Aleksi Pirvola

Production of synthetic natural gas from different biomass
Tekijä Aleksi Pirvola

Työn nimi Synteettisen maakaasun tuottaminen eri biomassoista

Maisteriohjelma Advanced Energy Solutions Koodi ENG30

Työn valvoja Professori Mika Järvinen

Työn ohjaaja(t) Tekniikan tohtori Timo Laukkanen

Päivämäärä 28.3.2020 Sivumäärä 66+1 Kieli englanti

Tiivistelmä


Avainsanat Kaasutus, bio-SNG, biomassa, prosessisimulointi, Aspen Plus
**Abstract**

Increasing the share of renewable transport fuels is stated as a target in the Renewable energy directive (RED II), which obliges all EU nations to provide solutions for cleaner transportation. Bio-SNG, a synthetic natural gas produced from biomass, offers a solution for large-scale production of biofuels. This thesis presents the technology required for bio-SNG production via gasification and catalytic methanation. Potential biomasses were analyzed for bio-SNG production in Finland. Performance of two biomass is studied with Aspen Plus process simulation.

Research of bio-SNG production technology showed that the technology is mostly mature although there is little experience from the process itself. Aspen model of two biomasses proved to be reasonably accurate illustration of a real demonstration plant. Biomass analysis concluded that the superior feedstocks for bio-SNG production are forestry byproducts, followed by wheat straw from agriculture. Aspen model concluded that the performance of woody biomass and straw is similar, while straw has slightly better process efficiencies.

**Keywords** Gasification, bio-SNG, biomass, process simulation, Aspen Plus
Forewords

The subject of gasifying biomass first raised my interest during my bachelor studies when I studied the subject at Gasum Oy. Since then, I have thought about writing my thesis on bio-SNG. It became a quite relevant topic as the European regulations demand all countries to increase the production of renewable fuels. Thanks to great teachers at Aalto University, I learned to model energy processes with simulation software and decided to apply it to my thesis. Writing this thesis was a tough challenge but I am very glad I chose this topic.

First, I would like to thank my thesis advisor Timo Laukkana, who was very supportive during the whole process. His knowledge of the process and Aspen modelling was important in constructing the model in the latter part of this thesis. I would also like to thank professor Mika Järvinen who supervised this thesis and guided me to contact Timo in the first place.

I would also like to thank my previous colleagues at Gasum Oy, Viljami Kinnunen and Inkeri Kauppi for introducing me to the subject of producing bio-SNG and the recent developments in industry in producing it from biomass.

Lastly, I would like to thank my parents, Juha-Pekka and Katriina, for always being on my side and supporting me especially during this last year. Thanks also to my mates at Aalto University, who have accompanied me during the whole journey in my studies at Aalto.

Espoo 28.3.2020

Aleksi Pirvola
# Table of content

Tiiivistelmä
Abstract
Forewords
Table of content ........................................................................................................ 5

1 Introduction ........................................................................................................... 7
   1.1 Background and motivation .......................................................................... 7
   1.2 Aim of the thesis ......................................................................................... 8
   1.3 Methods and scope .................................................................................... 8
   1.4 Structure of the thesis ................................................................................. 9

2 Production of bio-SNG ....................................................................................... 10
   2.1 Origins of synthetic natural gas ................................................................... 10
   2.2 Gasification technology ............................................................................. 11
       2.2.1 Fluidized bed gasification .................................................................... 13
       2.2.2 Direct fluidized bed gasification ......................................................... 13
       2.2.3 Indirect fluidized bed gasification ....................................................... 15
   2.3 Gas cleaning technology ............................................................................. 19
       2.3.1 Particulate matter ............................................................................... 19
       2.3.2 Tar ....................................................................................................... 20
       2.3.3 Sulfur .................................................................................................. 21
   2.4 Methanation technology ............................................................................. 22
   2.5 Efficiency of bio-SNG plant ...................................................................... 26
   2.6 Commercial and demonstration bio-SNG plants ...................................... 28

3 Biomass for bio-SNG production ..................................................................... 32
   3.1 Forest biomass .......................................................................................... 33
   3.2 Agricultural biomass .................................................................................. 36
   3.3 Industrial and municipal waste ................................................................. 38
   3.4 Renewable energy directive .................................................................... 40
   3.5 Availability of biomass in Finland ............................................................ 42

4 Simulation of bio-SNG process ...................................................................... 46
   4.1 Bio-SNG process model ............................................................................ 46
   4.2 Biomass for simulation ............................................................................. 53

5 Results ..................................................................................................................... 54
   5.1 Efficiency and energy balance ................................................................... 57
   5.2 Gas composition ....................................................................................... 58

6 Conclusions ......................................................................................................... 61
Bibliography ........................................................................................................... 63
Appendix. Aspen model for bio-SNG production ............................................... 67
# Abbreviations

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>ASU</td>
<td>Air separation unit</td>
</tr>
<tr>
<td>BFB</td>
<td>Bubbling fluidized bed</td>
</tr>
<tr>
<td>Bio-SNG</td>
<td>Substitute/synthetic natural gas produced from cellulosic biomass</td>
</tr>
<tr>
<td>CFB</td>
<td>Circulating fluidize bed</td>
</tr>
<tr>
<td>DFB</td>
<td>Dual fluidized bed</td>
</tr>
<tr>
<td>ECN part of TNO</td>
<td>Energy Research Centre of the Netherlands</td>
</tr>
<tr>
<td>EU</td>
<td>European Union</td>
</tr>
<tr>
<td>Gobigas</td>
<td>Gothenburg Biomass Gasification</td>
</tr>
<tr>
<td>HGF</td>
<td>Hot gas filtration</td>
</tr>
<tr>
<td>HTS</td>
<td>High temperature shift</td>
</tr>
<tr>
<td>ILUC</td>
<td>Indirect land use change</td>
</tr>
<tr>
<td>kWth</td>
<td>Thermal kilowatt</td>
</tr>
<tr>
<td>LTS</td>
<td>Low temperature shift</td>
</tr>
<tr>
<td>MDEA</td>
<td>methyl-diethanolamine</td>
</tr>
<tr>
<td>MSW</td>
<td>Municipal solid waste</td>
</tr>
<tr>
<td>MWth</td>
<td>Thermal megawatt</td>
</tr>
<tr>
<td>ppm</td>
<td>Particles per million</td>
</tr>
<tr>
<td>PR-BM</td>
<td>Peng-Robinson cubic equation of state with Boston-Mathias alpha function</td>
</tr>
<tr>
<td>RDF</td>
<td>Refuse derived fuel</td>
</tr>
<tr>
<td>RED</td>
<td>Renewable Energy Directive</td>
</tr>
<tr>
<td>SNG</td>
<td>Substitute/synthetic natural gas</td>
</tr>
<tr>
<td>SRF</td>
<td>Solid recovery fuel</td>
</tr>
<tr>
<td>vol-%</td>
<td>Volume percentage</td>
</tr>
<tr>
<td>VTT</td>
<td>Teknologian tutkimuskeskus VTT Oy</td>
</tr>
<tr>
<td>WGS</td>
<td>Water-gas shift</td>
</tr>
<tr>
<td>WGSR</td>
<td>Water-gas shift reactor</td>
</tr>
<tr>
<td>wt-%</td>
<td>Weight percentage</td>
</tr>
</tbody>
</table>
1 Introduction

1.1 Background and motivation

Global warming and methods to reduce it are discussed widely and often in almost all countries, political instances and media. Commonly, the focus is either on reducing the consumption of energy in households, industry and transportation, increasing energy efficiency, or changing the production methods of energy. Energy production is starting to shift from traditional fossil fuels such as coal and natural gas to renewable technologies such as wind power and photovoltaics for electricity generation and ground heat, heat pumps and biomass combustion for heat production. Alternative fuels for transportation can be used to mitigate the emissions it produces. Renewable electricity, hydrogen and biofuels can be used to replace traditional fuels such as gasoline and heavy fuel oils.

Biofuel production in Europe has been widely addressed recently in the European Commission. The new Renewable Energy Directive (RED II) was introduced in June 2018. The directive sets limits on the biofuels that are produced from food and feed crops and requires the member nations to increase their share of renewable transport fuels. The timeline for achieving the targets set in RED II are illustrated in Figure 1. By the end of 2023, all countries in European Union must produce 7 – 14 % of their transport fuels from renewable sources and limit their use of food and crop-based biofuel use to 7 %. (European Commission 2018). Therefore, the production of biofuels such as biogas and renewable diesel must be increased in the EU in the coming years.

Figure 1. EU RED II phasing-out timeline for crop based biofuels (European Commission 2018)

In Finland, the national target is to increase the share of biofuels in transportation gradually to 30 % by 2030. This target is mainly targeted for renewable diesel, as the amount of ethanol
mixed in gasoline cannot be increased further from current. Furthermore, biogas is not included in the national biofuel distribution target as it is exempted from taxation. In their report, the Ministry of Employment and Economy suggested in 2018 that biogas should be included in taxation and in the national biofuel target (Työ- ja elinkeinoministeriö 2018). Biogas is currently produced by anaerobic digestion from biowaste and sewage sludge in Finland. In addition, synthetic natural gas produced from biomass (bio-SNG) has been researched recently. Bio-SNG is comparable to natural gas flowing in Finnish transmission network. It consists mainly from methane and contains little impurities. A few demonstration plants in Sweden and the Netherlands have been erected to develop technology for producing bio-SNG from biomass feedstocks.

Producing bio-SNG is considered a potential alternative for anaerobic digestion of biowaste. However, the technology required for producing bio-SNG is much more complicated than with anaerobic digestion. Thus, in order to produce it with a feasible cost, a cheap feedstock is fundamental. Feedstocks for producing bio-SNG are limited also by the new RED II, which determines the feedstocks that are acceptable for a biofuel to be considered a renewable fuel. Furthermore, the increasing amount of biorefineries in Finland increases competition for feedstocks such as wood chips and wood side streams from forest industry. A solution for producing bio-SNG for competitive cost is utilizing alternative feedstocks generally ignored in the energy industry. Biomass such as straw or a feedstock that is locally available in large amounts could prove to be beneficial compared to more expensive and competed feedstocks.

1.2 Aim of the thesis

The aim of this thesis is to study the technical potential of different biomass feedstocks to produce bio-SNG through gasification and methanation processes by comparing their performance in a simulated process. By comparing different alternative biomass feedstocks, this thesis will highlight the most potential feedstocks for bio-SNG production in terms of cost and technical feasibility. The end result will be a comparison of the different potential feedstocks for producing bio-SNG, including the energy and mass balance based on the simulation, the energy efficiency of the process and the cost of producing bio-SNG.

1.3 Methods and scope

In the thesis, recent progress in biomass gasification technology is studied from literature, focusing on the technology that has been used for producing bio-SNG as a product. Commercial methanation technologies are listed to choose the best suited option for the simulation. Biomass feedstocks available in Finland are studied from multiple perspectives. Their geographical availability, chemical and physical properties and cost are included in the research. Finally, the simulation of the process is performed with Aspen plus -software. The simulation process is based on the most suitable combination of gasification and methanation for producing bio-SNG.

The scope of this thesis is limited to relatively small scale of bio-SNG production plants with output of under 50 MW of bio-SNG. The gasification technology will be limited to fluidised
bed gasifiers. This is because most of the experimental data from producing bio-SNG focuses on bubbling or circulating fluidised beds. The cleaning or production of tar during the gasification is not considered, nor are cleaning processes of other gas impurities such as hydrogen sulphides or carbon dioxide.

1.4 Structure of the thesis

The rest of the thesis is arranged as follows. Chapter 2 reviews the literature illustrating the gasification and methanation processes used in producing bio-SNG. Chapter 3 describes and reviews the different biomass feedstock that can be used in producing bio-SNG. Chapter 4 describes the process that is chosen for the simulation part. Chapter 5 illustrates the results obtained from the simulation/calculations with the selected process and feedstocks. Chapter 6 draws the most fundamental conclusions of the work done in the thesis.
2 Production of bio-SNG

Synthetic natural gas production from biomass utilizes mature technology that originates from several decades into the past. Multiple technologies have been developed to decompose solid matter containing carbon and hydrogen and to join these components together to form methane. This chapter reviews the recent developments of these technologies that are the most fundamental in producing synthetic natural gas from biomass.

2.1 Origins of synthetic natural gas

The concept of producing synthetic natural gas is originally based on making SNG from coal through gasification and methanation processes. The idea of producing SNG from coal arose in the United States in the 1960s when the demand of natural gas increased. To prepare for shortages of natural gas, producing SNG from the United States’ vast resources of coal became a viable option. Furthermore, the oil crisis in the 1970s accelerated the development of the SNG production technology further. Another incentive to produce SNG during ‘70s was to produce cleaner energy carrier from locally abundant coal reserves (Schildhauer, Biollaz 2016). Especially countries with large coal reserves have since shown interest in producing SNG from coal, including Germany, and United Kingdom. These efforts resulted in one coal-to-SNG plant to be erected in the United States. The Great Plains Synfuels plant in North Dakota was commissioned in 1984. The plant produces 4.8 million cubic meters of SNG per day, which accounts for approximately 1.5 GW of SNG. (Kopyscinski, Schildhauer et al. 2010).

The North Dakota plant has remained the only large commercial plant in western countries for a few reasons. After the initial interest in SNG in 1970s, the oil price dropped, new natural gas reserves were discovered in the North Sea, and new gas pipelines were built from Russia to Europe. Despite some resurfaced interest in SNG in the early 2000s, the Great Plains plant is the only coal-to-SNG plant in commission in Europe or the Americas. Rapid increase in shale gas production worldwide has decreased the attraction to erect more plants converting coal to natural gas. However, China has been keen on constructing more SNG production from coal, as it has large reserves of coal and little natural gas production. It also requires cleaner fuels to limit the local emissions in the more densely populated areas. Thus, China has built three large coal-to-SNG plants and is planning to build more. (Schildhauer, Biollaz 2016). Chinese National Development and Reform Commission has declared that coal-to-gas production of SNG should reach 1200 TWh by 2020 (International Energy Agency 2016).

However, the interest towards SNG production from biomass has increased in Europe during the recent two decades. There are several reasons for this development: First, the recent renewable energy directive from European Commission requires its member countries to increase the share of renewable fuels used in transport in the coming years. Secondly, many countries pursue increasing the share of domestic biomass in their energy production to decrease emissions, energy security and imported fuels (Schildhauer, Biollaz 2016). Furthermore, SNG production offers easier form transportation of biomass-based energy than traditional lingo-cellulosic biomasses such as wood. In addition, bio-SNG can be used as a
balancing energy source in the more volatile energy networks and as a form to store renewable energy for long periods of time.

Some demonstration projects focused on bio-SNG production have been commissioned in Europe. MILENA-project in the Netherlands and Gobigas-project in Sweden are the largest examples of applying similar technology used in coal-to-SNG plants to produce SNG from biomass. The technology has been studied in some projects in Finland as well, for instance during a collaboration between Gasum, Helen and Metsä Fibre in early 2010s. These projects demonstrating bio-SNG technology will be presented in chapter 2.6.

2.2 Gasification technology

Gasification is the most important single step in producing synthetic natural gas. Therefore, it will be a fundamental aspect of this thesis. Gasification is a complicated thermodynamic process that consists of several reactions between different phases and reactants. It can be described as a process which transforms the solid feedstock to different gaseous products in high temperature. The process can be performed in atmospheric pressure, or it may be pressurized up to several dozen atmospheres. In addition to the solid matter, a gaseous gasifying agent is injected into the gasifier. This gasifying gas can be either air, oxygen or steam. The resulting gaseous product is typically called syngas or product gas.

Gasification process can be divided into three different stages: drying, pyrolysis, and gasification. These process steps are illustrated in Figure 2, which also depicts the different products that are formed in each step. The initial stage of the gasification is drying at low temperatures of under 100 °C. During this stage, the moisture evaporates from the feedstock. The evaporation of water and the evaporated water requires large amounts of energy, which makes low moisture content of the feedstock crucial (van Loo, Koppejan 2008). During pyrolysis, the dried feedstock releases its volatile compounds as tars and gases such as hydrogen, carbon monoxide and carbon dioxide. Tars are heavy hydrocarbons, typically aromatic compounds with 1 to 5 rings. The remaining solids produce carbon-rich char. Most of the pyrolysis reactions take place at temperatures between 200 and 400 °C.
Ultimately, the gasification stage takes place at 800 – 1100 °C in the presence of oxidizing (gasifying) agent. During this stage of thermal devolatilization, most of the remaining char and tar are gasified into small gaseous products. The composition of the product gas varies greatly depending on the process parameters. Variables such as gasifying temperature, pressure, moisture content of the feedstock and gasifying agent have a large effect on resulting syngas and they must be chosen accordingly depending on the designed use of the product gas.

Most important reactions in the gasification stage are:

### Table 1. Gasification reactions (Schildhauer, Biollaz 2016)

<table>
<thead>
<tr>
<th>#</th>
<th>Name of the reaction</th>
<th>Reaction</th>
<th>Reaction heat (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Partial oxidation</td>
<td>C(s) + 0.5O₂(g) ↔ CO(g)</td>
<td>-111</td>
</tr>
<tr>
<td>2</td>
<td>CO combustion</td>
<td>CO(g) + 0.5O₂(g) ↔ CO₂(g)</td>
<td>-283</td>
</tr>
<tr>
<td>3</td>
<td>Carbon combustion</td>
<td>C(s) + O₂(g) ↔ CO₂(g)</td>
<td>-394</td>
</tr>
<tr>
<td>4</td>
<td>Reverse Boudouard</td>
<td>C(s) + CO₂(g) ↔ 2CO(g)</td>
<td>+172</td>
</tr>
<tr>
<td>5</td>
<td>Water gas</td>
<td>C(s) + H₂O(g) ↔ CO(g) + H₂(g)</td>
<td>+131</td>
</tr>
<tr>
<td>6</td>
<td>Water-gas shift</td>
<td>CO(g) + H₂O(g) ↔ H₂(g) + CO₂(g)</td>
<td>-41</td>
</tr>
<tr>
<td>7</td>
<td>Steam reforming</td>
<td>CH₄(g) + H₂O(g) ↔ 3H₂(g) + CO(g)</td>
<td>+206</td>
</tr>
</tbody>
</table>

The product gases in all the reactions in Table 1 are carbon monoxide, carbon dioxide and hydrogen. The reactions 1 – 3 illustrate partial oxidation and combustion reactions which are exothermic. Reactions 4, 5 and 7 are endothermic, while the water-gas shift reaction is slightly exothermic. The reactions requiring oxygen are exothermic, which is an important factor in the heat balance of the gasification process. If air or oxygen is chosen as the gasifying agent, the oxidation reactions can produce enough heat to devolatilize the solid char. However, if the gasifying agent is steam, the concentration of oxygen is lower. Therefore, an auxiliary source of heat is needed to gasify the char into gaseous products. This is one of
the most fundamental difference in the available gasification technologies. If the heat for
gasification is produced with only the feedstock and the gasifying agent, the process is called
direct or autothermal gasification. When an extra heat source is needed for the gasification,
the process is indirect or allothermal (Schildhauer, Biollaz 2016).

2.2.1 Fluidized bed gasification

Fluidized bed gasification is the most common form of gasifying biomass. First applications
of fluidized beds in biomass combustion emerged in the 1960s (van Loo, Koppejan 2008),
and they have since been used in many biomass combustion and gasification plants in Fin-
land. The technology has several advantages over traditional combustion methods, most of
which are related to the physical and chemical characteristics of biomass.

Fluidized beds consist of a reactor with a perforated floor with nozzles to blow air, oxygen
or steam upwards. An inert bed material is used in the reactor to ensure an even heat distri-
bution. The bed material is typically granulated silica or dolomite, although olivine has
been used in some biomass applications (Schildhauer, Biollaz 2016). The bed material forms
an even layer to the bottom of the reactor. Fuel for gasification is fed on top of the bed
material, which is fluidized by air, oxygen or steam. This fluidization of the bed mixes the
fuel and bed material forming an even mixture. The combustion temperature of fluidized
beds varies from 650 to 900 °C. This is due to the low ash melting temperatures of typical
biomass feedstocks: low temperature prevents the ash from agglomerating inside the reactor.
(van Loo, Koppejan 2008).

There are two types of fluidized beds used for biomass gasification: circulated (CFB) and
bubbling fluidized beds (BFB) and combinations of the two. In circulating fluidized beds,
the fluidization velocity is larger, which leads to higher turbulence of the fuel and bed ma-
terial particles. This ensures higher heat transfer inside the reactor and increases the fuel
flexibility concerning biomass type and moisture content. Bubbling fluidized beds have
lower fluidization velocity than CFB. The advantage of using BFBs is the possibility to use
moister fuels with larger particle size. Both of these technologies are susceptible to the ag-
gglomeration of bed material with biomass ash if too high temperatures are used. (van Loo,
Koppejan 2008)

2.2.2 Direct fluidized bed gasification

Direct gasification is a gasification technology which uses partial oxidation (combustion) to
gasify solid feedstock into product gases. Direct gasification can be operated with both bub-
bling and circulated fluidized beds. It uses pure oxygen as fluidizing agent, which reacts with
part of the feedstock releasing heat as a result. Steam can be added into the gasifier in addi-
tion to oxygen, which increases hydrogen content in the product gas. (Rauch, Hrbek et al.
2014)

Oxygen required for the process is typically produced with cryogenic air separation units
(ASUs), which separate oxygen from air. The oxygen used for SNG production process
needs to be almost pure, since nitrogen content of the product gas must be very low. Low
nitrogen content in product gases are enriched towards the end of the process, where it is
more expensive to remove. ASUs require large amounts of electricity (0.2 – 0.3 kWh/kgO2) and have large investment costs (Niskanen 2012).

All of the heat needed for the gasification process is produced in the gasifier during direct gasification. Oxygen is fed into the gasifier with lower concentration that is required for stoichiometric combustion of biomass. Therefore, the biomass is only partially combusted, and the rest reacts with other gases such as steam and carbon oxides. Oxygen feed is controlled in order to produce enough heat for the gasification reactions. Too low amount might lead to too low gasification temperatures and incomplete conversion of biomass. However, too high oxygen content leads to high amounts of carbon dioxide and high temperatures, which can cause issues with ash agglomeration in the fluidized bed reactors. Normal gasification temperature in autothermal gasification with a fluidized bed reactor is between 850 and 900 °C.

Direct fluidized bed gasifiers can be used under pressurized conditions, which is beneficial in larger scale applications. Direct gasifiers have a range of several hundred megawatts of thermal input, which would be difficult to achieve with atmospheric conditions. The size of the gasifiers can be reduced with pressurization (Schildhauer, Biollaz 2016). Therefore, material and investment costs can be decreased with direct gasifiers. Pressurization of gasification also saves energy from compression of product gas after gasifier (Higman, Burgt 2008).

The composition of syngas produced with direct gasification varies with the feedstock and the process parameters. In general, direct gasification of biomass produces synthesis gas with relatively low content of tar, 1 – 20 g/m³. Most of the product gas consists of carbon dioxide, carbon monoxide and hydrogen. Rauch et al. (2014) reviewed the typical gasification technologies and the product gas compositions with oxygen and steam fluidization. The typical dry product gas consists of 20 – 30 vol-% of carbon monoxide and hydrogen and 25 – 40 vol% of carbon dioxide. Methane content is between 5 and 10 vol-%.

Industrial size fluidized bed reactors suitable for gasification are provided by Andritz, Valmet and Sumitomo SHI FW, formerly Foster Wheeler. In Finland, these gasifiers have been used to replace the use of fossil fuels by gasifying biomass. In Vaasa, circulating fluidized bed have been built to gasify woody biomass such as bark, forest residues and stumps. The 140 MWth CFB gasifier by Valmet has been in use since 2013, and the product gas is used to replace coal use in pulverized coal boiler. This Vaskiluoto gasification plant is currently the largest existing biomass gasifier. In Lahti, two 80 MWth atmospheric gasifiers were built in 2012 to gasify RDF and contaminated wood. In addition, biomass gasifiers have been built at paper and pulp mills to produce product gas for lime kilns. Three gasifiers utilizing bark and wood residues have been built to replace oil and gas used to heat the kilns. (Isaksson 2018).
2.2.3 Indirect fluidized bed gasification

Allothermal or indirect gasification of biomass is another significant technology that has been developed for producing syngas for biofuel production. The process has multiple important differences to direct gasification: reactor setup, heat source, gasification agent, and product gas composition.

Indirect gasification technology is based on the concept of separating the gasification and combustion reactions. The biomass is first introduced in the gasification reactor, where it is partially gasified in high temperature. Opposed to direct gasification, relatively high amount of carbonaceous char remains unreacted during the process. The remaining char flows to combustion reactor, where it is combusted with air. In both of these reactors, an inert bed material is used to fluidize the bed and transfer between the fuel particles. Indirect gasification utilizes the bed material also for heat transfer between the combustion and gasification reactors. When char is combusted, it heats the bed material to a high temperature. The heated bed material is transported to the gasifier, where it releases the heat to the biomass fuel.

Due to the relatively complex mass flows between the reactors, indirect gasification is typically considered only in atmospheric pressures. This limits the scale of bio-SNG production plants to under 200 MWth, as reactor size would increase significantly with several hundred megawatt fluidized bed reactors. However, the procurement and supply chain of biomass naturally limits the scale of biomass use in power plants, which reduces the significance of the problem. (Schildhauer, Biollaz 2016).
Separating the gasification and combustion processes also allows the use of other gasification agent than pure oxygen. In indirect gasification, superheated steam can be used to fluidize the gasifier. Therefore, one of the benefits of the process is the ability to produce syngas suitable for methanation without requiring production of pure oxygen (Rauch, Hrbeck et al. 2014). Oxygen production in industrial scale requires expansive air separation units, which can be replaced with relatively inexpensive steam flow. Using steam as fluidizing agent also produces more hydrogen-rich product gas, which is beneficial for methanation process after gasification.

The product gas comprises of the same compounds as with direct gasification: carbon monoxide, carbon dioxide, hydrogen, and methane. Rauch et al. (2014) determines the approximate indirect gasification product gas to contain 20 – 25 vol-% of carbon monoxide 20 – 25 vol-% of carbon dioxide, 30 – 45 vol-% of hydrogen and 6 – 12 vol-% of methane. The tar content is estimated to be 1 – 10 g/Nm³. Therefore, there is a clear distinction between direct and indirect gasification product gas. With allothermal gasification, the higher amount of hydrogen and lower amount of carbon dioxide are produced in the gasifier.

A few indirect gasification technologies have been demonstrated in small-scale. All the technologies associated with indirect gasification have been aimed to produce biofuels via catalytic synthesis. Valmet, formerly Metso, delivered a dual-fluidized bed gasification equipment to Gothenburg for a bio-SNG demonstration plant. The gasifier comprised of a circulated fluidized bed that was used for combustion and a bubbling fluidized bed that was utilized for gasifying the biomass. This project Gobigas used Valmet’s technology to gasify woody biomass with 32 MWth gasification system. Another indirect gasification process has been developed by Energy Centre of the Netherlands (ECN). The gasifier concept called MILENA utilizes a novel combination of bubbling and circulating fluidized bed, where the biomass is combusted and gasified inside a single vessel. The technology has been demonstrated in India, where a 4 MWth gasifier was used to produce clean product gas from waste biomass. (Waldheim 2018).

The Gobigas gasifier concept is illustrated in Figure 4. The concept was studied in Chalmers University of Technology, where the dual fluidized bed gasifier (DFB) has been studied since 2007. In the gasifier, the primary feedstock is first fed to the gasifier (14), and the remaining char is transferred to the CFB combustor (1). The bed material from the combustor is directed into the BFB gasifier to heat the feedstock and to decompose it to product gases. The flow of the bed material is controlled by having different height pipes connecting the CFB and BFB reactors in the particle distributor (1). This way, the flow of bed material can be controlled to bring enough heat to the gasifier. Surplus bed material is flown back into the combustion section. The bed material used was olivine, which was chosen for its reaction activity enhancing properties. (Thunman, Seemann et al. 2018).
The gasifier in demonstration plant of Gobigas-project was tested with wood pellets and chipped wood during 2014 – 2018. Composition of product gas with wood pellets as feedstock is provided in Table 1. The fluidization agent used during the tests was superheated steam, and the gasification temperature was 870 °C. The results show that the hydrogen concentration is high, nearly 40 vol-% on dry basis; CO-concentration 24.0 vol-%; CO2-concentration 19.9 vol-%; and CH4-concentration 8.6 vol-% (Thunman, Seemann et al. 2018). Therefore, the results from Gobigas demonstration plant are similar as Rauch et al. (2014) estimated. All the concentrations are within the estimated range except CO2, which has a slightly larger concentration than Rauch et al. predict. The tar content in product gas is under 20 g/Nm³, mainly consisting of benzene and naphthalene.

Table 2. Product gas composition from Gobigas gasifier with wood pellets (Thunman, Seemann et al. 2018)

<table>
<thead>
<tr>
<th>Component</th>
<th>Wet gas (vol-%)</th>
<th>Dry gas (vol-%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H2</td>
<td>27.7</td>
<td>39.9</td>
</tr>
<tr>
<td>CO</td>
<td>16.6</td>
<td>24.0</td>
</tr>
<tr>
<td>CO2</td>
<td>13.8</td>
<td>19.9</td>
</tr>
<tr>
<td>CH4</td>
<td>6.0</td>
<td>8.6</td>
</tr>
<tr>
<td>C2H4</td>
<td>1.4</td>
<td>2.0</td>
</tr>
<tr>
<td>C2H2, C2H6, C3H6</td>
<td>0.2</td>
<td>0.3</td>
</tr>
<tr>
<td>Inertization gas, N2</td>
<td>3.7</td>
<td>5.3</td>
</tr>
<tr>
<td>H2O</td>
<td>30.2</td>
<td>-</td>
</tr>
</tbody>
</table>
MILENA gasifier concept is illustrated in Figure 5. The most significant difference to the DFB gasifier used in Gothenburg is the reactor design which combines the combustion and gasification reactors into single vessel. The combustion reactor, a bubbling fluidized bed, surrounds the circulated fluidized bed gasifier. This reduces the heat losses related to combustion process. Biomass is fed to the bottom of the CFB riser, where most of it is gasified with steam. The solid char and product gases are separated at the top of the reactor, where the solid char flows downwards to the BFB combustion reactor with the bed material. The bed material can be either olivine or sand. (Waldheim 2018).

![MILENA gasifier diagram](image)

*Figure 5. MILENA gasifier (Waldheim 2018)*

MILENA gasifier is designed to be used with waste material such as RDF or SRF as well as different biomass. With woody biomass, the gasifier is operated at 850 °C, while it is slightly lower for waste feedstock. The typical dry gas composition of MILENA gasifier is illustrated in Table 3. The data is for woody biomass with steam as fluidizing agent. The product gas composition differs significantly from Gobigas results as well as Rauch et al. (2014) estimations. The hydrogen concentration is lower, whereas the carbon monoxide and methane concentrations are high. In addition, the tar content of the gas is relatively high, 40 g/Nm3. (Waldheim 2018). The differences in gas composition might be due to a slightly different temperature during gasification and lower amount of fluidization steam. Furthermore, the heat transfer characteristics are different than in the gasifier used in Gobigas which might affect the volatilization of biomass in MILENA.
Table 3. MILENA gasifier product gas with woody biomass (Waldheim 2018).

<table>
<thead>
<tr>
<th>Component</th>
<th>Dry gas (vol-%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂</td>
<td>24</td>
</tr>
<tr>
<td>CO</td>
<td>34</td>
</tr>
<tr>
<td>CO₂</td>
<td>17</td>
</tr>
<tr>
<td>CH₄</td>
<td>15</td>
</tr>
<tr>
<td>C₂H₄</td>
<td>5</td>
</tr>
<tr>
<td>CₓHᵧ, NH₃, H₂S</td>
<td>1</td>
</tr>
<tr>
<td>N₂</td>
<td>3</td>
</tr>
</tbody>
</table>

2.3 Gas cleaning technology

Syngas produced by gasification of biomass contains various impurities in addition to the primary components. Tars, particulate matter, ammonia, sulfur compounds, hydrogen chloride and alkali metals are typical impurities that are encountered in the product gas. These compounds need to be removed from the gas in order to produce synthetic natural gas from the syngas. Otherwise, the impurities will reduce the performance of the downstream equipment (Schildhauer, Biollaz 2016). Compounds containing sulfur can poison catalysts used in methanation stages and tars can condense in heat exchanger causing equipment fouling. Furthermore, carbon dioxide must be separated from the product gas to meet the quality requirements for natural gas in the gas network. The most important gas cleaning technologies in terms of bio-SNG production via gasification are discussed in this section.

2.3.1 Particulate matter

Removal of particulate matter is one of the most crucial gas cleaning phases in biomass gasification processes (Simell, Hannula et al. 2014). Particulate matter is always present in gasifier product gases. They can cause erosion and fouling in the downstream equipment and thus must be removed. Particles of carbonaceous char, ash, and bed material such as olivine or dolomite exist in the gases after gasifier. Ash particles are formed of salts that can be either gaseous, liquid or solid (Schildhauer, Biollaz 2016).

Technologies to remove particulate matter include scrubbers, cyclones, electrostatic precipitators and different filters. Some of these technologies can be operated only at cooler temperatures than which the gas leaves the gasifier. Scrubbers operate typically at room temperature and most bag filters must be used in temperatures under 300 °C. Decreasing the temperature of the product gas before filtration can lead to problems with tar condensation. Low temperatures can cause the condensation of tars in the dust cake, which decreases the lifetime of the filtration system. Cyclones are commonly used in biomass gasification, as they are normally integrated to fluidized bed boilers. (Schildhauer, Biollaz 2016). Utilizing fluidized bed reactor results in high dust load in product gas, which is why removal of particulate matter is important in bio-SNG processes.

A technology that has been developed to increase the efficiency of particulate matter removal is hot gas filtration (HGF). Hot gas filtration enables cleaning of gases in temperatures close to the gasifier, around 800 – 900 °C. Therefore, heat exchangers are not exposed to the dust
load from gasification as the product gas can be filtered without primary cooling after gasifier. Due to the high operation temperature of HGF systems, there is no condensation of tar in the filter. Furthermore, hot gas filtration mitigates the amount of alkali, heavy metal and chloride compounds in the cleaned gases. HGF systems have been developed by VTT and several other researchers since 1980s. Simell et al. (2014) reviewed the research made by VTT concerning the gas cleaning processes. They concluded that the presence of steam in the gases increased the performance of the HGF system, which might be beneficial for indirect gasifiers that use steam as gasifying agent. However, both Simell et al. and Heidenreich (2013) conclude that more long-term testing is needed to prove the benefits of the technology.

2.3.2 Tar

Tar is a major issue for biomass gasification processes with synthetic fuel conversion which require very low concentration of tar. Although there is no exact definition for tar, they are normally defined as aromatic hydrocarbons with one or multiple rings. These compounds have to be removed from the gases as they can condense in temperatures of under 450 °C (Schildhauer, Biollaz 2016). Primarily, the tar formation can be limited by utilizing a catalytically active bed material. Dolomite and olivine has been shown to reduce tar in product gases and hinder sintering of the bed in fluidized bed reactors (Kaisalo 2017).

Two efficient technologies for tar removal have been recognized in biomass gasification: scrubbers utilizing organic solvents and catalytic reforming of tar into smaller hydrocarbon compounds (Simell, Hannula et al. 2014). Scrubbers with organic solvents collect tar compounds as aerosols which are formed as the tar starts to condense at lower temperatures. A wet scrubber process utilizing scrubbing oil has been developed by the Energy Research Centre of the Netherlands (ECN part of TNO). The process, called OLGA, separates light and heavy tars from the product gases in different stages. The process comprises of multiple scrubbers, as is illustrated in Figure 6. In the first stage, heavy tars are collected by scrubbing oil. Lighter tars are collected by the wet scrubber in the second stage. The heavy tars from the first stage are separated from the oil and recycled to the gasifier, while the oil used in the second stage is regenerated in the third stage of the process with steam. The steam and lighter tars from third reactor are recycled to gasifier. (Waldheim 2018).

TU Wien has developed a wet scrubber system for indirect gasification process utilizing rapeseed methyl ester, a biodiesel, for removing tar from product gas. Scrubber condenses the steam and tars in the gas, while tars are absorbed by the bio-oil. The oil is separated from water with gravity, and the tar-containing oil is recycled to the combustion reactor. (Waldheim 2018).
Catalytic steam reforming of tar is an efficient alternative for utilizing a scrubber for tar removal. It takes place at similar temperature as the product gas from the gasifier. Catalysts used for reforming are typically produced from nickel and precious metals. Nickel catalysts benefit from its low cost, commercial experience from steam reforming of methane and their activity in reforming tar. However, nickel is susceptible to coke formation. Precious metals are less prone for coke formation but are more expensive than nickel. (Kaisalo 2017). Furthermore, nickel is prone to sulfur poisoning in temperatures under 900 °C (Schildhauer, Biollaz 2016). Steam reforming of tar with catalysts is based on the dehydrogenation of tar on the catalyst surface, where the hydrocarbons in tar react with steam to form small gas compounds such as hydrogen, carbon monoxide and carbon dioxide. Most of the catalytic reforming of tar has been performed on small scale laboratory studies, which limits the current potential of the technology in large scale applications. Guan et al. (2016) conclude that in order to develop a feasible catalytic reformer of tar, more research is needed into coking mechanisms of heavy tar and novel catalyst materials for preventing the coking of catalyst.

### 2.3.3 Sulfur

Sulfur is especially harmful for catalysts used in methanation technology even in small amounts. Therefore, most of the sulfur in product gas must be removed before catalytic methanation of gasification syngas. Processes available for sulfur removal are wet scrubbing and solid sorption. Generally, both of these processes can absorb most of the H2S in the product gas. However, COS and other sulfur containing hydrocarbons are not removed efficiently and they may be detrimental for liquid solvents in wet scrubber and materials used in sorption. Hydrodesulfurization process can be used to hydrogenate the sulfur compounds into H2S, which can be then removed by scrubbers or sorption. (Schildhauer 2016). Hydrogen sulfide amounts to 90 % of typical biomass gasification product gas, while rest of the sulfur is mainly COS. Typical liquids used in wet scrubbers are amine based chemical solvents such as monoethanolamine (MEA), diethanolamine (DEA) and methyldiethanolamine (MDEA) or physical solvents such as methanol that is used in Rectisol process. The chemical
washing is based on absorbing and hydrolyzing H2S in the gas whereas physical washing is based on dissolving sulfur compounds into methanol. (Waldheim 2018).

Additionally, solid sorption can be used to reduce concentration of hydrogen sulfide from product gas. Zinc oxide is one of the most common adsorption material, as it can decrease the sulfur content in the gas to under 1 ppm. Other materials available are for example manganese oxide, copper chromite and copper oxide but they are not as efficient in adsorbing sulfur. Importantly, these materials are not regenerable and must be replaced after use. This sets limits to the feasible amount of sulfur in the upstream gas, as replacing the adsorbing material is costly. Therefore, other methods are used in bulk sulfur removal before solid sorption. Waldheim (2018). Furthermore, zinc oxide can be used for decreasing the sulfur content when especially low H2S concentration is needed in methanation purposes.

Activated carbon can also be used for sulfur removal. Most of other impurities such as tar and particulate matter must be removed before utilization of carbon beds as they could block the carbon beds. Activated carbon needs humidity between 30 – 90 % in the gas in order to dissolve and adsorb into the carbon bed. The decrease in sulfur concentration can be decreased as low as 1 ppm, however this depends on the conditions in the carbon beds such as relative humidity and inlet H2S concentration (Waldheim 2018). Therefore, activated carbon can be used in gasification processes that use steam as gasifying agent and have higher humidity after gasifier. Alkali salts can be used to improve the adsorption of sulfur compounds.

### 2.4 Methanation technology

Methanation technology has been developed over 100 years since it was first discovered by Sabatier and Senders in 1902. Sabatier and Sanders discovered that nickel and some other precious metals can be used to catalyze the methanation reaction from hydrogen and carbon monoxide to form methane and water. Later in the 1970s, during the oil crisis, many processes to produce synthetic natural gas from gasification product gas were developed. Since then, these processes have been used in several pilot and commercial plants. Therefore, many mature technologies exist for producing SNG from syngas. (Kopyscinski, Schildhauer et al. 2010).

Before methanation, the product gas typically undergoes a water-gas shift reaction (WGS), which converts the gas to the suitable for production of synthetic natural gas.

\[
CO(g) + H_2O(g) \leftrightarrow CO_2(g) + H_2(g), \Delta H = -41.1 \text{ kJ/mol}
\]

In this slightly exothermic reaction, carbon monoxide and water react to form hydrogen and carbon dioxide. The goal of water-gas shift reaction is to generate a gas in which the H2/CO-ratio is over 3, which is a requirement for the methanation reaction input gas. WGS reaction is typically achieved with catalysts in elevated temperatures. Two commercial processes exist: high temperature shift (HTS) utilizes iron oxide catalysts in temperature range of 350 to 450 °C, and low temperature shift (LTS) utilizes copper–zinc oxide catalysts in range of 190 to 250 °C. In addition, molybdenum catalysts have been developed for processes that include sulfur in the gas stream. (Ratnasamy, Wagner 2009).
Methanation process is based on exothermic reaction of hydrogen and carbon oxides. There are several reactions that take place during methanation. CO methanation converts carbon monoxide and hydrogen into methane and water. CO2 methanation reaction is a linear combination of slightly endothermic reverse water-gas shift reaction and CO methanation (Rönsch). Both of these methanation reactions are exothermic, which emphasizes the importance of heat management during the process. However, carbon monoxide can also react on the catalyst surface to form solid carbon, which can lead to catalyst deactivation. Hydrogen or steam can react with the carbon on the catalyst to convert it back to gaseous products. (Kopyscinski, Schildhauer et al. 2010).

\[
\begin{align*}
CO(g) + 3 H_2(g) & \leftrightarrow CH_4(g) + H_2O(g), \Delta H = -206 \text{ kJ/mol} \\
CO_2(g) + 4 H_2(g) & \leftrightarrow CH_4(g) + 2 H_2O(g), \Delta H = -164 \text{ kJ/mol} \\
2CO(g) & \leftrightarrow C(s) + CO_2(g), \Delta H = -173 \text{ kJ/mol}
\end{align*}
\]

Methanation utilizes metals as catalyst material in the reactors. Nickel catalysts are most often used due to their relatively low cost and good activity in methanation reactions. The activity of catalyst material is fundamental to the performance of methanation reaction. Therefore, deactivation of catalyst should be avoided. Rönsch et al. (2016) list the most common deactivation methods in methanation catalysts. Nickel catalyst poisoning with sulfur compounds, thermal degradation in high temperatures and fouling with carbon and coke deposits are among the most typical catalyst deactivation methods in SNG production with catalytic methanation.

Catalyst poisoning can occur if hydrogen sulfide is present in the gas. Hydrogen sulfide can react with nickel oxide to form nickel sulfides, which decreases the catalytic activity for methanation. Therefore, wet scrubbers and solid zinc oxide sorption are often used to reduce sulfur concentration in gasification product gas. In addition to sulfur removal, tar cleaning is another crucial phase for methanation process. Large hydrocarbon compounds in tar can be adsorbed into the catalyst surface in temperature range of 500 and 600 °C to form coke deposits. Furthermore, thermal degradation of nickel catalyst can occur in high temperatures of over 500 °C, when sintering of nickel can lead to decrease of surface area and activity. However, catalysts materials for high temperature operation have been developed to diminish the sintering of nickel. (Rönsch, Schneider et al. 2016)

Most of the commercial methanation processes are based on adiabatic fixed-bed reactors that use nickel catalysts. In these processes, at two or more reactors are placed in series with gas cooling between the reactors (Rönsch, Schneider et al. 2016). Gas temperatures are increased from about 300 °C up to 600 – 700 °C in first reactor, depending on the thermal durability of the catalyst material. Methanation reactions are limited by high temperatures, which is why intermediate gas cooling is necessary between methanation reactors to reach high methane output. High temperatures are controlled by recycling gas after methanation stages. This dilutes the input gas to the fixed-bed reactor in which the recycled gas is directed, which decreases the output temperature. Gas recycling is optimized to be as low as possible, as large gas volume increases the power needed to compress the gases and increases the size requirement of the methanation equipment. Methanation processes typically utilize high pressures which allows more methane to be produced as is stated by Le Chatelier’s principle. (Haldor Topsoe)
Haldor Topsøe, Air Liquide, Johnson Matthey and Wood Group provide methanation processes based on adiabatic methanation reactors. Haldor Topsøe methanation illustrated in figure 7, named TREMP, is based on 3 to 4 fixed-bed reactors with intermediate gas cooling and gas recycling after first stage. TREMP methanation utilizes high temperatures of near 700 °C. Therefore, the adiabatic fixed-bed reactors can simultaneously be used to generate superheated steam, which can be used for electricity generation or other purposes. (Haldor Topsøe). Air Liquide offers a methanation technology developed by Lurgi, which consists of two adiabatic fixed-bed reactors with gas recycling and cooling. Johnson Matthey’s and Wood Group’s concepts are based on three reactors. Wood Group’s methanation concept VESTA was developed by Foster Wheeler and Clariant. It differs from the other processes in that it does not utilize gas recycling to control reactor temperatures but adds steam to the reactors instead. (Rønsch, Schneider et al. 2016).

Adiabatic fixed-bed methanation process is utilized in several large scale commercial plants. Most of the recent applications are based in China, where coal is gasified to produce SNG. Both Haldor Topsøe’s TREMP and Johnson Matthey’s process has been utilized in scale of several million cubic meters of methane annually in multiple plants. (Rønsch, Schneider et al. 2016)

Besides adiabatic fixed-bed methanation, some other processes have been considered in converting gasification product gas into synthetic natural gas. Isothermal fixed-bed reactor was considered by Linde in methanation process. In this single vessel, the heat generated by methanation reactor was directly used to generate steam (Kopycsinski, Schildhauer et al. 2010). The benefit of isothermal reactor is the temperature control, which decreases thermal degradation of catalysts. Furthermore, it allows more simple processes compared to processes based on adiabatic reactors. However, no large scale applications of isothermal methanation currently exist. Additionally, fluidized bed reactors have been studied in methanation processes. Fluidized beds can be operated as nearly isothermal reactors and are efficient in removing the heat produced by methanation reactors. However, the catalyst particles and reactor walls face large mechanical stress, which decreases the lifetime of both
catalyst material and reactor. Fluidized bed methanation has been most recently demonstrated at Güssing by Paul Scherrer Institute, where a 1 MW SNG plant was erected. (Rönsch, Schneider et al. 2016). Recently, there has been efforts to develop fluidized bed methanation processes in China, where production of synthetic natural gas is increasing. Many studies concerning catalyst development and research have been conducted which are related to Chinas effort to increase its SNG production from coal in coming years.
2.5 Efficiency of bio-SNG plant

Biomass conversion into synthetic natural gas involves multiple process units in order to decompose solid biomass into good quality bio-SNG. The efficiency of biomass drying, gasification, gas cleaning and methanation is an important aspect in the design of a feasible and profitable bio-SNG plant. The early applications of SNG production utilized inexpensive feedstock such as coal and lignite, which meant that the efficiency of the plant wasn’t as crucial as it is today (van der Meijden, Christiaan M, Veringa et al. 2010). However, biomass is typically more expensive and has increasing demand in commercial applications. Therefore, highly efficient process is required to reduce the operational costs of the plant.

There are many different alternatives in measuring the efficiency of a bio-SNG plant. The most typical method is to assess the chemical efficiency of the conversion processes and thermal efficiency of the process as a whole or as single units. Chemical efficiency measures the change in chemical energy that is available in a substance in the input and output of a process. In bio-SNG processes, chemical efficiency is typically used to evaluate the performance of gasifier and the complete process. Cold gas efficiency evaluates the chemical energy that a gasifier converts from solid biomass into gaseous products (Alamia, Larsson et al. 2017):

$$\eta_{CG} = \frac{m_{PG} \cdot LHV_{PG,daf}}{m_{BM} \cdot LHV_{BM,daf}} \quad (2.1)$$

where $m_{PG}$ and $m_{BM}$ are the mass flows of the product gas after gasifier and biomass input to the gasifier, and $LHV_{PG,daf}$ and $LHV_{BM,daf}$ are the dry ash free lower heating values of product gas and biomass. Cold gas efficiency includes only the chemical conversion, so enthalpy changes are not included. Another key performance indicator is the biomass to SNG conversion efficiency, which measures the chemical energy of the product SNG divided by the chemical energy of input biomass:

$$\eta_{SNG} = \frac{m_{SNG} \cdot LHV_{SNG,daf}}{m_{BM} \cdot LHV_{BM,daf}} \quad (2.2)$$

Thermal efficiency of the plant is used to illustrate the total useful energy that can be extracted from the plant compared to all the energy inputs. This includes all the chemical, electric, and heat energy that is associated with the plant:

$$\eta_{PLANT,j} = \frac{\sum_{j=1}^{n} E_j}{\sum_{k=1}^{n} E_k} \quad (2.3)$$

where j is the energy output and k is the input to the process. Besides synthetic natural gas, the plant may produce district heat and electricity, which are included in the thermal efficiency. Energy inputs to the process include external heat sources, electricity to the compressors and naturally the chemical energy of the feedstock.

Efficiency of biomass to SNG processes has been studied by many articles. Many of the studies focus on the gasification section, since the composition of the product gas defines
the design of process units downstream of the gasifier. Van der Meijden et al. (2010) studied
the efficiencies of three bio-SNG plants based on different gasification technologies: pres-
surized direct and atmospheric pressure indirect fluidized bed gasification and pressurized
entrained flow gasifier. The feedstock that was chosen to model the processes was dried
wood. They used cold gas efficiency of the gasifier and SNG conversion efficiency to assess
the processes which were simulated with Aspen plus. The process based on atmospheric
pressure indirect gasification was found to have higher cold gas (81.1 % on LHV basis) and
SNG conversion efficiency (66.8 % on LHV basis) compared to direct (CGE efficiency 74.9
%, biomass to SNG 58.1 % on LHV basis) and entrained flow processes (CGE efficiency
77.4 %, biomass to SNG efficiency 54.3 % on LHV basis).

Bio-SNG plants consume and produce energy in different forms, which are difficult to ana-
lyze in terms of energy quality purely with thermal efficiency of the plant. Therefore, some
studies have focused on alternative ways to assess the process instead of calculating energy
efficiencies. Heyne et al. (2013) used an exergy analysis to analyze atmospheric indirect and
pressurized direct fluidized bed gasification. The processes were modelled with a generic
biomass containing no ash or moisture. The paper focuses only on the gasification section,
leaving out the downstream equipment. Exergies of the process are calculated as the chem-
ical and physical exergy of each input and output streams. Heyne et al. conclude that the
exergy efficiency of indirect gasifier is in the range of 70.0 and 76.6 % whereas the direct
method reaches efficiencies of 70.7 – 78.9 %. The cold gas efficiencies are 78.8 – 81.2 %
for atmospheric indirect gasification and 78.7 – 83.0 for pressurized direct gasification.

Kohl et al. (2014) compared three different methods in assessing the energy efficiency of a
complete bio-SNG process utilizing direct gasification of woody waste biomass from forest
industry. The article compares using thermal efficiency, exergy efficiency and primary en-
ergy efficiency as methods to analyze the performance of a bio-SNG plant. The primary
energy efficiency widens the scope of efficiency into the consumption of primary energy
and can be applied to analyze the impact of a process to the global primary energy use. The
article concludes that using thermal efficiency can give too optimistic results especially for
processes with high electricity use. Exergy efficiency is seen as a good method to analyze
bio-SNG processes in detail and to recognize potential places for improving the process.
Thermal efficiency is a relatively simple method compared to assess the performance of bio-
SNG plant and is thus widely used as the primary method in analyzing the process. With
Aspen plus and spreadsheet modeling Kohl et al. reached a thermal plant efficiency of 78.4
%, primary energy efficiency of 76.4 % and exergy efficiency of 52.3 %. The exergy effi-
ciency of the gasifier was 77.3 %, which is in the same range as with Heyne et al. The cold
gas efficiency of the gasifier was calculated as 80 %.
2.6 Commercial and demonstration bio-SNG plants

Production of synthetic natural gas from biomass with gasification and catalytic methanation has thus far focused on small scale projects. These projects have utilized gasification, gas cleaning, and methanation technology to demonstrate and optimize the process for larger scale production of bio-SNG. Most of the industrial development has taken place in Europe, where a few projects have been operational since early 2000s. First process for producing SNG from biomass was constructed in Güssing, Austria in 2009. Since then, a large demonstration plant has been built to Gothenburg, Sweden and large commercial plants have been planned in Sweden, Finland and Netherlands that are pending the final investment decision.

Table 4. List of bio-SNG projects

<table>
<thead>
<tr>
<th>Plant</th>
<th>Gobigas</th>
<th>Güssing</th>
<th>Swindon</th>
<th>Ambigo</th>
<th>E.ON</th>
</tr>
</thead>
<tbody>
<tr>
<td>Country</td>
<td>Sweden</td>
<td>Austria</td>
<td>UK</td>
<td>Netherlands</td>
<td>Sweden</td>
</tr>
<tr>
<td>Year of commission</td>
<td>2014</td>
<td>2009</td>
<td>TBD</td>
<td>TBD</td>
<td>TBD</td>
</tr>
<tr>
<td>Status</td>
<td>Mothballed</td>
<td>Mothballed</td>
<td>In construction</td>
<td>Pending investment decision</td>
<td>Pending investment decision</td>
</tr>
<tr>
<td>Scale (SNG)</td>
<td>20 MW</td>
<td>1 MW</td>
<td>&lt; 0.5 MW</td>
<td>2 – 3 MW</td>
<td>200 MW</td>
</tr>
<tr>
<td>Fuel</td>
<td>Wood pellets, wood chips</td>
<td>Woody biomass</td>
<td>RDF</td>
<td>Biomass, waste</td>
<td>Forest residues, bark, stumps</td>
</tr>
<tr>
<td>Gasification method</td>
<td>Indirect fluidized bed</td>
<td>Indirect fluidized bed</td>
<td>Direct fluidized bed</td>
<td>Indirect fluidized bed</td>
<td>Direct fluidized bed</td>
</tr>
<tr>
<td>Methanation method</td>
<td>Adiabatic fixed bed</td>
<td>Fluidized bed</td>
<td>Adiabatic fixed bed</td>
<td>Adiabatic fixed bed</td>
<td>Adiabatic fixed bed</td>
</tr>
</tbody>
</table>

The first bio-SNG demonstration plant in the world was constructed in Güssing, Austria, next to a 8 MWth combined heat and power plant that utilized gas engines for electricity production. A 1 MW SNG synthesis plant was built in 2009 to utilize a side stream of the gasifier. The gasification technology utilized at Güssing was similar to later constructed Gobigas plant in Sweden: a dual fluidized bed with woody biomass as a feedstock. The primary gas cleaning technology was based on a wet scrubber utilizing oil to absorb most of tar and impurities. Methanation technology was based on a fluidized bed process which was developed by Paul-Scherrer Institut. The demonstration plant successfully produced good quality synthetic natural gas, which was utilized in a natural gas filling station as transportation fuel. The process reached a cold gas efficiency of 61 %. (Schildhauer, Biollaz 2016)

The most prominent demonstration of bio-SNG production process has been the Gothenburg Biomass Gasification project (Gobigas) in Sweden. The plant was built by Göteborg Energi and commissioned in November 2014. Gobigas project focused on producing bio-SNG from biomass with high moisture content with high efficiency. Thermal power of the plant was 32 MW and it produced 20 MW of synthetic natural gas with maximum capacity operation. After being operated for over three years, the plant was mothballed in March 2018 due to too high operating costs. A larger commercial plant was originally planned to be built after the demonstration plant, but the uncertainties concerning legislation and
prices of biomass and bio-SNG have prevented the construction thus far. A process scheme of Gobigas plant is illustrated in Figure 8. (Thunman 2018)

Most of the operation at the Gothenburg plant was with wood pellets. However, pellets are not feasible as feedstock for a larger scale bio-SNG plant due to their high cost compared to other woody biomass such as forest residue and wood chips. Thunman et al. (2018) report, that gasifying wood chips and bark is possible with modifications to the process. They emphasize the effect of moisture content to the process and suggest that integrating a dryer is essential for stable operation of the plant.

The gasification technology used in Gobigas is based on indirect dual fluidized bed gasifier with steam fluidization. A 32 MWth gasifier was provided by Valmet, which consists of a bubbling fluidized bed gasifier and a circulating fluidized bed boiler for combustion. The gasification process is similar as was used in Güssing bio-SNG plant, where the indirect gasification process was previously researched. Biomass is gasified in the BFB boiler at 870 °C with superheated steam (320 – 350 °C), and the unreacted char is combusted in the latter CFB boiler. Olivine that is used as a bed material in the boiler transfers heat to the gasification reactions from the combustion of char.

![Figure 8. Gobigas process scheme modified from Thunman et al. (2018).](image)

After gasification, gas is cooled to approximately 200 °C. Bag filters filled with limestone are used to remove most of the particulate matter. Removal of tar and aromatic hydrocarbons is achieved with a wet scrubber and active carbon beds. The scrubber utilizes rapeseed methyl ester (RME) to absorb large polyaromatic hydrocarbons. Smaller hydrocarbons such as benzene and naphthalene and impurities such as hydrogen sulfide are collected by activated carbon beds. After compression to 16 bars, syngas undergoes further conditioning phases, where the remaining impurities are removed. Hydrodesulfurization and zinc oxide bed decreases sulfur concentration to under 10 ppm. Methanation is based
on Haldor Topsoe’s TREMP process, which utilizes four adiabatic fixed beds and nickel catalysts. (Thunman, Seemann et al. 2018)

Gobigas demonstration plant achieved a cold gas efficiency of 71.7 %, methane efficiency of 61.8 % and total plant efficiency of 57.7 %, with wood pellets that contained 8 % moisture. The aim of the project was to reach over 65 % methane efficiency. The methane efficiency is lower than expected, resulting largely from large heat losses from the gasification section that has quite poor insulation. This led to large amounts of recycled gasification gas to the combustion section in order to generate enough heat for the gasification. (Alamia, Larsson et al. 2017).

A bio-SNG demonstration plant is scheduled to be built in the Netherlands during the next years. This plant, located in Alkmaar, is based on the technology developed by Energy Research Centre of the Netherlands. The MILENA indirect gasification, OLGA gas cleaning and ESME methanation process will be used to demonstrate a 4 MWth SNG plant. The process is illustrated in Figure 9. The indirect gasification is operated at around 800 °C, and produces a nitrogen free, hydrocarbon rich product gas with high tar content.

Figure 9. Alkmaar bio-SNG plant process scheme, modified from Schildhauer & Biollaz (2016)

The gasifier’s structure allows pressurization up to 7 bars, which enables easier scaling up of the process. The relatively long residence time also enables the use of waste fuels such as SRF, RDF and REF. OLGA gas cleaning utilizes a multiple step wet scrubbing process around 100 °C, which allows recycling of tar compounds into the gasifier. After tar removal, the gas is dried, compressed and heated to above 350 °C. Hydrodesulfurization and zinc oxide bed are used to remove most of the sulfur compounds from the syngas. To convert smaller aromatic hydrocarbon compounds such as benzene and aliphatic compounds such as ethylene, a catalyzed steam reformer is used at over 400 °C. Before methanation, carbon dioxide will be removed with a solid adsorbent material at high temperature and pressure, which uses less energy compared to conventional CO2 separation processes with solvents. The ESME methanation step is based on commercial adiabatic fixed bed process with intermediate cooling. (Schildhauer, Biollaz 2016)
The only plant currently under construction is a demonstration project Gogreengas in Swindon, UK. The new 500 kWth bio-SNG plant will be the first demonstration plant to produce synthetic natural gas from refuse derived fuel (RDF). It is based on direct fluidized bed gasification with plasma gas cleaning and commercial adiabatic fixed bed Vesta methanation process from Wood Group (gogreengas.com projects status pdf). The fluidized bed will gasify the RDF feedstock in temperature of approximately 800 °C with steam and oxygen. After gasification, the product gas containing tar is directed to a plasma converter developed by Advanced Plasma Power. The converter uses several hundred kilowatts to power an electric arc, which heats the gases to over 1000 °C. During the conversion, most of the impurities including tar and aromatic and aliphatic hydrocarbons are converted into hydrogen, carbon dioxide, carbon monoxide and methane. Before catalytic methanation, gas is cooled with a waste heat boiler, particulate matter is filtered and wet scrubber and CO2 scrubber are used to remove most of steam, remaining aromatic hydrocarbons and carbon dioxide. (Materazzi, Taylor 2019)

The largest commercial planned bio-SNG plant has been considered to be constructed in Malmö or Landskrona by E.ON. The plant was designed to be based on 300 MWth direct fluidized bed gasification with catalytic tar reforming and adiabatic fixed bed methanation. The designed process would produce 200 MW of bio-SNG into the natural gas grid, approximately 50 MW of district heat and 20 MW of electricity. Thus, the bio-SNG efficiency would be in range of 60 – 65 % and total plant efficiency roughly 80 %. The feedstock is considered to include woody biomass such as forest residues, bark, stumps and recycled wood (European Commission 2016). However, no investment decision has been made due to uncertainties in future of biofuel related policies in Sweden and the price of oil. The plant has been granted a financial grant from European Commission, which requires an investment decision before 2020. (Waldheim 2019)
3 Biomass for bio-SNG production

Biomass is a broad concept that covers different products, waste and residue of biological origin that includes ligneous biomass from forest industry, agricultural animal and vegetal biomass, aquacultural biomass, and the biodegradable fraction of industrial and municipal waste (EU directive 2018/2001). This wide range of biomass sources have very different physical and chemical characteristics which must be studied carefully before utilizing them as fuel for thermochemical conversion process such as gasification. The quality and type of biomass that is used for energy production or conversion processes influence the whole gasification process including pre-treatment and process equipment such as boiler type and downstream gas cleaning.

Most important physical parameters for biomass fuels are the moisture content, calorific value, bulk and energy density of the fuel and the dimensions of the fuel particles. Moisture content of combustible biomass can vary from under 10 % of dried or torrefied wood pellets to a moisture of about 60 % of fresh bark (van Loo, Koppejan 2008). Fuels for combustion are typically dried before combustion or gasification in order to improve the calorific value of the fuel and to make adjusting of the process easier (E3003 luentomoniste). Energy and bulk density of the fuel is important for the logistics of the biomass. More dense fuels can be transported more efficiently and thus be collected from further away from the combustion or gasification plant. Availability of feasible biomass can cause bottlenecks for the size of a biomass plant if potential sources are scarce.

Size of the fuel particles is important for fluidized bed boilers which are the most used technology for gasification. The required particle size of the fuel is under 80 mm for bubbling fluidized beds and under 40 mm for circulating fluidized beds. (van Loo, Koppejan 2008) Additionally, ash content and ash sintering temperature is a significant factor for gasifying fuels in fluidized beds. High content of ash can lead to high concentrations of ash particles in product gas which increases the load for particle cleaning. Low sintering temperature can cause agglomeration of ash in boilers and formation of ash deposits on heat exchanger surfaces. Volatile matter in biomass varies and affects the thermal decomposition characteristics of the fuel. The thermal decomposition is important especially for indirect gasification processes, which are based on the concept of separating the devolatilization and char combustion phases. (van Loo, Koppejan 2008).

Chemical attributes affecting the gasification characteristics are mainly the concentrations of carbon, hydrogen and oxygen in the fuel. These determine the calorific value of the biomass fuel. Feedstocks containing high amounts of carbon and hydrogen have higher heating values. The concentration of other elements such as nitrogen, sulfur and chlorine affect the gaseous emissions formed during combustion and gasification process. In terms of producing synthetic natural gas, ammonia and hydrogen sulfide are the most troublesome effects of high concentrations of nitrogen and sulfur in the fuel. Chemical composition of biomass ash varies depending on the feedstock which alters the characteristics of the ash in high temperatures. Most important ash constituents are calcium, magnesium, potassium, natrium, phosphorus and silica. The concentration ratios between these ash elements largely determine the behavior of ash in gasification applications.
3.1 Forest biomass

Woody biomass is the most important source of bioenergy in Finland. Most of the forest biomass used for energy production is by-products from forest industries. Bark, sawdust and black liquor produced from pulp and sawmills form a major share of these products. The second largest source of energy are forest chips which are produced as a side product of logging. Forest chips are produced from small-diameter trees, stumps and logging residues such as tree tops, needles and branches that are unusable for forest industry (Routea, Asikainen et al. 2013).

![Pie chart showing wood fuel energy use in Finland in 2018.](image)

**Figure 10. Wood fuel energy use in Finland in 2018. (Luke 2019a)**

In 2018, the total industrial wood fuel use consisted of 46.7 TWh of black liquor, 38.7 TWh of solid wood fuels and under 2 TWh of other waste such as oils and sludges. Black liquor is a liquid product of pulp industry and is combusted at the pulp mills in large recovery boilers for energy production. Since black liquor is already highly utilized in Finland and it has poor gasification properties such as high ash content and low ash sintering temperatures, it is not considered as a potential feedstock in this thesis.

Solid wood fuels, which include both the by-products of forest industry and logging, are more suitable for biofuel production through gasification. Bark and sawdust are produced at sawmills that process roundwood, mostly from spruce and pine. They accounted for half of the energy production with solid wood fuels in 2018 with total use of 10.1 million cubic meters (Luke 2019b). Their moisture content is high, typically over 50%. Basic density of bark from spruce and pine varies between 300 and 340 kg/m3, and sawdust between 380 and 480 kg/m3. Basic density includes the mass of dry wood mass per volume of fresh wood. Ash content of bark is slightly higher than with sawdust: sawdust contains generally under 1 wt-% of ash, whereas bark has 1 to 3 wt-% of ash. (Alakangas, Hurskainen et al. 2016).
Therefore, ash content of sawdust depends highly on the amount of bark that is left amongst the sawdust during processing at sawmill (van Loo, Koppejan 2008).

In addition to energy use, sawdust can be used at pulp mills as lower quality feedstock or as raw material in particleboard and fiberboard mills (Heinimö 2008). Therefore, the utilization of sawdust as a feedstock in energy production is lower than the rate of its generation. However, bark has no other alternatives for utilization than as raw material in energy generation. Thus, most of produced bark is currently combusted at the sawmills to produce process heat.

The third major source of solid wood fuels are forest chips. They are produced from the surplus wood that is left during logging: logging residues, trees with small diameter, and stumps. The residues left from logging are tree tops, branches and needles. All the residues, stumps and small trees are collected and comminuted to small chips prior to application as a fuel. The utilization of forest chips increased from little over 2 million m³ to 8 million m³ during 2004 – 2013. Since then, the amount of used forest chips in heating and power plants has remained stable between 7 and 8 million m³. Routa et al. (2013) report, that the technical potential of forest chips production is between 14 and 20 million m³. Anttila et al. (2014) estimate in their study that the total technical potential of forest chip production is approximately 20 million cubic meters which is equal to approximately 40 TWh. Therefore, forest chips are one of the most potential sources of solid biofuels in the coming years, considering that the solid wood fuels from forest industry are already largely utilized as energy or other applications.

The moisture content of forest chip feedstock depends on the timing of the collection of the wood after logging. If the residues are collected simultaneously with logging, the moisture content is similar with fresh wood, varying between 50 and 60 %. If they are left in forest for the duration of summer months, their moisture decreases to under 30 % which increases the efficiency of transportation and reduces the need for drying. However, up to third of the mass of residues is lost due to loss of needles. (Alakangas, Hurskainen et al. 2016). As the

---

**Figure 11. Solid wood fuel energy use in heating and power plants during 2004 – 2018. (Luke 2019b)**
moisture content varies, the density of forest residues used for production of forest chips is dependent of the collection time. Comminuted forest chips from spruce have an approximate basic density of 400 – 465 kg/m³, while for pine the basic density is 385 – 405 kg/m³.

Whereas the logistics of forestry by-products such as sawdust and bark are relatively straightforward, the supply chain of forest residues is more complicated and requires careful designing. The supply chain of forest residues can be determined by the timing of comminution of the residue material. Three different types of supply chains exist for forest residues which are determined by the location of comminution of residues to chips. The most common method is to collect the forest residues and comminute them at the forest site at roadside with a mobile chipper. After comminution, the chips are transported with trucks to the plant for utilization. Around 70 % of all forest residues transported to energy use were supplied with this method. The advantage of roadside chipping are its flexibility, low investment costs, and the large amount of experience with the method in the Nordics. However, the supply chain may be susceptible to long waiting times if chipping and transportation are not operated efficiently at the logging site. (Routea, Asikainen et al. 2013).

Other options in arranging the logistics are comminuting the residues at a separate terminal or at the plant. Terminal comminution involves transportation of unprocessed forest residues to a terminal, where the residues are processed into chips and then stored and transported to a plant. The investment costs of this option are higher, and the volumes of processed biomass must be high in order to terminal comminution to be feasible. Furthermore, the transportation of residues from logging sites is not as efficient as it is with comminuted chips, which increases the costs. Terminal reduces the possible waiting at the roadside because the comminution and transportation are not dependent on each other. In 2013, about 7 % of logging residues were comminuted at a terminal. Plant comminution is the second most common option to handle logging residues with a 23 % share of forest residue processing. The investment costs are high as it requires a chipping system installed at the plant. The largest upside of comminuting forest biomass at the plant is the increased control over the logistic system and improved fuel quality control. Besides the high costs concerning the chipping equipment, the transportation of unprocessed low-density residues over long distances may increase the cost of the feedstock procurement. (Routea, Asikainen et al. 2013).

The ash content in forest chips from logging residues is generally slightly higher than wood fuels produced from stemwood. This is largely due to the amount of bark and needles used for chips, which contain higher amounts of ash. Alakangas et al. (2016) report that comminuted chips from logging residues have ash concentrations of 1.33 – 6 wt-% on dry basis, while chips from small diameter wood and stumps contain 0.5 – 0.6 wt-% of ash. Van Loo & Koppejan (2008) report ash content of 1.0 – 2.5 wt-% for forest chips containing bark.

The volatile matter in woody biomass varies. Alakangas et al. (2016) report that the volatile content of different wood species varies between 75 and 83 % on dry basis. Van Loo & Koppejan (2008) estimate the volatile matter of typical woodchips in the range of 76.0 and 86.0 % on dry basis and coniferous bark between 69.6 and 77.2 %. The Phyllis2 database updated by the Energy Centre of the Netherlands (ECN) report that on average, pine contains 84.3 % of volatile matter on dry basis. Furthermore, they report that on average, spruce and pine bark consist of 74.2 % and 73.2 % of volatile matter respectively. The amount of volatile matter in bark-free wood feedstock is slightly higher than with agricultural biomass.
Typical ultimate analyses of solid woody biomass are listed in table 5. The carbon content of wood is generally higher than with other biomass. Notable differences between the different parts of wood exist in carbon, oxygen and sulfur concentrations between stemwood and bark. The carbon content of stemwood is slightly lower and oxygen content higher than in bark, and thus the calorific value of bark can be higher than with stemwood. Furthermore, sulfur content is higher with spruce and pine bark and is the highest in the needles (Alakangas, HurSkainen et al. 2016, Werkelin, Skrifvars et al. 2005). Therefore, bark and forest chips produced from logging residues contain the highest amounts of sulfur of solid wood feedstocks for gasification. The chlorine concentrations of wood are low especially when compared to agricultural biomass.


<table>
<thead>
<tr>
<th></th>
<th>C</th>
<th>H</th>
<th>O</th>
<th>N</th>
<th>S</th>
<th>Cl</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wood in</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>general</td>
<td>48–50</td>
<td>6.0–6.5</td>
<td>38–42</td>
<td>0.5–2.3</td>
<td>0.05</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td></td>
<td>47.1–51.6</td>
<td>6.1–6.3</td>
<td>38.0–45.2</td>
<td>0.1–0.3</td>
<td>0.01</td>
<td>0.004</td>
</tr>
<tr>
<td>Pine</td>
<td>51.8</td>
<td>6.1</td>
<td>41.2</td>
<td>0.3</td>
<td>0.01</td>
<td>0.004</td>
</tr>
<tr>
<td>Spruce</td>
<td>52.0</td>
<td>6.3</td>
<td>41.4</td>
<td>0.1</td>
<td>0.1</td>
<td>0.0006</td>
</tr>
<tr>
<td>Bark, pine</td>
<td>49.2</td>
<td>5.9</td>
<td>44.8</td>
<td>0.2</td>
<td>0.1</td>
<td>0.0001</td>
</tr>
<tr>
<td>Bark, spruce</td>
<td>54.5</td>
<td>5.9</td>
<td>39.2</td>
<td>0.4</td>
<td>0.07</td>
<td>0.0002</td>
</tr>
<tr>
<td></td>
<td>53.5</td>
<td>5.8</td>
<td>38.7</td>
<td>0.4</td>
<td>0.03</td>
<td>0.009</td>
</tr>
<tr>
<td></td>
<td>53.0</td>
<td>5.8</td>
<td>40.9</td>
<td>0.5</td>
<td>0.04</td>
<td>0.0002</td>
</tr>
<tr>
<td></td>
<td>50.4</td>
<td>5.8</td>
<td>40.8</td>
<td>0.5</td>
<td>0.03</td>
<td>0.03</td>
</tr>
</tbody>
</table>

The main ash constituent in wood fuels from pine and spruce are calcium, potassium, and silica. Werkelin et al. (2005) report that the concentration of these constituents with forest residues are 0.9 – 6.5 g/kg, 1.5 – 2.7 g/kg, and 0.19 – 19.2 g/kg per dry fuel respectively. With bark the reported concentrations are 4.1 – 11.0 g/kg of calcium, 1.2 – 2.2 g/kg of potassium and 0.07 – 11.9 g/kg of silica. Alakangas et al. (2016) report that the concentrations for spruce bark are 8.4 g/kg of calcium, 2.0 g/kg of potassium and 0.2 g/kg of silica and for pine bark 6.4 g/kg of calcium, 3.2 g/kg of potassium and 0.06 g/kg of silica. These values are similar as van Loo & Koppejan (2008) report, although the silica content is reported to be higher for spruce bark.

The reasonably high amount of calcium in the wood ashes increases the ash sintering temperature. Furthermore, wood ashes contain some magnesium which also has an increasing effect on ash sintering temperature (van Loo, Koppejan 2008). Therefore, the wood ashes start to sinter at higher temperatures than other biomass feedstock such as straw or grass. Higher ash sintering temperatures simplifies the operation of gasification reactor and reduces the possibility of ash agglomeration. Potassium in the ash reacts with most of the chlorine forming KCl, which reduces the amount of hydrogen chloride in the product gases (Porbatzki, Stemmler et al. 2011, Werkelin, Lindberg et al. 2011).

### 3.2 Agricultural biomass

The use of agricultural biomass in energy production in Finland is significantly lower than with woody biomass. This is largely due to the large amount of forest area in Finland compared to the agricultural areas. The techno-economical potential of agrobiomass in energy
use is currently 4.3 TWh which expected to increase to 6.2 TWh by 2050 (Pahkala, Lötjönen 2015). In comparison, the techno-economic potential of forest chips is almost ten times as high in Finland. The potential of agricultural biomass in future depends strongly on average diet. If vegetarian diet becomes more common, more land is freed for bioenergy production instead of producing fodder for cattle. The estimated potential of bioenergy production with energy crops in Finland varies between 0 and 89 TWh depending on the scenario (Koljonen, Koreneff et al. 2012). Agrobiomass consists of inedible byproducts of cultivated food crops and energy crops which are cultivated especially for energy use. The use of agricultural biomass in energy production focuses currently mostly on the byproducts from agriculture which do not reserve land from food production.

Straw from cereals is produced simultaneously when grains are harvested. The resulting straw is either left at the field or baled if it is used as feedstock for combustion or gasification. The amount of straw that can be utilized depends on the harvesting method. Typically, 30 cm of the straw is left at the field, which is equivalent to 29 – 53 % of the total mass (Pahkala, Lötjönen 2015). Straw composes 70 – 90 % of the potential of agricultural byproducts. The alternative uses for straw are as fodder or as litter which is currently the most common application. Approximately 20 % of produced straw is used in these applications (Koljonen, Koreneff et al. 2012). In addition to using agricultural byproducts, reed canary grass has been cultivated to be used in energy production. The production of reed canary grass was at its highest in 2010 with nearly 20 000 of arable area but has since decreased as a result of increase in price of cereal crops (Pahkala, Lötjönen 2015). Miscanthus has been used in energy production in Denman and Southern Sweden, and it is the most utilized energy crop in Europe (Alexopoulou 2018). However, it can’t sustain the cold climate of Finland. Since the use of reed canary grass and miscanthus is minimal in Finland, the focus of this chapter will be on the byproducts of agriculture i.e. straw.

The density of straw is low, which is one of the adverse factors in its application as gasification feedstock. The bulk density of straw bales is 100 – 150 kg/m3 (Alakangas, Hurskainen et al. 2016), which reduces the feasible transportation range of straw to gasification site. Pelleting straw is a possible option for increasing the density, as the bulk density of straw pellets is 450 – 750 kg/m3. Furthermore, the moisture content can be high, depending on the timing of the harvest. If the straw is collected during autumn, rain can increase the moisture content to over 50 %. The typical range of moisture content is between 30 and 60 %. (Alakangas, Hurskainen et al. 2016). Agricultural biomass requires a separately designed supply chain from forest biomass in order to be feasible as an energy feedstock. After harvesting of crops, straw is collected and baled to increase density. Baling of straw can be made simultaneously with harvesting or straw can be left at the field in order to improve its fuel quality by leach off impurities with rain. Straw bales are either stored at the farming area or transported directly to plant for utilization. (van Loo, Koppejan 2008).

<table>
<thead>
<tr>
<th>Type</th>
<th>C</th>
<th>H</th>
<th>O</th>
<th>N</th>
<th>S</th>
<th>Cl</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wheat straw</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry</td>
<td>45.5 – 46.9</td>
<td>5.5 – 6.1</td>
<td>39.2 – 43.4</td>
<td>0.39 – 0.79</td>
<td>0.05 – 0.11</td>
<td>0.05 – 0.25</td>
</tr>
<tr>
<td>Dry, ash free</td>
<td>48.9</td>
<td>5.9</td>
<td>44.1</td>
<td>0.72</td>
<td>0.15</td>
<td>0.005</td>
</tr>
<tr>
<td>Barley straw</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry</td>
<td>45.1 – 46.5</td>
<td>5.3 – 6.1</td>
<td>40.1 – 43.7</td>
<td>0.33 – 0.71</td>
<td>0.08 – 0.16</td>
<td>0.0 – 0.8</td>
</tr>
<tr>
<td>Dry, ash free</td>
<td>47.5</td>
<td>6.1</td>
<td>45.4</td>
<td>0.8</td>
<td>0.1</td>
<td>0.005</td>
</tr>
<tr>
<td>Reed canary grass</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry</td>
<td>46</td>
<td>5.7</td>
<td>-</td>
<td>1.3</td>
<td>0.17</td>
<td>0.5</td>
</tr>
<tr>
<td>Dry, ash free</td>
<td>49.0</td>
<td>6.1</td>
<td>44.0</td>
<td>0.8</td>
<td>0.09</td>
<td>0.001</td>
</tr>
<tr>
<td>Miscanthus</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Dry, ash-free</td>
<td>49.6</td>
<td>5.6</td>
<td>43.8</td>
<td>0.54</td>
<td>0.06</td>
<td>0.002</td>
</tr>
</tbody>
</table>

The carbon content in agricultural biomass is slightly lower than with biomass from coniferous trees, and the oxygen concentration can be little higher. Sulfur and chlorine content is typically higher than with woody biomass, which is an important factor in synthetic methanation utilizing catalyzed metals which sulfur can deactivate. The chlorine content of cereal straw can be reduced by washing it before baling. Washing is performed by leaving the straw at the field during rain after harvesting, which reduces the concentration of watersoluble chlorine and alkali compounds (Alakangas, Hurskainen et al. 2016). The reactivity of straw is similar as with wood fuels. Volatile matter in straw varies between 70 and 81 wt-% of dry matter (van Loo, Koppejan 2008).

Agrobiomass has higher ash content than woody biomass, between 2 and 10 wt-% while it is highest for wheat straw (Oakey 2015). Wheat straw is best suited for high temperature applications such as gasification because of its high ash sintering temperature amongst the cereal straws. Wheat ash contains high amounts of silica, over 65 wt-% of ash, which decreases sintering at low temperatures. Barley straw ash contains more potassium and calcium than wheat, which makes it unfavorable for gasification applications because of very low ash sintering temperatures. Similar to chlorine, also alkali compounds in straw ashes reduce if it is kept outside during rain. Therefore, straw that is collected during autumn has generally higher ash sintering temperatures (Alakangas, Hurskainen et al. 2016).

### 3.3 Industrial and municipal waste

Solid waste produced from different sources can be considered as possible fuels for gasification. However, waste fuels are generally considered to be feasible only in heat and power generation rather than utilizing the gasification gas for fuel synthesis. Applications of waste gasification typically produce product gas for combustion in a boiler. Solid waste from different sources have several factors that can hinder their utilization for fuel synthesis processes. Waste-derived fuels are heterogeneous in their size and chemical properties and may contain large amounts of hazardous elements such as heavy metals and impurities such as chlorine and sulfur.
One of the sources of solid waste is municipal solid waste (MSW). Production of MSW has varied between 2.2 and 2.9 million tonnes between 1997 and 2017. During this time period, utilization of MSW as energy and material has replaced landfilling almost completely. Legislation concerning the prioritizing of waste utilization alternatives and landfilling have affected the increased energy production with municipal solid waste. Waste legislation (Jätelaki 646/2012) dictates that primarily waste production must be prevented, after that reused, recycled, utilized as energy and ultimately landfilled. Landfilling legislation (Valtioneuvoston asetus 331/2013) dictates that landfilled waste must not contain over 10% of biodegradable material. Therefore, incineration and gasification of MSW has increased dramatically during the last two decades. In addition to MSW, commercial and industrial waste and construction waste are most suitable for thermochemical utilization to produce energy (Alakangas, Hurskainen et al. 2016). Commercial and industrial waste consists mostly of paper, cardboard, plastic, textile, wood rubber, metal and inert material such as stone and glass. Construction waste includes wood, plastic, metals and inert materials such as concrete (Nasrullah 2015).

MSW and other solid waste is difficult to be used directly as a fuel without any pre-treatment. Therefore, solid recovery fuels (SRFs) are produced from solid wastes to improve the physical and chemical properties of waste fuels. Nasrullah (2015) discusses the production of SRF from different waste sources in Southern Finland in his dissertation. Production of solid recovery fuels includes processing the waste streams at a mechanical treatment plant, where the recyclable materials are collected, waste is shredded, and non-combustible materials are sorted out. SRF is classified according to EN 15359, which states the properties of standardized solid recovery fuels based on the calorific value and the amount of chlorine and mercury in the fuel and divides the SRF to five classes according to its quality.

After producing SRF from waste, the physical properties of the fuel are more homogeneous and suitable for combustion or gasification. However, the properties of SRF are very different to each other depending on the waste sources that are used in production of the fuel. Per
Alakangas et al. (2016), the moisture content of SRF is between 5 and 30 % and the bulk density between 180 and 210 kg/m³. The ash content of solid recovery fuels is 1 – 16 % on dry basis. The net calorific values of produced SRF is in the range of 17 – 37 MJ/kg on dry basis. Nasrullah (2015) produced SRF from three different sources: commercial and industrial waste (C&IW), construction and demolition waste (C&DW) and from municipal solid waste (MSW). In these SRFs, the moisture content varies between 15.0 and 25.0 wt-% depending on the waste feedstock. Ash content is between 9.0 and 12.5 wt-% and net calorific value in range of 20.0 – 25.0 MJ/kg on dry basis. The volatile matter in SRF varies between 70 and 89 wt-% according to different sources (Alakangas, Hurskainen et al. 2016, ECN.TNO 2019).


<table>
<thead>
<tr>
<th>SRF type</th>
<th>Moisture (wt-%)</th>
<th>NCV, dry (MJ/kg)</th>
<th>Ash (wt-%)</th>
<th>Volat. matter (wt-%)</th>
<th>C (dry wt-%)</th>
<th>H (dry wt-%)</th>
<th>O (dry wt-%)</th>
<th>N (dry wt-%)</th>
<th>S (dry wt-%)</th>
<th>Cl (dry wt-%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SRF C&amp;IW (I)</td>
<td>25.0</td>
<td>25.0</td>
<td>12.5</td>
<td>-</td>
<td>57.4</td>
<td>8.0</td>
<td>17.8</td>
<td>0.5</td>
<td>0.3</td>
<td>0.6</td>
</tr>
<tr>
<td>SRF C&amp;DW (I)</td>
<td>16.5</td>
<td>20.0</td>
<td>9.0</td>
<td>76.6</td>
<td>50.0</td>
<td>6.4</td>
<td>31.6</td>
<td>1.0</td>
<td>0.3</td>
<td>0.4</td>
</tr>
<tr>
<td>SRF MSW (I)</td>
<td>15.0</td>
<td>22.4</td>
<td>9.8</td>
<td>79.4</td>
<td>53.0</td>
<td>7.4</td>
<td>28.0</td>
<td>0.6</td>
<td>0.2</td>
<td>0.6</td>
</tr>
<tr>
<td>SRF (II)</td>
<td>9.1</td>
<td>23.1</td>
<td>5.9</td>
<td>-</td>
<td>56.0</td>
<td>7.4</td>
<td>-</td>
<td>0.63</td>
<td>0.16</td>
<td>0.19</td>
</tr>
<tr>
<td>SRF (II)</td>
<td>28.5</td>
<td>21.5</td>
<td>9.5</td>
<td>-</td>
<td>52.9</td>
<td>7.3</td>
<td>-</td>
<td>0.71</td>
<td>0.13</td>
<td>0.71</td>
</tr>
<tr>
<td>SRF (III)</td>
<td>28.5</td>
<td>25.3</td>
<td>8.3</td>
<td>88.4</td>
<td>61.7</td>
<td>8.4</td>
<td>32.5</td>
<td>0.52</td>
<td>0.14</td>
<td>-</td>
</tr>
</tbody>
</table>

Carbon content in SRF fuels is high, over 50.0 wt-%, and oxygen content relatively low, under 35 wt-%. Therefore, the calorific values of the fuel is high compared to forest and agricultural biomass. SRF contains high amounts of sulfur and chlorine, which are troublesome considering their potential as fuel for bio-SNG production. Their concentration is at least one magnitude higher than with wood or straw fuels. Nasrullah (2015) studied the effect of different materials in waste to the concentration of harmful elements in SRF. Especially SRF containing rubber was found to have high chlorine concentrations. Moreover, hard plastics and textiles were found to contain impurities and toxic elements such as cadmium, mercury and lead.

The ash content in SRF is relatively high while ash softening temperatures are quite high. Therefore, ash agglomeration in the furnace is not a significant problem in waste gasification. The main concerns of gasifying waste are related to the concentration of chlorine and heavy metals. Large amounts chlorine in SRFs can lead to corrosion of gasification reactors. The concentration of chlorine is included in the classification system of solid recovered fuels, which dictates that the cleanest SRF fuels should contain less than 0.2 wt-% of chlorine on dry basis. This amount is in the same range that cereal biomass contain chlorine.

### 3.4 Renewable energy directive

European Commission drafted a new renewable energy directive (REDII) during 2018, which must be introduced to national legislation during the 2020s. The directive 2018/2001/EU includes new targets for the share of renewable energy of energy consumption and for the share of advanced biofuels in transportation. Additionally, sustainability
criteria have been drafted for solid biofuels used in energy production while they have already been included for biofuel production in the previous renewable energy directive. The new target for the share of renewable energy in energy consumption in EU is 32% and the national target for the share of advanced biofuels in road and rail transport is 14% for each of EU member countries. The objective of the directive is to meet these targets by 2030.

The sustainability criteria set for transport biofuels dictates the acceptable feedstocks for liquid and gaseous fuels. The main objective of these criteria is to limit the use of energy crops in production of biofuels that may replace food production by indirect land use change (ILUC). Since the farming area used for energy crop production replaces food production, agricultural areas may be extended to areas with high carbon stock (European Commission 2019). This reduces natural carbon stocks and accelerates climate change. To avoid this phenomenon, particular feedstock is listed for the production of advanced biofuels. These feedstocks include algae, biowaste from households and industry, agricultural byproducts such as straw, forestry byproducts including forest residues resulting from logging and byproducts from forest industry such as bark and sawdust, and different oils and sludges excluding oils from energy crops (2018/2001/EU).

Therefore, the RED II dictates the acceptable feedstocks that are counted towards the national targets of advanced biofuel production. As solid biowaste from households and industry are already processed efficiently by anaerobic digestion to produce biogas, the most promising feedstocks to produce advanced biofuels in future in Finland are arguably the forest industry and agricultural byproducts. Municipal solid waste derived fuels are not included in the listed feedstock accepted for producing biofuels in the directive, thus decreasing its feasibility in biofuel production in coming years.

Currently the most advanced biofuels in Finland are from waste oils which are used to produce paraffinic diesel. Most of the renewable diesel is produced in Porvoo refinery through hydrotreatment with NExBTL process by Neste (Neste 2016). The product is a drop-in fuel, which may be used as a direct alternative for fossil diesel fuel. In addition, there is ethanol production from forest industry byproducts and industrial biowaste from food industry. Energy company St1 has been constructing these ethanol production facilities in Finland. (St1 2019).
3.5 Availability of biomass in Finland

Producing bio-SNG in industrial scale requires large amounts of biomass. For instance, a plant producing 100 megawatts of bio-SNG requires several hundred thousand tons of biomass. Biomass from forestry or agricultural by-products contain typically large amounts of moisture which makes long distance transportation unfavorable. Furthermore, the density of forest feedstocks and straw is low compared to coal or wood pellets used in energy production. Therefore, the local availability of biomass is fundamental to the feasibility of a bio-SNG production plant.

In Finland, the largest producer provinces of biomass by-products are Central Finland and Lapland, where logging and processing of coniferous trees is significant. In Central Finland, production of logging residues is especially high in addition to sawdust and other residues produced by stemwood processing. In Lapland, the large amounts of processed wood in sawmills result in high quantities of residual feedstocks. All the biomass feedstocks produced as by-products from agriculture, forestry, and logging are illustrated in Figure 13. The availability of two of the most potential feedstocks, logging residues and straw, are illustrated in figures 14 and 15, respectively. The estimated availability of biomass is measured as the annual technical potential of biomass extraction considering some environmental factors. The straw potential excludes the amount of straw that is needed for soil protection at farming sites i.e. the amount of straw that is left at the field. The forest residue availability includes the restrictions from protected forest areas and soil and water protection at the forest sites.
Figure 13. Total available biomass (dry mass) in Finland from forestry and agricultural byproducts in 2020. (S2Biom 2016)
In bio-SNG production, the natural gas network sets limitations, too. With an existing network the investment cost related to construction of new pipelines can be avoided. In Finland, the current gas network is located in Eastern and Southern Finland. The eastern-most location is in Imatra in South Karelia, where natural gas is transported from Russia to Finland. This increases the feasibility of a bio-SNG in Eastern Finland, as the availability of biomass is high in addition to existing gas infrastructure. In Central Finland and Lapland where the availability of logging residues is also high, the transportation of synthetic natural gas would require investments either long distance transportation or liquefaction if the preferred application of bio-SNG would be as transportation fuel.

Figure 14. Logging residue (dry mass) availability in Finland in 2020. (S2Biom 2016)

The straw availability is dependent on food production in Finland. Most of cereal crops are cultivated in southern parts of Finland and the potential of straw as a feedstock for bio-SNG production is largest in these provinces. The total mass of available straw is highest in Lapland of all the provinces. Generally, the dry mass of available straw is quite high even when compared to woody biomass. However, the real potential of its use as a feedstock is limited by its low density and high transportation and collection costs. Therefore, the highest potential of straw as a feedstock is in Southern Finland in Helsinki metropolitan area and Pirkanmaa, where largest amounts of straw can be collected in a dense area. Elsewhere the distances related to straw transport become long and the gas infrastructure does not allow injection of bio-SNG to the gas grid.
Figure 15. Cereal straw (dry mass) availability in Finland in 2020. (S2Biom 2016)
4 Simulation of bio-SNG process

Process simulation is typically a simplified illustration of a real-world system including modeling of different processes, unit operations, and chemical and physical phenomena. Through simulation, complex systems can be examined with less cost and effort. The development of computer science and computing power during 20th and 21st century led to creation of several simulation tools for industry to model processes. Specific software has been developed for different industries and purposes. In energy industry, simulation software can be used in studying the efficiencies of different processes, examine energy or exergy flows between the system, environment and process equipment, and to detect possibilities to improve the process to reach higher efficiency and profit.

The experimental section of this thesis is to compare the characteristics of a bio-SNG process with most promising biomass feedstocks. This part presents the simulation of the selected bio-SNG process and the methods used to reach the results. The simulation tool used in the thesis is Aspen Plus v11, which is based on the original simulation software released in 1981. Aspen Plus is a software developed by Aspentech Inc. Aspen Plus was created to be a widely used chemical and process engineering software for simulation purposes that could eliminate the need for many individual software which were owned by several different entities. (Aspen Technology Inc 2020b)

4.1 Bio-SNG process model

Aspen model that was created in this thesis is illustrated in Figure 16. The entire Aspen model can be seen in Appendix 1. The model is based on the bio-SNG demonstration plant constructed in Gothenburg. This demo plant was selected as the layout for the simulation in consequence of its relevance to current bio-SNG research and market. The Gobigas plant has generated plenty of research related to bio-SNG production and is a significant step towards the construction of a full-size plant. Furthermore, the amount of information and data available enables more accurate validation of the model as a whole and as individual units.

Most of the units in the process are modelled in Aspen’s flowsheet to reach as realistic results as possible. However, there are numerous intricate mass flows and reactions that are approximated more precisely outside the Aspen model. In some cases, there is exact information available of the modelled unit to reach arguably more precise results with a spreadsheet calculation than with Aspen modeling. Therefore, due to the complexity of the process, some units are simplified in the flowsheet and calculated as an Excel spreadsheet instead. These units include the initial drying that is integrated to the process, activated carbon beds which separate most impurities from the syngas, separation of carbon dioxide, and the ultimate unit which models drying of the bio-SNG. For these units, the mass flows are simplified in the flowsheet and energy consumption and efficiencies are acquired from literature.

The property method used in the model is the Peng-Robinson cubic equation of state with Boston-Mathias alpha function (PR-BM). This equation of state can be used for modelling processes including gases and petrochemicals which are nonpolar or slightly polar. The Boston-Mathias alpha function enables more accurate modeling in high reduced temperatures with the Peng-Robinson equation of state (Aspen Technology Inc 2020a) As the bio-SNG
The liquid-liquid equilibrium is calculated with the free water method, which assumes the water to be a clean liquid in addition to possible hydrocarbons. The steam tables applied for the thermodynamic properties of water are the NBS steam tables. Conventional and nonconventional components are defined in the model. The conventional components include gasification products containing carbon, hydrogen, nitrogen, sulfur and oxygen. The non-conventional components are defined as the solid materials in the process: biomass and ash. General coal enthalpy method is used to model the enthalpy of ash and biomass and IGT coal density model for density. Proximate, ultimate, and sulfur analysis for the nonconventionals are included in the simulation phase.
Biomass drying

Figure 17. Biomass drying in Aspen model.

The biomass drying process is modelled as energy-free unit in the Aspen model with a stoichiometric reactor, a separator, and a calculator block with Fortran code that is based on the Aspen Plus guide for solid modelling (Aspen Technology Inc 2013). The wet biomass is fed into the RStoic reactor with heated dry air. This reactor decreases the moisture content of the biomass to 20 % while the moisture is converted to steam. The remaining dried solid biomass and drying air is separated, and the exhaust gas is extracted from the process.

The energy consumption of the drier is calculated with Excel spreadsheet. The value of heat consumed in the process is based on Henrik Holmberg’s (2017) material on industrial biomass drying and evaporation. Holmberg presents different alternatives used in biomass drying in industry. In this model it is assumed that a belt dryer is used in the drying process. He mentions that the approximate consumption of heat is 4000 to 6000 kJ of heat per every kilogram of water evaporated from the feedstock. For the spreadsheet calculations, a consumption of 5000 kJ/kgH2O is assumed.

Gasification & combustion

The arguably most crucial part of the simulation is the combined process of gasification and combustion, which is illustrated below in figure 18. The bubbling bed gasifier is modelled using two reactor blocks, a separator block, and a calculator block. The first block, a yield
reactor, modifies the flow of dried biomass into separate molecular flows based on the proximate and ultimate analysis. The calculator block is used to define the exact amounts of different molecules created when the dried biomass is decomposed. The decomposed flow of biomass is directed to the second block, a Gibbs free energy reactor. This calculates the fractions of different gases that are created during steam gasification of biomass in temperature of 870 °C and a pressure of 1 bar based on the minimization of Gibbs free energy. To imitate the unreacted fraction of char in the gasification phase, a certain amount of char is set as an inert material.

![Diagram of gasification and combustion process in Aspen model.](image)

**Figure 18. Gasification & combustion process in Aspen model.**

The solid char is separated from the gaseous products in the separator block and directed to the combustion section. The principal point of this process is to create heat for the gasification through combustion of two products: the unreacted solid char from biomass gasification and recycled gaseous products from gasification. Therefore, a fraction of the product gases is directed to the combustion reactor alongside with the char. The combustion is modelled by a RGibbs reactor at a temperature of 900 °C and a pressure of 1 bar. The gas and char are combusted with a stream of preheated air. Heat from the combustion is directed to the gasification section. The flue gases from the combustion block are used to heat the steam for gasification block.

One major simplification made in the gasification process is the absence of tars in the gaseous products. This is due to the complexity of the tar formation processes, which are difficult to model accurately in Aspen Plus. Therefore, tar cleaning is not included in the process as separate block. However, other impurities including sulfuric compounds and ammonia are included in the model.
Gas cleaning & compression

The gas cleaning and conditioning blocks are the most simplified sections of this model. As most of the cleaning processes are complex, they are not modelled specifically in this thesis. Instead, the most energy intensive cleaning processes are included in the spreadsheet calculations, such as the removal of carbon dioxide. In the model, most impurities are removed prior to the compression of the gases. These removed impurities include all gaseous compounds containing nitrogen and sulfur. Additionally, most of steam in the stream is condensed in this block and removed from the process. This primary gas cleaning block is modelled as a separator block.

Figure 19. Gas conditioning & cleaning in Aspen model

After first gas cleaning block, the gas is compressed to 16 bars. The compression is performed in a 6-stage compressor, where the produced heat is used to produce superheated steam. The isentropic efficiency of the compression stages is assumed to be 0.8 and the mechanical efficiency 0.95. The inlet temperature to the compression stages is 250 °C and the outlet temperature is 350 °C. The compressed gas is directed to a water-gas-shift reactor (WGSR), where the molecular fraction of hydrogen is increased. In the WGSR the ratio of hydrogen and carbon monoxide is increased to 3.01. This is due to methanation, where this ratio must be over 3 to fully utilize the methanation process in methane generation. The WGSR is modeled as a stoichiometric block where the modelled reaction is the water gas shift reaction:

\[ CO + H_2O \rightarrow CO_2 + H_2 \]  

(4.2)

where the heat of reaction is -41.1 kJ/mol. The temperature of the gaseous stream is increased slightly as the reaction is exothermic.

Ultimately, the carbon dioxide is mostly removed from the process. As stated before, this is a very complex process and is simplified in this model. Therefore, the energy consumption is calculated in the spreadsheet calculations based on the values found in the literature. The
CO2 removal technology is assumed to be similar to the GoBiGas bio-SNG plant, which uses an amine absorption process. The process is simplified as one separator block in the model. The energy consumption of an amine absorption process utilizing methylethanoamine (MDEA) as a solvent is reported to be around 3.0 MJ of heat per kilogram of removed carbon dioxide (Alamia, Ösk Gardarsdóttir et al. 2017). This value is based on experimental data from the Gobigas plant. The heat is consumed during the regeneration of the MDEA solvent. The electricity consumption of the CO2 removal process is assumed to be negligible.

**Methanation**

The methanation process is the final stage where gasification syngas is transformed into containing mostly methane. The methanation process is based on the catalyzed adiabatic 4-stage Haldor Topsoe methanation. The process is very exothermic and controlling the temperature and heat of the process is emphasized. The methanation process consists of four adiabatic stoichiometric RStoic reactors (MET1 to MET4 in figure 20) with 4 coolers which transfer the generated heat for further utilization. The reaction in methanation blocks are

\[ 3H_2 + CO \rightarrow CH_4 + H_2O \]  

(4.3)

where the heat of reaction is -206 kJ/mol.

![Figure 20. Methanation in Aspen model](image)

In order to increase the fraction of methane as high as possible, some of the gas is recycled after second methanation reactor. The recycled gas is mixed with the gas coming to the first methanation step. The adiabatic methanation reactors are modelled between 250 and 700 °C and in 16 bars. The temperature of methanation reactors is limited to this because of the
chemical nature of the methanation reactor and the functional temperature of the nickel catalysts. The pressure required is high as it increases the conversion of methanation. After the last methanation reactor, the temperature of the gas is slightly over 300 °C. The whole methanation process is illustrated in figure 20.

**Bio-SNG drying**

Bio-SNG drying is modelled as a single separator block in the model, creating two different streams: one containing all the water still remaining in the syngas and the other methane and the other remaining gases such as carbon monoxide, nitrogen and hydrogen. For bio-SNG drying no consumption of energy is assumed for the sake of simplicity of the model.

**Heat production**

There are many major sources of surplus heat generated in the production process of bio-SNG. Some of these can be utilized directly in the process, such as in biomass drying. Finding applications for the produced heat is critical for the total energy efficiency of the process. In this model, the methanation sub-process is the largest source of surplus heat. The syngas entering this process is heated four times in succession and cooled each of these times. Furthermore, the temperature in which the syngas is heated in the adiabatic reactors varies from 700 °C during the first reactor to approximately 350 °C of the final reactor. Therefore, the methanation process can be utilized to produce superheated steam in addition to syngas with high methane concentration. The heat streams from methanation (HX1 to HX5 in figure 20) are directed to a separate heater, where liquid water is used to produce high pressure superheated steam. The produced steam is pressurized to 80 bars in temperature of 550 °C.

In addition to the methanation process, six stage compression creates significant heat. During this process, the gas is compressed from initial 1 bars to 16 bars and heated from 250 to 350 °C while being cooled between the stages. Additionally, cooling is required after the impurities are removed from the syngas among with surplus steam and before the final process of the bio-SNG plant which is bio-SNG drying. These offer further possibilities to extract heat from the process and improve the efficiency. These streams of surplus heat are utilized to create district heat: the heat streams from compression, impurity cleaning, and drying are mixed in the model to create 105 °C water of 10 bar pressure.

In the gasification and combustion sub-processes, the high temperature syngas from the gasifier and flue gas from combustion is used to heat input streams to the process. The syngas from gasification is applied to preheat the combustion air for the combustion process and the flue gas from combustion is utilized to generate superheated steam for the gasification process. The remaining heat that can be utilized from the flue gas after steam generation is directed to the district heat generation.

The heat needed for biomass drying is extracted from the same sources that are used to produce district heat. Therefore, no outside source of heat is needed for drying biomass. The amount of heat that is needed for drying is calculated in the spreadsheet and is based on the approximation that it consumes 5000 kJ of heat per kilogram of water evaporated from biomass (Holmberg, Ramm-Schmidt 2017).
Energy efficiency calculations

The energy efficiency calculations will be based on the equations 2.1 – 2.3. In this model, the cold gas efficiency and biomass to SNG efficiency will be calculated as they are introduced in chapter 2.6. The total plant energy efficiency will consider all the energy heat and electricity that is consumed and the heat and bio-SNG that is produced. Thus, the equation for the total plant energy efficiency becomes:

4.2 Biomass for simulation

The selected biomass for the Aspen Plus simulation was made based on the information about their suitability for gasification process and availability in Finland which were researched in this thesis. The feedstocks selected for modeling were spruce and wheat straw. Spruce was selected due to its high availability and high rate of utilization of forest chips in power plants. Although forest chips may contain many different parts of spruce, the properties of spruce stemwood are used in the model to approximate the possible composition of forest chips produced during spruce fellings.

Wheat straw was selected as the other modelled biomass. It has most auspicious characteristics amongst the agricultural feedstocks that were studied in this thesis. Wheat straw is adequately available in Finland, although in less amounts than wood-based biomass. The combustion characteristics are arguably superior to barley straw due to higher ash melting temperatures. While it has generally quite high ash content, it was seen as the preferred choice for purposes of the Aspen model.

The chemical properties of spruce forest chips and wheat straw is mainly based on the report of Alakangas et al. (2016) and the Phyllis2 database generated by ECN part of TNO. The following chemical attributes were selected as the input feed for the wet biomass in the model:

<table>
<thead>
<tr>
<th>Table 8. Proximate analysis</th>
<th>Spruce forest chips</th>
<th>Wheat straw</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture (wet, wt-%)</td>
<td>45</td>
<td>45</td>
</tr>
<tr>
<td>Fixed carbon (dry, wt-%)</td>
<td>19.1</td>
<td>19</td>
</tr>
<tr>
<td>Volatile matter (dry, wt-%)</td>
<td>76.4</td>
<td>73.5</td>
</tr>
<tr>
<td>Ash (dry, wt-%)</td>
<td>4.5</td>
<td>7.5</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Table 9. Ultimate analysis</th>
<th>Spruce forest chips</th>
<th>Wheat straw</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ash (dry, wt-%)</td>
<td>4.5</td>
<td>7.5</td>
</tr>
<tr>
<td>Carbon (dry, wt-%)</td>
<td>46.9</td>
<td>46</td>
</tr>
<tr>
<td>Hydrogen (dry, wt-%)</td>
<td>5.6</td>
<td>5.7</td>
</tr>
<tr>
<td>Nitrogen (dry, wt-%)</td>
<td>0.17</td>
<td>0.6</td>
</tr>
<tr>
<td>Chlorine (dry, wt-%)</td>
<td>0</td>
<td>0.1</td>
</tr>
<tr>
<td>Sulfur (dry, wt-%)</td>
<td>0.02</td>
<td>0.1</td>
</tr>
<tr>
<td>Oxygen (dry, wt-%)</td>
<td>42.8</td>
<td>40</td>
</tr>
</tbody>
</table>
5 Results

The modelled process was simulated with two scenarios: 1000 kg/h feed of wet forest chips and straw, both containing 45 % moisture (w.b.). This constitutes as approximately 2.33 MW of biomass input based on the lower heating values of the biomass for both cases. The mass flows into the process were similar in both processes to ensure the comparability of the model. Therefore, steam-to-biomass ratio, drying heat demand and electricity demand were nearly equal.

The results were analyzed by compiling energy flows from the most important components. The energy balance figures (21 & 22) show the most important flows of energy including the chemical energy in biomass feed, biomass char, produce gas and synthetic natural gas. The electricity demand of gas compression is also included; however, the electricity demand of water pumps is not shown as it is negligible. The heat flows between different process steps are illustrated including the heat for drying and for gasification, and the heat collected from cooling steps in different parts of the process.

Nearly every step of the model was simulated with Aspen Plus, excluding the heat for drying and the regeneration steam needed in the CO2 stripping process which were calculated with Excel. These were concluded to be more accurately represented by directly utilizing values from literature. Biomass drying has been extensively researched over the past years, and reliable data for its energy consumption is available. The CO2 removal process used in this model and at the Gobigas demonstration plant is an advanced and complicated process involving chemical solvents. Therefore, it was seen as necessary to use values from literature for energy consumption for CO2 stripping instead of including it in the Aspen model.
Figure 21. Energy balance of bio-SNG production plant with spruce forest chips
Figure 22. Energy balance of bio-SNG plant with straw
5.1 Efficiency and energy balance

Efficiencies of the plant are calculated similarly as presented in chapter 2.5. The most important efficiencies considered are cold gas efficiency, biomass-to-SNG efficiency and plant efficiency. The efficiency of the plant is calculated as the sum of chemical energy of the bio-SNG, produced superheated steam and district heat divided by the sum of the chemical energy in dry ash-free biomass, electricity and heat consumed in the regeneration of MDEA in the CO2 stripping process:

\[
\eta_{\text{PLANT,}j} = \frac{\sum_{j=1}^{n} E_j}{\sum_{k=1}^{n} E_k} = \frac{\dot{m}_{\text{SNG}} \cdot \text{LHV}_{\text{SNG}} + E_{\text{STEAM}} + E_{\text{DH}}}{\dot{m}_{\text{BM}} \cdot \text{LHV}_{\text{BM}} + E_{\text{ELEC}} + E_{\text{REGEN}}} \tag{5.1}
\]

The calculated efficiencies are presented in table 10. The cold gas efficiencies in both modelled cases are high, over 90%. This is due to the gas composition after gasification and the indirect gasification. There are little to none tar or hydrocarbons larger than methane in the simulation gas composition after gasifier, which increases the efficiency compared to demonstration circumstances. The reported cold gas efficiency of Gobigas plant is 71.7% on dry ash free fuel basis which is noticeably lower. In the case of the Aspen model, there are no major heat losses during the gasifying stage. At Gobigas, the heat losses from the gasifier were large, contributing a large part of the losses to the cold gas efficiency. Alamia et al. (2017) report, that the cold gas efficiency could be increased to 83.5% with feasible improvements to the demonstration plant. This is considerably closer to the values achieved with the Aspen model in this thesis.

Further factors affecting the efficiencies compared to the demonstration plant are the differences in char gasification rates and steam to biomass ratios. The char gasification rates in the model are relatively low compared to the demonstration plant. The reported value at Gobigas plant was 54%, whereas the model operates with lower values. The steam-fuel ratio i.e. the ratio of mass flow of water in biomass and fluidization steam to the dry ash-free mass flow of biomass also affects the efficiencies. The steam-fuel ratio of Gobigas is approximately 0.59 based on the experimental data from Alamia et al. In the model the steam-fuel ratio is 0.98 for both studied cases. Taking both char gasification and steam-fuel ratio into consideration, there is more hydrogen and oxygen and less carbon gasified into volatile gases in the model’s gasifier compared to the demonstration plant. The mass flow of the produced gas is also higher with the model’s cases, which may increase the cold gas efficiency of the studied model, especially if considerably more hydrogen and carbon monoxide gas is produced.

The cold gas efficiency for straw is calculated as 96.8% which is 4.3% higher than with forest chips. This may be due to the fact that straw has more carbon and hydrogen and less oxygen on mass basis in a dry ash free fuel compared to forest chips, which increases the lower heating value of the gasified gas from straw. Therefore, the cold gas efficiency of straw is somewhat higher.

Biomass-to-bio-SNG efficiencies are closer with the two cases. The biomass-to-bio-SNG efficiency for straw is 55.9%, which is larger by 2.3% compared to forest chips. At Gobigas, the reported value for bio-SNG efficiency was 61.8%. A major reason for the lower bio-SNG efficiencies of the model can be assumed to be the lower concentration of methane in
the gas after gasification. The Gibbs free method in calculating the gasification results with Aspen led to almost none methane in the syngas after gasifier. The reasonably low char gasification rate in the model also diminishes the amount of methane produced in the gasifier compared to Gobigas.

| Table 10. Efficiencies of Aspen models and Gobigas (Alamia et al. 2017)* |
|-----------------|-----------------|-----------------|
|                 | Case forest chips | Case straw | Gobigas |
| Cold gas efficiency (%) | 92.7            | 96.8        | 71.7*   |
| BM-to-SNG efficiency (%)  | 54.7            | 55.9        | 61.8*   |
| Plant efficiency (%)      | 78.6            | 80.5        | 57.7*   |

The plant efficiencies calculated with equation 5.1 are 78.6 % and 80.5 for forest chips and straw, respectively. Plant efficiency of straw is 2.3 % higher than with forest chips, as is the case with biomass-to-bio-SNG efficiency. The plant efficiency considers all of the inputs and outputs of the plant, including the produced heat and consumed electricity. With both cases, the values of produced district heat and steam are similar. Therefore, it is clear that the total plant efficiency of both cases is similar to the bio-SNG efficiency, too.

When the plant efficiency is compared to Gobigas, it should be noted that the energy flows taken into consideration are very different. In Alamia et al. (2017) paper, the efficiency considers only the bio-SNG as a viable product, whereas energy is consumed also by the rapeseed oil methyl ester (RME). In this thesis, also the superheated steam and district heat are considered as products from the process and taken into the efficiency calculations. Furthermore, the modelled process is rather exothermic, there is considerable amount of heat produced from gas cooling and during methanation. This explains the difference in total plant efficiency in the model and Gobigas demonstration plant.

5.2 Gas composition

The gas composition after gasification in the model is illustrated in table 11 with the demonstration data from the Gobigas plant. The main gas constituents are listed for both the wet and dry producer gases. The main differences of the model compared to the data from Gobigas are the concentrations of hydrogen, methane, and nitrogen. The amount of hydrogen in the model is larger by nearly 20 vol-% compared to the demonstration data. Furthermore, there is very little methane produced in the model. This is most likely due to the simulation method used in the gasification section. The Gibbs free energy model that minimizes the free energy in the gasifier does not favor the methane yield, which in turn leads to low methane and high hydrogen concentrations.

Concentrations of carbon monoxide, carbon dioxide and water are similar in the model and the demonstration plant data. The model results in slightly lower CO and CO2 concentrations and a little higher amount of H2O. However, they are within 3 vol.-% in the gas compositions after gasification.

There is little nitrogen in the model gas composition compared to the model. The only nitrogen in the Aspen process results from the biomass itself, whereas there is some nitrogen gas
fed into the Gobigas process as an inert gas for safety reasons. The use of nitrogen as an additive in the process is not favorable in a bio-SNG process, as the concentration will increase towards the end of the process. The resulting bio-SNG could have too much nitrogen for it to be of sufficient quality for grid injection.

Table 11. Gas composition after gasification (Thunman et al. 2018)*

<table>
<thead>
<tr>
<th>Gas (vol-%)</th>
<th>Gobigas, wood chips, wet</th>
<th>Model, wood chips, wet</th>
<th>Model, straw, wet</th>
<th>Gobigas, wood chips, dry</th>
<th>Model, wood chips, dry</th>
<th>Model, straw, dry</th>
</tr>
</thead>
<tbody>
<tr>
<td>H2</td>
<td>27.7*</td>
<td>40.0</td>
<td>40.8</td>
<td>39.9*</td>
<td>59.2</td>
<td>60.0</td>
</tr>
<tr>
<td>CO2</td>
<td>13.8*</td>
<td>11.4</td>
<td>10.9</td>
<td>19.9*</td>
<td>16.9</td>
<td>16.1</td>
</tr>
<tr>
<td>CO</td>
<td>16.6*</td>
<td>16.1</td>
<td>16.1</td>
<td>24.0*</td>
<td>23.9</td>
<td>23.6</td>
</tr>
<tr>
<td>H2O</td>
<td>30.2*</td>
<td>32.5</td>
<td>31.9</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>CH4</td>
<td>6.0*</td>
<td>0.003</td>
<td>0.001</td>
<td>8.6*</td>
<td>0.005</td>
<td>0.006</td>
</tr>
<tr>
<td>N2</td>
<td>3.7*</td>
<td>0.054</td>
<td>0.19</td>
<td>5.3*</td>
<td>0.080</td>
<td>0.28</td>
</tr>
<tr>
<td>CxHx</td>
<td>0.2*</td>
<td>0</td>
<td>0</td>
<td>0.3*</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>Tar</td>
<td>0.1*</td>
<td>0</td>
<td>0</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

The concentration of tar and larger hydrocarbons formed in Gobigas is rather low and contains only a small fraction of the chemical energy from the gasified biomass. Tar formation was not included in the model, and the high temperature gasification combined with the calculation method used for gasification did not result in practically any large hydrocarbons in the producer gas.

The simulated process yields similar values for most of the gas constituents in the producer gas after gasification. The largest differences are formed in the model due to the utilization of Gibbs free energy method which results in small methane yield. A slightly lower temperature during gasification would most likely increase the amount of methane produced. However, a lower temperature would most likely lead to a decrease in gasification reactivity of biomass (Alakangas et al. 2016). Therefore, a lower temperature is not considered a realistic method to increase the methane yield in the model.

Char gasification and steam-fuel ratio also affects the methane yield in the gasifier. The char gasification is higher in the demonstration case, which leads to higher number of hydrocarbons in the produced syngas. Furthermore, there is more steam utilized as fluidization gas in the model and the gasified biomass is more moist which results in higher steam-fuel ratios in the model. Thus, these two factors combined decrease the amount of produced methane and other larger hydrocarbons.

Other impurities considered in the model were sulfur and chlorine, which can appear in the syngas as hydrogen sulfide (H2S) and as hydrogen chloride (HCl). The concentration of these impurities was measured after the gasifier. The concentration of hydrogen sulfide was 77.7 ppm in dry gas for forest chip gasification and 404.1 ppm for straw. The amount of sulfur in dry straw is almost one order larger than with wood. Therefore, the larger amount of sulfur in produced syngas is no surprise. There was no chlorine assumed in the gasified forest chips, thus the concentration of chlorine compounds in produced syngas was zero. As for straw, there were 182.8 ppm of chlorine in the syngas after gasification.
Table 12. Composition of bio-SNG in the model (SFS-EN ISO 20765-1 2018)*

<table>
<thead>
<tr>
<th></th>
<th>Case forest chips</th>
<th>Case straw</th>
<th>Pipeline quality standard (SFS-EN ISO 20765-1)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH4 (vol-%)</td>
<td>99.3</td>
<td>98.4</td>
<td>70 – 100*</td>
</tr>
<tr>
<td>H2 (vol-%)</td>
<td>0.15</td>
<td>0.14</td>
<td>0 – 10*</td>
</tr>
<tr>
<td>N2 (vol-%)</td>
<td>0.38</td>
<td>1.31</td>
<td>0 – 20*</td>
</tr>
<tr>
<td>CO (vol-%)</td>
<td>0.17</td>
<td>0.15</td>
<td>0 – 3*</td>
</tr>
<tr>
<td>H2O (vol-%)</td>
<td>0</td>
<td>0</td>
<td>0 – 0.015*</td>
</tr>
<tr>
<td>CO2 (vol-%)</td>
<td>0</td>
<td>0</td>
<td>0 – 20*</td>
</tr>
<tr>
<td>O2 (vol-%)</td>
<td>0</td>
<td>0</td>
<td>0 – 0.02*</td>
</tr>
</tbody>
</table>

The composition of bio-SNG produced at the plant is presented in table 12 with the according standard for pipeline natural gas quality, SFS-EN ISO 20765-1. The amount of methane is high, as the model is optimized to convert most of hydrogen and carbon monoxide to methane during catalytic methanation. However, some amounts of hydrogen (forest chips 0.15 vol-%, straw 0.14 vol-%) and carbon monoxide (forest chips 0.17 vol-%, straw 0.15 vol-%) is assumed to remain in the produced synthetic natural gas. The concentration of both hydrogen and carbon monoxide are well within the quality standards, as they allow a concentration of 10 vol-% of hydrogen and 3 vol-% of carbon monoxide.

Nitrogen concentration is lower for forest chip bio-SNG (0.38 vol-%) than for straw (1.31 vol-%). Straw simulated with the model contains over three times as much nitrogen than forest chips, which is shown in the bio-SNG composition. However, as there is no further source of nitrogen, the amount is lower than the standards require for both cases. In the Gobigas demonstration plant, nitrogen was used as a purge gas in the gasification and combustion processes. In the model, this was not considered as it would increase the concentration of nitrogen significantly.

The water remaining in the syngas after methanation is assumed to be removed completely before the final product, bio-SNG. This is assumed to be feasible, as the gas is cooled before grid injection which would condense most water from the bio-SNG. Removal of remaining water is assumed to be made before final grid injection. In the standard for gas quality, the amount of allowed water is very low, under 0.015 percentage of gas volume or molar amount. Therefore, removal of all water is practically necessary in order to produce grid quality bio-SNG.

Overall, the model is a relatively close approximation of the Gobigas demonstration plant in terms of the gasification based on the data presented in table 11.
6 Conclusions

The process to produce bio-SNG via gasification of biomass and methanation of syngas remains quite untested. There have been only a handful of demonstrations and lab-scale experiments to convert cellulosic biomass into synthetic natural gas, most of them have been in Europe. Although the process to produce bio-SNG is unproven, the technology used in the process is reasonable seasoned. The gasification technology is equal to the bubbling or circulating bed combustion that is used widely in biomass combustion in heat and electricity production, and so is the biomass drying. Most of the gas cleaning and conditioning equipment have been used in other applications before, such as the water-gas shift reactors, which are commonly used in chemical industry to produce various products. Carbon dioxide scrubbing is in use in power plants to reduce greenhouse emissions and catalytic methanation has been applied in producing SNG from coal. Largest difficulties lie in tar cleaning or conversion from the syngas and in improving the total efficiencies of the process to make it more profitable.

With the inclusion of different biomass into the new renewable energy directive, which dictates the acceptable feedstocks for liquid and gaseous transport fuels, there is hope of bio-SNG production and research accelerating in near future. Waste biomass such as straw and forestry by-products combined with international and regional support systems might increase the rate of development for new facilities producing SNG from lignocellulosic feedstocks.

Modelling a bio-SNG process is challenging due to its complexity. The process can be divided into subProcesses which are itself relatively complex and demand proper research to apprehend. Therefore, compiling an accurate model of a complete bio-SNG production process is difficult. Simplifications to the model can lead to large errors in results as the phenomena that are being investigated may not be presented properly. Furthermore, the process involves complex gaseous and liquid compounds, properties of which are being continuously researched. Modelling tar formed during gasification is a major challenge, which needs to be researched in the future for proper understanding of the process.

In this thesis the focus was to replicate the largest bio-SNG demonstration plant, Gobigas, as an Aspen model and compare different waste and biomass with it. Most important components such as gasification and methanation were modelled as accurately as possible, and some simplified such as tar formation and CO2 stripping. Most unreliability was caused by calculation methods used in Aspen Plus and lack of modelling tar. However, the results calculated with Aspen Plus were reasonably close approximation of the demonstration plant data. Gas composition of syngas after gasification was within 3 vol-% with most gas compounds with the model and Gobigas, with hydrogen and methane concentration differing the most. Results of the model were the most different compared to Gobigas with cold gas efficiencies, which results from differences in parameters such as char gasification and steam-fuel ratio and from the large losses experienced in the demonstration plant. The measured cold gas efficiencies for the Aspen model were 92.7 – 96.8 % compared to 71.7 % of Gobigas, whereas the biomass-to-bio-SNG efficiencies for the model were 54.7 – 55.9 % compared to 61.8 % of Gobigas.

Wheat straw and spruce forest chips were selected for the simulation as they had most suitable chemical properties and their availability and potential are at least reasonably high in
Finland. The performance of both straw and forest chips was studied in Aspen with comparing the efficiencies and gas composition. Straw was seen to outperform spruce in efficiency calculations, with 1.2 – 4.1 % larger efficiency results in Aspen model. Although both straw and spruce are similar in their elemental analyses, the slightly higher carbon and hydrogen and lower oxygen content boost the efficiencies of straw in bio-SNG production. Both produced good quality bio-SNG which met all the quality standards for natural gas injected into grid. Therefore, purely on performance basis, straw is a superior feedstock.

However, the availability and physical properties hinder the potential of straw in bio-SNG production. The production volumes are significantly lower than with woody byproducts and straw lacks the existing infrastructure for procurement to industrial size power plant in Finland. The low density of straw and high moisture content are further negative factors compared to forestry products such as forest chips. Therefore, larger industrial plants producing bio-SNG from straw are not likely to be profitable in Finland.

Industrial and municipal solid waste was also studied in this thesis as a feedstock for bio-SNG production. The amount of waste has been increasing as landfilling of waste is decreasing due to legislation, which brings pressure to find new alternatives for waste utilization. However, waste gasification and methanation into SNG faces technical and legislative issues. Solid recovery fuels and other solid waste contain large quantities of impurities such as heavy metals and chlorine which would require intensive modifications to gas cleaning processes in order to reach acceptable levels of gas quality. Furthermore, industrial or municipal solid wastes are not considered as acceptable feedstocks for transport fuels in the EU RED II directive and are not accepted as such towards the national targets for renewable fuel targets.
Bibliography


Appendix. Aspen model for bio-SNG production