# Process Integration Opportunities for Synthetic Natural Gas (SNG) Production by Thermal Gasification of Biomass

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Göteborg, Sweden 2010

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# ABSTRACT

Synthetic natural gas (SNG) from gasified biomass is one promising option to produce renewable transport fuels. This thesis presents a process integration study investigating thermal gasification of biomass for the production of SNG and identifies critical conversion steps for the overall process performance. A base case process consisting of an indirect gasification unit followed by tar reforming, amine-based $CO_2$ separation, isothermal methanation and, finally, compression, $H_2$-purification by membrane separation and gas drying is presented. Based on the lower heating value (LHV) of the wet fuel feedstock, the estimated conversion efficiency from biomass to SNG is 69.4 %. The process mass and energy balances are obtained by using flow-sheeting software and are analysed by using pinch methodology.

The integration studies performed highlight the significant potential for improvement of the overall process performance offered by integrated feedstock drying. In particular, steam drying and low-temperature air drying – using available process excess heat – are shown to influence the process performance favourably.

The integration of SNG production with existing combined heat and power (CHP) steam power plants is proven to be a promising option to efficiently convert excess heat of the SNG process to electricity. The process integration study performed shows that an increased level of thermal integration leads to an increase in electricity production attributed to the SNG process (100 $MW_{LHV}$ dry fuel input) from 2 to 4.9 MW when using steam drying for feedstock drying, and from 0.5 to 5.6 MW for air drying, without any negative effects on SNG yield.

Alternative integration opportunities for biomass gasification not aiming at SNG production specifically, but at replacing fossil fuels for power production, are also highlighted. Biomass gasification integrated to a fossil natural gas combined cycle plant results in high biomass-specific electrical efficiencies of up to 49.6 %.

**Keywords:** process integration, synthetic natural gas, renewable energy, biomass, energy systems

# List of Publications

This thesis is based on the following papers.

I. Heyne, S., Harvey S. (2009). Methane from biomass: process-integration aspects. Proceedings of the Institution of Civil Engineers – Energy, 162 (EN1), pp. 13-22.

II. Heyne, S., Harvey S. (2009). Production of Synthetic Natural Gas from biomass – process integrated drying. Proceedings of the 22$^{nd}$ International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems (ECOS), Foz do Iguaçu/Brazil, pp. 1877-1886.

III. Heyne, S., Thunman, H., Harvey, S. (2010). Extending existing CHP plants for SNG production – a process integration study. *To be submitted*.

IV. Pihl, E., Heyne, S., Thunman, H., Johnsson, F. (2010). High efficiency power production from biomass by integration and hybridization with combined cycle gas turbine (CCGT) plants for natural gas. *Accepted for publication in Energy*.

Stefan Heyne is the main author of the first three appended papers and contributed by means of calculations to the gasification part of paper IV, for which Erik Pihl is the main author. Simon Harvey was the main supervisor of and Henrik Thunman co-supervised the work.

# Table of Contents

# 1 Introduction

The production of synthetic natural gas (SNG) from gasified biomass is one of the alternative pathways for the reduction of anthropogenic greenhouse gas (GHG) emissions. SNG produced from renewable resources results in reduced emissions of $CO_2$ when replacing fossil natural gas in conventional applications, such as gas turbine power cycles. An even higher potential for GHG emission reduction can be achieved by using biomass based SNG as a vehicle fuel, both for conventional internal combustion engine applications, as well as future fuel cell applications. Thus, biofuels in general are expected to contribute substantially to the GHG emission reduction within the transportation sector. Biomass is a limited resource, therefore biomass-based SNG must compete with other biomass applications within the energy sector based on an economic, infrastructural as well as an environmental perspective. Aiming at a sustainable energy supply system in the future, it is necessary to identify the most efficient conversion pathways for biomass and to introduce suitable process performance indicators to help decision-makers in restructuring the current energy supply structure in a positive way.

## 1.1 Background

Anthropogenic GHG emissions have increased drastically over the last century and emission reduction is a major challenge for both industrialised and developing countries. Biomass based energy supply is expected to be an important contribution to this necessary reduction. In some of its energy supply scenarios, the International Energy Agency (IEA) estimates that the use of biomass may reach today's level of oil consumption by 2050 (IEA, 2008). In 2007, the share of biomass in the total primary world energy supply was 9.8% (IEA, 2009), most of it used for basic heating and cooking applications while only a small share was used for high efficiency energy conversion options, such as combined heat and power (CHP) applications.

Within the European Union (EU) the share of renewables in the primary energy consumption in 2008 was 8.2%, with biomass accounting for about two-thirds of this share (EurObserv'ER, 2009). In a directive on the promotion of the use of energy from renewable sources (EU, 2009), the EU set a mandatory target of a 20% share of renewable energy in the overall energy supply and – in addition – a 10% share of renewable energy supply within the transport sector in the year 2020. In 2008, the share of renewable fuels within the transport sector was 3.4% on an energy basis, with biomass-based gas providing a marginal 0.3% of that share as biogas is used almost exclusively in Sweden for transport (EurObserv'ER, 2009). Fossil natural gas consumption as a vehicle fuel in Europe is estimated to 7.4 million tons of oil equivalent (Mtoe) in 2009 (Boisen, 2009). As a reference, road based transport energy consumption in the EU-27 countries was about 309 Mtoe in 2006 (EU, 2010).

A large potential for a further increase of the share of biofuels is expected with the introduction of second-generation of biofuels based on thermal gasification, such as dimethyl ether (DME), methanol, Fischer-Tropsch (FT) diesel, hydrogen or SNG. A recent analysis in a European framework indicates that a large share of the EU fuel demand can be covered by domestically produced biofuels (Londo et al., 2010). According to this study, the introduction of second-generation biofuels can be effectively promoted by the use of biomass in other energy applications, such as the co-firing of biomass in coal power plants. Synergy effects, including the build-up of an effective infrastructure for biomass within the energy sector based on these kinds of applications, are estimated to be more important than the precursor role of the first generation of biofuels. It is also clearly stated that policy instruments will play an important role in the promotion of renewable fuels within the transport sector.

## 1.2 Objective

This work focuses on using process integration tools to identify efficient pathways for the production of SNG by thermal gasification of biomass. A number of products and energy services resulting from the SNG production process are accounted for, as illustrated in Figure 1. The availability and amount of the different products/services depend to a large extent on both the process feedstock and layout.

A variety of applicable technologies exist to carry out the different conversion steps within the SNG production process. The combination of these different technologies results in a number of process alternatives with varying product spectra. So far, no plant has been built on an industrial scale and there is a lack of knowledge of how to combine the different sub-processes into an optimized and well-integrated process. Using process integration tools – in particular pinch methodology – this study investigates thermal integration opportunities between the different sub-processes and the resulting consequences for overall process efficiency.
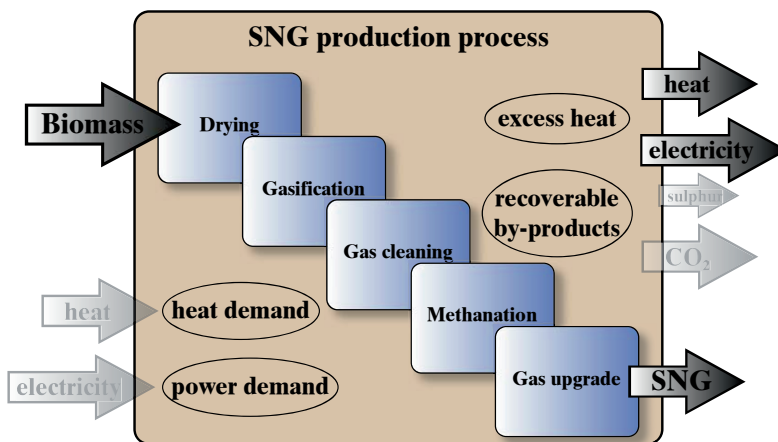


*Figure 1:* SNG production process scheme illustrating possible inputs and products/services.

The focus of the evaluation of different process pathways is on:

- the conversion efficiency of biomass into SNG

- the use of recoverable process heat for the cogeneration of power and heat

In order to facilitate this evaluation, it becomes necessary to carefully define appropriate performance indicators. These performance indicators should be of a general character, allowing for a fair comparison of SNG with other biofuel alternatives.

Models for obtaining the mass and energy balances for the different process steps within the SNG production process are developed and used as a basis for sensitivity analysis and process optimisation. Based on this investigation critical process steps for process heat integration and SNG yield are identified. Process

integration tools are used to point out the potential of overall process performance improvements by changes involving these critical process steps.

As the introduction of second generation biofuels may be facilitated by making use of existing infrastructure, the integration of the SNG production process with existing biomass-based combined heat and power plants using indirect gasification technology is investigated in detail. Alternative opportunities to integrate biomass gasification by replacing fossil fuels to produce power are also highlighted.

This thesis is the result of ongoing work, and future work within the project will aim at further steps for process evaluation of alternative production pathways for biomass to SNG, such as:

- the economic performance against the background of possible future energy market scenarios

- the $CO_2$ emission consequences related to the production and use of SNG

These aspects are, however, not covered in this report, but are subject to future work.

## 1.3   Thesis Outline

This thesis presents the results obtained so far within the research project. In Chapter 2, the basic process steps within the SNG process are presented and a résumé of published research on SNG production from biomass is given. The methodological approach used in this thesis is detailed in Chapter 3. The results of the performed investigations are then presented and discussed in Chapter 4 and rounded up by concluding remarks in Chapter 5. Finally, the planned further work is presented in Chapter 6.

# 2 Overview of Process Options for the Production of SNG by Thermal Gasification

The production of methane from biomass can be achieved in a variety of ways. Today's established technology ferments wet biomass and upgrades the resulting gas to produce grid-quality methane – often referred to as biogas. Examples of possible feedstock materials for this type of process include sewage or industrial waste water, or wet manure from agriculture. In 2005, the overall production of biogas by fermentation in Sweden amounted to 1.3 TWh (Clementson, 2007), while the near future potential is estimated at around 14-17 TWh per year (Svensson et al., 2009). Compared to the overall primary energy consumption in Sweden, which amounted to 618 TWh in 2008 (SEA, 2009), this potential corresponds to about 2.5%. To further increase the production of methane from biomass, the technology of choice is thermal gasification, which allows for a larger spectrum of feedstock – in particular lignocellulosic biomass – in addition to a substantially higher production capacity. Methane produced from gasified biomass, which meets natural gas grid specifications is hereafter referred to as Synthetic Natural Gas (SNG). Conservative estimates of the production potential for SNG by thermal gasification in Sweden are around 59 TWh per year; optimistic estimates on a larger timescale – taking into account advanced forest management – range up to 200 TWh (Svensson et al., 2009).

The interest in the production of SNG by thermal gasification has grown recently in the context of research on second generation biofuels. However, the process concept itself is not new; the production of SNG by means of coal gasification was heavily investigated during the oil crisis in the 1970s and 1980s. The most prominent example of a commercial facility for SNG production is the Great Plains Gasification Plant (Panek & Grasser, 2006). This plant has been in constant operation since 1984 and has a production capacity of about 2 GW SNG from coal

feedstock. The challenge of adapting this process concept to a different feedstock – namely biomass – mainly resides in the scale of production. This production is quite a great deal smaller because of the lower energy density and geographical availability of biomass. In addition, different types of impurities compared to coal-based SNG production have to be removed during gas processing, such as organic sulphur compounds. Further details will be given in the corresponding sections of this chapter.

## 2.1 Process Description

A general description of the SNG production process is illustrated in Figure 2. The generic setup can essentially be applied to almost any second generation biofuel with only slight modifications.



**Figure 2:** *General process steps to produce SNG by thermal gasification of biomass.*

The incoming biomass needs to be dried prior to gasification, where it is then converted from a solid state to a gaseous phase, along with solid mass residuals in the form of ash and non-gasified char. The gas phase – in the following referred to as product gas – then needs to be cleaned from any impurities, including particles, tars, sulphur compounds etc. before it can be catalytically converted to a mixture containing mainly $CH_4$ and $CO_2$. The final step of upgrading the gas is needed to provide grid quality SNG: the $CO_2$ is removed and the gas dried and compressed. A brief overview of the different technology alternatives and related critical aspects of the process integration is provided below.

### 2.1.1 Drying

The natural moisture content of woody biomass is usually around 50 wt-%. Prior to gasification using low quality heat, drying is used to reduce this moisture content, thereby reducing the energy input at the elevated temperature level of the gasification process. Increases in the overall thermal efficiencies of the combustion and gasification processes of about 5-15% can be achieved when drying the fuel prior to combustion or gasification (Amos, 1998). The optimal moisture content for biomass gasification depends on the gasification technology and the subsequent

product gas usage, but is between 10-20 wt-% for most gasification systems (Cummer & Brown, 2002).

The energy demand for moisture evaporation alone is around 2650 kJ/kg $H_2O$ (assuming the moisture is incoming at 0°C and leaving as saturated vapour at 80°C). Additional energy input is needed in drying equipment for heating up the biomass, compensating for losses, and material/gas transport.

Basically, three different drying techniques are applied within the bioenergy sector:

- Steam drying

- Flue gas drying

- Low-temperature air drying

A rough estimation of the energy demand for flue gas dryers amounts to 3200 kJ/kg $H_2O$ (Wimmerstedt & Linde, 1998). In contrast, for a steam dryer, the net heat demand is considerably lower (theoretically as low as 600 kJ/kg $H_2O$), since it is possible to recover a large amount of the input heat by condensation of the evaporated biomass moisture. A steam dryer is, therefore, of particular interest to heat integration. This advantage is counteracted by the higher investment costs; a fluidised bed type steam dryer compared to a stand-alone flue gas dryer capable of removing 25 t $H_2O$/h is estimated to result in about 40% higher investment costs (Wimmerstedt & Linde, 1998).

A common problem associated with the biomass drying process is the emission of volatile organic compounds (VOC) emitted from the wet material alongside the water vapour. Higher temperatures lead to higher emissions, and depending on the dryer type and the nature of the organic material, flue gases and water condensates contain a certain amount of impurities requiring treatment (Svoboda et al., 2009). The condensate mainly contains organic acids, terpenes and alcohols, while the flue gases are contaminated with non-condensable terpenes and aromatic compounds (Boström & Ljungqvist, 2000).

Given that a lower drying temperature leads to lower emissions and given the attractiveness of using low quality heat for the drying process, there is currently significant interest in the development of low-temperature air drying systems. Two different concepts for air drying – single-stage drying with recycle and multi-stage drying – have been estimated to result in energy demands just above 2700 kJ/kg $H_2O$ with heat sources at temperatures as low as 80°C (Holmberg &

Ahtila, 2005). The Swedish company Svensk Rökgasenergi AB offers a low-temperature air drying system with inlet air temperatures at 70ºC (SRE, 2010). Using low grade heat – that is usually discarded into the environment – for drying purposes instead of high temperature flue gases often generated by combustion of additional biomass represents a large potential for energy savings. It is estimated to increase the biomass potential within the Swedish forest industry by up to 25% without any additional wood cuttings (Johansson et al., 2004). This optimistic scenario is based on assumptions that not only the heating value is increased, but also that the loss of solid combustible mass by biological degradation is substantially decreased by the fuel drying process. More moderate estimates of the energy saving potential of drying are about 10% (Spets, 2001).

### 2.1.2 Gasification

The dried biomass is then fed into a gasification unit. For conventional thermal gasification three technology alternatives exist, that are depicted in Figure 3.
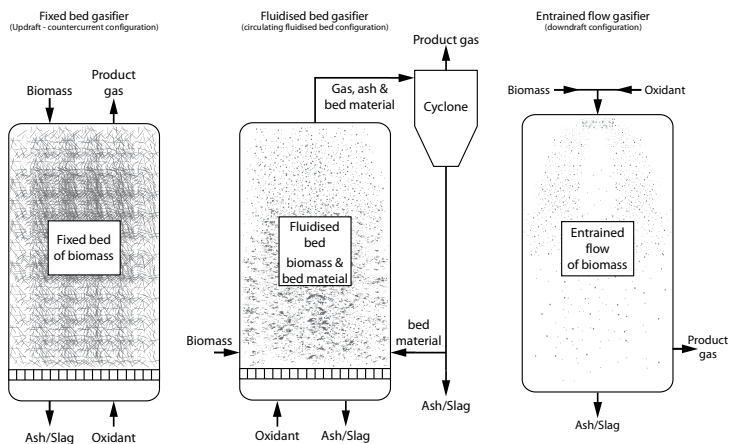


*Figure 3*: *Different types of thermal gasification technology.*

There also exist gasification technologies adapted to wet biomass feed – namely hydrothermal gasification at conditions close to the critical point for water (374 °C, 221 bar) – that has been proven on a laboratory scale (Waldner & Vogel, 2005). In a recently published systems analysis study, this technology has been identified as promising for the future production of SNG (Luterbacher et al., 2009), but due to its early stage of development and limited amount of published data, the hydrothermal gasification technology is not yet considered in this work.

The various technologies considered require specific feed qualities, have different ranges of operation, and – most importantly – have a certain range of capacity. Indicative values for the different parameters are given in Table 1. Fixed bed gasification is mainly suited for small scale applications, while fluidised bed and entrained flow gasification are more suitable for medium to large scale applications.

*Table 1*: Gasification technology characteristics.

|  | **Fixed bed** | **Fluidised bed** | **Entrained flow** |
|---|---|---|---|
| Input particle size [mm] | 10 – 300 | < 50 | < 0.1 |
| Outlet gas temperature [℃] | 400 – 1000 | 700-1200 | 1200-1500 |
| Operating pressure | from atmospheric pressure (1 bar) to ~25 bar | | |
| Gasification medium | air, steam, oxygen, flue gas, product gas | | |
| Plant size [$MW_{th}$ input] | < 50 | 10 – 100 | 100 – 1000 |

The different operational ranges and setups also result in differing qualities of the product gas with respect to gas composition, heating value and level of purity (e.g. tar content). Besides the gasification technology, the gasification medium influences these properties to a major extent. Fixed bed and entrained flow gasification are direct gasification technologies with an oxidising medium (e.g. air or oxygen) partly combusting the biomass in order to provide the necessary heat for gasification. Fluidised bed gasification allows for indirect gasification, with the heat for gasification being transferred to the biomass by means of the circulating bed material. A separated combustion chamber then heats up the circulating bed particles. The non-gasified char from the gasification reactor provides additional fuel supply to the combustion chamber. A simplified flow diagram involving indirect gasification is illustrated in Figure 4. Indirect gasification produces a gas with very low nitrogen content, making it suitable for synthesis of biofuels or chemicals. In the case of direct gasification, oxygen-production is necessary in order to obtain a nitrogen-free gas.
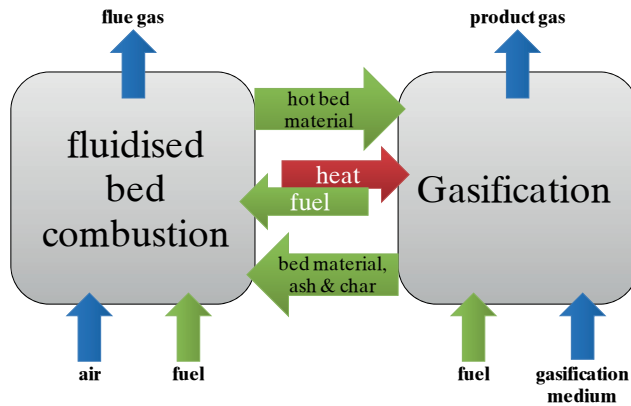
***Figure 4****: Simplified flow diagram of indirect gasification.*

Several biomass gasification projects are currently in progress with differing use of the product gas, ranging from heat and power production to various applications involving biofuel synthesis. In Värnamo, Sweden, a demonstration power plant based on oxygen-blown fluidised bed gasification to generate CHP with a fuel capacity of 18 MW was in operation from 1996 to 2000 (Ståhl, 2001). Plans to adapt the unit to produce biomass-based transport fuels or other higher-value chemicals are underway (Albertazzi et al., 2005). In Güssing, Austria, an 8 MW indirect gasification unit using steam as gasification medium – the fast internally circulating fluidised bed (FICFB) gasifier – has been in operation since 2001 (Hofbauer et al., 2002; Proll et al., 2007). The FICFB gasifier was used as a demonstration unit to produce SNG from a slip stream (Seemann et al., 2004) and in 2009, a process development unit was commissioned that produces up to 1 MW of SNG from product gas (Bio-SNG, 2009). The Energy Research Centre of the Netherlands (ECN) has developed a fluidised bed steam gasification process aiming to produce SNG as well. Laboratory and project scale units have been successfully operated, and a 10 MW demonstration plant will be taken into operation in 2012 (van der Meijden et al., 2009). In Freiberg, Germany, an entrained flow gasification plant has been commissioned to produce 18 000 $m^3$ FT diesel from biomass a year (Hoffmann, 2008). At the Chalmers University of Technology, an indirect gasification concept has recently been developed and demonstrated on a pilot-scale, and similar to the flow scheme illustrated in Figure 4, the gasifier is built as extension of an existing fluidised bed boiler (Thunman & Seemann, 2009). This concept makes it possible to use existing infrastructure in the

form of biomass heat and power plants, thus reducing the risk of investment compared to a stand-alone gasification plant.

### 2.1.3 Gas Cleaning

The product gas resulting from the gasification process mainly consists of carbon monoxide, hydrogen, carbon dioxide, methane and water vapour. However, the presence of trace components makes several gas cleaning steps necessary prior to synthesizing methane. The constituents present in the product gas and needing treatment are particulate matter (ash, bed particles), higher hydrocarbons and tars, sulphur and nitrogen compounds, as well as alkali metals.

Particulates may be removed from the product gas by cyclones, hot gas or fabric filters, as well as by scrubbing separators. Particle separation also reduces the tar content of the product gas and the extent of such removal is dependant on the separation technology applied (Han & Kim, 2008).

Tar removal is necessary in order to avoid excessive fouling of the heat exchanger equipment. It is possible to reduce tar formation during the gasification process by using catalytic bed material (Pfeifer et al., 2004), but several post-gasification product gas cleaning technologies are available as well. Scrubbing has been demonstrated as feasible to remove product gas tar both below and above the dew point (Zwart et al., 2009; Rauch & Hofbauer, 2003). Water condensation in the scrubbing unit makes a phase separation of the condensate necessary and yields waste-water contaminated with organic compounds, thereby favouring the scrubbing technology above the dew point. However, scrubbing generally implies thermal losses during the process as part of the sensible heat of the product gas is lost. Catalytic reforming of tars at higher temperatures might overcome this problem but has not yet been proven on an industrial scale. Chemical looping reforming (CLR) has been proven at a laboratory scale to produce synthesis gas (Ryden et al., 2006) and could potentially be applied for tar reforming (Lind et al., 2010).

Sulphur compounds – mainly hydrogen sulphide – present in the product gas are highly poisonous to catalysts and need to be reduced to very low concentrations prior to the synthesis step. Regenerative sulphur guards may be used to reduce sulphur concentrations to well below 1 ppm (Forzatti & Lietti, 1999). Washing techniques are also available to remove sulphur, even making recovery via the Claus process possible (Vogel et al., 2006). However, the recovery of sulphur is

only economically feasible for large scale units since sulphur compounds are only present in minor concentrations in product gas resulting from wood biomass.

For removing alkali traces in the product gas, both washing techniques and techniques based on solid sorbents – "alkali getters" – are available. The latter operate at high temperatures and are either based on chemisorption or physical adsorption (Turn et al., 1998). The influence of this removal process on the overall energy balance of the SNG process can be considered negligible due to the small amount of alkali present in the product gas based on wood biomass with low ash content. This can, however, change if wood waste material, sludge or other contaminated feedstock were used for gasification. Leaching of the feedstock prior to gasification is an alternative technology employed to remove alkali (Cummer & Brown, 2002)

### 2.1.4 Methanation

Clean product gas may then be catalytically converted to methane in the following step. The main reaction occurring during methanation is the conversion of carbon monoxide and hydrogen to methane and water. As documented in the literature (Hayes et al., 1985) and as is obvious from Eq. (1), the optimal ratio of hydrogen to carbon monoxide prior to methanation is 3/1.

$$CO + 3\,H_2 \leftrightarrow CH_4 + H_2O \qquad \Delta H_r^{298\,K} = -205.9 \text{ kJ/mol} \qquad (1)$$
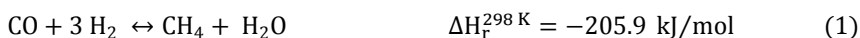
For product gas from biomass gasification, however, this ratio is often less than two, making a water-gas shift (Eq. (2)) necessary.

$$H_2O + CO \leftrightarrow H_2 + CO_2 \qquad \Delta H_r^{298\,K} = -41.1 \text{ kJ/mol} \qquad (2)$$

Carbon dioxide is another possible source of methane from the product gas and can be converted according to Eq. (3). This is, however, a linear combination of Eqs. (1) and (2).

$$CO_2 + 4\,H_2 \leftrightarrow CH_4 + 2\,H_2O \qquad \Delta H_r^{298\,K} = -164.8 \text{ kJ/mol} \qquad (3)$$

The highly exothermic character of the methanation reaction (Eq. (1)) makes it an interesting source of process heat recovery within the SNG process. Two different reactor concepts have been developed for methanation with most of the development having occurred in connection with coal-to-SNG projects of the 1970s and 1980s (Kopyscinski et al., 2010). Methanation is either carried out in a series of adiabatic fixed bed reactors with inter-cooling and optional product recycle

(Harms et al., 1980; Moeller et al., 1974), or in a single fluidised bed reactor at isothermal conditions (Friedrichs et al., 1982). Commonly used catalysts are Ni-based. The two alternative technologies are illustrated in Figures 5 and 6.

The Paul-Scherrer-Institute (PSI) has adopted the isothermal fluidised bed methanation technology for biomass-based product gas in a once-through reactor and has successfully operated the pilot-scale in the Güssing biomass gasification plant (Seemann, 2006; Bio-SNG, 2009).



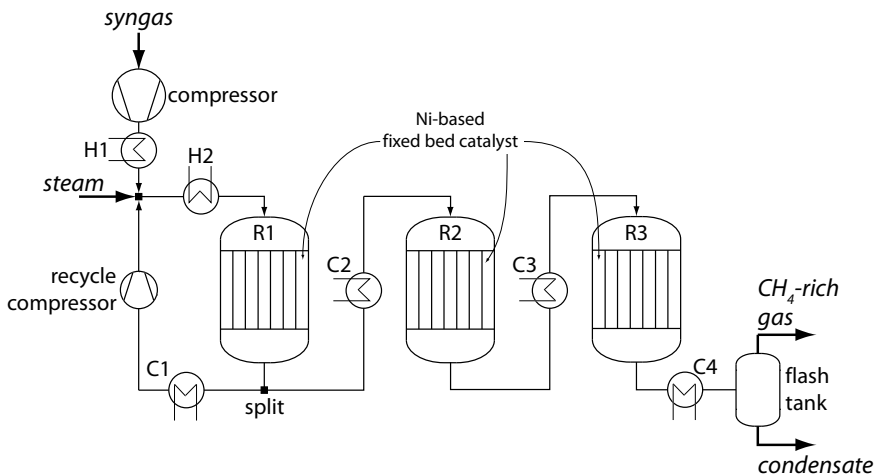*Figure 5: Adiabatic fixed bed methanation with inter-cooling and recycle (TREMP technology). H1 & H2: heater, C1-C4: cooler, R1-R3: adiabatic methanation reactor (flowsheet adapted from Harms et al. (1980)).*
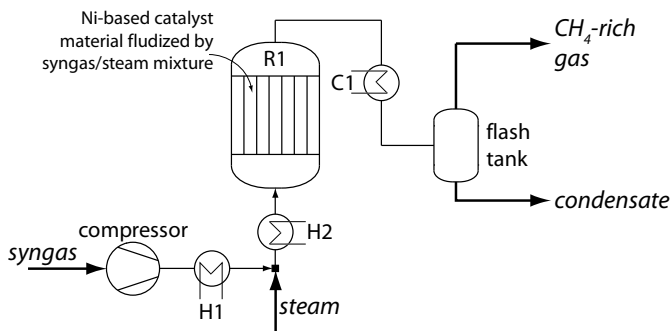


*Figure 6: Isothermal fluidised bed methanation (COMFLUX technology). H1 & H2: heater, C1: cooler, R1: isothermal methanation reactor (flowsheet adapted from Friedrichs et al. (1982)).*

From a process integration perspective the main differences between the two technologies are the temperature levels where excess heat is released and the operating pressure. For the adiabatic process, temperatures up to 650 °C are reached, limited by catalyst sintering problems at higher temperatures (Rostrup-Nielsen et al., 2007). Fluidised bed methanation is operated at temperatures around 300 °C. Lower temperatures would be even more favourable from a thermodynamic viewpoint, but are, however, limited by catalyst activation and carbon deposition problems (Seemann, 2006). A higher pressure is favourable to yield methane as the number of moles is reduced from four to two according to Eq. (1). Pressure effects are considerably more marked at higher temperatures (Deurwaarder et al., 2005). Consequently, adiabatic fixed bed methanation needs to be operated at elevated pressure in contrast to isothermal methanation, which achieves high conversion efficiencies even at atmospheric pressures. A careful integration of the methanation process into the overall SNG process is crucial to obtain an optimal overall process performance.

### 2.1.5 Gas Upgrade

The gas produced by methanation is a mixture of methane, carbon dioxide and water, with remaining traces of nitrogen, hydrogen and carbon monoxide. In order to feed SNG into the natural gas grid, certain quality requirements need to be fulfilled. Quality requirements differ between countries, but a harmonisation of gas quality within the European Union is in progress (EASEE-gas, 2005). Table 2 presents the recommended quality requirements. A number of trace components present in gas originating from biomass (e.g. CO, $H_2$ and $NH_3$) are, however, not mentioned.

The most important gas upgrade step is the separation of $CO_2$. This is an energy intensive process with several technical solutions available that differ considerably in heat and power demand. Table 3 gives an overview of the most common techniques and a qualitative indication of their energy demand, methane recovery and investment costs. When integrating the $CO_2$ separation within the SNG process, the energy demand may change substantially depending on the amount of available excess heat and pressure levels of the other process steps. This is, in particular, applicable to the heat demand; as an example, one could imagine covering the large heat demand of an amine-based absorption (MEA) by means of using the reaction heat released during methanation.

**Table 2:** *Natural gas quality requirements according to a recommendation of the European Association for the Streamlining of Energy Exchange – gas (EASEE-gas, 2005)*

| Parameter | Unit | Min | Max |
|---|---|---|---|
| Wobbe index (WI)[1] | kWh/m$^3$ | 13.60 | 15.81 |
| Relative density RD[2] | m$^3$/m$^3$ | 0.555 | 0.700 |
| Total sulphur S | mg/m$^3$ | - | 30 |
| H$_2$S + COS (as S) | mg/m$^3$ | - | 5 |
| Mercaptans | mg/m$^3$ | - | 6 |
| Oxygen O$_2$ | mol-% | - | 0.001 |
| Carbon dioxide CO$_2$ | mol-% | - | 2.5 |
| Water dewpoint | °C at 70 bar | - | -8 |
| Hydrocarbon dewpoint | °C at 70 bar | - | -2 |

[1] The Wobbe index WI is defined as the higher heating value divided by the square root of the relative density RD $\left( WI = \frac{HHV}{\sqrt{RD}} \right)$

[2] The relative density RD is defined as the gas density in relation to the density of air at standard conditions (0°C, 1.01325 bar) $\left( RD = \frac{\rho_{gas}}{\rho_{air}} \right)$

**Table 3:** *Qualitative characterisation of CO$_2$ separation processes (Johansson, 2008; ISET, 2008; Reppich et al., 2009).*

| Technique | Heat demand | Power demand | Methane recovery | Investment costs |
|---|---|---|---|---|
| Pressurised water scrubbing (PWS) | none | moderate | high | low |
| Amine based absorption (MEA) | very high | low | very high | high |
| Physical absorption (Selexol) | moderate | moderate | high | moderate |
| Pressure swing adsorption (PSA) | none | moderate | high | moderate |
| Membrane separation | none | high | moderate | very high |
| Cryogenic separation | none | high | very high | very high |

Finally, the gas needs to be dried of any remaining moisture and then compressed. Drying is usually achieved by a glycol wash unit using, frequently, triethylene glycol (TEG) as a solvent, alternatively it may be done by means of a temperature swing adsorption (TSA) process using silica gel or aluminium oxide (GPSA, 2004). The compression level is dependent on the location of the production site and the

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grid into which the SNG is fed. Local distribution networks in Sweden have a pressure up to 10 bar, while large transmission pipelines are pressurised at above 100 bar.

## 2.2  Related Work

The overall process chain from biomass to SNG has been analysed by several research groups, often related to experimental or pilot-scale projects. Mozaffarian & Zwart (2003) have compared different process alternatives to produce SNG based on pressurised oxygen-blown CFB gasification and atmospheric indirect steam gasification. Excess heat from the process is used to generate steam at 40 bar, which is then expanded to generate power in a condensing steam turbine. No systematic approach for the heat integration is reported. Indirect gasification is pointed out as a superior technology in all cases studied and the overall conversion efficiencies for biomass into SNG are estimated at up to 70% on a lower heating value (LHV) basis. Integrated drying is not considered, but the biomass feedstock is assumed to have been dried before entering the process. Cost estimates of the indirect gasification option range between 16.2 and 40 €/MWh$_{SNG}$. Even hydrogasification (using hydrogen as gasification agent) and coproduction of SNG and FT diesel are pointed out as promising and even more efficient alternatives by Mozaffarian & Zwart in their study.

A more recent study presents a comparison between three different gasification systems for SNG production (van der Meijden et al., 2010) – pressurised oxygen-blown gasification, indirect steam gasification and entrained flow gasification using oxygen. As pretreatment, only torrefaction is considered in the case of the entrained flow gasifier, but dry wood at 15 wt-% moisture is assumed to be the feedstock for all process alternatives. Again, indirect gasification is identified as the most promising technology from the perspective of conversion efficiency to produce SNG.

An exergetic analysis of the indirect gasification to produce SNG (Jurascik et al., 2010) identifies the gasifier, methanation step and $CO_2$ separation unit as the largest sources of loss. The overall exergetic efficiency of the process on a dry fuel basis (13.8 wt-%) – comparing the exergetic value of produced SNG, steam and additional excess heat to all exergetic input (fuel & electricity) – varies from 69.5 - 71.8% for gasification temperatures and pressures between 650 - 800°C and 1 - 15 bar, respectively.

A study by Duret et al. (2005) combines process modelling and process integration techniques to investigate an SNG production process based on the FICFB indirect gasification and isothermal methanation. Multi-stage membrane separation is selected for the SNG upgrade. The thermal efficiency of wood to SNG of 57.9% is determined for the process with a thermal input between 10 - 20 $MW_{LHV}$. Heat integration based on pinch technology is used to assess the opportunities for steam generation for power production. The electricity produced from the process excess heat is not sufficient for the estimated power demand of the SNG production process, and 7% of the power demand must be purchased from the grid. Feedstock drying is not considered and the incoming wood is assumed at 17 wt-% moisture.

Development of the former work extends the analysis to the drying step and presents a thermo-economic model to optimise SNG production using genetic algorithms (Gassner & Maréchal, 2009). Based on a generic superstructure, different technology alternatives to produce SNG may be combined, and subsequent automated process-integration based on pinch methodology provides the maximal amount of mechanical power that can be produced from excess heat under given conditions. By varying the operating parameters of the different process steps, the most promising combinations of technology can be selected based on predefined process performance indicators. Overall energy and exergetic efficiencies in the range of 69-76% and 63-69%, respectively, are obtained. A broad range of production costs for SNG is estimated in the study – 76 - 107 and 59 - 97 €/$MWh_{SNG}$ based on a thermal input of 20 and 150 $MW_{LHV}$ into the process, respectively.

## 2.3 Biomass-Based SNG Production on Industrial Scale

No large scale plant for SNG production by means of thermal gasification of biomass has entered operation as of today. However, demonstration plants have been successfully operated at ECN and Güssing/Austria, as mentioned in Section 2.1.2. At ECN, a large scale plant for SNG production (50 $MW_{LHV}$ input) is planned as future development of an intermediate size CHP plant (10 $MW_{LHV}$ input) based on ECN gasification technology. This CHP plant – to be built in 2012 – is projected to use gas engines for electricity production from the cleaned product gas (van der Meijden et al., 2009). The only planned industrial scale activity to produce SNG from biomass is the Gothenburg Biomass Gasfication project (GoBiGas) (Gunnarsson, 2009). Based on the Güssing demonstration plant to a considerable extent, Göteborg Energi AB is planning to produce 20 $MW_{LHV}$ of

SNG in an initial unit in 2012. Based on this experience, improvements will be implemented in a second plant, extending the capacity to 100 $MW_{LHV}$ by 2016. The first stage plant will only produce heat as a by-product, but for the large scale plant, a combined heat and power production unit making use of the available process excess heat is projected. The knowledge of the optimal integration of the different sub-processes is of substantial interest to determine economically viable process alternatives. The costs of large scale future plants are expected to decrease substantially with additional large-scale experience. Several obstacles to industrial activities still need to be resolved in order to incite large scale production of second generation biofuels in general:

- there are only a few or no suppliers that provide turnkey-ready plants
- volatile fuel market prices make it difficult to ensure the profitability of biofuel processes
- policy instruments promoting biofuels need to be clearly defined to allow for long term planning

All these aspects imply a high financial and technical risk to companies engaged in biofuel production activities. Again, process integration may contribute to reducing these risks.

# 3 Methodology

The aim of this work is to identify energy efficient pathways by which SNG may be produced from biomass. As this process is not yet established on an industrial scale, scant data on the different process steps is available. This makes it necessary to use modelling tools to estimate mass and energy balances. Using these energy balances, a systematic integration of the process steps may be conducted by means of pinch methodology. Figure 7 illustrates the basic idea of the process integration methodology presented in this work.
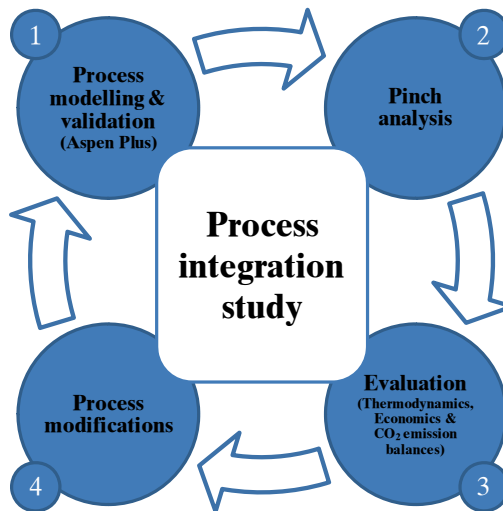


**Figure 7:** *Schematic representation of the process integration methodology applied in this work.*

An iterative process analysing the influence of changed operating parameters and alternative technologies for selected process steps within the SNG process leads to an improved process design. The integration with existing infrastructure on both a

heat integration as well as a technical level is investigated in particular. To evaluate the different process alternatives, process performance indicators allowing a fair comparison with biofuel alternatives other than SNG are defined. Finally, the most promising process alternatives are evaluated against the background of potential future energy market scenarios in order to identify economically viable production pathways for SNG with good $CO_2$ emission reduction potential. The different elements of methodology will be described in this section.

## 3.1   Process Modelling

In order to obtain the mass and energy balances of the SNG production process alternatives investigated, the flow-sheeting software Aspen Plus was used in this work. Aspen Plus provides an extensive physical property database, equations of state for different conditions and a number of built-in models for common process unit operations. In addition, it is possible to adapt the built-in models using calculation blocks of Fortran or Excel. For gas phase processes, the Peng-Robinson cubic equation of state employing the Boston-Mathias alpha function extension was used, while steam table data was applied to processes only involving water or steam. For distillation and scrubbing units involving electrolytes, the electrolyte non-random two liquid (ELECNRTL) model was applied. Modelling data were validated to a maximum possible extent based on published data.

The following species were taken into account as being present in the raw product gas:

- main components: $CO$, $CO_2$, $H_2$, $H_2O$, $CH_4$

- char, hydrocarbons & tars: $C$, $C_2H_4$, $C_6H_6O$, $C_{10}H_8$, $C_{16}H_{10}$

- sulphur compounds: $H_2S$, $COS$, $C_4H_4S$, $CH_4S$

- nitrogen compounds: $NH_3$, $HCN$, $NO$

- trace components: $N_2$, $O_2$, $HCl$

The gasification was modelled as a two-step process involving pyrolysis followed by gasification. The pyrolysis step is a simple decomposition of the solid biomass into a gaseous phase and char and ashes in solid form. The gas composition can be determined with the help of elemental balances and results from pyrolysis experiments using single wood particles (Thunman et al., 2001) to set the ratio between major components present in the volatile gas phase. The ratio of several

trace components (in particular tars, sulphur and nitrogen compounds) was difficult to determine based on literature data as little data is available. The ratios were set to fixed values with $H_2S$ and COS being the main species of sulphur compounds and $NH_3$ the major nitrogen compound. In fact, the idea of including all these trace species in the modelling effort is not to arrive at exact figures for their concentrations, but rather to get an inspiration of where in the process chain these compounds might cause problems. From an energy integration point of view the presence of these compounds is essentially insignificant.

The gasification step was modelled as a Gibbs equilibrium reactor with the following five species being accounted for in the equilibrium reactions: C, CO, $CO_2$, $H_2O$, $H_2$. All other species are assumed to be inert and the carbon conversion is set to a fixed value. By no means does this represent the complex network of reactions going on during gasification; it has, however, been shown to well represent the composition of the indirect steam gasification product gas resulting from the FICFB gasifier (Hofbauer & Rauch, 2000). For the base case process considered in this work, the steam-to-biomass ratio was set at 0.6 and a mixture of recycled product gas and steam was considered for fluidisation.

The product gas resulting from indirect gasification at 800-900ºC contains relatively large amounts of tar. These tar compounds are prone to condense during product gas cooling on heat exchanger surfaces and cause fouling of equipment. A novel technique for removing these tars – chemical looping reforming (CLR) – was implemented in the model. Supplying a small amount of oxygen by means of a catalytic carrier material to the product gas, the tars are selectively oxidised while the rest of the product gas, in particular methane, remains unaffected. A schematic flowchart of the process is illustrated in Figure 8. The chemical looping reforming is assumed to operate at 650 ºC and all tars are assumed to be converted to CO and $H_2$.
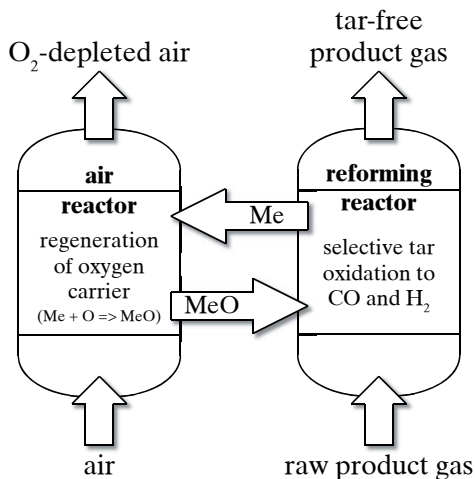
*Figure 8: Schematic flowchart of chemical looping reforming (CLR).*

The further processing of the product gas consists of a water scrubber mainly removing $NH_3$, a two step $CO_2$ removal using monoethanol amine (MEA), two isothermal fluidised bed methanation steps, compression, removal of hydrogen traces by a membrane process and finally – drying the resulting SNG. Except for the methanation step, the processes are primarily based on literature data and no detailed modelling has been performed to date. The methanation was modelled according to a Gibbs equilibrium reactor assuming a simultaneous water-gas shift and methanation reaction (see Eqs. (1) and (2)). A temperature approach was used for the equilibrium calculations in order to account for non-idealities of the reactor and steam is added prior to methanation in order to adjust the $H_2/CO$ ratio. The main modelling assumptions of the base case SNG production process are provided in Table 11 (c.f. Appendix).

## 3.2  Process Integration

Process integration refers to the application of systematic methodologies that facilitate the selection and/or modification of processing steps, and of interconnections and interactions within the process, with the goal of minimizing resource use. In general, process integration refers to a holistic approach to analyse the overall process performance, rather than on optimising single process steps. In this study, two different approaches to process integration are assessed. The first approach refers to the thermal integration between different process heat streams according to pinch technology, whereas the second approach refers to an

integration on a more practical level – namely the integration of the SNG production process with existing infrastructure in the form of fluidised bed boilers for combined heat and power production.

### 3.2.1 Pinch Technology

Pinch technology is based on the first and second law of thermodynamics and refers to the combination of pinch analysis and process design based on pinch rules. It is widely used to determine the minimum heating and cooling demand of various industrial processes and to identify potential process improvements. Savings up to 35% in energy consumption are achievable by analysing and improving these processes with the help of pinch analysis (CANMET, 2003). The basics of pinch analysis have been developed by Bodo Linnhoff and his research group who also published the first pinch analysis user guide (Linnhoff et al., 1994). This user guide has been revised and a new version has been published by Kemp (2007).

Initially, pinch technology was developed for the design of new, and improvement of existing heat exchanger networks, but its use has been extended for several other applications, including investigation of optimal integration features of new process steps in retrofit situations (Smith, 2005). Various studies have shown the usefulness of this methodology for retrofit projects in, for example, the pulp and paper industry (Hektor, 2008; Olsson, 2009). Even analogous applications to heat exchange have been analysed using pinch-like methodologies. Examples include the reduction of freshwater use by improving process & waste water management and hydrogen network analysis (CANMET, 2003).

A common representation of the thermal integration potential of a process in pinch technology is the "Grand Composite Curve" (GCC). The GCC is a representation of all heat streams of a process, illustrating the opportunities for heat exchange, as well as the minimum external heating and cooling demand for the process. Figure 9a is an illustrative example of a GCC. The temperature on the y-axis does not represent the actual temperature of the different streams but is an interval temperature shifted by a certain value (shifted upwards for cold streams needing heating and downwards for hot streams needing cooling) in order to take into account the temperature difference (driving force) necessary for heat exchange between streams.

A variation of the GCC illustrated in Figure 9b is the Carnot-based GCC in which the temperature in the y-axis has been replaced by the Carnot efficiency $\theta$ defined as

$$\theta = 1 - \frac{T_{ref}}{T} \tag{4}$$

where $T_{ref}$ is the temperature of the reference state (usually taken as the ambient temperature) and $T$ the actual temperature (or the interval temperature in the case of the GCC). This curve allows for screening of the potential power production of a given process, as the integral area below the curve (shaded area in Figure 9b) represents the maximum theoretical amount of mechanical power that might be produced from the process excess heat by means of a Carnot cycle. However, this overestimates the power production by, for example, a steam power cycle from the excess heat, as losses in heat transfer and non-idealities of the steam cycle are not accounted for. Nevertheless, it can be used as an easily computable and comparative measure between different process alternatives and their respective GCC representations.
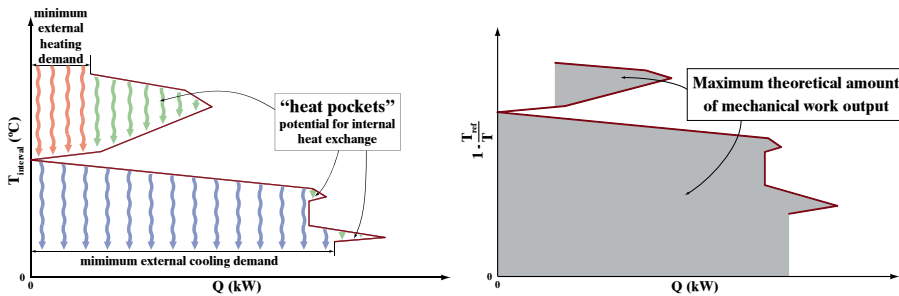


*Figure 9: a) Grand Composite Curve illustrating external heating and cooling demand and internal heat exchange potential, b) Carnot-based Grand Composite Curve (example taken from (Smith, 2005)).*

Another useful tool in pinch technology – in particular for analyzing the integration of certain sub-processes into the remainder of an industrial plant – is the so-called background/foreground analysis. The sub-process to be analyzed is represented by a separate curve (foreground) while all heat streams of the remaining plant form the background. Using this kind of representation, it is possible to determine the optimal layout of the foreground process for a given background. Within the SNG production process, this tool can be effectively used to analyse alternative technologies for a certain sub-process, but, in particular, for optimizing the steam power cycle to produce heat and power from the available

excess heat of the SNG production process. Figure 10 illustrates an example of such a background/foreground analysis of a steam power cycle. The solid curve represents the background process, while the dashed line represents the CHP steam cycle with its various pressure levels and internal heating. The example actually represents a SNG production process integrated to a CHP steam power plant. The background curve represents all heat streams of the SNG production process and the flue gas heat and district heating load of the CHP plant, while the foreground curve represents the steam cycle. In Figure 10 an additional staged dotted curve is added, representing the power produced by the different turbine sections of the steam power cycle. The temperature stages of the dotted curve correspond to the inlet, outlet and intermediate pressure levels of the steam turbine, the width of each stage corresponding to the thermodynamic power output of each stage. The dotted curve of steam turbine power is balancing the heat streams of the steam cycle (dashed curve) as a logical consequence of the overall energy balance: the heat taken up by the steam cycle is partially converted to power in the turbine, while the remainder of the heat is supplied to the background process. This representation gives a more realistic notion about the potential for power production by heat recovery than the Carnot-based evaluation, however, it requires a more detailed analysis.
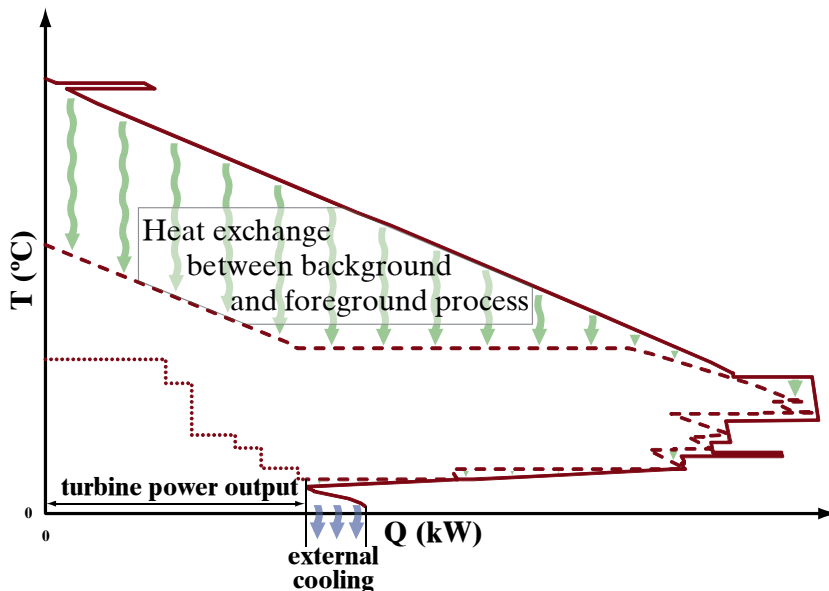


**Figure 10:** *Illustrative background/foreground GCC representation with a steam power cycle as foreground process.*

It is possible to specify the integration problem of the steam network for power production with the background process as a mixed integer linear programming (MILP) optimization problem (Maréchal & Kalitventzeff, 1997), but in this work the integration has been done manually. The concept of heat-cascading is applied to the integration, maximising the steam production at high pressure in the first place and using steam extractions at the lowest pressure level possible to provide heat to the background process. Graphically, this corresponds to minimizing the area in between the background and foreground curves in Figure 10.

### 3.2.2 Integration with Existing Infrastructure

In this work, pinch analysis is used for the thermal integration of the SNG process steps, and the efficient recovery of process heat to produce steam for turbine-generated power is in focus. Since excess heat is a significant by-product of the SNG process, it is important to efficiently use this heat to render the process economically viable. In this regard, the concept of extending existing CHP steam power plants based on circulating or bubbling fluidised bed (CFB/BFB) technology to integrate the SNG process (Thunman et al., 2007) is especially promising. This type of process integration not only applies to thermal integration by heat exchange according to the pinch methodology, but also to integration aspects on a construction level. In particular, the heat balance between the combustion and gasification chamber and changes in the steam flow through the turbine have to be considered and further analysed. Figure 11 illustrates a schematic chart of the integration between the SNG process and steam power plant.
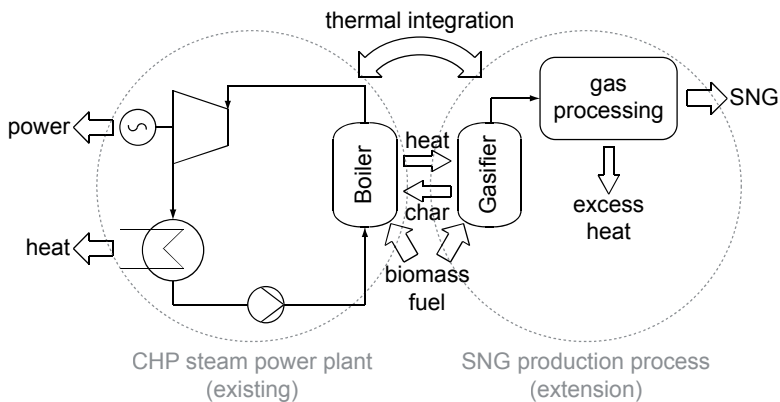


***Figure 11:*** *Schematic process layout for the integration of SNG production with an existing CHP steam power plant.*

The balance between boiler and gasifier needs to be controlled by the circulation of bed particles between the two reactors. The fuel feed to the boiler is altered as additional fuel is supplied in form of non-gasified char. As the boiler provides heat to the gasification unit, its steam production decreases, thereby reducing the steam flow through the turbine. This however, may be counteracted by increased thermal integration between the two processes – thereby making use of the available excess heat from the SNG process to produce steam.

## 3.3   Process Evaluation

In order to identify the most promising process alternatives, it is necessary to define specific criteria for ranking these options. This is done both from a thermodynamic viewpoint aiming at the maximum production of useful products based on a certain feedstock, as well as from an economic viewpoint in order to generate economically viable alternatives.

### 3.3.1   Performance Indicators

Thermodynamic performance indicators may be defined in numerous ways. Three indicators commonly used within production processes of biofuels based on biomass are cold gas efficiency, overall energetic efficiency, and overall exergetic efficiency. A qualitative overview of the advantages and drawbacks of these indicators is provided in Table 4. The cold gas efficiency $\eta_{CG}$ is defined as the energetic value of the resulting product in relation to the thermal input of the feedstock. Applied to the SNG process – using the lower heating values as a basis – this translates into:

$$\eta_{CG,LHV} = \frac{\dot{m}_{SNG} \cdot LHV_{SNG}}{\dot{m}_{fuel} \cdot LHV_{fuel}} \tag{5}$$

where $\dot{m}$ represents the mass flow and $LHV$ the lower heating value of SNG and fuel input, respectively. The thermal efficiency $\eta_{th}$ relates the thermal energy content of all useful products and services to the thermal energy input:

$$\eta_{th} = \frac{\sum_i \dot{m}_i \cdot LHV_i + \sum \dot{Q}^-}{\sum_k \dot{m}_k \cdot LHV_k + \sum \dot{Q}^+} \tag{6}$$

where $\dot{m}$ and $LHV$ are the mass flow and heating value of fuel(s) $i$ produced from and fuels $k$ used as input into the process, respectively. $\dot{Q}^-$ and $\dot{Q}^+$ denote useful thermal heat leaving or entering the system.

According to (Tsatsaronis, 2007), the exergetic efficiency $\varepsilon$ – neglecting potential and kinetic energies/exergies – can be defined as

$$\varepsilon = \frac{\sum_i \dot{m}_i \cdot (e_i^{PH} + e_i^{CH})}{\sum_j \dot{m}_j \cdot (e_j^{PH} + e_j^{CH})} \tag{7}$$

where $e^{PH}$ and $e^{CH}$ denote the specific physical and chemical exergy of the inputs $i$ and products $j$ of the system under consideration. The exergy value of a stream is always related to a reference state – usually the conditions in the environment – and represents the maximum amount of mechanical work that theoretically can be produced from that stream.

**Table 4:** *Process performance indicators available for biofuel production processes and their advantages and drawbacks.*

| Performance indicator | Advantages | Disadvantages |
|---|---|---|
| **Cold gas efficiency** (fuel conversion efficiency) | • Easy to calculate • Clear idea of fuel yield | • Does not account for by-products • No information about the overall process efficiency |
| **Thermal/energetic efficiency** (1st law efficiency) | • Possible to account for by-products • Relatively easy to calculate | • Detailed information about system boundaries necessary • Limited information about the overall process efficiency |
| **Exergetic efficiency** (2nd law efficiency) | • Possible to account for by-products • Information on overall process efficiency | • Efficiency related to a reference state (definition necessary) • Detailed information on system boundaries necessary |

Note that these indicators are not directly comparable but are dependent on the definitions of system limits; in addition, they often have to be analysed in combination with an economic framework to obtain a realistic overview. By-products and services, such as electricity and heat production, is often accounted for in varying ways by different studies, thereby making a comparison between different process alternatives difficult. A clear definition of system boundaries and background system assumptions is necessary in order to arrive at a fair comparison between alternative pathways for fuel production. This work aims at a concise way of defining performance indicators.

### 3.3.2 Energy Market Scenarios and CO₂ Consequences

To identify economically robust SNG production alternatives, the Energy Price and Carbon Balance Scenario (ENPAC) tool (Harvey & Axelsson, 2010) may be used. This tool allows the determination of consistent future energy market scenarios based on fossil fuel price and $CO_2$ emission charge estimates. Using the extreme values of projected future costs of these two entities, the tool uses built-in energy market models to determine prices – and the willingness to pay, respectively – for different services and products, including district heat, wood biomass and biomass-based transportation fuels. Even marginal electricity production technologies are determined, allowing for a systematic evaluation of $CO_2$ emission consequences of processes delivering multiple products and services such as the SNG process. Using the ENPAC tool to map the range of potential future energy market scenarios, viable SNG production process alternatives performing well from both an economic and $CO_2$ savings perspective for a range of scenarios can be identified. The influence of process changes on economic performance and $CO_2$ emission consequences may also be analysed against the background of different scenarios. The scenarios may be adapted to different time frames – currently ranging until 2050 – making them even useful to identify process alternatives that are attractive over a longer time frame. The application of these tools for a profound evaluation of the SNG production process is part of future work, while the results presented in this thesis are mainly restricted to pinch-based evaluations and process efficiency.

# 4 Results & Discussion

A base case process design was defined to serve as a basis for future evaluation of process modifications. The drying of feedstock has been evaluated in detail including its integration into the SNG process. The integration with an existing CHP steam power plant for the efficient use of the excess heat of the SNG process, as well as alternative options for using gasification technology to replace fossil fuels within the power sector, have been investigated. The results of these studies – treated in papers II to IV – are presented and discussed in this chapter.

## 4.1 Base Case Process Design

For the base case process design indirect gasification in a circulating fluidised bed was chosen, since this technology was pointed out by several former studies as being the most favourable for SNG production due to the high initial methane content of the product gas. In addition, this technology may be integrated with the existing energy conversion infrastructure in the form of steam power boilers, as previously discussed. The drawback of the indirect gasification technology is the high tar content of the product gas, a condition requiring additional treatment. A chemical looping reforming (CLR) step was assumed for the tar reforming. This is not yet a mature technology, but preliminary experimental tests conducted at Chalmers (Lind et al., 2010) have shown promising results; this overall process integration study may be used to further evaluate the potential of CLR to improve the overall efficiency of the SNG process. A filter and wet scrubber are added to remove particles, ammonia and to some extent sulphur compounds. Two amine-based $CO_2$ separation steps – that will also remove the $H_2S$ present in the product gas – are assumed, as well as two stages of isothermal methanation. The second methanation step mainly serves to reduce the CO concentration down to levels necessary for grid injection. A compression stage prior to the second methanation stage ensures the necessary pressure for final SNG delivery ; this pressure is set to

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10 bar necessary to inject the natural gas into a local network. For the final upgrading of the raw SNG, a membrane unit for $H_2$ removal and a triethylene glycol wash for drying purposes are added. A schematic flowsheet of the basic process design is illustrated in Figure 12 and details on the key modelling assumptions for the different units are given in the Appendix (Table 11).
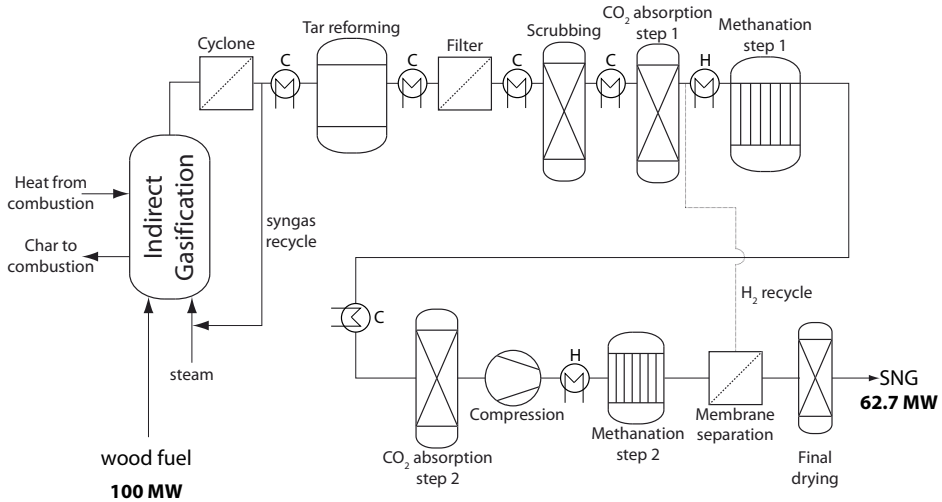


***Figure 12:*** *Flowsheet of the base case SNG production process.*

The process is designed for a dry fuel input (20 wt-% moisture) corresponding to 100 $MW_{LHV}$. A part of the syngas is recycled in order to reduce the amount of steam necessary for fluidisation. The calculated dry gas composition after gasification is given in Table 5. The SNG production of this process amounts to 62.7 $MW_{LHV}$ when assuming a dry biomass input at 20 wt-% moisture prior to gasification (Paper III). This results in a cold gas efficiency of 62.7% on a dry fuel basis. This, however, only represents the energy output in the form of SNG compared to the dry fuel input. The heaters (H) and coolers (C) illustrated in Figure 12 do not represent actual heat exchangers but only indicate that heat excess/supply from streams is available/necessary. Heat demand or excess heat from the different process steps is not illustrated. The way heat is supplied or used is not defined a priori, but all heat stream data is collected and compiled to make a pinch analysis.

***Table 5:*** *Calculated dry gas composition of the raw product gas based on the ASPEN Plus model (Paper III).*

| Component | Unit | Value | Component | Unit | Value |
|---|---|---|---|---|---|
| CO | vol-% | 22.4 | COS | ppm | 64 |
| $H_2$ | vol-% | 39.3 | $C_4H_4S$ | ppm | 24 |
| $CO_2$ | vol-% | 23.6 | $CH_4S$ | ppm | 24 |
| $CH_4$ | vol-% | 10.6 | $NH_3$ | vol-% | 0.12 |
| $C_2H_4$ | vol-% | 3.56 | HCN | ppm | 48 |
| $N_2$ | vol-% | 0.29 | NO | ppm | 48 |
| tars ($C_6H_6O$, $C_{10}H_8$, $C_{16}H_{10}$) | $g/Nm^{3}$ [1] | 1.28 | HCl | ppm | 89 |
| $H_2S$ | ppm | 129 | Total dry gas flow | $Nm^3/h$[1] | 39.4 |

[1] at 0°C and 1 bar

Figure 13 represents the resulting Grand Composite Curve (stream data is given in the Appendix - Table 12) for the base case process design (without any drying step included), showing a large amount of excess heat available from syngas cooling and methanation. It also shows that it is, for example, possible to supply the heat necessary for the $CO_2$ separation reboiler internally by means of heat transfer from the methanation unit and from syngas cooling.
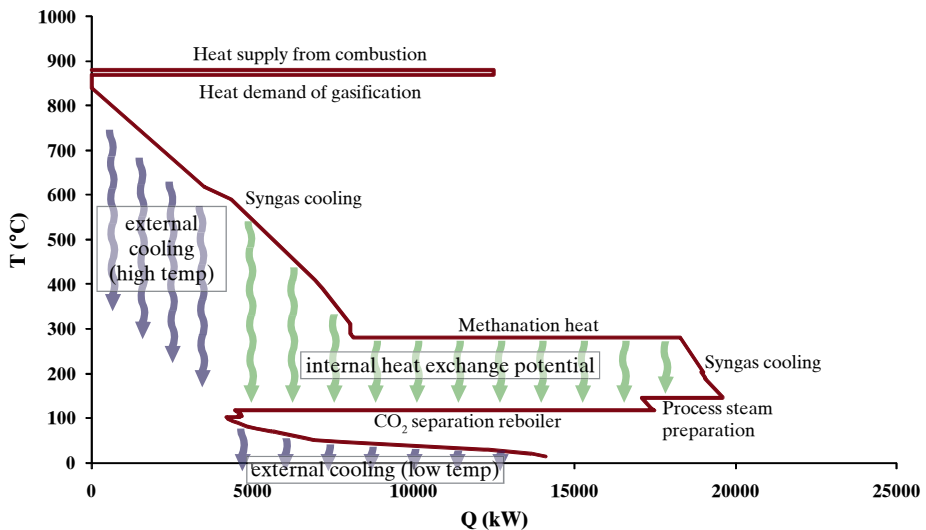


***Figure 13:*** *Grand Composite Curve for the base case SNG process with indication of the major heat sinks/sources.*

The horizontal lines at high temperatures represent the heat demand of the gasification reaction at the gasification temperature (850 °C for the base case), as

well as the supply of this heat from the combustion reactions in the separate boiler operating at higher temperatures (900 °C). In reality, this heat is transferred by circulating bed material heated up in the combustion reactor and releasing heat in the gasification reactor.

The large amount of excess heat at high temperature levels needs to be efficiently converted to valuable energy services, such as electricity and district heating. Table 6 gives the basic performance data for the process and an estimation of the power production potential of the SNG process assuming a maximum heat recovery and conversion of the heat into electricity based on the Carnot efficiency. As discussed in Section 3.2.1, this indicates the maximum amount of electric power that may be theoretically produced from the process heat. Referring to Figure 13, this would imply making use of the heat pocket – according to the illustration used for the internal heat exchange – to generate power. On a practical level, the reaction heat from methanation would be used to generate steam, while extracting steam from a turbine would be necessary for driving the $CO_2$ separation reboiler and for generating process steam. It has to be kept in mind that 7.3 MW of mechanical power estimated to be produced by the process overestimates the real potential and that a more detailed integration analysis is necessary to obtain a more precise estimate.

*Table 6: SNG process performance data.*

| Parameter | Unit | Value |
|---|---|---|
| Biomass fuel input (20 wt-% moisture) | [$MW_{LHV}$] | 100 |
| Heat supply from combustion | [MW] | 9.4 |
| SNG output | [$MW_{LHV}$] | 62.7 |
| non-gasified char to combustion unit | [$MW_{LHV}$] | 26.1 |
| Recoverable process excess heat | [MW] | 14.1 |
| Heat losses[1] | [MW] | 6.5 |
| Carnot-based power production potential from recoverable process excess heat[2] | [MW] | 7.3 |

[1] including losses in sensible heat (e.g. char and ash)
[2] as detailed in Section 3.2.1 ($T_{ref} = 20$ °C)

The overall energy balance is complicated further by the fact that the gasification unit is supplied externally with heat from a combustion unit, while at the same time, non-gasified char from the gasifier is returned to the combustion side acting

as additional fuel (c.f. Figure 4 in Section 2.1.2). Therefore, the figures given in Table 6 only provide an approximate estimation of the overall process performance, emphasising the need to define appropriate performance indicators.

The conversion of excess heat to mechanical power requires the integration of a steam cycle with the process. In addition, a combustion unit to provide heat for the gasification is necessary, making the integration with an existing combustion unit to produce power a promising alternative, as demonstrated in Section 4.3.1. Another important application for available excess heat within the SNG process is drying moist wood biomass to the desired moisture content prior to gasification. The results of these investigations are presented in the following section.

## 4.2  Case Study on Integrated Drying

Three different drying technologies for thermal integration within the SNG process have been investigated. The fuel was assumed to be dried from its initial moisture content of 50 wt-% to 10 wt-% prior to gasification. An integration of the gasification with an existing boiler was assumed, both fed with a thermal load of 100 $MW_{LHV}$ of biomass. The process heat from the SNG process for low-temperature air and steam drying, as well as flue gases from the combustion unit for drying in a flue gas dryer, were assumed to be available. The operating conditions of the drying technology were selected based on performed sensitivity analysis and pinch analysis for optimal thermal integration with the SNG process. Figure 14 illustrates the schematic flowsheet for the various drying technologies and the temperature levels at which heat may be supplied and recovered, respectively. The temperatures indicated are specific to the analysed case (Paper II) and may differ based on the specific background process.
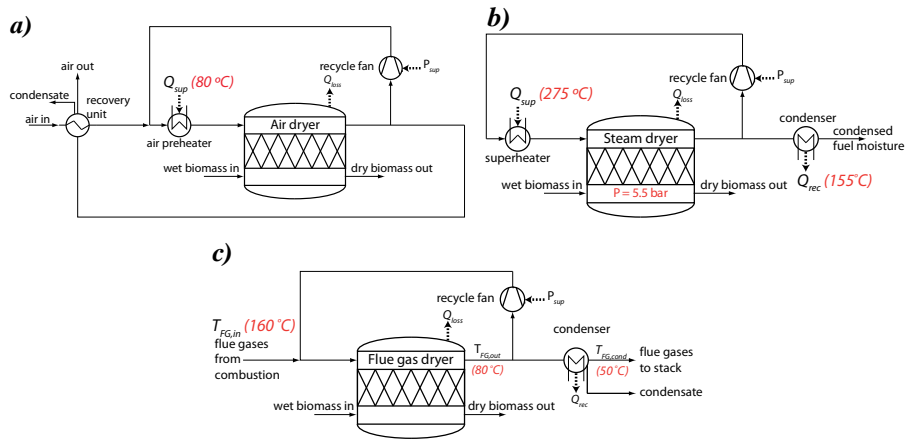
**Figure 14:** *Drying technologies investigated for integrated feedstock drying. a) low-temperature air dryer; b) steam dryer; c) flue gas dryer (adapted from Paper II).*

Figure 15 illustrates the integration of steam and low-temperature air drying with the SNG process. As heat streams from the combustion process are not included in Figure 15, the flue gas dryer is not represented. No detailed modelling of the combustion and power generation was conducted in Paper II, therefore flue gases were assumed available at 160ºC, representing a common flue gas temperature level after air preheating. The mass flow of flue gases was determined based on the thermal size of the boiler and the air-to-fuel ratio.
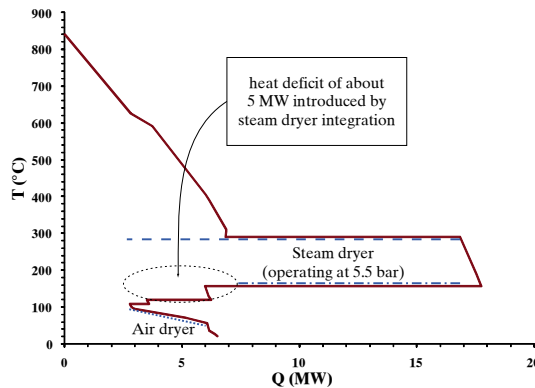


**Figure 15:** *Illustration of the integration opportunities for steam drying and air drying within the SNG production process. Solid curve – Grand Composite Curve of SNG process; dashed curve – heat demand for steam drying; dash-dotted curve – recoverable heat from steam dryer condenser; dotted curve: air dryer heat demand(based on Paper II).*

The steam dryer integration results in an internal heat deficit of about 5 MW within the SNG process that would have to be covered by another heat source such as steam extraction from the associated steam power cycle. The heat deficit of the steam dryer is mainly due to heat losses from the dryer amounting to 5% of the thermal dryer load, as well as to the fact that the solid biomass is heated to saturation temperature – this part of the heat not being recoverable in the steam condenser. At first sight, the heat provided to the solid biomass during the drying step appears to be a loss; however, the gasification heat demand decreases accordingly. This has, however, not been accounted for in this background/foreground analysis in which the SNG production process constituting the background was assumed to remain static as a simplification. Table 7 presents the amount of dry fuel that may be supplied by means of the different technologies when only considering the excess heat from the SNG process and the flue gases. A comparison with the SNG process performance using external drying – requiring additional fuel supply – is made. Drying all biomass with the help of an external stand-alone dryer results in a drop of conversion efficiency from the biomass fuel input to SNG (cold gas efficiency) of 7.4 percentage units.

*Table 7: Performance parameters for the integrated drying alternatives evaluated (Source: Paper II).*

| Parameter | Unit | Air dryer | Steam dryer | Flue gas dryer |
|---|---|---|---|---|
| Dry fuel supply necessary for SNG process | kg/s | 5.78 (for all alternatives) | | |
| Drop in cold gas efficiency for SNG process in case of external drying | % | 7.4 (for all alternatives) | | |
| Dry fuel supply possible by thermal integration | kg/s | 1.58 | 5.51 | 2.18 |
| Percentage points gained on cold gas efficiency by thermally integrated drying | % | 1.8 | 6.7 | 2.5 |
| fraction of efficiency drop recovered | % | 24.3 | 90.5 | 33.8 |

According to the study, this drop can be best compensated for by the steam drying system. Both flue gas drying and air drying still to a substantial degree require external drying. The heat deficit caused by the steam dryer (cf. Figure 15) has not been accounted for in this study. Theoretically, it would be possible to combine all three technologies as they are not competing with each other for heat sources, thereby covering all dry fuel supply for the SNG process and, potentially, even part of the boiler fuel demand; however, from a practical view point, this is not realistic. Consequently, in order to get an overall picture of the integration of the drying

process, it is necessary to also evaluate the heat streams within the associated steam power cycle, as done in Paper III. These streams can serve as heat sources for drying and as sinks for converting recoverable excess heat into mechanical work output.

## 4.3   Integration with Existing Power Plants

As previously discussed, the use of existing infrastructure can be a positive vector to promote second generation biofuels and renewable energy in general. The results of studying the integration of biomass gasification technology with existing heat and power production infrastructure will now be presented with a focus on SNG production, but also on illustrating options for gasification within the power sector – namely natural gas combined cycle power plants.

### 4.3.1   Integration of SNG Production with Existing Steam CHP Power Plants

The base case SNG process has been considered for integration with an existing biomass CHP steam power plant, as described in Section 3.2.2 (cf. Figure 11). The thermal input to both the power plant and the gasification unit was assumed to be $100\ MW_{LHV}$. A schematic flowsheet of the CHP steam power plant is illustrated in Figure 16.
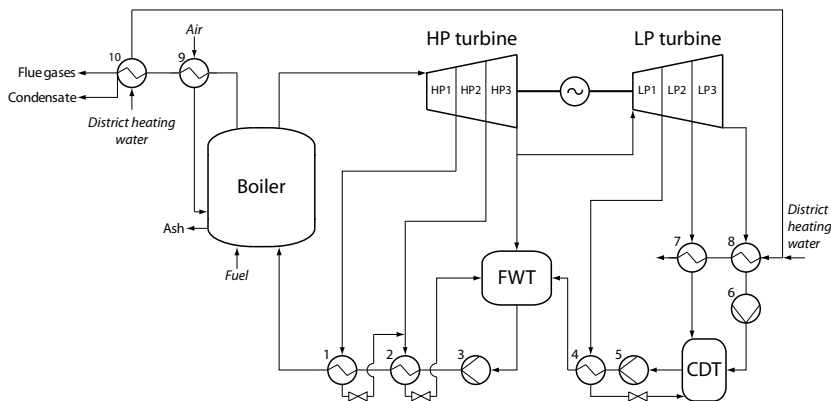


***Figure 16:*** *Schematic flowsheet of the biomass CHP plant design.*
*1 - high temperature feedwater preheater, 2 - low temperature feedwater preheater, 3 - feedwater pump. 4 - condensate preheater, 5&6 - feedwater pump, 7 - high temperature DH condenser, 8 - low temperature DH condenser, 9 - air preheater, 10 - flue gas condenser, FWT - feedwater tank, CDT - condensate tank*

The steam turbine has two pressure sections – a high (HP) and a low pressure (LP) section – each using three steam extractions. The extractions are used for internal preheating to increase the overall efficiency, and to produce district heat from condensing the last two steam extractions of the low pressure turbine section. A heat stream representation of the power plant, also indicating the power produced in the turbines, is illustrated in Figure 17. The solid line represents the flue gas heat from the boiler, the heat load from district heating water, as well as air preheating. The steam cycle heat streams are represented by the dashed curve, the upper part being the evaporation and superheating in the boiler. The ridged part of the dashed curve represents the internal preheating steps within the steam cycle, while the two lower horizontal lines represent the load in the steam condensers to produce district heat (units 7 and 8 in Figure 16). The dotted line is an illustration of the power production in the different turbine segments (HP1-3 and LP1-3). The temperature level of each turbine section represents the corresponding pressure of the outlet. As illustrated in Figure 17, the heat streams of the air preheating and district heat produced by condensing the flue gases (units 9 and 10 in Figure 16) have not been considered for integration with the SNG production process, but are considered unchanged.
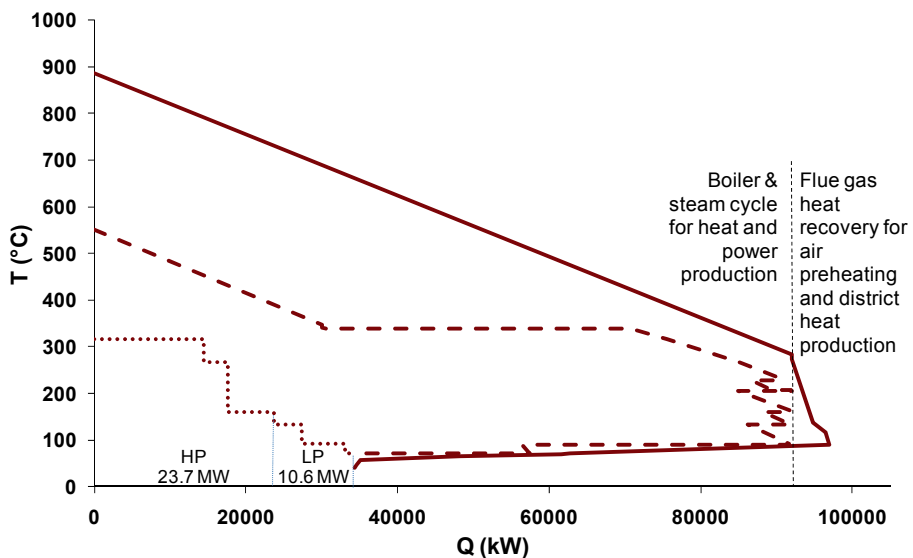


***Figure 17:*** *Pinch representation of biomass CHP steam power plant. Solid curve: heat streams from boiler and district heating system, dashed curve: steam cycle heat streams, dotted curve: power production in turbine stages (Paper III).*

For the SNG process to be integrated with the CHP steam power plant, two drying technology alternatives – steam drying (case 1) and low-temperature air drying (case 2) – have been evaluated. As the potential for flue gas drying (described in Section 4.2) was rather low, and as the flue gas heat recovery was used for both air preheating and for supplying district heat in the CHP steam power plant investigated, flue gas drying was discarded as a drying alternative.

Different levels of thermal integration between the steam power cycle and the SNG process have been investigated. An illustration of these two different levels is given in Figure 18. The figure serves as an illustrative example with a simplified representation of the steam cycle. The first level of thermal integration investigated (case 1A and 2A) is a balancing integration that only maks use of the high temperature excess heat from the SNG process for increasing the steam production. Internal heat recovery within the SNG process is assumed for the rest of the available process excess heat. The second level of integration investigated (cases 1B and 2B) makes use of the heat pockets by means of heat-cascading. Excess heat from the SNG process is used for high-quality steam generation. As a heat deficit within the SNG process is produced, internal heat recovery is not possible any more, and lower quality steam from the steam cycle has to be used to cover the SNG process heat demand.
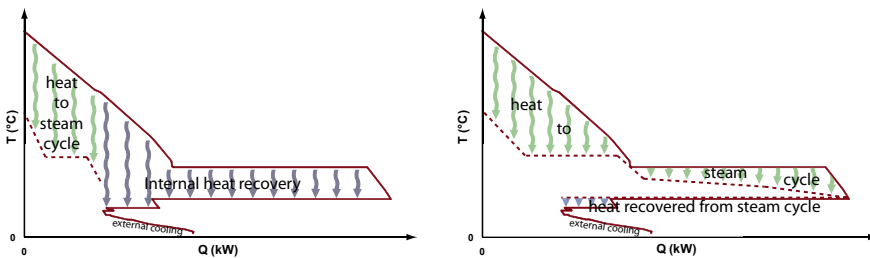


**Figure 18:** *Illustrative example of the two levels of thermal integration between the SNG process and the steam cycle. a) balancing integration, b) maximum integration using heat-cascading. Solid curve: SNG heat stream representation, dashed curve: simplified steam cycle heat stream representation.*

The Grand Composite Curves (GCC) of the SNG production process and the two drying alternatives are illustrated in Figures 19 and 20, respectively. The light grey shaded areas of Figures 19 and 20 represent the balancing integration cases in which the steam power cycle is integrated with the SNG process to balance the external heat excess/demand alone. The dark grey shaded area represents the

maximum possible thermal integration between the two processes by using the concept of heat-cascading.



**Figure 19:** *GCC for the SNG process alternative with steam drying. Light grey shaded area: case 1A – balancing integration, dark grey shaded area: case 1B – maximum integration (Paper III).*



**Figure 20:** *GCC for the SNG process alternative with air drying. Light grey shaded area: case 2A – balancing integration, dark grey shaded area: case 2B – maximum integration (Paper III).*

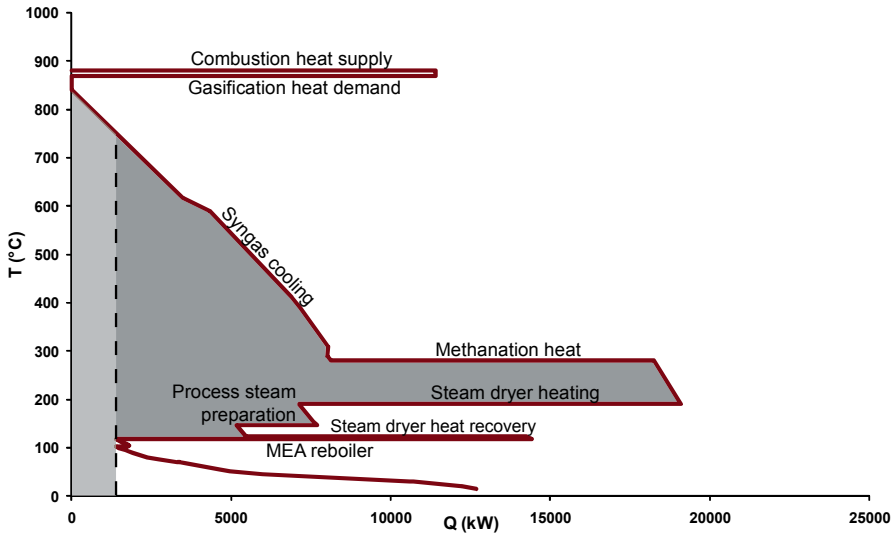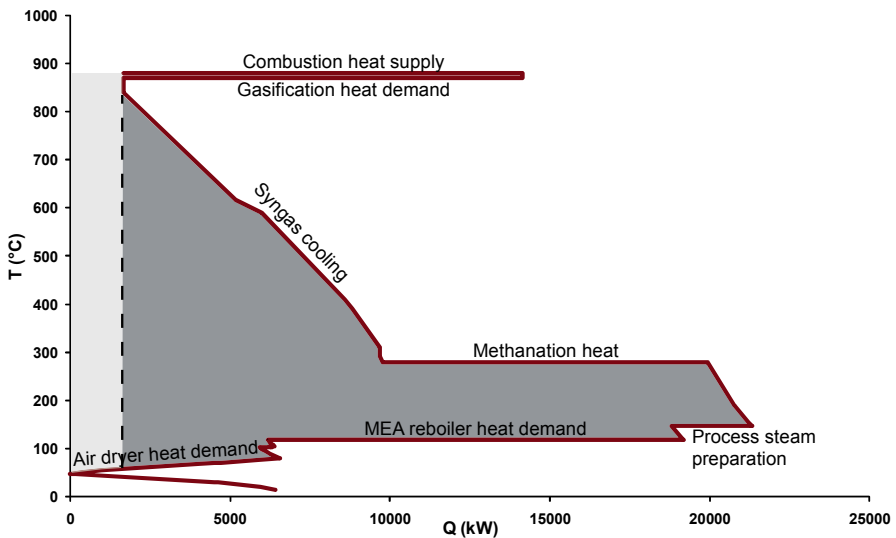As an example, Figure 21 illustrates the increased level of integration of the SNG process with air drying (case 2B). The solid line represents the heat streams from both the CHP steam power plant and the SNG process while the dashed line illustrates heat streams of the steam cycle integrated to a maximum extent with the SNG process, thereby making use of the large heat pocket represented by the dark grey shaded area in Figure 20. As the existing CHP steam power plant is associated with considerably larger heat loads, the heat pocket of the SNG process only appears as a relatively small nose in Figure 21.
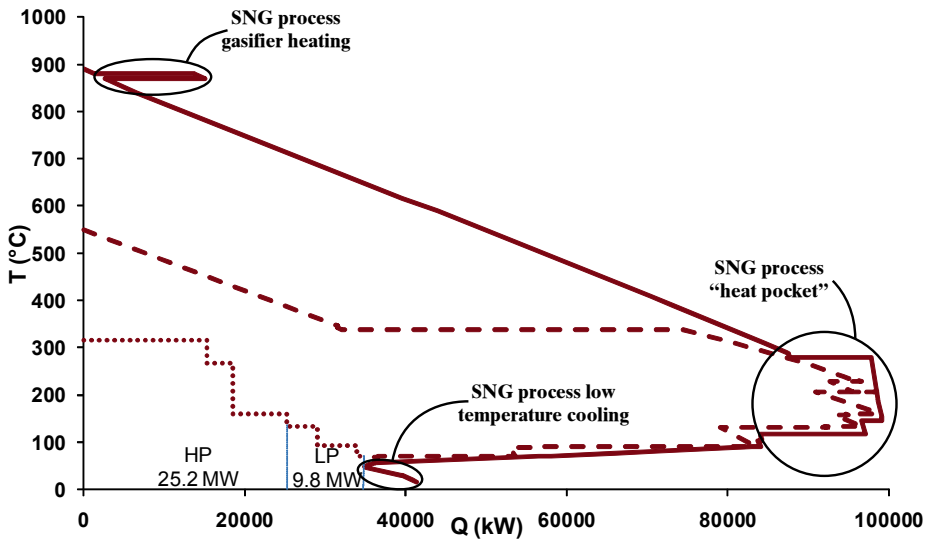


*Figure 21:* *Pinch representation of the increased integration case between the SNG process with air drying and the CHP steam power plant (case 2B). Solid curve: heat streams from boiler, SNG process and district heating system, dashed curve: steam cycle heat streams, dotted curve: power production in turbine stages (Paper III). The process streams representing the SNG process are highlighted.*

The power production in the turbines increases slightly for case 2B illustrated in Figure 21 compared to the CHP stand-alone operation (c.f. Figure 17), but the overall process' net electricity production decreases due to a higher internal consumption for the integrated process. This net production decrease is mainly attributable to the decreased energy supply to the steam cycle by the boiler. The boiler now supplies heat to both the steam production and the gasification reaction; because of the constant boiler load assumed, the steam production is lower than during the stand-alone operation. However, since non-gasified char

from the gasification is used as additional fuel in the boiler, the external fuel supply also goes down.

Figure 22 illustrates the decrease in heat and power production from the steam cycle because of the integration of the SNG production process. It also clearly illustrates the positive effect of an increased integration between the two processes on the electricity production. For the SNG process using steam dryer (cases 1A & B), a relative increase of 11.8 % can be obtained by maximising the thermal integration, while the increase for the SNG process with air drying (cases 2A & B) amounts to 21.9 %. For both alternatives, the increased thermal integration leads to higher production of electricity at the cost of lower production of district heat.
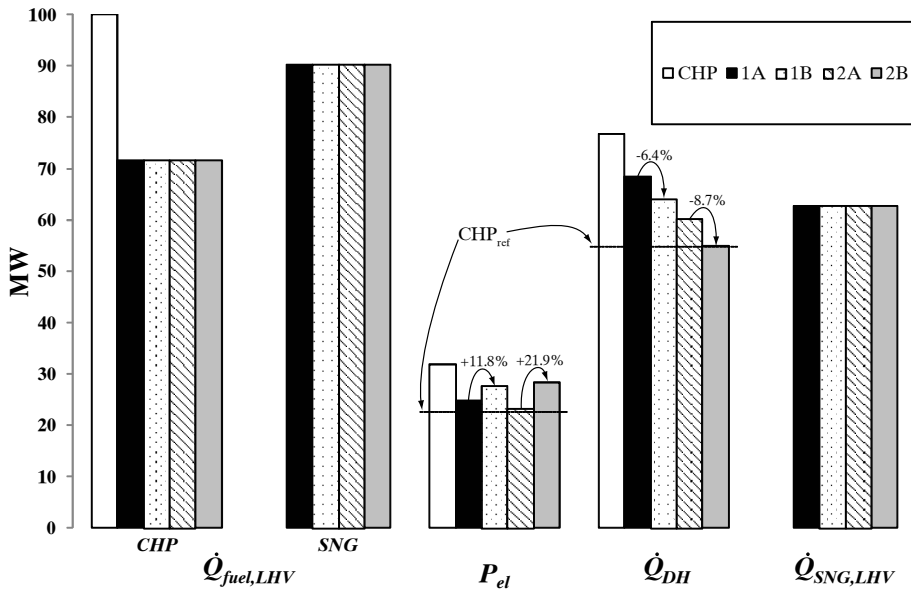


**Figure 22:** Fuel input ($\dot{Q}_{fuel,LHV}$), electricity ($P_{el}$), heat ($\dot{Q}_{DH}$) and SNG ($\dot{Q}_{SNG,LHV}$) production for the integration cases studied (case 1A & B and 2A & B) and the stand-alone CHP steam power plant (Paper III). $CHP_{ref}$ indicates the theoretical amount of electricity and district heat produced from the biomass input to the CHP boiler alone for the four cases of integrated SNG production.

Figure 22 also shows that all SNG process alternatives are net electricity and district heat producers because the output is higher than the theoretical case of combined heat and power produced by a stand-alone CHP plant with corresponding biomass fuel input (indicated by the dashed lines in Figure 22

($CHP_{ref}$)). It is possible to isolate the electricity and district heat production from the SNG process by assuming constant conversion efficiencies for the CHP plant both for the integrated and stand-alone cases. For the electricity production allocated to the SNG production $P_{el,SNG}$, this can be expressed as

$$P_{el,SNG} = P_{el} - \dot{Q}_{fuel,CHP} \cdot \eta_{el,CHP} \tag{8}$$

where $P_{el}$ is the overall net electricity production, $\dot{Q}_{fuel,CHP}$ the fuel supply to the CHP boiler and $\eta_{el,CHP}$ the electrical efficiency of the stand-alone CHP plant. Accordingly, the district heat production from the SNG process $\dot{Q}_{DH,SNG}$ can be estimated as

$$\dot{Q}_{DH,SNG} = \dot{Q}_{DH} - \dot{Q}_{fuel,CHP} \cdot \eta_{q,CHP} \tag{9}$$

where $\dot{Q}_{DH}$ is the overall district heat production and $\eta_{q,CHP}$ the heat efficiency of the stand-alone CHP plant, relating district heat production to thermal fuel input. Using these numbers, the beneficial effect of increased thermal integration on the performance of the SNG production process becomes even more evident: in the case of the steam dryer (1A & B), higher thermal integration leads to increased electricity production from the SNG process $P_{el,SNG}$ of a factor of 2.5, while in the case of the air-dryer it increases by a factor of more than 10 (2A & B). Table 8 gives an overview of the performance of the four different cases investigated.

*Table 8: Performance indicators and changes in fuel supply and output of the heat and power cycle for the integration cases studied (Paper III).*

|  | CHP | Case 1A | Case 1B | Case 2A | Case 2B |
|---|---|---|---|---|---|
| $P_{fuel,CHP}$ [MW$_{LHV}$] | 100 | 71.55 | 71.55 | 71.55 | 71.55 |
| $P_{fuel,SNG}$ [MW] | - | 90.33 | 90.33 | 90.33 | 90.33 |
| $P_{el}$ [MW] | 31.74 | 24.72 | 27.63 | 23.19 | 28.28 |
| $Q_{DH}$ [MW] | 76.81 | 68.42 | 64.06 | 60.11 | 54.91 |
| $P_{SNG}$ [MW$_{LHV}$] | - | 62.7 | 62.7 | 62.7 | 62.7 |
| $\eta_{tot}$ [%] | 108.6 | 96.3 | 95.4 | 90.2 | 90.1 |
| $P_{el,SNG}$ [MW] | - | 2.0 | 4.9 | 0.5 | 5.6 |
| $Q_{DH,SNG}$ [MW] | - | 13.5 | 9.1 | 5.2 | 0 |

When allocating all electricity production to the biomass fuel input to the CHP steam power plant alone, the electricity production efficiency in the cases with increased integration reaches 38.6 % for the SNG process with steam drying (case 1B) and and 39.5 % for air drying (case 2B), respectively. This can be compared to the CHP stand-alone electricity production efficiency of 31.74 %. Of course, this

number should be interpreted with precaution since the additional fuel supplied to the power boiler in the form of non-gasified char and increased steam production by thermal integration – both energetically based on the fuel input into the SNG process – are not accounted for in these numbers. The problem of allocating fuels, products and services requires a rigorous definition of performance indicators in order for this process to be comparable to others on a common basis.

### 4.3.2 Integration of Biomass Gasification in Combined Cycle Gas Turbine (CCGT) Power Plants

As an example of alternative applications of biomass gasification within the existing infrastructure, the replacement of fossil natural gas within combined cycle gas turbine (CCGT) power plants is illustrated. The amount of biomass fuel input in relation to the natural gas input in the cases investigated is relatively small for all integration alternatives, leaving the gas turbine part virtually unchanged. To evaluate the different integration alternatives and compare them to stand-alone options, a marginal electrical efficiency for the biomass fuel input $\eta_{bio}$ is defined:

$$\eta_{bio} = \frac{P - \eta_{ref} \cdot \dot{Q}_{NG}}{\dot{Q}_{bio}} \tag{10}$$

where $\eta_{ref}$ is the electrical efficiency of a reference stand alone CCGT plant, $\dot{Q}_{NG}$ is the thermal input of natural gas, $\dot{Q}_{bio}$ the thermal input of biomass and $P$ the total power production from the integrated plant. The idea behind Eq. (4) is to assume that the efficiency of the gas turbine cycle remains unchanged, and ro allocate the difference in power production between a theoretical stand-alone CCGT plant and the integrated solution with similar natural gas input $\dot{Q}_{NG}$ to the biomass thermal input alone.

The following cases have been investigated (for details refer to Paper IV):

- Reference CCGT plant (***Ref***)

- Reference stand-alone biomass CFB steam power plant (***BioSt***)

- Hybrid plant using gas turbine flue gases either as combustion air in the biomass boiler, for biomass drying, or as economiser heat source in the steam cycle (***Hyb***)

- FICFB biomass gasification integrated with a CCGT plant using either air or gas turbine flue gases to fluidise the combustion section of the gasifier (***Gasif***)

In the case of integrating a gasification unit – the focus of this discussion – product gas is used to substitute part of the fossil natural gas in the gas turbine. This blended product gas amounts to at most 5.5 % on an energy basis in the different cases investigated, allowing for a gas turbine operation that remains unchanged. From Table 9 can be seen that the marginal electrical efficiency of the gasification option is substantially higher than the electrical efficiency of the stand-alone steam plant. The highest efficiency for the gasification integration alternatives is up to 14.4 %-points higher compared to the stand-alone operation of a biomass CHP steam plant.

**Table 9:** *Electrical efficiency of the reference cases and electrical efficiency allocated to biomass use in the integrated cases (based on Paper IV).*

| Integration option | $\eta_{el,bio,\,LHV}$ $(\eta_{el})$[1] [%] |
|---|---|
| Reference CCGT (*Ref*) | 57 – 57.1 |
| Reference stand-alone steam plant (*BioSt*) | 35.5 – 37.9 |
| Hybrid combined cycle (*Hyb*) | 37.9- 41 |
| Gasification (*Gasif*) | 46 – 49.6 |

[1] For the reference cases the efficiency refers to the electrical efficiency $\eta_{el}$ for stand-alone operation, fossil based (NG) for CCGT reference case, biomass based for the steam plant reference.

An economic analysis of the performance of the different integration alternatives was conducted based on an extensive compilation of investment cost data gathered from both literature and equipment manufacturers. Based on the levelized cost of electricity (CoE) as performance indicator, the production cost for electricity from biomass are lowest for integrated gasification (Gasif) among all alternatives studied. Figure 23 shows CoE as a function of the wet fuel price for a selection of integration alternatives. The costs of production per MWh of electricity were estimated based on two different interest rate assumptions, with the gasification alternatives always being the most favourable from an economic perspective. Comparable costs for electricity production from coal range from 44 to 61 EUR/MWh which means that the co-combustion of biomass with coal is most likely the cheaper option. However, no $CO_2$ emission charge was accounted for in these cost estimations. In addition, for regions with well developed natural gas infrastructure and little or no coal in the energy supply mix, the integration of gasification with CCGT plants is a very promising alternative for the efficient introduction of renewable sources of energy into electricity production.
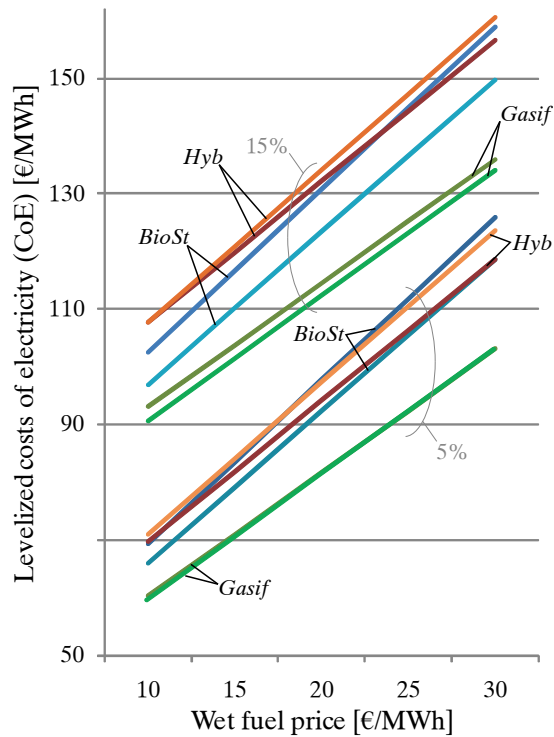
***Figure 23:*** *Cost of Electricity (CoE) in EUR/MWh as function of wet fuel cost in EUR/MWh$_{th}$ (LHV) for selected configurations (for details refer to Paper IV). Results are shown for an estimated lifetime of 25 years and discount rates of 5% (lower six lines) and 15% (upper six lines), respectively. (based on Paper IV).*

In the configurations analysed, the resulting product gas was not converted to SNG, but was rather cleaned to turbine specifications and directly used. This is possible as long as the gas turbine can accommodate the blend-in of product gas into the natural gas. To further replace natural gas, it might be necessary to convert the product gas into SNG, something that most likely might have a negative effect on electrical efficiency because of the conversion of chemical energy in the product gas to heat during the methanation step.

# 5 Conclusions

The results presented in this work clearly illustrate the importance of a sound integration approach to identify efficient SNG production process alternatives, making use of the available excess heat to produce power and deliver district heat. A basic setup for the production of SNG has been developed based on indirect gasification, which offers a high potential for integration with existing energy infrastructure. The chemical looping tar reforming stage assumed in the studies allows for efficient heat recovery from the product gas. Because of the large amount of excess heat available, the use of an amine based $CO_2$ separation technique is feasible without external heat supply.

Various alternative techniques for the drying of biomass prior to gasification have been assessed. In using available excess heat from the process, drying can be accomplished with little or no external heat supply depending on the cases studied. To optimally integrate the drying step the interaction with the CHP steam power cycle used for heat recovery and supply must be accounted for.

In particular, the integration of SNG production with an existing biomass CHP steam power plant has been investigated. An increased level of thermal integration between the two processes leads to significant increases in electricity production. Low-temperature air drying results in higher electricity production at an elevated level of thermal integration compared to steam drying, while the SNG production is unaffected by the choice of drying alternative. The production of district heat from the CHP plant was considerably reduced when integrating an SNG production process. This is in particular the case for low-temperature air drying, since this technique has a heat demand at a similar temperature level as the district heating system. From this perspective, low-temperature drying could also be seen as an opportunity for the alternative use of low temperature heat in case the demand for district heating should decrease.

Biomass gasification is a key technology with which to promote renewable energy supply. Its use to replace natural gas in combined cycle power plants shows an interesting potential to efficiently produce renewable electricity at reasonable costs, even though a conversion to SNG is not carried out in this case.

# 6 Further Work

This report is the result of the progress made so far within a PhD project and – based on these results - there are a number of aspects that still need to be investigated in greater depth, in addition to aspects that have not yet been the object of inquiry. Figure 24 illustrates the focus of further work to be done. A base case model for the SNG production process has been developed and analysed using pinch technology. A process evaluation has already been to some extent performed by estimating the SNG yield from biomass and the interaction between the steam power cycle and the SNG process when these two processes are integrated. The focus of further work will be on defining appropriate performance indicators for process evaluation and – based on these indicators and the results of the pinch analysis – to examine potential modifications to the process.
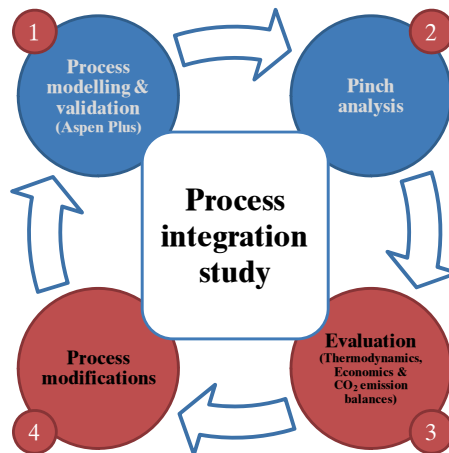


*Figure 24: Methodology representation highlighting (red) the focus points of further work.*

As Figure 24 shows, the process integration study is a continuous loop of iterations for the purpose of improving the process. This implies that – based on the results obtained from the process evaluation – key conversion steps within the SNG process that have a major influence on the performance of the process will be considered for a more detailed modelling to assess the potential for process improvements by modifying these key steps.

## 6.1   Advanced Models for Key Conversion Steps

The key step to produce SNG from biomass is obviously the gasification step which is converting solid biomass to product gas. Indirect gasification is the most promising technique with which to produce SNG because of its high inherent $CH_4$ content in the product gas. The operating conditions of the gasifier, including pressure, temperature, and gasification medium, strongly influence the composition of the product gas, in particular the $CH_4$ content and tar formation. Consequently, it becomes necessary to arrive at a model reflecting the influence of these parameters to determine promising operating regimes from an overall SNG production perspective. In addition, the char balance between the gasification chamber and the combustion chamber is an important parameter that affects the balance between the combustion heat released in the boiler and the product gas yielded by the gasification unit. In using a more detailed gasification model reflecting these aspects, it becomes possible to investigate the implications of changes in the char balance to overall system performance. In collaboration with the experimental and detailed modelling work conducted by the Energy Technology Group at Chalmers, an advanced model for energy integration purposes will be developed. This model will on the one hand allow for an investigation of the impact of changing operating parameters and eventually reactor design on the product gas composition and heat demand for the gasification, and on the other hand for an examination of the implications for the overall process design.

Similarly, the technology of tar removal is a crucial step – mainly from a technical viewpoint (tars lead to equipment fouling), but also from a heat integration perspective (tar scrubbing reduces the potential for heat recovery from product gas). The chemical looping reforming technology assumed in this study is currently undergoing development by the Energy Technology Group at Chalmers and the model used within process simulations will be updated and improved as the experimental work moves forward, in order to give a more realistic picture of the

potential of this technique for the SNG production process. A comparison to techniques that have been proven feasible within other demonstration projects will be conducted as well.

## 6.2 Process Modification and Optimisation

Besides the two process steps mentioned above, there are several technological alternatives available for the remainder of process steps to convert biomass to SNG, in particular for separating $CO_2$ after methanation. The amine based absorption assumed so far efficiently makes use of excess heat generated by the process, allowing a sound integration. The use of technologies such as membrane separation or pressure swing adsorption will increase the electricity consumption within the process, but at the same time an increased amount of process heat will be released, thereby improving the opportunities for steam generation and subsequent power production. The integration of these alternative technologies will be examined and evaluated from both a heat integration as well as an economic perspective.

## 6.3 Process Performance Evaluation

Appropriate process performance indicators of a general character will be defined and allow for the process to be optimised based on thermo-economic considerations. Based on these indicators, it will be possible to quantitatively evaluate the various SNG production process alternatives integrated with existing energy infrastructure, such as a CHP steam power plant, in comparison to a stand-alone SNG process plant.

Another important procedure for identifying process improvements is the evaluation of SNG production processes based on economic and environmental indicators. By ranking different process alternatives based on possible future energy market scenarios consistently reflecting future fuel market prices and $CO_2$ emission charges, as well as marginal electricity production techniques, will allow identifying economically robust process alternatives and their corresponding environmental performance with respect to $CO_2$ emission reductions.

# 7 Nomenclature and Abbreviations

| | |
|---|---|
| *ar* | as received |
| *daf* | dry ash-free fuel |
| *df* | dry fuel |
| $e^{CH}$ | specific chemical exergy |
| $e^{PH}$ | specific physical exergy |
| $\dot{m}$ | mass flow |
| $P$ | power |
| $\dot{Q}_{NG}$ | natural gas thermal power |
| $\dot{Q}_{bio}$ | biomass thermal power |
| *wt-%* | weight percent (mass based) |
| $\Delta H_r$ | enthalpy of reaction |
| $\eta_{bio}$ | marginal electrical efficiency for biomass fuel |
| $\eta_{CG}$ | cold gas efficiency |
| $\eta_{el}$ | electrical efficiency |
| $\eta_q$ | heat efficiency |
| $\eta_{ref}$ | electrical efficiency for reference stand-alone plant |
| $\eta_{th}$ | thermal efficiency |
| $\varepsilon$ | exergetic efficiency |
| $\rho$ | density |

**Abbreviations**

| | |
|---|---|
| BFB | bubbling fluidised bed |
| CCGT | combined cycle gas turbine |
| CFB | circulating fluidised bed |
| CHP | combined heat and power |
| CLR | chemical looping reforming |
| CoE | costs of electricity |
| DH | district heat |
| DME | dimethyl ether |
| ECN | Energy Research Centre of the Netherlands |
| ELECNRTL | electrolyte non-random two-liquid model |
| ENPAC | Energy Price and Carbon Balance Scenario |
| EU | European Union |
| FICFB | fast internally circulating fluidised bed |

| | |
|---|---|
| FT | Fischer-Tropsch |
| GCC | Grand Composite Curve |
| GHG | greenhouse gas |
| HHV | higher heating value |
| HP | high pressure (steam) |
| IEA | International Energy Agency |
| LHV | lower heating value |
| LP | low pressure (steam) |
| MEA | monoethanol amine |
| MILP | mixed integer linear programming |
| Mtoe | million tons of oil equivalents |
| PR-BM | Peng Robinson cubic equation of state with Boston-Mathias alpha function extension |
| PSA | Pressure swing adsorption |
| PSI | Paul-Scherrer Institute |
| PWS | pressurised water scrubbing |
| RD | relative density |
| SNG | synthetic natural gas |
| TEG | triethylene glycol |
| TSA | temperature swing adsorption |
| VOC | volatile organic compounds |
| WI | Wobbe index |

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# Acknowledgements

"Not everything that can be counted counts,

and not everything that counts can be counted"

Albert Einstein (1879 - 1955)

The work presented here is about putting numbers on things that definitely count for something. At this point I would like to thank everybody for the countless help and support in my quest.

First of all I would like to thank my supervisors Simon and Henrik for giving me the opportunity to work on this project and guiding me in my research. The interesting discussions and your constructive criticism are very much appreciated.

The financial support by the Swedish Energy Agency, Göteborg Energi AB and E.ON Sverige AB is gratefully acknowledged. Getting insights into the GoBiGas project and discussing different aspects of SNG production with people at Göteborg Energi is a valuable input for this project. Thank you Ingemar Gunnarsson, Lars Andersson, Malin Hedenskog and all the other people involved at Göteborg Energi.

At the Energy Technology department, a lot of practical information and help for my modelling work has been provided. Thank you, Martin, for the good time we had together both at work and everywhere else. Special thanks also to Erik Pihl who wrote an excellent article that I had the honour of contributing to.

One thing that definitely counts when doing a PhD is the working environment. I'd like to thank all the present and former co-workers at VoM for creating an excellent working atmosphere that is very inspiring and fun at the same time: innebandy, betting competitions, tyska maffian, countless discussions during fika,

Dressed-up Friday, and "En Svensk Klassiker" only gives a glimpse of the spectrum of VoM activities. Thanks in particular to the people that had to bear with me while sharing the office: Daniella, Mathias and – recently – Johan. I really enjoy(ed) your companion.

At countless occasions during my long way to and along this thesis I had to rely on the support of my family. I am very grateful for the true love and support you give me. Thank you for bringing me up the way you did and always being there for me.

And finally what really counts: Ramona, I owe you so much for always supporting me and being there when I need you. Being parent to Emil together with you is the best thing that ever happened to me. Thank you for being patient when I had periods of intensive work, and reminding me of what really counts in life at the same time. I love you.

# Appendix

**Table 10:** *Wood fuel (forest residues) properties used as input for the simulations.*

| Ultimate analysis | |
|---|---|
| C [wt-% df] | 50.30 |
| H [wt-% df] | 5.43 |
| O [wt-% df] | 41.57 |
| N [wt-% df] | 0.47 |
| S [wt-% df] | 0.04 |
| Cl [wt-% df] | 0.01 |
| Ash [wt-% df] | 2.18 |
| **Proximate analysis** | |
| Moisture content [wt-% ar] | 50 |
| Volatile matter [wt-% df] | 77.82 |
| Fixed carbon [wt-% df] | 20 |
| Ash [wt-%df] | 2.18 |
| **Heating value** | |
| LHV [MJ/kg df] | 19.54 |
| LHV [MJ/kg ar] | 8.55 |
| HHV [MJ/kg df] | 20.72 |

*Table 11: Basic modeling assumptions for key units within the SNG process base case*

| Indirect gasification unit | |
|---|---|
| Operating temperature [°C] | 850 |
| Pressure drop [bar] | 0.1 |
| Heat loss [fraction of thermal input] | 0.02 |
| Steam-to-biomass ratio [kg $H_2O$/kg dry fuel] | 0.6 |
| Pyrolysis modelling | Yield reactor based on (Thunman et al., 2001) |
| Gasification modelling | • Gibbs equilibrium reactor at operating temperature<br>• reactive species: C, CO, $CO_2$, $H_2$ & $H_2O$<br>• WGS at equilibrium<br>• 70% carbon conversion |

| Tar reforming unit (Chemical looping reformer) | | Scrubbing unit (water scrubber) | |
|---|---|---|---|
| | | $NH_3$ removal efficiency [-] | 0.99 |
| Pressure drop [bar] | 0.05 | Pressure drop [bar] | 0.02 |
| Operating temperature [°C] | 625 | Water recycle cooling temperature [°C] | 20 |
| Reactions | complete reforming of tars to CO & $H_2$ | Waste water stripper | operating a 1 bar, off-gases to be burnt in combustion boiler |

| $CO_2$ absorption (MEA unit) | | Methanation | |
|---|---|---|---|
| $CO_2$ separation efficiency | 0.95 | Operating temperature [°C] | 300 |
| Pressure drop [bar] | 0.05 | Pressure drop [bar] | 0.05 |
| Energy demand [MJ/kg $CO_2$ separated] | 3.7 (@115 °C) | Reaction modelling | Gibbs equilibrium reactor ($T_{approach}$ = 320°C) |
| Recoverable energy [fraction of energy demand] | 20% (@90 → 40 °C) | Steam addition | adjusted to obtain $H_2$/CO = 3 taking into account simultaneous WGS reaction |

| Membrane separation | | Compressors & Fans | |
|---|---|---|---|
| Inlet pressure [bar] | 10 | isentropic efficieny | 0.72 |
| Pressure drop [bar] | Permeate: 8 bar Retentate: 0.5 bar | mechanical efficiency | 0.98 |
| Split ratio [permeate/feed flow] | $H_2$:0.999 $CH_4$: 0.005 | intercooling temperature[1] [°C] | 80 - 120 |

| Pumps | | SNG delivery conditions | |
|---|---|---|---|
| pump efficiency | based on efficiency curve for water[2] | Pressure [bar] | 10 |
| mechanical efficiency | 0.98 | Temperature [°C] | 30 |

[1] in case of multi-stage compression
[2] default in ASPEN Plus

**Table 12:** *Stream data for the base case SNG process.*

| Stream type | $T_{start}$ [ºC] | $T_{target}$ [ºC] | Q [kW] | ΔT/2 [ºC] | Comment |
|---|---|---|---|---|---|
| Cold | 15 | 144 | 640 | 5 | Steam preparation for gasification 1 |
| Cold | 144 | 144 | 2523 | 2.5 | Steam preparation for gasification 2 |
| Cold | 144 | 400 | 632 | 10 | Steam preparation for gasification 3 |
| Hot | 900 | 900 | 12478 | 20 | Heat supply from combustion |
| Cold | 850 | 850 | 12478 | 20 | Heat demand for gasification |
| Hot | 850 | 600 | 3927 | 10 | Gas cooling after gasification |
| Hot | 627 | 400 | 3282 | 10 | Syngas cooling after reforming 1 |
| Hot | 400 | 200 | 2672 | 10 | Gas cooling after reforming 2 |
| Hot | 200 | 80 | 1499 | 10 | Gas cooling after reforming 3 |
| Hot | 109 | 81 | 345 | 10 | Gas cooling after reforming 4 |
| Hot | 56 | 20 | 3348 | 5 | Water scrubber recycle cooling |
| Cold | 100 | 100 | 397 | 2.5 | Waste water stripper reboiler |
| Hot | 100 | 25 | 315 | 5 | Scrubber waste water cooling |
| Hot | 56 | 40 | 1369 | 10 | Gas cooling after scrubber |
| Cold | 115 | 115 | 9009 | 2.5 | MEA 1 reboiler |
| Hot | 90 | 40 | 1802 | 10 | MEA 1 partial condenser |
| Cold | 58 | 300 | 1675 | 10 | Gas heating prior to methanation 1 |
| Cold | 15 | 103 | 21 | 5 | Steam preparation methanation 1 |
| Cold | 103 | 103 | 128 | 2.5 | Steam preparation methanation 2 |
| Cold | 103 | 300 | 22 | 10 | Steam preparation methanation 3 |
| Hot | 300 | 300 | 9992 | 20 | Methanation 1 reaction heat |
| Hot | 300 | 64 | 1338 | 10 | Gas cooling after first methanation 1 |
| Hot | 64 | 30 | 1272 | 10 | Gas cooling after first methanation 2 |
| Cold | 115 | 115 | 4008 | 2.5 | MEA 2 reboiler |
| Hot | 90 | 40 | 802 | 10 | MEA 2 partial condenser |
| Hot | 132 | 80 | 179 | 10 | Compressor cooling stage 1 |
| Hot | 157 | 100 | 201 | 10 | Compressor cooling stage 2 |
| Cold | 179 | 300 | 490 | 10 | Gas heating prior to methanation 2 |
| Hot | 300 | 300 | 150 | 20 | Methanation 2 reaction heat |
| Hot | 300 | 86 | 814 | 10 | Gas cooling after second methanation 1 |
| Hot | 86 | 30 | 396 | 10 | Gas cooling after second methanation 2 |
| Cold | 200 | 200 | 55 | 2.5 | TEG dryer reboiler |