Integration aspects for synthetic natural gas production from biomass based on a novel indirect gasification concept

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Abstract

An innovative indirectly heated biomass gasification unit has been recently built at Chalmers University of Technology as an integrated extension of a standard circulating fluidised bed (CFB) boiler for heat and power production. The gasification medium can be varied between steam, oxygen, combustion flue gases or recirculated syngas. In this paper a process for production of synthetic natural gas (SNG) based on this biomass gasification technique is proposed and investigated with emphasis on evaluation of possible heat integration options. Special attention is given to possible options for cogeneration of heat and power. The increase in electricity production from the power cycle is achieved by two means: combusting the non-reacted char from gasification in the boiler and extracting high temperature excess heat from the syngas to SNG conversion steps. It is shown that the amine-based CO_2 separation stage is a large heat sink. The reduction of the steam demand for the CO_2 absorbent regeneration stripper is of crucial importance to have a maximum of high temperature excess heat available from the gasification process to be used in the steam power cycle. The cold gas efficiency for SNG production comparing biomass input to SNG output is about 60 % for the proposed process. This performance indicator however does not consider the electricity production increase. The balance between SNG yield and increased electricity production is mainly dependant on the gasification efficiency since the amount of char from gasification that is used in the boiler directly influences the yield of synthetic natural gas.

Introduction

The emission of anthropogenic greenhouse gases has increased drastically over the last century and its reduction is a major challenge to the industrialised world. Biomass based energy is one of the options pursued to reduce fossil CO_2 emissions, but the current fuel production options from biomass are criticized as they are often based on food crops, thereby competing with land use for food production. In addition, efficiencies of the current production routes are often not satisfactory. Research is therefore focusing on the so-called second generation of biofuels based on non-food biomass. One of the key technologies for the production of these fuels is gasification. Based on the resulting syngas from the gasification stage there are numerous options for fuel production such as hydrogen, methanol, ethanol, dimethylether, Fischer-Tropsch diesel or synthetic natural gas (SNG) that achieve higher efficiencies compared to current biomass fuel conversion routes. SNG is sometimes also referred to as bio-methane or biogas but the term synthetic natural gas will be used throughout this paper.

An innovative indirectly heated biomass gasification unit has been recently built at Chalmers as an integrated extension of a standard circulating fluidised bed (CFB) boiler. In this paper, the production of SNG from the resulting syngas is investigated, with a focus on

assessing the potential for integration between the steam power cycle and the gasification stage and syngas treatment for SNG production.

Gasification technology

Biomass gasification has been studied extensively during the last decades. Several research projects with pilot plant installations have demonstrated feasibility of this technology. In Güssing (Austria) a 8MW$_{th}$ Fast Internal Circulating Fluidised Bed (FICFB) gasification demonstration plant has been in operation since 2001 [1, 2]. In Värnamo (Sweden) a demonstration power plant for biofuel-fired combined heat and power generation based on pressurized gasification was operated during the period 1996-2000 [3, 4]. The gasification plant in Värnamo has thereafter been modified for testing as a unit for production of high quality syngas for downstream conversion to transportation fuels or other high value chemicals [5]. The Energy Research Centre (ECN) in the Netherlands is also working on biomass gasification technology [6]. Carbona Technology have recently built demonstration projects for gasification [7-9], and the Technical University of Denmark is developing the so-called Viking gasification technology [10], to name only a selected choice of projects. All of these technologies are stand-alone gasification solutions, and all of them have faced some operational difficulties. The interest from industry in investing in these technologies is therefore still limited due to the high risks involved.

The gasification concept developed at Chalmers University of Technology in contrast is designed as an extension of an existing CFB boiler as often used for biomass or coal based power production. The gasifier is installed in the return leg of the boiler where a particle cooler is usually used to extract heat from the circulating bed particles. The installation of a gasifier in this way makes it possible to operate the power cycle based on the combustion in case of operational difficulties with the gasifier. The risk of such an extension is thereby reduced to a large extent. Furthermore, possible synergy effects can be the joint use of several sub-systems as fuel and ash handling, the water/steam system and parts of the flue gas clean-up equipment for both the gasification and combustion section.

Figure 1 shows the schematic flow sheet of a CFB boiler with an integrated gasification unit. The combustion fuel is fed to the riser part of the boiler; heat is transferred to the gasification side by the hot bed material. Non-converted biomass and char from the gasification process are returned to the combustion chamber. Both sections are separated by a particle seal. Steam, air, oxygen, flue gases, or the syngas itself can be used as fluidisation medium in the gasification reactor vessel. It is an indirect gasification process similar to the FICFB concept used in Güssing [2] but with the combustion and gasification section inverted and a higher flexibility in case of operational problems. In case the gasification and syngas treatment line have to be stopped, it still is possible to run the power cycle by extracting all heat for the production of steam. The installation then operates as a standard CFB boiler.
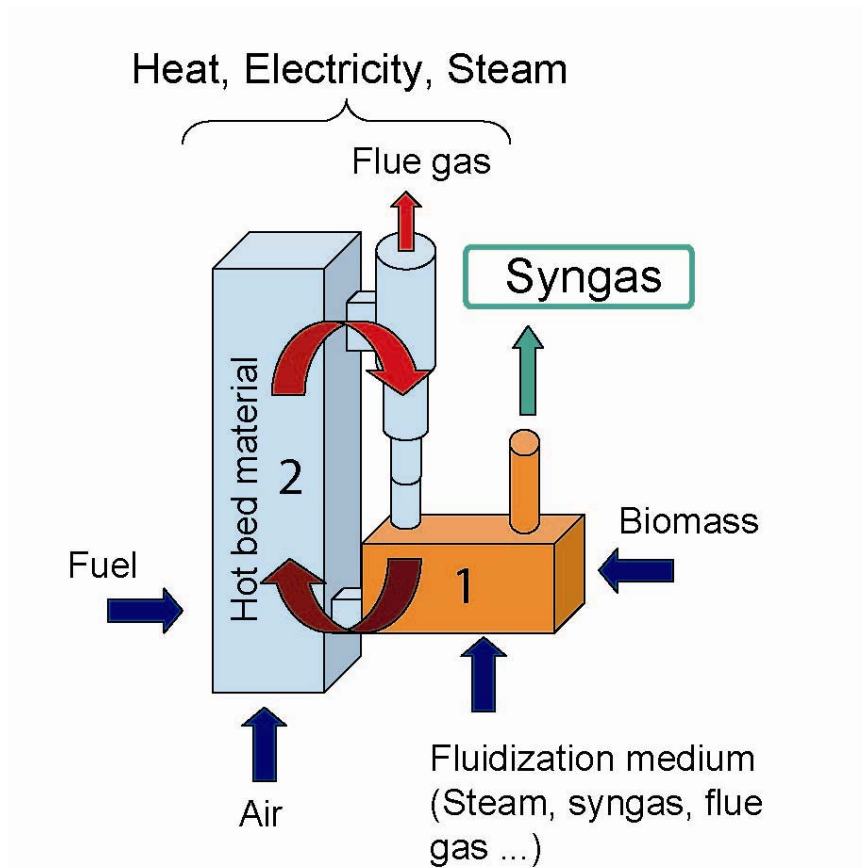
Figure 1: Schematic flow sheet of a biomass gasfication unit (1) integrated with a CFB boiler (2)

The potential of this novel gasification concept is illustrated by the overall number of boilers suitable for such an extension in Europe and North America represented in Figure 2. A realistic size for such an installation can be a gasifier fuel energy supply rate of around 100 MW and a combustion unit of comparable size. As shown in the figures there are more than 250 boiler units installed in the size range of 61 to 180 MW with an overall cumulative capacity of approximately 30 GW. Depending on the location of the boilers (biomass supply) and the application the syngas is used for (gas treatment equipment) even the larger boilers may be of interest for this concept.
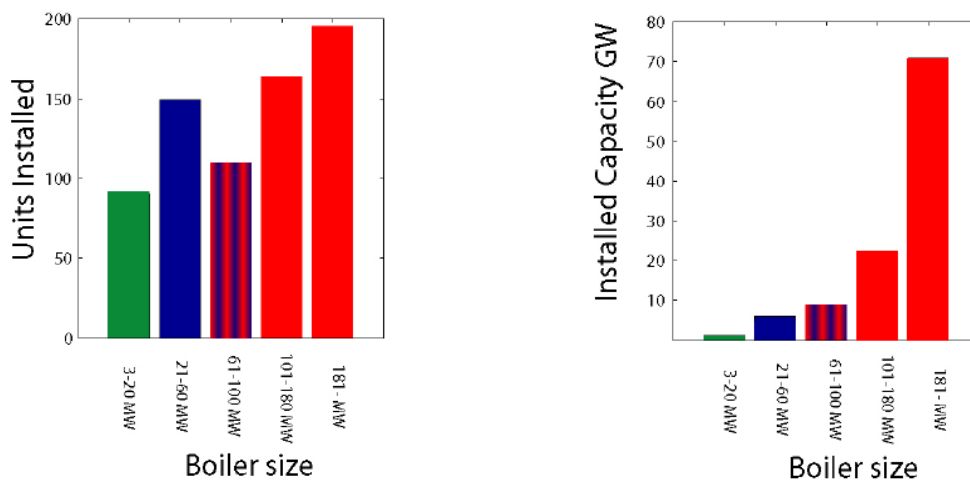

Figure 2: Installed fluidised bed boilers in Europe and North America (boilers delivered by Metso Power, Foster Wheeler and Alstom).

SNG production process

The aim of this paper is to illustrate opportunities for heat integration between SNG production via gasification and the existing power cycle as described above. SNG production from biomass is currently being investigated by several research groups [11-13]. Van der Drift [11] proposed a biomass-to-SNG concept based on an indirectly heated gasifier with a conversion efficiency estimated at around 70%. Gassner [12] used multi-optimisation techniques to generate a set of optimal SNG production pathways based on energy and exergy efficiencies as well as SNG yield and grass root costs. Felder [13] has investigated the ecological impacts of SNG for heating and as transportation fuel for cars using life cycle analysis based on the Güssing gasification technology [2]. Combined heat and power production was not considered as an option in that study. The performance of SNG is shown to be systematically better than fossil fuel use for all cases considered, its superiority depending on the weighting of fossil fuel consumption. The best option according to Felder's study is the use of SNG as a transportation fuel.

All studies emphasise the importance of heat integration and mention problems with tar formation during gasification. Tar compounds contained in the syngas can condense on equipment surfaces causing fouling. This limits the opportunities for effective energy recovery from the hot syngas leaving the gasifier. There are numerous techniques to treat the tars in the syngas: at the Güssing plant in Austria, a rapeseed oil scrubber is used to clean the syngas, the tar charged stream is then fed to the combustion unit of the gasifier. In-bed catalytic tar reduction has also been investigated within the Güssing project [14]. A similar technique based on oil-scrubbing is proposed by ECN [6]. For the Carbona process, a Ni-based catalyst for selective tar cracking at high temperatures (around 900°C) is used [9]. In the present study, chemical-looping reforming (CLR) is considered for the high temperature removal of tars. This technique has been successfully implemented for reforming of natural gas [15]. An oxygen carrier is circulated in a fluidised bed reactor taking up oxygen in one compartment (air reactor) and selectively reforming the tar components contained in the syngas in the other reactor compartment (fuel reactor). The principle of CLR is shown in Figure 3. In this study a complete reforming of all tars in the CLR unit to H_2 and CO is assumed. This represents an overestimation of the tar conversion and for the process design and operation tar slip may be problematic for downstream equipment such as scrubbers and water treatment, but from an energy viewpoint the error in the tar conversion estimation will not be significant due to the low concentration of tars.
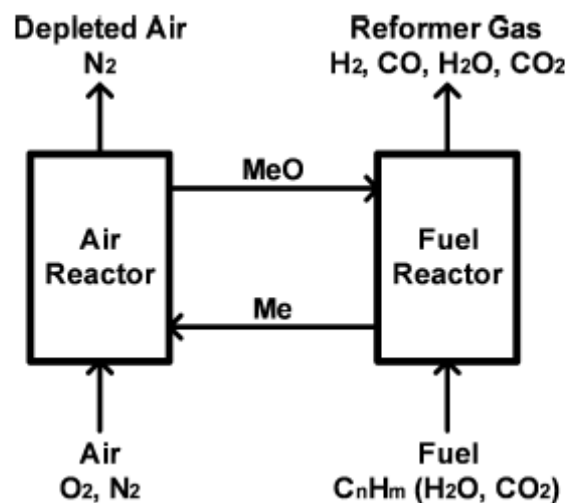

Figure 3: The principle of chemical-looping reforming [15].

The overall process set-up for the gasification process and the gas treatment and conversion to SNG is shown in Figure 4. The gas cleaning section consists of a tar reforming step (CLR), a water scrubber and filter section to take care of the particulate matter as well as sulphur and alkali compounds. Before the first methanation step, CO_2 is separated by an amine-based absorption process (MEA). In order to reduce the gas volume to be compressed, a second CO_2 separation stage is implemented before the second methanation stage at high pressure. The non-converted hydrogen and carbon monoxide are separated from the gases and recycled to the first methanation reactor before the product gas undergoes final drying.
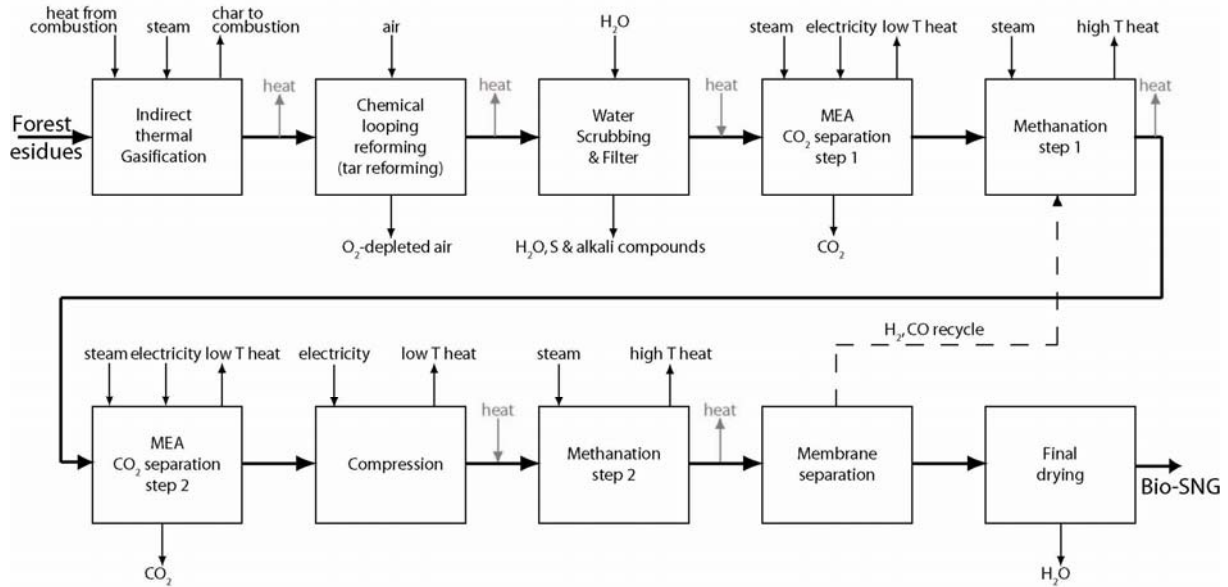

Figure 4: Proposed concept for the production of synthetic natural gas from biomass gasification.

It should be emphasised that the proposed process layout for production of SNG from biomass described above should not be considered as an optimal process configuration, but rather as a base case that is able to handle all non-desirable by-product substances. The goal of this study is mainly to illustrate the opportunities for integration of the gasification concept with the power cycle. Of course, the opportunities for process integration are directly dependant on the choice of the individual process steps acting as heat source or sink to a different extent. The current process design therefore constitutes a reference case for future evaluations of different process options.

Process simulation

In order to evaluate the process a simulation flowsheet was set up using the commercial software package Aspen Plus [16]. The simulation is based on the flowsheet presented in Figure 4. The property estimation method used for all simulation steps was the Peng-Robinson cubic equation of state.
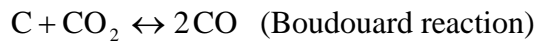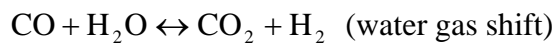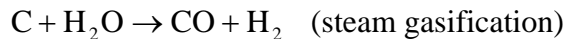
The gasification medium chosen for the process was the syngas together with a certain amount of injected high pressure steam to compensate for the pressure drop across the fluidised bed. The incoming biomass feedstock composition is specified in Table 1.

Table 1: Biomass feedstock composition

Proximate analysis		Ultimate analysis (wt-% dry ash free)	
Fixed carbon (wt-% dry fuel)	20	C	51.42
Volatile matter (wt-% dry fuel)	77.82	H	5.55
Ash content (wt-% dry fuel)	2.18	O	42.49
		N	0.48
Moisture content (wt-%)	10	S	0.04
LHV (MJ/kg dry fuel)	19.54	Cl	0.01

An estimation of the syngas composition leaving the gasifier is obtained by simulating a decomposition of the incoming biomass followed by an equilibrium reactor. The decomposition calculations are based on experimental measurement data of devolatilisation gases from wood [17]. A mass balance for each element present in the feedstock as well as an overall energy balance is established and several constraints based on the measurement data such as the molar ratio of CO/CO_2 ratio in the released gases, the char yield from pyrolysis or the heat of devolatilisation are used to solve the system of equations. More details are given in Table 2.

The equilibrium reactor accounts in a second step for the following reactions of the released gases, the char, and the fluidising medium:

$$C + H_2O \rightarrow CO + H_2 \quad \text{(steam gasification)}$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \quad \text{(water gas shift)}$$

$$C + CO_2 \leftrightarrow 2CO \quad \text{(Boudouard reaction)}$$

where the char (simulated as carbon) is assumed only to be involved in the steam gasification and Boudouard reactions to an extent of 20 %. This value has been adjusted to obtain a overall gasification char yield similar to the data reported from the laboratory experiments in [17]. There is however some uncertainty in this estimation. This constraint directly influences the overall carbon conversion efficiency of the gasification and thereby the possible yield of SNG in the end. A comparison to experimental data from steady-state operation of the gasifier will be necessary in the future to properly adjust this parameter.

The fluidising medium consists of recycled syngas and injected steam. As a last step in the gasification island, the bed material along with the non-converted char is separated from the gas stream and led back to the combustion unit. The recycle stream necessary for fluidisation is separated from the particle free syngas.

The key assumptions for modelling the different process steps are listed in Table 2. The resulting SNG composition and process efficiency data are given in Table 3. The cold gas efficiency of the process defined on a lower heating value basis is 59.4 %. This value compares the biomass input to the SNG production on a lower heating value basis assuming perfect internal heat exchange and not taking into account the electricity consumption of the process. This is a comparatively low value compared to the estimated 70 % by van der Drift [11]. The lower value can on the one hand be attributed to the conservative estimate of the carbon conversion efficiency in our study. Based on the model parameters only a fraction of the fixed carbon actually reacts to form a gaseous component and a large extend of the fixed carbon fraction in the biomass is separated from the syngas along with the bed material to be recycled to the combustion unit. This interaction is an important aspect in the overall energy balance considerations. But on the other hand the estimate by van der Drift [11] is based on a scale-up of a lab scale installation (5 kg/h biomass feed) and might be somewhat optimistic.

The process electricity consumption is estimated to be around 4 MW and can be mainly attributed to the CO_2 separation process and the compression of the syngas. It should however be noted that the electric power consumption of the other process units has not been

considered in this study. As mentioned above, the proposed process design should not be considered as optimal but rather as a base case that can be used as a reference in the future.

Table 2: Key process modelling assumptions.

Unit	Key assumptions – modelling parameters

Gasifier	• 100 MW$_{th}$ of biomass input (based on LHV)
	• T = 850 ºC
	• P = 1 bar
	• Decomposition constraints [17]:
	   – Char yield: 0.2 kg char/kg dry ash free fuel
	   – Molar ratio of CO/CO_2 in gases: 3/1
	   – Light tar compounds: CH_4/C_2H_4 (molar ratio 3/1)
	   – Molar ratio light tars/CO_2: 3/2
	   – Heavy tar compounds: C_6H_6, C_7H_8, $C_{10}H_8$ (present in equal molar fractions)
	   – Heat of devolatilisation: 1 MJ/kg dry fuel
	   – Char LHV: 33 MJ/kg
	   – Sulphur from feedstock completely converted to H_2S
	   – Chlorine from feedstock completely converted to Cl_2
	• Equilibrium reactor: 20 % char fraction available for steam gasification and Boudouard reaction:

Chemical looping reactor	• T = 600 ºC • Complete tar reforming to CO and H_2
Water scrubber and filter	• Complete particle removal • Complete sulphur & alkali compound removal

MEA absorption unit	• 95 % CO_2 separation efficiency
	• energy consumption [18]:
	   3.7 MJ/kg CO_2 (steam at 150ºC)
	   1.0 MJ/kg CO_2 (electric power)
	• boiler and condenser load assumed to be equal in MEA regeneration column
	• temperature level in the condenser of the regeneration column: 110 ºC (40 ºC temperature drop from bottom to top)

Methanation	• P= 1 bar (stage 1) / 10 bar (stage 2) • T = 300 ºC • Temperature approach to equilibrium (20 ºC) in order to account for deviations from chemical equilibrium in a real reactor
Compressor	• Number of stages: 3 (inter-cooled) • Stage isentropic efficiency: 0.72 • Mechanical efficiency: 0.98
Membrane separation process	• 99.9 % H_2 separation efficiency • 50 % CO separation efficiency • 0.1% CH_4 loss with permeate

Table 3: Process performance indicators.

Biomass input (MW$_{LHV}$)	100
SNG output (MW$_{LHV}$)	59.4
Gasification carbon conversion efficiency (moles carbon in syngas / moles carbon in feed)	0.689
SNG cold gas efficiency (LHV based)	0.594
Electric power consumption (MW)	4.04
Minimum heating demand (MW)	0
Minimum cooling demand (MW)	15.95
Pinch temperature (ºC)	840
SNG composition (vol-%)	
CH$_4$	97.18
CO$_2$	1.66
N$_2$	1.13
CO	0.01

Integration considerations

The heat stream data calculated using process simulation software was extracted to conduct a Pinch analysis of the proposed process. For heat exchanging, individual temperature difference contributions were selected for the different streams, i.e. $\Delta T_{min}/2 = 5$ K for liquids, boiling/condensating streams, and water vapour and $\Delta T_{min}/2 = 10$ K for gaseous streams. The resulting Grand Composite Curve (GCC) is represented in Figure 5. It can be clearly seen that there is a high potential for high pressure steam production via the syngas cooling. Another important heat source is the heat of reaction from the methanation reactor. Major heat sinks are the steam production for the methanation process as well as the steam needed for the fluidised bed reactor. The largest heat sink is the steam consumption in the stripper of the CO$_2$ absorption step. The MEA stripper condenser heat load represents an interesting opportunity for export of low temperature heat from the process. This heat could be used for example in a district heating network, if available.
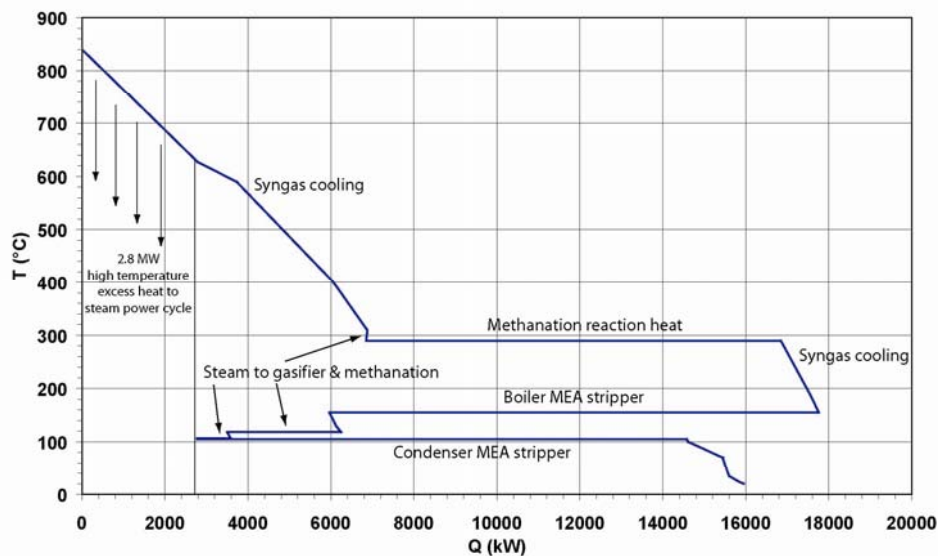

Figure 5: Grand Composite Curve for base case SNG production process.

The steam needed for the regeneration of the absorbent (Boiler MEA stripper in the figure above) in amine-based absorption can vary significantly depending on a number of different factors. The choice of absorbent, the separation efficiency, the lean solvent loading, the amine solvent concentrations, as well as the stripper operation pressure are major parameters. Abu-Zahra et al. [19, 20] have conducted a parametric study of the technical and economical performance of CO_2 capture based on MEA. They simulated the whole capture process using Aspen Plus. A minimum thermal load of 3.0 MJ/kg CO_2 was found for a lean MEA loading of 0.3 and using a 40 wt-% MEA solution and a stripper operating at 2.1 bar. The separation efficiency for CO_2 used in this study was 90 %. The value of 3.7 MJ/kg CO_2 used in this study (see Table 2) is an average value based on the study in [18] where the energy demand for CO_2 separation for a number of different coal power generation plants is investigated. The range indicated in reference [18] for the thermal energy consumption is 3-5 MJ/kg CO_2 separated. Exact numbers are hard to define as the energy consumption is dependant on many parameters and the optimisation is constrained by technical limits such as increased corrosion risk associated with higher MEA solution concentration.

In order to illustrate the importance of a low steam consumption for the CO_2 separation process, two extreme cases are represented in Figure 6. The left hand side figure corresponds to a low specific steam consumption of 3.0 MJ/kg CO_2 whereas the the right hand figure corresponds to high specific steam consumption case of 5.0 MJ/kg CO_2. It can be clearly seen that there is a strong influence on the overall energy balance of the SNG process. In the extreme case of a strongly increased steam demand for the CO_2 separation, external heating (1.37 MW for this case) is necessary to sustain the process. This illustrates the importance of an optimal design of the CO_2 separation step. Low steam consumption for the MEA absorption leads to a higher amount of high temperature excess heat to be transferred to the steam power cycle. The integration benefits with the steam cycle therefore are strongly dependant on the steam demand of the CO_2 separation unit. This of course also rises the question if other CO_2 separation technologies might be more appropriate and might be the object of future investigations.

An important aspect for the CO_2 separation technology to be considered as well is the scale of the SNG plant. MEA absorption is only economically feasible in large scale applications. This also renders the concept of extending an existing power cycle with the gasification unit more interesting as will be discussed later.
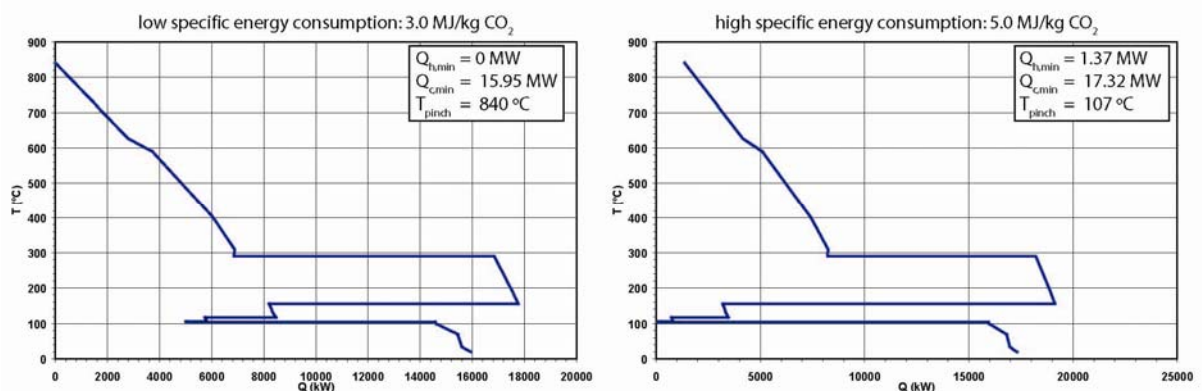

Figure 6: Influence of the steam load for the MEA absorption step on the SNG process.

The heat stream analysis presented so far focuses only on the part of the process starting with the syngas leaving the gasifier. However, since the gasification concept is designed as an extension of an existing boiler for power production it is interesting to investigate the interactions between the power cycle and the gasification process. The heat production inside

the boiler is reduced by the heat used for heating, pyrolysing and gasifying the biomass, but the recycled char compensates this effect and even increases the heat generation in the boiler. In addition the excess heat from the SNG production process can be used to generate steam for the power cycle as well. A simplified analysis of the heat flow of the base case as illustrated in Figure 7 results to

- 6.1 MW of heat transferred from the combustion section to the gasification process via the hot bed material
- 26.4 MW of combustion energy returned to the combustion section with the non-gasified char
- 2.8 MW of high temperature excess heat from the syngas available for steam production in the power cycle
- This results in a net heat flow to the steam power cycle of 23.1 MW

Assuming an electricity production efficiency of 0.4, the resulting increase in electricity production is 9.24 MW_{el}. This more than compensates for the estimated electric power consumption of the SNG production process. A more detailed investigation of the optimum integration between the power cycle and the SNG process should be done to obtain more reliable results. The balance is of course also a trade-off between SNG production and electricity production. Assuming a higher carbon conversion efficiency, the SNG production will increase whereas the electricity production will most likely decrease due to a smaller amount of char recycled to the combustion unit.
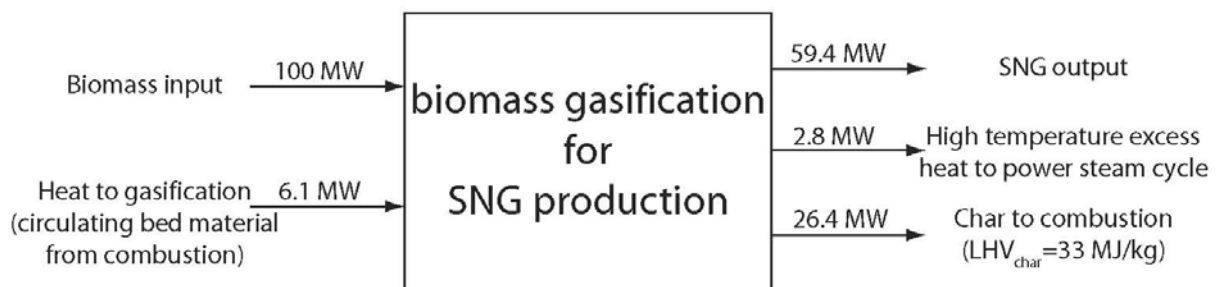

Figure 7: Heat energy balance for the base case SNG production process.

Another important synergy effect for this gasification concept can be the CO_2 separation process. When the combustion process is equipped with a post-combustion CO_2 separation unit a large benefit considering infrastructure and investment costs can be expected. This statement is also valid for other common process stages as fuel and ash handling or the common use of the steam system.

Conclusions
A process for the production of SNG by biomass gasification has been proposed based on a novel concept for gasification recently developed at Chalmers University of Technology. The gasification unit is designed as an extension of an existing CFB boiler. Important interactions between the power generation and SNG production processes have been identified. High temperature excess heat from the gasification can be efficiently used to increase the electricity production of the power cycle. For the base case studied the increase in electricity production from the power cycle has been shown to exceed the electric power consumption of the SNG production process causing a net increase of electricity output. The CO_2 separation process – chosen as MEA based absorption – is a large heat sink and needs to be optimised for optimum operation. The optimal design of this separation stage also

determines the opportunities for heat integration and power production. High steam consumption of 5 MJ/kg CO_2 in the MEA regeneration unit can even lead to a heat deficit in the SNG process. Decreasing the steam need of the CO_2 separation stage is therefore crucial.

A large potential for synergy effects lies in the common use of the CO_2 separation stage for both the flue gases from the combustion as well as the syngas. A more sophisticated approach to the use of excess heat from the SNG process for power production as currently proposed, as well as the trade-off between production of electricity and SNG will be object to future work.

Acknowledgments

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