

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Bio-SNG from Thermal Gasification - Process Synthesis, Integration and Performance

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Heat and Power Technology
Department of Energy and Environment
CHALMERS UNIVERSITY OF TECHNOLOGY

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ABSTRACT

Biomethane or synthetic natural gas (Bio-SNG) produced from gasified renewable woody biomass is a promising option for replacing fossil natural gas. The complete interchangeability with natural gas in all its conventional applications such as in the power generation, transportation and chemical industry sector is of particular interest.

This work presents results from a comprehensive process integration study of different process alternatives for Bio-SNG production from gasified biomass. The influence of the main conversion steps in the process chain – drying, gasification, gas cleaning, methanation, and gas upgrade – on the overall process performance is investigated. Process bottlenecks and both heat and material integration opportunities are highlighted. Using future energy market scenarios the energetic, economic, and carbon footprint performance of the investigated processes are evaluated from a system perspective clearly showing the sensitivity of the obtained results to underlying assumptions.

It is shown that drying of the biomass feedstock prior to gasification using excess process heat – using steam drying or low-temperature air drying technology – is an important aspect for improving the process energy efficiency. The results also indicate that indirect and direct gasification technologies perform equally well within the overall Bio-SNG production process. Existing infrastructure in the form of biomass-fired combined heat and power plants based on fluidised bed combustion technology presents interesting opportunities for integrating indirect gasification for Bio-SNG production, with beneficial effects on the cogeneration of electricity from the Bio-SNG process excess heat. The choice of methanation technology between fixed and fluidised bed is not a critical one with respect to process integration, since both technologies allow for efficient heat recovery and consequent cogeneration. For gas upgrade, in particular removal of CO₂ from the product gas, amine based separation is shown to achieve better energy efficiency and economic performance than membrane based or pressure swing adsorption processes. Preliminary estimations of Bio-SNG costs are significantly higher than current natural gas prices, thus dedicated and long term policy measures are necessary in order to stimulate Bio-SNG production. The process integration aspects presented in this thesis can contribute to reducing production costs by increasing energy efficiency and in consequence increasing economic robustness of Bio-SNG process concepts.

KEYWORDS: process integration, synthetic natural gas, biomass, gasification, energy systems, performance indicators, process modelling.

Appended papers

This thesis is based on the work described in 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. DOI: 10.1680/ener.2009.162.1.13.
- II. Heyne, S., Harvey S. (2009). Production of Synthetic Natural Gas from biomass – process integrated drying. *Proceedings of the 22nd International Conference on Efficiency, Cost, Optimization, Simulation and Environmental Impact of Energy Systems (ECOS)*, Foz do Iguacu/Brazil, pp. 1877-1886.
- III. Heyne, S., Thunman, H., Harvey, S. (2012). Extending existing combined heat and power plants for synthetic natural gas production. *International Journal of Energy Research*, 36 (5), pp. 670-681. DOI: doi:10.1002/er.1828.
- IV. Heyne, S., Seemann, M., Harvey, S. (2010). Integration study for alternative methanation technologies for the production of synthetic natural gas from gasified biomass. *Chemical Engineering Transactions*, 21, pp. 409-414. DOI: 10.3303/CET1021069.
- V. Heyne, S., Harvey, S. (2013). Assessment of the energy and economic performance of second generation biofuel production processes using energy market scenarios. *Applied Energy*, 101, pp. 203-212. DOI: 10.1016/j.apenergy.2012.03.034.
- VI. Heyne S., Harvey, S. (2013). Impact of choice of CO₂ separation technology on thermo-economic performance of Bio-SNG production processes. Accepted for publication in *International Journal of Energy Research*. DOI: 10.1002/er.3038.
- VII. Heyne S., Thunman, H., Harvey, S. (2013). Exergy-based comparison of indirect and direct gasification in the framework of Bio-SNG production. Submitted for publication in *Biomass Conversion and Biorefinery*.

Co-authorship statement

Stefan Heyne is the main author of all appended papers. Professor Simon Harvey supervised the work with contribution to all papers. Professor Henrik Thunman co-supervised the work and contributed substantially to Papers III and VII with conceptual input and modelling guidance. Assistant Professor Martin Seemann assisted with input about the methanation technologies investigated in Paper IV.

Paper I is a journal publication based on a poster presentation during a summer school organized by the UK Energy Research Centre. Papers II and IV are conference proceedings that were included in order to present the whole spectrum of conversion steps within the Bio-SNG process and the associated process integration aspects within this thesis.

A substantial part of the material presented in this thesis is based on the author's Licentiate thesis:

Heyne, S. (2010). Process Integration Opportunities for Synthetic Natural Gas (SNG) Production by Thermal Gasification of Biomass. Licentiate thesis. Department of Energy and Environment, Chalmers University of Technology. Gothenburg, Sweden.

Related work not included in this thesis

- Pihl, E., Heyne, S., Thunman, H., Johnsson, F. (2010). Highly efficient electricity generation from biomass by integration and hybridization with combined cycle gas turbine (CCGT) plants for natural gas. *Energy*, 35 (10), pp. 4042-4052. DOI: 10.1016/j.energy.2010.06.008.
- Heyne, S.; Thunman, H., Harvey, S. (2008). Integration aspects for synthetic natural gas production from biomass based on a novel indirect gasification concept. *11th Conference on Process Integration, Modelling and Optimisation for Energy Saving and Pollution Reduction (PRES), 24 - 28 August 2008, Prague, Czech Republic.*
- Arvidsson, M.; Heyne, S.; Morandin, M.; Harvey, D. (2012). Integration opportunities for substitute natural gas (SNG) production in an industrial process plant. *Chemical Engineering Transactions*, 29, pp. 331-336. DOI: 10.3303/cet1229056.

The article by Erik Pihl *et al.* focuses on biomass gasification for integration in natural gas power plants and is not dedicated to Bio-SNG production. It was therefore decided to not include it in this thesis. The second article is a conference paper that presents an early version of the base case SNG process used in Paper III. It also is the basis for the integration considerations presented in Paper II. Including it was not considered instrumental to improving the quality of the thesis and it therefore was discarded. The last article is mainly the work of Maria Arvidsson and is rather considered a continuation of the current work investigating further possibilities for integration of Bio-SNG production.

Other work by the author:

- Lind, H , Heyne, S , Johnsson, F. What is the efficiency of a biorefinery? (2012)
In: System perspectives on biorefineries. Sandén, BA (ed.). Chalmers
University of Technology, Göteborg.
- Heyne, S, Liliedahl, T, Marklund, M (2013) Biomass Gasification – A Synthesis
of Technical Barriers and Current Research Issues for Deployment at Large
Scale. f³ – The Swedish Knowledge Centre for Renewable Transportation
Fuels. Report 2013:5, Gothenburg, Sweden.

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1 Introduction

1.1 Background

Natural gas is the world's third most important energy source accounting for about 21% of primary energy supply¹. In the European Union natural gas is the second most important source of primary energy, with a share of about 25% of total supply (Eurostat, 2012). The future demand for natural gas use is estimated to increase further on a global level with a moderate increase for Europe. Figure 1 illustrates the historic development of natural gas use on a global and European level as well as projections according to the International Energy Agency (IEA)'s World Energy Outlook 2011 (IEA, 2011a).

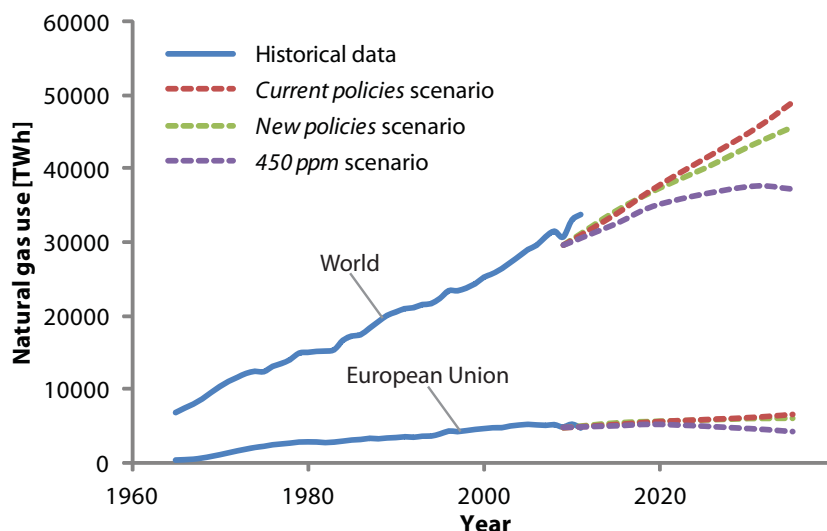


FIGURE 1. Natural gas use for the European Union and on a global level (historical data from BP World Energy Statistics 2012 (2012), projections from IEA World Energy Outlook 2011 (IEA, 2011a)).

¹ 29529 TWh (2539 Mtoe) natural gas, corresponding to 20.9% of the total primary energy demand of 141095 TWh (12132 Mtoe) in 2009 (IEA, 2011a)

The *New Policies* scenario represents the core scenario of the IEA World Energy Outlook assuming implementation of new policy measures according to recent government commitments, while the *Current Policies* scenario assumes no additional policy measures besides those in place as of mid-2011. Finally, the *450 ppm* scenario is based on the goal of limiting the increase in global mean temperature to 2°C above pre-industrial levels, illustrating a possible pathway of policy measure implementation in order to reach that goal (IEA, 2011a). Except for IEAs *450 ppm* scenario (implying the largest efforts for CO₂ reduction) all scenarios imply an increased use of natural gas over the coming 20 years. A recent development that may change the predictions considerably is the increasing exploitation of non-conventional gas resources, mainly by hydraulic fracturing (or fracking). In a dedicated scenario accounting for these gas resources IEA forecasts a substantially larger gas supply on a global level with gas overtaking coal in global primary energy supply by 2030 and reaching a level of 25% of world energy supply by 2035 (IEA, 2011b). However, environmental concerns related to the extraction of these non-conventional gas resources (e.g. ground water contamination by leakage of fracking chemicals) may well lead to more stringent regulations which could hinder the growth rate of non-conventional natural gas market share. A general consensus from all scenarios is that natural gas will play an important role in the future energy supply mix.

Natural gas in Europe is mainly used in heat and power generation plants, for heating and cooking in the residential and service sector, as well as in industry. As illustrated in Figure 2, the distribution of industrial sectors using natural gas in Europe is widespread with the chemical, mineral, and food industries having the largest share. In addition to the energy use of natural gas in industry (980 TWh/yr), an additional 153 TWh/yr (i.e. about 13.5% of the total natural gas use in industry) are used for non-energy purposes (mainly as process feedstock in the chemical industry), according to statistical data for 2010 (Eurostat, 2013).

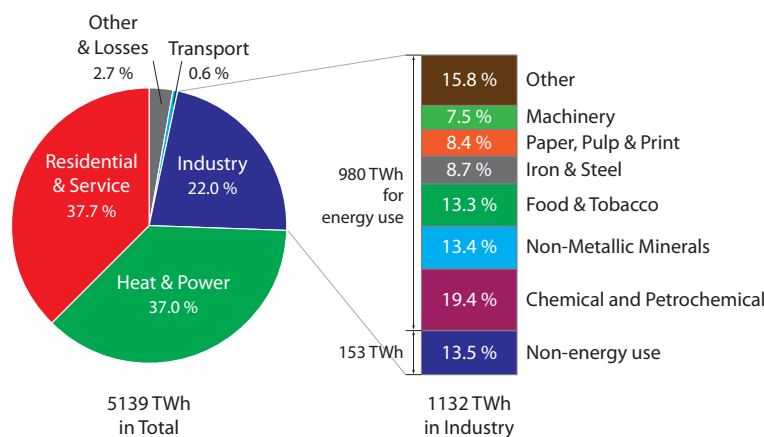


FIGURE 2. Natural gas use in Europe 2010 (Eurostat, 2013).

In Sweden, the share of natural gas in primary energy supply is considerably lower compared to the European average, corresponding to only about 3% of the total primary energy supply of 618/yr TWh in 2010 (Energimyndigheten, 2012). Natural gas is mainly used in the heat and power sector and in industry. Of the 5 TWh/yr of natural gas used in industry, about 25% are used for non-energy purposes serving as process feedstock, mainly in the chemical and petrochemical industry. The distribution of natural gas use in Sweden is illustrated in Figure 3.

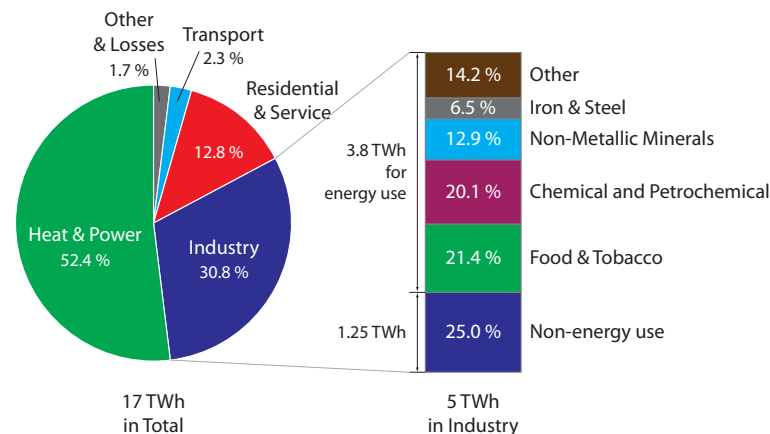


FIGURE 3. Natural gas use in Sweden 2010 (Eurostat, 2013).

Europe in general and Sweden in particular are net importers of natural gas with an energy dependency for natural gas of about 62.4% (Europe) and 100% (Sweden) in 2010². Europe, including Sweden, has therefore a lot to benefit from replacing fossil natural gas with domestic and renewable alternatives. Both reduced fossil CO₂ emissions and energy dependency are major incentives for substitution of fossil natural gas with renewable alternatives. Natural gas is mainly composed of methane (mole-% above 90%, with a strong variation depending on the origin of the gas), therefore a renewable alternative for replacing natural gas is biomethane, namely methane (CH₄) originating from biomass. A number of different terms are used in connection to biomethane and for the sake of clarity the common terminology – that also will be used within this thesis – is summarized below:

- *Biomethane*
General term for high purity methane produced from biomass feedstock.
- *Biogas*
Gas mixture containing methane originating from anaerobic digestion of biomass feedstock, primarily waste streams such as manure, sewage sludge,

² Energy dependency is defined as the ratio between net import and gross inland consumption of natural gas (Eurostat, 2012)

or organic waste. Raw biogas is often of low purity and consists of a mixture of mainly methane and carbon dioxide.

- *(Bio-)SNG*
synthetic (or substitute) natural gas at high purity, produced by a catalytic synthesis process from syngas, i.e. a mix of hydrogen, carbon monoxide (and/or carbon dioxide). Syngas is obtained by thermal gasification of e.g. coal or biomass.

Biomethane can basically replace fossil natural gas in any of its applications. Biomethane can be distributed via the natural gas grid or by road transport in compressed or liquefied form (CNG/LNG). The natural gas grid infrastructure in Europe is well developed, as illustrated by Figure 4. Sweden's natural gas infrastructure is restricted to the southwest of the country, but liquefied natural gas is considered a promising option for overcoming limitations for natural gas distribution. A recent study investigating options for increasing natural gas use for heavy duty road transport concluded that full coverage of Sweden could be accomplished by installing only 18 additional LNG refilling stations in addition to existing and planned ones (Stenkvist *et al.*, 2011), basically uncoupling the dependence of natural gas usage on existing gas grid infrastructure. In addition, a proposal for a directive on the deployment of alternative fuels infrastructure recommends an improvement in natural gas refilling stations ensuring a maximum distance of 150 km between CNG stations and 400 km between LNG stations for all EU member countries by 2020 (European Commission, 2013). If this proposal is accepted, future distribution of biomethane can be assumed to be essentially unrestricted.

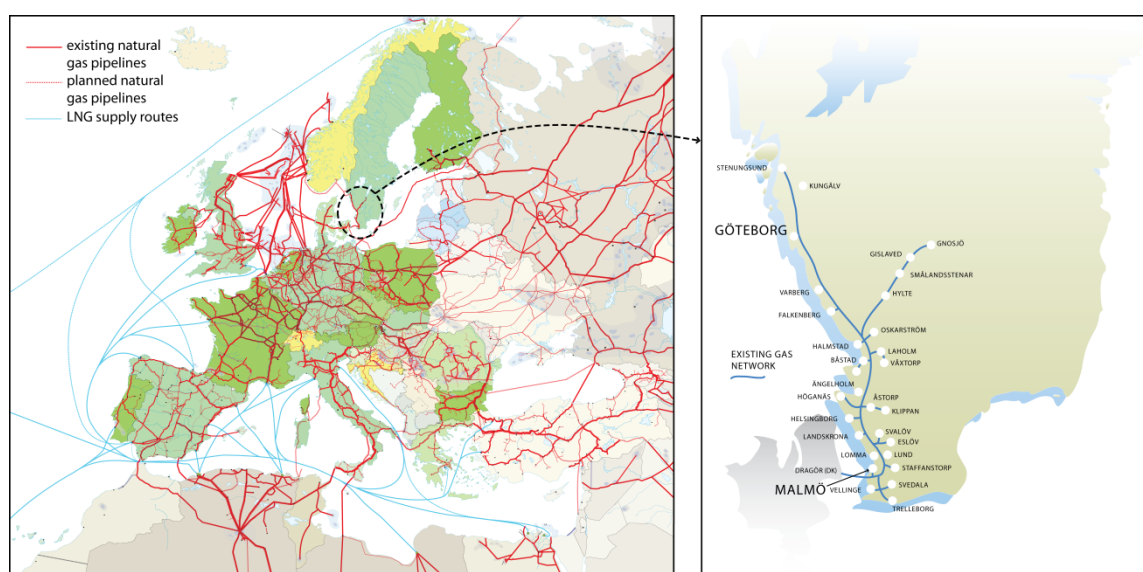


FIGURE 4. Natural gas grid infrastructure in Europe (left – modified from (ENTSOG, 2012)) and Sweden (right – modified from (Swedegas, 2012)).

The transport sector is of particular interest for biomethane as well as for other biofuels. IEA estimates that by 2050 about 27% of the transport fuel needs – corresponding to nearly 8900 TWh/yr³ – could be covered by biofuels on a global level, resulting in avoided emissions of about 2.1 Gt of fossil CO₂ per year. The amount of biomass necessary for biofuel production can be produced without land use competition and competition for food and fibre given a sound policy framework and technological improvements in biomass conversion processes (Eisentraut *et al.*, 2011). In Sweden, a recent governmental directive proposes a change in transport sector energy supply aiming at being independent from fossil fuels by 2030 (Regeringen, 2012). The major pathways for achieving this ambitious goal are electrification and biofuels, as well as increased efficiency in the transport system.

Estimations on the future development of the transport energy demand based on a EU study (European Commission, 2010) illustrated in Figure 5 indicate a slight increase in the total value with decreasing demand in the private and public road transport sector as well as an increase in the heavy duty transport and aviation in Europe. For Sweden the total transport energy demand is estimated to continue to grow within the near future and thereafter contract back to the 2010 year level by 2030. Trends within specific transportation sectors are expected to be similar to those of the rest of Europe.

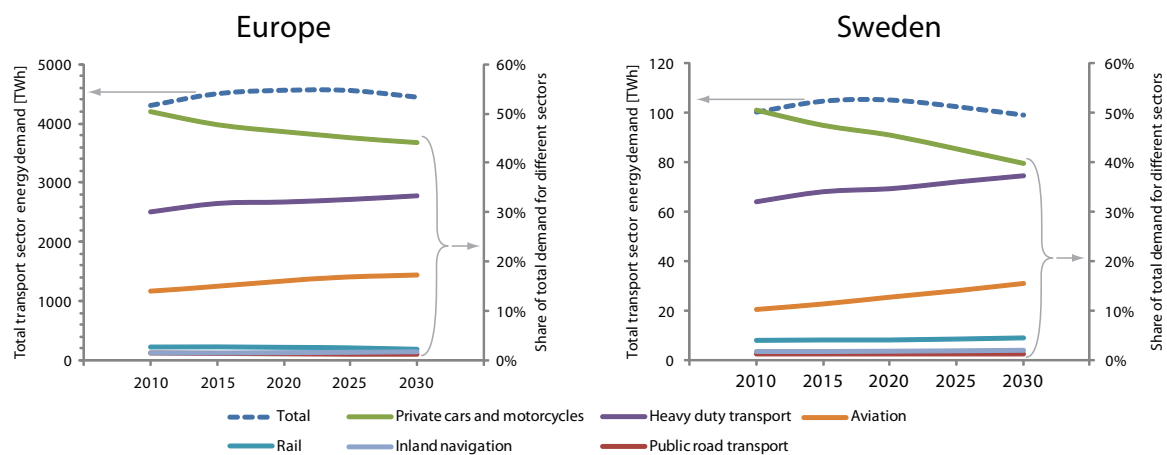


FIGURE 5. Transport sector energy demand predictions for Europe and Sweden based on the EU reference scenario (European Commission, 2010)

The total Swedish transport energy demand in 2010 (including international transport, not included in Fig 5) amounted to 122.1 TWh. As illustrated in Figure 6, production of upgraded biogas from fermentation provided 0.6 TWh in 2010

³ Estimated total transport fuel demand in 2050: 116 EJ (32222.2 TWh) with a contribution of 32 EJ (8888.9 TWh) from biofuels (Eisentraut *et al.*, 2011)

corresponding to about 12% of the renewable transportation fuel use (Energimyndigheten, 2012), the remainder being covered by ethanol and biodiesel.

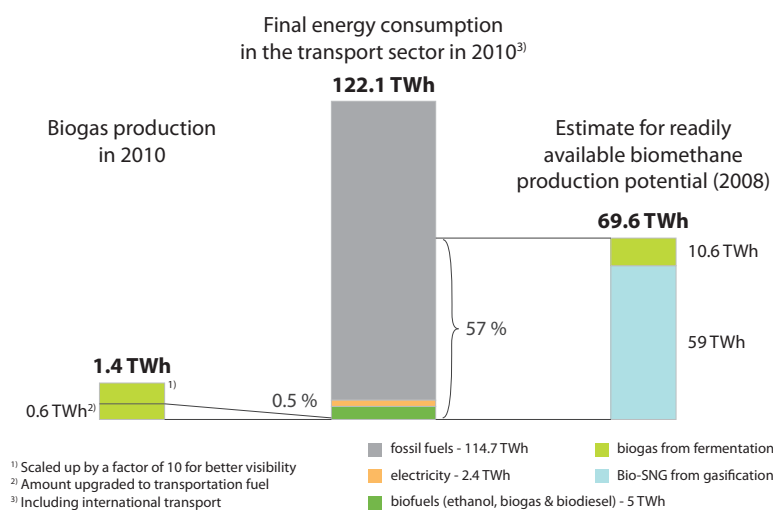


FIGURE 6. Current production and production potential of methane from fermentation and gasification in relation to the transport sector's final energy consumption in Sweden (based on (Energimyndigheten, 2012; Lineé *et al.*, 2008)).

These numbers could be substantially increased by further increasing biogas production from fermentation as well as by introducing Bio-SNG production via gasification. 57% of the Swedish transport fuel demand could theoretically be covered by renewable methane. However, there will be competition from other biofuel market segments (e.g. ethanol, biodiesel, DME) for both the feedstock (forest residues) and market share, thus such a large share of natural gas driven vehicles is unlikely to occur. According to estimates from 2008, there are currently biogenic feedstock resources for production of 10.6 TWh/yr of biomethane from fermentation as well as forest residues for producing 59 TWh/yr of Bio-SNG from gasification available in Sweden at present. In the future, using new technologies, the biomethane production potential from biomass fermentation could possibly increase to about 15 TWh/yr. Improved forest management is estimated to increase the availability of biomass lignocellulosic feedstock. If this increased feedstock is used to produce Bio-SNG, the potential from forest residues could increase to 89 TWh/yr by 2025 (Lineé *et al.*, 2008). In such a scenario, biomethane supply would be dominated clearly by Bio-SNG from thermal gasification which would account for more than 85% of the supply. Bio-SNG production by gasification in conclusion has a considerably larger potential to replace fossil natural gas compared to biogas from fermentation.

When considering alternative processes for generation of biomass-based fuels or products a crucial aspect is the choice of performance indicators to rank the different alternatives. It is a challenging task to define indicators that are capable of covering all the dimensions implicated with biofuel production and varying

performance indicator definition or underlying assumptions may change results considerably (Lind *et al.*, 2012). The different dimensions investigated may vary from energy performance aspects, to environmental aspects such as for example greenhouse gas emissions, land use change effects or water use, as well as to economic and social aspects. It is hardly possible to define a universal indicator suitable for comparison of a large variety of processes but certain assumptions have to be made and most importantly clearly presented when analysing different processes.

This thesis presents a thorough investigation of a number of key Bio-SNG production processes. The work focuses on process synthesis and includes process models for the different conversion steps in the production chain, an in-depth analysis of process integration options in order to maximize conversion efficiency, as well as an evaluation of the performance of the process configurations investigated.

1.2 Thesis Outline

Starting from the historical development of SNG production processes as well as the current status of industrial scale production plants, the details on the conversion steps and technologies within SNG production followed by a review of research activities focusing on integration methodologies and studies of Bio-SNG production processes are presented in Chapter 2. Thereafter, the objectives and scope of the current work are defined based on the identified research needs in Chapter 3. Chapter 4 includes an overview of the methodology adopted in this thesis. Important results from the appended papers are presented and discussed in Chapter 5. The order of presentation of the results is structured according to the process steps within the Bio-SNG production process. The influence of the different conversion steps on process performance is illustrated, and finally the integration of the overall process to existing infrastructure as well as system level performance indicators are presented. Chapter 6 summarizes the conclusions that can be drawn from the obtained results and finally, Chapter 7 outlines future areas of research efforts identified based on the current work.

Figure 7 illustrates the focus areas of the appended papers as well as their interrelations in the context of the Bio-SNG production process.

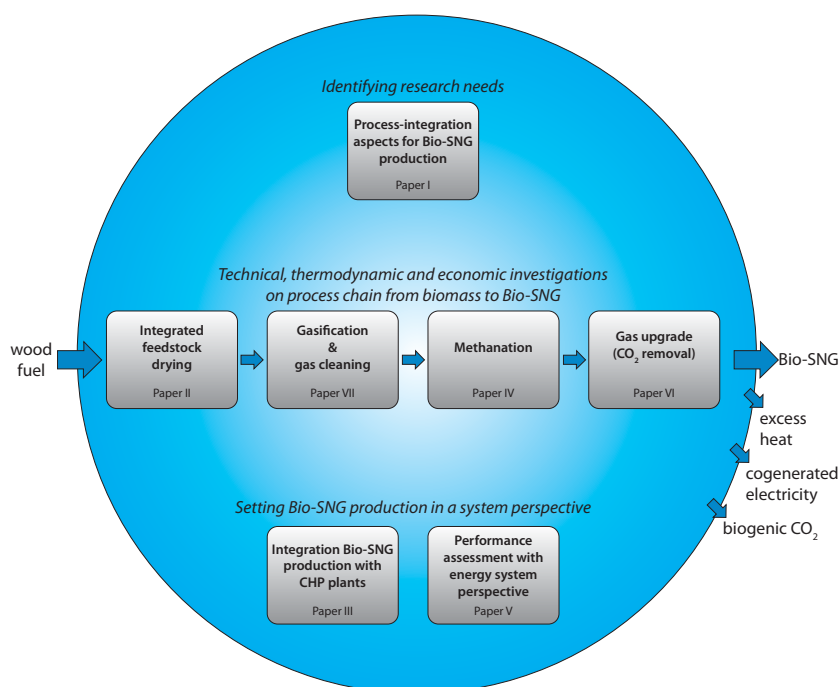


FIGURE 7. Content of and links between papers included in this thesis.

In Paper I research needs in relation to process integration aspects for Bio-SNG production are identified. Papers II, IV, VI and VII investigate different conversion steps within the Bio-SNG process and their importance for overall process performance. Paper III evaluates the consequences of integrating Bio-SNG production into existing infrastructure and Paper V assesses the energy and economic performance as well as the carbon footprint for Bio-SNG production in particular and biofuels in general, also accounting for the background energy system the process is to be implemented into.

2 Technology Background and Literature Review

The general process setup for Bio-SNG production by thermo-chemical gasification is illustrated in Figure 8. Overall, the process releases substantial amounts of excess heat. This excess heat can be partly used to satisfy heat demands in nearby processes or district heating networks. It can also be partly converted to electric power. The cogenerated heat and power constitute value-added products and services which can be sold, thus generating additional revenue.

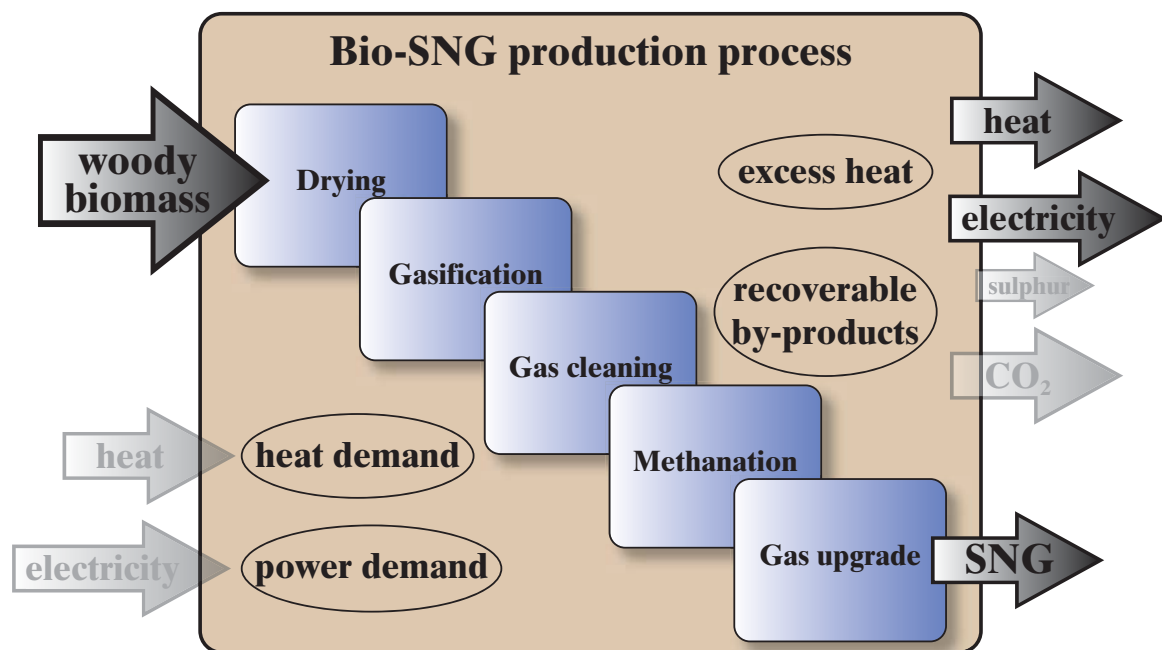


FIGURE 8. Bio-SNG production process with illustration of additional export (as well as possible import) of by-products and services.

With Bio-SNG, electricity, heat, and possibly carbon dioxide and sulphur as product spectrum, the process represents a biorefinery concept according to IEA's definition, namely "the sustainable processing of biomass into a spectrum of marketable products and energy" (IEA, 2009). Carbon dioxide could be considered as a product if biomass based processes are included in a future carbon capture and storage (CCS) framework (i.e. if the capture and storage of biogenic dioxide generates a revenue). Sulphur can be recovered by the well-known Claus

process but the amount of sulphur needs to be considerable to make the process economically viable. Woody biomass has a very low sulphur content so sulphur recovery is not an option for that feedstock, but might become when considering herbaceous crops or waste biomass where sulphur contents can be up to 1% approaching sulphur content levels of bituminous coal (Klass, 1998). One of the key issues for biorefineries is the efficient conversion of biomass material to products and services, demanding a sound heat and material integration of the different sub-processes to form a viable process in terms of energetic, economic and environmental performance (Demirbas, 2009). As the overall process is exothermal, heat import should be avoided. Inappropriate process design may however lead to import of external heat in order to satisfy the heat demand of sub-processes such as e.g. CO₂ separation during gas upgrade. In a similar manner, electricity is needed within the process but if process excess heat is used efficiently for cogeneration of electricity, import of electricity can be avoided. The integration of the different process steps is not a trivial task due to the large number of process alternatives and – in consequence – the large number as well as the varying nature of the process parameters to be handled. Process modelling and optimization studies in combination with process integration methods are needed to systematically analyse and improve the performance of biorefinery processes (Damartzis & Zabaniotou, 2011). In this chapter, an overview of the technology alternatives and modelling approaches for the different conversion steps is presented, followed by a review of industrial scale activities for SNG production. Finally a review of literature focusing on process integration aspects within Bio-SNG production is presented.

2.1 Conversion Steps for Bio-SNG Production

2.1.1 Drying

The natural moisture content of woody biomass is usually around 50 wt-%. Natural drying at the place of harvest or dedicated storage of biomass can reduce the moisture content considerably. Prior to gasification or combustion, a drying process is generally used to reduce the feedstock moisture content using low quality heat, thereby avoiding providing drying heat at the elevated temperature level of the thermal conversion process. Increases in energy efficiency of e.g. combustion boilers of about 5-15% can be achieved when drying the fuel prior to combustion (Amos, 1998). The optimal feed moisture content for biomass gasification depends on the gasification technology and the subsequent product gas usage, but is between 10-20 wt-% for most gasifiers (Cummer & Brown, 2002).

The energy demand for moisture evaporation alone is around 2650 kJ/kg H₂O (assuming the moisture enters the drying process at 0°C and leaves as saturated vapour at 80°C, imaginable representative temperature level for the exhaust of an air dryer). Additional energy input is needed in the 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:

- Low-temperature air drying
- Steam drying
- Flue gas drying

A rough estimation of the energy demand for flue gas dryers amounts to 3200 kJ/kg H₂O (Wimmerstedt & Linde, 1998). In contrast, for a steam dryer, the net heat demand is considerably lower (theoretically as low as 600 kJ/kg H₂O), 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 for heat integration opportunities. 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 ton/h of H₂O is estimated to result in about 40% higher investment costs (Wimmerstedt & Linde, 1998). Such a dryer could for example supply a biomass boiler with a thermal load of about 175 MW_{LHV(20 wt-%moisture)} when assuming drying from 50 to 20 % moisture content prior to combustion.

A common problem associated with biomass drying is the emission of volatile organic compounds (VOC) emitted from the wet material in addition to 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 condensate flows contain a certain amount of impurities requiring treatment (Svoboda *et al.*, 2009). The condensate mainly contains organic acids, terpenes and alcohols, whereas the flue gases are contaminated with non-condensable terpenes and aromatic compounds (Ek *et al.*, 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 H₂O with heat sources at temperatures as low as 80°C (Holmberg & Ahtila, 2005). Opcon Bioenergy for example offers a low-temperature air drying

system with inlet air temperatures at 70°C (Opcon, 2013). 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 for the pulp and paper industry where the heating value is increased with excess heat from kraft boilers that else would be lost to the environment and also accounts for the fact 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 and Gas Cleaning

After drying, biomass is fed into a gasification unit for the major conversion step within the Bio-SNG process converting solid biomass into product gas. For conventional thermal gasification three technology alternatives exist, that are depicted in Figure 9.

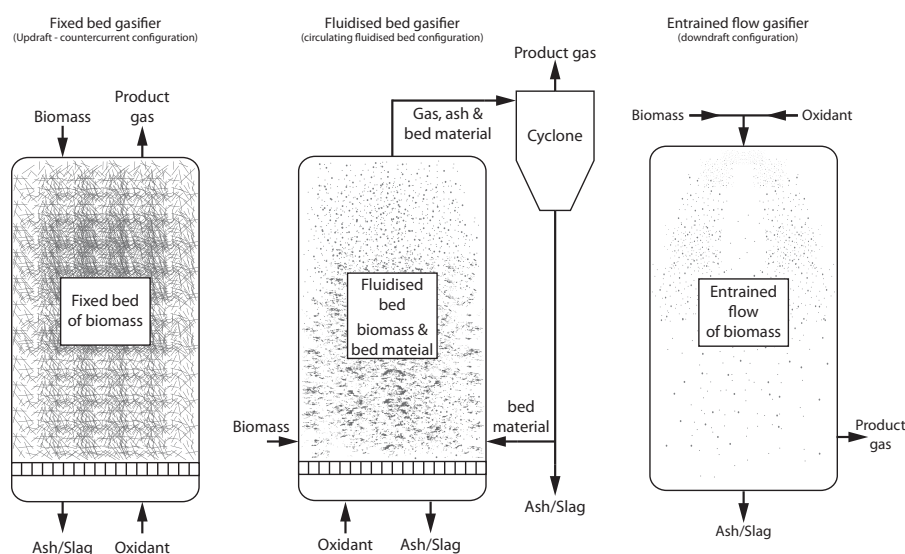
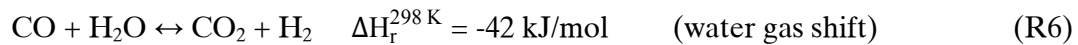
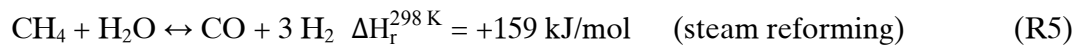
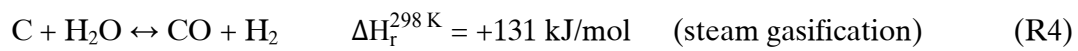
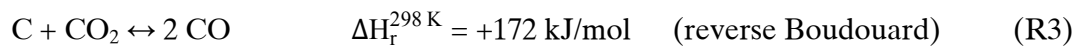
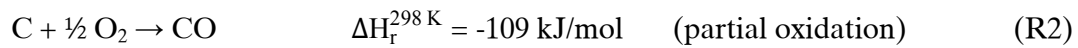
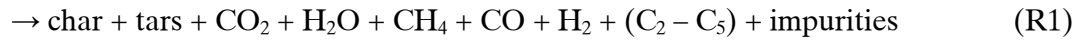


FIGURE 9. Different types of thermal gasification technology.

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 have been proven on a laboratory scale (Waldner & Vogel, 2005). Luterbacher *et al.* (2009) investigated this technology for production of Bio-SNG and identified it as a promising future technology. However, due to its early stage of development and limited amount of published data, the hydrothermal gasification technology is not considered in this work.

Thermal gasification of biomass basically proceeds in three major steps. First the remaining fuel moisture is evaporated, followed by pyrolysis converting the biomass into a char as well as permanent and condensable (water and tars) gases. The major chemical reactions occurring in gasification are listed here (adopted from Heyne *et al.* (2013)):

Biomass feedstock \rightarrow



The first reaction (R1) represents the pyrolysis step while the remaining reactions (R2-R6) are heterogeneous and homogeneous gasification reactions.

The various gasification technologies considered for realizing these conversion reactions require specific feed qualities, have different ranges of operation, and – most importantly – have a certain range of capacity. Indicative values for these different parameters are given in Table 1. Fixed bed gasification is mainly suited for small scale applications, whereas 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 [°C]	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]	< 10	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 can be operated in the same way using oxygen for direct gasification, but even 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 10. 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.

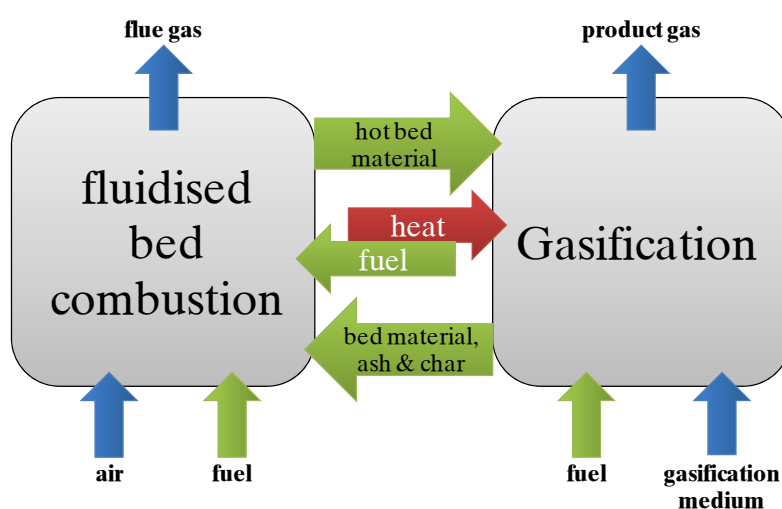


FIGURE 10. Simplified flow diagram of indirect gasification.

In the framework of Bio-SNG by thermal gasification, the two concepts considered for large-scale production are indirect and direct gasification in fluidised beds, both technologies available in the 100 MW range, with direct gasification being considered more suitable for the largest scales as the technology is less complex to operate under pressurised conditions. There also exist a number of small-scale concepts (e.g. heat-pipe reformer for SNG production (Tremel *et al.*, 2012)) that have not been considered in this work. For indirect gasification pressurising the gasification process implies pressurising two vessels (see Figure 10) and, in addition, managing the pressure differential between these two vessels. No commercial pressurised indirect gasification units exist, and it is unlikely that this will be the case in the medium term (Heyne *et al.*, 2013). Fluidised bed gasification has the advantage of being available at larger scale in relation to fixed bed gasification, and gives considerably higher methane content in the product gas – leading to higher conversion efficiency – compared to entrained flow gasification. Pressurisation is sometimes considered necessary to keep the increase in vessel volume moderate when scaling-up in thermal size, thereby gaining in economy of scale.

A general goal for biomass gasification for SNG production is a high methane content out from the gasifier, leading to better performance as losses in the methanation section can be avoided. However, the aim of high methane yields conflicts with associated high tar yields that are undesired. Considering modelling aspects of biomass gasification in fluidised beds, the range of models proposed in the literature varies from black-box to computational fluid-dynamic models. Gómez-Barea and Leckner (2010) present a comprehensive review of available models and conclude that most models are well capable of representing the specific reactor types they have been developed for. However, little data is available for validation of detailed models and in addition, validation at large scale is a major issue that is yet unresolved. Models used for gasification are therefore often specific to a reactor type of experimental equipment used and comparison between model results may in consequence be biased. Too many parameters such as feedstock composition, size, and shape, reactor geometry and operation concept, bed material choice and others make it difficult to apply general approaches that are at an acceptable level of detail for tackling the above mentioned task within a process integration study such as conducted within this thesis.

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. Gas cleaning – and in particular tar cleaning – is still considered by many as the Achilles heel of biomass gasification for downstream synthesis applications (Heyne *et al.*, 2013).

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 dependent 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 water dew point (Rauch & Hofbauer, 2003; Zwart *et al.*, 2009). Water condensation in the scrubbing unit makes a phase separation of the condensate necessary and yields waste-water contaminated with organic compounds, thereby favouring scrubbing technology that operates above the dew point. However, scrubbing generally implies thermal losses during the process as part of the sensible heat of

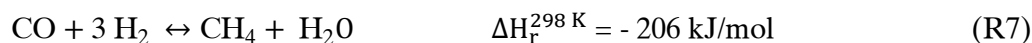
the product gas is lost. Catalytic reforming of tars at higher temperatures might overcome this problem but has not yet been proven on an industrial scale. Chemical looping reforming (CLR) has been proven at a laboratory scale to produce clean synthesis gas (Ryden *et al.*, 2006) and could potentially be applied for tar reforming within biomass gasification for biofuel production processes (Dutta *et al.*, 2011; 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. 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 (Vogel *et al.*, 2006).

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 are used for gasification. Leaching of the feedstock prior to gasification is an alternative technology used to remove alkali (Cummer & Brown, 2002)

2.1.3 Methanation

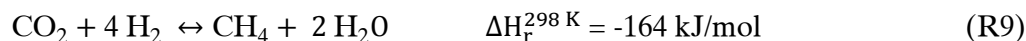
Clean product gas is catalytically converted to methane in the following step. The main reaction occurring during methanation is the conversion of carbon monoxide and hydrogen to methane and water. As documented in the literature (Hayes *et al.*, 1985) and as is obvious from reaction (R7), the optimal ratio of hydrogen to carbon monoxide prior to methanation is 3:1.



For product gas from biomass gasification, however, this ratio is often less than 2:1, making a water-gas shift (reaction (R8), already mentioned in gasification reactions as reaction (R6)) necessary.



Carbon dioxide is another possible source of methane from the product gas and can be converted according to reaction (R9). This is, however, a linear combination of reactions (R7) and (R8). The reaction illustrates that complete conversion of the carbon stock in the product gas (represented by CO and CO₂) can be converted in case enough hydrogen can be supplied.



The highly exothermic character of the methanation reaction (R7) makes it an interesting source of recoverable process heat within the Bio-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 undertaken during 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. Experimental results for catalytic conversion of methane indicate that the theoretical chemical equilibrium is approached very closely (Seemann, 2006). The two alternative technologies are illustrated in Figures 11 and 12.

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 (Bio-SNG, 2009; Seemann, 2006).

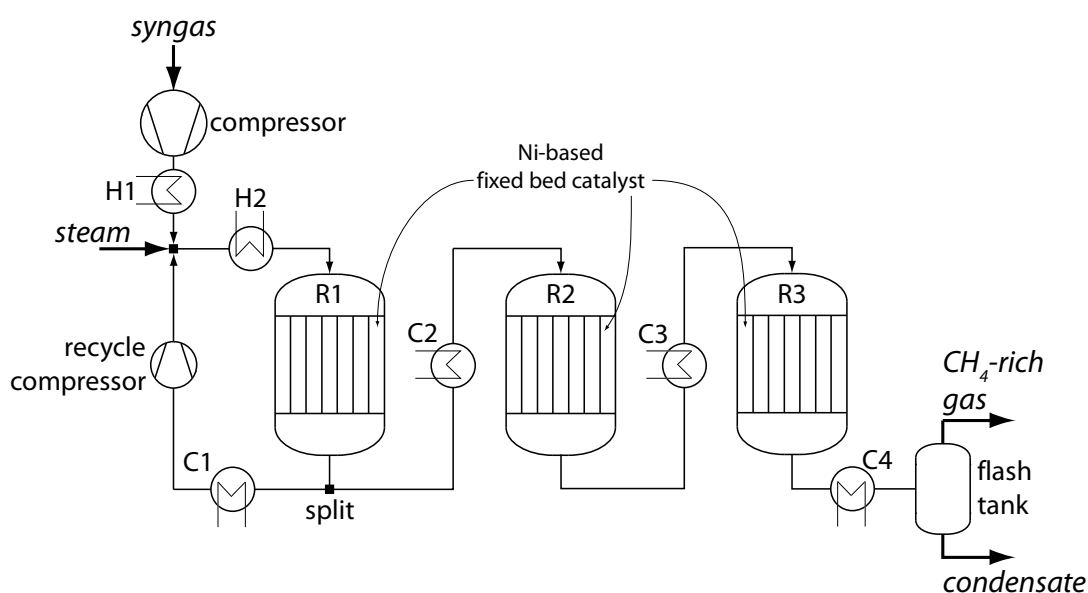


FIGURE 11. Adiabatic fixed bed methanation with inter-cooling and recycle (TREMPE technology). H1 & H2: heater, C1-C4: cooler, R1-R3: adiabatic methanation reactor (flowsheet adapted from Harms *et al.* (1980)).

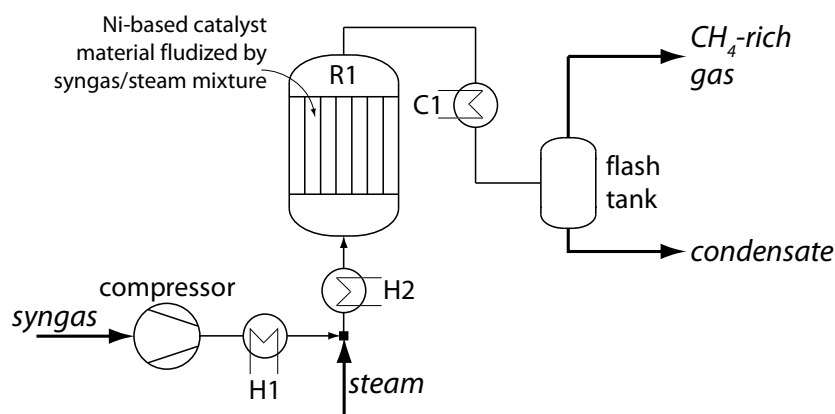


FIGURE 12. Isothermal fluidised bed methanation (COMFLUX technology). H1 & H2: heater, C1: cooler, R1: isothermal methanation reactor (flowsheet adapted from Friedrichs *et al.* (1982)).

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 for methane yield as the number of moles is reduced from four to two according to reaction (R7). 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 Bio-SNG process is crucial to obtain an optimal overall process performance.

2.1.4 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 Bio-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, H₂ and NH₃) are, however, not mentioned.

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) ^a	kWh/m ³	13.60	15.81
Relative Density RD ^b	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

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

^b 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) ($RD = \frac{\rho_{gas}}{\rho_{air}}$)

The most important gas upgrade step is the separation of CO₂. 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 technologies and a qualitative indication of their energy demand, methane recovery and investment costs.

TABLE 3. Qualitative characterisation of CO₂ separation processes (ISET, 2008; Johansson, 2008; Reppich *et al.*, 2009).

Technique	Heat Demand	Power Demand	Methane Recovery	Investment Costs
Pressurised Water Scrubbing (PWS)	none	moderate	high	low
Physical Absorption (Selexol)	moderate	moderate	high	moderate
Pressure Swing Adsorption (PSA)	none	moderate	high	moderate
Amine Based Absorption (MEA)	very high	low	very high	high
Membrane Separation	none	high	moderate	very high
Cryogenic Separation	none	high	very high	very high

When integrating the CO₂ separation within the Bio-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.

Finally, the gas needs to be dried of any remaining moisture and then compressed. Drying is usually achieved by a glycol wash unit using 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 grid into which the Bio-SNG is to be fed. Local distribution networks in Sweden have a pressure of up to 10 bar, whereas large transmission pipelines are pressurised up to 80 bar.

2.2 Industrial Scale Production of SNG

Production of SNG from solid fuels (primarily coal) was initially driven by the oil crisis in the 1970s leading to a number of pilot and demonstration plants (Kopyscinski *et al.*, 2010). The Great Plains Gasification Plant (Panek & Grasser, 2006) is the only plant that was built on commercial scale and that is still in operation today. The plant was started up in 1984 with a gas production capacity of about 1.5 GW_{SNG}. The plant has operated continuously since start-up and its capacity was gradually increased by process changes to reach 2 GW_{SNG} by 2004⁴. During normal operation the plant is supplied with 18000 t/d of lignite coal, corresponding to about 3.4 GW. In addition to SNG the plant produces a number of by-products such as naphtha, phenol, krypton, xenon, liquid nitrogen, and carbon dioxide. The latter is transported by pipeline and used for enhanced oil recovery (Panek & Grasser, 2006).

Investigations of Bio-SNG production processes based on thermo-chemical gasification of woody biomass started around the millennium shift when interest in large-scale production of renewable electricity, transportation fuels and materials increased. However, no industrial scale plant for Bio-SNG production is in operation to date. A number of biomass gasification concepts exist which have been demonstrated at varying scale and level of development and which are also considered for Bio-SNG production. One prominent example of biomass gasification technology is the fast internally circulating fluidised bed (FICFB) gasifier developed at TU Vienna with its demonstration plant in Güssing, Austria that was started up in 2001 (Hofbauer *et al.*, 2002). It is an indirect gasification

⁴ 170 million standard cubic feet of SNG per day (4.62 million Nm³/day) with an average heating value of 972 BTU/scf (37.8 MJ/Nm³) (Panek & Grasser, 2006)

technology operating similarly to the general concept illustrated in Figure 10. The Güssing gasifier has a 8 MW_{th} fuel feed thermal capacity. The product gas is used for co-generation of heat and power in internal combustion engines. A number of CHP plants based on the FICFB technology are in operation in Austria (Oberwart, Villach) and Germany (Senden) (Rauch, 2012). In the plant in Güssing, methanation tests were performed using a slip-stream of the product gas (Seemann, 2006; Seemann *et al.*, 2004) and a demonstration Bio-SNG plant with a $1 \text{ MW}_{\text{SNG}}$ production capacity was installed and has been operated successfully since 2009 (Bio-SNG, 2009).

A number of industrial scale projects for Bio-SNG production are planned based on the indirect gasification concept developed and demonstrated in Güssing. The Gothenburg Biomass Gasification (GoBiGas) project (GoBiGas, 2012; Gunnarsson, 2011) is currently under construction with a $20 \text{ MW}_{\text{LHV,SNG}}$ plant based on the FICFB gasification technology that is to start full scale operation in November 2013. The methanation and gas upgrade section in the GoBiGas plant is provided by Haldor Topsøe using a fix bed adiabatic methanation technology. This is different from the Güssing plant, where isothermal methanation in fluidised bed has been applied. The feedstock for gaining first operational experience will be wood pellets with the goal of changing to wood chip forest residues once the main process has been demonstrated. After evaluation of the operational experiences an extension of the plant to a total production capacity of $100 \text{ MW}_{\text{LHV,SNG}}$ is envisaged.

In France, the Gaya Bio-SNG production project based on the FICFB gasification technology is under development (Mambré, 2010; Perrin, 2012) with a thermal feedstock input of $0.6 \text{ MW}_{\text{th}}$. The project is designed as a development and testing platform that is planned to be run during a 5-year testing period with the aim of identifying favourable process options and conditions for scale-up to industrial size.

Another gasification technology based on the FICFB concept is the Adsorption Enhanced Reforming (AER) gasification technology that was successfully tested at the Güssing plant in 2007/2008 (Koppatz *et al.*, 2009). The concept uses a CaO-containing bed material and operates in the lower temperature range for fluidised bed gasification ($600\text{-}700 \text{ }^\circ\text{C}$). This allows situ carbonation of the bed material forming CaCO_3 , and CO_2 is thus removed from the product gas in the gasification reactor. On the combustion side, the carbonation reaction is reversed forming CaO and the CO_2 is released with the flue gases. The resulting product gas from the gasifier has a considerably higher H_2 content and in particular a H_2/CO ratio in the range of 3:1 making it particularly suitable for methanation. Based on the successful tests in Güssing a $10 \text{ MW}_{\text{th}}$ plant was planned to be built in Germany

(Marquard-Möllenstedt *et al.*, 2009) but due to drastic increases in wood fuel prices the project was abandoned for economic reasons (Bomm, 2011).

The Energy Research Centre of the Netherlands (ECN) has demonstrated Bio-SNG production based on its MILENA indirect gasification technology at pilot scale (0.8 MW_{th} input). The major difference compared to the Güssing gasifier is the fact that both combustion and gasification are integrated in a single vessel. Biomass is gasified with steam in an inner annular space in a fluidised bed. The unconverted char is burnt with air in the lower part of the vessel surrounding the gasification reactor for supplying the necessary heat (van der Meijden *et al.*, 2009). ECN in collaboration with the HCV group is going to build a plant in Alkmaar for gasification of 11.6 MW_{th} of waste wood for production of Bio-SNG based on the MILENA technology. The plant will use the major part of the cleaned product gas for production of green electricity using a boiler connected to a steam power cycle, and about 10% of the gas will be upgraded to Bio-SNG. The plant is scheduled for start-up in 2014 (van der Meijden *et al.*, 2011, van der Drift, 2013).

In Finland, a consortium between Gasum, Helsingin Energia and Metsä Fibre is studying possibilities to build a 200 MW_{SNG} Bio-SNG plant adjacent to a pulp mill. No choice of technology is done so far and an investment decision is planned to be taken in 2014 (Siitonen *et al.*, 2012).

Direct oxygen-blown gasification technology is the core of the Bio2G project being considered by E.ON with the purpose of building a large scale Bio-SNG production plant with capacity of 200 MW_{SNG} in Sweden (Fredriksson Möller, 2011). Due to uncertainties regarding support policies and in consequence a high economic risk the project was put on hold at the end of 2012 (Hennius, 2012).

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 no suppliers that provide turnkey-ready plants
- considerable biomass fuel market price variations and rather high price levels in relation to fossil alternatives 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 high financial risks for companies engaged in biofuel production activities. One pathway for reducing the financial risks is through technical process improvements in the process chain. Process integration may

contribute to reducing these risks by guiding the technology development by pinpointing crucial aspects for specific sub-processes for the overall process energy performance .

2.3 Process Synthesis and Integration Studies on Bio-SNG Production

A number of published studies have investigated the Bio-SNG process covering a varying spectrum of aspects from energy conversion efficiency and economic performance to process synthesis and integration with varying levels of detail.

Mozaffarian and Zwart (2003) compared different process configurations for production of Bio-SNG with pressurised oxygen-blown CFB gasification and atmospheric indirect steam gasification as key conversion technologies. Excess process heat was assumed to be used for cogeneration of electricity but no detailed process integration approach was applied. The study concluded that indirect gasification is the technology of choice leading to cold gas conversion efficiencies of up to 70% on a lower heating value (LHV) basis. Cost estimations for this technology determined in the study of Mozaffarian and Zwart (2003) are in the range of 16-40 €/MWh_{SNG}. Another study pointing out indirect gasification as superior to other gasification technologies for Bio-SNG production was published by van der Meijden *et al.* (2010). This study focuses on the energetic performance of different process configurations. Indirect gasification is stated to result in considerably higher net biomass to SNG conversion efficiency (67 % on a LHV basis, including the electricity balance and assuming biomass feedstock with a 15 wt-% moisture content), compared to 58% and 54 % for direct oxygen-blown and entrained flow gasification, respectively.

Exergetic analyses of Bio-SNG production based on indirect gasification technology point out the gasifier, methanation step and CO₂ separation unit as the largest sources of exergy losses (Jurascik *et al.*, 2010; Vitasari *et al.*, 2011). The overall exergetic efficiency of the process on a dry fuel basis (13.8 wt-%) is shown to vary from 69.5 - 71.8% for the investigated range of gasification temperatures and pressure, i.e. 650 - 800°C and 1 - 15 bar (Jurascik *et al.*, 2010).

Gassner proposed a methodology for synthesis of thermo-chemical processes for production of biofuels (Gassner, 2010) that is illustrated in detail for Bio-SNG production (Gassner & Maréchal, 2009, 2012). Starting from a superstructure with different technology options for the main process steps in Bio-SNG production, a genetic optimisation algorithm is used to identify optimal process solutions with respect to specified performance indicators, such as specific investment cost,

exergy efficiency, or chemical efficiency. The latter efficiency is defined by Gassner & Maréchal as being a weighted fuel-equivalent efficiency where cogenerated or consumed services such as heat or electricity are replaced by the equivalent amount of natural gas using reference conversion efficiencies. Each set of technology and operating parameters generated by the algorithm is analyzed using an energy-integration model maximising combined fuel, heat and power production of the process using a mixed integer linear programming approach. The nature of the genetic algorithm allows multi-objective optimisation resulting in Pareto curves indicating the trade-off between competing objective functions, for example specific investment cost and energy efficiency. Applying this methodology to Bio-SNG production results in overall energy efficiencies – accounting for the energy content of all streams leaving and entering the system – in the range of 69-76% and production costs in the range of 76 - 107 and 59 - 97 €/MWh_{SNG} based on a thermal input of 20 and 150 MW_{LHV} into the process, respectively (Gassner & Maréchal, 2009). Contrary to the above-mentioned studies, Gassner & Maréchal identify direct oxygen-blown gasification to be the most promising technology for Bio-SNG production (Gassner & Maréchal, 2012). A selection of other findings from the studies from Gassner and Maréchal are that air and steam drying are advantageous technologies for pretreatment prior to gasification, hot gas cleaning in particular is beneficial for pressurised gasification in connection to methanation with CO₂ removal downstream of the methanation unit. For large scale pressurised gasification they conclude that physical absorption is the most suitable CO₂ separation technology for achieving high overall process energy efficiency. In addition Gassner and Maréchal stress that energy recovery and cogeneration contribute significantly to energy and cost-efficient process design and that integration with existing facilities might be an option since small scale steam cycles are usually inefficient and expensive.

Rönsch (Rönsch, 2011; Rönsch & Kaltschmitt, 2012) compares indirect and direct gasification for SNG production with different options for converting the process excess heat to electrical power and identifies O₂-blown gasification as being slightly more advantageous with respect to exergetic efficiency and economic performance. However, the environmental performance evaluation measured as emission of CO₂-equivalents per MJ_{SNG} produced is in favour of indirect gasification for SNG production.

Nyström *et al.* (2007) investigated Bio-SNG production and a number of other biomass gasification concepts in connection to co-location or integration with a natural gas turbine combined cycle (NGCC) plant and an existing district heating network. Bio-SNG production for transportation fuel purposes was identified as the economically most robust solution among the transportation fuel processes investigated (DME, Bio-SNG and hydrogen), given the assumption that policy

support for transportation biofuels is in place. No heat or material integration with the combined cycle plant was assumed for Bio-SNG production and using Bio-SNG as fuel for the NGCC plant was stated to result in low overall energy efficiency from biomass to electricity and heat. Using the biomass gasification product gas directly in the gas turbine was identified as a preferable alternative. The latter concept also was illustrated economically viable by Pihl et al. (2010). Integration of Bio-SNG process with petrochemical processes has been investigated by Arvidsson *et al.* (2012) for replacing fossil natural gas feedstock. The case study investigating the replacement of the site's total demand in natural gas of 162 MW_{th,LHV} shows that the process excess heat from Bio-SNG production can be effectively used to cover the industrial site's steam demand (19 MW) or alternatively cogenerate electricity to cover the Bio-SNG process' electricity demand (21 MW). By maximally utilizing the thermal integration possibilities it was shown that it is possible to cover the site's net steam demand and produce additional 30 MW of electricity, but at the cost of an additional import of 36 MW_{LHV} of biomass fuel.

3 Objectives and Scope

3.1 Objectives

This work focuses on using process integration methods and tools to identify efficient and cost-effective pathways for the production of Bio-SNG by thermal gasification of biomass. The approach adopted accounts for the energy and monetary flows associated with all process product and energy service streams delivered by the Bio-SNG production process.

A variety of applicable technologies exist to achieve conversion within the Bio-SNG production process. The combination of these different technologies results in a number of possible process configurations with varying product spectra. So far, no plant has been built on an industrial scale and there is a lack of knowledge about 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.

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

- the conversion efficiency of biomass to SNG
- the use of recoverable process heat for the cogeneration of power and useful heat
- the economic performance as well as the carbon footprint

In order to facilitate this evaluation, it is necessary to carefully define appropriate performance indicators. These performance indicators should be of a general character, allowing for a fair comparison of Bio-SNG with other biofuel production process options.

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 process synthesis. The modelling of the different sub-processes is based on extensive literature data studies as well as published experimental data, when available. Wherever possible, a model capable of reflecting the influence of changes in operating parameters on the energy and material balance performance was developed. When this was not possible due to limited information on the processes, or the complexity of the modelling effort was judged beyond the scope of this work, standard performance as published in the literature was implemented in the models. A base case Bio-SNG production process flowsheet is proposed, and by varying or replacing different sub-processes in the base case, bottlenecks with respect to energy efficiency are identified. Based on this investigation critical process steps for process heat integration and Bio-SNG yield are identified. Process integration tools are used to identify opportunities for 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, possible integration of the Bio-SNG production process with existing biomass-based combined heat and power plants is investigated in detail.

In order to be able to evaluate the performance of a Bio-SNG process implemented within an existing background energy system, it is important to clearly define the system boundaries and the underlying assumptions for the evaluation. Using energy market scenario tools, a consistent definition of possible future background energy systems can be achieved. Using the energy, economic, and carbon footprint performance data of the available energy conversion technologies within these energy market scenarios, a consistent assessment of the Bio-SNG process alternatives investigated is presented including indirect effects related to the changed consumption and production of fuels and energy services within the energy system. This will be illustrated by the use of different appropriate performance indicators.

3.2 Scope

The scope of this thesis has been limited to currently available technologies for large-scale Bio-SNG production. Technologies considered for small-scale application or being at early development state as mentioned in Section 2 were therefore excluded. Due to these imposed restraints, the major subject of investigation of this work was focused on direct and indirect fluidised bed

gasification technologies as central conversion step within the Bio-SNG process as well as all associated process steps.

The focus has been on evaluation of process concepts based on indirect gasification as a number of studies identify this technology as superior with respect to energy conversion performance compared to alternative gasification technologies in the framework of Bio-SNG production (Mozaffarian & Zwart, 2003; van der Meijden *et al.*, 2010). In addition, indirect gasification as a potential retrofit technology for existing combustion units (Thunman & Seemann, 2009) has been investigated within this work. With ready access to indirect gasification experimental research at Chalmers University of Technology, allowing immediate access to key information about important trends for this technology (Thunman & Seemann, 2009), adopting this technology pathway as the main focus of this work was a natural choice. A comparative analysis based on exergy efficiency for Bio-SNG production concepts based on indirect and direct fluidised bed gasification completes the analysis.

Considering feedstocks suitable for the Bio-SNG process, only woody biomass in the form of forest residues has been considered (composition and heating values see Appendix) as woody biomass is considered to be the main supply for biofuel generation for transportation purposes in Sweden in the medium term. Investigating lower quality wood fuels such as demolition wood or refuse derived fuels is of course a topic of interest, but the implications are mainly on a technological level (e.g. risk for bed agglomeration in fluidised bed, increased gas cleaning demand) and effects on process performance from an energy efficiency perspective are hard to evaluate on a quantitative level. In addition they are not expected to change the basic integration issues for the Bio-SNG process that have been identified within this work.

The timeframe for the processes considered within this study is around 2030 where biomass gasification is expected to be available at large scale. The system boundary for the evaluation of the process performance basically is the Bio-SNG process from moist wood fuel to Bio-SNG. However, in order to account for the co-generated by-products (in particular heat and electricity) as well as to evaluate the carbon footprint performance an expansion of the system boundary beyond the process itself is necessary. Using an energy market scenario tool a number of consistent plausible future energy system backgrounds at European level are developed in order to evaluate the process performance on a system level with reference to the corresponding background energy service for each product or service. This approach even allows assessing the sensitivity of the process performance to background energy system parameters. Even though the process evaluation is far from being a full-scale life cycle performance analysis, it is still

considered a transparent and valuable approach to illustrate implications of Bio-SNG production on an energy system level on energy, economic and carbon footprint performance.

4 Methodology

The aim of this work is to identify energy efficient pathways by which Bio-SNG may be produced from biomass. As this process is not yet established on an industrial scale, little information is available on how to combine and operate the different process steps. This makes it necessary to use modelling tools to estimate mass and energy balances. Using these balances, a systematic investigation of integration opportunities between the major process steps may be conducted by means of pinch analysis methodology. Figure 13 illustrates the basic idea of the process integration methodology adopted in this work.

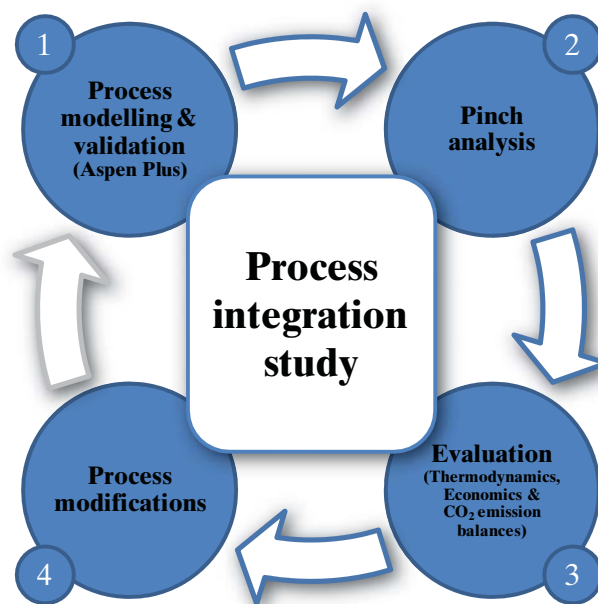


FIGURE 13. Schematic representation of the process integration methodology applied in this work.

Starting from a first process model, a Pinch analysis can be performed based on the mass and energy balances obtained. This allows determining the overall energy balance of a process with maximum internal heat recovery, including fuel yield and cogeneration of heat and power. Appropriate performance indicators can be used to rank different process alternatives with respect to varying criteria. Based on the

results from the Pinch analysis and the process evaluation, possible process modifications as well as eventual process bottlenecks can be identified and be subject to an additional investigation, starting again with modelling of the proposed process changes. Within this work, the focus was on pointing out process bottlenecks based on consistent process performance evaluation. Consequently, the process integration studies performed do not include repeated iterations of the process integration methodology as represented in Figure 13.

The heat and material integration of biomass gasification for Bio-SNG production with existing infrastructure is investigated in particular. To evaluate the different process alternatives, process performance indicators allowing a fair comparison with alternative biofuel production options 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 Bio-SNG with good CO₂ emission reduction potential. The different elements of methodology are described in this section.

4.1 Process Modelling

In order to obtain the mass and energy balances of the Bio-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 modify the built-in models using calculation blocks written in 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. The Peng-Robinson equation of state was chosen based on the fact that it is a standard property method for oil, gas and petrochemical applications. For distillation and scrubbing units involving electrolytes, the electrolyte non-random two liquid (ELECNRTL) model was applied. Modelling results were validated to a maximum possible extent based on published data. Biomass is modelled within Aspen Plus as a non-conventional solid with its properties being determined by the approximate and ultimate composition. For the heating value implementation that is of major importance for the energy balance calculations, an adoption of a general correlation for higher heating value estimation for fuels (Channiwala & Parikh, 2002) to the Aspen Plus input format was used when the heating value was not known from elemental analysis.

The following species were taken into account – except for the simplified gasification modelling in Paper VIII - as being present in the raw product gas:

- main components: CO, CO₂, H₂, H₂O, CH₄
- char, hydrocarbons and tars: C, C₂H₄, C₆H₆O, C₁₀H₈, C₁₆H₁₀
- sulphur compounds: H₂S, COS, C₄H₄S, CH₄S
- nitrogen compounds: NH₃, HCN, NO
- trace components: N₂, O₂, HCl

The alkali components present in biomass ash such as sodium or potassium have not been accounted for and ash is thus treated as an inert component in all simulations. Higher hydrocarbons are represented by ethylene C₂H₄, a simplification also adopted by Gassner and Maréchal (2009). The choice of tar components (phenol C₆H₆O, toluene C₁₀H₈ and pyrene C₁₆H₁₀) was determined by the objective of representing oxygenated and aromatic compounds of varying complexity. In reality, tars are a mixture of hundreds of organic compounds and their measurement is a serious challenge in biomass gasification applications (see e.g. Milne *et al.* (1998), Neves *et al.* (2012)). For the level of detail of this process integration study and the handling of the tar treatment technologies, the representation with the three chosen species is considered sufficiently detailed.

For the process integration studies conducted within this work, a base case Bio-SNG production process was developed and used for investigation of process-integrated drying options as well as comparison of stand-alone operation to integration with an existing combined heat and power plant. Variations of the process were undertaken in order to investigate the influence on the process energy performance and in consequence the economic and carbon footprint performance. Table 4 gives an overview of the use and modification of the base case in Papers II to VII appended to this thesis. A description of the base case process and the sub-processes is given thereafter.

TABLE 4. Overview of models used in Papers II to VII.

Paper	Process Aspects Investigated	Process Model Used
II	Biomass Drying	Base Case ^a
III	Overall Process	Base Case
IV	Methanation	Modified Base Case ^b
V	Overall Process	Base Case
VI	CO ₂ removal	Modified Base Case ^c
VII	Gasification	Simplified Gasification Model

^a earlier version of base case with less components and simpler modelling approach in operations downstream the gasifier resulting in a slightly different energy balance.

^b the main methanation step is changed between adiabatic fixed bed and isothermal fluidised methanation.

^c all CO₂ separation is accomplished after methanation with three different CO₂ separation technologies compared.

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 discussed previously. 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) show 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 CO₂ separation steps – that will also remove the H₂S 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 10 bar, i.e. the pressure level of a local distribution network. For the final upgrading of the raw SNG, a membrane unit for H₂ removal and a triethylene glycol wash for drying purposes are added. A schematic flowsheet of the basic process design is illustrated in Figure 14 with a simplified illustration of the associated combustion boiler and steam cycle. Details on the key modelling assumptions for the different units are given in the Appendix (Table 13).

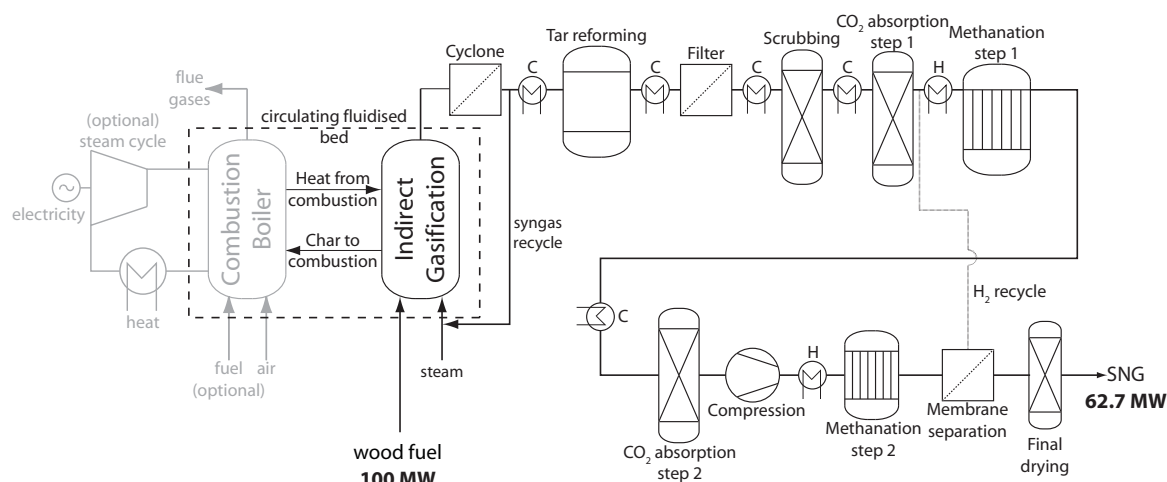


FIGURE 14. Flowsheet of the base case SNG production process (also illustrating the combustion boiler and a simplified scheme of the optional steam cycle).

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 14 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 in order to perform pinch analysis of the proposed process.

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 ₄ H ₄ S	ppm	24
CO ₂	vol-%	23.6	CH ₄ S	ppm	24
CH ₄	vol-%	10.6	NH ₃	vol-%	0.12
C ₂ H ₄	vol-%	3.56	HCN	ppm	48
N ₂	vol-%	0.29	NO	ppm	48
Tars (C ₆ H ₆ O, C ₁₀ H ₈ , C ₁₆ H ₁₀)	g/Nm ³ ^a	1.28	HCl	ppm	89
H ₂ S	ppm	129	Total Dry Gas Flow	Nm ³ /h ¹⁾	39.4

^a at 0°C and 1 bar

Relevant details of the modelling approach for the different technologies are presented in the following chapters for each specific conversion step and can be found in the corresponding papers.

Modelling of the gasification process has been handled in two ways: a more detailed approach representing indirect gasification that has been used for the base case model as well as a simplified modelling approach with a minimal number of species that was used for an exergy-based comparison between indirect and direct gasification.

For the base case the gasifier unit 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 since little data is available. The ratios were set to fixed values with H₂S and COS being the main sulphur compound species and NH₃ 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 to make it possible to get an indication of where in the process chain these compounds might cause problems when applying more sophisticated models in the downstream sub-processes. From an energy integration point of view the presence of these compounds is essentially insignificant and the detailed treatment of the trace components was not modelled.

The gasification step was modelled as a Gibbs equilibrium reactor with the following five species being accounted for in the equilibrium reactions: C, CO, CO₂, H₂O, H₂. 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 satisfactorily represent the composition of the indirect steam gasification product gas leaving the FICFB gasifier (Hofbauer & Rauch, 2000). Steam addition to the gasification process efficiently reduces the tar formation, with a value of steam-to-biomass ratio of 0.5 being sufficient according to Hofbauer and Rauch (2000). Gassner and Maréchal (2009) adopted a value of 0.6 that also was used for the base case in this study. The fluidisation is assumed to be accomplished with a mixture of recycled product gas and fresh steam.

The second approach to gasification modelling – the simplified model – is represented in Figure 15. The basic concept upon which the model is based is that the conversion of the biomass feedstock is similar in indirect and direct gasification with a changing amount of elemental carbon, hydrogen and oxygen (remaining elements are neglected). In order to exclude effects of equipment specific differences between the two technologies on the results, the gasification process is modelled using a simplified stoichiometric model accounting for five species only: CO, H₂, CH₄, CO₂, and H₂O. The first step in the simplified reaction scheme for determining the product gas composition is the decomposition of the biomass fraction entering the gasifier maximised so as to favour CO yield. After potential steam reforming of the CH₄ present by addition of steam, a gas composition with maximum CO concentration (case *CO max*) is obtained. By further addition of water for a complete water gas shift reaction, it is possible to gradually increase the H₂/CO ratio of the product gas with the limiting case corresponding to all CO being converted to H₂ (case *H₂ max*). The aim of this major simplification is to reduce effects of e.g. different bed materials and reactor setups on the comparability of the two technologies that might otherwise result in a biased comparison of the two gasification technologies. Carbon conversion is assumed to be complete in both cases.

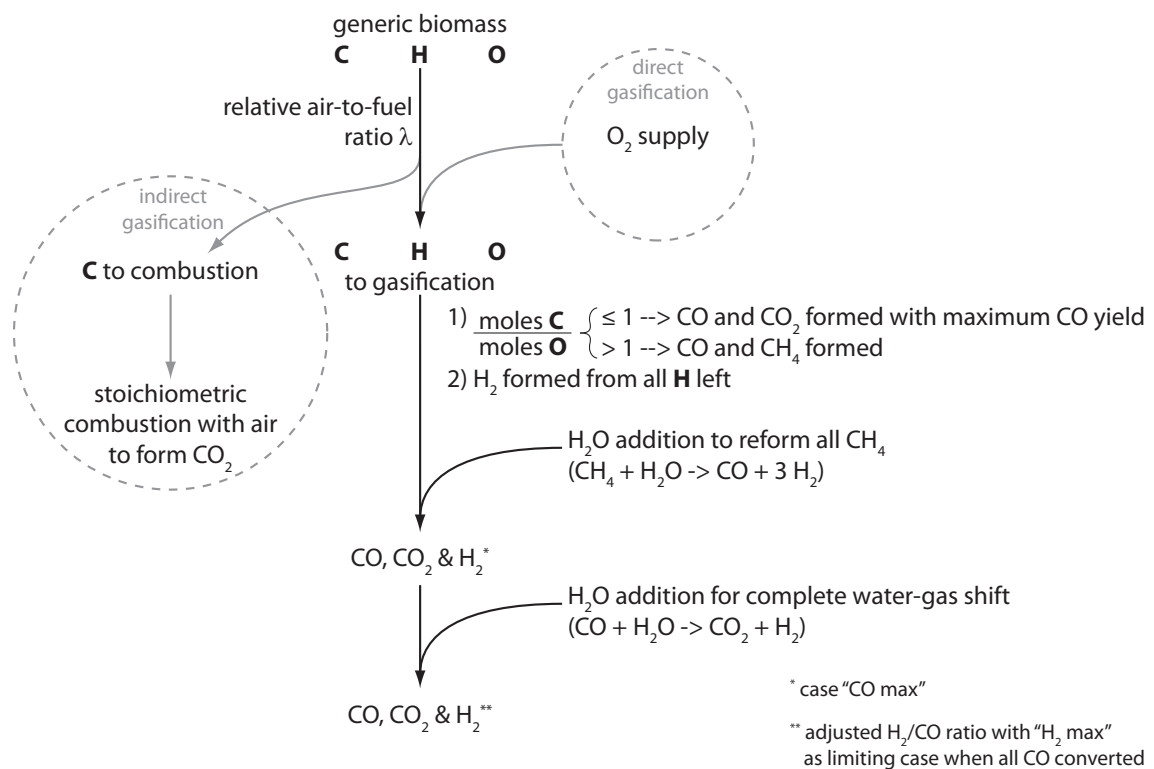


FIGURE 15. Schematic representation of the simplified gasification model.

The product gas resulting from gasification at 800-900°C 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 16. The chemical looping reforming is assumed to operate at 650 °C and all tars are assumed to be converted to CO and H₂. The operating temperature of the CLR assumed in this work is at the lower end of the appropriate range for catalytic tar reforming processes but due to the assumed ideality of the tar conversion, the influence of the temperature level at which the conversion is taking place is negligible.

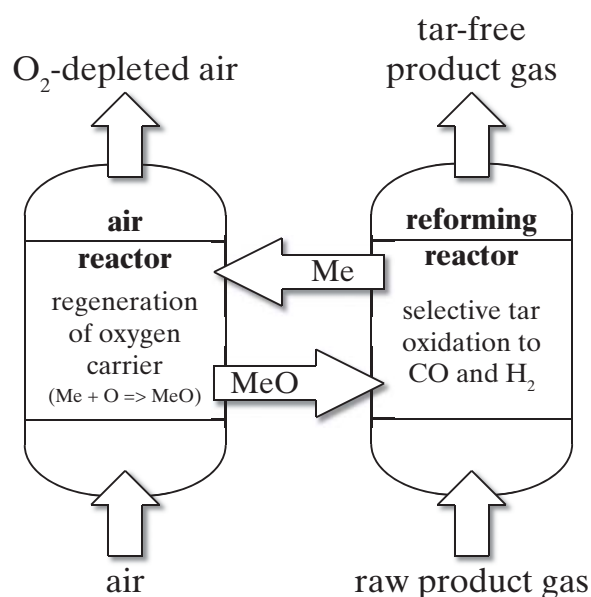


FIGURE 16. Schematic flowchart of chemical looping reforming (CLR).

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 mass, energy and species balances of these processes are primarily based on literature data since no detailed modelling has been performed. The methanation was modelled according to a Gibbs equilibrium reactor assuming a simultaneous water-gas shift and methanation reaction (see reactions R7 and R8). A temperature approach to equilibrium was used for the composition calculations in order to account for deviation from thermodynamic equilibrium in the real 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 (c.f. Appendix).

Considering the uncertainty within the process modelling simplifications and assumptions, effects on the overall process performance and the resulting conclusions need to be considered. When using literature based performance data, multiple sources were consulted whenever possible, aiming at acquiring data representing average performance for the respective technology in order to make the results comparable. Furthermore, Rönsch (2011) conducted a modelling-based study on Bio-SNG production comparable to the work presented here, in which they used a Monte Carlo analysis to investigate the propagation of uncertainties in the model to the final results. For the example of the gasification and methanation parameter influence on the exergy flow of the produced Bio-SNG it was shown that the uncertainty in input model data was strongly damped. A 5% standard deviation in input modelling parameters translated to a normally distributed

exergy efficiency result with a standard deviation of less than 0.9% (Rönsch, 2011). As the nature of the modelling approach is similar in this study, it is to be expected that even here the individual parameter uncertainties will not influence the overall energy performance results significantly and the results can be considered robust.

4.2 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. The IEA uses the following definition for process integration: “Systematic and general methods for designing integrated production systems, ranging from individual processes to total sites, with special emphasis on the efficient use of energy and reducing environmental effects” (Gundersen, 2000). In general, process integration refers to a holistic approach with focus on analysing the overall process performance or parts of a whole process, 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 using pinch technology methodology, whereas the second approach refers to an integration on a more practical level – namely the integration of the Bio-SNG production process with existing infrastructure in the form of fluidised bed boilers for combined heat and power production.

4.2.1 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 energy efficiency improvements. According to some sources, savings of up to 35% in energy consumption are achievable by analysing and improving these processes with the help of pinch analysis, see for example CANMET (2003). The basics of pinch analysis were originally 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 heat exchanger networks as well as improvement of existing heat exchanger networks. Its use has been extended to 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 and 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, the minimum external heating and cooling demand, as well as the internal heat exchange in so-called heat pockets that is necessary for achieving the minimum heating and cooling demand. The heat pockets also represent an opportunity for thermal integration of e.g. a steam cycle recovering heat from the process at the high temperature level and providing heat for the process at a lower heat level as will be illustrated later. Figure 17a is an illustrative example of a GCC. The temperature on the y-axis does not represent the actual temperature of the different streams, instead it is a shifted temperature scale since stream temperatures are 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 17b is the Carnot-based GCC in which the temperature in the y-axis has been replaced by the Carnot efficiency θ defined as

$$\theta = 1 - \frac{T_0}{T} \quad (1)$$

where T_0 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 17b) represents the maximum theoretical amount of mechanical power that might be produced from the process excess heat by means of a Carnot cycle operating with a heat source at temperature T and rejecting heat to the environment at temperature T_0 . 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 based on their respective GCC representations.

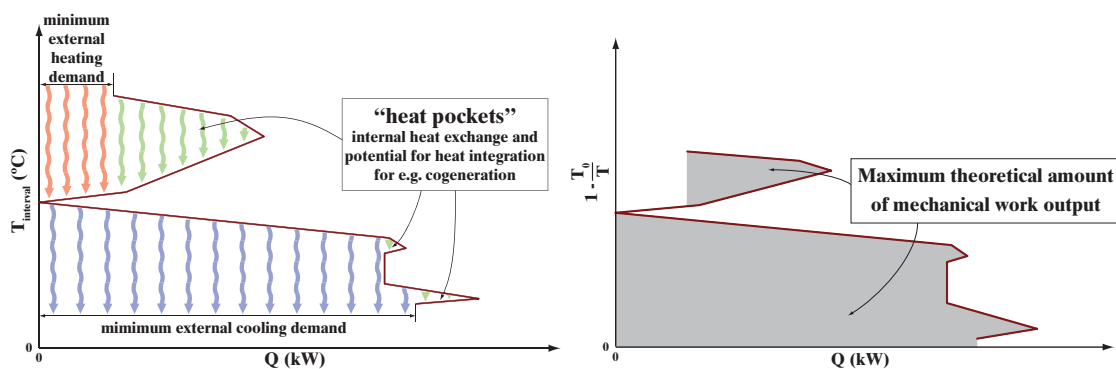


FIGURE 17. a) Grand Composite Curve illustrating external heating and cooling demand and internal heat exchange potential, b) Carnot-based Grand Composite Curve (example taken from (Smith, 2005)).

Another useful tool in pinch technology – in particular for analyzing the integration of certain sub-processes into the remainder of an industrial plant – is background/foreground analysis, also known as split-GCC analysis. The GCC of the sub-process to be analyzed is represented by a separate curve (foreground) while the GCC of 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 maximal integration with a given background process using a sequential procedure. First, the heat integration opportunities between the background and foreground process are identified from the given background/foreground curve based on given process data. In a second step, the foreground process is modified for achieving maximal heat integration to the background curve. Within the Bio-SNG production process, this tool can be used effectively to analyse alternative technologies for a certain sub-process. In particular, this representation is useful for identifying opportunities to optimize a steam power cycle for production of heat and power from the available excess heat of the Bio-SNG production process. Figure 18 is an illustrative example of such a background/foreground analysis of a steam power cycle operating using excess heat from a background process. The red curve represents the background process heat streams, whereas the blue curve represents the CHP steam cycle heat streams. By recovering excess heat from the process (shaded area), steam is generated that can be expanded in the turbine. Increasing the level of thermal integration in order to use more than the freely available excess heat (i.e. excess heat that would otherwise be discharged to the environment), the internal heat transfer within the background process (crossed area) is reduced and steam generation is maximised. To balance the heat deficit thereby created in the background process, steam extraction or condensation of the turbine exhaust is needed for providing heat to the process (area with circles). As an additional output of these curves the generated turbine power output can be read as the difference in energy uptake and release by the steam cycle. This representation thus 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.

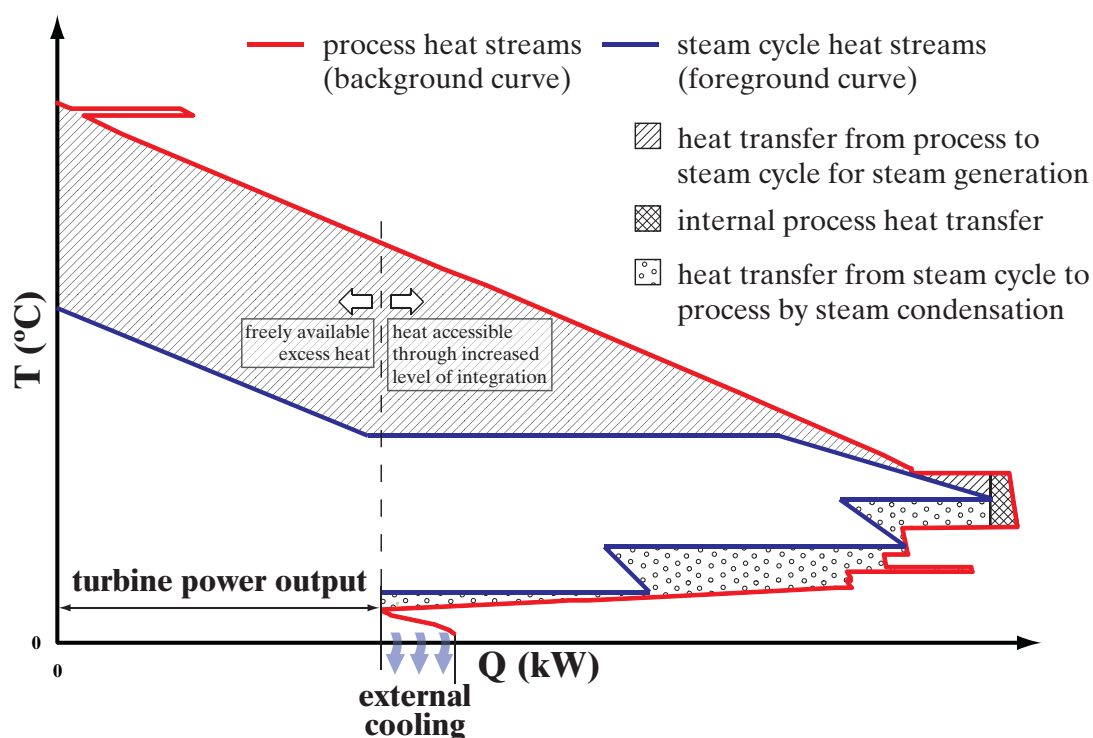


FIGURE 18. Illustrative background/foreground GCC representation with a steam power cycle as foreground process.

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 18.

4.2.2 Integration with Existing Infrastructure

In this work, pinch analysis is used for the thermal integration of the Bio-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 Bio-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 indirect gasification for cogeneration of heat, electricity and transportation fuels is especially promising (Thunman *et al.*, 2007). 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 19 illustrates a schematic chart of the integration between the Bio-SNG process and steam power plant.

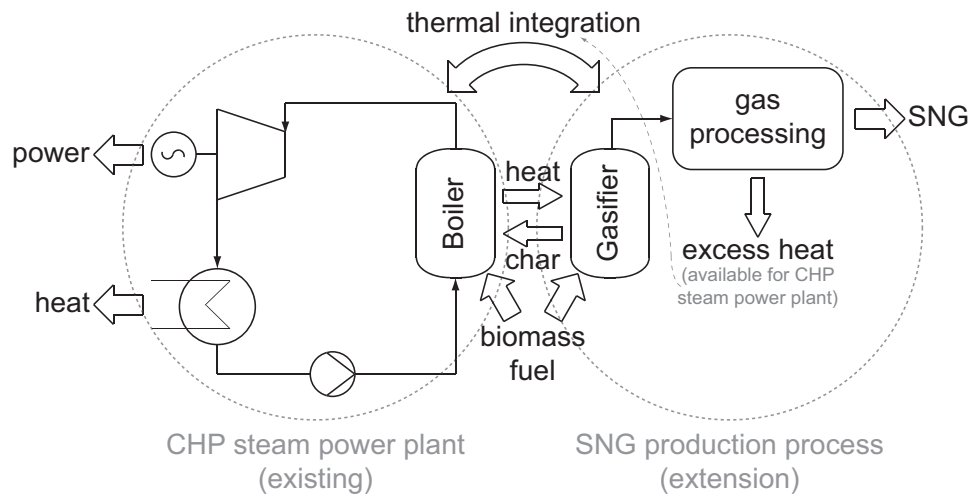


FIGURE 19. Schematic process layout for the integration of Bio-SNG production with an existing CHP steam power plant.

The balance between boiler and gasifier is 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 Bio-SNG process to produce steam.

4.3 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. Even carbon footprint indicators based on CO₂ equivalent emissions to all associated material and energy streams are defined.

4.3.1 Performance Indicators

Thermodynamic performance indicators

Thermodynamic performance indicators may be defined in numerous ways. Three indicators commonly used to characterize processes for conversion of biomass feedstock to biofuels include the cold gas efficiency, energy or thermal efficiency,

and exergy efficiency. The cold gas efficiency η_{cg} is defined as the energetic value of the resulting product in relation to the thermal input of the feedstock:

$$\eta_{cg} = \frac{\dot{m}_i \cdot LHV_i}{\dot{m}_k \cdot LHV_k} \quad (2)$$

where \dot{m} represents the mass flow and LHV the lower heating value of product i (e.g. Bio-SNG) and fuel input k , respectively. The energy efficiency η based on the first law of thermodynamics relates the energy content of all useful products and services to the energy input:

$$\eta = \frac{\sum_i \dot{m}_i \cdot LHV_i + \sum \dot{W}^- + \sum \dot{Q}^-}{\sum_k \dot{m}_k \cdot LHV_k + \sum \dot{W}^+ + \sum \dot{Q}^+} \quad (3)$$

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 whereas \dot{W}^- and \dot{W}^+ denote similarly the mechanical power (mainly in the form of electricity) entering or leaving the system.

In a similar way the exergetic efficiency η_{ex} can be defined based on the exergy flows within the process:

$$\eta_{ex} = \frac{\sum_i \dot{m}_i \cdot e_i + \sum \dot{E}_w^- + \sum \dot{E}_q^-}{\sum_k \dot{m}_k \cdot e_k + \sum \dot{E}_w^+ + \sum \dot{E}_q^+} \quad (4)$$

The specific exergy e_i of a fuel or material flow i is the sum of the physical, chemical, kinetic and potential exergy in relation to a defined environment. A common practice is to use atmospheric conditions as reference environment as for example proposed by (Kotas, 1985; Szargut *et al.*, 1988) and given in Table 6.

TABLE 6. Standard reference conditions for exergy analysis (Kotas, 1985).

Relevant species for this study	Standard Partial Pressure in the Environment [bar]	Standard chemical exergy e_{ch}^0 [kJ/kmol]
CO ₂	0.000294	20 170
H ₂ O (g)	0.0088	11 760
N ₂	0.7583	720
O ₂	0.204	3 970
Reference temperature T_0 [K]		298.15
Reference pressure P_0 [bar]		1.10325

For energy analysis of steady-state processes the kinetic and potential exergy are usually neglected. The physical exergy e^{ph} is defined according to:

$$e^{ph} = (h - h_0) - T_0(s - s_0) \quad (5)$$

The chemical exergy of a gaseous mixture e^{ch} can be determined by

$$e^{ch} = \sum_i x_i e_{i,ch}^0 + RT_0 \sum_i x_i \ln(x_i) \quad (6)$$

with $e_{i,ch}^0$ being the standard chemical exergy of component i in the mixture as a function of the defined reference environment and x_i the molar fraction of component i . For gas species present in the reference environment the standard chemical exergy is a function of their partial pressure, whereas for other species the value can be calculated based on the reference species and Gibbs free energy of formation. Tabulated values for standard components are also given in the literature (Szargut *et al.*, 1988). The chemical exergy of biomass can be determined based on the heating value according to the method by Szargut (Szargut *et al.*, 1988).

The exergy flow of work \dot{E}_w is equal to the mechanical power or electricity and the heat exergy flow \dot{E}_q is related to heat energy flow by the Carnot factor according to:

$$\dot{E}_q = \theta \cdot \dot{Q} = \left(1 - \frac{T_0}{T}\right) \cdot \dot{Q} \quad (7)$$

Exergy represents the maximum theoretical amount of work that can be extracted from a system in exchange with the environment by fully reversible processes (Kotas, 1985). Exergy based performance indicators can therefore be efficiently used to illustrate thermodynamic limits for various processes and concepts, avoiding the problem of the energy efficiency that values all energy flows on an equal basis.

Note that these indicators are dependent on the definitions of system boundaries; 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, are 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.

In order to investigate the performance of new biomass-based processes integrated in existing energy systems a system level efficiency η_{sys} comparing all energy streams leaving and entering the process on a primary energy basis has been applied in this work. Eq. (8) provides an example of a suitable definition that accounts for electricity (\dot{W}_{el}) and heat (\dot{Q}) as possible by-products or input to the process:

$$\eta_{sys} = \frac{\sum_i \dot{m}_i \cdot LHV_i + \frac{\dot{W}_{el}^-}{\eta_{el,ref}} + \frac{\dot{Q}^-}{\eta_{q,ref}}}{\sum_k \dot{m}_k \cdot LHV_k + \frac{\dot{W}_{el}^+}{\eta_{el,ref}} + \frac{\dot{Q}^+}{\eta_{q,ref}}} \quad (8)$$

This definition makes it necessary to account for the energy efficiencies of the reference processes ($\eta_{el,ref}$ and $\eta_{q,ref}$) within the background energy system the process is to be integrated into. Figure 20 illustrates the underlying concept for the system level efficiency definition. Note that \dot{W}_{el} and \dot{Q} can only figure either in the numerator or denominator of eq. (8) as only net flows are accounted for. Distribution losses associated with export and import flows are not accounted for. The definition of the reference technologies with their respective efficiencies is a non-trivial task. For this purpose, a consistent definition of the energy market background is necessary taking into account the various interdependencies of energy services, fuel prices and technologies. A tool developed within the author's research group was used for this purpose and is described in further detail in Section 4.3.2.

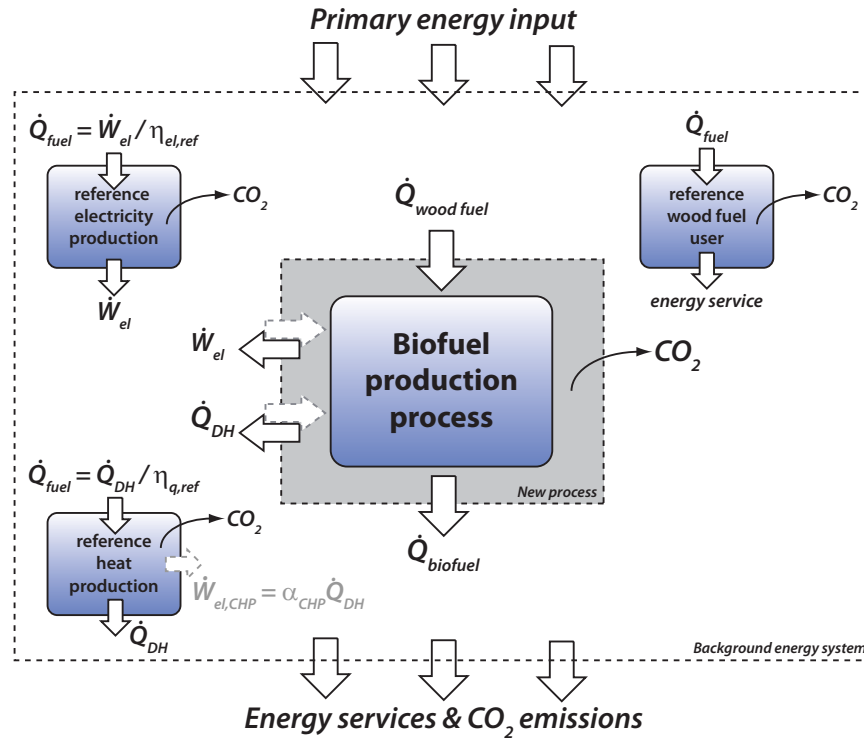


FIGURE 20. Schematic representation of the underlying concept for performance indicator definition.

Carbon footprint indicators

With given background technologies it is in addition possible to evaluate the carbon footprint consequences for a process that is considered for implementation in an existing energy system. The change in CO₂ emissions within the energy system $\Delta CO_{2,\text{sys}}$ can be evaluated per energy unit of biofuel supplied n_{biofuel}^- according to:

$$\Delta CO_{2,\text{sys}} = \frac{\Delta n_{\text{bio}} \cdot c_{\text{bio}} - \Delta n_{\text{el}} \cdot c_{\text{el}} - \Delta n_{\text{q}} \cdot c_{\text{q}} - n_{\text{biofuel}}^- \cdot c_{\text{fossil fuel}}}{n_{\text{biofuel}}^-} \quad (9)$$

This definition accounts for the emissions associated with the increased use of biomass ($\Delta n_{\text{bio}} \cdot c_{\text{bio}}$), the co-production of electricity and heat ($\Delta n_{\text{el}} \cdot c_{\text{el}}$ and $\Delta n_{\text{q}} \cdot c_{\text{q}}$) with associated indirect CO₂ emission effects of replacing the reference technologies with corresponding specific emissions ($c_{\text{el/q}}$), as well as the CO₂ consequences of replacing a fossil fuel alternative ($n_{\text{biofuel}}^- \cdot c_{\text{fossil fuel}}$). For Bio-SNG the fossil fuel alternative within this work has been natural gas. An important underlying assumption considering this analysis is the fact that biomass is considered a limited resource in the energy market scenarios. Biomass use in a novel process has to be analysed against the most likely alternative application for

biomass use in a given scenario. More details on the energy market scenarios and underlying assumptions will be presented in Chapter 4.3.2.

Economic indicators

Finally the economic indicators used throughout this work are the investment opportunity and the production costs per energy unit of biofuel. The investment opportunity IO is a simplified measure avoiding the necessity of determining capital costs of equipment but rather identifying the economic margin for investment based on the estimated fuel market price levels and the process consumption/production. Specific to the biofuel production the investment opportunity can be expressed as:

$$IO = \frac{\text{revenues} - \text{operating cost}}{\text{biofuel production}} \quad (10)$$

The investment opportunity IO is a crude screening tool used for primary selection of economically interesting process options. In order to be able to make a more qualified judgement for process selection, investment cost estimations are necessary. Based on available investment costs, the specific production costs per energy unit of biofuel c_{biofuel} can be determined as a function of annualized investment costs aC_F , operating costs and potential revenues. By-products that are to be considered include cogenerated heat and electricity. If it is assumed that biomass-based processes are included in the CO₂ emissions trading system, a revenue for capturing and storing CO₂ of biogenic origin can be accounted for in the revenues term. c_{biofuel} can thus be defined according to:

$$c_{\text{biofuel}} = \frac{aC_F + (c_{\text{op}} - c_{\text{sales}}) \cdot t_{\text{op}}}{\dot{m}_{\text{biofuel}} \text{LHV} \cdot t_{\text{op}}} \quad (11)$$

With c_{op} being the operating costs (labour, operation and maintenance, as well as material and commodity costs), c_{sales} the specific revenues from sales of the biofuel and eventual co-generated services and products, both costs being specific to time. t_{op} is the annual operating time of the plant. For the specific case of Bio-SNG production these production costs can then be compared to the costs for fossil natural gas c_{NG} for a given energy market scenario revealing the subsidy level c_{subsidy} necessary to make Bio-SNG production a viable alternative from an economic perspective.

$$c_{\text{subsidy}} = c_{\text{SNG}} - c_{\text{NG}} \quad (12)$$

4.3.2 Energy Market Scenarios and Estimation of CO₂ Emission Balances

As mentioned in the previous section, a tool developed at the Heat and Power Technology division – the Energy Price and Carbon Balance Scenario (ENPAC) tool (Harvey & Axelsson, 2010) – was used for defining consistent energy system backgrounds for analysing the performance of the proposed Bio-SNG production process schemes. The basic input and output data for the tool is shown in Figure 21.

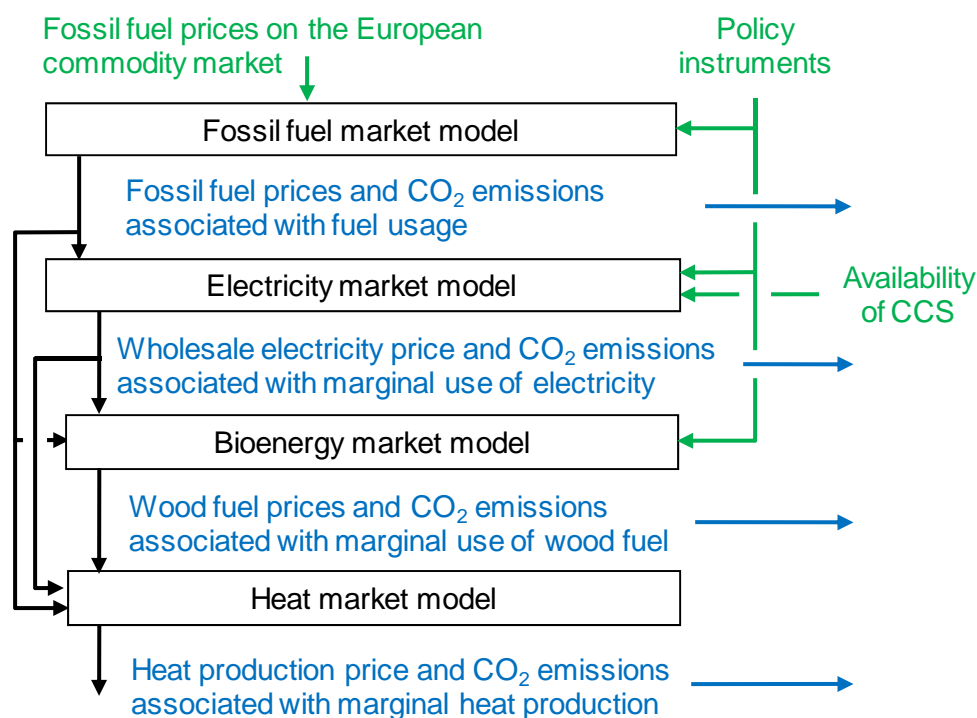


FIGURE 21. Representation of the major in- and output for the ENPAC tool (taken from Harvey and Axelsson (2010)).

This tool allows the determination of consistent future energy market scenarios based on fossil fuel price and CO₂ emission charge estimates in a European framework. Using the projected future costs of these two entities (from e.g. IEAs World Energy Outlook (IEA, 2011a)), 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 the energetic and economic performance, as well as CO₂ emission consequences of processes delivering multiple products and services such as the Bio-SNG process. Figure 22 illustrates the general concept of the ENPAC tool with different scenarios representing cornerstones of possible future developments assuming two levels for both CO₂ charge and fossil fuel prices.

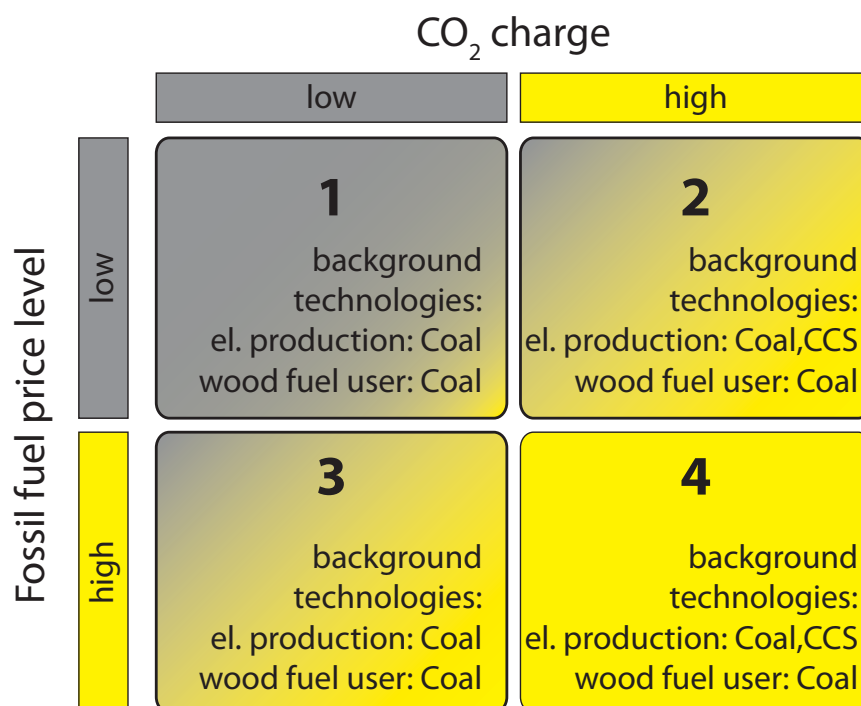


FIGURE 22. General concept of the Energy Market Scenario tool.

Figure 22 only serves for illustrative purposes and the input to the scenarios used in this work is not restricted to two price levels for CO₂ charge and fossil fuel prices only. Table 7 gives sample numerical values for the scenarios used in Paper V (note that the four scenarios do not correspond to the ones represented in Figure 22). The time perspective adopted for all developed scenarios is of medium term with projection for 2030 chosen as reference. The major focus of the tool is not to give exact numbers on the different energy commodity price levels but to cover the possible future energy market developments with scenarios that comprise consistent price levels for the different services and fuels. The use of the tool can be interpreted as a packaged sensitivity analysis on the influence of fuel prices on the performance of the process under consideration. Considering the carbon footprint evaluation the ENPAC tool uses a well-to-tank perspective accounting for all fuel emissions from the source to its application in the form of CO₂ equivalents.

TABLE 7. Energy market scenario data used in Paper V. Input values are based on IEAs World Energy Outlook 2011 (IEA, 2011a).

Parameter	Unit	Scenario 1 <i>current policies^a</i>	Scenario 2 <i>new policies^a</i>	Scenario 3 <i>450 ppm^a</i>	Scenario 4 <i>average^b</i>
<i>Fossil Fuel Price Level (Input)</i>					
Crude Oil	€ ₂₀₁₀ /MWh _{LHV}	63.2	55.2	45.6	54.4
Natural Gas	€ ₂₀₁₀ /MWh _{LHV}	36.1	33.5	27.8	31.9
Coal	€ ₂₀₁₀ /MWh _{LHV}	11.6	10.9	7.4	9.5
CO ₂ Charge (Input)	€ ₂₀₁₀ /t CO ₂	30.2	30.2	71.7	51
<i>Renewable Electricity Production Subsidy (Input)</i>	€ ₂₀₁₀ /MWh _{el}	20	20	20	20
<i>End User Prices and Policy Instruments</i>					
Wood Fuel (Forest Residue) c_{fuel}	€ ₂₀₁₀ /MWh _{LHV}	30.7	30.0	41.3	36.0
Electricity (incl. CO ₂ Charge) c_{el}	€ ₂₀₁₀ /MWh _{el}	68.2	66.9	86.3	78.9
Reference Electricity Production Technology		Coal	Coal	NGCC	Coal
Natural Gas (incl. CO ₂ Charge) c_{NG}	€ ₂₀₁₀ /MWh _{LHV}	48.1	45.5	49.1	48.6

^a Scenario names according to IEA's World Energy Outlook 2011 (IEA, 2011a) that form the major input to the current scenarios.

^b Average between extreme values of IEA's World Energy Outlook 2011 (IEA, 2011a) scenarios' fuel price and CO₂ charge levels.

5 Results & Discussion

5.1 Influence of Different Process Steps on Process Performance

In this section the results of the investigations on overall process performance changes with modifications in the Bio-SNG process setup are presented and discussed. The basic process setup presented in Figure 14 is the basis for the majority of the results with modifications as indicated in Table 4. The available excess heat for the base case as well as heat integration potential is illustrated in the Grand Composite Curve in Figure 23. The stream data for the base case is given in Table 14 in the Appendix.

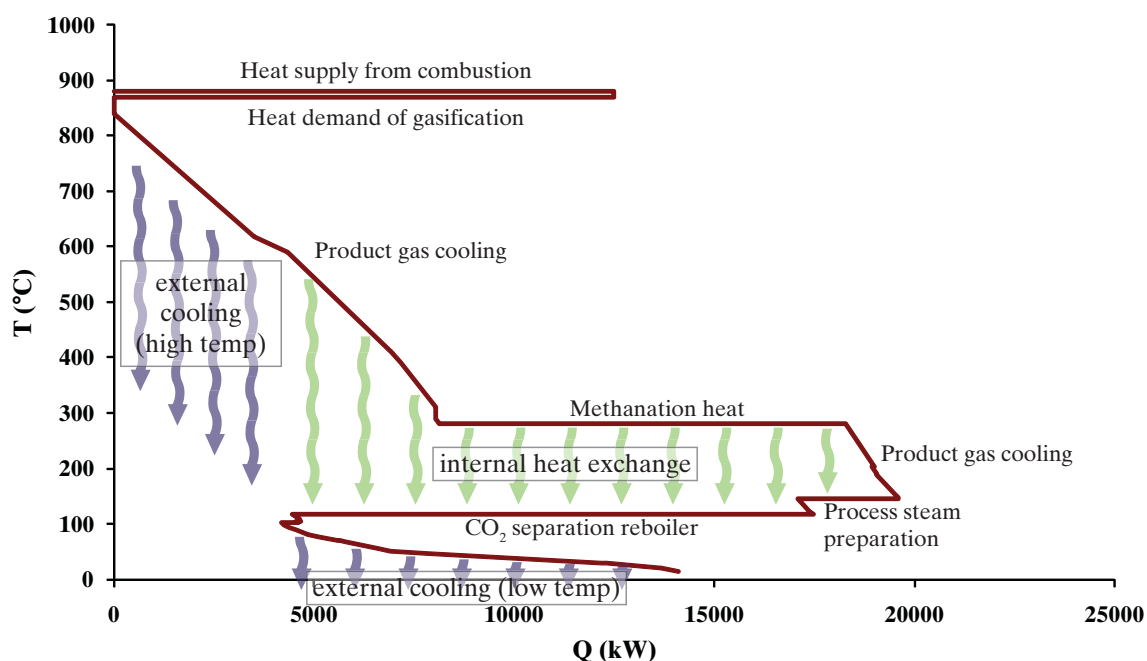


FIGURE 23. Grand Composite Curve for the base case Bio-SNG process with indication of the major heat sinks/sources.

5.1.1 Drying

Three different drying technologies for thermal integration within the Bio-SNG process have been investigated. In Paper II biomass 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 reactor with an existing fluidised bed boiler was assumed, the fuel feed to both gasification and combustion being assumed to be $100 \text{ MW}_{\text{LHV}}$ at 10 wt-% moisture. Excess heat from the Bio-SNG process suitable 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 Bio-SNG process that is present in Paper II. Figure 24 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 will differ when integrating the drying technologies to another background process.

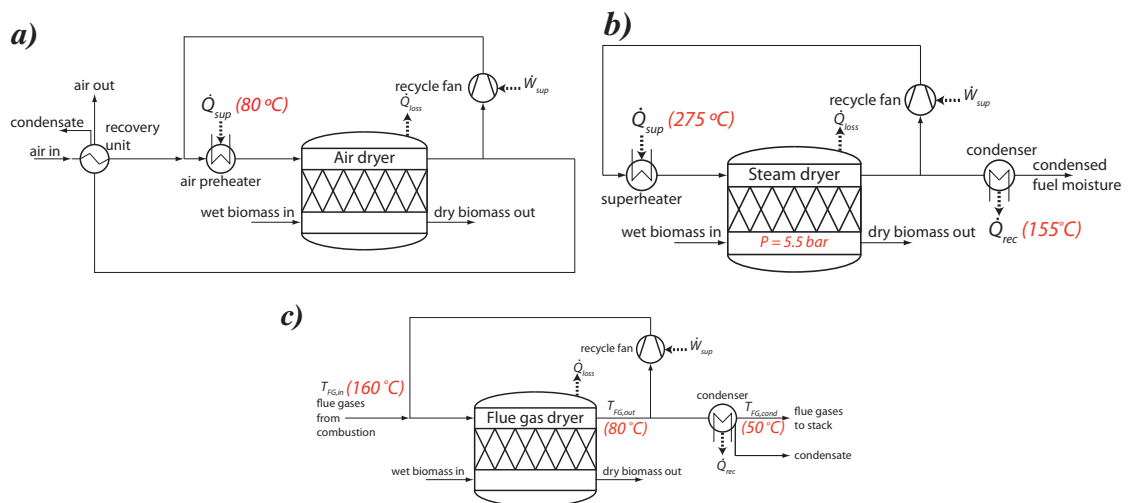


FIGURE 24. Drying technologies investigated for integrated feedstock drying. a) low-temperature air dryer; b) steam dryer; c) flue gas dryer (from Paper II).

Figure 25 illustrates possible heat integration of steam and low-temperature air drying with the Bio-SNG process. As heat streams from the combustion process are not included in Figure 25, the flue gas dryer using the boiler flue gases is not represented. No detailed modelling of the combustion and power generation was conducted in Paper II, therefore boiler flue gases were assumed available at 160°C , representing a common flue gas temperature level after air preheating.

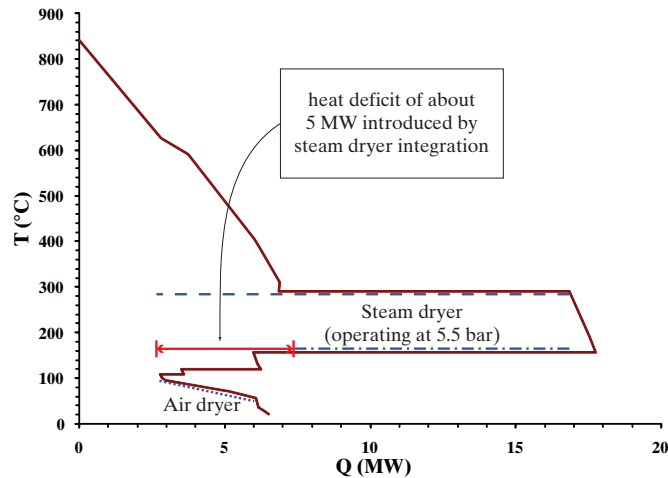


FIGURE 25. Illustration of the integration opportunities for steam drying and air drying within the Bio-SNG production process. Solid curve – Grand Composite Curve of Bio-SNG process; dashed curve – heat demand for steam drying; dash-dotted curve – recoverable heat from steam dryer condenser; dotted 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 Bio-SNG process caused by the difference in heat supply needed and heat recovered by condensation of the evaporated moisture. This deficit would have to be covered by another heat source such as steam extraction from the associated steam power cycle but has not been accounted for in Paper II. The Bio-SNG production process constituting the background was assumed to remain static as a simplification. The three drying technologies can supply 13.7% (air dryer), 47.7% (steam dryer) and 18.9% (flue gas dryer) of the total process dry fuel feed, respectively. 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 Bio-SNG of 7.4 percentage units from 59.4 to 52% when accounting for the additional biomass fuel supply needed for the external dryer. 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. However, the heat deficit caused by the steam dryer (cf. Figure 25) has not been accounted for in this study. 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.

5.1.2 Gasification and Gas Cleaning

In order to present a clear picture of the difference in performance for the two fluidised bed gasification technologies an exergy-based analysis using a simplified gasification reaction scheme was used. The major parameter investigated was the gasification pressure. The conversion in the downstream methanation process being improved by elevated pressure, operating the gasification unit under pressurised conditions may lead to improvements in overall energy performance compared to compressing the product gas from atmospheric gasification. The indirect allothermal gasification operates at atmospheric pressure with subsequent compression of the product gas whereas the direct autothermal gasification operates under pressurised conditions, supplying all incoming streams at the corresponding pressure. Varying both H_2/CO ratio and the relative air-to-fuel ratio λ for the gasification as additional parameters, a large range of operating conditions is screened. In order to reduce the influence of specific differences concerning reactor design and operating conditions (such as bed material choice) on the comparison, the simple stoichiometric model for the gasification step as described in Chapter 4.1 is used. This allows the two gasification technologies to be compared on a common basis. Figure 26 illustrates the two gasification concepts including simplified losses represented by pressure drop in the product gas chain for gas cleaning equipment and heat transfer exergy losses for heat exchange with a connected steam cycle.

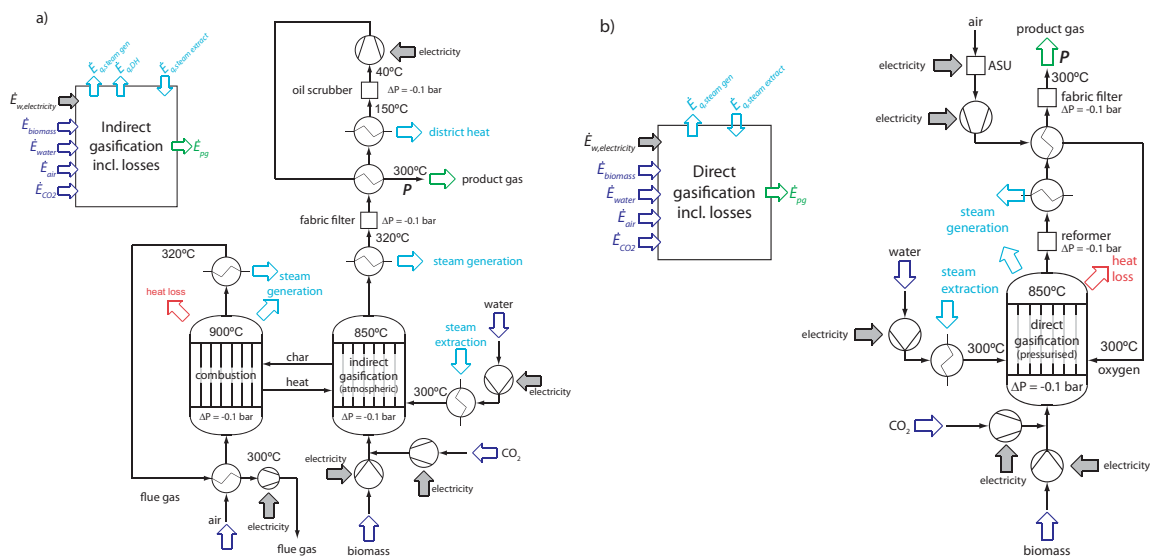


FIGURE 26. Indirect (a) and direct (b) gasification – system definition and associated exergy streams accounted for in efficiency calculations accounting for losses.

For an ideal case assumption without losses in heat exchange and compression processes, Figure 27 illustrates two cases of H_2/CO ratio in the product gas at a relative air-to-fuel ratio of 0.35. In these ideal cases both atmospheric and pressurised operation have been investigated for each of the two fluidised bed gasification technologies. The figure clearly illustrates that the increase in exergy efficiency with increasing pressure is limited to just above 3 %-points when the product gas delivery pressure is increased from 1 to 30 bar. The views at magnified y-scale given in Figure 27 also indicate that the pressurised systems – the complex pressurised indirect gasification system even outperforming direct gasification – perform considerably better than the atmospheric systems. The major increase in efficiency compared to 1 bar is achieved for a pressure in the range of 10 bar whereas a further increase in pressure only results in limited improvement in exergy efficiency.

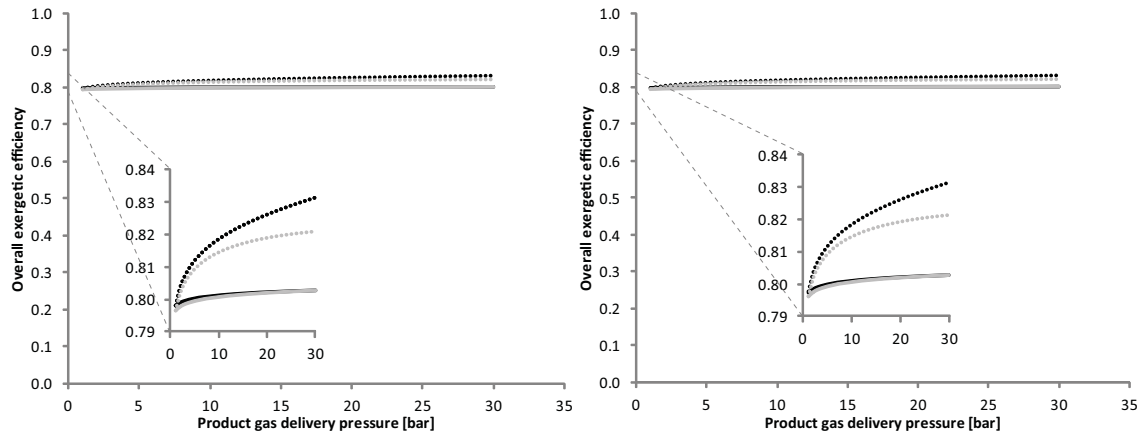


FIGURE 27. Exergy efficiency of ideal systems at $\lambda = 0.35$ for two H_2/CO ratios (left: $H_2/CO = 2$, right $H_2/CO = 3$). black lines – indirect gasification, grey lines – direct gasification, solid lines – atmospheric, dashed lines – pressurised.

Accounting for exergy losses in associated with heat exchange, compression, as well as supply of feed streams, the exergy efficiency of indirect gasification concepts is basically constant over the investigated pressure range as illustrated in Figure 28. Direct gasification exergy efficiency increases with pressure in a similar way to the ideal case, with pressurisation up to 10 bar achieving higher benefits while further increase in pressure only results in a small relative increase of the exergy efficiency. For the cases including losses a simplified estimation of the exergy losses associated with CO_2 removal has been performed as well. The exergy efficiency values for these cases $\eta_{ex,losses,CO_2}$ indicates that direct pressurised gasification is penalized harder and drops below the values obtained for indirect atmospheric gasification. This is due to the fact that the CO_2 concentration is substantially higher in the product gas from direct gasification as the products of the combustion reactions providing heat for gasification are present in the product

gas. In addition, using CO_2 as inertisation medium for the feeding system, a higher gasification pressure in the gasification reactors will lead to a higher CO_2 intake.

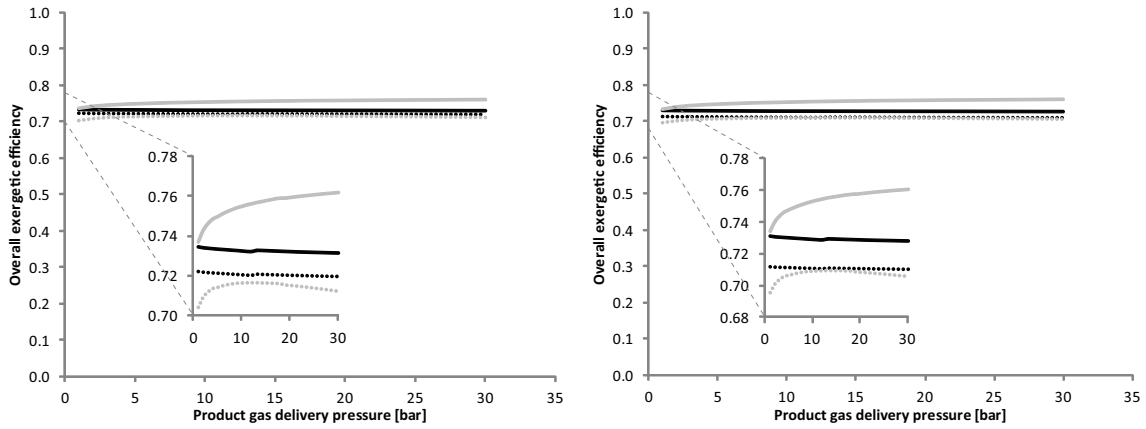


FIGURE 28. Exergy efficiency of the two systems including losses $\eta_{ex,losses}$ at $\lambda = 0.35$ for two H_2/CO ratios (left $\text{H}_2/\text{CO} = 2$, right $\text{H}_2/\text{CO} = 3$). Black lines – indirect gasification, grey lines – direct gasification. Dotted lines represent modified exergy efficiency $\eta_{ex,losses,CO_2}$ accounting for CO_2 separation exergy penalty.

It can be concluded that neither direct nor indirect gasification can be identified as intrinsically superior for Bio-SNG production based on the results presented. The key aspect for performance of Bio-SNG production processes is the efficient heat integration and cogeneration of power rather than the choice of gasification technology. In particular the conversion of high temperature process heat to steam for power generation contributes considerably to the exergy output from the process. Given the assumption that steam generation is not limited by material issues or heat recovery limitations related to tars or other impurities the internal electricity consumption of the gasification process can be covered as illustrated in Figure 29. The specific steam exergy output from the heat recovery w_{steam} exceeds the process' specific electricity consumption w_{gasif} over the whole pressure range. It might even be feasible to use excess exergy available and transfer it back to the gasification process further increasing the yield and efficiency in consequence.

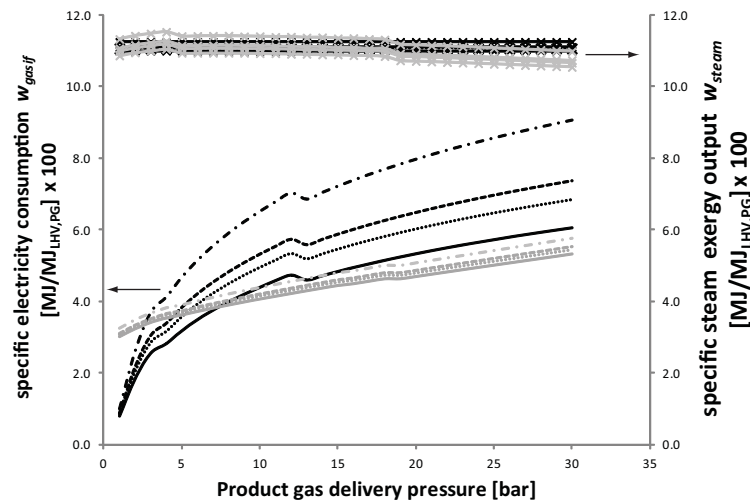


FIGURE 29. Specific electricity consumption w_{gasif} (plain curves) and steam exergy output w_{steam} (curves with markers) for the two gasification technologies at $\lambda = 0.35$ (black: indirect gasification, grey: direct gasification; Solid line – CO max, dotted line $H_2/CO = 2$, dashed line $H_2/CO = 3$, dash-dotted line H_2 max).

5.1.3 Methanation

An integration study investigating the potential power production from recovered process excess heat based on the Carnot efficiency indicates that both fixed bed adiabatic and fluidised bed isothermal methanation perform equally well for the given Bio-SNG process. An output of Bio-SNG of 63.3 MW_{LHV} per 100 MW_{LHV} dry fuel input and a ratio of about 1.2 between theoretical power production and overall power consumption is determined. The theoretical power production was estimated based on the Carnot based Grand Composite Curves for the two process alternatives, whereas the power consumption is derived from the process modelling results. Figure 30 illustrates the Carnot representation of the Grand Composite Curves for the two Bio-SNG process alternative based on indirect gasification. The grey shaded area represents the exergy recoverable, which is proportional to the potential electricity generation from process excess heat.

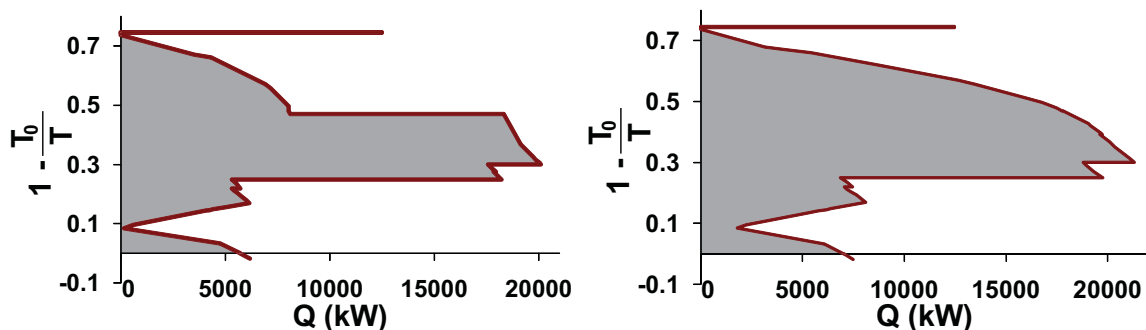


FIGURE 30. Carnot representation ($T_0 = 20^\circ\text{C}$) of the resulting grand composite curves for the Bio-SNG processes. Left: isothermal fluidised bed methanation. Right: adiabatic fixed bed methanation.

As presented in Table 8 the electric power generation potential for the process alternative using adiabatic fixed bed methanation is higher (8.3 MW) than for the process with isothermal methanation (6.6 MW) but in turn the estimated power consumption is higher as well. The ratio between power generation potential and estimated power consumption in consequence is similar, both being around 1.2, as already stated.

TABLE 8. Performance data for the Bio-SNG process using the two alternative methanation technologies.

Parameter	Isothermal Methanation	Adiabatic Fixed Bed Methanation
Dry Biomass Fuel Input ^a [MW _{LHV}]	100	100
Bio-SNG Production [MW _{SNG,LHV}]	63.3	63.3
Carnot-based Power Production Potential Absolute [MW] / Specific [kW/MW _{SNG,LHV}]	6.57 / 103.8	8.28 / 130.8
Process Power Consumption Absolute [MW] / Specific [kW/MW _{SNG,LHV}]	5.45 / 86.1	6.75 / 106.6
Ratio Power Production Potential/Power Consumption [-]	1.21	1.23

^a at 20 wt-% moisture

The two process alternatives do not differ significantly with respect to energy performance based on these results. It should however be noted that the difference between specific power production potential and consumption per produced energy unit Bio-SNG is larger for fixed bed methanation, resulting in possible benefits for that technology at larger scale. The gas quality in terms of CO concentration and Wobbe index generated does not differ significantly between the two methanation technologies, based on the given models. Fixed bed gasification needs to be operated at higher pressure (18 bar) in order to reach the desired high carbon monoxide conversion of 99.9% while fluidised bed gasification only needs slight pressurisation (2.5 bar). From a technical viewpoint fluidised bed gasification has the advantage of no need for recycle streams and a simultaneous water-gas shift reaction avoiding the need for an extra reactor vessel. In addition, fluidised bed methanation technology has been demonstrated to have reduced risk for carbon deposition and consequent catalyst deactivation due to the regenerative character of the fluidised bed (Seemann *et al.*, 2006). On the other hand, fixed bed gasification is a simpler reactor concept and large scale experience from coal gasification projects is available.

5.1.4 Gas upgrade

A detailed comparison of three alternative CO₂ separation technologies – amine-based chemical absorption (MEA), membrane-based separation, and pressure swing adsorption (PSA) – illustrates the advantages and drawbacks of the alternatives from a techno-economic viewpoint. The scale of the plant investigated is a thermal input to the gasifier of 100 MW_{th,LHV} at 20 wt-% moisture content with a preceding drying step resulting in a biomass thermal input of 90.3 MW_{th,LHV} at 50 wt-% moisture. Table 9 provides an overview of the energy flows associated with the three technologies for CO₂ separation, even considering biogenic CO₂ storage for the MEA and membrane cases. As for the PSA process alternative, the CO₂-rich off-gas is released at sub-atmospheric pressure, and the CO₂ storage option was not considered. Even though CO₂ from biomass-based processes is not currently considered within the emission trading system it might be in the time perspective of about 2030 adopted in this work.

TABLE 9. Overall energy balance for the different CO₂ separation technology alternatives (values in brackets refer to cases with CO₂ storage).

		MEA (with CO₂ Storage)	Membrane (with CO₂ Storage)	PSA
Fuel Input	MW _{LHV} ^a	90.3	90.3	90.3
SNG Production	MW _{LHV}	62.8	58.7	59.6
Gas Upgrade Section Power Consumption	MW	1.7 (3.1)	4.0 (5.4)	3.1
Overall Power Consumption ^b	MW	6.3 (7.7)	8.6 (10.1)	7.7
Power Generation	MW	10.7 (11.0)	14.3 (14.8)	13.8
Net Power Export	MW	4.3 (3.3)	5.6 (4.7)	6.1

^a 50 weight-% moisture

^b including gasification section, gas upgrade section and auxiliaries (taken as 2% of thermal fuel input)

Based on the energetic yield of SNG, amine-based absorption performs best among the three technology alternatives. The net power production is highest for the PSA case and lowest for the amine-based absorption. When evaluating the economic performance of the three technologies, MEA performs best among the three technologies, as illustrated in Figure 31. The figure also indicates that the differences between the three technology alternatives are moderate and that CO₂ capture and storage can even lead to increased Bio-SNG production costs, depending on the background energy market scenario assumed. The annuity factor used for the evaluation was 0.1, representing for example an estimated economic lifetime of 20 years and a fractional interest rate per year of 8%.

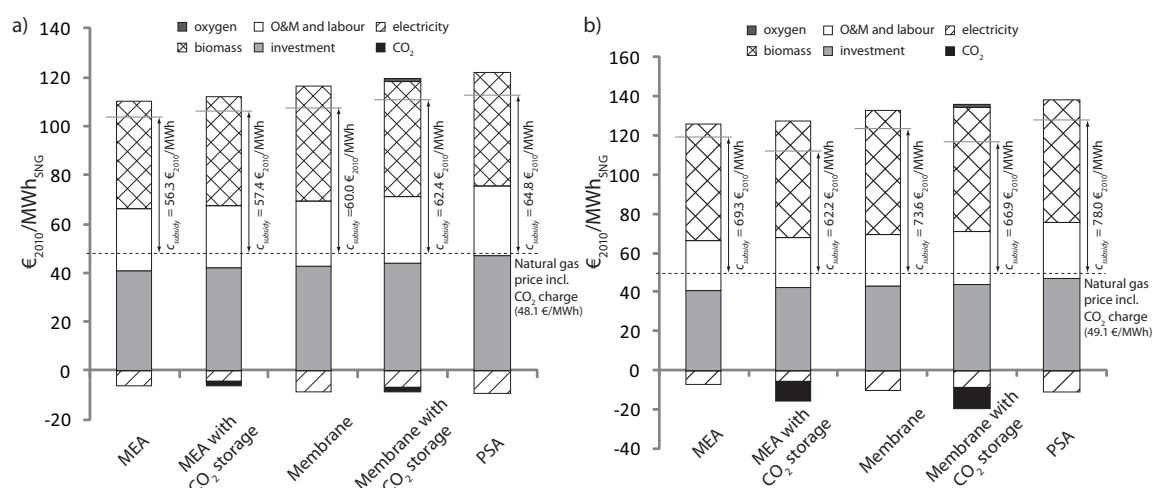


FIGURE 31. Contribution of the different cost items to the production costs for Bio-SNG for Scenario 1 (a) and Scenario 3 (b).

Common for all technology alternatives for CO₂ removal and in consequence for Bio-SNG production in general is the fact that the production costs by far exceed the natural gas price prevailing in the respective energy market scenario. Depending on both the scenario and the CO₂ separation technology a support for Bio-SNG production in the range of 56-78 $\text{€}_{2010}/\text{MWh}_{\text{SNG}}$ in some form is necessary to make the process concepts economically viable. It needs to be stated though that no taxes on fuel prices are included in the scenarios but only CO₂ charges. The largest contribution to the Bio-SNG production cost is the biomass feedstock cost of biomass and the investment cost, each standing for about one third. The breakdown of the total fixed capital investment cost is given in Figure 32. The contribution of the gas upgrade section including the CO₂ separation step varies between 13 and 22 % with PSA resulting in the highest contribution.

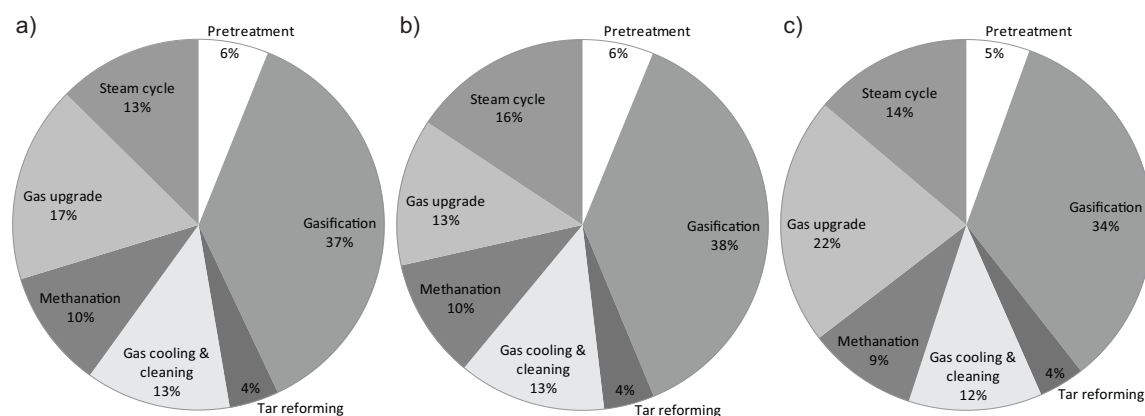


FIGURE 32. Distribution of total fixed capital investment cost (TFCI) between different plant parts for the three CO₂ separation technologies (all without CO₂ storage). a) MEA, b) membrane, c) PSA.

5.2 Integrated process concepts for Bio-SNG production

5.2.1 Rankine steam cycle integration

As illustrated in Section 5.1.3 a suitable tool for rapidly estimating the potential for cogeneration of electricity using process excess heat is the use of Carnot-based Grand Composite Curves. By assuming a conversion efficiency from exergy potential to real electricity production – that is based on a detailed steam cycle integration study (Paper III) – it is even possible to get a more realistic estimation of the cogeneration potential for electricity. This has been done for the study comparing different CO₂ separation technology alternatives for gas upgrade within the Bio-SNG process (see Section 5.1.4). In the framework of comparative process integration studies of the current level of detail this approach can be considered sufficiently accurate to lead to reliable conclusions. When aiming at a more detailed estimation of steam cycle integration, additional assumptions on the complexity and design of the steam cycle are necessary. The basic concept as well as results from a detailed study on integration of Bio-SNG production into existing infrastructure is presented in the next section.

5.2.2 Integration with existing infrastructure

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 Bio-SNG production, but also on illustrating options for gasification within the power sector – namely natural gas combined cycle power plants.

The Bio-SNG process has been considered for integration with an existing biomass CHP steam power plant, as described in Section 4.2.2 (cf. Figure 19). 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 33. It is based on a report on optimal design of biomass-fired heat and power plants (Steinwall *et al.*, 2002).

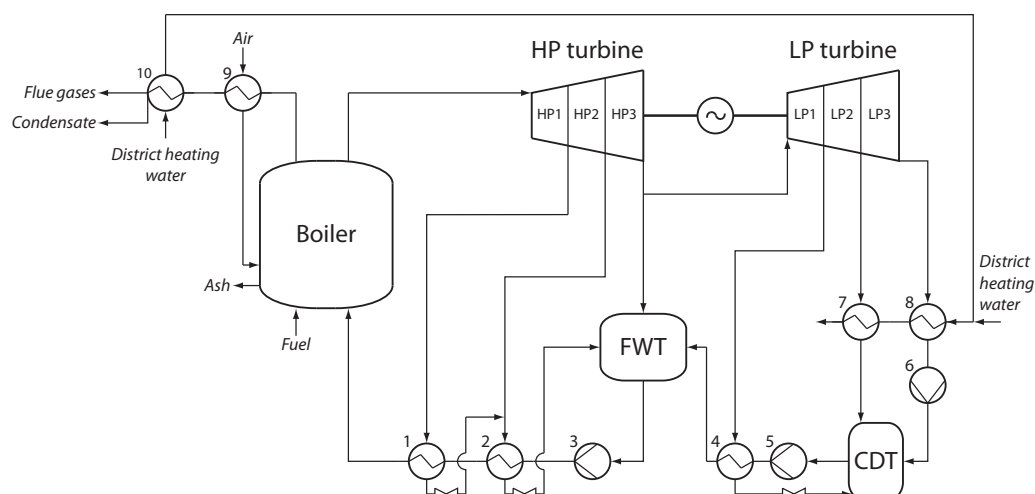


FIGURE 33. Schematic flowsheet of the biomass CHP plant design (based on (Steinwall *et al.*, 2002)). 1 - high temperature feedwater preheater, 2 - low temperature feedwater preheater, 3 - feedwater pump, 4 - condensate preheater, 5&6 - feedwater pump, 7 - high temperature DH condenser, 8 - low temperature 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 equipped with 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 34. 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 33). 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 34, the heat streams of the air preheating and district heat produced by condensing the flue gases (units 9 and 10 in Figure 33) have not been considered for integration with the Bio-SNG production process, but are considered unchanged.

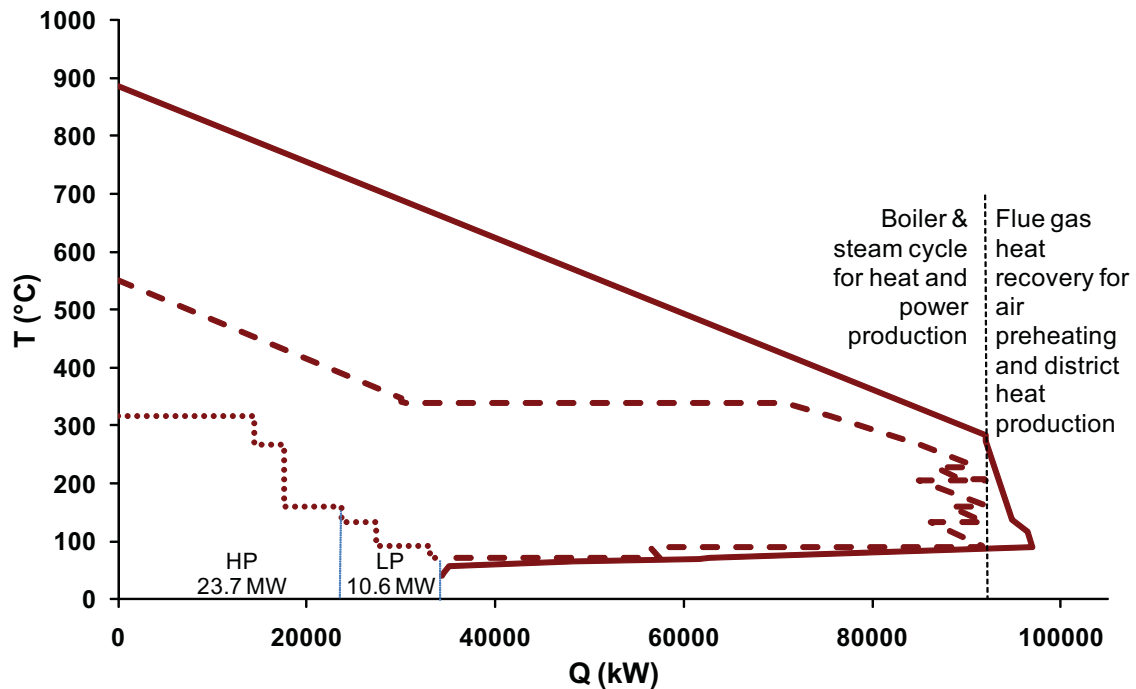


FIGURE 34. Pinch representation (GCC curves) of biomass CHP steam power plant. Solid curve: heat streams from boiler and district heating system, dashed curve: steam cycle heat streams, dotted curve: power production in turbine stages (Paper III).

For the Bio-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 5.1.1) 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.

Two different levels of thermal integration between the steam power cycle and the Bio-SNG process have been investigated. An illustration of these two different levels is given in Figure 35. 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 makes use of the high temperature excess heat from the Bio-SNG process for increasing the steam production. Internal heat recovery within the Bio-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 Bio-SNG process is used for high-quality steam generation. As a heat deficit within the Bio-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 Bio-SNG process heat demand.

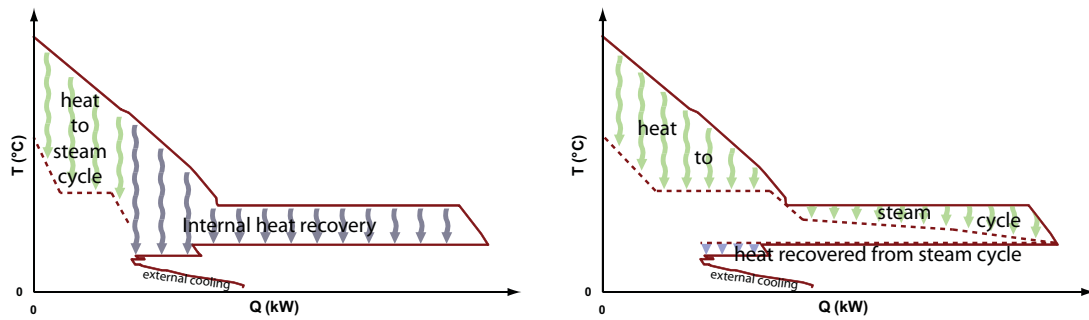


FIGURE 35. Illustrative example of the two levels of thermal integration between the Bio-SNG process and the steam cycle. a) balancing integration, b) maximum integration using heat-cascading. Solid curve: Bio-SNG heat stream representation, dashed curve: simplified steam cycle heat stream representation.

The Grand Composite Curves (GCC) of the Bio-SNG production process and the two drying alternatives are illustrated in Figure 36 and 37, respectively. The light grey shaded areas of Figure 36 and 37 represent the balancing integration cases in which the steam power cycle is integrated with the Bio-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.

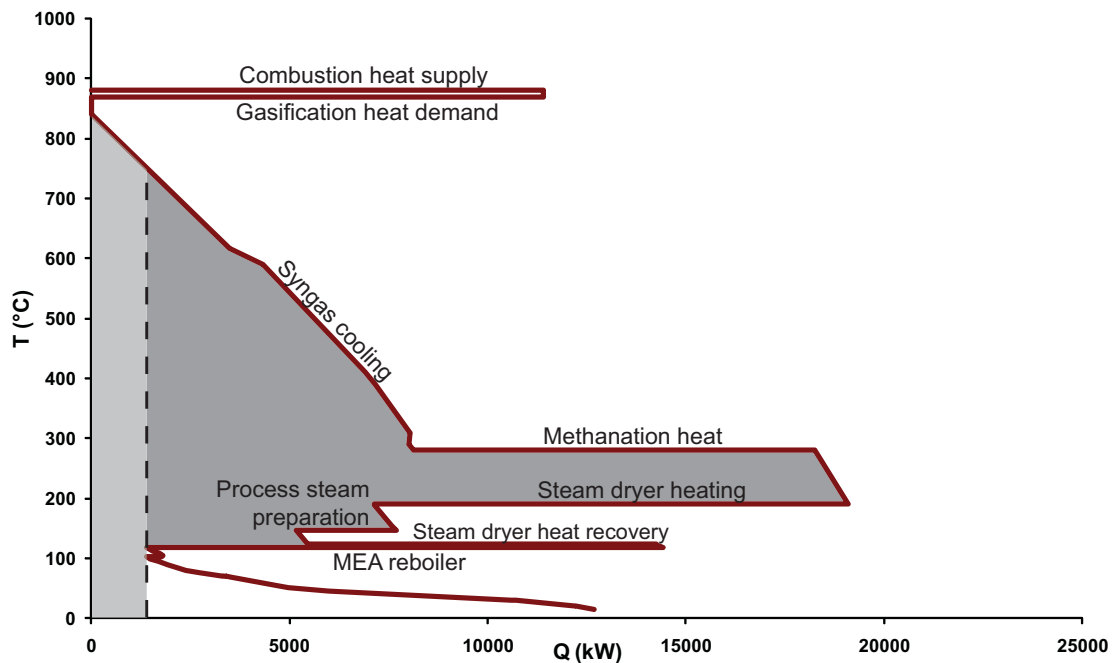


FIGURE 36. GCC for the Bio-SNG process alternative with steam drying. Light grey shaded area: case 1A – balancing integration, dark grey shaded area: case 1B – maximum integration (Paper III).

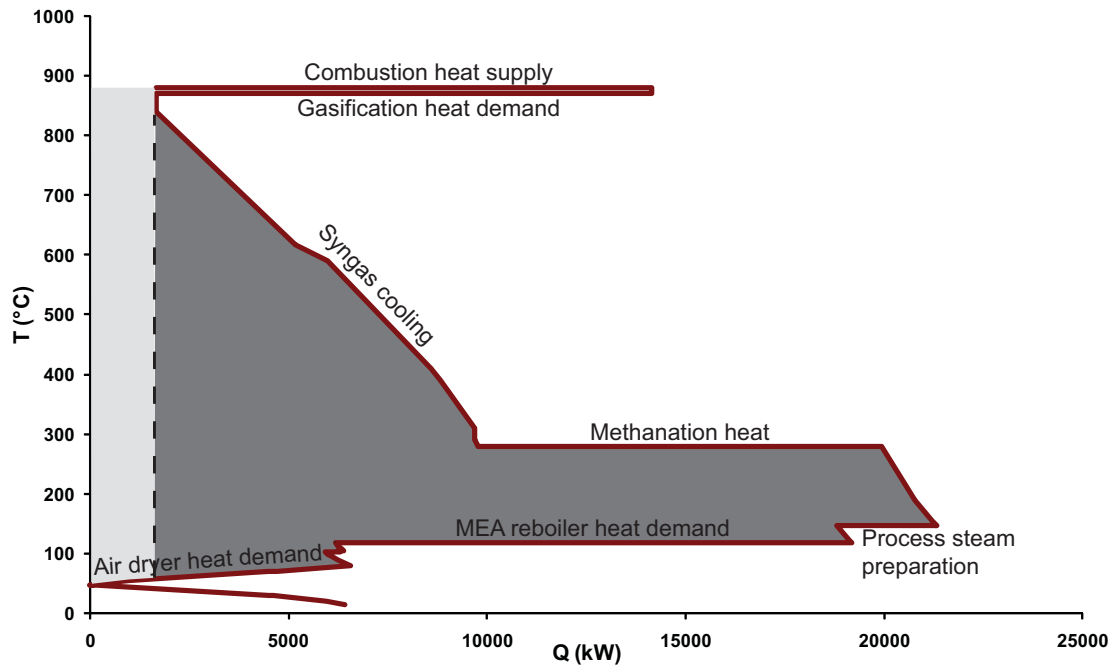


FIGURE 37. GCC for the Bio-SNG process alternative with air drying. Light grey shaded area: case 2A – balancing integration, dark grey shaded area: case 2B – maximum integration (Paper III).

As an example, Figure 38 illustrates the increased level of integration of the Bio-SNG process with air drying (case 2B). The solid line represents the heat streams from both the CHP steam power plant and the Bio-SNG process while the dashed line illustrates heat streams of the steam cycle integrated to a maximum extent with the Bio-SNG process, thereby making use of the large heat pocket represented by the dark grey shaded area in Figure 37. As the existing CHP steam power plant is associated with considerably larger heat loads, the heat pocket of the Bio-SNG process only appears as a relatively small nose in Figure 38.

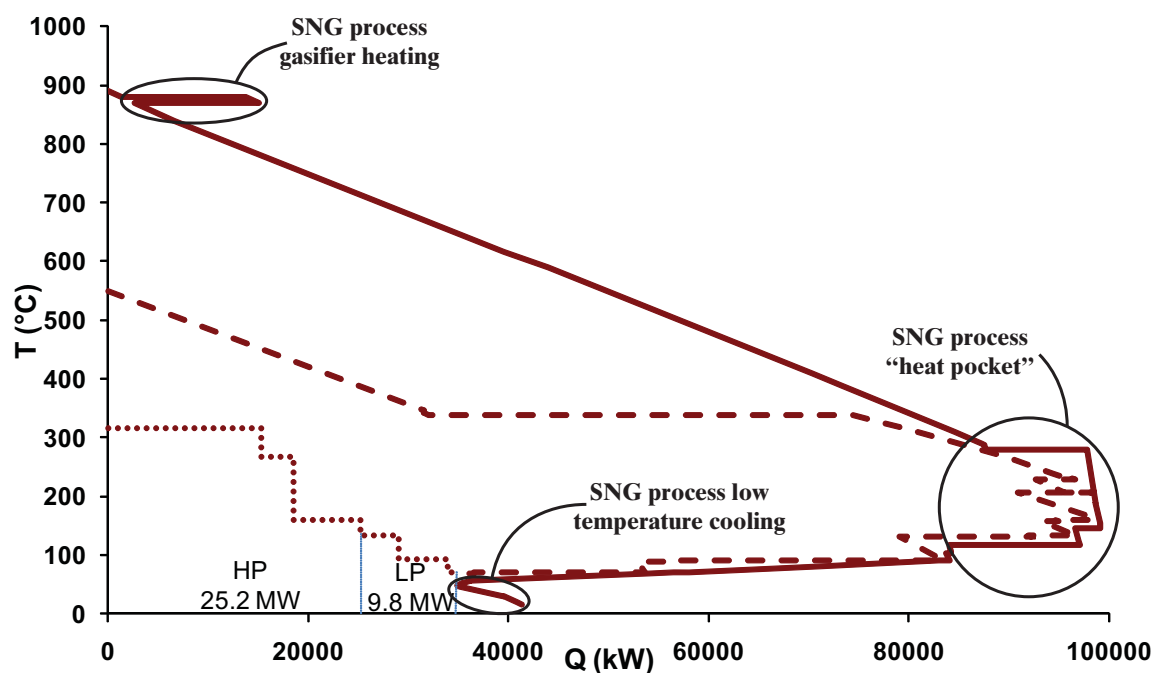


FIGURE 38. Pinch representation (GCC curves) of the increased integration case between the Bio-SNG process with air drying and the CHP steam power plant (case 2B). Solid curve: heat streams from boiler, Bio-SNG process and district heating system, dashed curve: steam cycle heat streams, dotted curve: power production in turbine stages (Paper III). The process streams representing the Bio-SNG process are highlighted.

The power production in the turbines increases slightly for case 2B illustrated in Figure 38 compared to the CHP stand-alone operation (c.f. Figure 34), 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 decreases.

Figure 39 illustrates the decrease in heat and power production from the steam cycle resulting from integration of the Bio-SNG production process. It also clearly illustrates the positive effect of an increased integration between the two processes on the electricity production. For the Bio-SNG process equipped with a steam dryer (cases 1A & B), a relative increase of 11.8 % can be obtained by maximising the thermal integration, while the increase for the Bio-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.

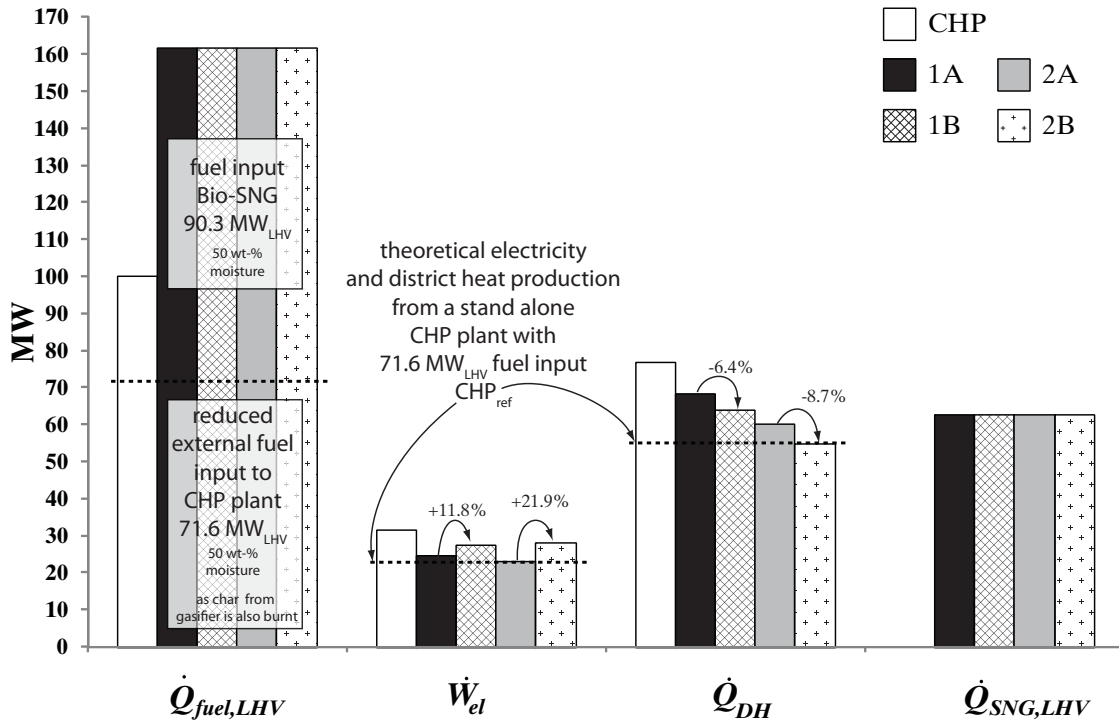


FIGURE 39. Fuel input ($\dot{Q}_{fuel,LHV}$), electricity (\dot{W}_{el}), heat (\dot{Q}_{DH}) and Bio-SNG ($\dot{Q}_{SNG,LHV}$) production for the integration cases studied (case 1A & B and 2A & B) and the stand-alone CHP steam power plant. CHP_{ref} indicates the theoretical amount of electricity and district heat produced from the biomass input to the CHP boiler alone for the four cases of integrated Bio-SNG production.

Figure 39 also shows that all Bio-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 39 (CHP_{ref})). It is possible to isolate the electricity and district heat production from the Bio-SNG process by assuming constant conversion efficiencies for the CHP plant both for the integrated and stand-alone cases. The electricity production allocated to the Bio-SNG production, $\dot{W}_{el,SNG}$, can be expressed as:

$$\dot{W}_{el,SNG} = \dot{W}_{el} - \dot{Q}_{fuel,CHP} \cdot \eta_{el,CHP} \quad (13)$$

where \dot{W}_{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 Bio-SNG process, $\dot{Q}_{DH,SNG}$, can be estimated as:

$$\dot{Q}_{DH,SNG} = \dot{Q}_{DH} - \dot{Q}_{fuel,CHP} \cdot \eta_{q,CHP} \quad (14)$$

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 Bio-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 Bio-SNG process $\dot{W}_{el,SNG}$ 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 10 gives an overview of the performance of the four different cases investigated.

TABLE 10. 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
$\dot{Q}_{fuel,CHP}$ [MW _{LHV}]	100	71.55 ^a	71.55 ^a	71.55 ^a	71.55 ^a
$\dot{Q}_{fuel,SNG}$ [MW]	-	90.33 ^b	90.33 ^b	90.33 ^b	90.33 ^b
\dot{W}_{el} [MW]	31.74	24.72	27.63	23.19	28.28
\dot{Q}_{DH} [MW]	76.81	68.42	64.06	60.11	54.91
\dot{Q}_{SNG} [MW _{LHV}]	-	62.7	62.7	62.7	62.7
η_{tot} [%]	108.6	96.3	95.4	90.2	90.1
$\dot{W}_{el,SNG}$ [MW]	-	2.0	4.9	0.5	5.6
$\dot{Q}_{DH,SNG}$ [MW]	-	13.5	9.1	5.2	0

^a reduced external fuel supply as the CHP boiler is supplied with non-gasified char from the indirect gasifier.

^b 90.33 MW_{LHV} at 50 wt-% moisture leading to 100 MW_{LHV} input to the gasifier after drying to 20 wt-% moisture.

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 Bio-SNG process with steam drying (case 1B) 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 Bio-SNG process – are not accounted for in these numbers.

5.3 Evaluation of integrated Bio-SNG process configurations

In the preceding sections the performance of different Bio-SNG process alternatives and the integration with another process (illustrated by integration with a CHP plant in this thesis) has been presented and discussed. This section extends the analysis to the surrounding energy system in order to evaluate the fuels and energy services entering and leaving the Bio-SNG process in relation to the existing energy conversion technologies in the surrounding energy system.

5.3.1 Energy efficiency

In order to illustrate the importance of the surrounding energy system for the energy performance, a comparative study on integrating Bio-SNG production in an existing energy system – both as stand-alone and integrated configurations – has been done. An important aspect was the clear definition of the system boundaries and the underlying assumptions for the evaluation. The life-cycle-perspective for the study (Paper V) was a well-to-tank perspective meaning that no specific application for the produced biofuel was considered. The case of Bio-SNG production that is used for illustration of the methodology in the results presented might be such an example as Bio-SNG is not limited to transport applications but might also replace fossil natural gas in any of its other applications within the power or chemical sector. Figure 40 illustrates the cases that have been investigated.

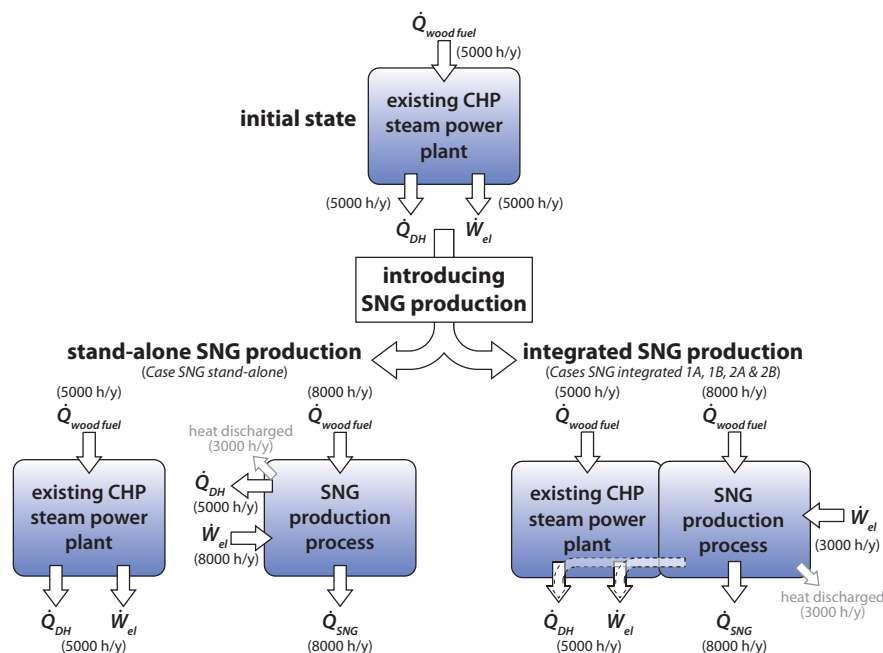


FIGURE 40. Illustration of the stand-alone and integrated operation for the SNG production process as extension of an existing CHP steam power plant.

An existing biomass CHP plant is either to be extended with Bio-SNG production or Bio-SNG production is implemented as stand-alone operation. The integration alternatives investigated are based on Paper III and four possible extensions with differing degree of thermal integration and two alternative biomass drying technologies prior to gasification are investigated. For the stand-alone case, a Bio-SNG plant with low temperature air-dryer is assumed. The Bio-SNG plant only provides heat to a district heating system with the CHP plant operating in the same way as before. The demand for district heat and electricity within the energy system is assumed to be constant. An increased generation from the new processes is thus assumed to result in reduced fuel consumption in the reference plants assumed for the production of the same energy services, and vice versa. Due to the limited operating time of the CHP plant that only supplies heat to the district heating system during cold periods (5000 h/y), excess heat is discharged to cooling water during 3000 h/y for all Bio-SNG cases. The limited operation of the CHP plant also implies that the integrated solutions can only cogenerate electricity during this period.

Table 11 illustrates the annual energy balances for the five alternatives as well as the change to the initial situation where only the biomass CHP plant is in operation. Annual energy figures are used instead of duties in order to account for the differing operating hours of the plants. Based on these figures it is possible to determine an energy efficiency η for the five cases according to eq. (3) (refer to Section 4.3.1). According to this performance indicator the stand-alone solution for Bio-SNG production achieves the highest energy efficiency of 91.5%. This number is the aggregated value for the Bio-SNG and the CHP plant as given in Table 11.

Table 11. Annual energy figures for the different process alternatives.

	<i>Wood Fuel</i>		<i>SNG</i>		<i>Electricity</i>		<i>District Heat</i>	
	GWh/y	Δ^a	GWh/y	Δ^a	GWh/y	Δ^a	GWh/y	Δ^a
<i>SNG Stand-alone</i>^b	1222	722	502	502	132	-27	485	101
<i>SNG Integr. 1A</i>^{b c}	1080	580	502	502	113	-45	342	-42
<i>SNG Integr. 1B</i>^{b c}	1080	580	502	502	128	-31	321	-64
<i>SNG Integr. 2A</i>^{b d}	1080	580	502	502	104	-55	301	-84
<i>SNG Integr. 2B</i>^{b d}	1080	580	502	502	131	-27	275	-110

^a change in annual production/consumption compared to initial state with existing CHP plant only

($Q_{wood\ fuel} = 500$ GWh/y, $W_{el} = 159$ GWh/y, $Q_{DH} = 384$ GWh/y)

^b the absolute energy figure numbers are for both the CHP and SNG plant

^c electricity consumption of 3.4 MW during SNG-only mode (3000 h/y)

^d electricity consumption of 4 MW during SNG-only mode (3000 h/y)

When extending the system boundaries to the surrounding energy system and applying the system efficiency concept described in Section 4.3.1, a system energy efficiency η_{sys} (eq. (8)) comparing all services and products output to the input on a primary energy basis can be determined and compared. Figure 41 illustrates the resulting values for the stand-alone solution and the four integrated cases, even giving the processes' energy efficiency η according to eq. (3). It can be seen that the ranking of the different solutions is dependent on the background energy system to a certain degree, meaning that dependent on the scenario, primary energy conversion to energy services and products differs for a given process.

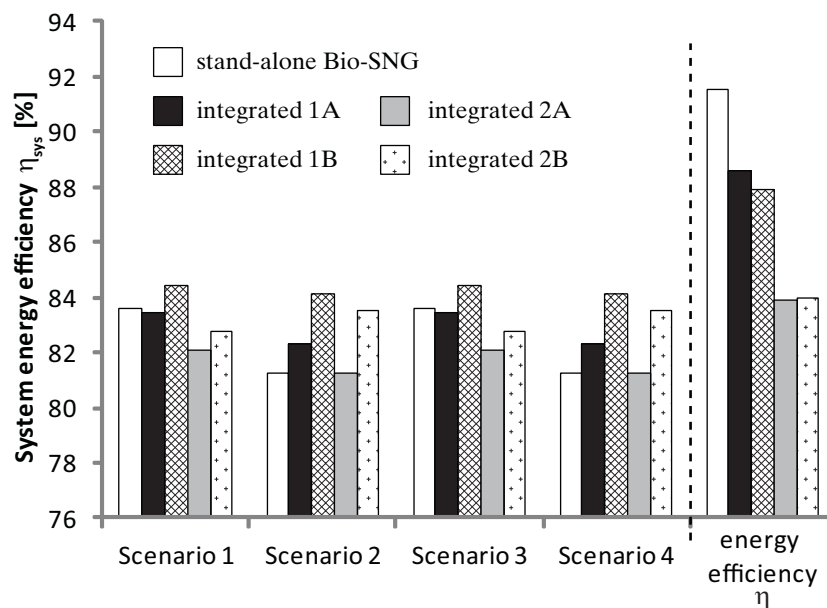


Figure 41. System energy efficiency η_{sys} and process energy efficiency η for the different Bio-SNG production cases in the four energy market scenarios.

For all scenarios an increased integration (going from cases A to B) leads to better system efficiency values due to an increased electricity production at the cost of decreased district heat supply. This is more pronounced for scenarios 2 and 4 that have a background electricity generation technology with lower efficiency (condensing coal power with Carbon Capture and Storage instead of standard condensing coal power). For scenario 1 and 3 three of the four integrated solutions even lead to decreased efficiency on a system level. The reason for that is the higher efficiency in the background electricity generation as well as the indirect effects on the energy system electricity balance due to the heat export. As the assumed background district heat delivery system is a combined heat and power plant, export of district heat will indirectly lead to a decrease in electricity generation from the background CHP plant that needs to be replaced. An underlying assumption for all these arguments is the fact that the electricity and heat demand in the considered energy system are assumed constant.

5.3.2 Carbon footprint

When analysing the CO₂ emission consequences of the introduction of Bio-SNG production it can be stated that all alternatives investigated in the study introduced in the previous section lead to an increase in annual CO₂ emissions for the given scenarios. For scenarios 1 and 3 where the reference electricity generation technology is coal-based condensing power without CCS, the integrated solutions result in a better performance, while for scenarios 2 and 4, a stand-alone plant is the better option. This is due to the substantially higher amount of external electricity production for the stand-alone solution that leads to lower CO₂ emissions when this electricity is produced in a power plant equipped with CCS technology. The associated CO₂ emissions for biomass use do not differ between the scenarios and therefore are not the reason for the different results between the scenarios. If the reference user of biomass is a coal condensing power plant, the emissions associated with additional use of biomass are approximated with emissions from coal combustion, regardless of whether or not the plant is equipped with CCS.

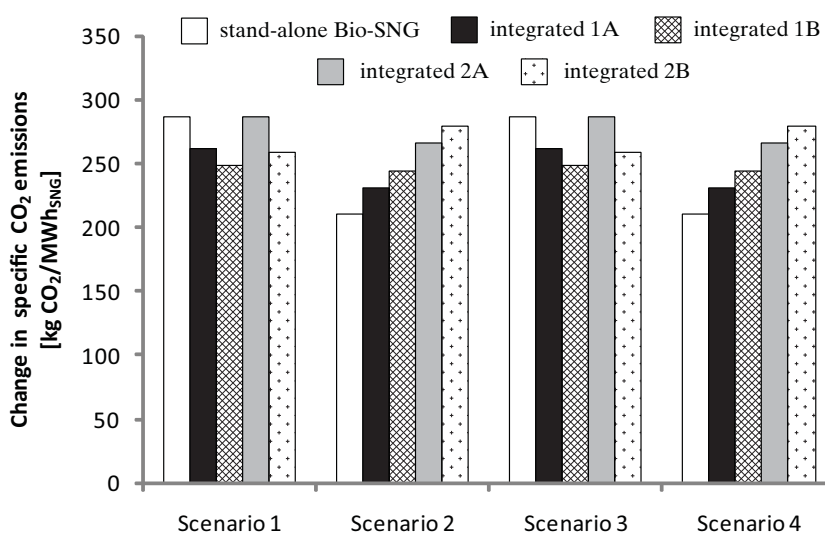


FIGURE 42. CO₂ consequences for the different Bio-SNG production processes in the four energy market scenarios.

The fact that all cases lead to an increase in CO₂ emissions can be explained by the fact that biomass is considered a limited resource, its use being associated with increased emissions from a reference process that is considered to be the most likely user of biomass for a given scenario. This reference user will have to replace the biomass that now used in a new process with a fossil alternative causing emissions that have to be accounted for. For all cases in this study, the reference user of biomass is assumed to be a coal power plant. This puts a large emission penalty on biomass to start with. In addition the SNG produced from biomass replaces fossil

natural gas having lower specific emissions than coal. The CO₂ balance for the Bio-SNG process can therefore hardly be positive with the given assumptions.

For the purpose of illustration, the specific change in CO₂ emissions is shown in Figure 43 for the case where the indirect emissions from the reference biomass user are not accounted for. This reflects the case where biomass is considered an unlimited resource. The emissions associated to biomass use are reduced to the greenhouse gas emissions related to harvest and fuel transport. It also results in negative specific emissions for the district heat c_q for the given study as this technology is biomass-based as well and the cogenerated electricity replaces fossil-based electricity. No CO₂ storage is taken into account for the figures represented in Figure 43 but nevertheless the change in specific emissions is negative. Adding CO₂ storage would further improve the results. In that case it would be important to not only take into account emissions due to fuel transport and harvest, but also account for indirect effects cause by land use change and carbon soil dynamics. Cherubini et al. (Cherubini & Ulgiati, 2010) indicated that these land use change effects can have a considerable influence on the greenhouse gas balance of biorefinery systems from crop residues. The fuel in mind for the proposed Bio-SNG process though will be forest residues where the land use change effects are considerably lower (Lindholm, 2010; Lindholm *et al.*, 2011). The greenhouse gas balance of harvesting forest residues accounting for soil carbon is shown to be heavily dependent – among other factors such as site productivity, geographical location – on the time-scale the evaluations are based on. In a long-term perspective little effect on soil carbon is to be expected from forest residues (Lindholm *et al.*, 2011).

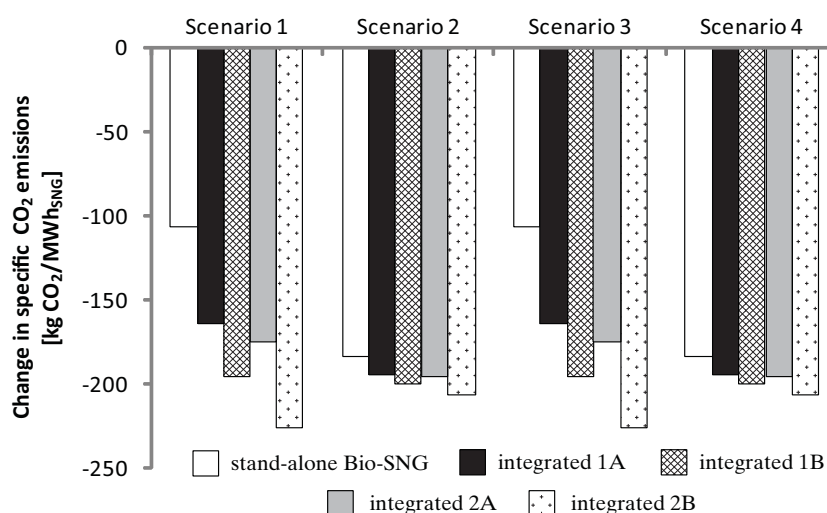


FIGURE 43. CO₂ consequences for the different Bio-SNG production processes considering biomass an unlimited resource.

5.3.3 Process economics

Economically, all SNG production alternatives are not feasible as such within any of the scenarios. The annual investment opportunity for the different cases given in Figure 44 shows very low values of several thousand € per GWh_{SNG} , making it impossible to finance such a project. The difference in investment opportunity ΔIO between integrated and stand-alone cases is negative for all cases and scenarios, rendering an integration of the two processes economically unattractive. These figures clearly demonstrate that the economic viability of SNG production (and biofuels in general) is largely dependent on the existence of specific support policies. No biofuel support policy has been assumed in the current study. The necessity of such a policy for rendering biofuel process alternatives economically interesting has been also been stated by Wetterlund and Söderström (Wetterlund & Söderström, 2010), among others. An additional factor influencing the investment opportunity of the integrated solutions negatively is the fact that the district heat delivery is decreasing. The decreased district heat production having to be compensated by external combined heat and power plants (e.g. a new CHP plant has to be built to cover the decreased heat delivery) puts high economical burdens on the integrated solutions. Such solutions therefore would only be viable in case of a decreasing heat demand on the end-user side or cheaper alternatives than building a new CHP plant for covering the deficit in DH production.

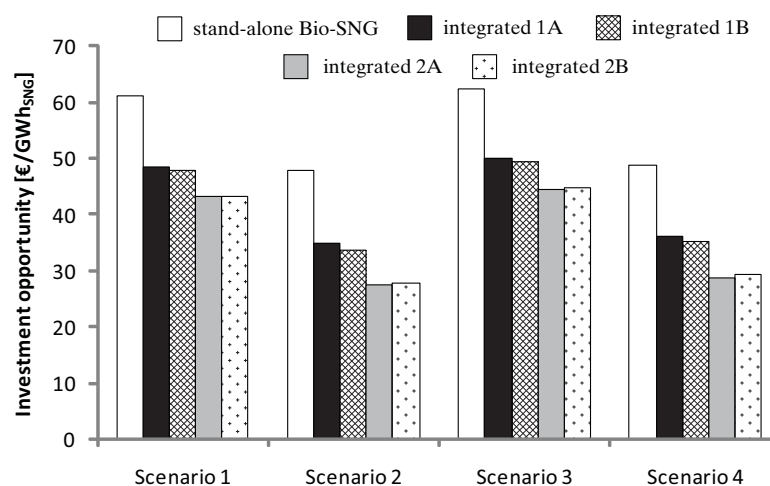


FIGURE 44. Specific investment opportunity IO for the different Bio-SNG production processes in the four energy market scenarios.

Based on the gas upgrade investigation performed and described in Section 5.1.4 a production cost estimate for Bio-SNG has been made. The results have already been presented in the corresponding section, but in order to set the results in a context a comparison to other studies investigating economic aspects of Bio-SNG production is presented in Figure 45. No correction for the currency time value has

been done in this figure as the uncertainty in the cost estimates is assumed to exceed the influence of the time value of money. In addition Figure 45 is intended to illustrate trends rather than to quantify specific numbers.

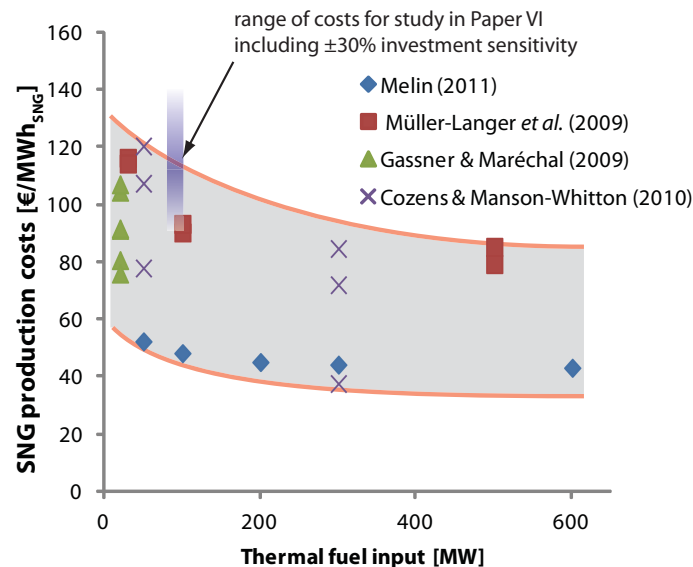


FIGURE 45: Bio-SNG production cost compilation from different sources (Cozens & Manson-Whitton, 2010; Gassner & Maréchal, 2009; Melin, 2011; Müller-Langer *et al.*, 2009).

The range of the production cost estimate presented in Paper VI is on the upper limit of the range reported in other literature studies. Since it was clearly stated that the estimate presented in this study is somewhat conservative, this is to be expected. The considerable differences in cost estimations for a single plant size presented by Cozens and Manson-Whitton (2010) are due to the use of different grades of biomass feedstock. Wood pellets result in highest costs, followed by wood chips, and finally, solid recovery fuel or refuse derived fuel give the lowest production costs. Hence, a substantial contribution to the costs for Bio-SNG is not only the capital investment but the fuel cost as well as costs for auxiliaries, in particular electricity that varies considerably between the different studies. Nevertheless, it is interesting to investigate the resulting price span reported in the given studies and relate it to the price projection for world market fossil natural gas given by IEA for 2030 of about 32-36 €/MWh_{LHV} (IEA, 2011a). The final local costs for natural gas might increase slightly due to transfer and transport costs, as well as specific taxes on natural gas; nevertheless this price level is below the lower range for Bio-SNG production given in Figure 45. Competing directly with the price of natural gas is hardly feasible for Bio-SNG, with the difference between biomass and natural gas price leaving little to no margins for investment (c.f. the low values for investment opportunity *IO* illustrated in Figure 44). Without specific incentives, price parity between natural gas and Bio-SNG will not be possible in the medium term. Alternatively, another reference fuel for setting the level of

maximum Bio-SNG production can be chosen. A study investigating the market opportunities for Bio-SNG presents a range of maximum possible production costs from about 37 to 101 €/MWh_{SNG} for a European context. The highest price levels for Bio-SNG are achieved when the competing product is assumed to be fossil gasoline. The transportation sector clearly is the focus application identified for Bio-SNG (Colmsjö & Nilsson, 2009). The highest level of that estimation is well in the range of the production cost estimates presented in Figure 45.

6 Conclusions

The results presented in this work clearly illustrate the importance of a holistic approach in order to identify efficient Bio-SNG production process alternatives, making use of the available excess process heat to produce power and deliver district heat. The main conclusions that can be drawn from the results of this thesis with respect to process synthesis, integration and performance are presented below.

6.1 Process synthesis

An important process step for Bio-SNG process synthesis is the feedstock drying prior to gasification. Low-temperature air drying and steam drying are both technologies that offer good opportunities for integration. Aiming at a maximum amount of cogeneration of electricity, low-temperature air drying performs slightly better than steam drying with 5.6 MW against 4.9 MW of potential for cogeneration of electricity from the Bio-SNG process excess heat for the investigated case with a wood fuel feed of 90.3 MW_{LHV} (50 wt-% moisture) producing 62.7 MW_{LHV} of Bio-SNG.

The two main thermo-chemical gasification technologies considered within this work are fluidised bed technology based. An exergy based comparison between indirect and direct fluidised bed gasification using a simplified gasification reaction model scheme indicates that the exergy efficiency from biomass to product gas prior to methanation is similar for both technologies when accounting for the necessary downstream CO₂ removal. The exergy efficiency of pressurised direct gasification is about 2-3 %-points higher than for atmospheric indirect gasification with subsequent pressurisation of the product gas for the maximum investigated pressure of 30 bar. However, when considering the penalty for downstream CO₂ removal, direct gasification is penalized more severely due to a higher CO₂ concentration in the product gas. The exergy efficiency value for both processes is in the same range, i.e. approximately 70-72% over the pressure range from 1 to 30 bar.

Methanation is the process step that is least critical from a process synthesis and integration perspective when choosing technology. Both available technologies perform similarly with respect to the ability to completely convert the product gas to methane and to make excess heat available for generation of steam. A comparative study estimating the co-generation potential for electricity from Bio-SNG production in relation to the process' power consumption indicates a ratio between cogeneration potential and electricity consumption of about 1.2 for both methanation technologies. Fixed bed adiabatic methanation might be considered slightly better suited for large scale production as the difference between specific power generation potential and actual specific power consumption is larger, allowing for increased cogeneration. Fluidised bed methanation on the other hand has been developed specifically for conversion of biomass based product gas.

Among the gas upgrade and CO₂ separation processes available at commercial scale, processes involving amine-based absorption perform best in comparison to membrane or PSA based separation technologies considering the conversion from biomass to SNG. From 90.3 MW_{LHV} wood fuel at 50 wt-% moisture, the process applying amine-based CO₂ separation produces about 63 MW_{LHV} of Bio-SNG, an increase of about 5% in comparison to the PSA-based process option, and 7% in relation to membrane based CO₂ separation.

6.2 Process integration

On an exergy-basis, the steam generation from excess heat corresponds to about 10% of the overall exergy leaving the gasifier unit, emphasising the importance of efficiently recovering this heat. Integrated solutions with e.g. existing biomass-fired combined heat and power plants may lead to favourable performance compared to stand-alone operation, depending both on the system boundaries chosen and the energy system background adopted. A general conclusion that can be drawn from the investigations in this thesis is the need to strive for a maximum thermal integration of the steam cycle. This refers to the increased use of high temperature process heat that is not freely available but used for internal heat transfer to a heat sink at lower temperature within the process. By recovering this high temperature heat to a steam cycle and providing heat to the process heat sink by e.g. steam extraction, increased cogeneration of electricity can be achieved. When not aiming at maximum integration but only making use of freely available excess process heat, the amount of cogenerated electricity from the Bio-SNG base case process investigated in this thesis drops to 0.5 MW (from 5.6 MW, steam dryer) and to 2.0 MW (from 4.9 MW, low-temperature air dryer), respectively.

6.3 Process Performance

Energy performance

Assumptions about energy market conditions in the surrounding energy system the Bio-SNG process is to be implemented into have a considerable influence on the process' system energy efficiency, describing the conversion of all process inputs to outputs on a primary energy level basis. For Bio-SNG production process alternatives integrated to an existing biomass CHP power plant, the choice of background energy system can alter the ranking of the process options according to the system energy efficiency. High values of electricity generation efficiencies in the background energy system can lead to a decreased performance of integrated Bio-SNG production process alternatives from an overall energy system perspective in comparison to stand-alone operation. For example, the system energy efficiency of 83.6% for a standalone Bio-SNG process exceeds the one for integrated process alternatives not fully exploiting the energy integration potential (82.1%), when the reference electricity in the background energy system is high ($\eta_{el,ref} = 51\%$ for the investigated case). However, integrated solutions with a maximum degree of energy integration are shown to outperform stand-alone solutions with respect to system energy efficiency even for energy background scenarios with high electricity generation efficiencies. Depending on the assumed energy system background, the system energy efficiency for solutions with maximum energy integration is 0.8 to 2.9% higher than for a stand-alone Bio-SNG process based on the investigated cases.

Economic performance

From an economic perspective the need for dedicated support schemes for biofuels in general and Bio-SNG in particular is clearly illustrated by the rather high production costs estimated, in the range of 103-127 €₂₀₁₀/MWh_{SNG}. A large share of the costs is related to the feedstock costs, illustrating the need for investigating low grade biomass feedstock for Bio-SNG production. Cogeneration of electricity enabling net export is shown to be a considerable contribution to cost reduction. The determined cost figures within this thesis can be used as a basis to investigate the effects of additional process energy improvements on the economic process performance.

Carbon footprint performance

In a similar manner, the carbon footprint of the Bio-SNG process is strongly dependent on the background energy system and its associated emissions. If Bio-SNG is primarily considered as a promising future transportation fuel option, it is

reasonable to calculate the carbon footprint performance by assuming that Bio-SNG replaces fossil alternatives such as gasoline or diesel. With the ambitious goals of reducing greenhouse gas emissions, there will be a large competition for biomass feedstock for replacing fossil fuels within numerous applications. Biomass in consequence will be a limited resource and the environmental benefits – mainly based on greenhouse gas emission reduction – from its use for a specific application need to be evaluated against a reference application. Prioritizing the use of biomass is a challenging task and complicated political issue. In the short term, the most substantial reductions of GHG emissions can be made by replacing coal in coal-fired power plants. Assuming coal-fired condensing power plants as competing user for the limited resource biomass, Bio-SNG production replacing fossil natural gas cannot outperform co-combustion of biomass fuel in coal power plants with respect to carbon footprint. Changing reference conditions with respect to both biomass reference usage and fuel replacement by Bio-SNG will however change these results.

7 Future research

7.1 Technical aspects

The foremost technical barrier for large scale deployment of biomass gasification for SNG production is the tar conversion or removal step. Available technologies inflict significant penalties on heat recovery for cogeneration as well as on the chemical energy content of the product gas. Commercial hot gas cleaning technologies are currently being developed for biomass gasification applications, which could lead to significant performance gains. From a technical perspective, it is important to determine whether a wet gas cleaning stage – implying that the product gas needs to be cooled and reheated prior to methanation – is still required in order to achieve sufficient particle removal. However, the exergy analysis performed within this work indicates that losses due to cooling and reheating product gas for cold gas cleaning including a scrubber are not that significant. Process data for a proven concept for hot gas cleaning and its evaluation from a process integration perspective are necessary in order to compare the two gas cleaning alternatives in more detail.

In addition to tars, the reducing environment in the product gas stream at the gasifier outlet and the presence of alkali metals and chlorine compounds imply a risk for corrosion when recovering high temperature excess heat for generation of steam for cogeneration of electricity (Källström, 1993; Viklund *et al.*, 2011). In connection to the tar problem, the material issue is an important question to be solved in order to fully exploit the process integration potential for biomass gasification based processes.

The gas upgrade for biomass gasification including reforming or removal of tars, trace component removal, and in particular CO₂ separation, often adopts technologies derived from solutions for fossil-based processes developed for refineries with significantly larger throughputs. When these processes are adopted at smaller scale, the specific costs become rather high as demonstrated in Section 5.1.4 for CO₂ separation technologies. The maximum scale of biomass based processes will never reach the range of fossil alternatives due to restrictions in

biomass logistics and gasifier sizing. New innovative technologies specifically adopted for biomass-based processes are needed that allow for building at scales in the several 100 MW range without putting a high penalty on capital and operating costs. Examples of technologies specifically adapted for biomass-based product gas include fluidised bed methanation technology or chemical looping reforming considered in part of the sections in this work.

Instead of removing CO₂ from the product gas, an interesting option is to add additional H₂ in order to completely convert the carbon in the biomass feedstock into a synthetic fuel, e.g. Bio-SNG. The Power-to-gas concept (ZSW, 2011), for example, uses renewable (inherently intermittent) electricity to produce H₂ which is thereafter used for methane synthesis from CO₂. System aspects and variations of this process concept could be an interesting topic of future research.

The focus of most studies investigating Bio-SNG production is on forest residues or other higher quality biomass fractions. With an increased demand for biomass fuels due to competing biomass users and competition for land-use for agriculture, lower quality fuels with higher level of impurities will have to be used. The impact of the increased amount of trace components in the product gas on the overall process design and energy performance is another topic that needs attention.

7.2 Process integration aspects

The heat recovery limitations for the product gas are a critical aspect from a process integration perspective that still needs investigation. Material issues have to be solved in order to be able to produce superheated steam using the hot product gas. Further investigations on how to exactly design the heat exchange network for example for indirect gasification units will doubtlessly be required. Further investigations will also be necessary to compare alternative investment options such as extending existing units or building combined and flexible combustion/gasification units as Greenfield plants.

Another interesting area of research is the integration of biomass gasification and Bio-SNG production with industrial production sites. Fossil natural gas use in the petro-chemical industry can for example be replaced by a renewable resource by introducing Bio-SNG production. Natural gas is commonly converted by steam reforming or partial oxidation to a syngas resembling the composition of biomass gasification product gas. With improved product gas cleaning and adjustment of the H₂/CO ratio the gas from biomass gasification could be used directly, avoiding chemical energy losses in the both the methanation step in Bio-SNG production and the steam reforming/partial oxidation on site. Practical advantages of using

Bio-SNG in the existing petro-chemical industry have to be weighed against energy efficiency aspects.

For Bio-SNG usage in the transportation sector, producing compressed or liquefied synthetic natural gas (CNG/LNG) is a way to decouple the fuel distribution from the natural gas grid infrastructure. Cryogenic processes for liquefaction inherently involve CO₂ separation, a mandatory step during production of Bio-SNG. Possible synergy effects from integrating liquefaction process with a Bio-SNG plant can be studied using process integration methodology. One possible such effect might be the use of low-temperature excess heat for chilling the raw gas prior to liquefaction.

7.3 Economic aspects

Policy instruments need to be in place in order to achieve market breakthrough for Bio-SNG and other biofuels. Market forces can be expected to deliver solutions to many of the technical barriers in the medium term. The design of these policy instruments however is not a trivial task due to the numerous interdependencies within the energy market and difficulties on choosing between directed policy measures specifically promoting a certain product and general policy measures allowing for market mechanisms to determine the most competitive processes. The most important aspect considering policies might though be their long term reliability. As early biomass gasification projects imply a high risk the long term profitability needs to be ensured in order for companies to invest. Developing proper strategies for the design of such policies is still a question that needs attention. This question is also related to the aspects of fuel replacement. The present work shows that replacing fossil natural gas with Bio-SNG is hardly economically viable under current conditions. Determining the appropriate level for policy instruments supporting Bio-SNG requires selecting the reference fossil fuel that is to be replaced by Bio-SNG, which obviously is a difficult choice for policy-makers. For example, considering Bio-SNG in the transport sector the reference chosen could be either natural gas, fossil gasoline or diesel, or even a competing biofuel alternative. The choice of reference will have implications on the economic situation as well as on the footprint of Bio-SNG and none of the choice is obvious per se.

Nomenclature and Abbreviations

a	annuity factor
ar	as received
daf	dry ash-free fuel
df	dry fuel
c_i	specific cost for i / specific CO ₂ -equivalent emissions for i
C_F	total capital investment cost
e^{CH}	specific chemical exergy
e^{PH}	specific physical exergy
\dot{E}	exergy flow
h	specific enthalpy
\dot{m}	mass flow
n_i	amount of i in energy
P	pressure
\dot{Q}	thermal power
R	ideal gas constant
s	specific entropy
t_{op}	annual operating time
T	temperature
\dot{W}	work flow/power/electricity
$wt-\%$	weight percent (mass based)
x_i	molar fraction of component i
ΔH_r	enthalpy of reaction
η	energy efficiency
η_{cg}	cold gas efficiency
η_{el}	electrical efficiency
η_q	heat efficiency
η_{ref}	electrical efficiency for reference stand-alone plant
η_{ex}	exergetic efficiency
Θ	Carnot factor
λ	relative air-to-fuel ratio
ρ	density

Abbreviations

AER	adsorption enhanced reforming
BFB	bubbling fluidised bed

CFB	circulating fluidised bed
CHP	combined heat and power
CLR	chemical looping reforming
CNG	compressed natural gas
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 Scenarios tool
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
IO	investment opportunity
LHV	lower heating value
LNG	liquefied natural gas
LP	low pressure (steam)
MEA	monoethanol amine
MILP	mixed integer linear programming
Mtoe	million tons of oil equivalents
NGCC	natural gas combined cycle
O&M	operation and maintenance
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
WGS	water gas shift
WI	Wobbe index

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Appendix

TABLE 12. 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 13. Basic modelling 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 ₂ O/kg dry fuel]		0.6	
Pyrolysis Modelling		Yield reactor based on (Thunman <i>et al.</i> , 2001)	
Gasification Modelling		<ul style="list-style-type: none"> Gibbs equilibrium reactor at operating temperature reactive species: C, CO, CO₂, H₂ & H₂O WGS at equilibrium 70% carbon conversion 	
Tar Reforming Unit (Chemical Looping Reformer)		Scrubbing Unit (Water Scrubber)	
Pressure Drop [bar]	0.05	NH ₃ Removal Efficiency [-]	0.99
Operating Temperature [°C]	625	Pressure Drop [bar]	0.02
Reactions	complete reforming of tars to CO & H ₂	Water Recycle Cooling Temperature [°C]	20
		Waste Water Stripper	operating a 1 bar, off-gases to be burnt in combustion boiler
CO₂ 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) adjusted to obtain H ₂ /CO = 3 taking into account simultaneous WGS reaction
Recoverable Energy [Fraction of Energy Demand]	20% (@90 → 40 °C)	Steam Addition	
Membrane Separation		Compressors & Fans	
Inlet Pressure [bar]	10	Isentropic Efficiency	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 ^a [°C]	80 - 120
Pumps		SNG delivery conditions	
Pump Efficiency	based on efficiency curve for water ^b	Pressure [bar]	10
Mechanical Efficiency	0.98	Temperature [°C]	30

^a in case of multi-stage compression^b default in ASPEN Plus

TABLE 14. Stream data for the base case Bio-SNG process.

Stream type	T_{start} [°C]	T_{target} [°C]	Q [kW]	ΔT/2 [°C]	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

