 Hydrocarbon yields

The yield of hydrocarbons is an essential parameter in the MILENA Excel model and influences the overall system efficiency. For electricity generation by gas engines or gas turbines and for the production of Bio-SNG, a high hydrocarbon yield is beneficial. For syngas application a low hydrocarbon yield is required, preferably close to zero.

Methane is the hydrocarbon that is present in the gas in the highest concentration. It was observed from data generated in the ECN CFB gasifier that methane yield is influenced by Equivalence Ratio (ER). In literature [8] a relation was found that agreed with ECN measurement. This relation is used in the MILENA model and was verified with data from MILENA experiments. Figure 7-7 shows the measured and theoretical yield of CH_4 as a function of ER. The line shown in the figure represents the theoretical CH_4 yield line. The right hand y-axis gives the methane yield on mol fraction basis.

The variation in ER is achieved by varying the amount of fluidization air to the gasifier riser. During steam gasification no air is sent to the gasifier riser. Another reason for variations in ER is the variation in the amount of oxygen transported by the bed material from the combustor to the riser (see 7.12).

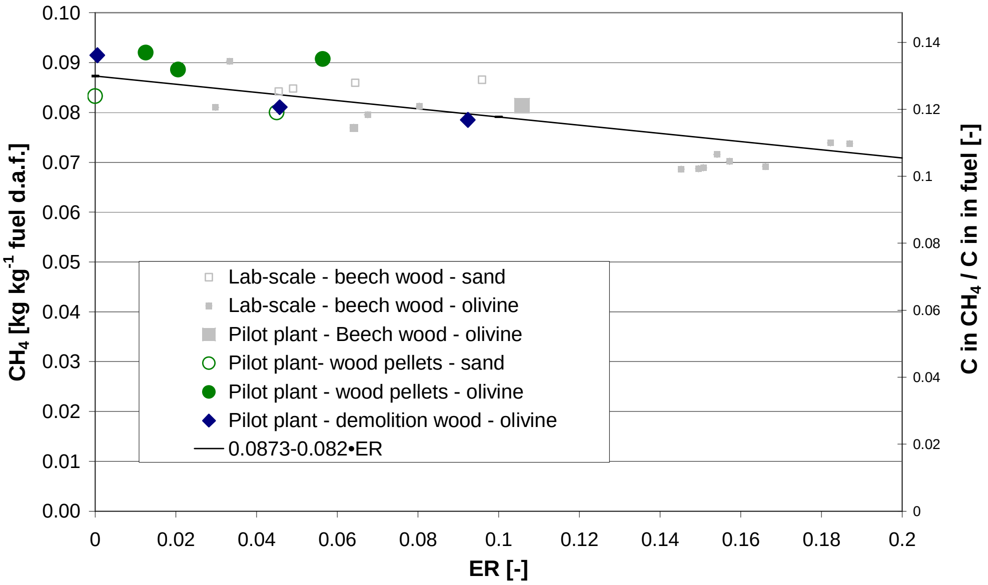


Figure 7-7: CH_4 yield in the MILENA gasifiers as function of ER.

As can be seen from the figure the CH_4 yield decreases with an increase in ER. An increase in ER from 0 to 0.1 gives a reduction in CH_4 yield of approximately 10%, so the oxygen or air that is added results in combustion of the fuel compounds present in the producer gas. It is uncertain to what extent the char is combusted as well. This empirical relation was obtained from literature [8]. The MILENA data points obtained from the lab-scale and pilot-scale MILENA are in accordance with the relation obtained from literature. Hence, there is no need to update the relation used in the MILENA model.

At typical operating temperatures of the MILENA gasifier the theoretical concentration of CH_4 and higher hydrocarbons should be close to zero, if the gas is at chemical equilibrium. At typical operating temperatures of fluidized bed gasifiers (850°C) the CH_4 concentration is always far higher than the theoretical equilibrium concentration. For Entrained Flow gasifiers, which are operated at approximately 1300°C the reported CH_4 concentration is close to the chemical equilibrium concentration (< 1vol.%). So it was expected that an increase in temperature would lead to a lower CH_4 yield. In the MILENA model this was solved by using a correction factor for the CH_4 yield. Figure 7-8 shows the correction factor that needs to be applied to the relation for CH_4 production as function of temperature. The figure shows no decline in CH_4 yield at increased gasification temperatures, but some tendency to increase. The correction factor varies between 0.9 and 1.1. No correction is applied in the MILENA model.

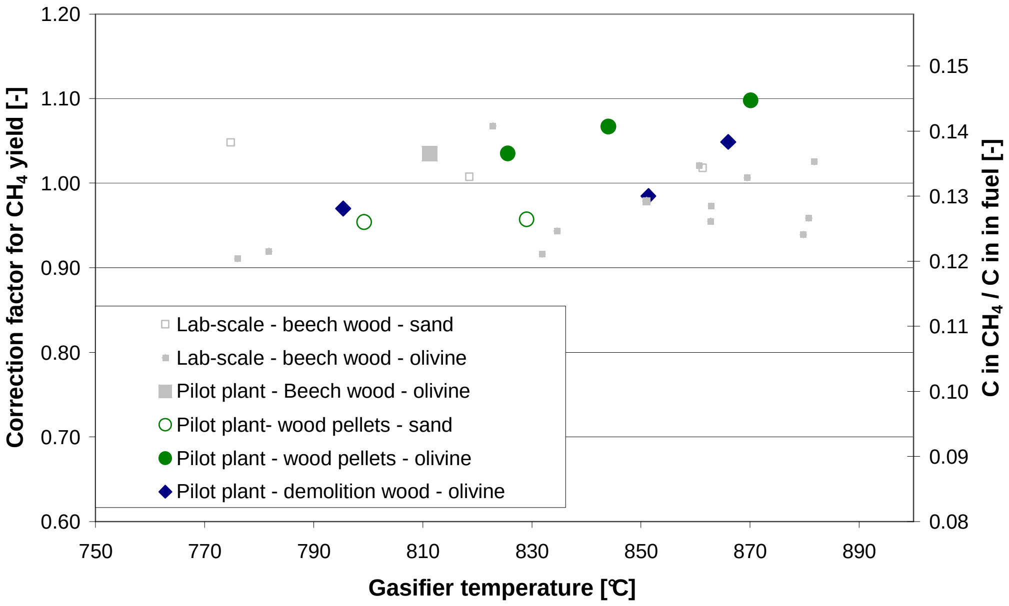


Figure 7-8: Correction for CH_4 yield as function of temperature.

It can be concluded that the original relation used to calculate the CH_4 yield is valid for the MILENA gasifier operated at ‘standard’ operating conditions (gasification temperature between 770 and 880°C and atmospheric pressure) using woody fuels and sand or olivine as bed material. No modifications to the MILENA model were required.

For the yield of higher hydrocarbons no relations were available from literature. From experiments done in the ECN CFB BIVKIN gasifier it was observed that the concentration of higher hydrocarbons is a fraction of the CH_4 concentration. Data produced with the ECN CFB gasifier were originally used in the MILENA model. Experimental data produced with the MILENA gasifiers were used to verify the relations.

On energy basis the cleaned MILENA producer gas contains approximately 16% of ethylene (C_2H_4). Ethylene from fossil origin is widely used in industry, so replacing fossil ethylene by bio-ethylene can have an interesting market potential. Figure 7-9 shows the relative ethylene yields at different temperatures. The lab-scale measurements show no significant effect of temperature on ethylene production. The pilot-scale measurements show a decline in ethylene yield when temperature is increased. The solid line shows the theoretical relation. Measurement data are close to the theoretical line, so the original relation needs no modification.

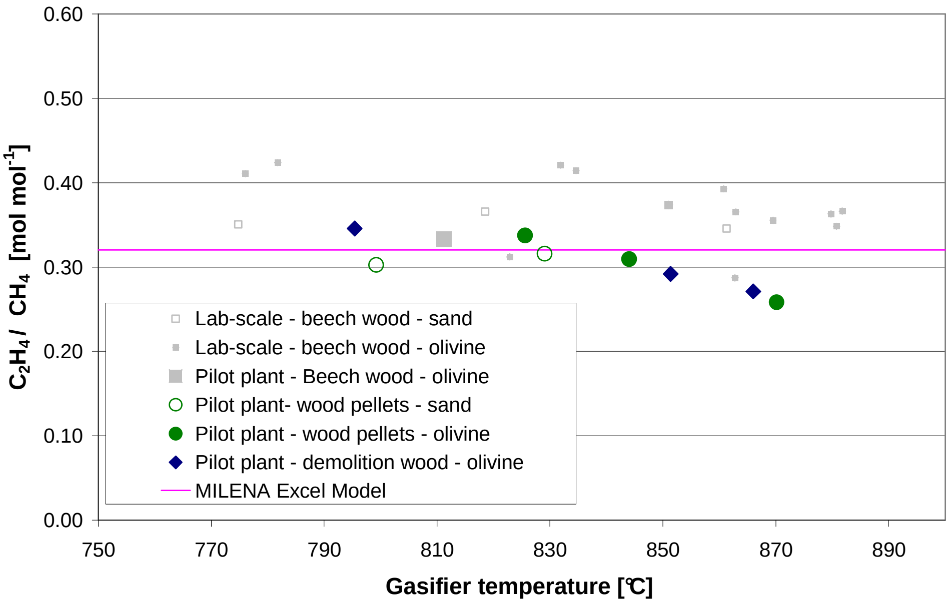


Figure 7-9: C_2H_4 yield relative to CH_4 as function of temperature.

The acetylene (C_2H_2) concentration is of minor importance to the mass and energy balance, but acetylene is a known soot precursor in downstream catalytic processes, so a reduction in concentration can be beneficial.

Figure 7-10 shows the measured acetylene yields. The solid line shows the theoretical relation. Acetylene is a relatively unstable molecule, and a decrease in concentration was expected at increased temperatures, whereas the measurements in Figure 7-10 do not show this. The lab-scale data show a higher yield. The pilot data are close to the fraction that was used in the model. Because the pilot-scale data are seen as more important, the fraction used in the MILENA excel model was not changed.

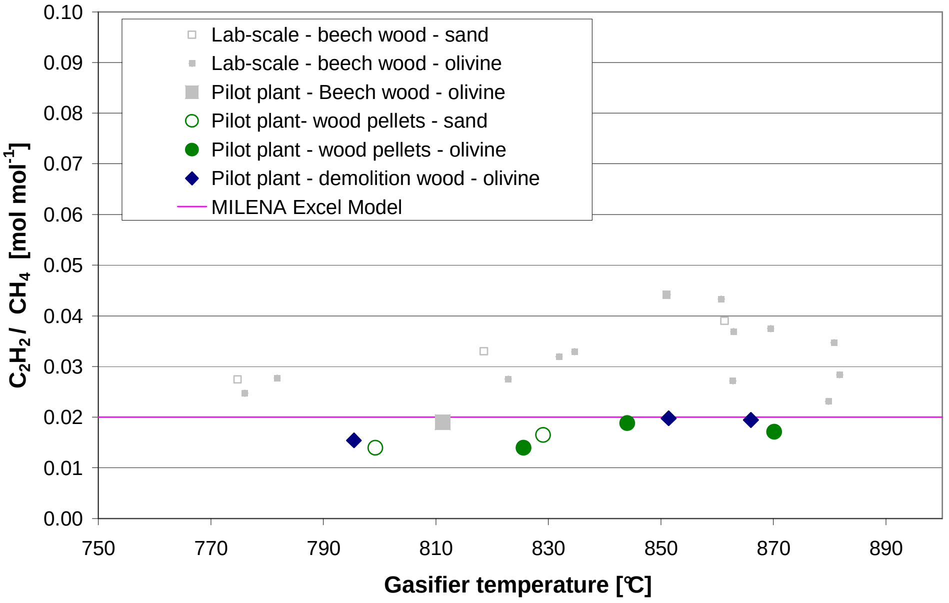


Figure 7-10: C_2H_2 yield relative to CH_4 as function of temperature.

The relative yield of ethane (C_2H_6) is also of minor importance because of its relatively low concentration, but the ethane concentration shows a remarkable trend (see Figure 7-11). The relative ethane yield clearly decreases when the gasification temperature is increased. Possibly ethane is cracked into ethylene and hydrogen.

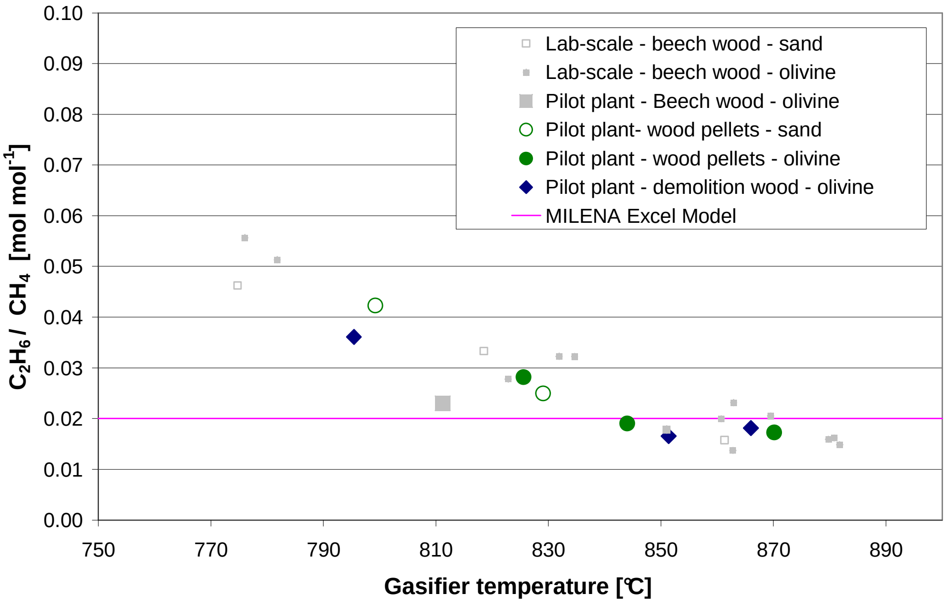


Figure 7-11: C_2H_6 yield relative to CH_4 as function of temperature.

The relation for ethane production was not changed in the MILENA model, because of the negligible influence on the component balance at typical operating temperatures.

Figure 7-12 shows the relative benzene (C_6H_6) yield. There is no significant difference in benzene yield between the fuels or bed materials. A small increase in benzene yield at increased temperatures can be observed. Benzene is a tar cracking product, so an increase is to be expected. However temperature dependency was neglected, the relation in the MILENA model was not changed.

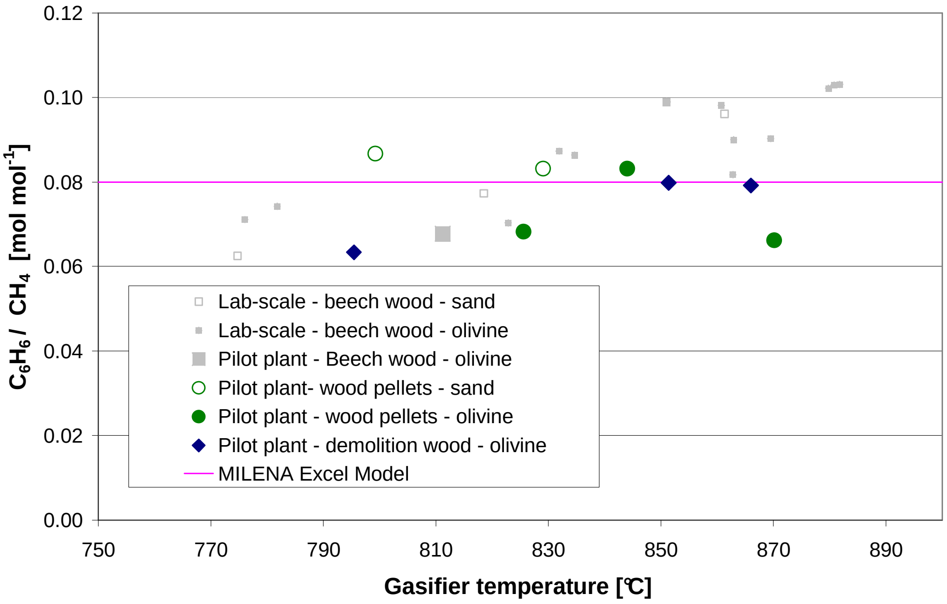


Figure 7-12: C_6H_6 yield relative to CH_4 as function of temperature.

Figure 7-13 shows the relative toluene (C_7H_8) yield. A decrease in toluene yield at increased temperatures was observed. The relation for toluene production was changed accordingly.

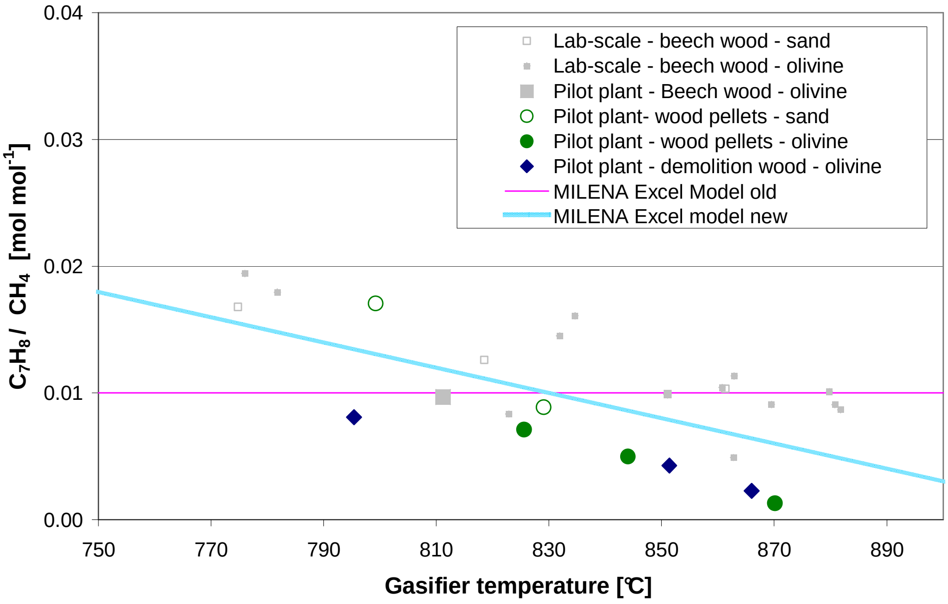


Figure 7-13: C_7H_8 yield relative to CH_4 as function of temperature.

Table 7.3 shows the relations for the yield in hydrocarbons which were used in the past and the modified relations based on measurements in the MILENA gasifiers. Propylene (C_3H_6) is present in the producer gas in low concentrations, but was not included in the previous model.

Table 7.3: Relations used to calculate gasifier outlet gas composition at atmospheric pressure.

Yield	Previously used in Model	MILENA measurements	Updated MILENA Model
CH_4 [kg kg^{-1} fuel d.a.f.]	0.0873-0.082•ER	-	0.0873-0.082•ER
C_2H_2 [mol mol^{-1} CH_4]	0.02	0.03	0.02
C_2H_4 [mol mol^{-1} CH_4]	0.32	0.33	0.32
C_2H_6 [mol mol^{-1} CH_4]	0.02	0.337 – 0.0004 • T	0.02
C_6H_6 [mol mol^{-1} CH_4]	0.08	0.08	0.08
C_7H_8 [mol mol^{-1} CH_4]	0.01	0.093-0.0001 • T	0.093-0.0001 • T
C_3H_6 [mol mol^{-1} CH_4]	-	0.008	0.008

The temperature value used in the temperature correction for toluene yield must be entered in °C.

7.6 Tars

The operation and the design of the MILENA gasifier are not optimized to minimize the tar content in the producer gas. A tar removal or tar reforming device is required to make the gas suitable for gas engine operation or Bio-SNG production. This means that the tar yield is less relevant than for gasifiers designed to produce no or low amounts of tar (e.g. the FICFB process). The OLGA tar removal technology is used in the MILENA lab-scale and pilot plant. The OLGA technology is also going to be applied in the demonstration plant. Tar production data are required to design the OLGA tar removal system. Especially the concentration of naphthalene is of relevance, because this compound is present in the gas in the highest quantity. Benzene and toluene are not included in the tar definition used in this thesis, as they are considered to be a valuable fuel for most applications.

For the design of the producer gas cooler it is required to know the tar dew-point. The cooler should be designed in such a way that wall temperatures are kept above the tar dew point. The tar dew point can be measured or can be calculated from the composition of the tar. The ECN website www.thersites.nl provides a useful tool to calculate the tar dew point. In general the calculated tar dew point of the raw gas from the MILENA is above 250°C. The relations used to calculate the dew point are only valid below 250°C. The wall of the cooler is kept above 400°C to prevent tar condensation.

Figure 7.14 shows an example of the compounds present in the MILENA producer gas. Naphthalene is by far the most common compound in the tar.

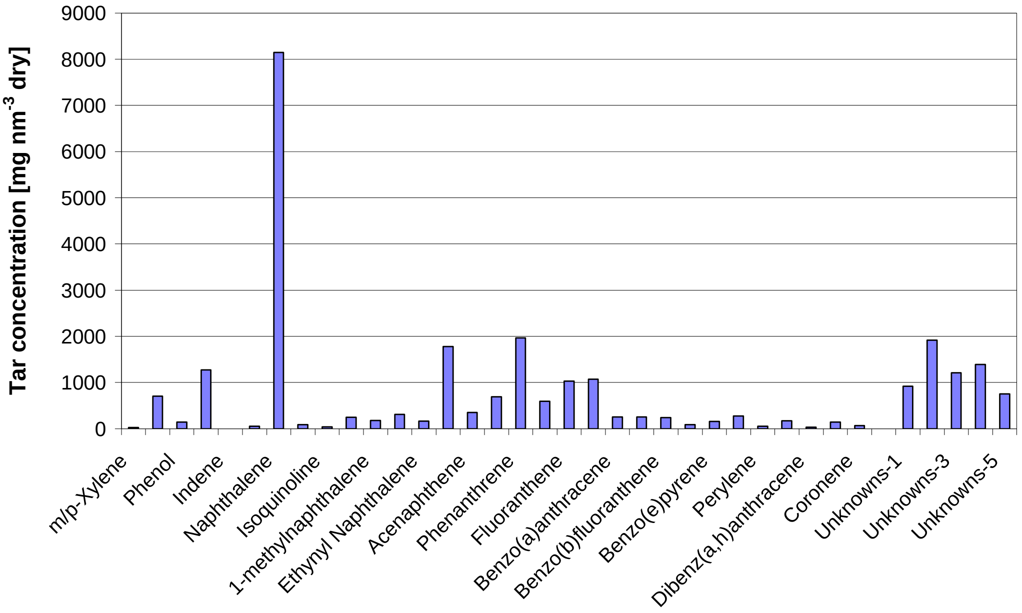


Figure 7.14: MILENA average tar composition for demolition wood and olivine.

The composition of the tar is also of relevance for the MILENA Excel model, because it influences the element and energy balances. Table 5.2 shows the measured compositions and the data used in the model. The composition shown in the most left hand column was used until the middle of 2010. The right hand column shows the average measured composition from lab-scale and pilot-scale tests. The average value was used in the modified MILENA model from July 2010.

Table 7.4: Tar composition on mass fraction basis

		Previously used in model	MILENA lab-scale measurements	MILENA pilot measurements	Updated MILENA Model
Fuel		-	Clean wood	Demolition Wood	-
C	[wt.%]	94.2	93.4	92.9	93.2
H	[wt.%]	5.8	6.1	6	6.1
O	[wt.%]	-	0.5	0.9	0.7
N	[wt.%]	-	-	0.1	-

The heating value of the tar is 38.1 MJ kg^{-1} on LHV basis and 39.4 MJ kg^{-1} on HHV basis.

Figure 7-15 shows the measured tar concentration as a function of temperature. The Solid Phase Adsorption (SPA) method was applied to measure the tar concentration [9]. The SPA method is useful for compounds from phenol to pyrene. Class 1 tars (7- and higher ring compounds) are not included, because they cannot be measured with the SPA method and the contribution to the total amount of tar is negligible.

Only experiments done with steam/dry biomass ratios below 0.5 are shown (this also includes experiments with air to the gasifier riser instead of steam). This is because one of the essential differences between MILENA and the FICFB concept is the low steam/dry biomass ratio. The steam versus dry biomass ratio for a typical commercial scale MILENA gasifier is 0.4. The amount of water in the biomass is included in the steam fraction of the ratio. Steam has a positive effect on the reduction in tar concentration, but the steam vs. dry biomass ratio is kept low to keep the energetic efficiency high.

During the tests different batches of olivine were used (from Austria and Norway). The Austrian batch seems more catalytically active. The gas composition is normally closer to CO shift equilibrium composition. It was also observed that it takes time (> 40 hours) to activate the bed material. Figure 7-22 shows an example for this activation. The H_2 concentration increases over time, probably due to an increased CO shift activity of the bed material. In the measurement data presented in this paragraph no difference was made between the types of olivine or the time the bed material was used in the reactor.

Figure 7-15 shows a decline in total tar concentration (excluding benzene and toluene) when the gasification temperature is increased.

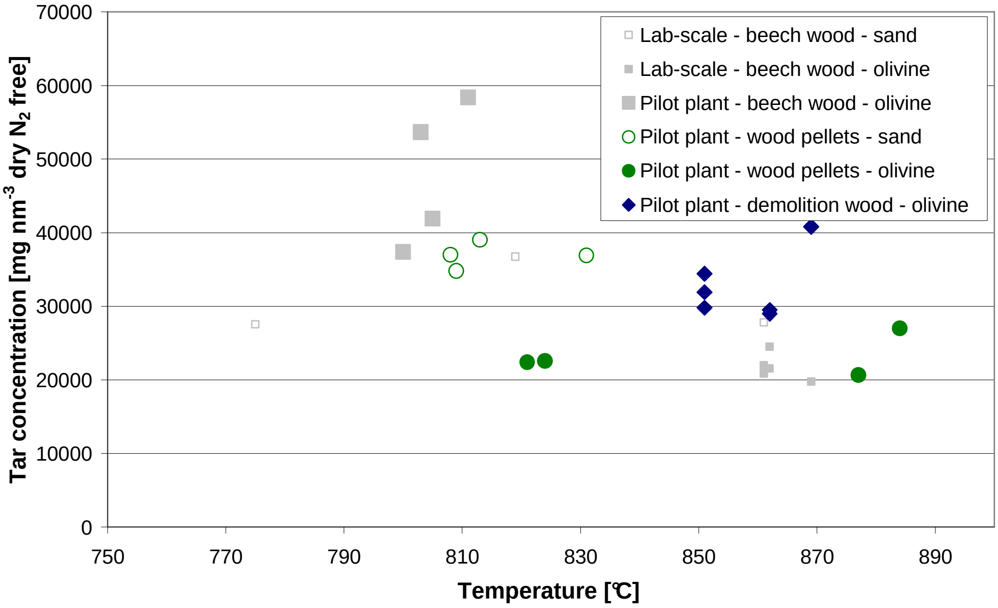


Figure 7-15: Total tar concentration as function of temperature.

Figure 7-16 shows the decline in class 2 tars (heterocyclic components) at increasing temperature; OH groups disappear with increasing temperatures. This agrees with measurements reported by others [10]. There is no clear difference between sand and olivine as bed material.

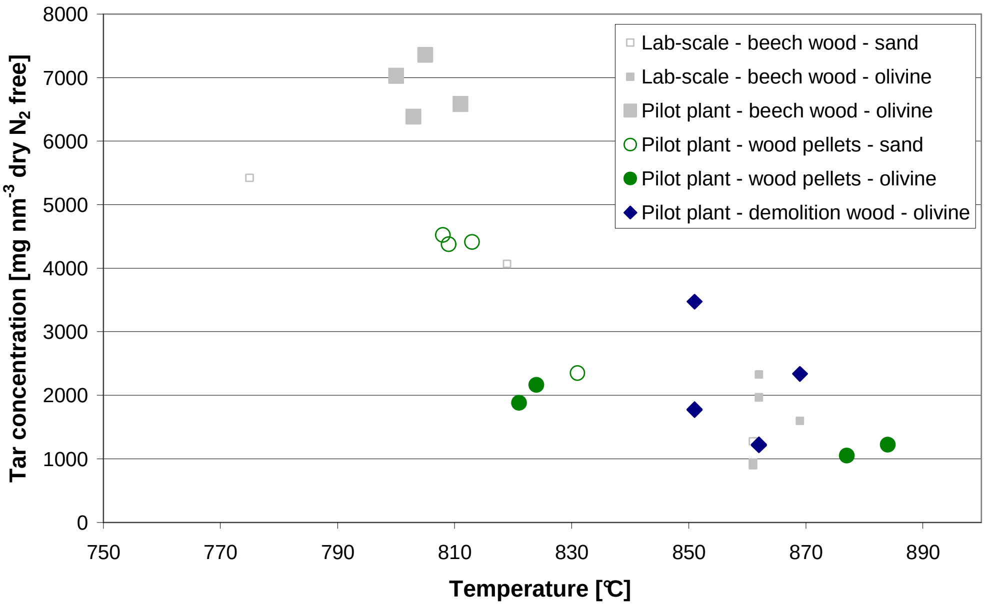


Figure 7-16: Concentration of class 2 tars as function of temperature.

The pilot plant and lab-scale installation seem to produce similar quantities of class 2 tars, despite the longer gas residence time in the pilot plant. The difference in fuel does not significantly influence the class 2 tar yield.

Class 3 tars are light aromatic hydrocarbons which are not important in condensation and water solubility issues. The concentration of class 3 tars (excluding Benzene and Toluene) is relatively low compared to the other classes.

Roughly 50 mass% of the class 4 tars (light poly-aromatic tars), is made up by naphthalene, also called white tar because of its color. Figure 7-17 shows the fraction of naphthalene in the total amount of tar produced. This graph clearly shows the relative increase of naphthalene at an increased temperature probably from the breakdown of heavy polyaromatic compounds.

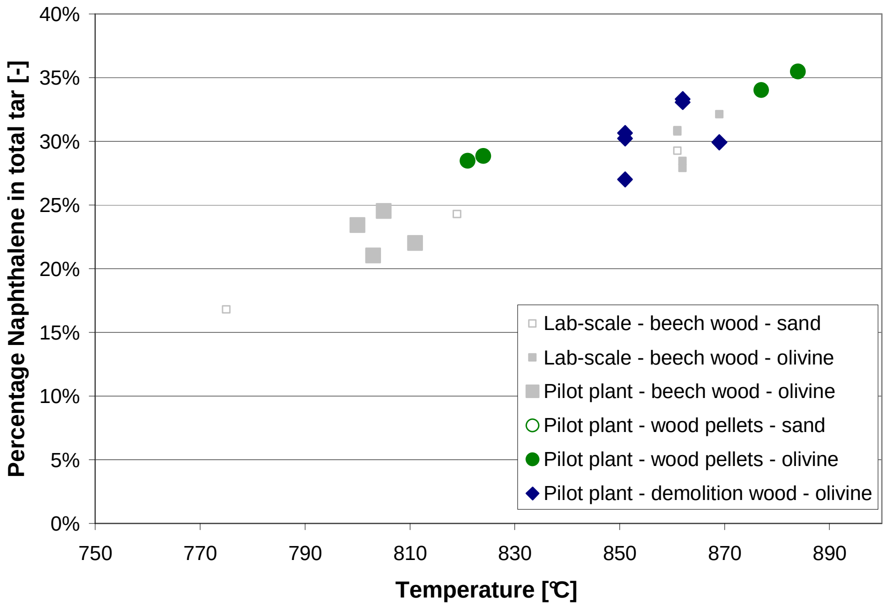


Figure 7-17: Mass fraction of Naphthalene in total tar as function of temperature

Class 5 tars (heavy aromatic hydrocarbons) are of relevance, because they determine the tar dew point of the gas. Figure 7-18 shows the measured concentration of class 5 tars. Finding the right operation conditions to reduce the amount of class 5 tars is of interest, because class 5 tars can cause fouling in the producer gas cooler that is required before the tar removal. Tar condensation is normally prevented by keeping the cooler wall temperature above the tar dew point. Figure 7-15 shows no clear trend regarding reduction of class 5 tars.

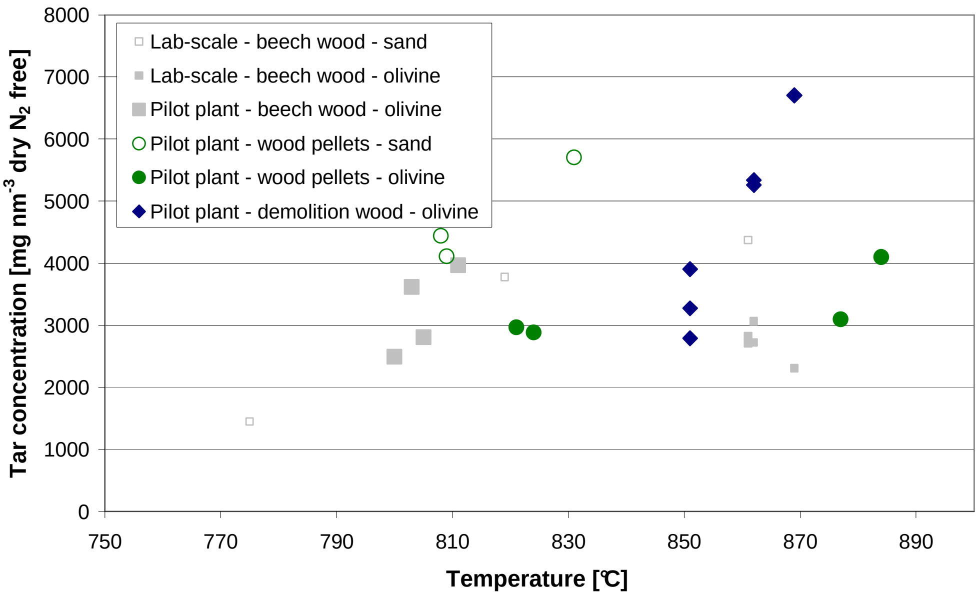


Figure 7-18: Concentration of class 5 tars as function of temperature.

7.7 Heat balance

The pilot-scale MILENA gasifier was designed for 800 kW_{th} biomass input, but most of the time it is operated at lower loads, because the cooling capacity in the gas cleaning test rig is limited.

Table 7.5 shows a typical heat balance for the MILENA pilot-scale installation (without gas cleaning). The heat balance shown is for a test using clean wood pellets as fuel and sand as bed material. The biomass input was 83% of the design capacity.

Table 7.5: MILENA pilot plant, typical heat balance

In	HHV [kW]	**Out**	HHV [kW]
Latent heat air	21	Latent heat producer gas	61
Biomass	667	Condensation heat water	23
Condensation heat water	10	Chemical energy producer gas	515
BTX + tar recycle	0	Latent heat flue gas	52
Natural gas	55	Condensation heat water in flue gas	7
		Heat loss	20
		Carbon/char to cyclone	11
		Chemical energy in tar	61
		Latent heat ash	0

Sum	754	750

Natural gas is fired in the combustor to thermally simulate the recycle of tars and hydrocarbons (some benzene and toluene) which are normally removed from the gas by the OLGA gas cleaning system.

The heat loss of the installation is around 15 kW, but was assumed to be higher for this case because the refractory was not thoroughly heated before the start of the tests.

The latent heat in the flue gas or producer gas is used to pre-heat the combustion air. Pre-heating of combustion air increased the combustor and gasifier riser temperature. A higher gasification temperature results in a higher carbon conversion, thus a higher production of chemical energy in the producer gas.

The heat balance accuracy for a good test is normally within 5%, when a well defined fuel like wood pellets is used.

7.8 Cold Gas Efficiency

The calculated Cold Gas Efficiency (CGE) for the pilot plant on LHV basis is defined as the calorific value of the tar free gas from the OLGA gas cleaning divided by the heating value of the biomass entering the gasifier (no integration of biomass dryer). It varies between 79% and 82% for clean wood with a moisture content of between 25 and 10% respectively. This definition includes the ammonia in the gas. Combustion air was preheated up to 400°C and a recycle of tar and carbon containing cyclone ash was assumed. An increase in cold gas efficiency is possible by integrating a biomass dryer, recycling tar to the gasifier riser instead of to the combustor or an increase in combustion air pre-heat temperature. Sufficient heat is available from the producer gas cooler and the flue gas cooler to supply the heat for the air pre-heating.

The calculated CGE for the pilot plant does not differ much from that of a commercial scale installation using the same fuel, because the main difference is the heat loss, which reduces with increasing scales. The heat loss of the pilot plant is already very modest (approximately 2% of the thermal biomass input), so this has not a large influence on the resulting CGE.

The tests with demolition wood B showed a relatively high content of ammonia in the producer gas. The corresponding heating value was approximately 1% of the total heating value of the producer gas. Therefore, removing the NH_3 (as is required for most gas engine applications), has a significant and negative effect on the CGE. In principle NH_3 is a suitable fuel for gas engines but it will lead to an increase in NO_x production [11].

7.9 Modified MILENA model

The relations obtained from the experiments for carbon conversion as described in paragraph 7.4 and for hydrocarbon yields as described in paragraph 7.5 were implemented in the up-dated MILENA model. The model was applied to calculate a standard case using clean wood with a moisture content of 25 wt.% (a.r.). The composition of the wood pellets as given in Table 7.1 was used. This case represents the desired operating conditions for Bio-SNG production.

The OLGA tar removal is part of the model, because the effect on the energy and material balances of the recycle streams (tars and carbon containing dust) from the OLGA cannot be neglected.

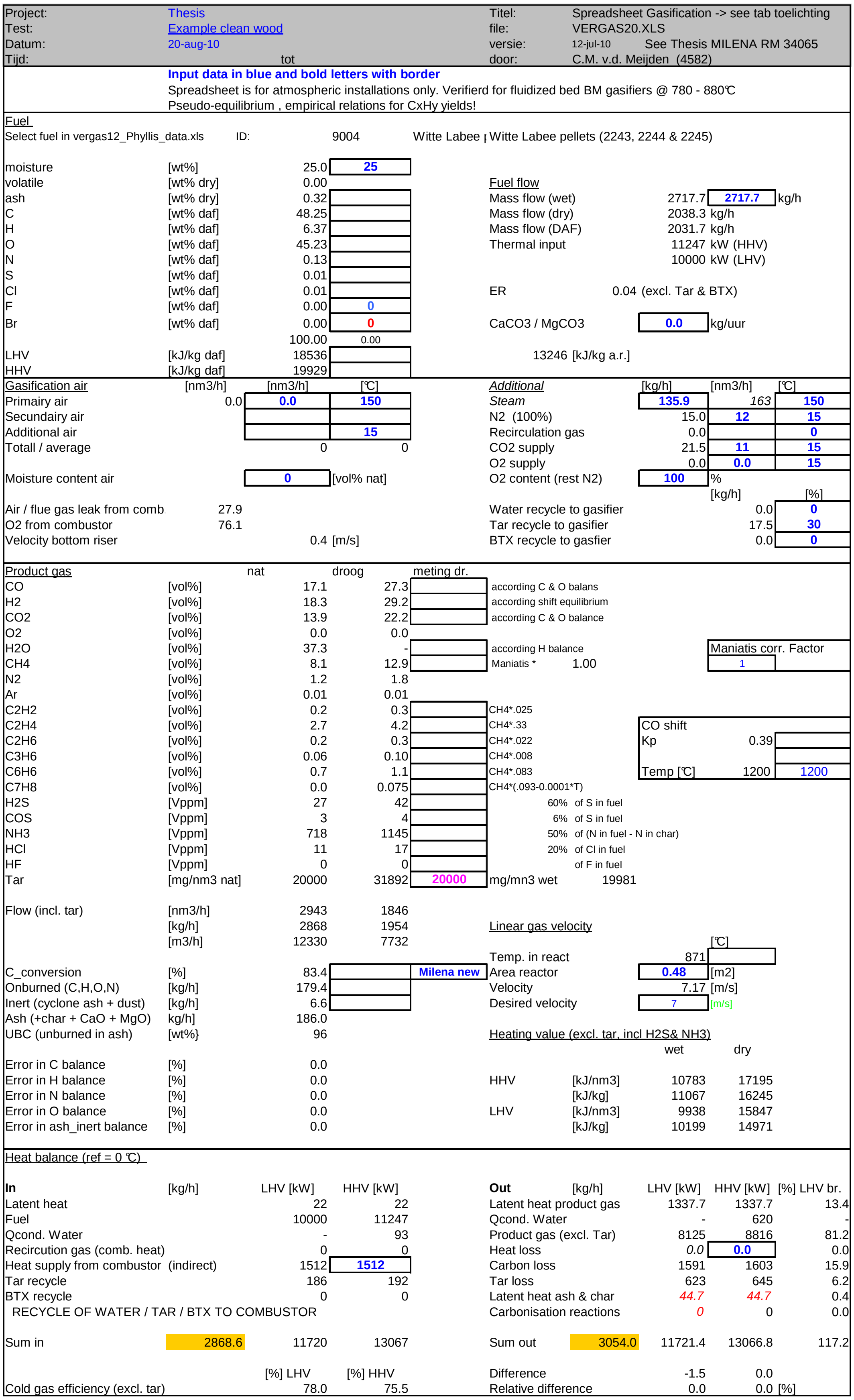
Steam is used as fluidization agent, to minimize the dilution of the producer gas with nitrogen. CO_2 is used as ‘inert’ purge gas. Olivine is used as bed material, transport of oxygen from the combustor to the gasifier riser is included (assumed to be 0.1 mass% of the circulating bed material).

The gasifier riser output using the modified relation is shown in Figure 7-19. The combustor output is shown in Figure 7-20.

A combustion air pre-heat temperature of 320°C was assumed. Tars and dust from the gas cleaning are returned to the MILENA combustor and gasifier riser.

The Cold Gas Efficiency based on the cleaned gas, defined as described in paragraph 2.7 is approximately 78% on LHV basis. This is calculated in an additional worksheet, which is used to describe the gas cleaning (not shown).

The concentrations of pollutants like NH_3, H_2S and COS are relatively low, because the concentrations of S and N in the used wood pellets are relatively low.



Project:	Thesis		Titel:	Spreadsheet Gasification -> see tab toelichting
Test:	Example clean wood		file:	VERGAS20.XLS
Datum:	20-aug-10		versie:	12-jul-10 See Thesis MILENA RM 34065
Tijd:		tot	door:	C.M. v.d. Meijden (4582)

Input data in blue and bold letters with border

Spreadsheet is for atmospheric installations only. Verifierd for fluidized bed BM gasifiers @ 780 - 880℃

Pseudo-equilibrium , empirical relations for CxHy yields!

Fuel

Select fuel in vergas12_Phyllis_data.xls ID: 9004 Witte Labee ‖ Witte Labee pellets (2243, 2244 & 2245)

moisture	[wt%]	25.0	**25**				
volatile	[wt% dry]	0.00		Fuel flow			
ash	[wt% dry]	0.32		Mass flow (wet)	2717.7	**2717.7**	kg/h
C	[wt% daf]	48.25		Mass flow (dry)	2038.3	kg/h	
H	[wt% daf]	6.37		Mass flow (DAF)	2031.7	kg/h	
O	[wt% daf]	45.23		Thermal input	11247	kW (HHV)	
N	[wt% daf]	0.13			10000	kW (LHV)	
S	[wt% daf]	0.01					
Cl	[wt% daf]	0.01		ER	0.04	(excl. Tar & BTX)	
F	[wt% daf]	0.00	**0**				
Br	[wt% daf]	0.00	**0**	CaCO3 / MgCO3		**0.0**	kg/uur
		100.00	0.00				
LHV	[kJ/kg daf]	18536			13246	[kJ/kg a.r.]	
HHV	[kJ/kg daf]	19929					

Gasification air	[nm3/h]	[nm3/h]	[℃]	*Additional*	[kg/h]	[nm3/h]	[℃]
Primairy air	0.0	**0.0**	**150**	*Steam*	**135.9**	*163*	**150**
Secundairy air				N2 (100%)	15.0	**12**	**15**
Additional air			**15**	Recirculation gas	0.0		**0**
Totall / average		0	0	CO2 supply	21.5	**11**	**15**
				O2 supply	0.0	**0.0**	**15**
Moisture content air		**0**	[vol% nat]	O2 content (rest N2)	**100**	%	
						[kg/h]	[%]
Air / flue gas leak from comb.	27.9			Water recycle to gasifier		0.0	**0**
O2 from combustor	76.1			Tar recycle to gasifier		17.5	**30**
Velocity bottom riser		0.4	[m/s]	BTX recycle to gasfier		0.0	**0**

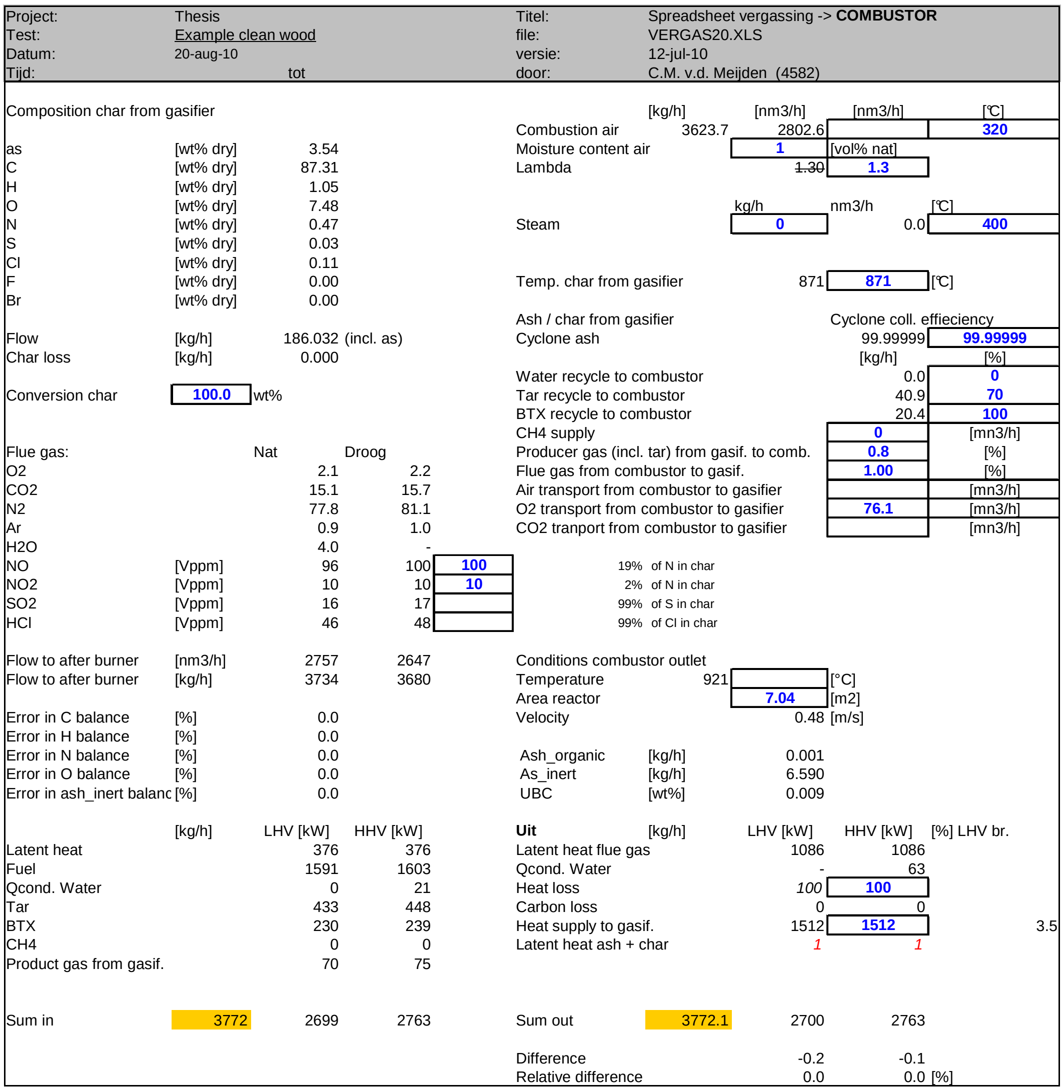
Product gas		nat	droog	meting dr.			
CO	[vol%]	17.1	27.3		according C & O balans		
H2	[vol%]	18.3	29.2		according shift equilibrium		
CO2	[vol%]	13.9	22.2		according C & O balance		
O2	[vol%]	0.0	0.0				
H2O	[vol%]	37.3	-		according H balance	Maniatis corr. Factor	
CH4	[vol%]	8.1	12.9		Maniatis * 1.00	1	
N2	[vol%]	1.2	1.8				
Ar	[vol%]	0.01	0.01				
C2H2	[vol%]	0.2	0.3		CH4*.025		
C2H4	[vol%]	2.7	4.2		CH4*.33	CO shift	
C2H6	[vol%]	0.2	0.3		CH4*.022	Kp 0.39	
C3H6	[vol%]	0.06	0.10		CH4*.008		
C6H6	[vol%]	0.7	1.1		CH4*.083	Temp [℃] 1200	1200
C7H8	[vol%]	0.0	0.075		CH4*(.093-0.0001*T)		
H2S	[Vppm]	27	42		60% of S in fuel		
COS	[Vppm]	3	4		6% of S in fuel		
NH3	[Vppm]	718	1145		50% of (N in fuel - N in char)		
HCl	[Vppm]	11	17		20% of Cl in fuel		
HF	[Vppm]	0	0		of F in fuel		
Tar	[mg/nm3 nat]	20000	31892	**20000**	mg/mn3 wet	19981	

Flow (incl. tar)	[nm3/h]	2943	1846				
	[kg/h]	2868	1954		Linear gas velocity		
	[m3/h]	12330	7732				[℃]
					Temp. in react	871	
C_conversion	[%]	83.4		**Milena new**	Area reactor	**0.48**	[m2]
Onburned (C,H,O,N)	[kg/h]	179.4			Velocity	7.17	[m/s]
Inert (cyclone ash + dust)	[kg/h]	6.6			Desired velocity	7	[m/s]
Ash (+char + CaO + MgO)	kg/h]	186.0					
UBC (unburned in ash)	[wt%}	96			Heating value (excl. tar, incl H2S& NH3)		
						wet	dry
Error in C balance	[%]	0.0					
Error in H balance	[%]	0.0			HHV [kJ/nm3]	10783	17195
Error in N balance	[%]	0.0			[kJ/kg]	11067	16245
Error in O balance	[%]	0.0			LHV [kJ/nm3]	9938	15847
Error in ash_inert balance	[%]	0.0			[kJ/kg]	10199	14971

Heat balance (ref = 0 ℃)

In	[kg/h]	LHV [kW]	HHV [kW]	**Out**	[kg/h]	LHV [kW]	HHV [kW]	[%] LHV br.
Latent heat		22	22	Latent heat product gas		1337.7	1337.7	13.4
Fuel		10000	11247	Qcond. Water		-	620	-
Qcond. Water		-	93	Product gas (excl. Tar)		8125	8816	81.2
Recircution gas (comb. heat)		0	0	Heat loss		*0.0*	**0.0**	0.0
Heat supply from combustor (indirect)		1512	**1512**	Carbon loss		1591	1603	15.9
Tar recycle		186	192	Tar loss		623	645	6.2
BTX recycle		0	0	Latent heat ash & char		*44.7*	*44.7*	0.4
RECYCLE OF WATER / TAR / BTX TO COMBUSTOR				Carbonisation reactions		*0*	0	0.0
Sum in	2868.6	11720	13067	Sum out	3054.0	11721.4	13066.8	117.2
		[%] LHV	[%] HHV	Difference		-1.5	0.0	
Cold gas efficiency (excl. tar)		78.0	75.5	Relative difference		0.0	0.0	[%]

Figure 7-19: MILENA Excel model input and output for gasifier riser.



Project:	Thesis	Titel:	Spreadsheet vergassing -> **COMBUSTOR**
Test:	Example clean wood	file:	VERGAS20.XLS
Datum:	20-aug-10	versie:	12-jul-10
Tijd:	tot	door:	C.M. v.d. Meijden (4582)

Composition char from gasifier					
as	[wt% dry]	3.54			
C	[wt% dry]	87.31			
H	[wt% dry]	1.05			
O	[wt% dry]	7.48			
N	[wt% dry]	0.47			
S	[wt% dry]	0.03			
Cl	[wt% dry]	0.11			
F	[wt% dry]	0.00			
Br	[wt% dry]	0.00			
Flow	[kg/h]	186.032 (incl. as)			
Char loss	[kg/h]	0.000			
Conversion char	100.0	wt%			
Flue gas:		Nat	Droog		
O2		2.1	2.2		
CO2		15.1	15.7		
N2		77.8	81.1		
Ar		0.9	1.0		
H2O		4.0	-		
NO	[Vppm]	96	100	100	19% of N in char
NO2	[Vppm]	10	10	10	2% of N in char
SO2	[Vppm]	16	17		99% of S in char
HCl	[Vppm]	46	48		99% of Cl in char
Flow to after burner	[nm3/h]	2757	2647		
Flow to after burner	[kg/h]	3734	3680		
Error in C balance	[%]	0.0			
Error in H balance	[%]	0.0			
Error in N balance	[%]	0.0			
Error in O balance	[%]	0.0			
Error in ash_inert balanc	[%]	0.0			

	[kg/h]	LHV [kW]	HHV [kW]
Latent heat		376	376
Fuel		1591	1603
Qcond. Water		0	21
Tar		433	448
BTX		230	239
CH4		0	0
Product gas from gasif.		70	75
Sum in	3772	2699	2763

	[kg/h]	[nm3/h]	[nm3/h]	[℃]
Combustion air	3623.7	2802.6		320
Moisture content air		1	[vol% nat]	
Lambda		~~1.30~~	1.3	
		kg/h	nm3/h	[℃]
Steam		0	0.0	400
Temp. char from gasifier		871	871	[℃]
Ash / char from gasifier			Cyclone coll. effieciency	
Cyclone ash			99.99999	99.99999
			[kg/h]	[%]
Water recycle to combustor			0.0	0
Tar recycle to combustor			40.9	70
BTX recycle to combustor			20.4	100
CH4 supply			0	[mn3/h]
Producer gas (incl. tar) from gasif. to comb.			0.8	[%]
Flue gas from combustor to gasif.			1.00	[%]
Air transport from combustor to gasifier				[mn3/h]
O2 transport from combustor to gasifier			76.1	[mn3/h]
CO2 tranport from combustor to gasifier				[mn3/h]

Conditions combustor outlet			
Temperature	921		[°C]
Area reactor		7.04	[m2]
Velocity		0.48	[m/s]
Ash_organic	[kg/h]	0.001	
As_inert	[kg/h]	6.590	
UBC	[wt%]	0.009	

Uit	[kg/h]	LHV [kW]	HHV [kW]	[%] LHV br.
Latent heat flue gas		1086	1086	
Qcond. Water		-	63	
Heat loss		*100*	100	
Carbon loss		0	0	
Heat supply to gasif.		1512	1512	3.5
Latent heat ash + char		*1*	*1*	
Sum out	3772.1	2700	2763	
Difference		-0.2	-0.1	
Relative difference		0.0	0.0 [%]	

Figure 7-20: MILENA Excel model input and output for combustor.

7.10 Distribution of trace elements and pollutants

All biomass fuel contains many different trace elements. The most relevant are chloride, sulphur and nitrogen. These elements end up in the producer gas, flue gas and ash. The distribution of these elements influences the heat and mass balance. Especially the effect of nitrogen can be significant. Emission limits and specifications of downstream equipment (e.g. a gas engine) impose restrictions on these elements in the gas.

Some types of biomass fuels (e.g. demolition wood) are polluted with stones, glass, nails, etc. These pollutants can have a negative impact on the hydrodynamic behaviour of the gasifier and need to be discharged from the system.

Fuel bound nitrogen, sulphur and chloride:

Table 7.6 shows an indicative distribution of fuel bound Cl, S and N between producer gas and flue gas. The distribution is derived from a collection of measurement data for clean wood and demolition wood. The values should be considered as indicative. They are intended to give an impression of the distribution only.

Table 7.6: Indicative distribution of the fuel bound elements N, S and Cl to gaseous compounds and ash.

N		S		Cl	
compound	fraction [%]	compound	fraction [%]	compound	fraction [%]
NH_3	50	H_2S	60	HCl	20
HCN	10	COS	6		
N_2	30	Thiophenes	6		
Tar	0,2	Tar	-		
Char	10	Char	23	Char	60
Fly-ash	-	Fly-ash	5	Fly-ash	20

The hydrogen cyanide (HCN) concentration in the producer gas was only measured twice during a test with demolition wood B. The average concentration was 4245 ppmV on dry basis. This was far higher then expected. The batch of demolition wood used during the test contained a relatively large fraction of particle board. The binders used in the particle board are probably the source of the high HCN concentration. Earlier tests in the ECN CFB gasifier BIVKIN using a mixture of chicken manure and wood showed a HCN yield that was approximately lower by a factor of 10. The HCN measurements will be repeated in the future to check whether the relatively high HCN concentration is representative for demolition wood B.

Thiophenes are always found in the gas in relatively low concentrations, but this sulphur containing compound is hard to remove with technologies commonly applied to remove H_2S. Therefore the concentration is relevant if the equipment placed downstream (e.g. catalysts) is sensitive to sulphur.

The amount of HCl in the gas is strongly influenced by the composition of the bed material and the ash of the biomass, because chloride reacts with several (ash) compounds at typical MILENA process conditions.

Bed pollutants:

Large pollutants in the fuel like glass, stones, nails and screws end up in the riser and are discharged from the system by a screw system. The mixture of bed material and pollutants is sieved to regain the bed material. Figure 7-21 shows an example of the bottom ash discharge from the MILENA riser after sieving (> 0.7 mm). The discharge is a mixture of bed material and large particles. Most of these particles are molten glass particles covered with a layer of bed material (olivine).

Figure 7-21: Typical example of ash particles discharged from riser.

The size of the bed pollutant particles was larger than expected. According to the fuel specification, particles above 25 mm should not be present in the fuel, but in practice this appeared hard to achieve. The large particles were the cause for several problems with the ash discharge system. This system was modified twice, to be able to discharge the bed pollutants.

7.11 Flue gas quality

The flue gas leaving the MILENA is normally sent to a stack after cooling and dust removal and has to meet the local emission standards. If a clean fuel is gasified the standards normally include NO_x, SO_x and dust emissions. The cleaned producer gas normally is put in the same category as natural gas or biogas. Therefore, after combustion of the producer gas, the typical emission limits for natural gas or biogas have to be fulfilled.

Table 7.7 gives an indication of the emissions from the lab-scale installation and the pilot plant when clean wood is used as fuel. The emissions of CO strongly depend on the air to fuel ratio. The concentration rises sharply when the O_2 content in the flue gas drops below 2 percent. The limits in the first column are given as an example. Local emission limits for biomass plants vary strongly.

Table 7.7: Measured flue gas emissions

		Limits	Lab-scale	Pilot plant
O_2	[vol.% dry]	11	4.2	5
CO_2	[vol.% dry]	-	13.4	13
CO	[mg Nm^{-3} @ 11 vol.% O_2 dry]	50	± 30	<10
NO_x	[mg/Nm^{-3} @ 11 vol.% O_2 dry]	70	± 120	± 70

In the pilot-scale installation a bag-house filter is used at a temperature between 100 and 200°C. The concentration of dust is low enough to meet the emission limits.

7.12 Behavior of olivine as bed material

According to literature [1] olivine needs pretreatment before it is active in tar cracking. In the MILENA this is done by keeping the olivine at 800°C for at least 12 hours in an oxidizing environment. However, according to literature higher temperatures are favorable [12]. The iron in the olivine precipitates at the surface of the particle because of this activation. Figure 7-22 gives a good impression for the time required to activate the catalyst. The figure depicts the measured concentrations of the main compounds during a MILENA lab-scale test using small beech wood particles. The test was done in November 2008. The test was started with fresh olivine from Austria. The H_2 and CO_2 concentrations increase over time. The CO concentration decreases, even after 95 hours of continuous operation when the gas composition was still changing. The change in gas composition is a result of the more active catalyst. After 100 hours the gas composition is close to the theoretical CO shift equilibrium. Similar behavior was observed during other lab-scale tests using olivine.

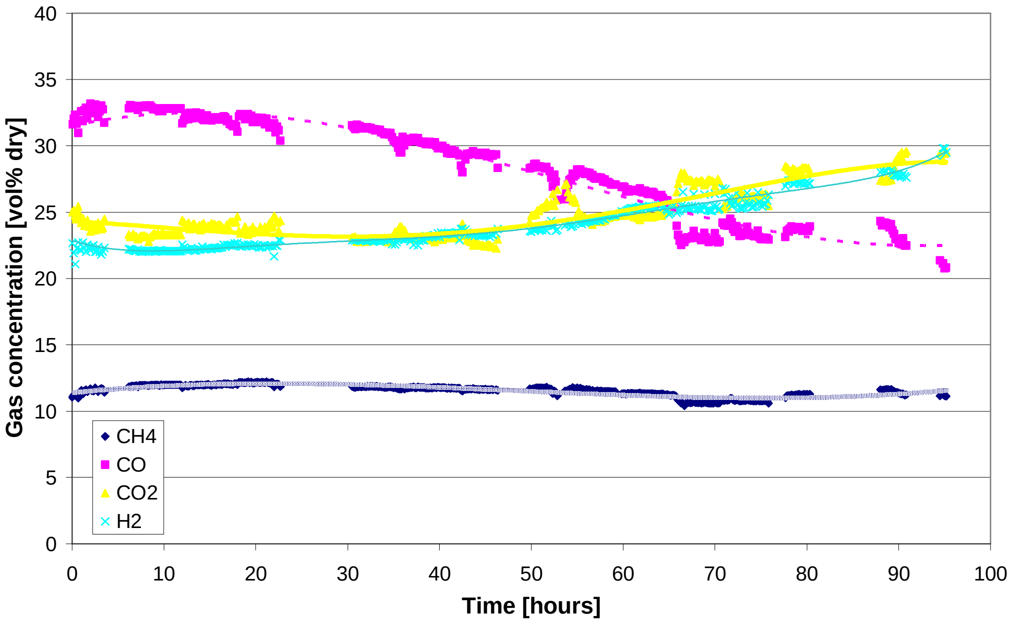


Figure 7-22: MILENA lab-scale duration test gas composition.

The influence of the type of bed material and operating conditions on tar production are described in paragraph 7.6.

Transport of oxygen from combustor to gasifier riser.

After the first tests with olivine it was noticed that the balance for oxygen over the gasifier riser and the combustor was incorrect. Further analysis of the results showed that transport of oxygen from the combustor to the gasifier riser was the most logical explanation. The iron in the bed material oxidizes in the combustor and reduces in the gasifier riser. This results in a net transport of oxygen from combustor to gasifier riser. These findings agreed with data in literature [2, 13], but exact figures or relations to quantify the amount of oxygen transported were not given in literature.

From the different tests the amount of oxygen transported by the olivine was calculated to be equivalent to an ER of 0 – 0.1 at typical MILENA solid circulation rates as described in paragraph 7.3. This equals to 0 - 0.3 mass% of oxygen added to the riser per kg of the circulating bed material. Figure 7-7 shows the methane yield as function of ER. The ER varies from 0 to 0.2. Part of the oxygen came from the circulating bed material and the other part was added as air. Oxygen transport was observed in the lab-scale installation with Austrian olivine and Norwegian olivine, but the amount varied strongly. During the pilot-scale experiments (mainly using demolition wood and Norwegian olivine), the amount of oxygen transport was much lower. It is not clear what causes the differences in the amount of oxygen transport. Further research is required to determine the influence of the oxygen transport.

For the design of the MILENA 10 MW_{th} demonstration plant the amount of oxygen transported was assumed to be 0.1 mass% of the circulating bed material for the case where olivine is used as bed material.

Consumption of olivine:

A major drawback of the use of olivine is the consumption of this bed material. Olivine is more sensitive to attrition than sand. The developers of the FICFB

Gasifier in Güssing have put a lot of effort in minimizing the consumption of olivine.

The first tests in the pilot plant with olivine showed an excessive los in bed material. This was caused by the large fraction of fines in the fresh Austrian olivine. For this reason it was decided to change to Norwegian olivine, which was available without fines. After the switch in bed material the loss in bed material from the combustor was reduced to an acceptable level of approximately 2 kg h^{-1}. A further reduction is expected from an increase in height of the freeboard as it is planned for the demonstration plant. The height in the freeboard of the pilot plant was restricted by the building were it is placed and was lower than theoretically required (see chapter 6).

7.13 Agglomeration

One of the major operational problems in fluidized bed combustors and gasifiers is agglomeration of the bed material. Bed agglomeration can result in de-fluidization of the bed which normally leads to local temperature deviations. This can result in local melting and will finally lead to a complete shutdown of the installation. Agglomeration can be divided into type I and type II agglomeration. Type I agglomeration is best described as deposition induced agglomeration and type II as melt induced agglomeration (see chapter 2.4). Inorganic alkali components from the fuel, mainly potassium (K) and sodium (Na), are the most likely source for type I agglomeration. Type II agglomeration was often observed after tests in the ECN CFB gasifier BIVKIN. The main reason for this type of agglomeration in the CFB gasifier was probably the high char content in the reactor. Because of the limited fuel conversion in a CFB reactor the remaining char tends to accumulate in the reactor. Figure 7-23 gives an impression of the amount of char accumulated in the ECN CFB BIVKIN gasifier when wood pellets were used as fuel. This sample was taken from the sand circulation system after cooling down of the reactor. The operation was shut down without going into combustion mode. The large particles are all char particles from wood pellets. As can be seen from the picture the volume of char is relatively high compared to the volume of bed material (the small 0.5 mm particles). The high char/bed material ratio increases the chance of local hot spots at locations where air is added. Local hot spots will result in local ash melt which will result in type II agglomeration. Char accumulated was reduced in the ECN CFB gasifier by milling the wood pellets.

Figure 7-23: Circulating bed material from ECN CFB gasifier BIVKIN after test using wood pellets as fuel. Sand of 0.4 – 0.6 mm was used as original bed material.

The design of the MILENA prevents char accumulation and local hot spot. Because the char is completely combusted in the BFB combustor. Type II agglomeration, caused by excessive accumulation of char is therefore prevented.

Another causes for type II agglomeration can be melting of fine ash particles which are in the biomass or melting of glass particles. Glass particles are a very common pollutant in demolition wood. Tests with demolition wood (B-wood) were done to investigate the behavior of the glass particles in the MILENA gasifier. Figure 7-24 shows a picture of the bottom-ash discharged from the riser.

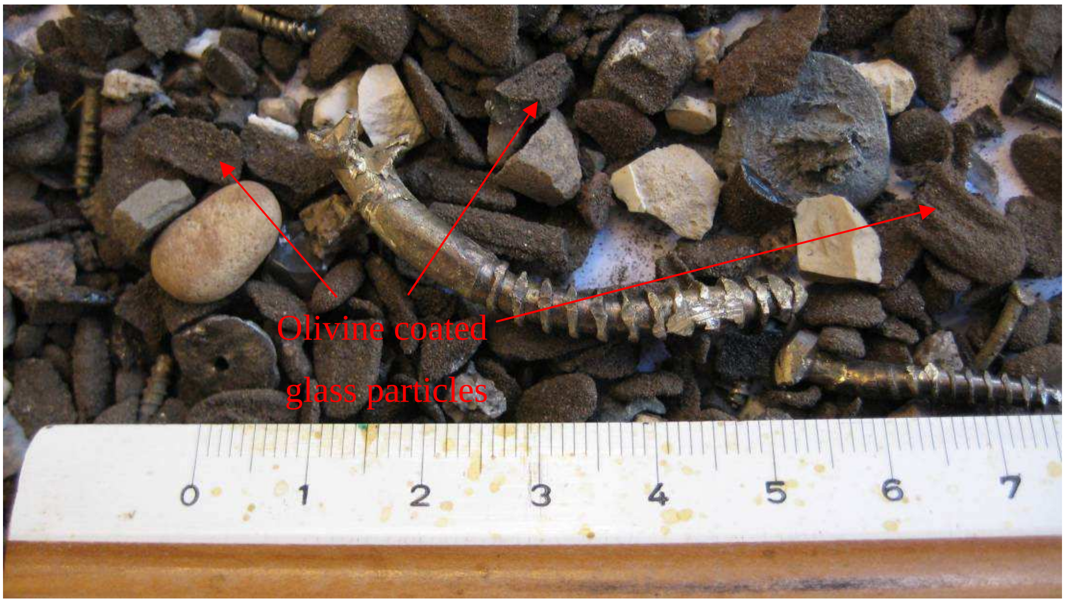


Figure 7-24: Bottom ash riser, produced during demolition wood B test.

Most of the glass particles were coated with a thin layer of bed material (olivine). This is shown in Figure 7-25 in more detail The coating prevents sticking of the molten glass particles to each other. The center of the particle consists of partly molten glass. The edges are covered with bed material.

Figure 7-25: Close-up of broken glass particle.

Figure 7-26 shows what happens with glass particles in the bed (from left to right). The raw glass particles are heated to process temperature and get sticky. The bed particles stick on the glass surface until the particles are completely covered with bed particles.

Figure 7-26: Various stages of bed material covering of glass particles.

During one demolition wood gasification test part of the fuel was screwed directly into the combustor/sand transport zone of the MILENA reactor, because of a mechanical failure of the riser reactor. After this test type II agglomerates where found in the reactor, but the phenomenon was not observed during normal operation.

Fast growing biomass crops like grass have a relatively high content of potassium and silicon. Even without an interaction with the bed material the ash from the biomass can form a melt at normal operating temperature, resulting in type II agglomeration. Several tests were done at relatively low gasification temperatures to see if the MILENA gasifier is suitable for such fuels. A summary of the results is given in Table 7.8.

Table 7.8: Agglomeration results.

Fuel [-]	Test duration [hours]	Tgasfier [°C]	Bed material [-]	Agglomeration [-]
Beech wood	> 1500	800 - 870	sand/olivine	No
Wood pellets (clean wood)	≈ 20	800 - 870	sand/olivine	No
Demolition wood	≈ 50	800 - 870	olivine	No/Possibly
Wood sewage sludge mixtures	≈ 20	800 - 870	olivine	No
Sewage sludge	< 10	≈ 800	olivine	Yes
Wood lignite mixtures	≈ 20	850	olivine	No
Grass	< 5	648 – 700	sand	Yes
Digestate pig manure + corn	< 5	649 – 700	sand	Yes

Straw	≈ 10	650 - 700	sand	No

Type II agglomerates were found after tests with demolition wood B in the pilot plant. During that test the ash discharge system became blocked. Glass particles accumulated in the reactor.

7.14 Alternative fuels

One of the attractive features of fluidized bed gasifiers is fuel flexibility. The development of the MILENA gasifier is focused on using woody fuels, because this type of fuel is widely available and the other indirect gasifiers (FICFB and SilvaGas) reported successful results using woody fuels. Other (biomass) fuels are suitable as well, at least in theory.

A limited test program was done using alternative fuels to get an impression for the suitability for gasification in the MILENA reactor. All tests described in this paragraph were done in the lab-scale installation.

Torrefied beech wood particles were tested once in the lab-scale MILENA. The fuel behaved similarly as dry wood.

Grass was tested in the lab-scale MILENA several times at different temperatures. At normal gasification temperatures (800 – 850°C) the bed material (quartz sand) agglomerated. The reactor wall clearly suffered from corrosion, probably due to the high chloride content of the used grass. Further lab-scale tests are planned at lower gasification temperatures (650 – 700°) to prevent these agglomeration problems.

A mixture of sewage sludge granules and clean wood was tested successfully in the lab-scale installation. Pure sewage sludge resulted in bed agglomeration.

Lignite/wood mixtures were tested more extensive. The results are described below.

<u>Lignite/wood mixture:</u>

Lignite is the type of coal that has most similarities with wood. It has a high content of volatile matter which makes it suitable for conversion in the MILENA reactor. The global reserves of lignite coal are enormous, but the fuel has a bad reputation because the mining of the fuel destroys local landscapes and many of the (older) power stations firing lignite do not meet present day emission standards.

Co-gasification of wood and lignite in the MILENA for the production of SNG can be attractive, because the production of SNG from biomass can be CO_2 negative if complete CO_2 sequestration is done. It was calculated that up to 35 weight percent of wood can be replaced with lignite without a net fossil CO_2 emission [14].

Because of the lower volatiles content compared to wood the yield of char is higher than the char yield for wood. This will result in excess heat production in the combustor. To limit this heat production a mixture of wood and lignite was used during lab-scale tests. Table 7.9 shows the fuel composition of the beech wood and lignite used during the tests.

Table 7.9: Composition of beech wood and lignite in the tests.

		Beech wood chips	Lignite
C	[wt. % a.r.]	45.7	55.0
H	[wt. % a.r.]	5.5	3.9
O	[wt. % a.r.]	38.6	25.1
N	[wt. % a.r.]	0.27	0.61
S	[wt. % a.r.]	0.054	0.260
Cl	[wt. % a.r.]	0.045	0.013
Ash	[wt. % dry]	0.9	3.8
Water	[wt. % a.r.]	9.0	13.4
Volatiles	[wt. dry]	82	49.2

Table 7.10 shows the measured gas composition for beech wood and a mixture of beech wood and lignite. The tests were done with Austrian olivine as bed material.

Table 7.10: Measured gas composition for beech wood and beech wood/lignite mixture

Percentage of lignite	[wt. %]	0	55
CO	[vol.% dr.]	37.2	23.3
H_2	[vol.% dr.]	21.2	36.2
CO_2	[vol.% dr.]	18.9	22.6
CH_4	[vol.% dr.]	12.1	8.2
C_2H_2	[vol.% dr.]	0.4	0.2
C_2H_4	[vol.% dr.]	4.6	2.9
C_2H_6	[vol.% dr.]	0.4	0.2
C_6H_6	[Vppm dr.]	8290	6787
C_7H_8	[Vppm dr.]	1332	1009
N_2	[vol.% dr.]	4.4	4.4
Class 2 tars	[mg Nm^{-3}]	4842	1674
Class 3 tars (excl. toluene)	[mg Nm^{-3}]	580	241
Class 4 tars (2 & 3 ring)	[mg Nm^{-3}]	13801	9157
Class 5 tars (4 & 5 ring)	[mg Nm^{-3}]	2666	1745
Unknown tars	[mg Nm^{-3}]	8450	3271
Total tar (excl. toluene)	[mg Nm^{-3}]	30340	16088

The gas quality improved with respect to the amount of tars, if the wood/lignite mixture is compared to pure wood. The most relevant parameter in the design of a commercial unit is the carbon conversion of the fuel mixture, because this strongly influences the heat balance. If too much heat is produced, external cooling of the MILENA combustor is required. From the tests it was observed that the carbon conversion declines when lignite is gasified. This was to be expected because the volatile content of lignite is lower than the volatile content of biomass.

The lab-scale tests showed that co-gasification of wood lignite mixtures is possible. A positive surprise was the lower tar concentration. Further testing on lab and pilot-scale is required to explain the reduced tar concentration and to produce more reliable data required for scale up of the process.

7.15 Upgrading of the gas into Bio-SNG

Research on upgrading of the MILENA producer gas into Bio-SNG is an ongoing activity at ECN. A lot of experimental work is done on removing trace levels of pollutants before the catalytic conversion of the gas into $CH_{4.}$ The experiences from this experimental work are used to predict to final Bio-SNG gas composition.

The output of the updated MILENA model as described in paragraph 7.9 was used to calculate the gas composition of Bio-SNG after methanation and upgrading. The calculated overall efficiency excludes electricity consumption and electricity production from excess heat.

The producer gas after the OLGA gas cleaning is further cleaned (complete removal of S and Cl) and the hydrocarbons are converted at atmospheric pressure into CH_4, CO, CO_2, H_2 and H_2O. Water is added to prevent formation of soot. CO_2 and water are partially removed from the gas and the gas is compressed to typical methanation pressures (e.g. 40 bar). Conventional methanation processes are used to convert the gas to Bio-SNG. After water removal the gas is ready for gas grid injection. Table 7.11 shows the calculation results.

Table 7.11: Final Bio-SNG composition

CO	[vol.%]	0.0
H_2	[vol.%]	1.2
CO_2	[vol.%]	0.4
O_2	[vol.%]	0.0
CH_4	[vol.%]	93.4
N_2	[vol.%]	4.6
LHV	[MJ nm^{-3}]	33.7
HHV	[MJ nm^{-3}]	37.4
Overall efficiency (LHV) basis	[%]	68.3

The overall efficiency on LHV basis can be increased to over 70% by integrating a dryer that uses low temperature waste heat from the process.

7.16 Discussion and concluding remarks

Oxygen transport from the combustor to the riser is still not well understood. The behavior of the MILENA gasifier when olivine is used is not predictable at the moment. Luckily the overall effect of oxygen transport on the mass and heat balance and the gas composition is relatively low. For the basic engineering of the MILENA demonstration plant this uncertainty was taken into account by making different cases: one with sand as bed material (no oxygen transport) and one with an ‘active’ olivine.

The carbon conversion as function of temperature was determined from experiments in the lab-scale installation and pilot plant. It must be noticed that during some of the experiments done in the pilot plant, fuel might have leaked into the combustor. This results in a lower calculated carbon conversion. New experiments are scheduled to verify this.

The main aim of the MILENA development was to arrive at a reliable biomass gasification technology that can be scaled up to over 100 MW and to produce a gas that can be upgraded to Bio-SNG with a high overall efficiency. The experiments done in the lab-scale installation and pilot plant have shown that the MILENA concept is capable of doing that. The design was made is such a way that the scale up is relatively easy and pressurized operation is possible. Experiments have shown that the process operates as expected on woody fuels. Experimental data were used to modify the MILENA model. The modifications were modest. The model is usable to predict the gas composition and the heat balance for the MILENA gasifiers operated between 770 and 880°C and atmospheric pressure using woody fuel. The model / relations can be used for comparable biomass fluidized bed gasifiers as well (e.g. BFB and CFB gasifiers), but experimental verification is recommended. The updated model shows that an overall efficiency of 68% can be achieved from wood with 25 wt.% moisture to SNG. The efficiency can be increased by integrating a dryer that uses waste heat.

Accumulation of char results in (type II) agglomeration. Accumulation of char can be prevented by combustion of the char as is done in the MILENA concept. Lab-scale and pilot-scale experiments have shown that agglomeration is not an issue in the MILENA reactor when clean woody fuels are used. Several tests with other fuels have shown that agglomeration can be an issue. Further tests are scheduled to identify the suitable fuels and operating conditions for the MILENA reactor.

Olivine is the standard catalytic bed material used to reduce the tar yield of fluidized bed biomass gasifiers. It was observed that different batches of olivine behave differently. The results in tar reduction in the MILENA are disappointing. This is probably due to the relatively short contact times between the catalytic bed material in the riser reactor and the producer gas compared to BFB reactors which are normally used in combination with olivine. It was observed that catalytic activity of olivine increased over time, but this was only observed in the lab-scale installation using clean wood. Better pretreatment/activation (higher calcination temperature) of the olivine will probably lead to better results. The origin of the olivine also influences the tar concentration in the producer gas, but this was not subject of the work described in this thesis. The behavior of olivine (and of other catalytic bed materials) will be a topic of future research.

Because of the relatively high tar content of the MILENA producer gas additional tar conversion or removal processes are required to make the gas suitable for gas engine operation or Bio-SNG production. The OLGA tar removal technology has been used successfully by ECN to remove the tars to an acceptable level.

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Chapter 8

Conclusions and outlook

8.1 Conclusions

Tests in the lab-scale and pilot-scale MILENA gasifiers have shown that the MILENA technology is suitable for conversion of woody biomass into a combustible gas with high efficiency. The residual ash is virtually carbon free (< 1 wt.% C), which means that the loss in heating value of the residues including the ash is close to zero. The yield of hydrocarbons (CH_4, C_2H_4 and C_6H_6) is relatively high, which has a positive effect on overall efficiency if the gas is used for power generation or production of Bio-SNG. The CH_4 concentration in the producer gas is higher than the target concentration of 12 vol.% when steam is used as gasification agent in the riser.

Measurements have shown that the gas exchange between combustor and riser gasifier can be below 1%, limiting the N_2 content in the producer gas to 1 – 3 vol%.

The Cold Gas Efficiency (CGE) on LHV basis, defined as the energy content of the cleaned gas (free of tars) divided by the energy content of the biomass entering the gasifier varies between 79% for clean wood with 25 wt.% moisture and 82% for wood with 10wt.% moisture. A further increase in cold gas efficiency is possible by integrating a biomass dryer, recycling tar to the gasification reactor, instead of the combustion reactor, or an increase in combustion air pre-heat temperature.

The scale up from the 30 kW_{th} lab-scale to the 800 kW_{th} pilot-scale installation was without major problems for the process related issues. The gas composition and heat balance were up to the expectations. All major problems were related to mechanical failures. Solving the problems related with thermal stresses took most of the time. They are still topic of an ongoing optimization program. Fuel and bottom ash handling is the other major challenge.

In several duration tests clean wood was used as fuel and quartz sand or olivine as bed material. Agglomeration of the bed material was not observed to be a problem. Demolition wood contains several pollutants which can cause bed agglomeration, but the first 50 hours of tests, using a relatively poor quality of demolition wood with a lot of plywood, particle board and glass pieces, showed that bed agglomeration can be prevented. A longer duration tests (1000 hours) is scheduled for the end of 2010 to prove that the MILENA gasifier can run for a prolonged period using a relatively difficult fuel (demolition wood B).

The tar content of the MILENA producer gas is relatively high (20 – 40 gram nm^{-3} dry). The use of a catalytically active bed material like olivine has only a modest effect on the tar concentration. This is probably due to the relatively short contact time between the gas and the catalyst compared to Bubbling Fluidized Bed reactors and the low steam to biomass ratio. The origin of the olivine also influences the tar concentration in the producer gas, but this was not subject of the work described in this thesis and requires more research. Tar removal technology like the OLGA is required to clean the gas before it can be used in gas engines or upgraded into Bio-SNG.

During tests with olivine as bed material, transport of oxygen from the combustor to the gasifier was observed. This is caused by oxidation and reduction of the iron in the olivine (chemical looping). The amount of oxygen transported varied strongly. More research is required to determine what parameters influence the transport of oxygen.

Many of the lab-scale tests and most of the pilot-scale tests were done with the OLGA tar removal technology connected to the gasifier to clean the gas. The relatively high concentration of tar can be sufficiently reduced by the OLGA gas cleaning technology. The removed tar is used in the combustor of the MILENA to provide part of the required heat.

The MILENA Excel model was verified using data from different tests at lab and pilot-scale, using different fuels and different bed materials. Only small modifications were required. The MILENA model can now be used for “normal” operating conditions (gasification temperature between 770 and 880°C and atmospheric pressure) using woody fuel.

Empirical relations for hydrocarbon yields are always required to model a biomass fluidized bed gasifier. From literature only data for CH_4 yield was available. The relations for hydrocarbon yields that were determined for the MILENA gasifier can also be used for comparable biomass gasification processes (e.g. BFB and CFB gasification), but experimental verification is always recommended.

The calculated overall efficiency from wood with 25 wt.% of moisture to Bio-SNG is 68% on LHV basis. A further increase to over 70% is possible by integrating a biomass dryer that uses low temperature waste heat from the process.

8.2 Short term outlook

HVC and ECN are preparing a MILENA demonstration gasifier, to be operated on demolition wood B (painted wood), in combination with OLGA gas cleaning. The cleaned gas will be used in a gas engine to produce heat and electricity. The Bio-CHP demo is considered to be a crucial intermediate step towards commercial Bio-SNG plants. After a successful CHP demonstration further scale-up to a 50 MW_{th} SNG demonstration unit is foreseen.

The basic engineering of the MILENA demo plant is finished. The final decision for realization will be taken after a 1000 hour duration test in the

MILENA and OLGA pilot-scale installation at ECN, using demolition wood as a fuel. The demo plant will be built in Alkmaar next to the waste incinerator and the biomass combustion plant of the HVC group.

Several suppliers of major parts of the MILENA gasifier were consulted to make an accurate cost estimate for a commercial scale MILENA reactor. This cost estimate has shown that a Bio-CHP configuration based on the MILENA technology is economically viable if the Dutch subsidy on sustainable electricity (SDE) is available for this demonstration plant. The MILENA demonstration was designed for a net electrical output of approximately 3 – 3.5 MW_e. Residual heat will be used in the local heat grid. The demonstration plant is not optimized to electrical efficiency, because residual heat can be used.

Special attention was given to the steel insert which separates the gasification and combustion zone. The steel insert is exposed to high temperatures (approximately 900°C) under oxidizing and reducing conditions. A thermo-mechanical analysis was made using finite element software to check whether the structure could support the mechanical stresses for a prolonged period of time. Several suppliers were consulted. Different commercial materials were offered that are expected to be enough corrosion resistant under the operating conditions. Now the expected lifetime of the insert is more than a year. The mechanical design is made such that replacement of the insert is easy. One of the aims of the demonstration project is to test what lifetime can be achieved.

Figure 8-1 shows the integrated demo-plant with the fuel bunkers, MILENA gasifier, OLGA gas cleaning, start-up flare, gas engines and containers to test the upgrading of the gas into Bio-SNG.

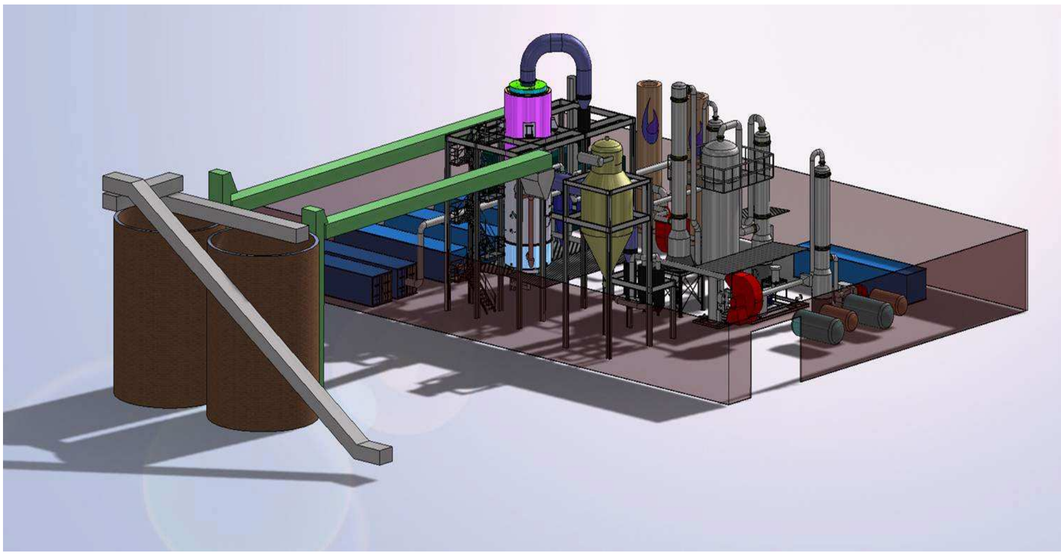

Figure 8-1: MILENA demo plant

After a successful demonstration period the MILENA Bio-CHP technology will be ready for commercial application. The next phase will be a commercial scale (50 MW_{th}) MILENA Bio-SNG demonstration.

8.3 Long term outlook

At the moment large scale biomass combustion plants (> 20 MW_e) have the highest efficiency to electricity (approximately 30%), but in practice use of the residual heat is difficult, because of the large scale. Small scale combustion plants have a higher overall efficiency (electrical + thermal), because their capacity is normally based on the local heat demand. Electrical efficiencies for small scale (<5 MW_e) biomass boilers are in general low with typical electrical efficiency between 10 and 25% on LHV basis.

Gasification offers the option to use gas engines for Combined Heat and Power production (CHP). The advantage of a gas engine is the high electrical efficiency, even at a small scale (< 5 MW_e). Power production is normally the economical driver for a Bio-CHP installation, so a high electrical efficiency is beneficial.

The electrical efficiency of Bio-CHP’s using gasification technology can vary significantly. The efficiency is strongly influenced by the type of biomass, water content of the biomass, scale of the installation, type of gasifier, type of gas engine, level of integration and emission limits. Figure 8-2 shows the calculated net electrical efficiencies for MILENA based Bio-CHP’s. The fuel is demolition wood with 20% moisture. The scale is based on the selection of two 2 MW_e gas engines Residual heat is used for district heating.

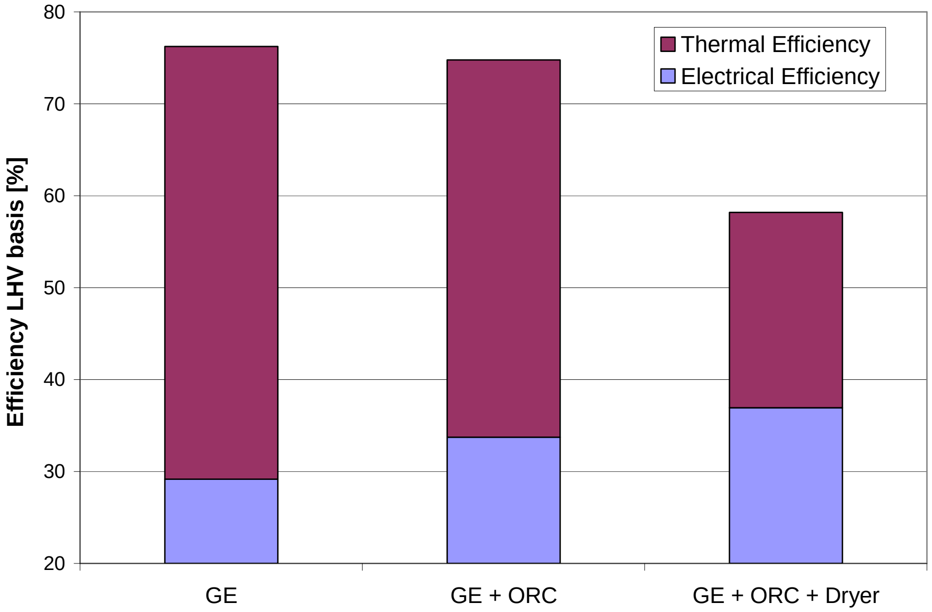

Figure 8-2: Calculated efficiencies of MILENA Bio-CHP configurations

The left column shows the efficiency assuming the producer gas is fired in a gas engine (GE). Adding an Organic Rankine Cycle (ORC) increases the electrical efficiency significantly. A further increase can be achieved by integrating a biomass dryer. The right column shows the calculated efficiencies assuming wood with 40% moisture is dried to 20%. The MILENA gas engine (+ORC) combinations offers a significantly higher efficiency than combustion based processes at a typical scale of 3 – 10 MW_e. For large scale applications a gas turbine in combination with a steam cycle needs to replace the gas engines, to further increase overall efficiency and reduce specific investment costs.

The production of Bio-SNG from woody biomass is the major driver for the development of the MILENA technology at ECN. The comparison of different biomass gasifiers in chapter 4 has shown the overall efficiency from wood to Bio-SNG will be significantly higher if an Indirect gasifier like the MILENA gasifier is used. The net overall efficiencies on LHV basis, including electricity consumption and pretreatment but excluding transport of biomass are 54% for Entrained Flow, 58% for CFB and 67% for Indirect gasification. Efficiency can be further increased by integration of a biomass dryer using waste heat from the process and by increasing the operating pressure of the gasifier.

Pressurized gasification can reduce or prevent energy consumption by a producer gas compressor. The investment cost of a large installation can also be reduced by pressurization because equipment will be smaller. A possible side effect is an increased yield of methane/other hydrocarbons at elevated pressure. Tests are required to verify this.

ECN plans to extend its work on MILENA pressurized gasification. The reactor concept is designed in such way that pressurization to a pressure between 3 and 7 bar is relatively simple, because the process takes place in one vessel.

The general interest is Bio-SNG is increasing. Several large utility companies are studying the option to produce Bio-SNG from woody biomass. A 20 MW_{th} demonstration using the FICFB gasification technology is prepared in Goteborg. The HVC ECN consortium plans to demonstrate a 50 MW_{th} SNG plant based on the MILENA gasification technology in 2015. Experimental work will focus on testing the required catalysts as well as final gas conditioning steps that are required for upgrading the gas into Bio-SNG.

Dankwoord (Dutch)

Vanaf deze plek wil ik iedereen bedanken die heeft bijgedragen aan de ontwikkeling van de MILENA vergasser en de realisatie van dit boekje.

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De realisatie van de lab-schaal en pilot-schaal MILENA installaties is mogelijk geworden door de inzet van de Engineeringafdeling van ECN, met bijzondere bijdragen van Frans, Ronald, Marcel, Nicolas, Ruud en Marco. Hiervoor mijn dank.

Geen biomassa proefinstallatie komt in bedrijf zonder de inzet van een team gedreven technici die alle ontwerpfouten van de 'uitvinder' weer mogen corrigeren. Voor al deze correcties, het meedenken bij het oplossen van problemen en alle nachtdiensten wil ik Jan, Marco, Herman, Dennis, Edwin, Johan, Ruud, Lucas en Stefan bedanken.

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Op naar de volgende MILENA!

Curriculum Vitae

Christiaan van der Meijden was born on November 21st, 1969 in Den Helder, The Netherlands. In the beginning of 1994 he finished his study Mechanical Engineering (BSc) at the Polytechnic of Alkmaar.

After his study he worked for two years as a commissioning and testing engineer for Stork Boilers in Hengelo.

From 1996 to 1999 Christiaan worked for the ECN Biomass Operator group as assistant group leader and from 2000 to 2004 as group leader. He was directly involved in the development of biomass gasification (Circulating Fluidized Bed) and gas cleaning equipment. The experiences gained with operating and modifying the pilot-scale gasification facilities resulted in the first (patented) designs for an allothermal gasification concept with an integrated tar reduction zone in 1998 (the STAR reactor). This concept was later adapted to the present MILENA design.

In 2004 he became a project leader in the group SNG and Syngas and became responsible for the development of the new MILENA biomass gasification technology. This has led to the realization of a lab-scale MILENA gasifier in 2004 and an 800 kW_{th} pilot plant in 2008.

At present Christiaan is the project leader of the experimental program in the MILENA pilot gasifier and is responsible for the engineering of the 10 MW_{th} MILENA demonstration plant.

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