GASIFICATION-BASED BIOREFINERY FOR MECHANICAL PULP MILLS

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To My Wife Li-hua LIU
To My Father Xing-gong HE
To My Mother Cai-xia LIU
To My Sister Li HE
To My Sister Jiao HE
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ABSTRACT

The modern concept of "biorefinery" is dominantly based on chemical pulp mills to create more value than cellulose pulp fibres, and energy from the dissolved lignins and hemicelluloses. This concept is characterized by the conversion of biomass into various biobased products. It includes thermochemical processes such as gasification and fast pyrolysis. In mechanical pulp mills, the feedstock available to the gasification-based biorefinery is significant, including logging residues, bark, fibre material rejects, biosludges and other available fuels such as peat, recycled wood, and paper products. This work is to study co-production of bio-automotive fuels, biopower, and steam via gasification in the context of the mechanical pulp industry.

Biomass gasification with steam in a dual-fluidized bed gasifier (DFBG) was simulated with ASPEN Plus. From the model, the yield and composition of the syngas and the contents of tar and char can be calculated. The model has been evaluated against the experimental results measured on a 150 KWth Mid Sweden University (MIUN) DFBG. The model predicts that the content of char transferred from the gasifier to the combustor decreases from 22.5 wt.% of the dry and ash-free biomass at gasification temperature 750 °C to 11.5 wt.% at 950 °C, but is insensitive to the mass ratio of steam to biomass (S/B). The H₂ concentration is higher than that of CO under normal DFBG operating conditions, but they will change positions when the gasification temperature is too high above about 950 °C, or the S/B ratio is too far below about 0.15. The biomass moisture content is a key parameter for a DFBG to be operated and maintained at a high gasification temperature. The model suggests that it is difficult to keep the gasification temperature above 850 °C when the biomass moisture content is higher than 15.0 wt.%. Thus, a certain amount of biomass needs to be added in the combustor to provide sufficient heat.
for biomass devolatilization and steam reforming. Tar content in the syngas can also be predicted from the model, which shows a decreasing trend of the tar with the gasification temperature and the S/B ratio. The tar content in the syngas decreases significantly with gasification residence time which is a key parameter.

Mechanical pulping processes, as Thermomechanical pulp (TMP), Groundwood (SGW and PGW), and Chemithermomechanical pulp (CTMP) processes have very high wood-to-pulp yields. Producing pulp products by means of these processes is a prerequisite for the production of printing paper and paperboard products due especially to their important functional properties such as printability and stiffness. However, mechanical pulping processes consume a great amount of electricity, which may account for up to 40% of the total pulp production cost. In mechanical pulping mills, wood (biomass) residues are commonly utilized for electricity production through an associated combined heat and power (CHP) plant. This techno-economic evaluation deals with the possibility of utilizing a biomass integrated gasification combined cycle (BIGCC) plant in place of the CHP plant. Integration of a BIGCC plant into a mechanical pulp production line might greatly improve the overall energy efficiency and cost-effectiveness, especially when the flow of biomass (such as branches and tree tops) from the forest is increased. When the fibre material that negatively affects pulp properties is utilized as a bioenergy resource, the overall efficiency of the system is further improved. A TMP+BIGCC mathematic model is developed based on ASPEN Plus. By means of this model, three cases are studied:

1) adding more forest biomass logging residues in the gasifier,
2) adding a reject fraction of low quality pulp fibers to the gasifier, and
3) decreasing the TMP-specific electricity consumption (SEC) by up to 50%.

For the TMP+BIGCC mill, the energy supply and consumption are analyzed in comparison with a TMP+CHP mill. The production profit and the internal rate of return (IRR) are calculated. The results quantify the economic benefit from the TMP+BIGCC mill.

Bio-ethanol has received considerable attention as a basic chemical and fuel additive. It is currently produced from sugar/starch materials, but can also be produced from lignocellulosic biomass via a hydrolysis–fermentation or thermo-chemical route. In terms of the thermo-chemical route, a few pilot plants ranging from 0.3 to 67 MW have been built and operated for alcohols synthesis. However, commercial success has not been achieved. In order to realize cost-competitive
commercial ethanol production from lignocellulosic biomass through a thermo-chemical pathway, a techno-economic analysis needs to be done.

In this work, a thermo-chemical process is designed, simulated, and optimized mainly with ASPEN Plus. The techno-economic assessment is made in terms of ethanol yield, synthesis selectivity, carbon and CO conversion efficiencies, and ethanol production cost.

Calculated results show that major contributions to the production cost are from biomass feedstock and syngas cleaning. A biomass-to-ethanol plant should be built at around 200 MW. Cost-competitive ethanol production can be realized with efficient equipments, optimized operation, cost-effective syngas cleaning technology, inexpensive raw material with low pretreatment cost, high-performance catalysts, off-gas and methanol recycling, optimal systematic configuration and heat integration, and a high-value byproduct.

Keywords: Gasification, Mechanical Pulping, Bio-fuels, Power Generation, Biomass Residues, ASPEN Plus
Acknowledgements

This thesis would not have come to be without the tireless work of Per Engstrand, Wennan Zhang, and Olof Björkqvist, my supervisors. I am very grateful to them for support, inspiration, and encouragement. I greatly appreciate the support (discussion, offering data, revising article) from our colleagues, a Research Engineer Ulf Söderlind, a doctoral Student Kristina Göransson, and a Mechanic Lars Strömmer.

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Contents

Acknowledgements vii

List of Papers xi

1 Introduction 1
   1.1 Biorefinery concept for this work . . . . . . . . . . . . . . . . . . . . . 1
   1.2 Gasification . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 2
   1.3 TMP + BIGCC . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 4
   1.4 Biomass to Ethanol . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 7

2 Process and Modeling 9
   2.1 Process evaluation approach . . . . . . . . . . . . . . . . . . . . . . . . . 9
   2.2 Gasification . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 10
      2.2.1 DFBG . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 10
      2.2.2 Simulation . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 11
   2.3 TMP + BIGCC . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 13
      2.3.1 TMP + PM . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 13
      2.3.2 BIGCC and its implementation in a TMP mill . . . . . . . . . . . 13
      2.3.3 Economic evaluation . . . . . . . . . . . . . . . . . . . . . . . . . 17
   2.4 Biomass to Ethanol . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 18
      2.4.1 Process . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 18
      2.4.2 Simulation . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 19

3 Result and Discussion 21
   3.1 Gasification . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 21
   3.2 TMP + BIGCC . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 24
<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.3 Biomass to Ethanol</td>
<td>30</td>
</tr>
<tr>
<td>4 Conclusions</td>
<td>33</td>
</tr>
<tr>
<td>4.1 Gasification</td>
<td>33</td>
</tr>
<tr>
<td>4.2 TMP + BIGCC</td>
<td>33</td>
</tr>
<tr>
<td>4.3 Biomass to Ethanol</td>
<td>34</td>
</tr>
<tr>
<td>Bibliography</td>
<td>35</td>
</tr>
</tbody>
</table>
List of Papers

This thesis is mainly based on the following papers:


List of Figures

1.1 Products from syngas ........................................ 2
1.2 TMP energy consumption and supply. (Three processes (BIGCC, Boiler, and CHP) are modelled respectively, and compared. The model is designed to ensure that the steam demand is just satisfied, as the results show in the graph. The results demonstrate that the BIGCC process is the most efficient of the processes studied, and therefore, by implementing the BIGCC system in a TMP mill, less electricity will need to be purchased, and even less, with the TMP SEC decreased.) .... 6

2.1 Process evaluation approach ................................ 9
2.2 Concept of a DFBG system ................................. 11
2.3 Flowsheet of a DFBG biomass gasification model ....... 12
2.4 Flowsheet of a typical TMP–PM process ................. 14
2.5 Flowsheet of a BIGCC model (∗ ASPEN Plus module) ... 15
2.6 Block flow diagram of a biomass-to-ethanol process .... 19

3.1 Effect of temperature on the gasification (experiment—MIUN gasifier, wood pellets, sand bed material, S/B=0.6, τ=5.0 s, dp=5.0 mm) ........ 21
3.2 Effect of S/B on the gasification (experiment—MIUN gasifier, wood pellets, sand bed material, 850 °C, τ=5.0 s, dp=5.0 mm) .............. 22
3.3 Effect of biomass moisture content on the gasification temperature (experiment—MIUN gasifier, wood pellets, sand bed material, τ=5.0 s, dp=5.0 mm) ........................................ 23
3.4 Fraction of biomass combusted to hold the gasification temperature (Experiment—MIUN gasifier, wood pellets, sand bed material, 850 °C, τ=5.0 s, dp=5.0 mm) ........................................ 23
3.5 Effect of residence time on the gasification (experiment—MIUN gasifier, wood pellets, sand bed material, 850 °C, S/B=0.6, dp=5.0 mm) ... 23
3.6 TMP energy consumption and supply. (Three processes (BIGCC, Boiler, and CHP) are modelled respectively, and compared. The model is designed to ensure that the steam demand is just satisfied, as the results show in the graph. The results demonstrate that the BIGCC process is the most efficient of the processes studied, and therefore, by implementing the BIGCC system in a TMP mill, less electricity will need to be purchased, and even less, with the TMP SEC decreased.) 24

3.7 Effect of the amount of biomass fed into the gasifier or boiler. (The electricity production from the BIGCC plant increases with the amount of biomass used, and reaches the demand of the TMP+PM mill when about 10.6 MWh biomass per tonne of pulp is used as feedstock to the gasifier (Fig. 3.7(a)). BIGCC produces much more electricity and much less heat than CHP (Figs. 3.7(a) and 3.7(b)). Both the net revenue and the IRR increase significantly with the amount of biomass feedstock, and they increase faster as the electricity price increases. The pulpwood price is more sensitive than the electricity price to the economic profitability of a TMP mill (Figs. 3.7(c) and 3.7(d)). The electricity certificate is an effective incitement (Figs. 3.7(c) and 3.7(d)). It will be more profitable to invest in a BIGCC plant than in a CHP plant if more biomass materials are used for heat and power production (Figs. 3.7(c) and 3.7(d)).) 25

3.8 Effect of feeding reject fibres into gasifier. (BIGCC produces twice the electricity that CHP does, but is still far from the TMP+PM demand (Fig. 3.8(a)). Both BIGCC and CHP produce much more steam than TMP+PM demand (Fig. 3.8(b)). Both the net revenue and IRR increase with the amount of reject fibres (Figs. 3.8(c) and 3.8(d)). Both the net revenue and the IRR values of TMP+BIGCC are much higher than those of TMP+CHP, and the gap becomes bigger when more reject fibres are used for heat and power production (Figs. 3.8(c) and 3.8(d)). The economic profitability of a TMP mill is dominated by the pulpwood price and/or the electricity price (Figs. 3.8(c) and 3.8(d)).) 26

3.9 Effect of reducing the TMP-specific electricity consumption. (The TMP+PM electricity consumption cannot be fully made up by the electricity produced from the BIGCC plant, even if the TMP SEC goes down by 50%. But the gap becomes fairly small, and much smaller in comparison with the case of a CHP plant (Fig. 3.9(a)). With the decreasing of the TMP SEC, less steam can be supplied from the TMP process. Thus, the steam supply to the TMP+PM mill needs to be compensated for by the BIGCC (or CHP) plant (Fig. 3.9(b)). Lower TMP SEC leads to higher economic profit (Figs. 3.9(c) and 3.9(d)). For both the net revenue and the IRR, TMP+BIGCC > TMP+CHP > TMP+Boiler (Figs. 3.9(c) and 3.9(d)). The economic profitability of a TMP mill is dominated by the pulpwood price and/or the electricity price (Figs. 3.9(c) and 3.9(d)).) 27

3.10 Ethanol production cost as a function of plant scale 30
<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.11</td>
<td>Capital cost breakdown</td>
<td>31</td>
</tr>
<tr>
<td>3.12</td>
<td>Production cost contribution chart</td>
<td>31</td>
</tr>
<tr>
<td>3.13</td>
<td>Sensitivity of higher alcohols credit to the ethanol production cost</td>
<td>32</td>
</tr>
</tbody>
</table>
List of Tables

2.1 Model input data [1, 2] ......................................................... 15
2.2 Data for economic analysis [3, 4, 5, 6] ................................. 17
Chapter 1
Introduction

Gasification of biomass is a promising technology for producing valuable biofuels or renewable electricity and may hence be a key technology for reducing the emissions of climate gases especially carbon dioxide.

Biomass is seen as an important source of renewable energy for the future. From the viewpoints of economics, environment, land use, water use, and chemical fertilizer use, there is a strong preference for using lignocellulosic biomass. The raw materials could be forest/agricultural/industrial/municipal organic residues. These raw materials are much more abundant than food crops and can be harvested with much less interference to the food economy and much less strain on land, air, and water resources.

Indirect gasification technology can use inhomogeneous and moist biomass as a source for gasification. This means that this technology is a good candidate to be integrated into a mechanical pulping mill where the gasifier can utilize low quality fiber material and wood residuals from the pulp and paper production process. The purpose of this work is to present a model based evaluation of indirect dual bed gasification technology and its integration into a mechanical pulping process.

1.1 Biorefinery concept for this work

International Energy Agency defines biorefinery as “the sustainable processing of biomass into a spectrum of marketable products (food, feed, materials, chemicals) and energy (fuels, power, heat)”. The modern concept of “biorefinery” in the forest industry is dominantly based on chemical pulp mills to create more value than pulp fibres, and energy from the dissolved lignins and hemicelluloses. This concept is characterized by the conversion of biomass into various biobased products. It includes thermochemical processes such as gasification and fast pyrolysis.

In mechanical pulp mills, the feedstock available to the gasification-based biorefinery is significant, and can be classified into five categories: 1) residues from the thinning and logging such as tree tops and branches (about 30% of the harvested trees);
bark, taking up 10–13% of the pulp wood flow; 3) reject fibres (deteriorate paper properties; 10–20% of the total pulp fibres); 4) wastewater treatment sludge from sedimentation or primary clarification operations and secondary/biological treatment operations; 5) other available fuels such as peat, the recycled wood, and paper products. This project is to study co-production of bio-automotive fuels, chemicals, biopower, and steam via gasification in the context of the mechanical pulp industry. In particular, the aim is to study the possibility of commercializing conversion of the above-mentioned biomass residues into three main fuels/chemicals - dimethyl ether (DME), FT-diesel, and ethanol-rich mixed alcohols - plus electricity and steam.

The prospect of obtaining various fuels and chemicals from biomass through various thermochemical pathways is promising, as seen in Fig. 1.1.

![Figure 1.1: Products from syngas](image)

The gasification process is able to convert solid biomass into a kind of gas that can be used, after gas cleaning, in gas boilers, gas engines, or fuel cells. The gas can also be converted into SNG or Green Gas, FT-Diesel, DME, or ethanol, etc. SNG can be injected into the gas grid or can be used as transport fuel (Bio-CNG).

### 1.2 Gasification

The DFBG (also referred to as indirect or allothermal gasifier), with steam as the gasification agent, can be used to produce a syngas of 12–20 MJ/Nm³ (LHV), which is the appropriate syngas for the downstream synthesis of transportation fuels and chemicals. The gasifier consists of two separate reactors: a steam gasifier used to convert biomass into syngas, and an air combustor used to burn the char and to provide
the necessary heat for biomass steam gasification. A typical DFBG is composed of a BFB (bubbling fluidized bed) and a CFB (circulating fluidized bed). In comparison with the auto-thermal pressurised gasifier, the advantages of a DFBG are [7]:

- no oxygen demand to obtain nitrogen-free syngas
- low temperature operation
- low investment cost
- no or simple pre-treatment of biomass
- easy feeding of biomass
- suitable for the biomass-based S/M scale bio-automotive fuel plants

Reduction of tar/CH$_4$ in the syngas is a major challenge to the commercialization of DFBGs for synthetic fuels production. Tar condenses with decreasing temperature (below 400 $^\circ$C), which leads to knotty problems to the downstream processes. The problems are e.g., fouling and blocking of filters and pipeline passages, dirty working environment, heavy waste water treatment, poisoning of catalyst in synthesis reactors, etc. Moreover, tar results in a part of energy lost from the syngas.

For the R&D of DFBG technology, a great deal of effort has been contributed to the experimental study. Very little work can be found on the mathematic simulation, especially, the simulation on tar evolution in biomass gasification.

In coal industry, a chemical percolation devolatilization (CPD) model was developed to model coal devolatilization based on characteristics of the chemical structure of the parent coal. The direct use of chemical structure data as a function of coal properties is useful to justify the model on a theoretical basis rather than an empirical basis. Fletcher [8] made a contribution to the CPD model development. The predictions of the amount and characteristics of syngas and tar fit well with available measurement data. The application of CPD model to biomass has not been seen to a certain extent.

ASPEL Plus is often used to simulate the coal/biomass conversion processes. Sotudeh-Gharebaagh et al. [9] used it to predict coal combustion in a CFB combustor regarding combustion efficiency, emission levels of CO, SO$_2$, and NO$_x$, as well as concentration profiles of O$_2$ and CO. Lee et al. [10] presented the kinetic expressions for steam–char and oxygen–char reactions in the atmospheric coal gasification in a fluidized bed reactor. These expressions were cited by Nikoo et al. [11] in the biomass gasification simulation with ASPEL Plus. Doherty et al. [12] developed a simple model with ASPEL Plus for the simulation of biomass gasification with steam and air in a CFB gasifier. Parameter sensitivity analyses, especially the effect of air pre-heating on gasification, were performed. The result is plausible. Corella et al. [13] set up a model for atmospheric biomass gasification in a CFB gasifier. In this model, the hydrodynamics and reaction kinetics aspects were considered, and the issues on the tar formation and reduction were discussed. On the basis of this model, Corella
et al. [13] investigated the axial profiles of temperature, and the concentrations of ten different substances (H₂, CO, CO₂, tar, char · · ·). However, the details have not been presented.

In this work, biomass gasification with steam in a DFBG is simulated with ASPEN Plus. To deal with the simulations on biomass pyrolysis and the evolutions of tar and char, some subroutines are designed based on relevant empirical equations from literature. The submodels are set up based on the thermodynamics and reaction kinetics. The yield and composition of the syngas, and the contents of tar and char can be calculated from the model. The concerned variables are: structure of the gasifier, gasification temperature (T), steam to biomass mass ratio (S/B), biomass moisture content (M), gasification residence time (τ) and biomass/char particle size (dₚ). The model is evaluated against the experimental results measured on a 150 KWth MIUN (Mid Sweden University) DFBG.

1.3 TMP + BIGCC

Paper mill as that using thermomechanical pulp (TMP) process has a very high wood-to-pulp / wood-to-paper yield (>95%), but consumes a large amount of electricity (about 2 MWh per tonne of pulp). As the TMP-process normally is integrated with the paper production, and the electricity utilized in the refining is converted to steam and hot water, one can say that the electric power is utilized twice, first to produce the pulp and then to produce steam for drying paper. Chemical pulp mill as that producing bleached softwood kraft can be self-sufficient in both heat and electric energy use, but has a low wood-to-pulp yield (45%). About half of the pulp wood, i.e. lignin, extractives, and most of the hemicellulose, is dissolved to become black liquor which is incinerated to produce steam and electricity. This way, black liquor has become the most important biomass resource in the pulp and paper industry and also the largest bioenergy source in Sweden. There has been a focus in the technology development on black liquor gasification for cogeneration of heat and power [14, 15, 16, 17, 18]. On the other hand, no development work has been done for gasification-based cogeneration of heat and power in the context of a TMP mill.

The pulp quality possible to achieve by means of mechanical pulping processes is a prerequisite for the production of printing paper and paperboard due especially to their important functional properties of these products, such as printability and stiffness. In the Nordic countries, a larger share of paper production uses mechanical pulp than anywhere else in the world [19]. Improved energy-efficient refining has been an important research subject for a long period of time [20, 21, 22, 23, 24, 25, 26]. FSCN at Mid Sweden University, together with Scandinavian forest and supplier companies, PFI and other Scandinavian universities, is working with a large research program with the goal of showing how to design the TMP and CTMP mills of the future, with 50% reduced specific electricity consumption (SEC) compared to that with the present state-of-the-art technology. Today, a large part of the biomass such as the logging residues (branches and tree tops), is normally not utilized as a source
of bioenergy. Furthermore, the possibility of utilizing the fraction of fibre materials that has a negative influence on the final product properties as a source of bioenergy has not been evaluated. Normally, only the bark, shavings, and bio-sludge are utilized in the present bark boilers of CHP systems of mechanical pulp mills. It is also important to study the potential of achieving cost-effective and energy-efficient heat and power production from those biomass residues, especially for a TMP mill.

The back-pressure system with a steam turbine is popular in TMP plant. This system has low electricity generation efficiency of less than 30%. In comparison, a gas turbine system has electricity generation efficiency up to 40%, while a biomass integrated gasification combined cycle (BIGCC) may reach the efficiency up to 50%. Implementation of a BIGCC in a TMP mill may create significant potential for self-sufficient production of electricity and for more economic benefit to the TMP mill.

Ong’iro, et al. [27, 28] developed several models to study the effects of design and operation parameters on the energy efficiency of a coal-based integrated gasification combined cycle (CIGCC) and an integrated gasification humid air turbine cycle respectively. A thermodynamic model of a combustion power plant was also developed with ASPEN Plus to evaluate the thermodynamic feasibility of cogeneration with higher ratio of electricity to heat. Using ASPEN Plus, Emun et al. [29] simulated a CIGCC, and pointed out that the overall energy efficiency can approach 45%. Eriksson and Kjellström [3] performed a techno-economic analysis over a CHP plant integrated into a wood-based ethanol production process. They claimed that utilization of the residues from an ethanol production plant through a CHP system was the most promising. For a CHP plant, the annual fixed operation cost was assumed to be 2% of the total investment, and the capital investment was only about 10% of the total cost of a wood-based ethanol production plant [3].

In the present work, a BIGCC model is set up with ASPEN Plus. This model is specially developed for a combined TMP+BIGCC system. The syngas is produced from the biomass residues collected at the pulpwood logging site, and the rejects from the TMP mill by means of a dual fluidized bed gasifier (DFBG) system. The obtained syngas will fuel the gas turbine cycle in the BIGCC system to generate electricity, while the steam used in the PM process is extracted from the steam cycle.

A techno-economic analysis is performed on the basis of this model. Three cases are studied:

1) adding more forest biomass logging residues in the gasifier,
2) adding a reject fraction of low quality pulp fibres to the gasifier, and
3) decreasing the TMP-specific electricity consumption (SEC) by up to 50%.

The production profit and the internal rate of return (IRR) are calculated. The results will quantify the economic benefit from a TMP+BIGCC mill in comparison with a TMP+CHP mill.

The energy balance over the BIGCC (or Boiler or CHP) process is studied by applying the model developed in this study. The steam consumed in the PM process will be obtained from the TMP and BIGCC (or Boiler or CHP) processes. Compared with the CHP, the BIGCC produces much more electricity, because the BIGCC system has a higher electricity generation efficiency of 45%. As a result, by implementing the BIGCC system in a TMP mill, less electricity will need to be purchased, and even
Figure 1.2: TMP energy consumption and supply. (Three processes (BIGCC, Boiler, and CHP) are modelled respectively, and compared. The model is designed to ensure that the steam demand is just satisfied, as the results show in the graph. The results demonstrate that the BIGCC process is the most efficient of the processes studied, and therefore, by implementing the BIGCC system in a TMP mill, less electricity will need to be purchased, and even less, with the TMP SEC decreased.)

less, with the TMP SEC decreased (Fig. 3.6). The residual low grade energy from the BIGCC (or Boiler or CHP) process is utilized in DH system.
1.4 Biomass to Ethanol

In the transport sector, energy consumption accounts for more than 30% of the total energy supply in developed countries, which is 98% dependent on fossil fuel and difficult to be replaced with sustainable and CO₂ neutral bio-automotive fuel in the transport fuel market. Bio-ethanol is the major bio-automotive fuel today, and is rapidly expanding as a fuel additive in addition to its use as a chemical raw material. It is already commonly used in gasoline blended with 10% ethanol, and can be blended up to 85% by flexible fuel technology [30]. Through on-board reforming to hydrogen, ethanol is also suitable to be applied in future fuel cell vehicles [31]. As an alternative to methyl tert-butyl ether (MTBE) which raises serious health risks, ethanol enhances octane rating of gasoline and reduces emissions of CO, VOC (volatile organic carbon) and particulates [31].

Ethanol is commercially produced via two routes, hydrolysis-fermentation of sugars from corn or sugar cane and hydration of petroleum-based ethylene. The ethylene hydration route is unattractive for large-scale ethanol production, which is dependent on imported crude oil and the volatile price of crude oil. Bio-ethanol feedstock can be divided into two major groups, sugar/starch materials and lignocellulosic biomass. Ethanol produced from sugar/starch materials is referred to as the 1st generation bio-automotive fuel [7]. However, with regard to raw material potential, sustainability, fossil fuel consumption, farmland occupation, conflict with productions of food and high value chemicals, as well as net CO₂ emission, bio-ethanol produced from lignocellulosic biomass is more attractive, which is referred to as the 2nd generation bio-automotive fuel. Production of the 2nd generation bio-automotive fuels is too expensive at present due to complex conversion process and its demands on large scale and big investment. However, the production cost is expected to drop down as the lignocellulosic biomass feedstock is cheap, abundant, and does not compete with food production. Lignocellulosic biomass is envisaged to provide a significant portion of the raw materials for bio-ethanol production in the medium and long-term.

Lignocellulosic biomass can be converted into bio-ethanol by gasification-synthesis, gasification-fermentation, and hydrolysis-fermentation. In the hydrolysis-fermentation route, the biomass-to-ethanol conversion efficiency is fairly low since 25 - 30 wt % biomass as lignin cannot be broken down, and hydrolysis of cellulose and hemicellulose is difficult. A significant portion of 5-carbon sugars from hemicellulose hydrolysis cannot be completely metabolized into alcohol. Further achievements strongly depend on the development of cheaper and more efficient enzymes and microorganisms [32, 33, 34, 35]. The techno-economic assessment of production of ethanol (hydrolysis-fermentation) from lignocellulosic biomass has been made [36, 37, 38, 39, 40, 41, 42].

In the gasification-synthesis route, biomass is gasified into raw syngas which is further reformed, cleaned, compressed, heated and converted into mixed alcohols from which ethanol and higher alcohols are obtained through a series of separation and
purification treatments. This route has the following advantages — short reaction time, inexpensive and abundant raw materials, nearly complete biomass conversion and so on. However, commercialization of catalytic synthesis of ethanol from syngas is under development [43].

On the other hand, catalytic synthesis of methanol from H$_2$ + CO is an existing commercially available technology. It is evaluated [44] that MeOH yield is 0.31 Kg MeOH per Kg dry biomass (Japanese cedar 400 t$_{\text{dry/d}}$), the production cost lies between 12.8 and 78.0 ¥/m$^3_{\text{MeOH}}$, and the energy recovery rate is 31% (higher than the wood-to-power generation efficiency 22%). The lowest production cost is achieved when the shortfall of heat and power is compensated by an auxiliary CHP plant fed with biomass, char, off-gas, etc.

The present work aims at commercial ethanol production via thermo-chemical route in the near future. The thermo-chemical process is designed, simulated and optimized. ASPEN Plus is the main software used in the simulation. Techno-economic evaluation is conducted in terms of ethanol yield, synthesis selectivity, carbon and CO conversion efficiencies, and ethanol production cost. Sensitivity of the important techno-economic parameters is also analyzed.
Chapter 2

Process and Modeling

2.1 Process evaluation approach

The process evaluation approach is schematically described (Fig. 2.1). ASPEN Plus
is an excellent modeling tool which is versatile and relatively easy to use. It can be used to model complicated chemical engineering systems. It consists of many built-in model blocks simulating various unit operations. Where more sophisticated block ability is required, additional information may be added into a block in a form of FORTRAN subroutines, or entirely new user blocks may be created. It has an extensive material property database where the diverse stream properties required to model the material streams are all available with an allowance for data addition. Some equations and data are also packaged in a specific Property Method set in ASPEN Plus. The involved substances and operation conditions are crucial in choosing a proper Property Method. Equations and data for the calculations can be accessed from the database. The simulation can be controlled with some FORTRAN routines and design specifications.

2.2 Gasification

Biomass gasification is a process that converts biomass into H₂, CO, CO₂, CH₄, etc. It could be regarded as an incomplete combustion process. In a gasifier, the biomass undergoes several processes: drying, devolatilization, combustion, and gasification (around 850 °C).

Gasifying agents could be air, O₂, H₂O, etc. Owing to its composition, the gas from air gasification is not suitable for synthesis purposes. Pure O₂ is expensive. For steam gasification, the gasification heat could be either externally or internally provided. The idea of supplying heat internally can be realized with a specially designed DFBG.

2.2.1 DFBG

A DFBG consists of two beds (Fig. 2.2). One is blown with steam to gasify biomass and to produce syngas. The other is blown with air to burn char to produce heat. Bed material is circulated to transfer heat from combustion bed to gasification bed. Flue gas is prevented from mixing with the produced syngas. Since air is not injected into the gasifier directly, N₂-free syngas can be obtained. The temperatures in the reactors are maintained by the char combustion and bed material recirculation. Some options for DFBG technology are possible from different combinations of the BFB and the CFB. So far, the most attractive design is supposed to have biomass gasification in the BFB and char combustion in the CFB from the viewpoints of particle circulation, fuel conversion, tar cracking, etc. Some typical combination (gasifier/combustor) examples are BFB/CFB (Güssing [45, 46, 47], Trisaia [48], CAPE [49], Chalmers [50, 51], MIUN [7, 1, 2], CFB/BFB (MILENA [52, 53]), and CFB/CFB (SilvaGas [54, 55], Taylor [54]). The measured H₂/CO ratios range from 0.45 (SilvaGas) to 3.5. The CH₄ concentration is generally around 10.0 vol. %. The tar contents range from 0.5 g/Nm³ (Trisaia, Güssing) to 32.0 g/Nm³ (MIUN, Milena). The measurement data are scattering in a wide range. Similar summaries can be found [1, 2, 56].
2.2 Gasification

A MIUN gasifier is schematically shown in Fig. 2.5. The gasifier consists of an endothermic steam-fluidized bed gasifier and an exothermal CFB combustor. The biomass treatment capacity is 150 kWth, that is, approx. 30 kg/h. The heat carrier is silica sand (150 µm, in diameter). The bed material circulation is controlled with the air velocity in the combustor, the steam velocity in the gasifier, the total solids inventory, and the aeration in a tube connecting the gasifier and the combustor at the bottom. The Process Logic Controller-operated feeding system is designed for constant biomass feeding. The biomass is fed into a pneumatic oscillatory vane feeder to achieve high precision. The fluidization agent in the gasifier is steam, and the syngas is drawn off from the top of the gasifier. The residual biomass char is transferred along with bed material into the combustor through the lower pressure lock. In the combustor, the fluidization agent is air participating in the char combustion (950–1050 °C). The hot bed material is separated from the flue gas through a cyclone and recycled into the gasifier through the upper pressure lock (loop seal pot) which is designed to prevent gas leakage. The heights of the gasifier and combustor are 2.5 m and 3.1 m, and the inner diameters are 300 and 90 mm, respectively. It is 0.7 m from the surface of bed material to the distributor, and about 0.2 m to the feeding point. The experiments were carried out at 750 °C, 800 °C, and 850 °C, respectively. The steam input into the gasifier was held at 4.0 kg/h. Wood pellets are from SCA BioNorr AB. The S/B ratios are 0.3, 0.6, and 0.9, respectively. The details on experiments have been reported elsewhere [1, 2].

2.2.2 Simulation

ASPEN Plus consists of various sub-models simulating various unit operations. It can be used to model complicated chemical engineering systems. Equations and data for the calculations are accessed from ASPEN Plus databases. Customized models can be created to extend the capability of ASPEN Plus. Here, the RGibbs module
was mainly adopted. The *RGibbs* module performs calculations according to the *Gibbs* free energy theory. Some subroutines are integrated for simulating biomass devolatilization and the evolutions of tar and char. Relevant empirical correlations [10, 11, 13, 57, 58] are adapted and employed in the subroutines. As illustrated in Fig. 2.3, this model consists of four sections. The bed materials are not considered in the phase and chemical equilibria calculations. $Q_{cg}$ denotes a stream of heat carried by bed material. With respect to tar and char, as nonconventional components, the calculations will be conducted independently from using the database in ASPEN Plus. $S/B$ is calculated according to:

$$S/B = \frac{M_{\text{steam}} + M_{H_2O \text{ in biomass}}}{M_{\text{dry biomass}}} \quad (2.1)$$

where, $\dot{M}$ denotes the mass flow (kg/s). A number of hypotheses for the model are listed:

- the process is of steady state;
- the biomass feedstock and bed materials are fed at an uniform temperature;
- ash as well as bed materials are inert;
- the char particles are spherical and of uniform size;
- the mixing of solid particles is perfect;
- the combustion and gasification of biomass are described by the shrinking core model.
2.3 TMP + BIGCC

In the present work, a BIGCC model is set up with ASPEN Plus. This model is specially developed for a combined TMP+BIGCC system. The syngas is produced from the biomass residues collected at the pulpwood logging site, and the rejects from the TMP mill by means of a dual fluidized bed gasifier (DFBG) system. The obtained syngas will fuel the gas turbine cycle in the BIGCC system to generate electricity, while the steam used in the PM process is extracted from the steam cycle.

2.3.1 TMP + PM

Pulp quality and energy economy are the key issues in any TMP plant. Energy consumption is high for any mechanical pulping method, and it is especially so for TMP. The final pulp quality is controlled by the energy input to the mechanical processing. A large total energy input seems to be necessary in order to increase the flexibility of the stiff wood fibres during refining. The largest energy input is in the peripheral area of the refining zone. The major part of the energy consumed in refining is converted to heat (steam). In an integrated mill, the steam can be recovered for heating purposes.

Fig. 2.4 presents the mass and energy balances over a TMP–PM line. The streams of wood, pulp, paper, electricity, steam, feedwater, warm water, biomass residues, effluent, etc. are considered.

A typical flowsheet of the TMP process is proposed (Fig. 2.4). Some of the input data to this model are from a TMP+PM plant, Hallsta, Holmen Paper [4].

A CHP plant represents an energy centre for the whole TMP+PM plant. Biomass residues from the TMP+PM plant are utilized through the CHP plant to produce electricity and steam needed by the TMP+PM plant. In this study, the application of gasification technology is considered. The original CHP plant is thus intended to be replaced by a BIGCC plant.

Available biomass residues from a TMP plant are significant and include: 1) logging residues, amounting to 30% of the tree; 2) bark, 12% of the stem, 3) bio-sludge, 2% of the pulpwood, and 4) reject fibres, 10% of the pulp, as shown in Fig. 2.4. Fig. 2.4 implies that 0.7 tonne of biomass residues are available from the production of each tonne of pulp. Additional forest biomass logging residues (including peat) can be obtained from other applications such as the production of chemical pulp, timber as construction material, and so on.

2.3.2 BIGCC and its implementation in a TMP mill

Coal-based IGCC technology has been effectively commercialized. Biomass-based IGCC technology, however, is in the pre-commercialization stage, with the syngas cleaning as the key issue. In this study, DFBG (Fig. 2.5), an indirect gasifier, is chosen for biomass gasification. It consists of a fluidized bed gasifier and a circulating fluidized bed riser as the char combustor to provide heat for the biomass gasification.
Figure 2.4: Flowsheet of a typical TMP–PM process
The main advantages of the gasifier are that it can easily treat various solid fuels, including high moisture biomass, and the biomass pre-treatment is fairly simple [5]. The detailed data on such a gasifier can be found in the literature [1, 2]. In accordance with the energy contents and conversion efficiencies, a routine is programmed to calculate how much syngas can be produced.

![Figure 2.5: Flowsheet of a BIGCC model (ASPEN Plus module)](image)

<table>
<thead>
<tr>
<th>Table 2.1: Model input data [1, 2]</th>
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<tbody>
<tr>
<td><strong>LHV biomass</strong> [MJ/kg]</td>
</tr>
<tr>
<td><strong>efficiency</strong></td>
</tr>
<tr>
<td>gasification</td>
</tr>
<tr>
<td>combustion</td>
</tr>
<tr>
<td>boiler</td>
</tr>
<tr>
<td>DH</td>
</tr>
<tr>
<td><strong>syngas yield</strong> [m$^3$/kg daf biomass]</td>
</tr>
<tr>
<td>composition [vol.%]</td>
</tr>
<tr>
<td>LHV [MJ/kg]</td>
</tr>
<tr>
<td><strong>flue gas</strong> [°C]</td>
</tr>
<tr>
<td><strong>working steam</strong> [bar]</td>
</tr>
<tr>
<td>[°C]</td>
</tr>
<tr>
<td>steam (in PM) [bar]</td>
</tr>
<tr>
<td>$^a$ DH=(biomass energy-steam and electricity from BIGCC to TMP+PM)×0.5</td>
</tr>
<tr>
<td>$^b$ hot inlet-cold outlet temperature difference, input data for a submodel of a heat exchanger combining a gas and steam turbine cycles</td>
</tr>
</tbody>
</table>

A detailed study on syngas cleaning for good-quality syngas was made by Göransson et al [1]. Eriksson and Kjellström [3] suggested that the additional investment for a separate gasifier and gas cleaning unit amounts to about 25% of the CHP investment.
The cleaned syngas will be combusted to generate electricity through a gas turbine system. The exhausted high temperature flue gas goes through a heat exchanger to generate the working steam (100 bar, 565 °C). The working steam is expanded through a steam turbine cycle from 100 bar to 0.1 bar to generate electricity. A certain amount of 2.5 bar steam is extracted for use in PM. The extracted steam and the steam from the refining process of TMP will meet the overall steam demand of PM.

For comparison, two other systems, TMP+Boiler and TMP+CHP systems, are also modelled. The waste heat after the production of electricity and steam is utilized by a district heating (DH) system at an efficiency of 50%. Table 2.1 lists the model input data.

Simulation

ASPEN Plus is an excellent modelling tool which is versatile and relatively easy to use. It can be used to model complicated chemical engineering systems. It consists of many built-in model blocks to simulate various unit operations. If more sophisticated block ability is required, additional information will be added into the block in a form of FORTRAN subroutines, or an entirely new user block will be created.

The BIGCC model (Fig. 2.5) is set up with several ASPEN Plus modules. Each module simulates a unit process. The electricity generation efficiency of a BIGCC depends on the efficiencies of gasification, compression, combustion, expansion, etc. The input data are collected from a variety of sources including experimental and literature data. The mass and energy balance is computed throughout every step in the BIGCC.

A compressor or a turbine is simulated with a module “Compr” which can calculate the power consumed or produced when the pressure ratio, isentropic, polytropic, and mechanical efficiencies, and clearance volume are given.

The combustor is modeled as a reactor in ASPEN Plus with the module “RGibbs” which executes equilibrium calculations by Gibbs free energy minimization. When a system does not reach a complete equilibrium state, “RGibbs” can also be used by specifying the extent of equilibrium.

The heat exchanger is simulated with the module “Heatx”, the pump with the module “Pump”, and the flow splitter with the module “FSplit”.

The accessed equations and data are packaged in a specific Property Method set in ASPEN Plus. The involved substances and operation conditions are crucial in choosing a proper Property Method. In modeling the gas turbine system, the Property Method PR-BM is adopted, and for the steam turbine system, the Property Method STEAM-TA is used.

The ASPEN Plus function block “Design Spec” is applied to predict how much steam can be produced from the heat exchanger. In essence, it is the multi-objective optimum design method adopted. The 100 bar, 565 °C steam is targeted to be produced. Simultaneously, the temperature of the exit flue gas is set to be 50.8 °C. The manipulated variable is “MASS-FLOW” (flowrate, kg/s). Similarly, for the module “FSplit” (simulating the distribution of the stream of steam), the function block “Design Spec”
is also employed. The original total stream of steam will be divided into two parts. The aim is to take out a certain amount of 2.5 bar steam to be used in PM. In optimization, the target value is the known amount of 2.5 bar steam. The manipulated variable is “FLOW/FRAC” (stream fraction).

### 2.3.3 Economic evaluation

The economic profitability of TMP+BIGCC is evaluated in terms of the net revenue (after subtracting the depreciation) and IRR. IRR is also called the discounted cash flow rate of return or the rate of return. IRR disregards the absolute amount of money to be gained, and is an indicator of the efficiency, quality, or yield of an investment. An investment is considered acceptable if its IRR is higher than an established minimum acceptable IRR. Generally, the higher the IRR, the better the investment.

<table>
<thead>
<tr>
<th>Table 2.2: Data for economic analysis [3, 4, 5, 6]</th>
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<tbody>
<tr>
<td>operation time</td>
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<tr>
<td>power generation efficiency</td>
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<td></td>
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<tr>
<td>specific capital investment</td>
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<td>paper</td>
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<td>heat</td>
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<tr>
<td>O &amp; M cost</td>
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<tr>
<td>electricity price</td>
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<tr>
<td>electricity certificate price</td>
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<tr>
<td>pulpwood</td>
</tr>
<tr>
<td>biomass to BIGCC</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>plant lifespan</td>
</tr>
<tr>
<td>discounted payback period</td>
</tr>
<tr>
<td>annual depreciation</td>
</tr>
<tr>
<td>the first 10 years</td>
</tr>
<tr>
<td>the next 20 years</td>
</tr>
<tr>
<td>profit tax rate</td>
</tr>
</tbody>
</table>

The data input to the model are listed in Table 2.2. The electricity generation efficiency of BIGCC or CHP is calculated from the corresponding model. For a TMP+PM plant, its average overall capital investment is about 1920 EUR/tpy (in paper production capacity) [6], in which, generally, to invest in a TMP+PM plant, at an existing industrial site, demands about 338 EUR/tpy. The overall unit capital investment for BIGCC is 2300 EUR/KWel [59]. The capacity of power generation is about 30 MWel.
The prices for some facilities are referred to [3, 60]. The operation and maintenance cost accounts for 7.0% of the capital investment of a combined TMP+BIGCC plant. Electricity is sold at about 50 EUR/MWh. An electricity certificate of 30 EUR/MWh is considered. The price of pulpwood is in the range of 15–25 EUR/MWh. The discounted payback period is set at 10 years. Available raw biomass materials from a TMP plant are free of charge. The overall cost for the logging residues is 10 EUR/MWh. The heat is sold at 80 EUR/MWh for DH. The plant lifespan is anticipated to be 30 years. In sensitivity analysis, the pulpwood is priced at 20 or 40 EUR/MWh, and the electricity at 50 or 80 EUR/MWh. The other conditions are kept unchanged.

Two depreciation methods are adopted: for the first 10 years the Double-Declining Balance method (DDB), and for the next years the Straight-line Depreciation Method. Tax is not added. Table 2.2 lists the data on this economic evaluation. The economic analysis routine is programmed with Excel.

2.4 Biomass to Ethanol

2.4.1 Process

In the gasification–synthesis route, biomass is gasified into raw syngas which is further reformed, cleaned, compressed, heated and converted into mixed alcohols from which ethanol and higher alcohols are obtained through a series of separation and purification treatments. Advantages of this route include a short reaction time, inexpensive and abundant raw materials, high conversion rate, etc [43, 44].

The process consists of five steps: biomass pretreatment, gasification, syngas cleaning and conditioning, alcohols synthesis, and separation. Fresh biomass needs pretreatment. Weighed wood chips are screened. Metal debris is removed using a magnetic separator. Particles larger than 10 mm are conveyed to a hammer mill for further size reduction. By direct contact with the hot flue gas, wet biomass is dried. Gasification is carried out in DFBG. Particles from raw syngas are separated. A small amount of MgO is added to raise the ash melting point and to avoid the agglomeration that would result from the interaction of biomass potassium with silicate compounds. The composition and quality of syngas from the biomass gasifier are dependent on a number of factors. Syngas with \( \text{H}_2/\text{CO} \) molar ratio of 1–2 is suitable for ethanol synthesis. Raw syngas contains various impurities which are potential threats to ethanol synthesis catalysts. Tars are cracked. Additional CO and \( \text{H}_2 \) are formed. Additional cooling is done via water scrubbing, and impurities such as particulates, residual tars, and ammonia are further reduced. Subsequently, in a five-stage compressor, the syngas is compressed to 29 bar. \( \text{NH}_3 \) and \( \text{H}_2\text{O} \) are condensed. Afterwards, acid gases (\( \text{CO}_2 \) and \( \text{H}_2\text{S} \)) removal is carried out in an amine system. Other sulfurous compounds—if contained in raw syngas, will be converted into \( \text{H}_2\text{S} \). The presence of some \( \text{CO}_2 \) (about 5 vol.%) is necessary for the function of a methanol synthesis catalyst, and may also be advantageous for ethanol synthesis. After cleaning, syngas is compressed and pre-heated to the synthesis conditions of 100 bar and 300 °C. Synthesis is completed in seconds or minutes, and the product is cooled and initially purified to separate out the unconverted syngas. Mixed alcohols
are de-gassed, dried, and separated into ethanol and higher alcohols. In a flash separator, alcohols are separated from off-gas, and off-gas is diverted to a gas cleaning section. Since the recycled gas is at high pressure, it is further heated and applied to generate extra electricity. The depressurized alcohols stream is dehydrated with a molecular sieve and then sent to the main distillation column. The overhead is further processed in the second column to get high-purity ethanol. The obtained methanol/water mixture is recycled for alcohols synthesis. The biomass-to-ethanol process is designed to be energy self-sufficient.

2.4.2 Simulation

Before setting up the model, an optimal thermo-chemical biomass-to-ethanol process is designed (Fig. 2.6). In the process, off-gas and MeOH from ethanol synthesis and separation steps are recycled to increase conversion efficiency. A CHP plant is integrated from economic and environmental points of view.

![Figure 2.6: Block flow diagram of a biomass-to-ethanol process](image)

The cost of equipment can be estimated by:

\[ I_1 = I_2 \times \left( \frac{Q_1}{Q_2} \right)^n \]  
\[ I_1 = I_2 \times \frac{C_1}{C_2} \]  

(I—investment; Q—production capacity or equipment size; CI—cost year index)

Here, “n” denotes the characteristic scaling exponent of either the overall plant or a single piece of equipment. Typically, “n” equals 0.6, that is, “0.6 exponent method”. “n” has different referential values for different processes and types of equipment. Equipment installation cost is estimated to be 1.45 times equipment purchase cost. Operation cost consists of fixed and variable operation costs. Labour cost equals national per capita income multiplied by a factor of 1.2. Variable operation cost is calculated according to the stream flow rate. Price of cost item equals the temporal market price at the time when economic analysis is performed. Biomass procurement cost is set as 60 €/dry tonne. The fixed operation cost is supposed to stay invariable within a certain range, while variable operation cost is varied with the
production scales. Since capital recovery charge is related to capital investment, “0.6 exponent method” is adopted to calculate the charge. Once total capital and operation costs are determined, ethanol production cost is calculated by “discounted cash flow analysis” when the NPV is zero.
Chapter 3

Result and Discussion

3.1 Gasification

A mathematic model based on ASPEN Plus is established to simulate biomass gasification in DFBGs.

As seen in Fig. 3.1(a), the syngas yield increases with temperature as a result of the enhanced gasification and reforming at a higher temperature. Tar content decreases from 50.0 g/m$^3$ at 750 °C down to about 13.0 g/m$^3$ at 900 °C attributed to tar thermal cracking and steam reforming. Char to be combusted is regarded as an intermediate product. The char fraction decreases from 22.5 wt.% at gasification temperature of 750 °C to 11.5 wt. % at 950 °C. The lower gasification temperature leads to higher char yield, and subsequently results in higher combustor temperature, hotter bed
material, and higher gasification temperature, so that less char will be produced.

As seen in Fig. 3.1(b), the concentration of CO increases with temperature, but that of H$_2$ reaches a maximum value at about 850 °C. This is because H$_2$ tends to take part in the hydrogenation reactions at higher temperatures. The H$_2$ concentration is higher than that of CO under normal DFBG operating conditions, but they will change positions when the gasification temperature is too high above about 950 °C. The CH$_4$ concentration is around 10.0 vol. % and not sensitive to temperature. CH$_4$ is highly stable, and much more difficult to be cracked or reformed than tar. Therefore, CH$_4$ concentration is far from its chemical equilibrium value which is close to zero. Significant reduction of CH$_4$ takes place at a temperature higher than 1250 °C or with the help of catalysts (e.g., nickel-based catalysts).

Figure 3.2: Effect of S/B on the gasification (experiment—MIUN gasifier, wood pellets, sand bed material, 850 °C, $\tau=5.0$ s, $d_p=5.0$ mm)

S/B values were calculated with an assumption that the dry biomass has 5.0 wt. % moisture. The tar content decreases from 50.0 g/m$^3$ at S/B = 0.3 down to about 10.0 g/m$^3$ at S/B = 1.2 attributed to the enhanced steam reforming of tar. The char fraction is insensitive to S/B as shown in Fig. 3.2(a). The syngas yield is in the range of 0.8–1.0 Nm$^3$/kg daf biomass, and the LHV is about 11.65 MJ/Nm$^3$. Both calculated data are fairly close to the measurement results from a pilot DFBG plant at Vienna University of Technology [47].

The H$_2$/CO ratio is a strong function of the S/B ratio, which is primarily controlled by the water–gas shift reaction. The H$_2$ concentration is higher than that of CO under normal DFBG operating conditions, but they will change positions when the S/B ratio is too far below about 0.15 as seen in Fig. 3.2(b). The CH$_4$ concentration is around 10.0 vol. % and not sensitive to the S/B ratio.

For an autothermal gasifier, gasification temperature can be held by adjusting the oxygen equivalent ratio. For a DFBG, extra fuel needs to be added in the combustor.
3.1 Gasification

Fig. 3.3 shows that the gasification temperature is difficult to be kept above 850 °C when the biomass moisture content is higher than 15.0 wt. %. Gasification temperature can be raised about 50 to 100 °C when the combusted biomass is 10.0 wt. % of the total biomass feedstock, as seen in Fig. 3.3.

In Fig. 3.4, the exact amount of the combusted biomass can be found out corresponding to different biomass moisture content when the gasification temperature is held at 850 °C in three cases of S/B = 0.3, 0.6, and 0.9, respectively.

The tar content in the syngas decreases significantly with gasification residence time which is a key parameter (Fig. 3.5). But, char content is insensitive to the residence time. The syngas yield is therefore increased.
3.2 TMP + BIGCC

Figure 3.6: TMP energy consumption and supply. (Three processes (BIGCC, Boiler, and CHP) are modelled respectively, and compared. The model is designed to ensure that the steam demand is just satisfied, as the results show in the graph. The results demonstrate that the BIGCC process is the most efficient of the processes studied, and therefore, by implementing the BIGCC system in a TMP mill, less electricity will need to be purchased, and even less, with the TMP SEC decreased.)

The energy balance over the BIGCC (or Boiler or CHP) process is studied by applying the model developed in this study. Fig. 2.4 shows how much biomass can be available. The steam consumed in the PM process will be obtained from the TMP and BIGCC (or Boiler or CHP) processes. The model is designed to ensure that the steam demand is just satisfied, as seen in the results (Fig. 3.6). For the TMP+PM mill studied, the TMP SEC is 2.5 MWh/bdt pulp, the PM SEC is 0.75 MWh/bdt pulp, and the steam consumption in PM is 1.38 MWh/bdt pulp. Compared with CHP, BIGCC produces much more electricity because the BIGCC system has a higher electricity generation efficiency of 45%. As a result, by implementing the BIGCC system in a TMP mill, less electricity will need to be purchased, and even less with the TMP SEC decreased (Fig. 3.6). The electricity generation efficiency of a full-scale BIGCC plant can approach 50%. The BIGCC plant associated with the TMP mill needs to supply steam to PM, which lowers the electricity generation efficiency. The residual low-grade energy from the BIGCC (or Boiler or CHP) process is utilized in the DH system (Table 2.1).

Independent biomass heat and power plants scaled up to 100 MW (in biomass energy input) have been popular in Sweden as well as other forest-rich countries. Forest logging residues, peat, biomass obtained from short rotation forestry, and industrial and household wastes, etc. are all available bioenergy resources that can be taken as feedstock to BIGCC plants. The electricity consumed in a TMP mill can be fully provided by a BIGCC plant without difficulty concerning biomass availability. This is evaluated based on the model. The calculation results are shown in Fig. 3.7.
3.2 TMP + BIGCC

Figure 3.7: Effect of the amount of biomass fed into the gasifier or boiler. (The electricity production from the BIGCC plant increases with the amount of biomass used, and reaches the demand of the TMP+PM mill when about 10.6 MWh biomass per tonne of pulp is used as feedstock to the gasifier (Fig. 3.7(a)). BIGCC produces much more electricity and much less heat than CHP (Figs. 3.7(a) and 3.7(b)). Both the net revenue and the IRR increase significantly with the amount of biomass feedstock, and they increase faster as the electricity price increases. The pulpwood price is more sensitive than the electricity price to the economic profitability of a TMP mill (Figs. 3.7(c) and 3.7(d)). The electricity certificate is an effective incitement (Figs. 3.7(c) and 3.7(d)). It will be more profitable to invest in a BIGCC plant than in a CHP plant if more biomass materials are used for heat and power production (Figs. 3.7(c) and 3.7(d)).)
Figure 3.8: Effect of feeding reject fibres into gasifier. (BIGCC produces twice the electricity that CHP does, but is still far from the TMP+PM demand (Fig. 3.8(a)). Both BIGCC and CHP produce much more steam than TMP+PM demand (Fig. 3.8(b)). Both the net revenue and IRR increase with the amount of reject fibres (Figs. 3.8(c) and 3.8(d)). Both the net revenue and the IRR values of TMP+BIGCC are much higher than those of TMP+CHP, and the gap becomes bigger when more reject fibres are used for heat and power production (Figs. 3.8(c) and 3.8(d)). The economic profitability of a TMP mill is dominated by the pulpwood price and/or the electricity price (Figs. 3.8(c) and 3.8(d)).)
As seen by the thick solid line in Fig. 3.7(a), electricity production from the BIGCC plant increases with the amount of biomass used, and it reaches the demand of the TMP+PM mill when about 10.6 MWh biomass per tonne of pulp is used as feedstock to the gasifier. However, a great amount of heat is also generated along with the electricity production. This huge supply of low-grade energy (steam and heat) must be directed to other uses such as DH. As can be clearly seen in the figure, BIGCC is superior to CHP, which produces much more electricity and much less heat than CHP.

Figure 3.9: Effect of reducing the TMP-specific electricity consumption. (The TMP+PM electricity consumption cannot be fully made up by the electricity produced from the BIGCC plant, even if the TMP SEC goes down by 50%. But the gap becomes fairly small, and much smaller in comparison with the case of a CHP plant (Fig. 3.9(a)). With the decreasing of the TMP SEC, less steam can be supplied from the TMP process. Thus, the steam supply to the TMP+PM mill needs to be compensated for by the BIGCC (or CHP) plant (Fig. 3.9(b)). Lower TMP SEC leads to higher economic profit (Figs. 3.9(c) and 3.9(d)). For both the net revenue and the IRR, TMP+BIGCC > TMP+CHP > TMP+Boiler (Figs. 3.9(c) and 3.9(d)). The economic profitability of a TMP mill is dominated by the pulpwood price and/or the electricity price (Figs. 3.9(c) and 3.9(d)).)
Figs. 3.7(c) and 3.7(d) show that both the net revenue and the IRR increase significantly with the amount of the biomass feedstock, and increase faster as the electricity price gets higher. The pulpwood price is more sensitive than the electricity price to the economic profitability of a TMP mill (Figs. 3.7(c) and 3.7(d)). The electricity certificate is an effective incitement (Figs. 3.7(c) and 3.7(d)). It is more profitable to invest in BIGCC than in CHP when more biomass is used for heat and power production. Plant scale and electricity are two strong factors dominating the implementation of BIGCC in a TMP mill.

Usually in TMP mills, up to about 10% of the pulp fibres have properties that are not good enough to remain in the final product, and they may be separated and removed as reject fibres. The reject fibres can be utilized as feedstock to biomass gasifiers. For instance, by taking away 10% of the fibres as reject fibres, the TMP SEC will be decreased. 20% TMP SEC reduction should be possible when taking out the 10% worst fibre material with respect to final paper product properties [61]. Fig. 3.7 shows the effects on the energy balance and economic profitability when the reject fibres are fed into the gasifier.

As seen in Fig. 3.8(a), the electricity yield from BIGCC increases slightly with the amount of reject fibres, but is far from the electricity demand of the TMP+PM mill suggested by the dotted line. Electricity yield of BIGCC has been more than double that of CHP. BIGCC is more efficient than CHP.

The vertical coordinator in Fig. 3.8(b) represents the steam and heat produced, including steam supplied by BIGCC to TMP+PM, and heat supplied to a district heating system. The amount of steam and heat increases as the amount of reject fibres transferred to the gasifier increases. Lower TMP SEC will lead to less steam supply from the TMP process. As a result, the heat and power plant needs to supply more steam. Obviously, CHP generates more steam and heat than BIGCC as much less electricity is generated by CHP.

Normally, the prices of electricity and pulpwood are 50 and 20 EUR per MWh respectively, as indicated by the thick solid line in Figs. 3.8(c) and 3.8(d).

Both the net revenue and the IRR values increase with the amount of the reject fibres. This can be explained by several factors: 1) more electricity is generated as more reject fibres are added; 2) the TMP SEC is decreased; 3) the paper yield is held constant in the calculation while more pulpwood is used. Both the net revenue and the IRR values of TMP+BIGCC are much higher than those of TMP+CHP, as shown in Figs. 3.8(c) and 3.8(d). This is because BIGCC produces much more electricity than CHP for the same amount of biomass residues, and has a lower specific investment cost than CHP (see Table 2.2). An increase in the market electricity price from 50 to 80 EUR/MWh obviously reduces the net revenue, as shown by the dash-dot line in Fig. 3.8(c). The pulpwood price is more sensitive than the electricity price to the economic profitability (Figs. 3.8(c) and 3.8(d)).

As discussed above, electricity consumption in TMP mill is a key factor in the high cost of pulp production. Reduction of TMP SEC is an attractive research area. For a future TMP mill, the SEC is expected to be decreased by 50%. Under this condition, a techno-economic analysis is done for a case in which a BIGCC plant is integrated into a TMP mill.
3.2 TMP + BIGCC

The reduction of the TMP SEC is indicated in Fig. 3.9(a) by the dotted line. The TMP+PM electricity consumption cannot be fully made up by the electricity produced from the BIGCC plant, even if the TMP SEC goes down by 50%. But the gap becomes fairly small, and much smaller in comparison with the case of a CHP plant.

Decreasing the TMP SEC decreases the supply of steam from the TMP process. Thus, the steam supply to the TMP+PM mill needs to be compensated for by the BIGCC (or CHP) plant, as shown by the dotted line in Fig. 3.9(b). Since a certain amount of 2.5 bar steam is extracted from the BIGCC system, the electricity generation efficiency of the steam turbine cycle will be lowered, which gives rise to a slightly lower electricity yield from the BIGCC plant as seen in Fig. 3.9(a). A clear increase in the steam and heat from BIGCC (or CHP) with the reduction of the TMP SEC can be seen in Fig. 3.9(b), which is attributed to the lower electricity yield and smaller amount of waste heat from the BIGCC (or CHP) plant.

In some TMP+PM mills, a backup boiler is used to supply steam to meet the steam demand of PM without self-production of electricity. In the TMP+Boiler case, only a proper amount of biomass will be fed to produce a certain amount of steam for PM. Such a TMP+Boiler case is shown by the inset figure in Fig. 3.9(b). As observed in the inset figure, the boiler needs more biomass residues as the TMP SEC decreases, and the internal biomass residues within the TMP mill will no longer be enough when the TMP SEC decreases by more than 23%.

As shown in Figs. 3.9(c) and 3.9(d), lower TMP SEC leads to higher economic profits in both the net revenue and the IRR. For both the net revenue and the IRR, TMP+BIGCC > TMP+CHP > TMP+Boiler. The TMP+BIGCC is a much more profitable combination. This is mainly attributed to the lower specific investment cost in the BIGCC plant. The investment of BIGCC is expected to be further decreased with more research and development towards commercialization of BIGCC technology. So far, BIGCC technology has not been fully commercialized. There exists a risk when BIGCC is applied to industries, including the pulp and paper industry.
3.3 Biomass to Ethanol

A mathematic model for techno-economic assessment of the thermo-chemical biomass-to-ethanol process has been developed mainly with ASPEN Plus. The assessment is made in terms of carbon and CO conversion efficiencies, ethanol yield, synthesis selectivity and ethanol production cost.

As shown in Fig. 3.10, obviously the production cost decreases with the plant size due to the economic scaling effect. In particular, with small scales up to 200 MW, the production cost decreases rapidly and levels off. This suggests that a thermo-chemical biomass-to-ethanol plant should be built at around 200 MW. The production cost at this scale is around 0.33 €/l ethanol, which might be reduced further to 0.25 €/l ethanol in the scale of 1000 MW. However, a 1000 MW ethanol plant based on biomass is not realistic due to limited feedstock availability. The biomass price is held constant in the present model calculation, but it will also rise dramatically for the large plant and leads to a high operation cost.

The economic scaling effect is mainly attributed to the capital investment. For a small ethanol plant, the capital investment is much higher than the operation cost, as shown in Fig. 3.10. The gap between the capital investment and the operation cost is becoming smaller and smaller as the plant is becoming larger and larger, and the process equipment is utilized more efficiently. The first huge capital investment plus the risk in the thermo-chemical conversion technologies is the biggest barrier to BTL commercialization.

The capital investment is broken down by the costs of individual processes, pretreatment, gasification, syngas cleaning, alcohols synthesis, alcohols separation and purification, power generation, and the utilities as shown in Fig. 3.11. The biggest share of total capital investment is allocated to the syngas cleaning including tars/CH₄ reforming by about 38%, followed by about 16% of biomass pretreatment and about 15% of alcohols synthesis. The capital cost distribution is not sensitive to the ethanol
3.3 Biomass to Ethanol

plant size.

![Pie chart showing cost breakdown]

**Figure 3.11: Capital cost breakdown**

The huge cost of syngas cleaning in response to a limited size of ethanol plant is the most important factor limiting commercial production of ethanol from biomass via a thermo-chemical conversion. This is a big drawback compared with biochemical conversion.

The total production cost is broken down as shown in Fig. 3.12. The costs of biomass raw material and syngas cleaning are the two biggest shares of the total production cost. As the ethanol plant is becoming larger, the raw material cost grows to make...
up a larger share of the total production cost, while the syngas cleaning and other capital investment cost shares get smaller. On the other hand, the mixture of higher alcohols is also produced as a valuable byproduct to ethanol. The higher alcohols credit exerts a negative effect on the ethanol production cost, and compensates for the high cost of biomass raw material. This cost compensation by the higher alcohols credit is much clearer when the plant size is bigger.

The degree of compensation depends on higher alcohols’ end-use. For a low-value use as an excellent gasoline additive, higher alcohols would have a selling price similar to that of fossil fuels (about 0.16 € /l) [60]. For a high-value use as chemicals, higher alcohols would have a selling price of about 0.81 € /l [60]. Fig. 3.13 shows the ethanol production cost against the higher alcohols’ selling price. It can be seen clearly that the production cost is reduced from 0.31 € /l when higher alcohols are used as a gasoline additive to about 0.19 € /l when higher alcohols are used as chemicals.

In summary, cost-competitive ethanol production can be realized with efficient equipment, optimized operation, cost-effective syngas cleaning technology, inexpensive raw material with low pretreatment cost, high-performance catalysts, off-gas and methanol recycling, optimal systematic configuration and heat integration, and a high value byproduct.
Chapter 4

Conclusions

4.1 Gasification

A mathematic model based on ASPEN Plus is established to simulate biomass gasification in the DFBGs. The model predicts that the content of char transferred from the gasifier to the combustor decreases from 22.5 wt.% of the dry and ash-free biomass at gasification temperature 750 °C to 11.5 wt.% at 950 °C, but is insensitive to S/B. The H2 concentration is higher than that of CO under the normal DFBG operation conditions, but they will change positions when the gasification temperature is too high above about 950 °C, or the S/B ratio is too low under about 0.15. The biomass moisture content is a key parameter for a DFBG to be operated and maintained at a high gasification temperature. The model suggests that the gasification temperature is difficult to be kept above 850 °C when the biomass moisture content is higher than 15.0 wt.%. Thus, a certain amount of biomass needs to be added in the combustor to provide sufficient heat for biomass devolatilization and steam reforming. Tar content in the syngas can also be predicted from the model, which shows a decreasing trend of the tar with the gasification temperature and the S/B ratio. The tar content in the syngas decreases significantly with gasification residence time which is a key parameter.

4.2 TMP + BIGCC

A TMP+BIGCC mathematic model is developed using ASPEN Plus. Based on the model, the energy balance and the techno-economic analysis of the TMP+BIGCC mill are done in comparison with other combinations of TMP+CHP and TMP+Boiler. The conclusions based on the calculation results are described below.

Electricity yield of BIGCC has been more than double that of CHP. BIGCC has a lower specific capital investment than CHP, which results in a shorter discount payback period and higher IRR. It is more profitable to invest in BIGCC than in CHP when more biomass is used for heat and power production and when the electricity price
is higher. The pulpwood price is more sensitive than the electricity price to the economic profitability of a TMP mill. Plant scale and electricity are two strong factors dominating the implementation of BIGCC in a TMP mill.

Both the net revenue and the IRR increase with the amount of the reject fibres since more electricity is self-produced on-site, the TMP SEC is reduced, and the paper output is held constant while the pulpwood is increased in the calculation. Rejection of low-quality pulp fibres and their reuse in BIGCC for self-production of electricity, and electricity certificate are highly attractive measures to raise the profitability of a TMP mill.

The TMP+PM electricity consumption cannot be fully made up by the electricity produced from the BIGCC plant, even if the TMP SEC goes down by 50%. But the gap becomes fairly small, and much smaller in comparison with the case of a CHP plant. Lower TMP SEC leads to higher economic profits in both the net revenue and the IRR. The profits are ranked as TMP+BIGCC > TMP+CHP > TMP+Boiler.

### 4.3 Biomass to Ethanol

A mathematic model for a techno-economic assessment of the thermo-chemical biomass-to-ethanol process has been developed mainly with ASPEN Plus. The assessment is made in terms of carbon and CO conversion efficiencies, ethanol yield, synthesis selectivity, and ethanol production cost.

Calculated results show that major contributions to the production cost are from syngas cleaning and biomass feedstock. A biomass-to-ethanol plant should be built at around 200 MW as the biomass input, which gives the ethanol production cost of 0.33 €/l in the context of a typical developed country. The capital investment is much higher than the operation cost and dominates the total ethanol production cost for small- and medium scale plants. This suggests that the first huge capital investment with technological risk is a big barrier to the commercialization of a thermo-chemical biomass-to-ethanol process. The costs of biomass raw material and syngas cleaning are the two biggest shares of the total production cost. As the ethanol plant becomes larger, the raw material cost makes up a larger share of the total production cost. This can be off-set to a certain extent by the higher alcohols credit, which might reduce the ethanol production cost from 0.31 €/l to about 0.19 €/l when the higher alcohols are used as valuable chemicals.
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Appendix A
Calculation Details

TMP energy consumption and supply
unit: [MWh/bdt pulp]

“Consumption” – energy of steam used in TMP and PM
♦ Original data from (Pettersson, 2005)

“Consumption” – energy of electricity used in TMP
♦ Original data from (Pettersson, 2005)

“TMP+BIGCC” – energy of steam supplied from TMP to PM
♦ [MWh/mo.] / [bdt pulp/mo.] = [MWh/bdt pulp] (4604 + 22906) / 19938 ≈ 1.4

“TMP+BIGCC” – energy of electricity used in PM
♦ [MWh/mo.] / [bdt pulp/mo.] = [MWh/bdt pulp] 22906 / 19938 = 1.1

“TMP+BIGCC” – energy of steam produced by BIGCC
♦ 1,37978 - 1,14886 ≈ 0.2

“TMP+BIGCC” – energy of electricity produced by BIGCC (re. Appendix B)
♦ 1,1

“TMP+BIGCC” – energy of electricity purchased
♦ 3,2628 - 1,0598 ≈ 2.2

“TMP+BIGCC” – energy of heat supplied to DH
♦ 0.9

Available biomass; data adapted from (Pettersson, 2005)
<table>
<thead>
<tr>
<th></th>
<th>MWh/mo.</th>
<th>MW</th>
</tr>
</thead>
<tbody>
<tr>
<td>100 bar steam needed</td>
<td>1,148,6</td>
<td>1,6</td>
</tr>
<tr>
<td>2,5 bar steam needed</td>
<td>437,8</td>
<td>6,0</td>
</tr>
<tr>
<td>sum</td>
<td>1,586,4</td>
<td>7,6</td>
</tr>
</tbody>
</table>

\[
\text{energy of heat supplied to DH} = \frac{(3,08144 - 1,0598 - 0,2915) \times 0,5}{0,9}
\]

\[\approx 0,9\]

**Biomass needed**

\[
\text{Biomass needed} = \left(\frac{\text{steam needed [MW]}}{\text{biomass combustion efficiency [%] / 100} \times \text{boiler efficiency [%] / 100}}\right) \times \text{[h/mo.] / [bdt pulp/mo.]} = \text{[MWh/bdt pulp]}
\]

\[
(29716,4 + 4358,4 + 106130,1) / 12 \times 1000 \times 18 / 3600 / (227498,9 / 12) = 3,1
\]

**TMP+BIGCC** – heat loss

\[3,08144 - 1,0598 - 0,2915 = 0,9\]

**TMP+Boiler** – energy of steam supplied from TMP

\[1,1 + \text{energy of steam produced by boiler}\]

\[\approx 0,2\]

**TMP+Boiler** – energy of electricity purchased

\[\approx 3,3\]

**TMP+Boiler** – energy of heat supplied to DH

\[\approx 0,05\]

**TMP+CHP** – energy of steam supplied from TMP

\[1,1 + \text{energy of steam produced by CHP}\]

\[\approx 0,2\]

**TMP+CHP** – energy of electricity produced by CHP (re. Appendix C)

\[\approx 0,5\]

**TMP+CHP** – energy of electricity purchased

\[3,2628 - 0,4742 = 2,8\]

**TMP+CHP** – energy of heat supplied to DH

\[(3,08144 - 0,4742 - 0,2915) \times 0,5 = 1,2\]

**TMP+CHP** – heat loss

\[3,08144 - 0,4742 - 0,2915 - 1,15789 = 1,2\]

**Biomass** – energy content of logging residues

\[2,3\]

**Biomass** – energy content of bark

\[0,7\]

**Biomass** – energy content of bio-sludge

\[0,1\]

**Biomass** – energy content of reject fibers

\[0,5\]

Appendix B
Model BIGCC

Flowsheet and Blocks (Figure below)

Software: ASPEN Plus

Property Method

gas turbine cycle          steam turbine cycle

PR-BM                      STEAM-TA

Gas turbine system

Compressor (air compression)

<table>
<thead>
<tr>
<th>Component</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>N2</td>
<td>0.0002207</td>
</tr>
<tr>
<td>CO2</td>
<td>0.7866666</td>
</tr>
<tr>
<td>CH4</td>
<td>0.0010094</td>
</tr>
<tr>
<td>Ar</td>
<td>0.0293954</td>
</tr>
<tr>
<td>C2H6</td>
<td>0.0000000</td>
</tr>
</tbody>
</table>

Combustor (combustion of syngas and air)

<table>
<thead>
<tr>
<th>syngas, 4.85 kg/s (input data)</th>
</tr>
</thead>
<tbody>
<tr>
<td>wood+bark</td>
</tr>
<tr>
<td>bark</td>
</tr>
<tr>
<td>bio-sludge</td>
</tr>
<tr>
<td>reject fibers</td>
</tr>
<tr>
<td>logging residues</td>
</tr>
<tr>
<td>[t/yr]</td>
</tr>
<tr>
<td>247636.9</td>
</tr>
<tr>
<td>29716.4</td>
</tr>
<tr>
<td>217920.5</td>
</tr>
<tr>
<td>4358.4</td>
</tr>
<tr>
<td>21356.2</td>
</tr>
<tr>
<td>192305.9</td>
</tr>
<tr>
<td>106130.1</td>
</tr>
</tbody>
</table>

Turbine (power generation)

<table>
<thead>
<tr>
<th>Wood-bark</th>
<th>bark</th>
<th>bio-sludge</th>
<th>reject fibers</th>
<th>logging residues</th>
</tr>
</thead>
<tbody>
<tr>
<td>247636.9</td>
<td>29716.4</td>
<td>217920.5</td>
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</tr>
<tr>
<td>106130.1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

[\text{kg/t} \times \text{s/h}] = \text{gasification efficiency} \% / \text{syngas MJ/kg} = \frac{\text{syngas} \times \text{MJ/kg}}{\text{syngas MJ/kg} / \text{t/mo.}} \times \frac{3600 \text{kJ}}{1000 \text{J}} \times \frac{1}{730} \times 18 \times 0.80 \approx 4.85
Electricity from the gas turbine system

\[ W = (\text{ABS}(W_{\text{TG}}) \times 0.96 - \text{ABS}(W_{\text{CG}})) - \text{ABS}(W_{\text{PS}}) \]

Where,

- \( W \) — electricity from the gas turbine system [W]
- \( W_{\text{TG}}, W_{\text{CG}}, \) and \( W_{\text{PS}} \) — output data from the blocks (Turbine, Compressor, and Pump), respectively

\[ W \approx 16446750.7 \text{ W} \]

**Pump** (pumping the feed water)

- Pump
- **Discharge pressure**: 10.0 bar

**Heat Exchanger** (steam production)

- **Temperature**: C 0.6
- **Pressure bar**: 10.0
- **Vapor Frac**: 0.000
- **Mass Flow kg/hr**: 2393.271
- **Mass Flow kg/hr**: 4234.915

**Steam turbine system**

**FSplit 1** (100 bar steam internally used)

- **Temperature**: C 170.7
- **Pressure bar**: 10.0
- **Vapor Frac**: 0.000
- **Mass Flow kg/hr**: 6156.444
- **Mass Flow kg/hr**: 77867.941

Steam 11.7 kg/s (input data)

\[ \approx 42124.615 / 3600 \]

**FSplit 2** (2.5 bar steam used in PM)

<table>
<thead>
<tr>
<th>Steam</th>
<th>Specification</th>
<th>Basis</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$25\text{BAAR}$</td>
<td>Split fraction</td>
<td>$25\text{BAAR}$</td>
<td>$100%$</td>
</tr>
</tbody>
</table>

- **Temperature**: C 50.0
- **Pressure bar**: 100.0
- **Vapor Frac**: 1.000
- **Mass Flow kg/hr**: 2249.387
- **Mass Flow kg/hr**: 4363.96

**Turbine 1**

**Turbine 2**

Electricity from the steam turbine system

\[ MW = (\text{abs}(W_1) + \text{abs}(W_2))/1000/1000 \]

Where,

- \( MW \) — electricity from the steam turbine system [MW]
- \( W_1, W_2 \) — output data from the blocks (Turbine 1 and Turbine 2), resp.

\[ MW \approx 11.08 \text{ MW} \]

Gasification efficiency

- el. from BIGCC [W] 27523599.9
- QIN [W] 61619535
- generation efficiency [%] 44.7

Electricity from the BIGCC system [W]

\[ W \approx 16446750.7 + 11076849.2 \]

QIN (absorbed in by the WM)

<table>
<thead>
<tr>
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<th>Specification</th>
<th>Basis</th>
<th>Value</th>
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<tbody>
<tr>
<td>$25\text{BAAR}$</td>
<td>Split fraction</td>
<td>$25\text{BAAR}$</td>
<td>$100%$</td>
</tr>
</tbody>
</table>

- **Temperature**: C 298.0
- **Pressure bar**: 106.0

GE = $W / \text{QIN} \times 100\%$
Appendix C
Model CHP

Flowsheet and Blocks (Figure below)

Pump (pumping the feed water)

steam produced from the boiler

\[ \frac{[MJ/kg] \times [t/mo.] \times [kg/t] / [h/mo.] \times [s/h]}{100 \times \text{boiler efficiency} \%} \times 10^{-6} \times \text{boiler efficiency} \% / 100 = [MW] \]

\[ 18 \times (29716,4 + 4358,4 + 106130,1) / 12 \times 1000 / (730 \times 3600) \times 99 / 100 \times 75 / 100 \approx 59,4 \]

59045,3728 / 3600 \approx 16,4 kg/s steam can be produced with the energy 59,4 MW

Boiler (steam production)

<table>
<thead>
<tr>
<th>Steam</th>
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<tbody>
<tr>
<td>100BAR</td>
<td>Split fraction</td>
<td>0.02422039</td>
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FSplit 1 (100 bar steam internally used)

<table>
<thead>
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<th>Stream</th>
<th>Specification</th>
<th>Basis</th>
<th>Value</th>
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</thead>
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<tr>
<td>100BAR</td>
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FSplit 2 (2.5 bar steam used in PM)

<table>
<thead>
<tr>
<th>Stream</th>
<th>Specification</th>
<th>Basis</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>25BAR</td>
<td>Split fraction</td>
<td>0.12512489</td>
<td></td>
</tr>
</tbody>
</table>

Turbine 1

<table>
<thead>
<tr>
<th>Duty specification</th>
<th>Discharge pressure</th>
<th>25 bar</th>
</tr>
</thead>
</table>

Turbine 2

<table>
<thead>
<tr>
<th>Duty specification</th>
<th>Discharge pressure</th>
<th>0 bar</th>
</tr>
</thead>
</table>

Gasification efficiency

\[ W = (\text{ABS}(W1) + \text{ABS}(W2)) \times 0.9 - \text{ABS}(W_{PS}) \]

\[ GE = W / (\text{ABS}(QIN) \times 0.9) \times 100\% \]

\[ GE \approx (120582,0 \times 12602535 \times 0.9 - 113705,702) / (52092858,7 \times 0.9) \times 100\% \approx 26\% \]

100BARPM=steam, expanded from 100 bar to 2.5 bar, and then is used in PM
100BART=100 bar steam to the turbine
CAIR=compressed air
Cl, CO, HI, HO=cold/hot inlet, outlet, resp.
FW=feed water
GE=generation efficiency
QIN=heat absorbed
WM=working medium
WTG=work, turbine, gas turbine system
WCG=work, compressor, gas turbine system
WPS=work, pump, steam turbine system