THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

Indirect gasification production of biomethane for use in heavy-duty state-of-the-art gas engines

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Department of Energy and Environment

CHALMERS UNIVERSITY OF TECHNOLOGY

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Abstract

The climate targets set for the European transport sector have stimulated intensive research by groups in academia, the energy industry, and vehicle manufacturing in the Gothenburg region into biomethane production *via* indirect gasification of lignocellulose biomass and the development of advanced gas engine technologies.

This work presents the results of a comprehensive study of biomethane production and utilization in heavy duty engines. The different steps in the biomethane chain (biomass drying, gasification process, and combustion) are assessed, and opportunities for improving the efficiency of utilization of biomass resources are evaluated. The biomethane chain is investigated through a well-to-wheel (WtW) analysis of the newly built GoBiGas plant (Gothenburg, Sweden), in combination with three state-of-the-art gas engines technologies: spark-ignited (SI); dual fuel (DF); and high-pressure direct injection (HPDI). Opportunities for improving the biomethane process are focused on the drying system and on the dual fluidized bed gasifier. An advanced drying system for the dual fluidized bed gasifier, which uses low-temperature steam as the drying medium and recovers the evaporated moisture as a gasification agent, is evaluated. A method for simulating the process that occurs in the dual fluidized bed gasifier using experimental data is introduced, with the aim of exploiting the extensive body of information derived from pilot and demonstration gasifiers in relation to process optimization and techno-economic analyses. The uncertainty that arises from the measurements is assessed stochastically and transferred to process parameters.

The WtW analysis shows that emissions from biomethane are reduced by 73%, 46%, and 68% when used in the SI, DF, and HPDI engines, respectively, as compared to using NG and LNG. The evaluation of the drying process reveals a theoretical energy efficiency of 95% when combined with a DFB gasifier and an exergy efficiency of 53%, values that are considerably higher than those obtained with other drying systems. Through interpolation and extrapolation of the experimental data, the proposed modeling method is demonstrated to be a flexible tool for simulating the gasifier under several operational conditions Comparisons of the data from different measurement set-ups demonstrate that a detection rate of ≥95% for the carbon in the produced gas is necessary to keep the uncertainty at <3% and to estimate the char conversion and oxygen transport rates in the gasifier.

Overall, the results of this study indicate that the current biomethane chain achieves considerable reductions in emissions compared to the use of fossil fuels, and that there is significant potential for further improvements.

Keywords: Biomass gasification, DFB gasifier, biomethane, SNG, dual fuel, process simulation, well-to-wheel, gas engines

List of publications included in the thesis

I. A. Alamia, I. Magnusson, F. Johnsson, H. Thunman. "Well-to-wheel analysis of bio-methane via gasification, in heavy duty engines within the transport sector of the European Union". Applied Energy, 2015 (accepted for review)

II. A. Alamia, H. Thunman, and I. Magnusson, "Fuel Quality analysis for biogas Utilization in Heavy Duty Dual fuel engines," in *20th European Biomass Conference & Exhibition*, Milan, 2012

III. A. Alamia, H. Ström, and H. Thunman, "Design of an integrated dryer and conveyor belt for woody biofuels," *Biomass and Bioenergy,* vol. 77, pp. 92-109, 6// 2015

IV. A. Alamia, H. Thunman, M. Seeman " Method for process simulation of dual fluidized bed gasifiers through experimental data", (submitted)

Professor Henrik Thunman is the principal academic supervisor and has organized and participated in the planning of the work. He has also contributed with discussions and editing of all four papers. Ingemar Magnusson is the industrial co-supervisor and has contributed with discussions and editing of Papers I and II. He also had a major role in the collection of the data for Paper I. Martin Seemann, who is the academic assistant supervisor, has contributed to the planning and editing of Paper IV.

Publication not included in the thesis

- A. Alamia, F. Lind, H. Thunman. "Hydrogen production from biomass gasification for utilization in oil refineries". ACS 244th National Meeting, Philadelphia, 2012

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Alberto Alamia

Mykonos, 21th September 2015

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1 Introduction

The ambition within the Swedish transport sector is to develop a fossil-free vehicle fleet by Year 2030, as expressed in the Integrated Climate and Energy Policy, props. 2008/09:162 and 2008/09:163 [1, 2] released by the Swedish Government in 2009. One of the main topics addressed in these directives is the production of efficient and renewable biofuels, which are considered to be crucial for reducing emissions in the heavy-transport sector, where electrical powertrains are not suitable. The commitment made towards such stringent climate goals demands a spectrum of solutions for the production of renewable biofuels, fuel distribution, and new vehicle technologies. The challenge is to develop those technologies that in combination generate significant reductions in greenhouse gas (GHG) emissions and promote efficient utilization of the renewable energy sources.

Driven by these aspirations, the joint efforts of academia, the energy industry, and vehicle manufacturers in the Gothenburg region have directed research towards the production of biomethane *via* indirect gasification of lignocellulose biomass and the development of advanced gas engine technologies. The local utility for heat and electricity in the City of Gothenburg (Göteborg Energi AB) has recently brought in operation the demonstration plant GoBiGas, which currently is the largest biomethane plant in the world, with a production capacity of 160 GWh biomethane/yr [3]. The Chalmers University of Technology is a research partner of Göteborg Energi AB in the development of the gasification process, with several experimental campaigns being conducted using the Chalmers pilot gasifier [4-6]. In the meanwhile, Volvo AB Advanced Technology and Research has developed advanced engine technologies for the combustion of gaseous fuels [7-9]. Chalmers has joined the research project on advanced gas engines through the ConGas project [10], in collaboration with both Volvo AB and Göteborg Energi AB. Within this project, a fuel-tolerant engine concept has been developed that is based on the dual fuel technology and that has performance profiles comparable to those of diesel engines in terms of drivability and efficiency, while complying with Euro 6 emission regulations. This thesis builds on the experiences gained from the research on indirect gasification and the ConGas project to evaluate and propose improvements to the entire chain, from biomass conversion to eventual combustion in the vehicle, hereinafter termed the ‘well-to-wheel’ (WtW) chain.

1.1 Aim of the work and outline

The overall aim of the thesis is to investigate various opportunities to optimize the WtW chain to achieve higher efficiency of utilization of biomass and a reduction in GHG emissions. The starting point of this investigation is the state-of-the-art WtW chain for biomethane production, as in GoBiGas Phase 1, involving fuel distribution in the form of compressed or liquefied gas and combustion in SI, DF and HPDI engines. In this thesis, four papers are presented; Figure 1 places them in the framework of the WtW chain for biomethane.

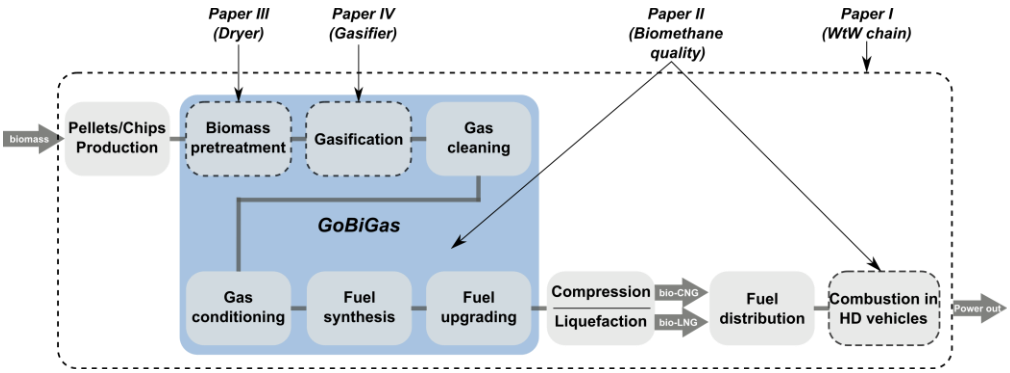

***Figure 1** – Overview of the thesis and the topics of the included papers.*

Evaluation of the current WtW chain is the objective of Paper I. The results therein provide estimations of the GHG reductions and the efficiency levels with respect to utilization of the biomass. Paper I is based on a case study of the GoBiGas plant involving three different gas engine technologies and using a state-of-the-art diesel engine as reference. Paper II deals with biomethane quality in the contexts of operability of the gas engine and emissions targets. Biomethane quality, which is strictly dependent upon its composition, can influence both the biomethane process and combustion in the gas engine. Papers III and IV focus on the biomethane process, tackling two areas of potential improvement. In Paper III, a new concept for a belt-dryer that can be used in dual fluidized bed gasifiers is introduced, which to the goal of reducing the energy and exergy losses associated with the pre-treatment of wood chips supplied to the biomethane plant. The proposed dryer uses low-temperature steam as the drying medium and recovers the evaporated moisture as a gasification agent. Additional aspects taken in consideration are: the storage of dry wood; inertization of the fuel; levels of emissions during the drying process; utilization of low-temperature heat; and integration with the rest of the plant. Paper IV deals with the modeling of the gasifier for evaluation of the biomethane process in a flow-sheet software. Modeling of a DFB gasifier is particularly demanding owing to the high degree of freedom associated with the operation of the double-reactor system, the complexity of the reactions, and the reactors' hydrodynamic profiles. The approach used in this work is based on the analysis of experimental data obtained from pilot and demonstration gasifiers. The modeling consists of two phases: 1) a mass and heat balance for the analysis of a database of experimental data; and 2) a flow-sheet model of the gasifier that can shift between different operational points and use interpolation/extrapolation of the data to simulate other conditions. In the data analysis, a stochastic approach was used to assess the uncertainty related to both measurement errors and incomplete characterization of the raw gas produced by the gasifier.

1.2 Framework for biomethane in the European Union

In the last decade, the European Commission (EC) has approved a set of policies and directives to reduce the dependency on oil of the transport sector and to achieve deep cuts in emissions [17-20]. The long-term target is a 60% reduction in emissions and oil dependency by Year 2050, as compared to the situation in Year 1990. The EU strategy to reduce emissions focuses on the introduction of alternative fuels, such as first- and second-generation biofuels (including biomethane), natural gas (NG) and liquefied natural gas (LNG). While NG and LNG yield only moderate reductions in GHG emissions, as compared with oil-based transportation fuels (diesel and petrol) [21], biofuels can achieve near-zero emissions, if one assumes that the entire biomass supply chain is carbon-neutral. In particular, the EC has regulated the introduction of first- and second-generation biofuels through Directive 2009/28/EC [22], which states that each Member State should achieve at least a 10% share of renewable energy, including biofuels, renewable electricity, and renewable hydrogen, across the entire transportation sector by Year 2020.

First-generation biofuels are produced by conventional technologies, such as biochemical reactors that use sugar cane as feedstock, and they are currently the most important alternative to oil, accounting for 4.4% of transport fuels in the EU [20]. However, first-generation biofuels confer weak climate benefits and have significant negative Land Use Change (LUC) effects [23]. In contrast, second-generation biofuels can be produced from low-value forest residues, such as the waste generated by sustainable forestry management. In addition, these fuels are produced using more advanced conversion technologies with higher conversion efficiencies, for example, gasification-based processes. The combination of energy efficiency and climate benefits means that second-generation biofuels are superior to first-generation biofuels. Consequently, the EC has updated Directives 2009/28/EC and 98/70/EC through amendment 2012/0288 [24], which limits the use of first-generation biofuels to a maximum of 5% and excludes *de facto* first-generation biofuels from post-2020 incentives.

To date, second-generation biofuels have suffered from the lack of an infrastructure for distribution and refueling. In recognition of these limitations, the EC has included second-generation biofuels in the “Clean Power for Transport” [20] (CPT) initiative, which is promoting the development of an infrastructure designed to ensure economies of scale for the supply of alternative fuels. In the CTP initiative, NG, LNG, and biomethane are considered to be the main substitutes for diesel and petrol used in heavy vehicles, and targets are set for the installation of refueling station networks by Year 2020. These targets proscribe a maximum distance between refueling stations of 150 km for NG in compressed form (CNG) and 400 km for LNG. CNG will be used mainly in light vehicles or city buses, while LNG will play a major role in long-haul vehicles. Therefore, most of the LNG stations will be installed along the trans-European core road network. Biomethane production could be developed without the need for a separate infrastructure, since it can be distributed through the network created for LNG and CNG. Therefore, biomethane is expected [25] to be a low-risk option for the introduction of second-generation biofuels to the heavy transport sector.

1.3 Development of a commercial biomethane WtW chain

Research on biofuels has focused on so-called second-generation biofuels, which are produced from residues, waste, lignocellulose biomass, and cellulose, as well as from non-food crops and algae. Second-generation biofuels can be produced by biological or thermochemical conversion (gasification or pyrolysis) of the energy sources. The latter is especially suitable for lignocellulose biomass, since it enables conversion of the lignin fraction. This thesis focuses on biomethane production based on the indirect gasification of lignocellulose biomass and utilization in heavy-duty (HD) vehicles. This biomethane chain is considered to be very promising in terms of its potential for reducing GHGs and its technical feasibility.

Indirect gasification is realized in dual fluidized bed (DFB) gasifiers, although other types of gasifiers can be used to produce methane. Gasification technologies fall into the following broad categories: entrained flow (EF); fluidized bed (EF); and DFB. DFB and FB gasifiers are indicated for biomethane production owing to the large fraction of methane present in the raw gas. However, the tar content of the gas can be high, and this will affect the operation of the biomethane plant. DFB gasifiers are allothermal (indirectly heated) gasifiers that entail two FB reactors exchanging heat and fuel through the circulation of a bed material. The reactors are separated by loop seals to avoid mixing of the two gas phases. Compared to an FB autothermal (directly heated) gasifier, the DFB technology enables the production of nitrogen-free gas, without requiring pure oxygen, thereby avoiding the associated energy penalty. DFB gasifiers have been extensively developed in the last decades [11]; some of the most significant gasifiers operating at commercial scale are: the SilvaGas [12] gasifier (1998, USA), the Güssing plant (2001, Austria) [13-15], and the new GoBiGas plant [3].

The GoBiGas project is divided into two phases, with a demonstration plant of 20 MW of biomethane in the first phase, and a commercial plant of 80–100 MW biomethane in the second phase. The first GoBiGas plant, which was completed in 2014 [16], is currently operating and is supplying biomethane to the local natural gas network. The collaboration between Chalmers and Göteborg Energi AB has enabled parallel strands of research on DFB gasification at the laboratory scale, pilot scale (Chalmers gasifier), and demonstration scale (GoBiGas) to acquire the required knowledge for the design of a full-scale process (Fig. 2).

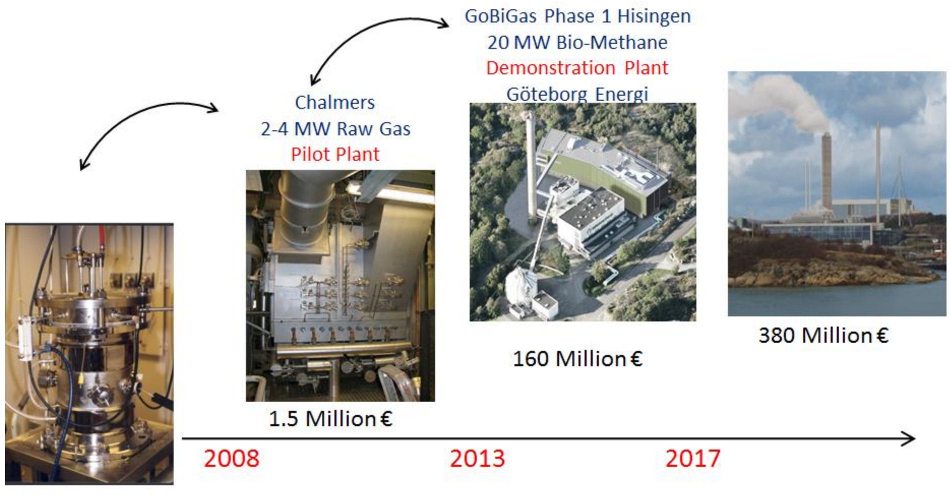

***Figure 2** – Research units involved in the development of the GoBiGas project.*

At the other end of the WtW chain, Volvo AB and other manufacturers (Iveco, Scania and Daimler) are developing their own gas engine technologies, which use both compressed and liquefied gases. At present, the gas engine technologies available on the EU market include: spark-ignited (SI); and dual fuel (DF). In addition, the high-pressure direct injection (HPDI) engine is expected to be commercialized soon, and it is already available for stationary applications in the US market [27]

.

1.4 WtW analysis of biomethane in heavy-duty vehicles

In a WtW analysis, the energy use and GHG emissions associated with the production of the fuel and its use in the vehicle or engine are assessed. The term WtW comes from the analysis of oil-based fuels, although it is also applied to biofuels. Compared to Life Cycle Assessment (LCA), WtW analysis has the same system boundaries but it does not include the consumption of materials and water, other pollutants, and end-of-life disposal. LCA requires more expansive datasets and involves more complex calculations, especially for developing processes that are based on new technologies. Furthermore, WtW is a far more common system for assessing fuels, and the existing literature provides a basis for comparison.

In the present work, the WtW chain is defined from the primary source, i.e., wood from the Swedish forest, to the crank shaft of the vehicle engine. WtW studies of light vehicles usually calculate the levels of energy use and emissions at the wheel, including in the model the powertrain, vehicle weight, and aerodynamics. For heavy-duty vehicles, this approach was considered to be less relevant, as the weight and load of such vehicles vary much more than those of light vehicles. Furthermore, some of the compared engine technologies are in the late stage of development, which means that very few representative vehicles are currently available on the market.

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***Table 1** – Overall scheme of the WtW, WtT and TtW analyses.*

NG	*Biomass*		*LNG*		
• Extraction • Compression (80 bar)	• Pellet production • Biomethane production		• Extraction & shipping	*WtT*	*WtT*
• Transport via pipeline	• Compression (30 bar)	• Liquefaction (-161°C)	• Terminal operations		
CNG	*Bio-CNG*	*Bio-LNG*	*LNG*		
• Injection in local network (30 bar) • Compression (300 bar) and refueling operations		• Transport by truck • Refueling operations			
• Final conversion in the engines - Spark-ignited and dual fuel (CNG, Bio-CNG, LNG, Bio-LNG) - High-pressure direct injection (LNG, Bio-LNG)				*TtW*	

The WtW chain is divided into two sections. The first section, which is referred to as 'well-to-tank' (WtT), accounts for the processing of primary resources, transport, fuel production or refinement, and final distribution to the refueling stations. The second section, which is called 'tank-to-wheel' (TtW), focuses on fuel conversion in the vehicles, possibly based on a specific driving cycle. In the present study, the TtW section is limited to fuel conversion in the engine (up to the crank shaft). Table 1 explains the system boundaries used to define the WtW chains for biomethane, NG, and LNG.

1.5 Gas engine technologies and the ConGas project

To achieve a comprehensive understanding of the WtW chain, three gas engine technologies are studied: SI, DF, and HPDI. The results are compared to state-of-the-art diesel engines. Traditionally, SI engines have enjoyed the largest market share, whereas the DF and HPDI engines have only recently been commercialized (Fig. 3). The DF and HPDI technologies are derived from a conventional diesel engine, using gas as the main fuel and a limited amount of diesel to ignite the combustion process. As part of the ConGas project, the DF technology was evaluated for a Volvo 13-litre engine. The objective of the tests was to develop a fuel-tolerant engine concept with diesel engine performance profile in terms of drivability and efficiency, while complying with Euro 6 emissions regulations. The project was carried out by Volvo AB Advanced Technology and Research in close collaboration with Chalmers University and Göteborg Energi AB.

The DF engine can be operated with either gas or diesel, which is highly beneficial in regions with poor gas-fueling infrastructures. The combustion concept is based on a conventional spark-ignited Otto engine. The gaseous fuel is injected at the inlet port and pre-mixed with air or exhaust gases during the compression stroke. Ignition of the charge is obtained by injection and auto-ignition of a small amount of diesel through a conventional diesel-injection system. The ignition event in a DF engine takes place in a larger fraction of the cylinder volume than in a spark-ignited engine [8], resulting in a high rate of heat release during the ignition phase. The combustion process is characterized by a premixed flame propagation, as in the SI engine. The diesel fraction in the fuel mixture is also a control parameter that is used to optimize the operability of the engine, and it varies significantly within the load. The upper load range is typically limited by knocking [26], resulting in high sensitivity to fuel quality. Under low-load conditions, emissions of unburned hydrocarbons are high, and this requires the development of a dedicated after-treatment system for catalytic oxidation of methane. The remaining emissions from the DF engine are considerably lower than those from a state-of-the-art diesel engine.

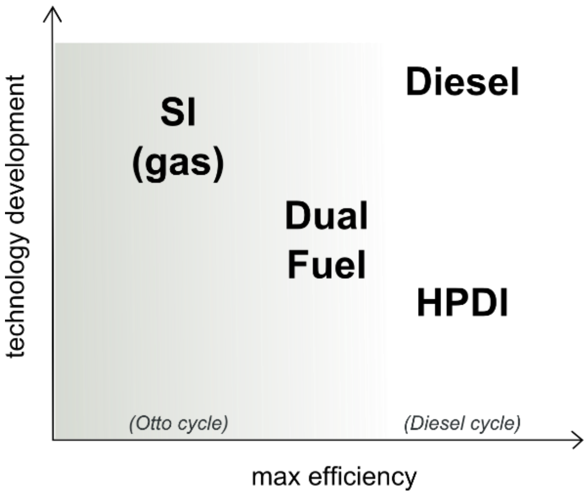

Figure 3 *– Overview of the engine technologies investigated.*

The HPDI technology is based on the dual fuel concept with direct in-cylinder injection of gaseous fuel providing the conditions for mixed limited combustion, in similarity to conventional diesel engines. The gas and diesel are supplied using the special high-pressure gas injection system produced by Westport Inc. [27]; owing to the high injection pressure, only liquefied gases can be used in HPDI vehicles. A major advantage of the HPDI engine over other gas engine technologies is the absence of the knocking restriction on the upper-load limit, which improves the efficiency at high loads. Furthermore, emissions of hydrocarbons are lower than in DF engines [7].

The reference efficiencies and diesel fractions in the fuel blends for state-of-the-art engines are listed in Table 3. The maximum efficiency of the SI gas engine is estimated to be 39%, as derived from several sources [28, 29]. It should be noted that this value refers to the most recent SI units, whereas older engines typically have significantly lower efficiencies.

Table 2– *Engine efficiencies (maximum and cycle) and diesel fuel fractions.*

Engine type	**Diesel**	**SI**		**HPDI**		**DF**	
Fuel type	*Diesel*	*CNG*	*LNG*	*Diesel + CNG*	*Diesel + LNG*	*Diesel + CNG*	*Diesel + LNG*
Cycle efficiency (%)	43	35	35	43	43	40	40
Max efficiency (%)	45	39	39	45	45	42	42
Diesel fraction (en.%)	100	0	0	5	5	30	30

The values shown for HPDI refer to the investigation conducted by Volvo [7], where the engine was compared with the corresponding diesel unit. In a report from the Swedish Gas Technology Center [30], an SI vehicle, a dual-fuel CNG truck, and a diesel vehicle were compared. The DF vehicle showed a high degree of methane slip, as the after-treatment system had not been completed at that time. Additional tests were carried out on the DF engine within the ConGas project using a single-cylinder research engine. It was found that the efficiency and the diesel fraction of the DF engine were more heavily dependent upon the load and the fuel quality [32, 33], as compared to the HPDI engine. Despite the fact that DF achieves a high efficiency and a low diesel fraction at medium load, operation of the engine is shifted towards higher diesel fractions at both low and high loads. Knocking is the limiting factor at high load; even if it can be partially controlled through the recirculation of exhaust gases, the fraction of diesel is increased for smooth operation of the engine. At low load, the level of emission of unburned gas is increased and more diesel fuel is injected to achieve more effective combustion [31]. During the tests, the DF engine was optimized so as to minimize the levels of emissions, in order to meet the Euro 6 regulations.

The gas quality for combustion in a DF engine was investigated based on three parameters related to the gas composition: lower heating value (LHV); methane number (MN); and flame propagation. The LHV represents the energy content of the fuel and it can be used to determine the maximum load and power of the engine. MN is commonly used to express the level of resistance to knock of gaseous fuels. In the present study, the AVL method [34] was applied to calculate the value of NM from the gas composition. Flame propagation under lean conditions depends on the set of fuel properties and engine parameters, and it is not related solely to the fuel composition. However, it has been shown [33] that fuels with high MN (typically those that include long hydrocarbons) achieve more complete combustion under lean conditions than does pure methane.

1.6 Current biomethane process – GoBiGas Phase 1

The production of biofuels, particularly biomethane, is a complex process that involves several stages before and after the conversion of the biomass into an energy dense gas. Figure 4 shows the scheme for the biomethane process based on the GoBiGas plant, which is fed with wood chips. The process can be divided into five macro-areas: drying, gasification, gas cleaning, methanation, and gas upgrading.

The first step, the drying of the wood chips, is crucial for process efficiency. As the water content of the biomass is usually about 50%, it must be decreased substantially prior to gasification in order to avoid increasing the high-temperature energy input of the gasifier [35, 36]. Nevertheless, biomass drying is an intense process that requires a substantial input of energy, which if it uses valuable heat reduces the overall efficiency of the process. Therefore, it is advantageous to integrate the drying system into the heat exchanger network of the biomass plant and to use low-temperature or waste heat.

Gasification of biomass yields a number of different products in the solid, liquid and gas phases, the distribution and composition of which depend on the gasification technology applied, in combination with the operational parameters. One of the advantages of the DFB gasifier over alternative technologies is the rather high fraction of methane in the produced raw gas (10%–15% vol.) [15, 37], which reduces the conversion losses in the subsequent methanation step. A negative aspect of DFB gasification is that it can yield high levels of tar and other organic compounds (OC) [37-39]. These compounds must be dealt with as part of the operation of the plant, as they can cause fouling of the downstream equipment and deactivation of the catalyst in the methanation step [40-42]. Therefore, they must be removed upstream of the biofuel synthesis steps in the gas-cleaning section. The total yield of tar and OC affects not only the efficiency of the process, but also its complexity. The technical and economic feasibilities of gasification processes can be susceptible to the performance of the gas-cleaning steps upstream of fuel synthesis [11].

Gas cleaning is the most complex part of the process, as it involves the separation of solid particles (entrained ash and bed material), sulfur compounds (hydrogen sulfide, carbonyl sulfide), alkali (chlorine), tars, and OC, which can cause fouling in pipes and heat exchangers. The particles are removed using cyclones and filters at a temperature higher than that needed for water condensation. A wet cleaning system for drying the raw gas is usually necessary. The wet cleaning process can be combined with tar removal by scrubbing the gas with oil or rapeseed methyl ester (RME), as in the GoBiGas case. Methods for reforming the tar and OC should be implemented if effective. Reforming can be achieved in the gasifier through the use of a catalytic component in the bed material or through an external reformer. In the latter case, the raw gas is introduced into a secondary reactor that contains active bed material for reforming the tars, although other measures, such as thermal cracking, can be used. The main advantages of reducing the tar content of the gas are reduced consumption of the scrubbing agent and increased chemical efficiency of the process, since the OC can contain up to 10% of the fuel energy [37]. In the GoBiGas process, the consumption of RME depends on the concentration of the removed tar, which consists of naphthalene and a small fraction of heavier compounds. Lighter tar compounds, such as benzene, toluene and xylenes (BTX), together with chlorine are sequestered in the active carbon, which is regenerated with steam, and subsequently injected into the after-burner.

Since sulfur components are highly poisonous for the catalysts used in the olefin reformer and the methanation section, they must be reduced to very low concentrations (i.e., ppb levels). The sulfur in the gas is removed by hydrodesulfurization (HDS) and subsequent H_2S sequestration using amine scrubbing, moreover sulfur guards are included. Recovery of sulfur at the current scale of biomass plants compounds is not economically feasible due to the low content of sulfur in the woody biomass, as compared to the content in raw oil and coal, which are the reference fuels for sulfur recovery processes.

Upstream of the methanation section, the clean gas is conditioned by reforming the hydrogenated olefin and shifting the ratio of hydrogen to carbon monoxide to 3 in a water-gas shift reactor. The excess CO_2 is removed by passage through an amine scrubber, and the conditioned gas is sent to the methanation reactors. The methanation reaction is highly exothermic (≈206 kJ/mol$_{CH4}$), and the cooling of the reactors is a source of recoverable heat within the process, along with the cooling of the raw gas and flue gases. The final gas upgrading involves the drying of the gas and compression at 30 bar, which is the pressure level required for injection into the local NG grid (in Sweden).

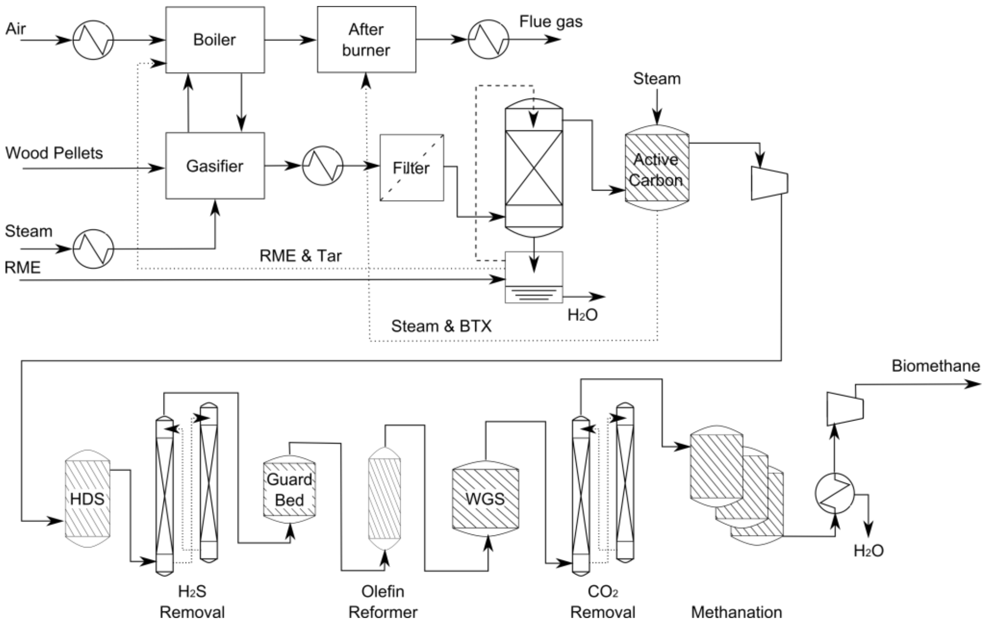

***Figure 4** – Schematic of the GoBiGas Phase I process.*

The combustion side of the gasifier comprises two reactors: a boiler and a post-combustion chamber. The post-combustion chamber allows lean combustion within the boiler, so as to limit oxidation of the bed material (i.e., oxygen transport to the gasifier) while burning other off-streams (tar off-gases, ammonia, hydrogen sulfide) in the process. This layout provides flexibility in the handling of the process streams and the heat balance.

In the boiler, the char that exits the gasifier is combusted together with the tar-rich scrubbing agent and eventually, some product gas is combusted to balance heat in the process. The flue gases are then sent to the heat exchanger section where air and steam are preheated. In Phase 1 of the GoBiGas project, no steam cycle is implemented for electricity production, and the excess heat from the process is used for district heating. In a larger plant with a dryer and electricity production, the excess heat from the plant would be significantly lower.

Energy balance of the GoBiGas-phase 1 process

The GoBiGas plant produces 20.5 MW of biomethane from an input of 32 MW of wood pellets (with the ambition to convert wood chips by the beginning of Year 2016), which are a byproduct of the Södra Cell combined pulp and paper plant in Värö. The drying of sawdust during pellet production requires low-temperature heat, which in the Värö plant is obtained through the combustion of wood residues in the pulp mill. Thus, for GoBiGas Phase 1, biomass drying is performed upstream of the plant. Figure 5 gives an overview of the steps involved in the biomethane production process. For the production of 1 MJ of biomethane, the process consumes 1.56 MJ of wood pellets, with the difference (0.56 MJ) being released as waste heat. Part of the waste heat (0.2 MJ) is directly recovered in the district heating (DH) network, with an additional 0.29 MJ being delivered to DH through heat pumps that consume 0.09 MJ_{el}. In addition to the wood pellets, the gasification process requires electricity at a rate of 0.037 $MJ_{el}/MJ_{biometh}$ and the RME used in the gas cleaning process corresponds to 0.024 $MJ_{RME}/MJ_{biometh}$. This solution, which combines heat pumps and DH, is favorable for Swedish conditions, whereby almost all towns and cities have DH networks. However, this is not the case in many countries in Europe and it remains to be seen if DH will be widely implemented. While electricity from the net is used in this plant, future plants of larger size, as proposed for the second phase of GoBiGas, could be equipped with steam cycles for electricity production, which together with the drying would minimize the level of excess heat.

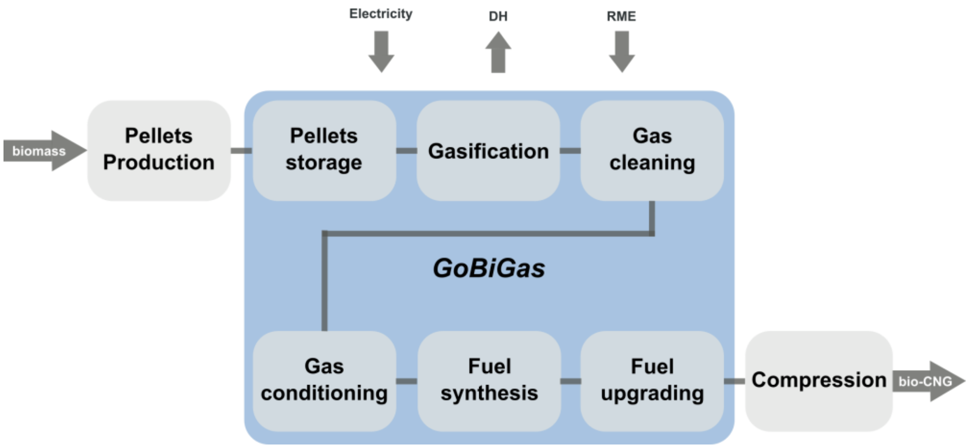

Figure 5 *– Boundaries of the GoBiGas Phase 1 plant and energy streams.*

In the present study, the efficiency of the plant is defined by the total efficiency (Eq. 1) and the chemical efficiency (Eq. 2). For Phase 1 of the GoBiGas plant, the design efficiency (η_{plant}) is 87% when DH by heat pumping is included, and is 74% without heat pumping. The design chemical efficiency η_{ch4} is 61.7% if on does not consider the electricity used by the heat pumps. The final compressions up to network pressure (30 bar) require an additional 0.011 MJ/MJ$_{biometh}$, which is not included in the calculation of the efficiencies:

$$\eta_{plant} = \frac{LHV_{biometh} \cdot m_{biometh} + DH}{m_f \cdot LHV_f + m_{RME} \cdot LHV_{RME} + El} \quad (1)$$

$$\eta_{ch4} = \frac{LHV_{biometh} \cdot m_{biometh}}{m_f \cdot LHV_f + m_{RME} \cdot LHV_{RME} + El} \quad (2)$$

1.7 Biomass drying for DFB steam gasifiers

Woody biomass at the point of delivery is usually in the form of chips or chunks with the largest dimension in the range of 10–80 mm [43], and a moisture content of 50%–60% depending on the season and the type of wood. If the biomass has a heating value (LHV) on a dry basis of <19 MJ/kg and a moisture content of 50% on a wet basis (w.b.), the heat demand for complete evaporation of the moisture and heating to a gasification temperature of 900°C corresponds to 22% of the LHV of the fuel. However, if the biomass is pre-dried to 10% w.b. moisture, the heat demand is only 2.5% of the fuel LHV. During gasification, this heat is provided by combustion of the fuel or product gas. By reducing the moisture content, a higher fraction of the biomass can be gasified and, thereby, the total efficiency of the process is increased. Drying is also beneficial in terms of restricting the dimensions of the gasifier and the ancillary equipment.

Nevertheless, biomass drying is an intensive process that requires a substantial input of energy, which influences negatively the total efficiency of the process, if valuable heat is used. Therefore, it is advantageous to use waste heat at low temperature and to integrate the drying system within the heat exchanger network of the biofuel plant. In addition to improving the efficiency of the process, the drying system should minimize the risk of fire and dust explosion, reduce the emissions of pollutants, and ensure homogeneous fuel feeding. A fire or explosion in the dryer can arise from the ignition of volatile organic compounds (VOC) that are released during the drying. Thermal degradation of the biomass starts at temperatures >100°C and becomes significant at temperatures >120–130°C depending on the type of biomass used [44]. The risk of fire is, however, increased for an unintended stop of the dryer during which volatiles can accumulate.

Biomass drying is currently carried out with a variety of drying technologies, the most common being: rotary dryers [45, 46]; fluidized bed dryers (including flash dryers and superheated steam dryers); and belt dryers [47]. Belt dryers are typically better suited to exploiting low-temperature heat (≤130°C), thereby limiting the risk of fire and harmful emissions, and in some cases, allowing heat recovery from the dryer. This technology is the basis for the drying system integrated with the DFB gasifier proposed in Paper III. The proposed concept consists of two consecutive belt dryers with the possibility for intermediate storage. The first stage uses a conventional belt dryer that employs low-temperature heat sources (<100°C) and air as the drying medium. Drying can typically be extended from an initial level in the fuel of 50% moisture to 10%–20% moisture using only waste heat from the remainder of the plant. The conveyor belt used to transport the biomass to the gasifier is substituted by the second belt dryer, which uses steam at a higher temperature (120°–150°C), which reduces the moisture content to just a few percent. The use of steam allows a higher drying temperature with negligible risk of fire and it allows discharging of the fuel directly into the charge hopper of the feeding system, thereby maintaining the biomass in a steam atmosphere. The moisture that is evaporated along the dryer is re-used without condensation, and the potential for heat recovery is significantly increased. The removed moisture is recycled as a gasification agent in the gasifier, thereby reducing the steam consumption in the gasifier and the emissions of OC from the dryer. Assuming that the moisture content of the biomass is reduced from 20% w.b. to 2% w.b., the ratio of the removed moisture to the dry biomass is approximately 0.23, corresponding to 25%–46% of the gasifier's steam demand.

1.8 Biomass conversion in a DFB gasifier

The DFB gasifier is the cornerstone of the biomethane process, since it converts the solid biomass into the raw gas, which is subsequently synthetized into biomethane. The performance of the gasifier determines the efficiency of the process, its complexity, and ultimately, the overall feasibility of the plant.

A DFB system is composed of two FB reactors: a gasifier fluidized with steam: and a combustor fluidized with air. The gasifier is typically the bubbling-bed type and the combustor is the circulating-bed type. Biomass is fed into the gasifier through the fuel feeding system using a purge gas, to prevent air contamination and back-flow of the raw gas. The reactors are separated by two loop seals that are fluidized with steam (Fig. 6), preventing gas mixing and enabling the production of nitrogen-free raw gas.

In the gasifier reactor, the volatile matter and a fraction of the char are converted to raw gas through a series of processes. The unconverted char is transported by the bed material to the boiler, where it is combusted. Circulation of the bed material controls the heat transfer between the reactors, thereby maintaining the heat balance between them. The heat produced in the boiler must be sufficient to cover all the endothermic processes in the system, i.e., the fuel conversion in the gasifier, the heating of the inlet streams, and the external heat demand required by the plant.

Some bed materials have catalytic and oxygen-carrying properties that are used to influence the composition of the raw gas. Some catalytic bed materials, such as olivine, bauxite, and ilmenite, can catalyze the fuel conversion reactions and reduce the yields of tar and OC. They also have oxygen-carrying properties when exposed to the respective oxidizing and reducing environments in the boiler and the gasifier. The oxygen released in the gasifier oxidizes the raw gas components, resulting in a reduction of the energy content, oxidation of OC species, and an increase in the carbon dioxide content. Overall, the oxidation of volatiles is an undesired effect, despite the reduction in tar yield [48], as it lowers the efficiency of the gasifier and increases the amount of CO_2 in the raw gas, necessitating an energy-demanding removal process.

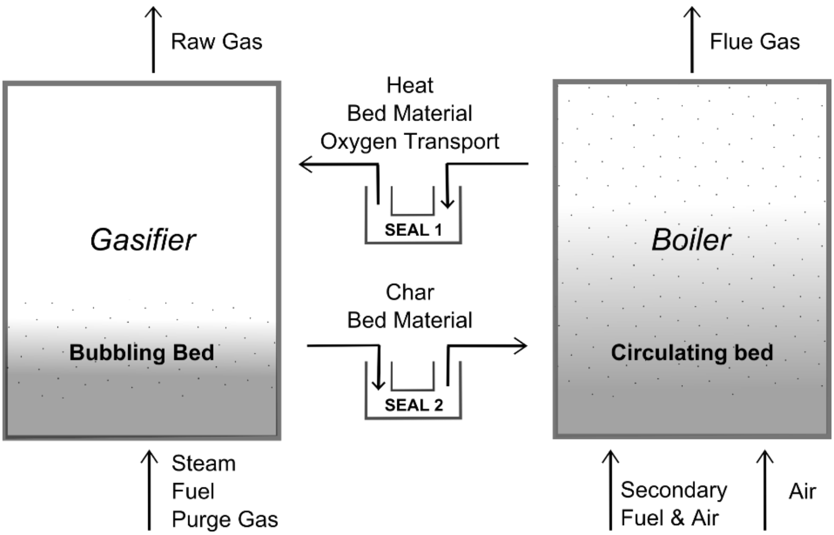

***Figure 6** – Schematic of the DFB gasifier.*

The thermochemical conversion of biomass in the gasifier entails different stages (Fig. 7) [49-51]. Initially, the fuel is dried and devolatilized within a short period of time, as compared with the residence time of the particles in the reactor [52, 53]. The rapid release of volatiles prevents the gasifying agent from interacting with the particles, and devolatilization occurs in an atmosphere of volatiles and water vapor derived from the particle, producing a mixture of permanent gases (PG) and OC. This first step is completed at a relatively low temperature (in the range of 450°–500°C [54]). Thereafter, different reactions occur either homogeneously or heterogeneously between the volatiles gases, tar, and char, converting the intermediate products to the raw gas [50, 54, 55]. Unlike devolatilization, char gasification is a slow process that requires a higher temperature and interaction between the solid phase and the steam. The fraction of char that is gasified (X_g) is often lower than the theoretical optimal value set by the heat balance, and controlling char gasification to achieve high efficiency is currently one of the challenges faced by gasification-based processes.

The raw gas is formed from a mixture of steam, PG, (i.e., H_2, CO, CO_2, CH_4, C_2H_x, C3Hx, C_4H_x), a fraction of undesired OC, which includes paraffin, olefins, alkynes, and tar. The conversion of OC to useful gas, in the gasifier or in external reactors, should be maximized since the OC can retain a significant fraction of the raw gas energy. Currently, research is focused on methods that employ catalytic bed materials in the gasifier or secondary catalytic reformers to improve the rate of conversion of the OC to useful gas components [38, 56-58].

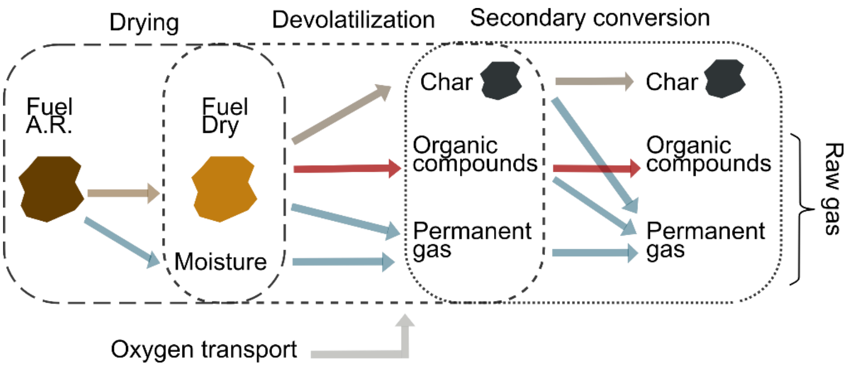

***Figure 7** – Biomass conversion steps in the gasifier.*

Mass balance of the gasifier

The overall fuel conversion can be approximated by a set of reactions that convert the biomass into the volatile products of tar and char (here approximated as pure carbon). In the gasifier reactor, the char can be gasified with H_2O or CO_2, although in the present study, it is assumed that the concentration of steam in the zone surrounding the char particle is sufficiently high compared to that of CO_2 that the char reactions with carbon dioxide can be neglected.

This reaction scheme describes also the mass balance for a simplified composition of the raw gas species. At the present state of research the evaluation of the DFB gasifier involves some empirical correlation based on experimental data, as the chemistry it is not known in detail. Understanding the fuel conversion process beyond the basic reaction scheme will require more and better measurements [59, 60].

Of special interest for the biomethane process are investigations into the yield and composition of the tar, the yield of methane, and the char gasification. Evaluations of these parameters in a comprehensive model require full closure of the mass balance of the gasifier, which cannot be achieved without special measures. Furthermore, the amount of oxygen transported by an active bed material is not detected by conventional measurements, introducing one additional variable into the mass balance equations. In modeling, the fuel conversion scheme can be adapted to the measurement data available, and the key parameter can be assessed within an uncertainty range that reflects the completeness and quality of the measurements. An example of this is provided in the present work, where a reaction scheme that is flexible in relation to different measurement set-ups is used to assess the parameters relevant for the process and associated uncertainty.

Table 3 *– Overall biomass conversion reactions*[1]

Char gasification reactions
$C + H_2O \rightarrow H_2 + CO$
$C + CO_2 \rightarrow 2CO$
Char combustion reactions
$C + O_2 \rightarrow CO_2$
$C + 1/2\, O \rightarrow CO$
Char reaction with metal oxide
$C + zMeO \rightarrow CO + (z-1)MeO + Me$
Overall conversion of OC [4]
$OC + \alpha_1 H_2O + \alpha_2 CO_2 \rightarrow \alpha_3 CO^* + \alpha_4 C_xH_y + \alpha_5 CH_4 + \alpha_6 CO + \alpha_7 H_2 + \alpha_8 C(s) + \alpha_9 CO_2$
Gas-phase reaction in the gasifier
$C_xH_y + xH_2O \rightarrow xCO + \left(\frac{x}{2} + y\right) H_2$
$CH_4 + H_2O \rightarrow CO + 3H_2$
$CH_4 + CO_2 \rightarrow 2CO + 2H_2$
$C_xH_y + \frac{x}{2} CO_2 \rightarrow xCO + \frac{y}{2} H_2$
$CO_2 + H_2 \rightarrow CO + H_2O$
$OC + zMeO \rightarrow OC^* + PG^* + Me$
$OC + PG + H_2O \rightarrow OC^* + PG^*$
$OC + PG \rightarrow OC^* + PG^* + C(s)$

[1]Where the α-coefficients are related to one of the OC and the terms OC^* and PG^*represent the composition of the left organic compounds and permanent gas.

Energy balance of the gasifier

The formulation of the heat balance presented here focuses on the calculation of the internal heat demand (*iHD*) of the gasifier reactor, the chemical efficiency, and the cold gas efficiency. The internal heat demand depends on both the heat required by the fuel conversion and the sensible heat for heating of the inlet streams. In a standalone DFB gasifier, *iHD* sets the theoretical maximum char gasification, i.e., the total yield of raw gas. The efficiency of the gasifier can be expressed as the raw gas efficiency η_{rg} (Eq. 3) and the cold gas efficiency η_{cg} (Eq. 4), corresponding to the fractions of the heating value of the fuel in the raw gas and in the PG, respectively. The *iHD* (Eq. 5) is a key parameter for the optimization of a gasification process, since it correlates with the maximum yield of raw gas, i.e., the maximum efficiency, with the temperatures of the inlet streams: steam, air, and fuel. For a gasifier that is coupled to a boiler and serving other processes, the maximum fraction of char that is gasified is limited by the external heat demand. The gasifier can also be coupled to a large boiler that is fed with secondary fuel or recirculated product gas to satisfy the external heat demand.

$$\eta_{rg} = \frac{\sum_{PG+OC} LHV_i \cdot m_i}{LHV_f} \quad (3)$$

$$\eta_{cg} = \frac{\sum_{PG} LHV_i \cdot m_i}{LHV_f} \quad (4)$$

$$iHD = -(Q_{s_{gasif}} + Q_{r_{gasif}}) = Q_{s_{boil}} + Q_{r_{boil}} \quad (5)$$

The theoretical dependency of the raw gas efficiency on char gasification is shown in Figure 8. The efficiency of the gasifier increases with char gasification, since more fuel is converted. The internal heat demand of the gasifier increases with char gasification due to the overall endothermic gasification reactions. When the internal heat demand cannot be met by the combustion of the char, any further gasification will require combustion of the product gas or secondary fuel to sustain the process. Since the combustion of gasification products reduces the efficiency of the gasifier, as shown in Figure 8, the optimal char gasification is set by the heat balance of the system. The combustion of product gas exerts a greater effect on biomethane plant efficiency (not shown), since at high gasification levels the concentration of methane is reduced and the subsequent fuel synthesis has to be extended. The feeding of the scrubbing products to the boiler represent at the same time a combustion of the gasification product (tar) and a feeding of a secondary fuel (scrubbing agent).

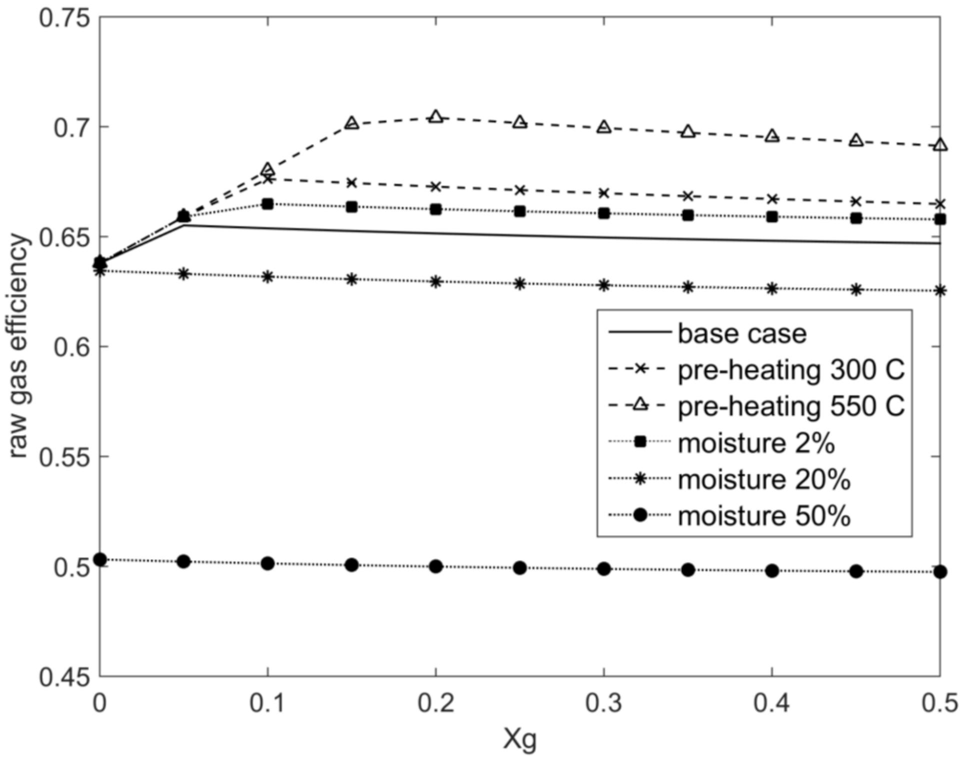

Figure 8 *– Raw gas efficiency as a function of char gasification (X_g).*

The plot is obtained by assuming that char gasification can be increased without changing the temperature in the gasifier, the fluidization or the bed material. If the increase in char gasification incurs an energy penalty other than the heat for gasification (e.g., higher steam demand), the efficiency will be lower. In contrast, the internal heat demand can be reduced and the raw gas efficiency improved by pre-heating the inlet steam and air, and drying the fuel. The effects of pre-heating (steam and air) and drying are shown in Figure 8. Compared to the base case with a low level of pre-heating (120°C), the theoretical raw gas efficiency can be increased by 7% by increasing the temperature of the inlet steam and air to 550°C. The effect of drying is equivalent to a transition from 20% w.b. to 2% w.b.

1.9 Possibilities for optimization of the process

A biomethane plant can be categorized as a bio-refinery with production of three main products: biomethane; electricity; and DH. However, the driving force for this process is the production of biomethane as a transport fuel. Therefore, the construction of a new plant is appropriate for a framework in which biomethane has a high market value. The investment needed for a steam cycle is susceptible to an economic trade-off between the price of DH and the price of electricity. Here, it is assumed that the value of electricity is higher than that of DH and that the installation of a steam cycle that is integrated into a heat recovery network can be justified.

The present study looks at the second phase of the GoBiGas plant from the perspective of maximizing the production of biomethane, as compared to the present situation. Therefore, the investigation aims to optimize the chemical efficiency of the plant. Several possibilities are available to improve the current process, some of which are investigated in this thesis (e.g., drying, in Paper III) and others will be subject of future work (Table 4).

So-called 'primary measures' for the optimization of the gasifier and gas cleaning act to reduce the tar yield and increase the methane content. This can be achieved by using active bed materials and advanced management of the alkali-containing fines from the fuel. Research on gasification is on-going, seeking to improve gasifier performances. The gas cleaning system can be redesigned in line with the improved performances of gasifiers, giving more design options. In this context, a flexible modeling tool for the gasification unit was developed in Paper IV with the objective of utilizing experimental data from research gasifiers of pilot and demonstration sizes to simulate a full-scale process.

The secondary measures focus on the layout and design of the biomethane plant. There are two main areas of intervention: optimized gas cleaning for the separation of valuable tar species; and optimized heat recovery. Tar species, such as benzene and naphthalene, account for 80%–90% of the total tars. These species can be separated as products, since they have a high market value and are considered as renewable in origin. This option can be enabled if it is deemed economically favorable, and may result in a different design of the gas cleaning system.

Table 4 *– Possible measures for improving the efficiency of the biomethane production process.*

	Measures to improve efficiency
Primary measures	Advanced management of fines and alkali from the fuel
	Active bed materials
	Optimized gas cleaning
Secondary measures	Separation of valuable tar products (benzene, naphthalene)
	Drying integrated into the plant
	Enhanced pre-heating of inlet streams
	Electricity production
	Enhanced pre-heating by electricity (internal use of the produced electricity)

The recovery of the excess heat in the plant should be optimized to increase the overall efficiency and economic performance of the plant. There are four main areas where the excess heat can be recovered: drying of the biomass; pre-heating; DH; and electricity production. Both drying and pre-heating reduce the internal heat demand of the gasifier and the boiler, enabling higher gasification rates and yields of raw gas. Several authors have demonstrated the influence of pre-heating on gasifier efficiency [37, 61, 62], proving that it should be pushed as far as possible. Currently, the pre-heating step is limited by the materials used in heat exchangers, which make it economically unfeasible to employ temperatures higher than 500°–600°C. The electricity production from the heat recovered in the biomethane plants is shown [63] to be higher than the internal demand, and this extra production can be sold as a product or be re-utilized in the process to achieve an even higher raw gas efficiency, e.g., by enhancing the pre-heating temperature of the steam and air. Investigations of heat recovery and electricity production will be parts of future studies.

2 Methodology

The WtW chain is a complex system that offers several possibilities for the design of the chain itself and the constituent processes. The general method used to investigate the WtW chain involves identifying sections of the chain that can be optimized to increase the overall efficiency (Fig. 8) and tackling the existing impediments to such improvement. The present study aims to adopt a WtW perspective that focuses on potential improvements in efficiency and emissions, while maintaining a close connection with the technical aspects that regulate the operational and performance levels of the processes (e.g., biomethane production, fuel distribution, and combustion in HD engines).

Paper I of this thesis focuses on the analysis of the existing WtW chain, to establish a reference point for further improvements and to identify areas for intervention. Various cases were compared based on different fuels (bio-CNG, bio-LNG, NG, LNG) and engine technologies (SI, DF, HPDI). The WtW emission intensities were calculated for each case and compared to the reference diesel case. The WtW efficiency for renewable fuels was assessed using a parameter called *biomass impact*, which expresses the emission saving from the diesel case that is specific for the energy input of the biomass. A sensitivity analysis of the results was carried out to elucidate the effects of engine efficiency, the diesel fraction in the fuel blend, CO_2 emissions from the European and Nordic electricity mix, and other factors.

Paper II investigates the quality of the biomethane for combustion in DF engines, which is the technology that is most sensitive to the composition of the fuel. The aim was to set boundaries for the biomethane composition within which the WtW efficiency could be improved. Fuel quality is considered to be a critical aspect, since it influences the efficiencies of both the biomethane process and the engines.

Paper III introduces a concept for the design of a steam dryer. The drying process is simulated through computational fluid dynamics (CFD) using a particle model of the biomass. Drying experiments were performed to validate the model. The integration of the dryer into the process was investigated through a flow-sheet simulation in the Aspen Plus software.

Paper IV deals with the modeling methodology for the gasifier. The approach used in this paper is based on the introduction of experimental data into the model and the related measurement uncertainties. The proposed method combines pre-treatment of the experimental data through stochastic analysis of the uncertainties and simulation of the gasifier based on the overall fuel reaction scheme, which can be implemented in the Aspen Plus flow-sheet software.

2.1 WtW emissions and utilization of biomass resources

WtW analysis, which is well-established for both fossil and renewable fuels, is mainly based on assessment of the emission intensities of the different WtW chains. The biomass feedstock and renewable fuels are considered to be emission-neutral when combusted, i.e., direct and indirect Land Use Changes (LUC and iLUC) are not considered, although for total emissions, we include contributions from methane (CH_4) slips from the engine and methane leakages along the WtW chain and emissions of N_2O from the engines. The emissions intensities along the WtW chain are expressed in grams of CO_2 equivalent per MJ of the LHV of the fuel (gCO_2e/MJ_{fuel}). However, results based on emission intensity are not considered to be sufficient for comparing different biomethane-related WtW chains. There are risks associated with neglecting the specific biomass consumption in the process and with overestimating the importance of cases that show low emissions but with high consumption of biomass. This is related to the availability of biomass resources, which cannot be considered to be unlimited given the land use implications, transportation logistics, and competitive applications of biomass. The *biomass impact* (BI) parameter is defined as the reduction in GHG emissions based on the utilization of biomass resources. The value of BI is calculated from the emissions saved when using biomass-derived fuel instead of diesel for an equivalent engine output, and it is expressed as gCO_2e saved per MJ of dry biomass.

The BI is calculated using specific biomass consumption, *sbc*, taking into account both the efficiency of the biomethane production plant, η_p, and the efficiency of the engine, η_e, based on the gaseous fuel consumption:

$$sbc\left[\frac{MJ_{biom}}{MJ_{out}}\right] = \left(\eta_p\left[\frac{MJ_{biometh}}{MJ_{biom}}\right]\cdot\eta_e\left[\frac{MJ_{out}}{MJ_{biometh}}\right]\right)^{-1} \quad (6)$$

$$\Delta em\left[\frac{gCO2e}{MJ_{out}}\right]_{saved} = em_{diesel}\left[\frac{gCO2e}{MJ_{out}}\right] - em_{case}\left[\frac{gCO2e}{MJ_{out}}\right] \quad (7)$$

$$BI\left[\frac{gCO2e}{MJ_{biom}}\right]_{saved} = \frac{\Delta em\left[\frac{gCO2e}{MJ_{out}}\right]_{saved}}{sbc\left[\frac{MJ_{biom}}{MJ_{out}}\right]} \quad (8)$$

where *em* is the emission intensity per MJ of engine output, and *Δem* is the emissions savings, calculated as the difference between the emission level in the diesel case and the emission levels of the other biomass process pathways.

2.2 Sustainability, emissions and feedstock

The present study focuses on the biomethane from thermochemical conversion of lignocellulose biomass, assuming that the burning of biomass is carbon-neutral. This is not a point for discussion if the feedstock is obtained from long-term managed forests that are maintained on a sustainable basis. In this case, the carbon dioxide emitted when the biofuel is combusted was previously sequestered from the atmosphere by biomass growth and it will be re-sequestered if the forest is managed sustainably. The question regarding direct and indirect emissions for LUC remains connected to the managed land, together with the demand and price of bioenergy. In the present study, we assume that no LCU emissions are associated with the biomass feedstock, which is a reasonable assumption to make for the situation in Sweden where the growing biofuel industry compensates for the reduced demand in the pulp and paper sector. At the present moment, the feedstock allowed in the biomethane process comprises wood pellets and chips, mostly derived from wood processing. However, future developments of the gasification technology will enable the utilization of tree tops and branches [GRenar Och Toppar (GROT)]. The exploitation of GROT for biofuel production will have an effect on the carbon dynamics in forest management. A report in the literature [64] reveals that the harvesting of felling residues leads to an initial decrease in the carbon stock of the soil, which stabilizes over time despite the increased biomass output. Therefore, GROT can be used in the future without considering LUC emissions.

Other emissions from biofuels that are not products of complete combustion are considered as pollutants and accounted for in the total WtW emissions. In this category falls methane leakages along the chain and combustion products that are not CO_2 or H_2O, i.e., methane, other unconverted hydrocarbons, and NOx.

2.3 Pre-design of the steam belt dryer concept

A multi-scale modeling approach is used to evaluate the viability of the proposed dryer design. Information about the drying process for a two-dimensional cut of the steam flow through the packed bed of wood particles is obtained from CFD, in which the evolution of the drying front inside an individual particle is modeled using a particle sub-model. The results from the CFD simulations are used in the macroscopic mass and heat balance (Aspen Plus) of the dryer to enhance the numerical predictions of the capacity of the dryer.

The evaporation temperature of the water in the sub-model was set according to the results of experimental investigations. The normalized weight losses and temperatures of 19 samples of wood chips were recorded during temperature-programmed evaporation in a thermogravimetric analysis (TGA). It was concluded that the amount of tightly bound water (evaporated at high temperature) was insignificant for the investigated fuel, and that all the water evaporates from the particle at a temperature ≤105°C.

2.3 Modeling of the DFB gasifier through experimental data in the flow-sheet simulation

The performance of the gasifier determines the efficiency, the complexity, and ultimately, the overall feasibility of the process. Therefore, any modeling that acts to optimize the biofuel process should include an accurate sub-model of the gasifier. Introducing DFB gasification into the flow-sheet software can be accomplished through thermodynamic or restricted thermodynamic equilibria [61, 65-67], semi-detailed kinetics mechanisms [49, 68-72], and experimental data from existing plants [63, 73-75]. Models that are based on thermodynamic equilibrium produce substantial deviations from the measurements, especially with regard to the yields of methane and tar, and in terms of the carbon conversion. Kinetic models have higher accuracy if the coefficients used in the reaction mechanism are correctly estimated [68]. However, a comprehensive model that includes all the chemical and physical interactions requires extensive knowledge of the process, which is currenly not available. Therefore, not all the required kinetic coefficients can be properly estimated.

Modeling using experimental data avoids the issues linked to the above-described approaches, although it requires an analysis of the measurements to enable simulation of the gasifier. The validity of such a model is never better than the quality and completeness of the available measurement datasets. The total uncertainty as to the variables that describe the fuel conversion must take into account the unclosed mass balance (undetected species) and the intrinsic uncertainty of the measurements. In the present work, a method to investigate the range of uncertainty related to the completeness and quality of the measurements is developed and demonstrated.

The aim of the model is to analyze the large amount of data available from pilot and demonstration gasifiers and to employ these data in process simulation, optimization, and techno-economic analyses [76, 77]. The modeling approach consists of two phases (Fig. 9): 1) a mass and heat balance for the analysis of a database of experimental results (inverse model); and 2) a flow-sheet model of the gasifier (direct model), which can shift between different operational points and use interpolation/extrapolation to simulate other conditions.

For example, specific bed materials, such as olivine, are often used to reduce the tar yield [48, 78-80]. However, the catalytic properties of the bed material are activated after 100 hours [81-83] and in the meantime, the gasifier delivers a different raw gas composition. The operation of the process during the activation period can be simulated using data from inert bed materials in the start-up phase, gradually switching to data from activated olivine though interpolation of the data-points.

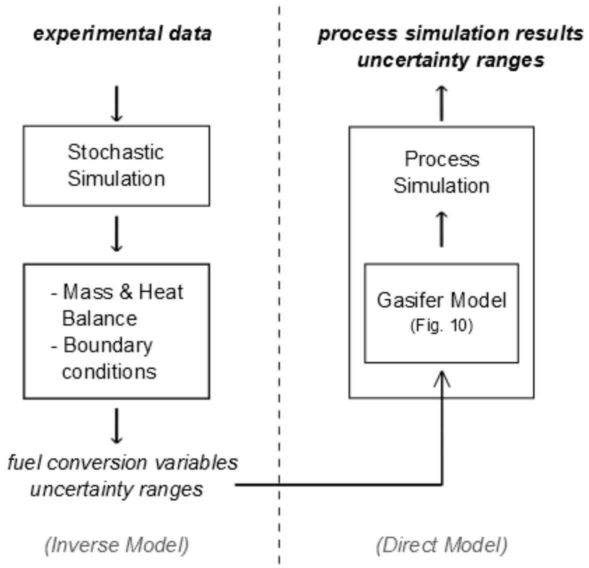

Figure 9 – *Method for utilization of the inverse and direct models.*

2.4 Evaluation of measurement systems for the gasifier

In the data analysis, the experimental data are introduced into the mass balance to calculate the fuel conversion variables that will be used subsequently to simulate the gasifier, along with their uncertainties. The fuel conversion variables used in the model are: the char gasification (X_g); the fraction of volatiles converted to the various raw gas compounds (Z_i); and the oxygen transport (λ_{Otr}). The calculation of the uncertainty linked to the fuel conversion variables must take in account the unclosed mass balance (undetected species) and the intrinsic uncertainties of the measurements. The total uncertainty is calculated using a stochastic simulation of the experimental data (measurements of PG, tar, char yield, fuel composition, etc...), and once resolved, the mass balance for each variation of the data. The mass balance is solved by guessing within valid ranges the two independent variables of char gasification (X_g) and oxygen transport (λ_{Otr}) and retaining the solutions that verify a set of boundary conditions, which includes: the composition of undetected compounds; the fraction of carbon in the raw gas detected by the measurements (*fCd*); and complete devolatilization of the biomass (see Paper IV, Section 3.4). The points on the X_g, λ_{Otr} plane that are solutions for the mass balance form the solution domain for which the mean value and standard deviation are calculated. The mean values are considered to be the most probable solution for the mass balance and the standard deviation represents the total uncertainty. For each solution of the mass balance, all the dependent variables in the mass and heat balance are calculated, along with their mean values and standard deviations.

2.5 Flow-sheet model of the gasifier

The flow-sheet model for simulation of the DFB gasifier is presented in Figure 10. The model is based on three types of fuel conversion variables (X_g, λ_{Otr}, Z_i) calculated from the mass balance of the experimental data, and four temperatures (T_{dev}, T_g, T_{RG} and T_c). In addition to these variables, the temperatures and flow rates of the inlet streams are required (air, steam, fuel). The DFB gasifier is divided into four blocks: blocks 1–3 for the gasifier, and block 4 for the boiler; each block calculates part of the fuel conversion reactions and the corresponding heat terms. Reactor sub-models with known stoichiometry should be used for the gasifier's blocks in the flow-sheet software, e.g., RStoic in Aspen Plus. The sub-model of the boiler can be based either on the known stoichiometry or on equilibrium. An additional calculation is required to obtain the values of λ_{ch}, λ_v and ΔH_{MeO}, which can differ depending on the software; in Aspen Plus, an extra calculator block is added.

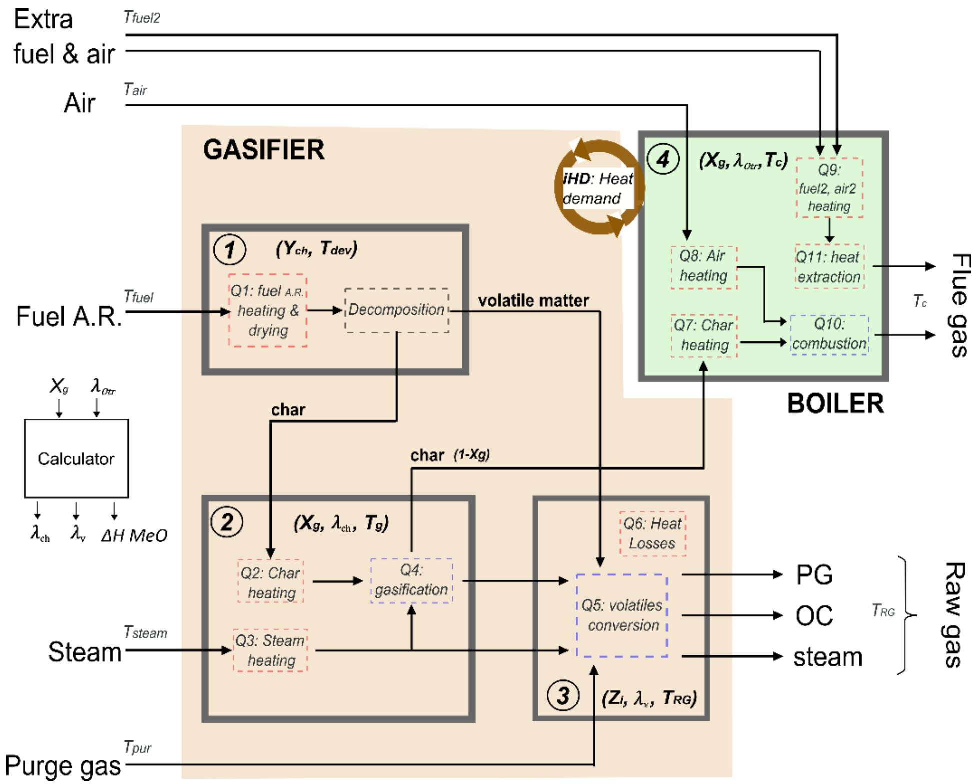

Figure 10 *– Flow-sheet scheme for the DFB gasifier mass and heat balance.*

The flow-sheet model has two objectives: 1) to simulate the gasifier at a single operational point and to evaluate the uncertainty of the process parameters; and 2) to use multiple operational points from a database or use interpolation and extrapolation for process optimization. In the inverse model, the uncertainty related to the fuel conversion variables is calculated. Thereafter, in the direct model it is transferred to the process, resulting in uncertainty ranges for parameters such as: yield of tar and OC or the carbon conversion, efficiency of the gasifier etc. The interpolation of an operational point in a database requires the making of assumptions regarding oxygen transport and the heat demand of the gasifier, as explained in Paper IV (Section 3.3).

3 Results and Discussion

3.1 WtW analysis

In Paper I, the WtW GHG emissions levels for heavy duty engines fueled with biomethane (bio-CNG and bio-LNG), NG, LNG are calculated and compared to those fueled by diesel. Figure 11 shows the results for the WtT and TtW GHG emissions and some results from the sensitivity analysis. The cases based on fossil fuels (CNG and LNG) yield reductions in emissions of 5–38 gCO_2e/MJ_{out}, corresponding to 2%–18% of the diesel emissions. The biomethane cases achieve emissions reductions in the range of 106–160 gCO_2e/MJ_{out}, corresponding to 75%, 50%, and 74% of the diesel emissions for the SI, DF, and HPDI engines, respectively. The influence of the fuel distribution form (LNG, CNG) is small compared to that of the engine technology. The calculations show decreases in WtW emissions of 73%, 46%, and 68% when biomethane is used in the SI, DF, and HPDI engines, respectively, as compared to operating these engines with NG or LNG. Among the biomethane users, the DF cases yield the weakest reductions in emissions despite high engine efficiencies, mostly due to the large fraction of diesel fuel in the energy mix.

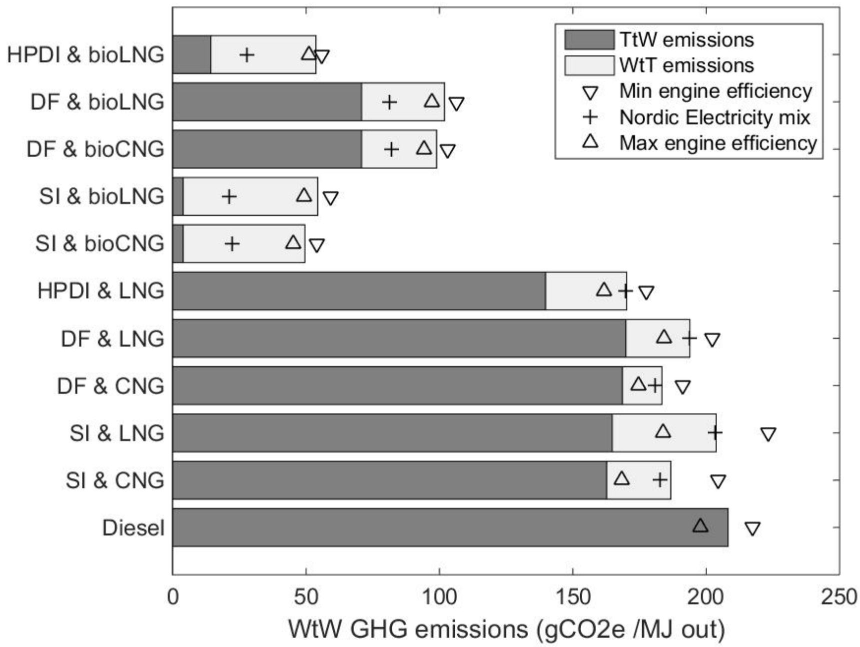

Figure 11 *- WtW, WtT and TtW emissions for different combinations of heavy-duty engines and fuel types.*

In the sensitivity analysis, the efficiencies of the engines were investigated, as well as the effect of switching from the European electricity mix to the Nordic mix. The differences in engine efficiency result in variable levels of emissions, as compared to the base case, corresponding to ±9% for SI and ±4.5% for DF and HPDI, with small differences between the fossil fuel and renewable fuel cases. The biomethane cases are particularly sensitive to the electricity mix, since the emissions from WtT are higher than those from TtW (Fig. 11). The switch from the EU mix to the Nordic electricity mix reduces the WtW emissions by 15–31 gCO_2e/MJ_{out}

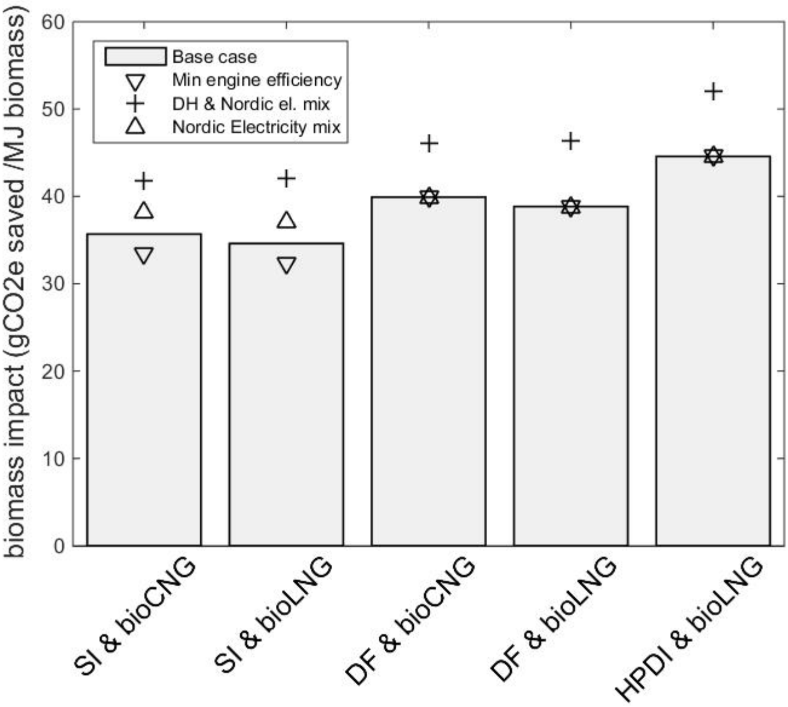

***Figure 12** – Biomass impacts.*

The impact of biomass on the biomethane-based cases is shown in Figure 12. The SI engines require overall more biomass resources per MJ_{out} than the other engines, owing to low engine efficiency, resulting in a BI of around 35 gCO_2e_{saved} per $MJ_{biomass}$, with either bio-LNG or bio-CNG. The DF technology produces lower levels of emissions per $MJ_{biomass}$ than SI owing to higher engine efficiency. The BI values for the DF cases indicate savings of around 40 gCO_2e_{saved}/MJ_{out} for both bio-LNG and bio-CNG, representing the highest BI value among the CNG cases. The most efficient utilization of biomass resources is obtained with the HPDI engine and bio-LNG, which combine high efficiency and a low fraction of diesel to yield a BI value of 45 gCO_2e_{saved} per MJ_{out}.

The effect that engine efficiency exerts on the BI value depends on the specific engine design. Since the BI is based on the comparison of each case with the diesel reference, there is no variation for engines with a diesel design (the same change in engine efficiency is applied to the diesel reference case). For an SI engine, the engine efficiency is different, yielding a biomass impact that ranges from -7% to +6% $gCO_2e/MJ_{biomass}$. The switch from the EU mix to the Nordic electricity mix corresponds to an extra saving of 5–7 gCO_2e per MJ_{out} in the BI.

Overall, improvements in engine performance tend to reduce the differences in emissions and BI between the cases, whereas low performance exacerbates these differences. Among the biomethane cases, the DF engine shows the highest variability in emissions and BI with engine performance. The results show that the DF technology with CNG has the potential to achieve a BI similar to that seen for the HPDI case if future developments increase engine performance. However, this may not be the case if the engine is operated under low-performance conditions. The HPDI engine retains a high BI in all the performance scenarios, thanks to its already optimized efficiency and low diesel fraction.

3.2 Biomethane quality

Twenty-two different compositions of NG [31] from a pipeline, LNG, and biogas have been investigated as being representative of the European gas market (Table 5). Figure 13 shows how the different compositions are distributed on the MN/LHV plane. The general trend shows that as the energy content of the gas increases, the resistance to knocking of the fuel decreases.

Achieving high tolerance to variations in the LHV and MN of the fuel should be a priority for engine manufacturers, since any gas engine will encounter a market with a variety of gaseous fuels. In particular, for DF engines, the strategies to control the knocking and the performance of the after-treatment systems require optimization, as they are crucial for attaining low levels of emissions and a high level of efficiency for the engine.

Table 5: *Compositions of NG from various pipeline and LNG sources[1].*

Vol %	***RUS1***	***DNK***	***NLD1***	***NOR1***	***ALG***	***LBY***	***NOR2***	***RUS2***	***NLD2***	***AUT***
CH_4	98.4	89.8	81.6	92.1	88.3	85.8	86.4	97.8	83.2	85.3
C_2H_6	0.6	5.8	2.7	4.1	6.8	6.9	8.4	0.9	4.0	3.1
C_3H_8	0.2	2.3	0.5	0.9	1.4	1.8	1.9	0.3	0.8	0.5
C_4H_{10}	0.1	0.9	0.2	0.5	0.3	0.7	0.4	0.1	0.2	0.1
C_5H_{12}	0.0	0.2	0.0	0.1	0.1	0.2	0.1	0.0	0.1	0.1
C_6H_{14}	0.0	0.1	0.0	0.1	0.0	0.1	0.0	0.0	0.00	0.0
N_2	0.3	0.4	14.0	1.5	2.4	3.2	0.9	0.8	10.1	9.2
CO_2	0.4	0.5	1.0	0.7	0.7	1.3	1.9	0.1	1.6	1.7
Vol %	***GER***	***SWE#***	***GER#***	***ALG****	***NGA****	***QAT****	***OMA****	***BIO1***	***BIO2***	***BIO3***
CH_4	87.6	90	95.0	87.6	90.4	89.3	86.7	97.5	75.0	80.0
C_2H_6	0.7	5.7	2.6	9.4	5.1	7.1	8.4	0.0	0.0	0.0
C_3H_8	0.1	2.2	0.7	2.0	3.0	2.5	3.3	0.0	0.0	0.0
C_4H_{10}	0.0	0.9	0.4	0.2	1.5	1.0	1.8	0.0	0.0	0.0
C_5H_{12}	0.0	0.2	0.2	0.1	0.0	0.1	0.1	0.0	0.0	0.0
C_6H_{14}	0.0	0.1	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
N_2	9.1	0.3	0.4	0.6	0.0	0.0	0.0	0.0	1.0	10.0
CO_2	2.5	0.6	0.5	0.1	0.0	0.0	0.0	2.5	24.0	10.0

[1]*Investigated compositions for pipeline NG: *, LNG; "BIO", biogas; #, internal market average gas.*

Hydrocarbons longer than methane have the effect of lowering the MN of the gas and eventually increasing the emissions from the engine, if the efficiency is limited by knocking phenomena. Therefore, in biomethane production, hydrocarbons such as ethylene and propylene (which are among the gasification products) should be reformed to syngas and converted to methane. Methane with a purity >95% is considered to be the optimal fuel for combustion in gas engines, as well as for injection into the pipeline. Therefore, the WtT and TtW sections of the chain can be optimized separately.

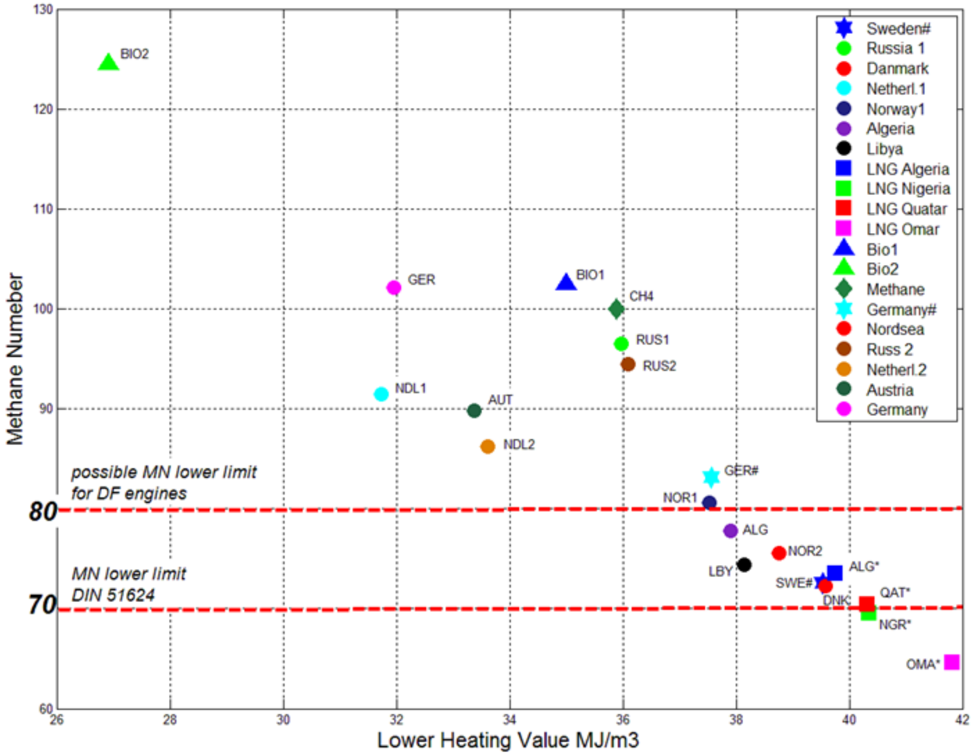

Figure 13 *– MN / LHV map of the European market*

3.3 Steam belt dryer

The proposed belt dryer is composed of three sections that use steam at different temperatures (Fig. 7 in Paper III). A higher temperature is required in the first section to avoid condensation of the drying media onto the biomass, and in the last section to achieve the low moisture content. A steam temperature of 155°C was considered sufficient in the first and last sections in the CFD simulation. Figure 14 reports the temperature profiles along the dryer (left panel) and the moisture content of the biomass (right panel). Both the biomass and the evaporated moisture (steam out) were pre-heated prior to the introduction in the gasifier, thereby increasing the energy efficiency of the gasification process.

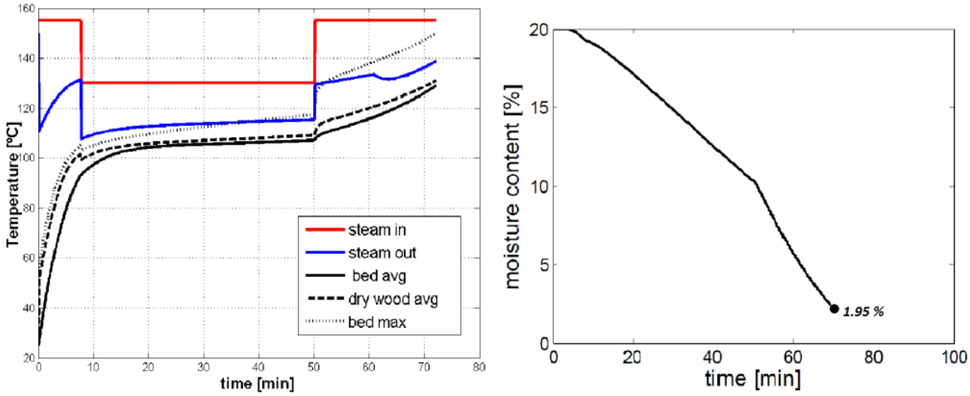

Figure 14 *– Temperature and moisture profiles in the integrated steam dryer.*

The energy balance of the drying system is reported in Figure 15 and includes the electricity required for steam circulation. Here, 93% of the energy input is provided as heat between 140°C and 165°C, in the heat exchanger upstream of the dryer. Most of the energy demand is used for moisture evaporation and wood heating. This heat is recovered in the system through re-use of the moisture as gasification steam and integration of the conveyor belt into the feeding system of the gasifier (i.e., pre-heating of the biomass). The theoretical energy efficiency of the dryer is 95%, since the energy loss is attributed solely to heat losses to the surroundings.

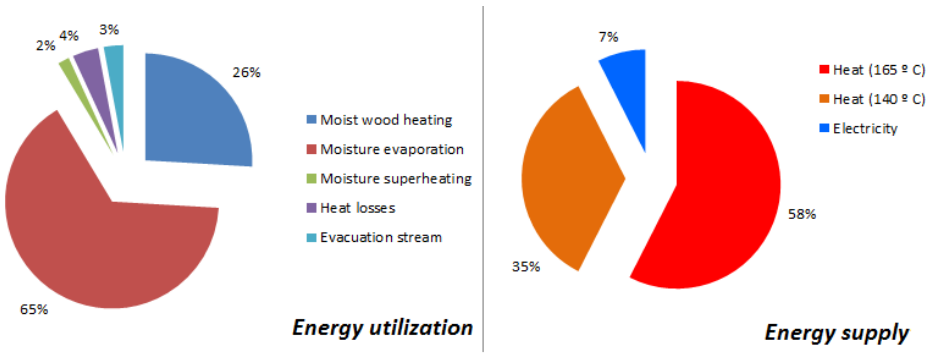

Figure 15 *- Energy balance of the dryer.*

A further energy analysis of the drying process was performed for Paper III and led to an energy efficiency of about 53%. The second-law efficiency of the system is considerably higher than those of other dryers that use air or steam [84-86]. This strong result is due to the recovery of the evaporated moisture as gasification steam, and the heat transfer that occurs with a moderately low temperature difference. This analysis underlines the importance of continuity between the drying step, the pre-heating equipment, and gasifier feeding. Cooling the wood and condensing the moisture after drying are not beneficial for the system, since the sensible heat cannot be recovered for any useful purpose.

The results of the belt design calculation are shown in Table 6. In biomass plants, the length of the conveyor belt is often considerably longer than 100 m due to the logistics of the plant and safety considerations. The results show that the proposed dryer is compatible with the conveyor belts that are currently used in biomass boilers and gasifiers.

***Table 6** – Belt width and belt speed in the two sections of the dryer for different total lengths of the dryer. The final moisture content is 4% w.b.*

Total length [m]	***Width [m]***	***Speed B1 [cm/s]***	***Speed B2 [cm/s]***
100	2.9	5.6	2.8
150	1.9	8.4	4.2
200	1.4	11.2	5.6

3.4 Analysis of the gasifier measurements for modeling

The solution of the mass balance for one measurement point from the Chalmers gasifier is reported in Figure 16. This point was measured using different techniques and the measurement set-ups are compared based on the expected solution of the mass balance (mean values) and the associated uncertainty. Figure 16 shows the mean values (dots) for the solution domain on the X_g, λ_{otr} plane, which is delimited by lines indicating the 95% confidence intervals.

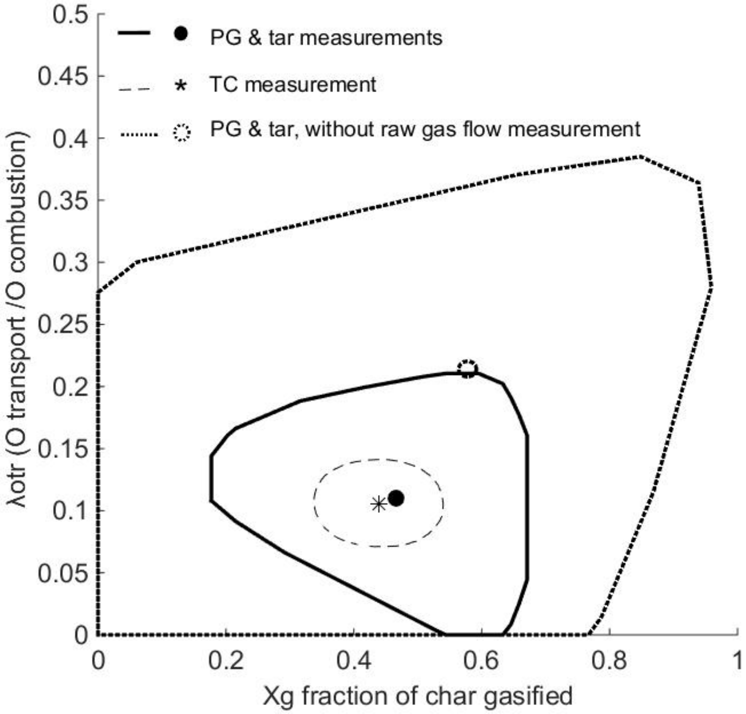

***Figure 16** – Mass balance solution for one measurement point* from the Chalmers gasifier.

The solid dot and solid line represent the solution for the standard measurement set-up with PG measurement, SPA tar measurement, and flow measurement of the raw gas (by helium injection). This measurement set-up (which does not close the carbon balance) is compared with the total carbon measurement set-up (asterisk and dashed line), which has a considerably smaller uncertainty. The results show clearly that the mean values of the two measurements are in good agreement, and that the stochastic approach used for the mass balance can be used to analyze incomplete measurement data.

The largest solution domain (dotted line and dotted circle) is obtained from the base measurement set-up without the raw gas flow measurement. In this case, the mass balance equations have one more degree of freedom and not only the uncertainty is increased. In addition, the mean values are no longer in agreement with the best available measurements. These results show that the raw gas flow is a key measurement in solving with confidence the mass balance; if it is not available some other information based on the experience (e.g., maximum tar amount) must be added to generate reliable results.

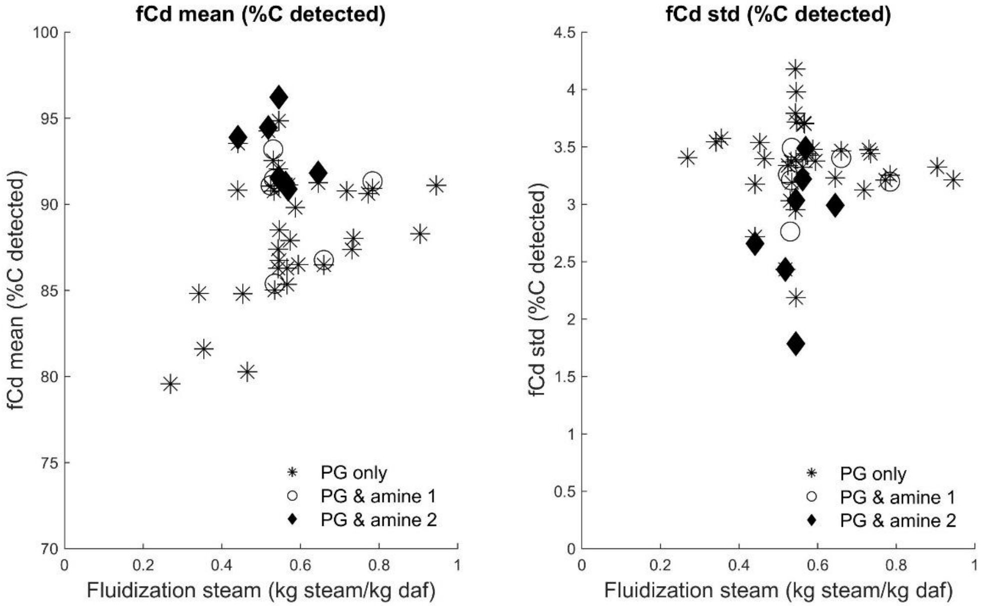

Figure 17 *- Effect of the measurement system on the uncertainty of the mass balance (silica sand and wood pellets).*

The data analysis enables the comparison of measurement set-ups based on their capacities to detect the carbon in the raw gas, through evaluation of the *fCd* (fraction of carbon detected) within the solution domain. Figure 17 compares three measurement set-ups based on the analysis of 37 experimental points obtained with silica sand and wood pellets at different gasification temperatures (T_g790°–830°C) and fluidizations (μ_{st} 0.25–0.95). The three measurement systems used are: PG only; PG and tar with amine 1; and PG and tar with amine 2. As the original amine (amine 1) did not absorb a considerable part of the benzene, the system was upgraded by introducing active carbon (amine 2). Tar sampling with amine 2 showed consistent improvement, increasing the mean *fCd* by around 95% and reducing the standard deviation to <3%. Under these conditions, an *fCd* value >95% can be considered as a target for the measurement system, and further upgrading will produce only marginal improvements.

3.5 Simulation of the gasifier

The results of the data analysis for a dataset of six operational points are reported in Figure 16. The fraction of volatiles converted (Zi) to each of the raw gas compounds that contribute to the energy content is normalized for the oxygen transport as well as for the fraction of char gasification (X_g). The six points are obtained as the temperature is varied at stable fluidization levels and *vice versa*. The distribution of Zi shows the difference in fuel conversion between the operational points. The main variability is in relation to the tar and the OC, which is compensated by different yields of hydrogen and carbon monoxide from the volatiles. The levels of intermediate hydrocarbons, such as methane and ethylene, are only slightly affected by the variation in the OC.

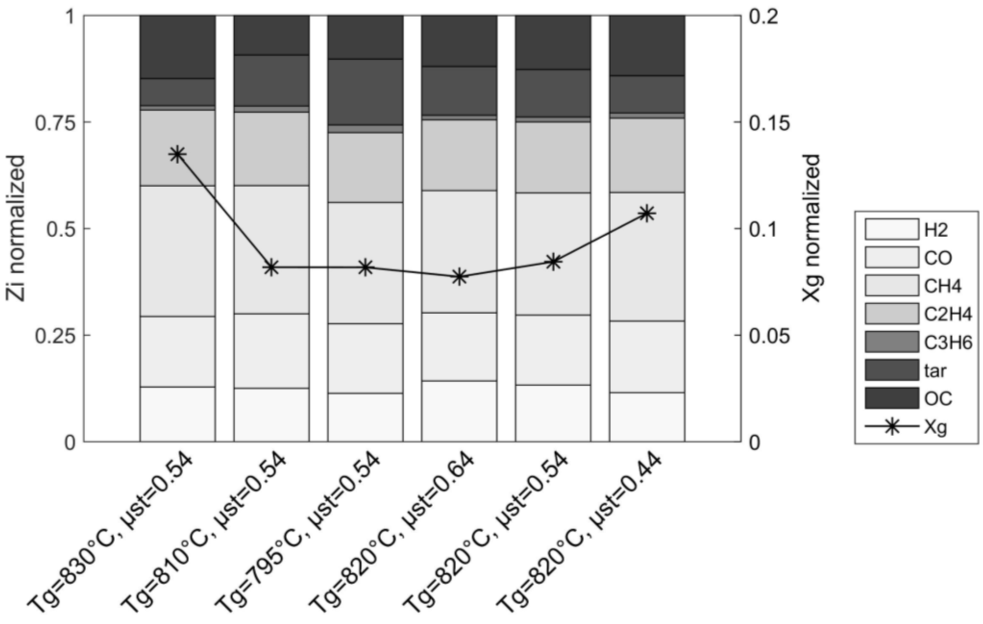

Figure 18 *– Fuel conversion variables for six operational points with silica sand and wood pellets. Shown are the fractions of volatiles converted (Zi) and the fractions of char gasification (Xg), both of which are normalized for the oxygen transport.*

This database (Fig. 18) can be used directly for simulation of the gasifier through the flow-sheet model (Fig. 10), which is used for interpolation and extrapolation of other operational points. An example of this is presented in Table 7, where two new points are obtained.

Table 7 *– Results of interpolation and extrapolation of points based on the data presented in Figure 18.*

	Tg=811°C, μ_{st}=0.56	***Tg=850°C, μ_{st}=0.56***
Variable	***Interpolation***	***Extrapolation***
X_g [-]	0.085	0.25
Z_{H2} [-]	0.12	0.13
Z_{CO} [-]	0.17	0.16
Z_{CH4} [-]	0.29	0.31
Z_{C2H4} [-]	0.17	0.18
Z_{C3H6} [-]	0.01	0.01
Z_{und} [-]	0.11	0.03
Z_{tar} [-]	0.09	0.17
H/C_{min} [-]	1.07	1.20
$R_{h,otr}$ [-]	0.09	0.10
η_{rg} [%]	0.72	0.78
η_{cg} [%]	0.61	0.69
iHD [MJ/kg_{daf}]	3.95	3.48
$Y_{OC,tot}$ [g/Nm^3]	73.6	56.8

Each extrapolated point is calculated with the assumption that the oxygen transport is proportional to the bed material circulation, i.e., to the internal heat demand for the gasifier (Paper IV, Section 3.4).

4 Conclusions

4.1 WtW and fuel quality

The WtW analysis of biomethane use in heavy duty engines reveals that with the current technology it is possible to achieve emission reductions of between 50% and 75% relative to the diesel case. The emission reductions increase to 70%–89% when switching from the EU to the Nordic market due to the lower emissions associated with electricity consumption in the biomethane chain.

Compared to fossil fuels, bio-CNG and bio-LNG, in the EU market, show emissions reductions of 73%, 46%, and 68% for the SI, DF, and HPDI engines, respectively, with small differences noted between bio-CNG and bio-LNG. These results reinforce the role of biomethane in the EU and Swedish strategies for the reducing emissions and dependency on oil.

The comparison of the different engine technologies based on the WtW analysis shows that HPDI achieves the best utilization of the biomass resources, even when a sensitivity analysis of the key parameter is performed. However, the HPDI engine can only use liquefied gas. Among the other engines that can be operated with CNG or bio-CNG, the DF technology has a stronger biomass impact than the SI engines.

The emissions associated with DF engines are susceptible to the quality of the gas, since this influences operation of the DF engine more than it does that of other technologies. A gas composition close to pure methane is considered optimal both for combustion in the DF engine and for the biomethane process. Nevertheless, the EU gas market offers such a variety of gas qualities that the manufacturers of DF engines will need to optimize their strategies for the control of knocking and after-treatment of the exhaust gas.

4.2 Integrated steam dryer

An optimized dryer design plant was investigated to increase the overall efficiency of the biomethane and its performance was estimated though simulations and experiments. Energy efficiency levels up to 95% can be achieved in the drying process by re-using the evaporated moisture as gasification steam and integrating the dryer in the conveyor belt into the feeding system. With this design, the energy supplied for heating and drying of the biomass is recovered in the inlet streams of the gasifier, thereby contributing to reducing the gasifier heat demand and increasing process efficiency. The preliminary design of the dryer/conveyor belt shows that the dimensions are compatible with conveyor belts currently in use in biomass plants. Therefore, the dryer is considered suitable for integration into future plants.

4.3 Model of the gasifier for the flow-sheet simulation

The possibility to use experimental data for simulating the DFB gasifier in flow-sheet models was investigated. The mass and heat balance of the gasifier was based on the inverse model, which calculates the fuel conversion variables from the experimental data, and on the direct model, which uses these variables to simulate the gasifier in the flow-sheet. The model was applied to several operational points from the Chalmers gasifier. The main conclusions are listed below.

- The total uncertainty arising from the measurement set-up was estimated using a stochastic approach. The results were validated against total carbon measurements and it is concluded that the method is accurate as long as measurements of the raw gas flow are available. Otherwise, some other information based on the experience (e.g., maximum tar amount) is required to evaluate the mass balance.
- Different measurement set-ups were compared based on the total uncertainty deriving from the measurements. Set-ups that detect ≥95% of the carbon in the raw gas are considered acceptable. However, total carbon measurements should always be applied, if available.
- It is shown that interpolations and extrapolations of the fuel conversion variables calculated in the data analysis are possible. In particular, the fuel conversion in the Chalmers gasifier can be extrapolated to simulate conditions that are not attainable in this reactor but that are applicable to an industrial-scale gasifier.

Overall, the inverse model and direct model are shown to be flexible tools to simulate the gasifier under several operating conditions, including those related to start-up and disturbances, and these models can be applied to process optimization, to improve biomethane efficiency and reduce WtW emissions.

5 Future work

The future work will focus in part on the analysis of experimental data from the GoBiGas Phase 1 plant, to provide a solid basis for the simulation of the second phase of the project. The database will be used for the development of a flow-sheet model, with built-in modules to enable comparisons of different gas cleaning designs and heat recovery strategies, as well as the optimization of single-/multi-product production. The overall goal is to propose designs that are suitable for processes based on indirect gasification of biomass, for the production of gaseous bio-fuels, such as biomethane and hydrogen. Optimization of bio-fuel production can lead to significant reductions in WtW emissions, and this is a necessary step towards achieving the high production targets set by the European Union.

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