

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

# Large-Scale Production and Use of Biomethane

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Energy Technology  
Department of Energy and Environment  
CHALMERS UNIVERSITY OF TECHNOLOGY  
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## ABSTRACT

Societal ambitions to create an economy based on renewable resources, require the development of technologies transforming these resources into energy-carrying products and biomaterials. Dual fluidized bed (DFB) gasification represents a key technology for achieving sustainability targets, as it is a scalable and highly efficient route for the conversion of biomass. The development of DFB technology has led to the construction of the GoBiGas (Gothenburg-Biomass-Gasification) demonstration plant, in 2014. The GoBiGas plant is a world-first advancement for large-scale production of biofuels as it represents a substantial scaling up of the gasification technology combined with downstream biomethane synthesis. However, to ensure the desired breakthrough of biomass-based products, it is necessary to improve the profitability of gasification plants, through increasing their size, efficiency and identifying opportunities with high economic feasibility for the transport, energy, and chemical sectors.

This thesis presents an exploration of potential improvements for the up-scaling of the biomethane process to a commercial scale. The work summarises and places in context the experience acquired in the research groups at Chalmers and Göteborg Energi AB, including the experience gained from the dedicated experiments in the Chalmers Gasifier and during the commissioning phase of the GoBiGas plant. A method for analysis of the experimental data is introduced, with the goal of improving the quality of the simulations of large-scale gasification processes. The method is applied to the evaluation of the DFB gasifier at the GoBiGas plant, which is presented in the thesis and used as references for further investigations. Some of the measures investigated to increase the profitability of a large-scale plant were proposed in this work, including: an advanced biomass steam dryer integrated with the gasifier, power-to-gas conversion via direct heating of the DFB gasifier and co-production of biomethane with intermediate products for other chemical industries. Furthermore, the utilization of biomethane as fuel for heavy duty vehicles was evaluated within a project in collaboration with Volvo AB. The well-to-wheel approach was applied to calculate the emissions related to three state-of-the-technologies: spark-ignited, dual fuel and high-pressure direct injection.

The evaluation of the GoBiGas plant shows that the gasifier reaches high fuel conversion, with char gasification of ~54%, and the fraction of the volatiles converted to methane of ~34%<sub>mass</sub>. Due to the relatively small scale the heat losses to the surroundings are significant, which affects the cold gas efficiency calculated in 71.7% LHV<sub>daf</sub> with dried biomass (8% moist). The simulation of the DFB gasifier within a large-scale optimised

process shows a cold gas efficiency up to 84% LHV<sub>daf</sub> using fresh biomass (40% moist) with an advanced drying systems. The chemical efficiency of such a plant is calculated in 72% LHV<sub>daf</sub>, which is more than 20pp higher than the current GoBiGas design. Owing to the efficient conversion of the biomass in the gasifier, the co-production of biomethane and other intermediate chemicals represents a feasible opportunity to increase the profitability of the plant. The chemical efficiency of such processes is estimated between 72% and 85% therefore, there is no substantial advantage to produce biomethane, unless biomethane is the desired end-product. The use of biomethane from GoBiGas plant, as fuel for heavy-duty vehicles, reduces the emissions by 30–41 gCO<sub>2e</sub> per MJ<sub>biomass</sub>, compared to diesel. The emission saving increase to 43–54 gCO<sub>2e</sub><sub>saved</sub>/MJ<sub>biomass</sub> if biomethane is produced at large scale. Following the demonstration at a commercial scale, biomethane is established as a biofuel with a high environmental benefit, although the gap between the current status and its potential application is highlighted.

**Keywords:** Biomass gasification, DFB gasifier, biomethane, SNG, dual fuel, process simulation, well-to-wheel, dual fuel, biorefinery, GoBiGas, power-to-gas, biorefinery

## Appended Papers

This thesis is based on the work described in the following papers. The papers are not listed in chronological order based on date of publishing, but rather in the order that makes it easier for the reader to follow the structure of this 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*, Vol. 170, pp 445-454, 2016
- 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”, *Energy & Fuels*, vol. 30 (5), pp 4017–4033, 2016
- V A. Alamia, A. Larsson, C. Breitholtz, H. Thunman, “Performance of large-scale biomass gasifiers in a biorefinery, a state-of-the-art reference”, submitted to *International Journal of Energy Research*, October 2016
- VI A. Alamia, S. Ö. Gardarsdóttir, A. Larsson, F. Norman, H. Thunman, “Comparison of decentralized and standalone thermochemical biorefineries at large scale” submitted to *Energy Technology*, November 2016

### Co-authorship statement

Alberto Alamia was the main author of Papers I-VI. Professor Henrik Thunman was the principal academic supervisor and has contributed with discussions and editing of all the papers. Dr. Ingemar Magnusson was the industrial assistant supervisor in the ConGas project (Paper I and II), he contributed with discussions and editing of Papers I and II and was responsible for the experimental part in Paper I. Professor Filip Johnsson has participated with discussion and editorial comments to Paper I. Dr. Martin Seemann, who was the academic assistant supervisor, contributed with method development, discussion and editing to Paper IV. Dr. Henrik Ström was responsible for the CFD simulations in Paper III and contributed to the editing. Dr. Anton Larsson has collected the experimental data at the GoBiGas plant and contribute to the editing of (Paper V and VI). Stefania Ö. Gardarsdóttir has developed the sub-models for the absorption processes in Paper VI.





## **Related work not included in this thesis**

- Alamia, F. Lind, H. Thunman. “Hydrogen production from biomass gasification for utilization in oil refineries”. ACS 244th National Meeting, Philadelphia, 2012
- A. Alamia , I. Magnusson, “Assessment of waste heat recovery for a power generation system based on Volvo dual fuel engines”, Report



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Alberto Alamia

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# 1 Introduction

The ambition to create a more sustainable society in the European Union drives the transition towards a circular economy [1-4], that is based on renewable energy and the reuse of materials. The implementation of these sustainability principles requires the development of technologies that transform renewable resources into energy-carrying products and biomaterials. In particular, biomass gasification represents a key technology for achieving sustainability targets, as it represents a scalable and highly efficient route for the production renewable hydrocarbons, especially biofuels (e.g., biomethane, ethanol dimethyl ether, hydrogen) and bio-based products (e.g., platform chemicals, biomaterials).

Given the ambition to attain sustainability in the heavy transport sector, a joint venture involving the energy industry, academia, and vehicle manufacturers in the Gothenburg region of Sweden has looked into the possibility of using biomethane. This co-operation has created the GoBiGas plant, which is a first-of-a-kind, industrial-sized demonstration unit that applies indirect gasification to produce biomethane. The production capacity of the GoBiGas plant is 160 GWh biomethane/yr [5]. The plant is owned by the local heat and electricity utility in the City of Gothenburg (Göteborg Energi AB), and was brought into operation in 2014. In the meanwhile, Volvo AB has developed two advanced engine technologies for the combustion of gaseous fuels [6-8], for use in heavy-duty applications.

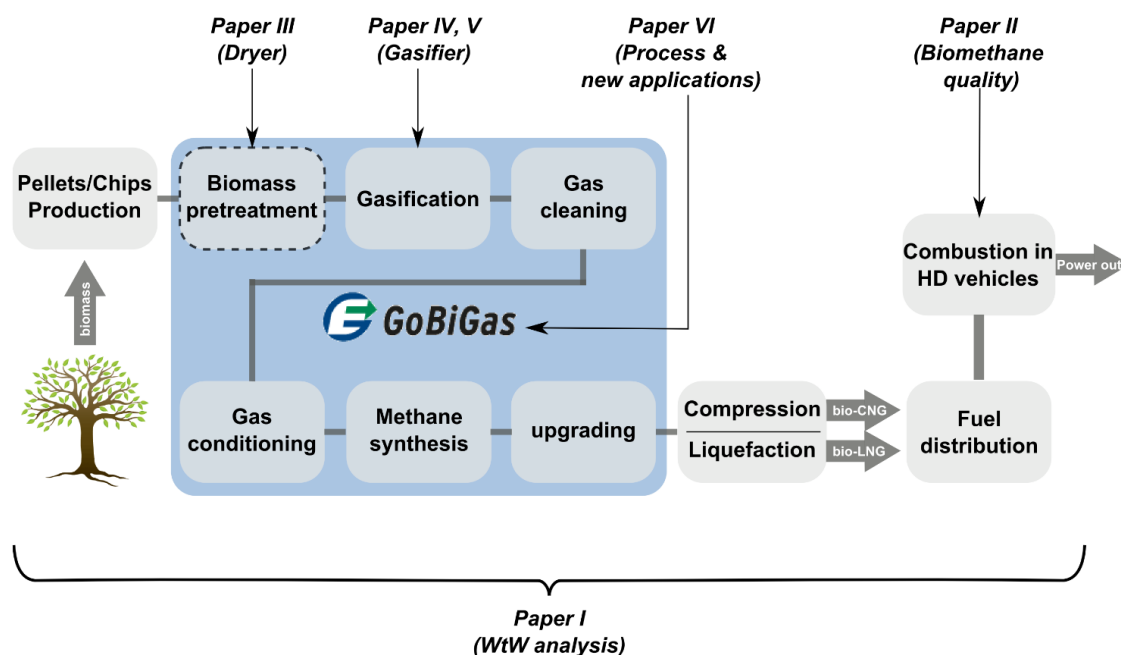
The establishment the GoBiGas process represents a world-first achievement for large-scale production of biofuels, as it is a substantial scaling up of the gasification technology and proves the feasibility of biomethane production on a commercial scale. However, to ensure the desired breakthrough of biomass-based products, it is necessary to improve the profitability of gasification plants, through increasing their size and efficiency, as well as identifying opportunities with greater economic feasibility for the transport, energy, and chemical sectors [9-11].

In this thesis, the current biomethane production chain and possibilities to improve the biomethane process for future applications on a large scale are evaluated. The work has been a collaboration between Chalmers University of Technology, Göteborg Energi AB, and Volvo AB, with the main focus being on the development and evaluation of the gasification process, combined with an evaluation of advanced gas engines [12].

## 1.1 Aim of the work and outline

The overall aim of the work was to investigate various opportunities to enhance the production and usage of biomethane on a large scale. The starting point for the work was an analysis of the current biomethane chain in the GoBiGas plant, combined with the use of biomethane in heavy-duty engines (Fig. 1). This was followed by an exploration of potential improvements for the up-scaling of the biomethane process to a commercial scale, with the focus on the gasification section. The work summarises and places in context the experience acquired in the research groups at Chalmers and Göteborg Energi AB, including the experience gained from more than 4,000 hours of dedicated experiments in the Chalmers Gasifier and during the commissioning phase of the GoBiGas plant[13-15].

In Paper I, the greenhouse gas (GHG) emissions and energy use in the biomethane chain are the subjects of a Well-to-Wheel (WtW) analysis, in which three gas engines technologies are investigated, and biomethane use is compared to the use of natural gas and diesel fuel. Paper II deals with biomethane quality, which is strictly dependent upon its composition, which influences the biomethane process, as well as the operability and emissions of the engine. In Paper III, the potential of implementing an advanced drying strategy for the biomethane process is investigated. Here, a new concept for a biomass belt dryer that can be used in dual fluidized bed (DFB) gasifiers is proposed. The proposed dryer uses low-temperature steam as the drying medium and recovers the evaporated moisture as a gasification agent, thereby reducing the energy and exergy losses associated with the pre-treatment of wood chips supplied to the biomethane plant. Other aspects that are taken in consideration include the: storage of dry wood; inertisation of the fuel; levels of emissions during the drying process; utilisation of low-temperature heat; and integration with the rest of the plant.



**Figure 1** – Overview of the thesis and the topics covered in the appended papers.



In Paper IV, a method for analysis of the experimental data from DFB gasifier is introduced, with the goal of improving the quality of the simulations of gasification processes. The approach is based on a stochastic analysis of the experimental data, enabling accurate estimations of the mass balances and associated uncertainties. The method uses experimental data obtained from the Chalmers gasifier for its validation. In Paper V, the method is applied to the gasification section of the GoBiGas plant using data from the first experimental campaign performed at the plant in the winter of 2015. Thereafter, the data are used to investigate the effects of various identified improvements of the process (i.e., drying, pre-heating, reduction of heat losses, and utilisation of additives in the reactor). Furthermore, a concept of power-to-gas conversion *via* direct heating of the gasifier is introduced and assessed.

In Paper VI, the commercial-scale process is assessed. Here, the focus is on optimising the overall process after the improvements identified in Paper V and the steam dryer concept (Paper III) are introduced. Furthermore, three principally different strategies for the implementation of the technology, *stand-alone*, *local drop-in*, and *decentralised*, are investigated and compared in terms of efficiency, production ranges, and implications for connection to the local energy system.

## 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 aimed at reducing the dependency on oil of the transport sector and achieving deep cuts in emissions [16-19]. The target [20] is a 40% reduction in emissions and oil dependency by Year 2030, as compared to the situation in Year 1990, and 60% by Year 2050, with a share of renewable energy of at least 27%. The EU strategy to reduce emissions in the heavy transport sector focuses on the introduction of alternative fuels, such as first- and second-generation biofuels (including biomethane), natural gas (NG), liquefied natural gas (LNG), and electricity. 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 [4], 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 using 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 [19]. However, first-generation biofuels confer weak climate benefits and have significant negative Land Use Change (LUC) effects [22]. 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 combined advantage 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 [23],

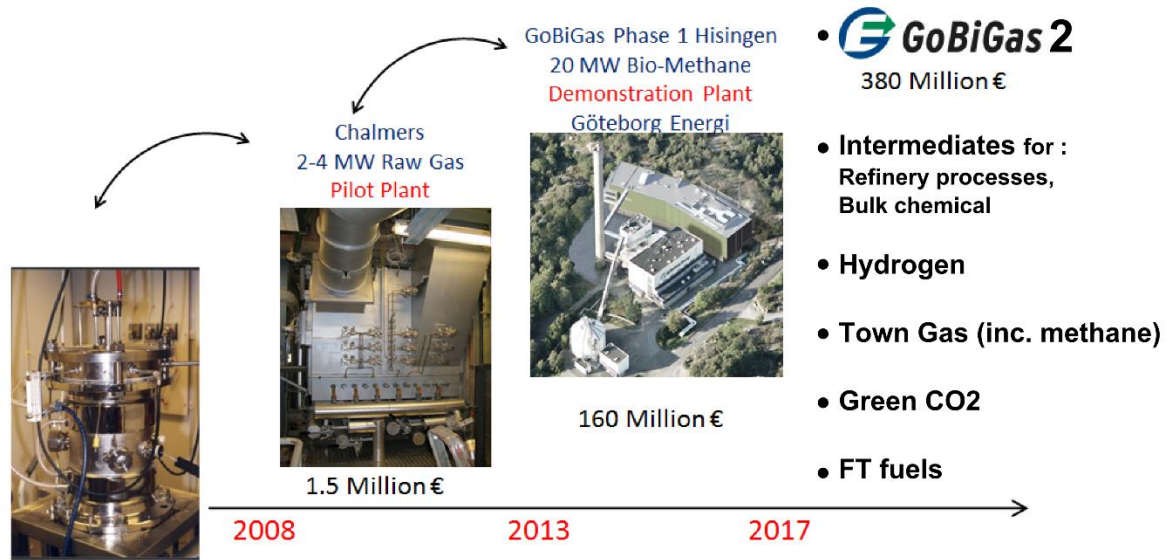
which limits the use of first-generation biofuels to a maximum of 5% and excludes *de facto* first-generation biofuels from post-2020 incentives.

To date, second-generation gaseous biofuels have faced serious challenges, such as high investment costs and lack of distribution infrastructures. In recognition of these limitations, the EC has included second-generation biofuels in the “Clean Power for Transport” [19] (CPT) initiative, which is promoting the development of an infrastructure that is designed to ensure economies of scale for the supply of alternative fuels. In the CPT 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 refuelling station networks by Year 2020. These targets proscribe a maximum distance between refuelling 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 powering long-haul vehicles and 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 [24] 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 plant

Second-generation biofuels can be produced from various feedstocks, such as residues, waste, lignocellulosic biomass, non-food crops, and algae, through biological or thermochemical conversion (gasification or pyrolysis). The latter is especially suitable for lignocellulosic biomass, since it enables conversion of the lignin fraction. Gasification technologies fall into three broad categories: entrained flow (EF); fluidised bed (FB); and DFB. DFB and FB gasifiers are identified as the preferred technologies for biomethane production owing to the large fraction of methane that is present in the produced gas. However, the tar content of the gas can be high, and this affects the operation of the biomethane plant. Compared to an FB auto-thermal (directly heated) gasifier, the DFB allo-thermal (indirectly heated) gasifiers enable the production of nitrogen-free gas, without requiring pure oxygen, thereby avoiding the associated energy penalty. DFB gasifiers have been extensively developed over the last few decades [25]; Some of the most significant gasifiers operating at commercial scale are: the SilvaGas [26] gasifier (1998, USA), the Güssing plant (2001, Austria) [27-29], and the new GoBiGas plant [5].

The planning of the GoBiGas project started in May 2005, together with an ambitious research program funded by government and industry, which also included the building of a 2–4-MW research gasifier on the campus of Chalmers University of Technology, commissioned in December 2007. The GoBiGas project is planned as two main phases, where the first phase includes the construction of a demonstration plant of 32 MW<sub>biomass</sub>. The second phase is for a commercial plant of >100 MW<sub>biomass</sub>. To date (November 2016), only the first phase has been realised and the second phase has been placed on hold. The plant built in the first phase of the project is designed to meet the following targets: production output of 20 MW of biomethane; operation for 8000 h/year; ≥65% chemical efficiency (biomass to biomethane); and a total energy efficiency (biomass plus district heating) of ≥90%.



**Figure 2** –Development of the GoBiGas project and possible future application

The construction of the first GoBiGas plant started in Year 2011 and was completed in November 2013, requiring 300,000 man-hours of engineering and 800,000 man-hours of construction, with associated labour costs of around 90 M€. The actual building contains: 5,000 m<sup>3</sup> of concrete; 800 tonnes of rebar; 1,300 tonnes of structural steel; 25 km of piping; 90 km of electric cables; 130 pumps, compressors, fans, and conveyers; 200 towers, reactors, heat exchangers, tanks, and vessels; 2,500 instruments; and 650 valves [15]. The total cost to date for the project is 165 M€, of which 24 M€ has been provided as governmental support through the Swedish Energy Agency (based on an exchange rate of 9.30 SEK per Euro). Table 1 provides a summary of the investment costs for the different parts of the process (described below in Fig. 3, Section 1.5), defined according to the main component, where the cost includes all the surrounding systems and equipment of the plant, including scale factors to enable estimations of the costs associated with plants of different scales.

The commissioning of the plant took 21 months, during which several major challenges were overcome. Two major breakthroughs occurred during the commissioning phase. First, potassium was added to saturate and stabilise the chemistry that controls the catalytic effect, to assure the quality of the produced gas, thereby avoiding any clogging of the raw gas cooler. Second, the bed height of the gasifier was lowered so that the fuel could be fed closer to the surface of the bubbling bed in the gasifier, thereby reducing the heat transfer and clogging of the fuel-feeding screw and enabling more than 1,600 hours of continuous operation. At the time of writing, October 2016, the plant is operational and delivers biomethane to the natural gas grid [30]. A decision about the construction of the second phase of the GoBiGas project is still on hold[31], despite the commitment of the involved partners (European Union, Government of Sweden, and Göteborg Energi) to pursue development of gasification processes. The main reasons for the delay in reaching a decision are linked to the economics of such a plant (the first of its type) and the current low prices of oil and NG.

**Table 1** – Break-down of the costs for the different parts of the process, including the estimated scale factor (SF), which is defined as:  $C/C_{ref} = (P/P_{ref})SF$ , where C is the Cost, P is the power, and “ref” indicates the values of the reference plant.

Process component	Cost (M€)	Scale factor
Gasifier-section (total)	32.8	
• Fuel feeding	8.25	0.62
• Gasifier	11	0.80
• Raw gas cooler, filter, and scrubber	4.5	0.79
• Flue gas cleaning	8.25	0.55
Methanation section (total)	65.5	
• Carbon beds	13.7	0.62
• Syngas compressor	13.7	0.60
• Hydrogenation and sulphur removal	7.2	0.62
• Shift and pre-methanation	10	0.62
• CO <sub>2</sub> separation	7.2	0.62
• Methanation and drying	13.7	0.62
Buildings and construction (total)	21	0.40

However, future gasification-based plants may have a range of applications that is much broader than just biomethane production. With reference to the sole Swedish industry, possible applications include liquid biofuels from forest residues (*via* catalytic processes or syngas fermentation), biofuels from tall oil, the lining, cracking, separation, and production of intermediate chemicals for the chemical and petrochemical sectors (Fig.2).

## 1.4 State-of-the-art of heavy-duty gas engines technologies

At present, the state-of-the-art gas engine technologies 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 commercialised soon, and it is already available for stationary applications in the US market [27]. The most recent technologies, DF and HPDI, are both based on a diesel engine design and dual fuel combustion concept, where the gaseous fuel is ignited by injection and auto-ignition of a pilot injection of diesel. Nevertheless, their performance and operation are substantially different. The DF engine can be operated with either a gas/diesel mixture or diesel, which is beneficial in regions with poor gas-fuelling infrastructures. The nature of the combustion is similar to that in a conventional, spark-ignited Otto engine, where the gaseous fuel is port-injected and pre-mixed with air and exhaust gases. The combustion process is characterised by pre-mixed flame propagation, as in the SI engine, and the upper-load range is typically limited by knocking. The HPDI technology is based on direct in-cylinder injection of a 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. [32]; 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 lower fraction of diesel [6] and the absence of the knocking restriction on the upper-load limit.

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

Engine type	<b>Diesel</b>	<b>SI</b>		<b>HPDI</b>		<b>DF</b>	
Fuel type	<i>Diesel</i>	<i>CNG</i>	<i>LNG</i>	<i>Diesel + CNG</i>	<i>Diesel + LNG</i>	<i>Diesel + CNG</i>	<i>Diesel + LNG</i>
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

Traditionally, SI engines have enjoyed the largest market share, whereas the DF and HPDI engines have only recently been commercialised. Therefore, filling the knowledge gap in the literature was in the scope of the present work. During the work for this thesis, Chalmers was involved in the development of the DF concept through a collaborative project with Volvo AB Advanced Research and Technology and Göteborg Energi. The project aimed to reach engine performance levels, in terms of drivability and efficiency, comparable to those obtained with diesel, while complying with Euro 6 emission regulations. The project, which was called ConGas, included an evaluation of the quality of the biomethane used for combustion in the DF engines (Paper II) and WtW analyses of biomethane use in DF and HPDI engines (Paper I), since Volvo AB is developing and manufacturing both these technologies. The results of the engine tests during the ConGas project have facilitated the integration of the scarce literature available for DF and HPDI engines and allowed the compilation of a reference table for the efficiencies and diesel fractions of the fuel blends to be used for state-of-the-art engines, as listed in Table 2 (Paper I).

The data for the DF and HPDI engines are compared with the maximum efficiency of the SI gas engine, which is derived from several sources [33, 34] as being around 39%. It should be noted that this value refers to the most recent SI units, whereas older engines typically have significantly lower efficiencies. Owing to the diesel engine design, the efficiencies of the DF and HPDI engines are higher than those of the SI engines, at the cost of a fraction of the fuel mix being from fossil sources. Thanks to the standard diesel injector, DF engines have greater fuel flexibility than HPDI engines [6] [35], although DF engines are more heavily dependent upon the load and the fuel quality [36, 37], as compared to the HPDI engine. The DF engine achieves high efficiency and a low diesel fraction at medium load, whereas the operation of the engine is shifted towards higher diesel fractions at both low and high loads.

## 1.5 Biomethane production process at the GoBiGas plant

The GoBiGas process (Fig. 3) can be divided in two macro sections: gasification, where the solid fuel is converted to the product gas (tar free); and methanation, where the product gas is refined to biomethane. The gasification section is a substantial scale-up from the design of previous research facilities and was built by Valmet AB, on a REPOTEC license from the design of the Gussing DFB gasifier [28, 38]. The methanation section is a scale-down from the industrial state-of-the-art designs of Haldor Topsøe [39, 40]. While the size of the methanation section is not optimal from the perspective of economics [15], a plant

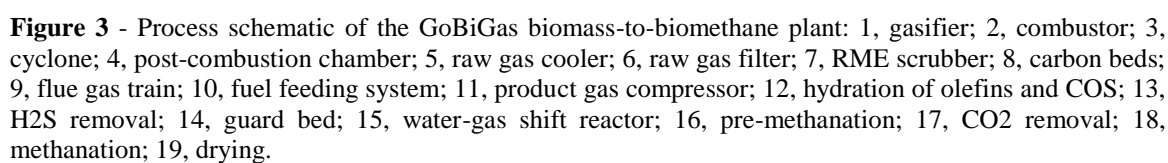
of this size was necessary to understand fully the process before the construction of a commercial plant of  $\geq 100 \text{ MW}_{\text{biomass}}$ . Due to its relatively small size, the GoBiGas plant does not include a biomass dryer or an advanced steam cycle for heat recovery. Instead, the electricity is bought from the grid, and the excess heat is sold as heat *via* the local district heating network.

The DFB gasifier at the GoBiGas plant can be operated with wood pellets, wood chips or forest residues. The fuel is fed to the gasification reactor (nr. 1, Fig. 3), wherein the major part is converted into gas through devolatilisation and partial gasification of the char. The remaining char is transported with the bed material to the combustor (nr. 2), where it is burnt to produce heat. The transfer of heat between the combustor and the gasifier is achieved through circulation of the bed material. The selected material is olivine, which is a natural magnesium-iron-silicate ore that is commonly used in DFB gasifiers due to its ability to reduce the yield of tar and its tendency not to agglomerate at these process temperature levels [14, 41]. However, to achieve the desired catalytic behaviour, olivine needs to be activated. In the literature, different approaches to activating olivine are described; the one used in the GoBiGas plant is based on the addition of potassium [14, 28] and recirculation of the fines containing ash compounds and carbon.

The bed material is separated from the raw gas in the cyclone (nr. 3) and re-circulated to the gasifier to provide the heat necessary for the endothermic processes. A post-combustion chamber (nr. 4) is used to combust the off-gases and slip-streams. The sensible heat in the flue gases is then recovered through heat exchange (nr. 9), to preheat the inlet streams and produce district heating. The produced raw gas is cooled (nr. 5) and particles are removed by passage through a textile-bag filter (nr. 6), before entering the tar scrubber (nr. 7) where rape methyl ester (RME) is used as the scrubbing agent. A continuous flow of fresh RME ( $0.03\text{--}0.035 \text{ MW}_{\text{RME}}/\text{MW}_{\text{biomass}}$ ) is fed to the scrubber to avoid saturation of naphthalene, which is the main tar component removed in this stage. The used RME and the extracted tar are fed to the combustor, providing more heat for the gasification process. Downstream of the RME scrubber, a fan enables re-circulation of the raw gas to the combustor, thereby providing extra heat to the gasification process if necessary. A minimum re-circulation of raw gas is required to stabilise the temperature in the reactor and to cope with fluctuations in the moisture content of the fuel. Light cyclic hydrocarbons, mainly benzene but also small fractions of toluene and xylene (referred to as BTX), remain in the gas and are removed through a series of three fixed beds filled with activated carbon. The plant has four active-carbon beds (nr. 8), enabling regeneration of one bed at all times, using steam. The heat and the tar components in the off-gases from the carbon beds can be recovered by condensation, although they are currently fed directly to the combustor.

The syngas from the gasification requires further cleaning and shift stages to acquire the level of purity and composition required for the final synthesis of methane. The pressure in the pre-methanation section is increased to 16 bar through a six-stage intercooled compressor, to meet the requirements for the hydration of olefins and COS in reactors nr. 12 and nr. 13. The pressure in the pre-methanation section is partially decreased in the  $\text{CO}_2$  separation stage (nr. 17), reducing significantly the consumption of heat in the re-boiler. Both the  $\text{H}_2\text{S}$  and  $\text{CO}_2$ -removal processes are based on a standard absorber-desorber set-up, with a lean-rich solvent heat exchanger between the columns. The  $\text{CO}_2$  that is separated in this process is of high purity, so it is compressed for utilisation as a purge gas and for secondary services; alternatively, these streams can be used for  $\text{CO}_2$  storage or can be

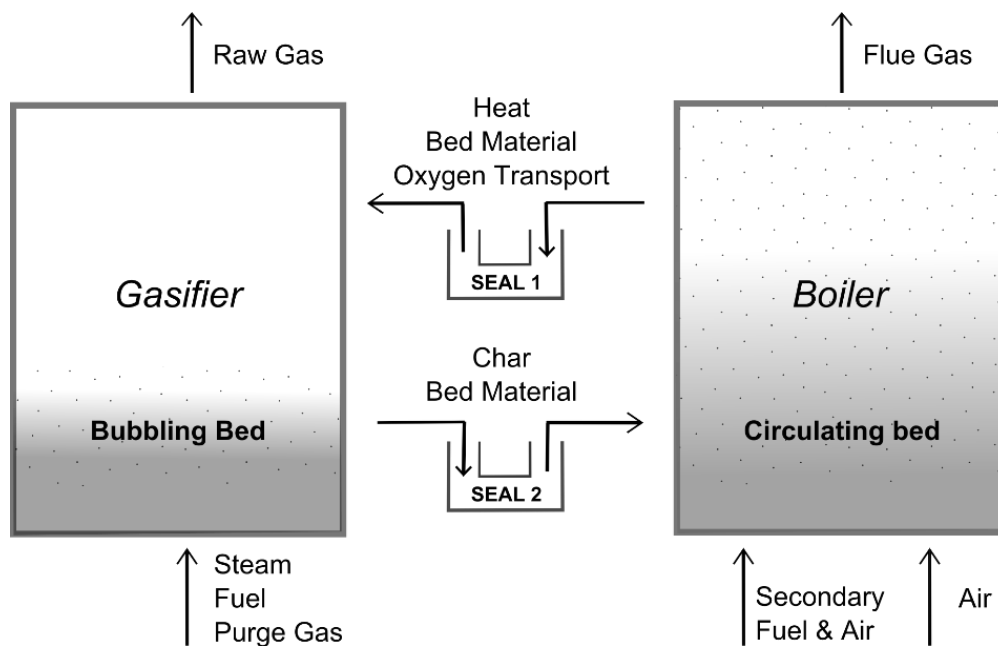
utilised elsewhere. The guard bed (nr. 14) located up-stream of the reactors has a sulphur-sensitive catalyst to protect it from possible contamination. During the hydration and  $\text{H}_2\text{S}$  removal steps, there are a number of heat exchangers and condensers that cool the gas and reduce the steam content to a few percent. Therefore, steam is re-injected into the syngas flow to drive the water-gas shift (WGS) reaction in reactor nr. 15. The WGS reactor is pressurised and operated at a temperature of around  $300^\circ\text{C}$ , and the  $\text{H}_2:\text{CO}$  ratio is increased from the original  $\sim 1.7$  to  $>3.0$ , which is optimal for the synthesis of methane. The syngas is directed to a pre-methanation reactor where parts of the carbon monoxide and carbon dioxide are converted to methane (nr. 16). This reaction is strongly endothermic, raising the temperature to close to  $680^\circ\text{C}$ . The final stage of the process entails Haldor Topsoe TREMP [39] methanation, which involves a series of four methanation reactors (nr. 18), with intermediate drying and without recycling of the gas [40]. The methanation process was developed for biomethane production and it is based on the technology used in coal gasification plants. Biomethane purity  $>96\%_v$  is achieved in four stages and drying of the gas in a temperature swing adsorption dryer (nr. 19). The final product is delivered at 6.0–6.5 bar to the compression station, where the pressure is increased to 30 bar and the gas is injected into the natural gas network.





## 1.6 Biomass conversion in a DFB system

The DFB gasifier is the cornerstone of the biomethane process, since its performance determines the efficiency of the plant, the extents of the gas cleaning and upgrading, and the availability of the whole process. DFB gasifiers 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. The gasification reactor is typically a bubbling bed fluidised with steam, and the combustor is a circulating bed that enables transport throughout the system. The biomass used as fuel is fed to the gasifier by the fuel feeding system, in which a purge gas is used to prevent air contamination and back-flow of the raw gas into the fuel feeding system. The gasification and combustor reactors are separated by two loop seals that are fluidised with steam (Fig. 4), preventing gas mixing and enabling the production of a nitrogen-free raw gas. The circulation of the bed material controls the heat transfer between the reactors, thereby maintaining the heat balance between them. Therefore, 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.



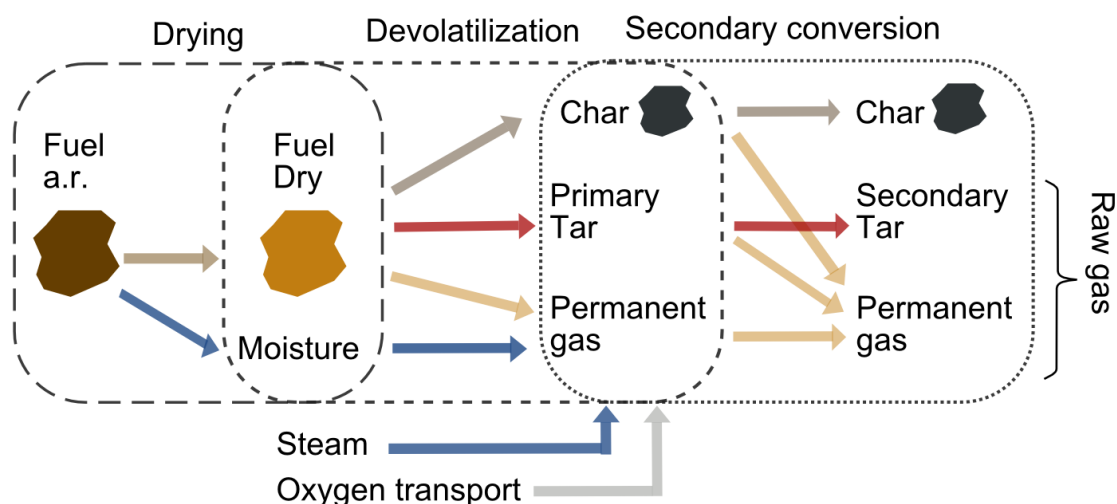
**Figure 4** – Schematic of the DFB gasifier.

The chemical conversion of the biomass, in the gasifier reactor, entails different stages (Fig. 5) [42–44]. Initially, the fuel is dried and devolatilised, usually within 1–2 minutes, which is shorter than the residence time of the particles in the gasification reactor [45, 46]. The rapid release of volatiles prevents the gasifying agent from interacting with the particles, and devolatilisation occurs in an atmosphere of volatiles and water vapour derived from the particles. The gas produced consists of a mixture of permanent gases (PG), water vapour, and primary tar. This first step is completed at a relatively low local temperature inside the fuel particle (in the range of 450°–500°C [47]). Thereafter, the volatiles gases undergo further heating and different reactions occur either homogeneously or heterogeneously between the volatiles gases, tar and char [43, 47, 48]. In particular, the primary tar species

are unstable at these temperatures and are converted into more stable secondary tar through reactions R6 and R12, which increase the yield of PG. The char fraction of the fuel (16%<sub>w</sub> - 20%<sub>w</sub>) contributes to the total PG yield by char gasification reaction, with mainly steam (R1). Unlike devolatilisation, gasification is a slow process that requires a higher temperature and interaction between the solid phase and the steam.

The resulting raw gas mixture may contain a few hundred different species, including problematic tar compounds that can cause clogging of the pipes and coolers, as well as deactivation of the catalyst in the down-stream equipment. Therefore, controlling the formation and destruction of these tar compounds during the gasification process has a major impact on the reliability of the plant. Other aromatic species with lower boiling points do not hinder the operation of the plant, although they must be removed prior to the final synthesis, which means cost and energy penalties. However, as they have a commercial value, to increase the economic feasibility of these types of plants, efforts should be made to optimise the process so that these components can be extracted in such a way to provide income for the plant.

The *in situ* measures used to reduce the tar yield include the use of active bed materials and the catalytic effect of ash compounds. In particular, olivine has been successfully used as the bed material in several gasification plants [14, 49-51]. The activity of olivine towards steam reforming of hydrocarbons has been partially attributable to its iron content, which becomes free reducible iron, through calcination [52]. This process of activation of the bed material occurs during the successive redox cycles between the two reactors, and has been explained in terms of the migration of the iron towards the particle surface [53].

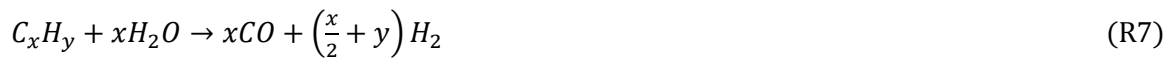
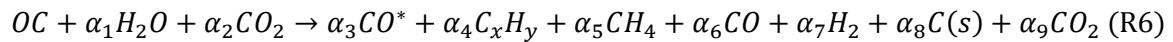


**Figure 5** – Biomass conversion steps in the gasifier reactor.

Ash components, such as Cl, Ca, K, Si, S, Al, Mg, and Na, are of great interest in gasification, since they are known to catalyse char gasification [54], WGS reaction [55] and tar decomposition [56], and they can significantly enhance the catalytic effect of bed material. The activation of the bed material has been partially explained in terms of an ash coating forming around the bed material particles [51, 52]. Nevertheless, the interactions that occur between ash compounds and the bed material particles are not yet fully understood [57], and ongoing research is focusing on the topic [58, 59].

These ash compounds species can come from the fuel, or externally introduced using additives rich in inorganics. In particular  $K_2CO_3$  is used for the activation of olivine in the GoBiGas plant [13, 14], and other additive (e.g. sulphur) have been tested in the plant [60]. Marinkovic has recently proposed that the catalytic activity of olivine is related to the potassium uptake and release that occurs between the two reactors [51, 61]. In particular, it is assumed that the potassium is taken up by the bed material at the combustion side (e.g., in the form of  $K_2SO_4$ ) and is released in the reducing atmosphere of the gasifier in the form of catalytically active  $KOH$  (g) or  $KCO_3$  (g).

Another property of the catalytic bed material is the capacity to transport oxygen from the oxidising atmosphere in the combustion chamber to the reducing atmosphere in the gasification reactor. The oxygen-transport capacity is associated with the metallic species, and this phenomenon has been investigated intensively in the field of Chemical Looping Combustion (CLC) [53, 62, 63]. The main effects of oxygen transport are oxidation of the gaseous compounds in the gasifier and increased conversion of the char [64]. Overall, while oxygen transport is undesirable, it is an intrinsic consequence of circulation of the bed material and ash species in the systems.



<sup>1</sup>Where the  $\alpha$ -coefficients are related to one of the organic compounds (OC) and the terms  $OC^*$  and  $PG^*$  represent the composition of the remnant organic compounds and permanent gas.

## 1.7 Possibilities for optimization of the biomethane process

Initial developments of gasification-based plants in Europe have focused on *stand-alone* and single-product processes for the production of electricity (Gussing) or biofuels, such as biomethane (GoBiGas) and hydrogen. The future energy plans in the EU include the introduction of large-scale, high-efficiency gasification plants. However, the absence of long-term national policies that would ensure stability for investors and the slow development of an infrastructure for the distribution of biofuels (e.g., LNG, CNG, and DME filling stations) and of various vehicle technologies (LNG or DME vehicles), have hindered the construction of the required facilities [11, 65, 66].

The profitability of *stand-alone* plants is dependent not only upon the value of the products, but also on the investment cost related to the complexity of the process [67, 68]. Developments in the technology and process designs are expected to reduce the complexity of the core sections of new plants, such as those for gasification and biofuel synthesis. However, other areas that are highly beneficial in terms of the overall efficiency will retain a high level of complexity, such as the handling and preparation (including the drying) of the biomass, and the steam cycle integrated in the heat recovery network. The profitability levels of new biomethane plants can be increased in two ways: 1) technical improvements, which increase the efficiency and reduce the complexity of the plant; and 2) the discovery of other economically feasible strategies for the generation from the same plant of a spectrum of products intended for the chemical, transport, and energy sectors (see Section 1.8).

A list of possible technical improvements to the biomethane process, separated into primary and secondary measures, are reported in Table 3. Primary measures focus on the performance of the gasifier, aiming to lower the tar yield, increase fuel conversion (gasification), and increase the methane content. These measures entail research on the activation of the bed material, the catalytic effect of alkali, the use of potassium- or sulphur-based additives, strategies for the re-circulation of fines, and reactor design. These areas are not directly investigated in this thesis, although the method for analysis of the experimental data (Paper IV) and the gasifier model (Paper IV and V) can take into account improvements from primary measures.

**Table 3** – Possible measures for improving the efficiency of the gasification process (based on current GoBiGas design)

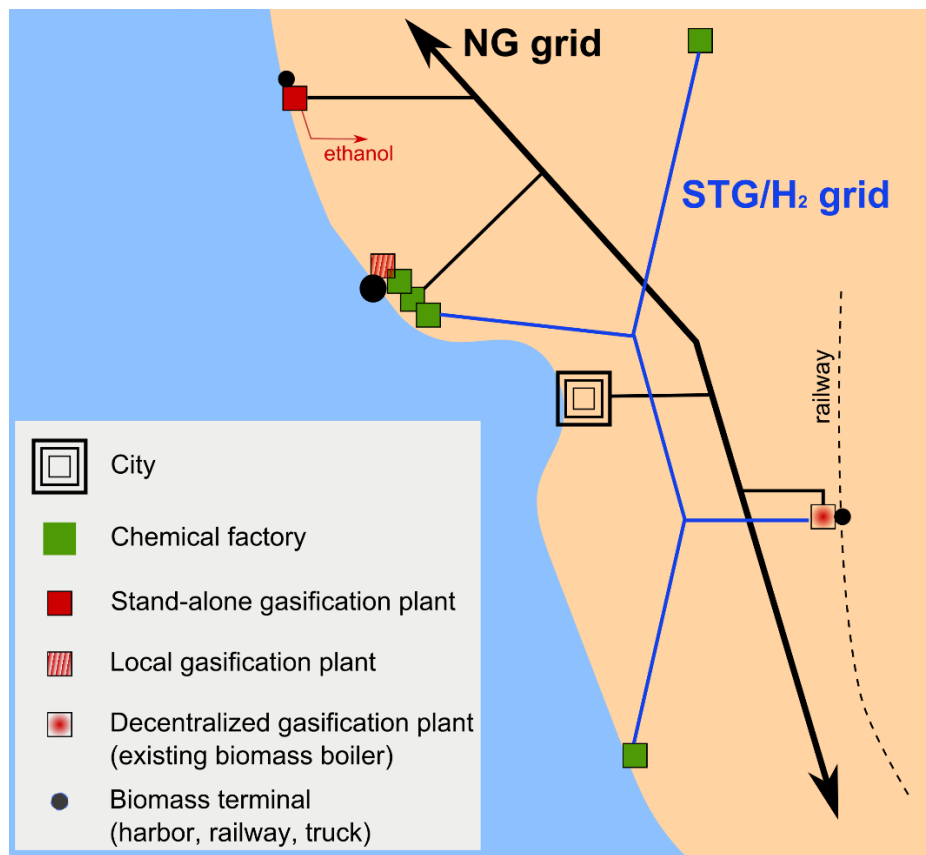
<b>Primary measures (not investigated)</b>	<ul style="list-style-type: none"> <li>• Advanced management of fines and alkali</li> <li>• Activation of bed materials</li> <li>• Use of additives</li> <li>• Improved reactor design</li> </ul>
<b>Secondary measures (investigated)</b>	<ul style="list-style-type: none"> <li>• Drying integrated into the plant</li> <li>• Enhanced pre-heating of inlets to the gasifier</li> <li>• Electricity production</li> <li>• Power-to-gas technologies</li> </ul>

The secondary measures focus on the management of the excess heat in the process and the production/use of electricity. A drying system can be installed to increase efficiency and fuel flexibility. The drying system should be integrated into the heat recovery network, which can include a steam cycle from the production of electricity (reducing the consumption of electricity from the grid). Furthermore, the pre-heating temperature of the air and steam delivered to the gasifier can be optimised. Another opportunity offered by gasification processes is the introduction of power-to-gas technologies to increase the production of biomethane. Power-to-gas technologies are of interest, since electricity can be added intermittently to a continuous process, enabling the conversion of intermittent renewable energy sources, such as wind or solar. Furthermore, the surplus of electricity generated from the excess heat in the process can be converted to methane (i.e., re-circulating energy in the process), thereby further increasing the efficiency from biomass to biofuels.

In the present work, two distinct power-to-gas technologies are investigated. The first relates to a traditionally proposed process based on the electrolysis of water, while the second look at the possibility of applying direct heating of the gasifier to reduce the combustion of char and re-circulated gases[60]. In the water electrolysis process, the produced hydrogen is injected after the WGS reactor, so as to adjust the  $H_2:CO$  ratio before methanation. Compared to standalone electrolysis processes the integration of water electrolysis in a biomethane plant is particularly favourable due to the fact that renewable CO and  $CO_2$  are already part of the syngas and that the methanation reactors are already in place. Furthermore, the pure oxygen that results from the electrolysis process can be used in the combustor to reduce the inlet air flow. Power-to-gas by direct heating of the DFB gasifier can be achieved by introducing resistance heaters in the DFB gasifier reactor or by further pre-heating the inlet gases[60]. The consequence is a reduction in the internal heat demand in the gasifier, decreasing the amount of char that needs to be combusted to fulfil the heat requirement, thereby making it available for syngas production *via* steam gasification. The main advantage of direct heating over electrolysis is the higher efficiency of the process, since almost all the electricity provided is stored as chemical energy in the gasification products. However, there is a stricter technical limitation linked to the direct heating option due to the maximum rate of char gasification achievable in the process.

## **1.8 Decentralization and introduction of new gasification processes**

Apart from biofuel production in *stand-alone* plants, gasification has a strong potential for using biomass in the production of bulk chemicals and in refinery processes. This requires the creation of a synergy between the chemical (and petrochemical) industry and the forest industries. Both these industrial sectors are associated with large production volumes with small margins, as well as large investments in process equipment and the development of expertise. Thus, their combined interest can contribute substantially to the decarbonisation of the chemical sector [69-71] and to the development of large-scale gasification facilities. The driving force lies in the added value for both industries without a requirement for extensive integration. Chemical industries benefit from renewable hydrocarbons for both emissions and marketing reasons, while the forest industry can increase considerably the value of their residues.



**Figure 6**— Schematic of the three different implementation strategies for biomass gasification plants: *stand-alone*; *local drop-in*; and *decentralised*.

The integration of the two processes is achieved within the gasification plant, where the biomass residues are converted to an intermediate chemical, which is then introduced into an existing value chain for fossil-based products [69]. The quality of the intermediate product and its position in the existing value chain have been investigated [69, 72, 73] in the recent years. In this thesis, the focus is shifted to the geographical location of the gasification plant, identifying two main strategies: *local drop-in*, being placed in proximity to the customers (i.e., a cluster of industries); and *decentralised*, being located at the biomass facility and supplying the intermediate chemical *via* a pipeline. Compared to the *stand-alone* biomethane process, both of the above-mentioned strategies have lower complexity and higher customer flexibility, which can increase the profitability of the plant. Figure 6 illustrates schematically the different geographical locations of the three types of plants.

*Local drop-in* plants have the advantage of synergy with the existing equipment for the synthesis of a chemical (e.g., methane reformers), thereby reducing the complexity of the gasification plant and increasing the flexibility towards customers' requirements. However, *local drop-in* plants require the construction of biomass handling and drying facilities that can handle large amounts of biomass, which comes with a considerable cost due to the low energy density and high moisture content of the fresh biomass [67, 74, 75]. Heat integration with an existing steam cycle is possible, and the gasification plant could benefit from excess heat from other processes. The reliability of the gasification plant could be an issue if

maintenance and un-planned shutdowns occur more frequently than in other chemical process plants.

The *decentralised* strategy, which disconnects the gasification plant from the final customers, sets a stricter limitation of the quality of the intermediate chemical. The main advantage is the flexibility of the geographical location. In particular, existing biomass boilers [74] and pulp mills [76] have the required infrastructure (biomass handling, dryers, steam cycles integrated into the heat recovery network) and can be retrofitted or up-graded to become highly efficient gasification plants. The main drawback of *decentralised* plants is the investment required for the construction of a connecting pipeline, although this cost can be counter-balanced by an increase in the number of customers by use in existing chemical processes. The complexity of the product synthesis section depends on the product quality and can be limited to a few cleaning steps, assuming that the customers can handle a gas composition that is similar to that of the product gas from gasification.

In this work, the qualities assumed for the intermediate chemical in *decentralized* and *local drop-in* plants are investigated, to visualize the possibilities associated with the concept. The main issue is to look at products that are different from pure methane. The rationales for this is that for most applications in chemical factories (except heat and electricity generation), methane first needs to be reformed to syngas, which increases the complexity and reduces the chemical efficiency of the system. Thus, the methane that is generated during gasification is, for these processes, a drawback rather than a benefit. Furthermore, the reforming of renewable methane is questionable, since it is a valuable product on the biofuel market. Therefore, two extreme cases are taken in consideration in this work: 1) an intermediate that contains methane, which is supplied to industries that are already equipped with a natural gas reformer or have equipment that can separate the methane before use; and 2) a methane-free intermediate obtained *via* separation during the biomethane process (see Fig. 6, and Paper VI).





# 2

## Methodology

The investigations of the different aspects of the production and use of biomethane in heavy duty engines, and the further development of the gasification-base process require the application of different methods. In particular, three important areas are looked at: 1) evaluation of the WtW chain (Papers I and II); 2) analysis of the experimental data and simulation of the DFB gasifier (Papers IV and V); and 3) evaluation of potential improvements to biomethane production (Papers III and VI). The combination of these results highlights not only the strengths and weaknesses of the current WtW chain for biomethane, but also possibilities for future developments.

The analysis of the existing WtW chain (Paper I) focuses on the use of biomass resources and the issue of GHG 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 II investigates the quality of the biomethane used for combustion in DF engines, which is the technology that is most sensitive to fuel composition. The aim was to identify the boundaries of the biomethane composition within which the WtW efficiency could be improved.

Paper IV proposes a method for the analysis of experimental data and modelling of the DFB gasifier. The proposed method combines pre-treatment of the experimental data with implementation in a flow-sheet software, such as Aspen Plus. In Paper V, the model developed in Paper IV is applied to the evaluation of the GoBiGas process. The result is a comprehensive evaluation of the gasification section using data from the first experimental campaign conducted at the plant. An algorithm for the simulation of the gasifier is proposed that enables the evaluation of possible secondary measures for improving the process (e.g., drying, pre-heating, reduction of heat losses and utilisation of additives in the reactor). Furthermore, the concept of power-to-gas conversion *via* direct heating of the gasifier is introduced and assessed.

Overall, Papers III, V, and VI investigate possible secondary measures (Table 3) for improvement of the biomethane process, as well as alternative strategies for the introduction of gasification processes into the existing energy system. In particular, Paper III introduces a concept for the design of a steam dryer. Paper VI focuses on the flow-sheet modelling of gasification-based processes on a large-scale, including the biomethane process and other possibilities for *decentralised* and *local drop-in* plants. The flow-sheets make use of the models developed previously (in Papers III, IV, and V) to simulate the

DFB gasifier and the integrated steam dryer. The simulations assess the efficiencies and production ranges of the investigated processes.

## 2.1 WtW analysis and utilization of biomass resources

WtW analysis is a well-established method for the estimation of emission intensities and the utilisation of primary energy resources for transportation fuels. Compared to a Life Cycle Assessment (LCA), a WtW analysis has the same system boundaries but does not include the consumption of materials and water, other pollutants or end-of-life disposal. WtW requires less-extensive datasets, which is an advantage when assessing developing technologies. In the present WtW analysis, biomass feedstock and renewable fuels are considered to be emission-neutral when they are combusted, which means that direct and indirect Land Use Changes (LUC and iLUC, respectively) are not considered, although for total emissions, the contributions from methane slips from the engine and methane leakages along the WtW chain are included. The WtW analysis is divided into: (i) the well-to-tank (WtT) part, which accounts for the energy expended and the associated emissions to deliver the finished fuel in the fuel tank; and (ii) the tank-to-wheel (TtW) part, which includes the final conversion of the fuel in the engine. For biomethane, the WtT part focuses on the production and distribution in compressed (bio-CNG) or liquid (bio-LNG) form, while the TtW part of the chain is concerned with the engine technologies. Each different combination of WtT and TtW is referred to as a WtW pathway.

The indicators used in the WtW analysis are the total WtW energy and the WtW GHG emissions, in similarity to the approach adopted in the JEC study [5]. However, results based exclusively on emission intensities are not considered to be sufficient for comparing biomass-based pathways. A third indicator is introduced, called the *biomass impact* (BI), which expresses the WtW emissions reduction from the diesel pathway per MJ of biomass [gCO<sub>2</sub>e/MJ<sub>biomass</sub>]. The biomass impact is calculated [Eq. (2)] as the ratio of the GHG WtW emissions reduction,  $\Delta\text{GHG}$  [Eq. (1)], to the specific biomass consumption  $sbc$  [gCO<sub>2</sub>e/MJ<sub>out</sub>], for each biomethane pathway. A comparison based only on the WtW GHG emissions and total WtW energy risks overestimating the importance of biomethane pathways with low GHG emissions but high specific consumption of biomass. Instead, the BI enables a comparison of biomethane pathways based on their positive effects on the emissions per unit of biomass, based on a single parameter.

$$\Delta\text{GHG} \left[ \frac{\text{gCO}_2\text{e}}{\text{MJ}_{\text{out}}} \right] = \text{GHG}_{\text{diesel}} \left[ \frac{\text{gCO}_2\text{e}}{\text{MJ}_{\text{out}}} \right] - \text{GHG}_{\text{biometh}} \left[ \frac{\text{gCO}_2\text{e}}{\text{MJ}_{\text{out}}} \right] \quad (1)$$

$$\text{BI} \left[ \frac{\text{gCO}_2\text{e}}{\text{MJ}_{\text{biom}}} \right] = \frac{\Delta\text{GHG} \left[ \frac{\text{gCO}_2\text{e}}{\text{MJ}_{\text{out}}} \right]}{sbc \left[ \frac{\text{MJ}_{\text{biom}}}{\text{MJ}_{\text{out}}} \right]} \quad (2)$$

The following assumptions form the basis for this approach: 1) biomass is considered CO<sub>2</sub>-neutral; 2) in the medium-to-long term, a mix of renewable- and fossil-derived energies will be present in the market or in the fuel blend or in the combustion process (e.g., DF engines) [18] ; and 3) the future availability of biofuels will be limited by the amount of sustainably grown biomass. Under these assumptions, it is clear that the biomass resources should be employed in those WtW pathways that achieve the maximal reduction in emissions.

**Table 4** – overall scheme of the WtW analysis

<i>CNG</i>	<i>Biomass</i>		<i>LNG</i>	<i>WtT</i>	<i>WtW</i>
<ul style="list-style-type: none"> <li>• Extraction</li> <li>• Compression (80 bar)</li> <li>• Transport via pipeline</li> </ul>	<ul style="list-style-type: none"> <li>• Pellet production</li> <li>• Biomethane production</li> </ul>	<ul style="list-style-type: none"> <li>• Compression (40 bar)</li> </ul>	<ul style="list-style-type: none"> <li>• Extraction &amp; shipping</li> <li>• Terminal operations</li> </ul>		
	<i>Bio-CNG</i>	<i>Bio-LNG</i>			
<ul style="list-style-type: none"> <li>• Injection into a local network (40 bar)</li> <li>• Compression (250 bar) and refuelling operations</li> </ul>		<ul style="list-style-type: none"> <li>• Transport by truck</li> <li>• Refuelling operations</li> </ul>			
<ul style="list-style-type: none"> <li>• Final conversion in the engines               <ul style="list-style-type: none"> <li>- Spark-Ignited and Dual Fuel (CNG, bio-CNG, LNG, bio-LNG)</li> <li>- High-Pressure Direct Injection (LNG, bio-LNG)</li> </ul> </li> </ul>				<i>TtW</i>	

The boundaries of the WtW analysis are presented in Table 4, which includes ten different pathways: five based on biomethane (bio-LNG and bio-CNG); and five based on natural gas (LNG and CNG). The diesel pathways used for comparison are not included in Table 4. A sensitivity analysis of the results was carried out to elucidate the effects of engine efficiency, the diesel fraction in the fuel blend, the CO<sub>2</sub> emissions from the European and Nordic electricity mix, and other factors.

## 2.2 Sustainability, emissions and feedstock

The present study focuses on the production of biomethane through thermochemical conversion of lignocellulose biomass, assuming that the burning of biomass is carbon-neutral. This is indisputable 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 has been previously sequestered from the atmosphere during 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 for bioenergy. In the present study, is assumed that no LCU emissions are associated with the biomass feedstock, which is a reasonable assumption to make for Sweden, where the growing biofuel industry compensates for the reduced demand in the pulp and paper sector. At the present moment, the feedstocks allowed in the biomethane process are wood pellets and wood chips, mostly derived from wood processing. However, future developments of the gasification technology will enable the utilisation 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. Previous studies [77] have revealed that the harvesting of felling residues leads to an initial decrease in the carbon stock of the soil, which stabilises over time despite the increased output of biomass. 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 are accounted for in the total WtW emissions. This category includes methane leakages along the chain and combustion products that are not  $\text{CO}_2$  or  $\text{H}_2\text{O}$  (i.e., methane), other unconverted hydrocarbons, and  $\text{NO}_x$ .

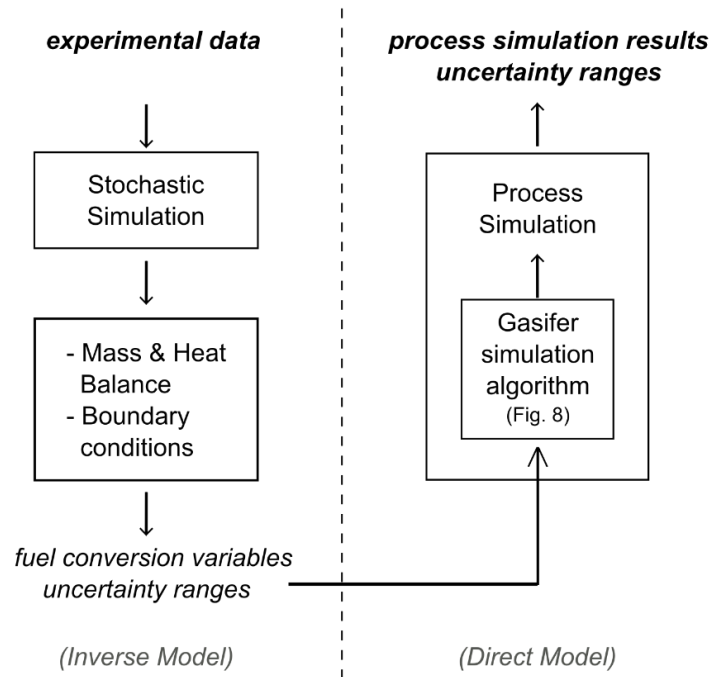
## **2.3 Pre-design of the steam belt dryer concept**

The viability of the proposed dryer design is evaluated using a multi-scale approach. Information about the drying process is obtained from a CFD analysis of a two-dimensional cut of the steam flow through the packed bed of wood particles, in which the evolution of the drying front inside an individual particle is modelled using a particle sub-model. The evaporation temperature of the water in the sub-model was set according to the results of experimental investigations. In a thermogravimetric analysis (TGA), the normalised weight losses and temperatures of 19 samples of wood chips were recorded during temperature-programmed evaporation. The results of the CFD simulations were used in the macroscopic mass and heat balance (Aspen Plus) to calculate the energy consumption levels and the energy and exergy efficiencies of the dryer. Furthermore, a pre-design of the geometry was calculated.

## **2.4 Stochastic analysis of experimental data from a DFB gasifier**

The performance of the gasifier determines the efficiency, the complexity, and ultimately, the overall feasibility of the process. Therefore, any modelling that acts to optimise biofuel production 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 [42, 78-82], semi-detailed kinetics mechanisms [42, 78-82], and experimental data from existing plants [74, 83-85]. The approach developed here is based on the analysis of the experimental data from pilot and demonstration gasifiers, so as to input these data into the process simulation, optimisation analysis, and techno-economic analyses [86, 87].

The goal is to overcome the problems faced by the other modelling approaches. In particular, thermodynamic equilibrium models produce substantial deviations from the measurements, especially with regard to the yields of methane and tar, as well the rate of carbon conversion. Kinetic models have higher levels of accuracy, assuming that the coefficients used in the reaction mechanism are correctly estimated [78]. However, a comprehensive model that includes all the chemical and physical interactions requires extensive knowledge of the process, which is currently not available.



**Figure 7** – Schematic showing utilization of the data analysis and simulation of the DFB gasifier.

The two main issues related to the use of experimental data in simulations are: (i) the estimation of the uncertainty linked to the measurements; and (ii) the simulation of the mass and energy balances of the gasifier under different operating conditions (e.g., pre-heating, reactor size, moisture content of the fuel). Therefore, the modelling approach consists of two phases (Fig. 7): 1) a stochastic analysis of the experimental data, to calculate the fuel conversion variables and the associated uncertainty (inverse model); and 2) an algorithm for the simulation of the mass and heat balances of the gasifier in flow-sheet models (direct model; see Section 2.5).

The analysis of the experimental data aims to calculate a set of variables that describes the fuel conversion in the gasifier: the char gasification ( $X_g$ ), the oxygen transport ( $\lambda_{Otr}$ ), and the fraction of volatiles converted to the various energy-carrying compounds in the raw gas ( $Z_i$ ). The uncertainty linked to the fuel conversion variables depends on the intrinsic uncertainties of the measurements and on the type of measurement set-up (with possible undetected species and an unclosed mass balance). The equation describing the mass balance has a different degree of freedom depending on the measurements available (Paper IV). In particular, the mass balance of the set-ups that measure only the concentrations (and not the yields) of the raw gas compounds has  $\geq 2$  independent variables, leading to high uncertainty. Instead, set-ups that measure the yields of raw gas compounds have a unique solution if the mass balance is closed (e.g., with total carbon measurements), and the mass balance has two independent variables ( $X_g$  and  $\lambda_{Otr}$ ) if there are undetected species (unclosed mass balance).

The value of the fuel conversion variable and its related total uncertainty are calculated using a stochastic simulation of the experimental inputs (measurements of PG, tar, char yield, fuel composition, fuel feed, etc.) to generate more than  $10^5$  variations. For each variation of the inputs, the valid domain of the independent variables (e.g.,  $X_g$  and  $\lambda_{Otr}$ ) is

calculated, thereby solving the mass balance and retaining those solutions that respect the boundary conditions for: 1) the compositions of undetected compounds; 2) the fraction of carbon in the raw gas detected by the measurements ( $fCd$ ); and 3) complete devolatilisation (see Paper IV, Section 3.4). The mean values and standard deviations of the total pool of solutions are calculated, corresponding to the expected fuel conversion variables and their levels of uncertainty.

## 2.5 Simulation of the DFB gasifier

An algorithm was developed to simulate the DFB gasifier using the set of fuel conversion variables, previously calculated from the analysis of the experimental data. One advantage of using this approach is that the heat balance can be extrapolated for different conditions [60, 88]. The method enables to transfer experimental knowledge from a smaller facilities to a larger plant, which can differ in heat losses, pre-heat of ingoing streams, moisture content of the feedstock and other parameters, affecting the efficiency of the process.

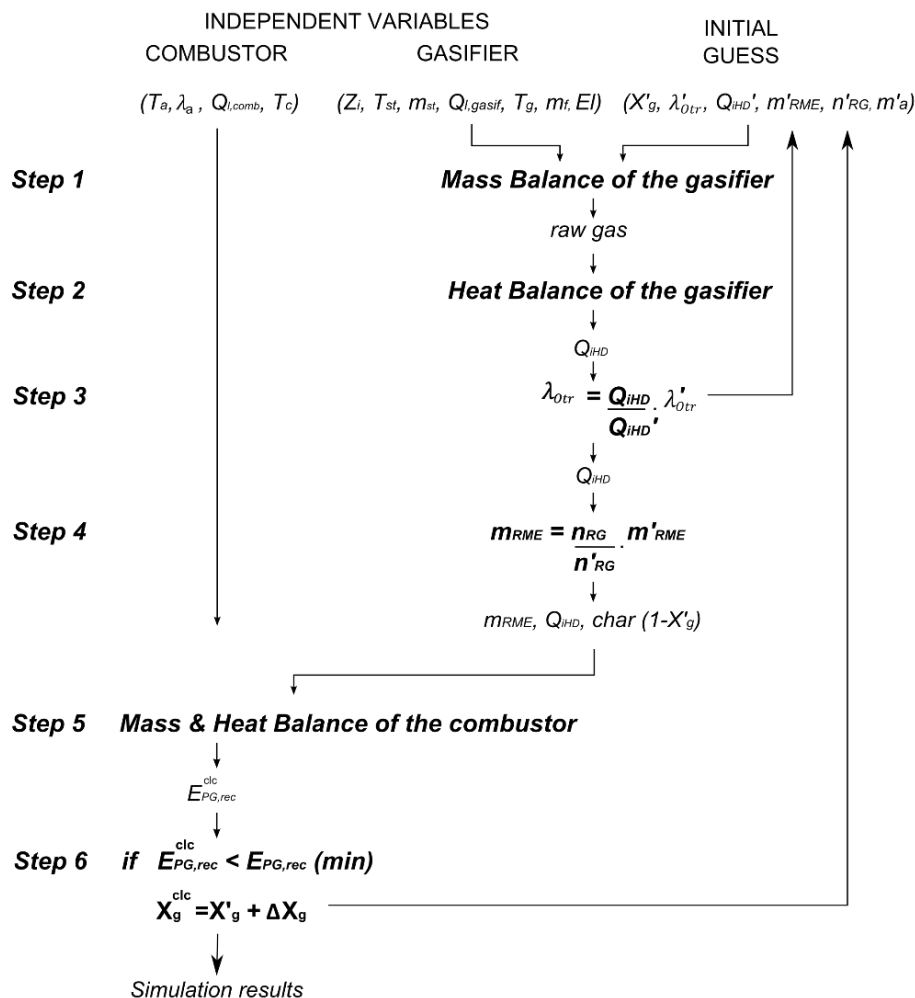


Figure 8 – Simulation algorithm for the DFB gasifier (mass and heat balance).

The simulation algorithm is presented in Fig. 8 and it is based on four main assumptions : 1) the circulation of bed material and the oxygen transport are linearly proportional to the internal heat demand of the gasifier (i.e., the oxidation level of the bed material from the combustor is equal for all cases); 2) the RME flow is linearly proportional to the mass flow of the wet raw gas; 3) the average re-circulated flow gas should at a minimum be 1% of the fuel input, to cope with process fluctuations, such as variations in the moisture content; 4) the char gasification can be varied within a valid interval (40% - 70%) to cope with variations in the heat balance; 5) the gas re-circulation is reduced to the minimum before increasing the char gasification.

Each simulation is defined by a set of independent variables and requires a set of initial values. The starting values are initially guessed and thereafter re-calculated through two iterative calculations, one linked to the mass and energy balances of the gasifier to derive the heat demand of the gasifier and  $\lambda_{Otr}$  (Step 3), and one linked to the mass and energy balances of the entire system to derive  $X_g$  (Step 6). To simulate a different chemistry in the reactor (e.g. due to additives), the  $Z_i$  values are recalculated based on the measured composition of the product gas (using the mass and heat balance equations in Paper IV). Variations of the heat balance are addressed in such a way that a decrease in internal heat demand is balanced by reduction of the re-circulation of the raw gas to the combustor to the minimum level. Beyond this point any further reduction of the heat demand can be compensated by a decrease in char combustion making more char available for gasification. The algorithm is also used for the simulation of direct heating of the DFB gasifier by electricity, as power-to-gas process. The electricity can be introduced in the simulations both as direct heating of the gasifier or combustor, or as further pre-heating of the steam and air.

## 2.6 Flow-sheet modelling of gasification-based processes

The investigation of gasification-based processes is performed using flow-sheet simulations of different plant designs. For a *stand-alone* plant, the focus is on evaluation at the commercial scale, with the introduction of a series of proposed improvements to the process. Designs for *decentralised* and *local drop-in* plants are analysed and compared to those for *stand-alone* plants, with the focus being on the achievable efficiencies and range of products. The investigated process designs are divided in four classes and presented in Table 5. The classification is based on the final product of the process. Designs of class A produce biomethane, class B designs produce methane and ethanol, class C designs produce methane and/or STG, and class D designs produce biomethane and hydrogen. The process analysis includes gasification and gas synthesis, as well as the steam cycle used for heat recovery and electricity production, which is not included in the current GoBiGas process. All the designs are investigated at a size of 100 MW<sub>biomass</sub> and the feedstock is 40% moisture w.b. A separate flow-sheet for the GoBiGas plant (32 MW<sub>biomass</sub>) was developed for validation of the model.

For a *stand-alone* plant, several aspects are evaluated to improve the efficiency over that of the current GoBiGas design (design A.1). Two drying systems are investigated: a single-stage air dryer that reduces the moisture content to 20% w.b. (design A.2); and a double-drying system with the steam dryer recovering the moisture as the gasification agent, as presented in Paper 3 (design A.3). Other aspects investigated are the possibility to introduce

power-to-gas technologies (electrolysis and direct heating [60]) and the co-production of methane and ethanol. Co-production of methane and ethanol is investigated *via* a biochemical pathway (syngas fermentation), which compared to catalytic synthesis, retains high efficiency, tolerates sulphur-containing impurities in the syngas, and is less effected by inert gases, such as methane [89, 90].

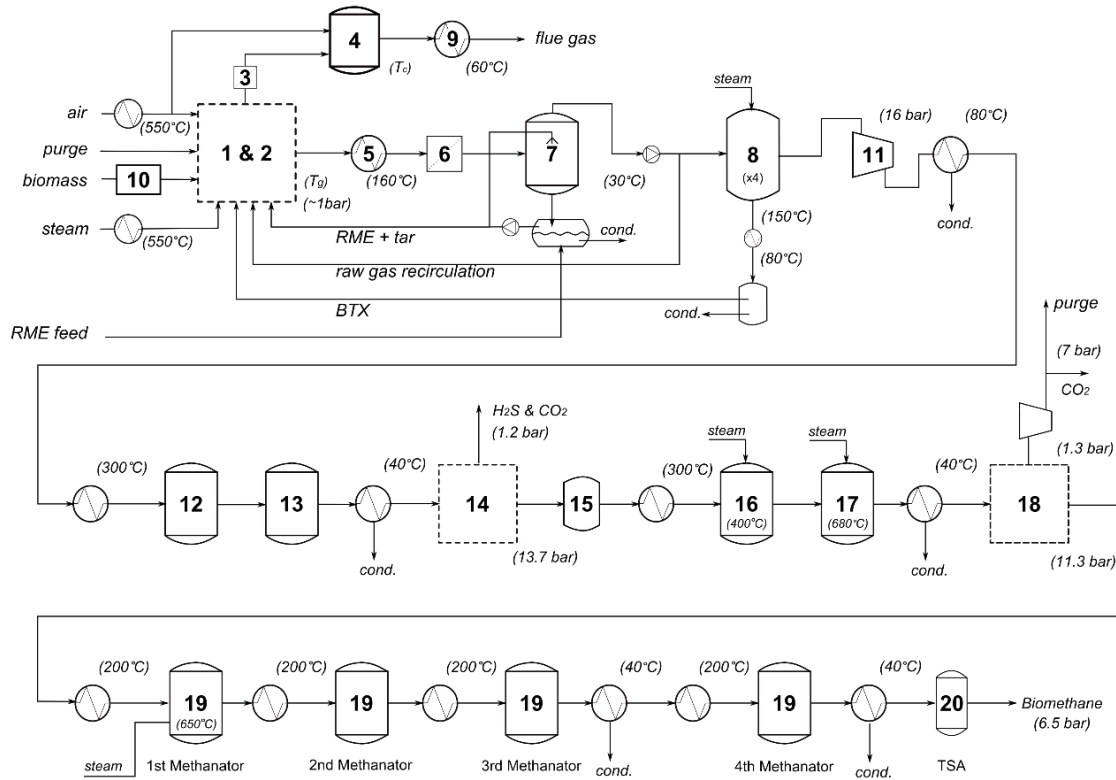
**Table 5:** Designs investigated, based on the final products

Designs	Product 1	Product 2	Strategy	Networks	Power-to-gas	Drying
A.1	Biomethane	-	standalone	NG, El	no	no
- A.2	Biomethane	-	standalone	NG, El	no	single-stage <sup>1</sup>
- A.3	Biomethane	-	standalone	NG, El	no	double-stage <sup>2</sup>
- A.4	Biomethane	-	standalone	NG, El	electrolysis	double-stage <sup>2</sup>
- A.5	Biomethane	-	standalone	NG, El	direct heating	double-stage <sup>2</sup>
B	Biomethane	Ethanol	standalone	NG, El, ethanol	no	double-stage <sup>2</sup>
C	STG	Biomethane*	local/ decentralised	NG*, El, STG,	no	double-stage <sup>2</sup>
D	Hydrogen	Biomethane	local/ decentralised	NG, El, hydrogen	no	double-stage <sup>2</sup>

\*Optional, <sup>1</sup> Air drying, <sup>2</sup> air and steam drying with moisture recovery as the gasification agent

For *local drop-in* and *decentralised* plants, two possible nitrogen-free intermediate products are considered: a sustainable town gas (STG), i.e., upgraded syngas from the biomass gasification; and hydrogen. STG is suitable for industries that are interested in replacing fossil carbon sources (especially natural gas) with renewable carbon. Hydrogen is considered to be an option that is more suited to refinery processes, which have high hydrogen demands for desulphurisation of their products and for ammonia production. Both of the intermediate products can be produced in *decentralised* and *local drop-in* plants, although STG is considered more suitable for decentralised production, due to a lower energy demand for compression. The hydrogen is co-produced with biomethane to avoid the reforming of the methane production from gasification, and to increase market flexibility. Hydrogen is separated from the syngas in the biomethane process *via* Vacuum Pressure Swing Adsorption (VPSA).





**Figure 9** - Process flow-sheet of the GoBiGas design at 100 MW<sub>biomass</sub>. Key to symbols: 1, gasifier (separate DFB sub-model [88]); 2, combustor (separate DFB sub-model); 3, cyclone; 4, post-combustion chamber; 5, raw gas cooler; 6, raw gas filter; 7, RME scrubber; 8, carbon beds; 9, flue gas train; 10, fuel feeding system; 11, product gas compressor; 12, olefins hydrogenator; 13, COS hydrolyser; 14, H<sub>2</sub>S removal (separate sub-model) ; 15, guard bed; 16, water-gas shift reactor; 17, pre-methanation; 18, CO<sub>2</sub> removal (separate sub-model) ; 19, methanation; 20, TSA drying.

The flow-sheet simulation is performed in Aspen Plus, and an example of the modelling scheme is given in Fig. 9 (for design A.1). Some of the components are simulated through dedicated sub-models, such as the DBG gasifier (nr. 1 and nr. 2) and the absorption processes (nr. 14 and nr. 18) and dryers (based on Paper III, and not reported in Fig. 9). In particular, the sub-model of the gasifier is built based on Paper IV, where the fuel conversion variables are obtained from the analysis of the experimental data from the GoBiGas plant, and the new operational parameters are calculated with the simulation algorithm proposed in Paper V.

## 2.7 Process indicators

The evaluations of the gasification section and the biomethane process require a definition of the different efficiencies. Table 6 reports the definition used in this thesis, where all the efficiencies are calculated using the lower heating value of dry ash-free biomass (LHV<sub>daf</sub>). Nevertheless, in Papers V and VI, the results are also presented based on the LHV of 50% moisture biomass. Regarding the gasification section, a distinction is drawn between raw gas efficiency ( $\eta_{RG}$ ) and cold gas efficiency ( $\eta_{CG}$ ), in that the former is considered to be the gas exiting the gasification reactor (including tar), while the latter represents only the gas leaving the gasification section (net of removed tar and gas re-circulation). The raw gas efficiency, which assesses the fuel conversion in the gasifier, can reach ~110% if all of the

char is gasified (endothermic reaction), with secondary fuels (e.g., RME) and/or electricity being fed to the combustor. Instead, the cold gas efficiency assesses the conversion of biomass in the entire DFB gasifier, taking into account the tar production and the required re-circulation of raw gas. Other energy inputs are present in the gasification section, namely the RME stream (which is subsequently combusted) and the electricity for power-to-gas conversion through direct heating. The total efficiency of the gasification section is assessed as  $\eta_{sect}$ , which is based on the cold gas efficiency but includes all the energy inputs.

The evaluation of the biomethane process is based on the chemical efficiency  $\eta_{ch}$ , and the total efficiency  $\eta_{tot}$ . The chemical efficiency is based on the sole biomass inputs and includes all the chemical products (biomethane, hydrogen, STG, and ethanol). The total electricity parameter is based on all the energy inputs and includes the production of electricity in the products. Regarding electricity, it must be pointed out that the process can be either a consumer ( $El_{in}$ ) or a producer ( $El_{out}$ ). The power-to-gas process is assessed using the marginal power-to-gas efficiency  $\eta_{p2g}$ . This value is calculated as the increment in biomethane production and the net variation in electricity consumption of the plant (Paper VI). The power-to-gas efficiency is strongly dependent upon the reference process, and it is appropriate to evaluate the introduction of power-to-gas technologies in an existing process. For an assessment of power-to-gas conversion in absolute terms, it is preferable to refer to the total efficiency (Paper V)

**Table 6** – definition of the efficiencies based on the LHV of dry ash-free biomass

	Based on biomass input	Based on all energy inputs
Raw gas efficiency	$\eta_{RG} = \frac{E_{RG}}{E_{biom}}$	
Cold gas efficiency	$\eta_{CG} = \frac{E_{CG}}{E_{biom}}$	
Gasification section efficiency		$\eta_{sect} = \frac{E_{CG}}{E_{biom} + E_{RME} + El_{sect}}$
Chemical efficiency	$\eta_{ch} = \frac{E_{CH_4} + E_{STG} + E_{H_2} + E_{other}}{E_{biom}}$	
Total efficiency		$\eta_{tot} = \frac{E_{CH_4} + E_{STG} + E_{H_2} + E_{ethan} + El_{out}}{E_{biom} + El_{in} + E_{RME}}$
Power-to-gas efficiency		$\eta_{P2G} = \frac{E_{CH_4} - E'_{CH_4}}{El_{in} - E'_{out}}$

*E* energy terms, RG raw gas, CG cold gas (product gas), 'reference process.

# 3

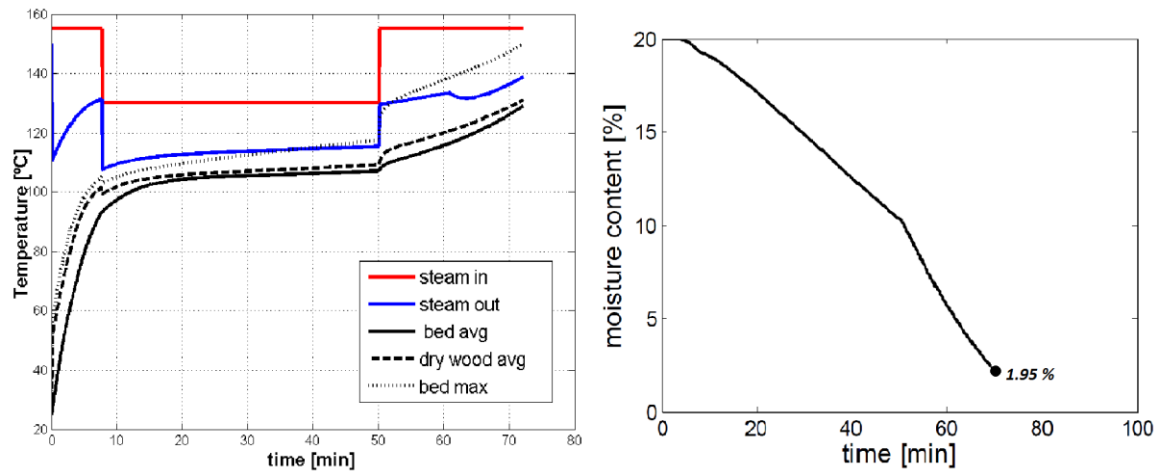
## Results and discussion

The investigations carried on during the work for this thesis covers several aspects of biomethane production and use in heavy-duty engines. Initially, these aspects are presented Paper III together with the pre-design of the steam dryer, as this concept is used subsequently in the evaluation of large-scale gasification processes. Thereafter, the results of the stochastic analysis of the experimental data from the Chalmers gasifier are introduced, as well as a comparison of the different measurement set-ups, including total carbon measurements for validation (Paper IV). The results obtained for the mass and heat balances of the gasification section of the GoBiGas plant are presented together with an evaluation of potential measures to improve the efficiency of the plant (Paper V). The focus is then shifted to the process level, with an investigation of the designs for *stand-alone*, *decentralised*, and *local drop-in* plants (Paper VI). In conclusion, are presented the results of the WtW study calculated with biomethane production optimized at large-scale plants (not present in paper I) and compared to those of biomethane produced at.

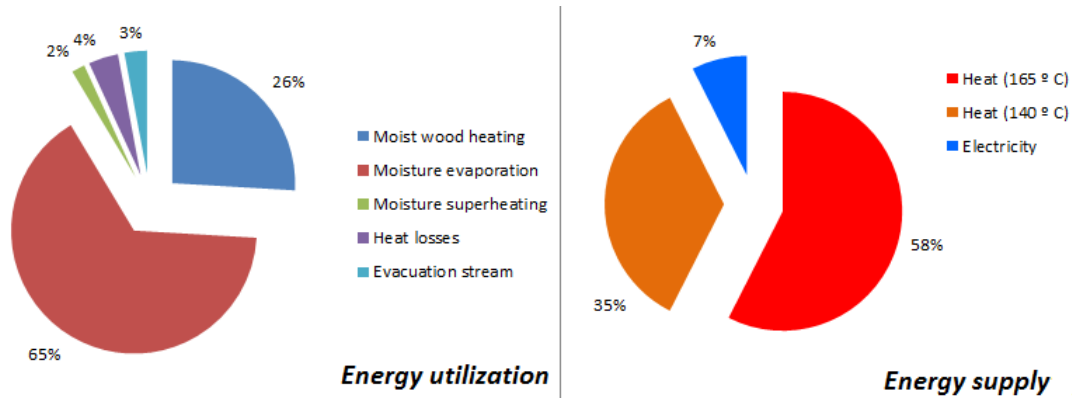
### 3.1 Steam belt dryer

The proposed belt dryer is composed of three sections that use steam at different temperatures. The CFD analysis shows that a higher temperature is required in the first and last sections to avoid condensation of the steam onto the biomass and to achieve a very low moisture content. Instead, most of the drying occurs in the intermediate section with a lower temperature of the drying agent. The results of the CFD simulation show that a steam temperature of 155°C is sufficient in the first and last sections, and that intermediate section can be operated with steam at 120°C. Figure 10 reports the temperature profiles along the dryer (left panel) and the moisture contents of the biomass (right panel).

The energy balance of the drying system is reported in Fig. 11 and includes the electricity required for steam circulation and heat losses. The heat is provided to the process in the exchanger up-stream of the dryer, at temperatures of between 140°C and 165°C. The electricity accounts for 7% of the total energy input. This heat is recovered in the system through re-use of the moisture as gasification steam and direct feeding of the biomass into the gasifier (i.e., pre-heating of the biomass). The calculated energy efficiency of the dryer is 95%, since the only energy loss is heat losses to the surroundings.



**Figure 10** – Temperature and moisture profiles in the proposed steam dryer.

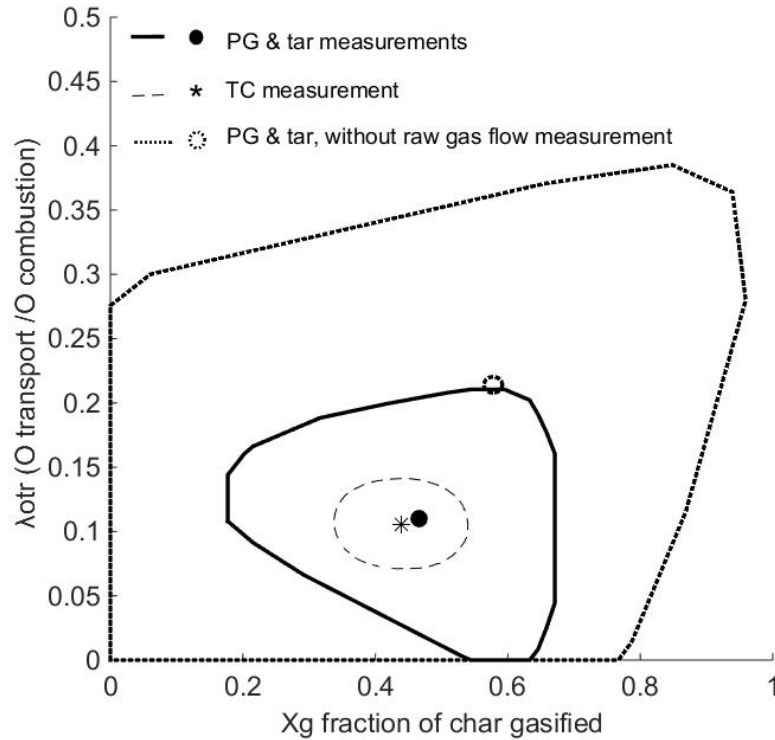


**Figure 11** - Energy balance of the steam dryer.

An exergy analysis of the drying process was performed for Paper III, showing 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 [91-93]. This 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.

### 3.2 Analysis of the Chalmers gasifier measurements

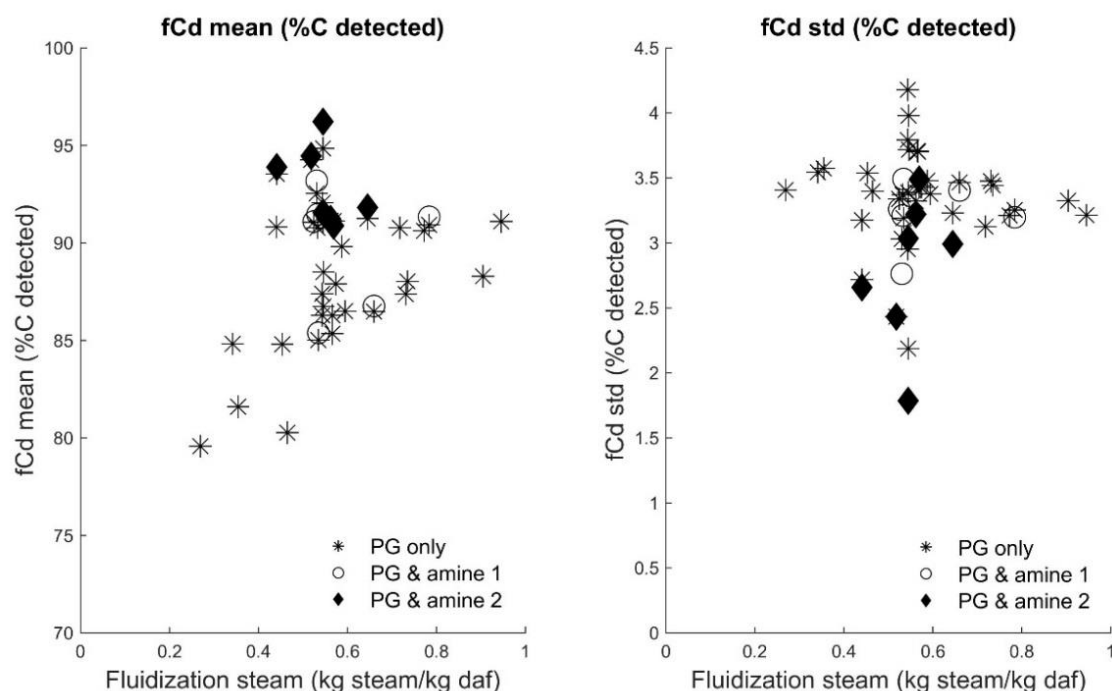
The results of the stochastic method applied to a measurement point from the Chalmers gasifier are reported in Fig. 12. The same point was measured with different techniques, including measurements of: total carbon (TC); permanent gases (PG) and tar with total flow; and permanent gases and tar without total flow. The mean values (dots) and uncertainty levels (lines indicating the 95% confidence intervals) related to the measurement technique are reported in the solution domain on the  $X_g, \lambda_{otr}$  plane. 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 (indicated by asterisk and dashed line), which has a considerably smaller degree of uncertainty.



**Figure 12** – Solution of the mass balance for one operational point, comparing three measurements setups (Chalmers gasifier).

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 analyse incomplete measurement data. The largest solution domain (indicated by 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 for 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.

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). Figure 13 compares three measurement set-ups based on the analysis of 37 experimental points at different gasification temperatures ( $T_g$  790°C–830°C) and fluidisations ( $\mu_{st}$  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 a 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 a measurement system without total carbon measurements.



**Figure 13** - Uncertainty due to the measurement setup (Chalmers gasifier, silica sand and wood pellets).

### 3.3 Evaluation of gasification section at GoBiGas

The stochastic analysis of the measurements was used to evaluate the mass and energy balances in the GoBiGas gasification section, using the data from the first measurement campaign (Paper V). The results, which are reported in Tables 7 and 8, refer to operation of the gasifier using wood pellets (8% moist w.b.) as the fuel, with gasification temperature of 870°C, and potassium-activated olivine as the bed material. The rate of char gasification is calculated as 53.8% with a standard deviation (SD) of 4.7 percentage points (pp), and the oxygen transport,  $\lambda_{otr}$ , is estimated as 4.9% (SD, 2.7 pp) of the volume of oxygen required for stoichiometric combustion of the fuel. Calculation of the conversion of volatiles shows that 34.1% of the volatile matter is directly converted to methane, which is favourable for the downstream synthetic processes. The percentage of volatiles converted to tar is 3.5%, and 5.8% is converted to BTX, such that in total 9.3% of the volatiles form unwanted hydrocarbons.

The heat losses to the surroundings correspond to 5.2% of the energy in the fuel, or about 1.6 MW, of which 0.4 MW are from the gasification side and 1.2 MW are from the combustion side. Compared to the heat lost in a typical circulating fluidised bed combustor, i.e., 1–2%LHV<sub>daf</sub>, the heat lost in the GoBiGas system is considerably higher, due to the simple system of insulation of the reactors (see Paper V). The high losses of heat affect the energy balance between the two reactors, requiring a high level of re-circulation of the raw gas,  $E_{rec,PG}$ , to maintain the temperature of the process, corresponding to 9.8% of the fuel LHV on a dry basis. The total heat demand of the GoBiGas gasifier is 18% of the energy of the ingoing fuel, whereby about half of the heat demand is covered by the re-circulated gas. The raw gas efficiency of the gasifier is calculated as 87.3% LHV<sub>daf</sub> (SD, 1.9 pp), with 71.7%LHV<sub>daf</sub> (SD, 1.8 pp) of the energy in the fuel being converted to permanent gases

and delivered to the methanation section i.e., ‘cold gas efficiency’. Including the energy input from the RME, the efficiency of the gasification section is 69.2%  $\eta_{\text{sect}}$  (SD, 1.6 pp).

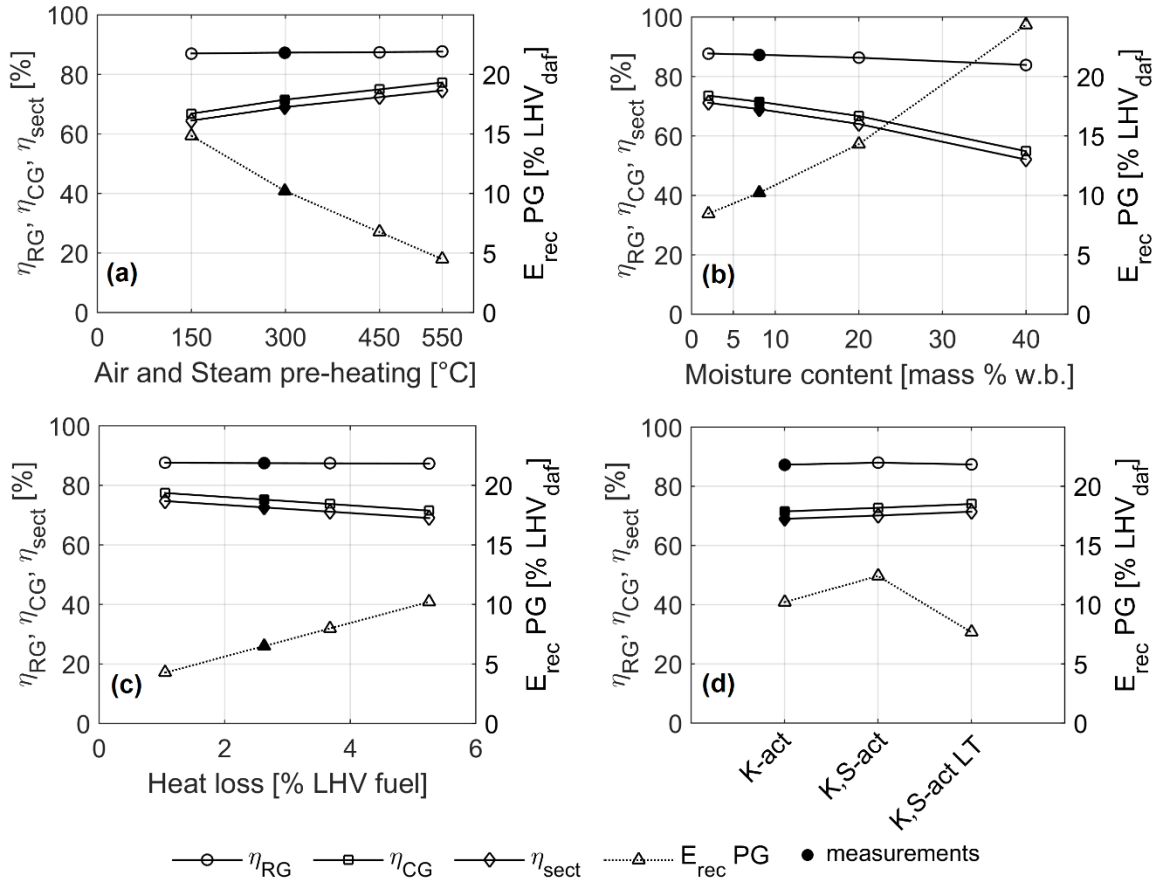
The sensitivity analysis of the performance of the GoBiGas gasifier aims to identify measures that could be used to improve the efficiency of DFB gasifiers using the GoBiGas gasifier as a reference. For this purpose, the air and steam pre-heating, the moisture content of the fuel, the heat losses of the system, the use of sulphur as an additive, and the introduction of electricity as a heat source were varied, as described in Section 2.5, and the results are presented in Fig. 14 and 15. The results are expressed as the raw gas efficiency  $\eta_{\text{RG}}$ , cold gas efficiency  $\eta_{\text{CG}}$ , gasification section efficiency  $\eta_{\text{sect}}$ , and product gas; the filled markers indicate the relevant reference points from GoBiGas (*K-act* case). Since all of these measures influence the heat demand in the boiler, they affect the required re-circulation of the raw gas, as well as the efficiency of the gasification section. Note that as soon as the level of re-circulated raw gas reaches the defined minimum, char gasification is increased to fulfil the heat balance, as described in Section 2.5, and this in turn increases the raw gas efficiency.

Since the GoBiGas plant requires a high level of re-circulation of the raw gas, owing to the considerable heat losses, most of the measures analysed affect only the re-circulation. Therefore, the only situation in which it is possible to derive a benefit from the significantly increased char gasification is when there is extensive introduction of electricity into the system. Pre-heating the air and steam from 300°C to 550°C (Fig. 14a) reduces the re-circulation of the raw gas to about 50% of the reference case, thereby increasing the cold gas efficiency from 71.7%LHV<sub>daf</sub> to 77.3%LHV<sub>daf</sub>. The reduction of heat losses has an effect similar to that of pre-heating, although the heat losses would need to be reduced by a factor of five to increase the  $\eta_{\text{CG}}$  to 77.4%LHV<sub>daf</sub> (Fig. 14c). The moisture content depends on the fuel that is being used and the drying process, which are dictated by the economics of the plant, considering both the operational and investment costs.

**Table 7** – Solution of the mass and energy balance of the GoBiGas gasifier, with dried biomass (8% moist).

<i>Mass balance</i>				<i>Energy balance</i>			
		<i>mean</i>	<i>SD</i>			<i>mean</i>	<i>SD</i>
$X_g$	[% mass]	53.8	4.7	$\eta_{\text{RG}}$	[%LHV <sub>daf</sub> ]	87.3	1.9
$\lambda_{\text{otr}}$	[% mass]	4.9	2.7	$\eta_{\text{CG}}$	[%LHV <sub>daf</sub> ]	71.7	1.8
$\lambda_{\text{ch}}$	[% mass]	0.9	0.5	$\eta_{\text{CH}_4}$	[%LHV <sub>daf</sub> ]	61.1	1.5
$\lambda_v$	[% mass]	7.8	3.8	$\eta_{\text{sect}}$	[%]	69.2	1.6
$Z_{\text{H}_2}$	[% mass]	25.2	1.2	$\eta_{\text{plant}}$	[%]	57.7	1.3
$Z_{\text{CO}}$	[% mass]	9.8	0.8	$Q_{\text{iHD}}$	[%LHV <sub>daf</sub> ]	18	1.0
$Z_{\text{CH}_4}$	[% mass]	34.1	0.2	$E_{\text{rec,PG}}$	[%LHV <sub>daf</sub> ]	9.8	0.2
$Z_{\text{C}_2\text{H}_4}$	[% mass]	13.8	0.1	$Q_{\text{l,tot}}$	[%LHV <sub>daf</sub> ]	5.2	0.6
$Z_{\text{C}_3\text{H}_6}$	[% mass]	0.02	0.0	Fuel	[kg <sub>daf</sub> /h]	5820	142
$Z_{\text{tar}}$	[% mass]	3.5	0.2	feed*			
$Z_{\text{btx}}$	[% mass]	5.8	0.3				

$X_g$ , Char gasification,  $\lambda_{\text{otr}}$  oxygen transport,  $Z_i$  volatile converted to the *i*-th compound, \* from the carbon balance

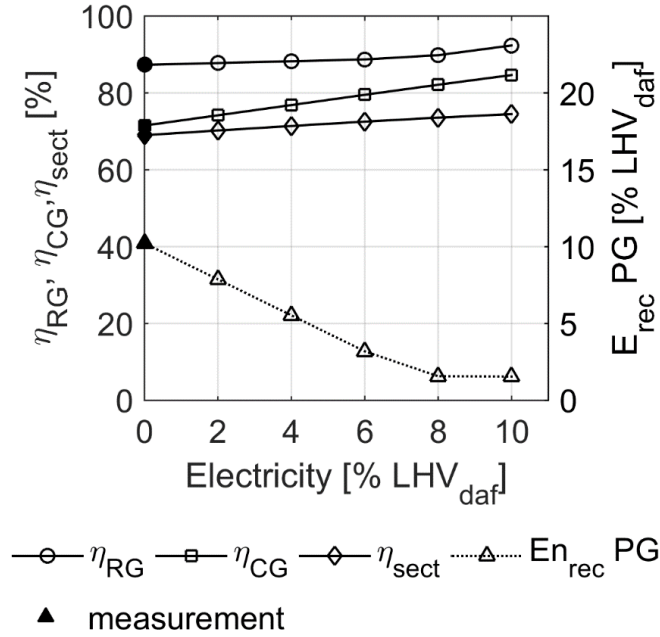


**Figure 14** - Sensitivity analysis of thermal measures.

A shift from wood pellets (8% moisture) to fresh wood chips (40% moisture, assuming the same chemical composition as the wood pellets) has the effect of reducing  $\eta_{CG}$  from 71.7%LHV<sub>daf</sub> to 56.3%LHV<sub>daf</sub> in the current design, while further drying of the fuel to 2% moisture could raise the cold gas efficiency by ~2 pp (Fig. 14b). This condition of extreme drying can be achieved with steam dryers, which are connected directly to the feeding system of the DFB gasifier, as suggested in Paper III [94, 95]. This type of dryer also pre-heats the biomass to a temperature of 80°–100°C, which further reduces the heat demand in the gasifier [95, 96].

Activation with potassium and sulphur affects the gas composition and reduces the tar content, enabling operation of the gasifier across a wider range of conditions. Figure 14d shows the results for the *K,S-act* case with low tar content and the same temperature levels as in the *K-act* case, and for the *K,S-act LT* case with the same tar content as the base case, but with the temperature in the reactors reduced by 50°C, both of which cases were experimentally tested in GoBiGas. In the *K,S-act* case, the lower yield of tar indicates that more energy is stored in the permanent gas, although this is partially compensated for by the higher level of re-circulation of the raw gas, which is used to counteract the lower tar flow to the combustor. In the *K,S-act LT* case, the lower temperature in the reactor reduces both the heat demand in the combustor and the rate of raw gas re-circulation, while the tar yield is similar to that in the base case. Overall, the cold gas efficiency is increased to 72.9%LHV<sub>daf</sub> for the *K,S-act* case and to 74.2% LHV<sub>daf</sub> for the *K,S-act LT* case.



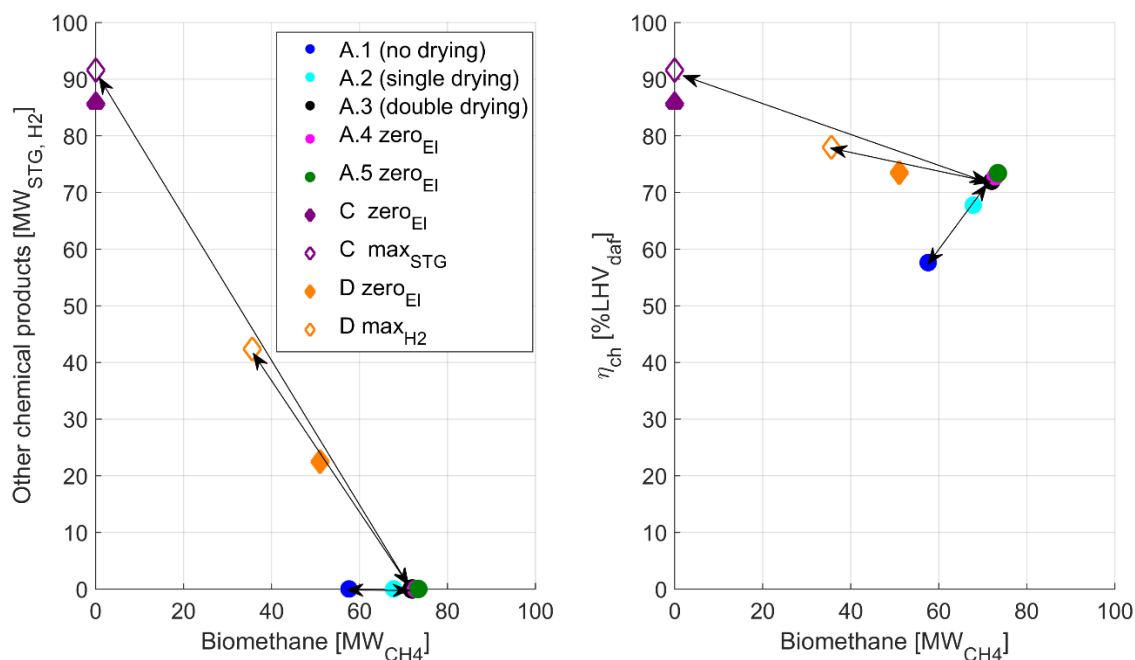


**Figure 15** –Effects of electricity introduction into the gasifier reactor (direct heating)

The introduction of electricity into the DFB gasifier affects multiple aspects of the process. Overall, the rate of re-circulation of the raw gas is reduced and it may reach the minimum value (Fig. 15). If more electricity is provided the gasification of char may increase. The minimum level of re-circulation of raw gas in this case is reached by introducing electricity corresponding to the 8% of the energy in the fuel, thereby achieving a cold gas efficiency of 82.1%LHV<sub>daf</sub>. An electricity input corresponding to 10% of the LHV of the fuel would enable char gasification to be increased from 53.8% to 60% and would increase the raw gas efficiency to 92.3%LHV<sub>daf</sub>. Unlike the other measures investigated, the introduction of electricity causes the cold gas efficiency and the efficiency of the gasification section to diverge (Fig. 15), since in the latter the electricity is accounted for as an energy input. In particular for the case in which electricity replaces 8% of the LHV of the fuel, the efficiency of the gasification section increases by ~4.5 pp, while the cold gas efficiency is increased by ~10 pp.

### 3.4 Process simulation of gasification *stand-alone, decentralized and local drop-in* plants

The data from the analysis of the GoBiGas plant (Paper V) are used to simulate gasification-based processes at large scale and to explore the potential of *stand-alone, decentralised, and local drop-in* plants. The results are presented in the form of chemical and total efficiencies, production ranges, and power-to-gas efficiency. The results show that it is possible for *stand-alone* plants to increase the chemical efficiency from the current level of 48%LHV<sub>daf</sub> [60] (at GoBiGas with 40% moist biomass) to ~72%LHV<sub>daf</sub> at the large scale (design A.3), i.e., by more than 20 pp. This improvement is due to the combination of advanced drying systems, high-temperature pre-heating of the inlet steam and air (up to 550°C), and better insulation of the reactors (heat losses <2%LHV<sub>daf</sub>).



**Figure 16** – Production ranges in the investigated plant designs,  $zero_{EI}$  net electricity production level equal zero

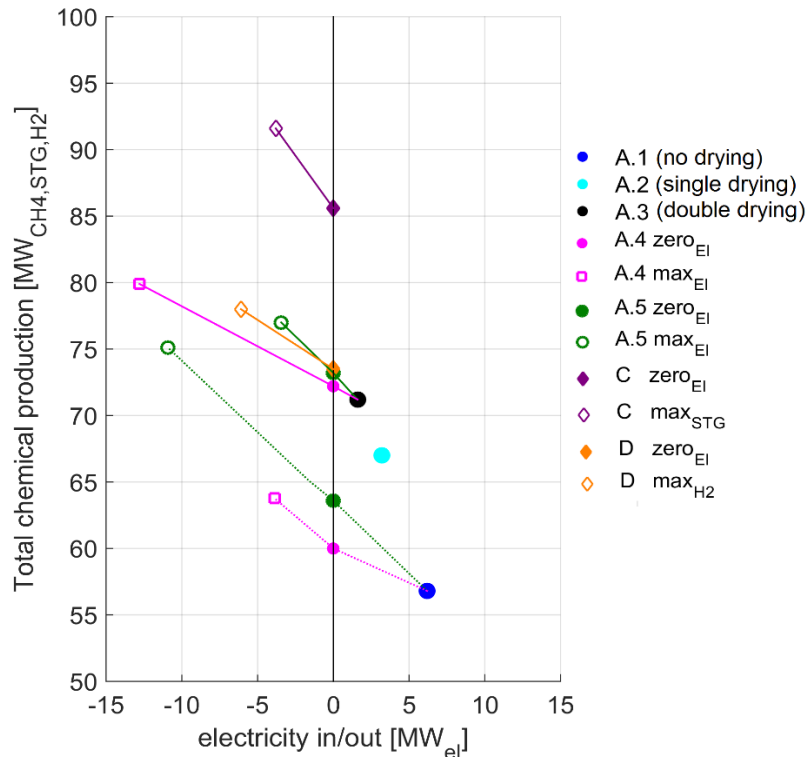
These measures optimize the biomass conversion in the gasifier, increasing the cold gas efficiency from 56.3%LHV<sub>daf</sub> of the GoBiGas plant with fresh biomass to ~84%LHV<sub>daf</sub>. With the introduction of a steam cycle, the total efficiency can be increased from 45% [60] (at GoBiGas) to 73% (design A.3). It is worth noting that in design A.3, the plant is a producer of electricity (net of the internal demand) rather than a consumer of electricity, as is the case for the GoBiGas plant.

These improvements can be reached in steps. Increasing the scale of the GoBiGas plant, with a reduction in heat losses, a higher pre-heating temperature, and the introduction of a steam cycle (design A.1) increases the chemical efficiency to 57%LHV<sub>daf</sub> and η<sub>tot</sub> to 63%. A major improvement is achieved by changing from design A.1 to design A.2, i.e., introducing an air dryer (reducing moisture content from 40% w.b. to 20% w.b.), which corresponds to an increase in chemical efficiency of around 10 pp, reaching 78%LHV<sub>daf</sub>, and a total efficiency of 70%. The introduction of the steam dryer (Paper III), to perform drying to <3% w.b. and recovering the moisture as a gasification agent (design A.3), increases the chemical efficiency by a further 3 pp.

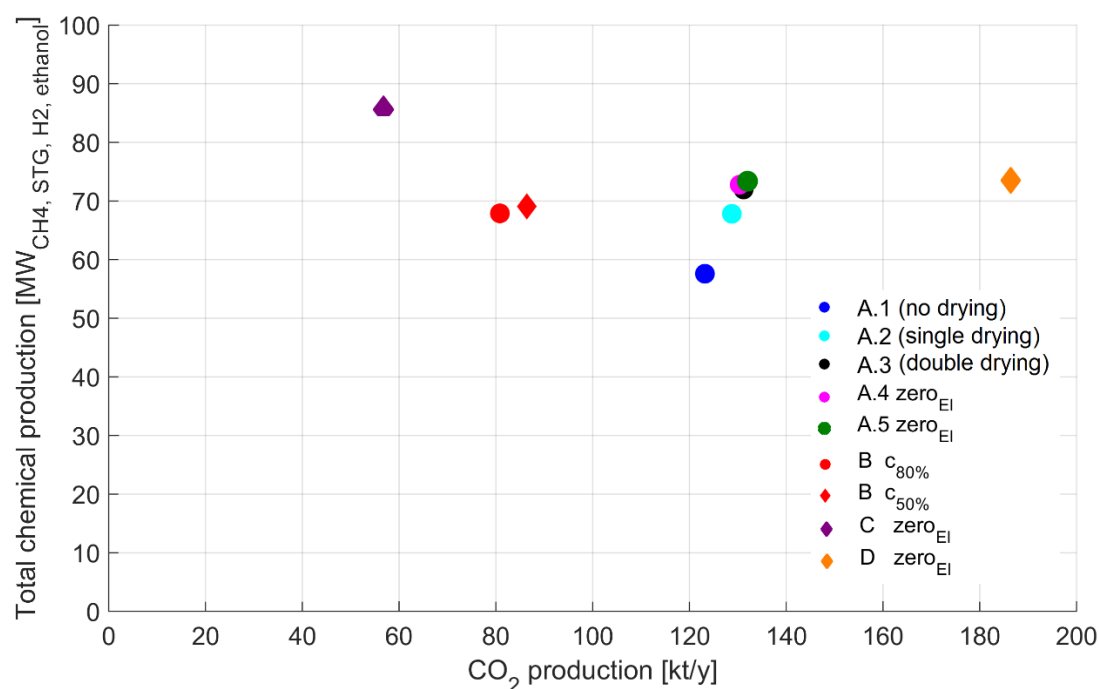
In *decentralised* and *local drop-in* plants that co-produce biomethane and STG or H<sub>2</sub>, the electricity demand of the plant can be a limiting factor. In particular, for STG plants, the electricity production is limited by the low level of excess heat in the process owing to the absence of methanation reactors, while for H<sub>2</sub> plants, the electricity demand is high owing to the compressors in the VPSA system. Therefore, the processes are investigated for two cases: a  $zero_{EI}$  case, in which the production is controlled so as to have zero consumption of electricity in the plant; and the  $max_{H_2/STG}$  case, in which electricity from the grid is used to maximise the production STG or hydrogen. The production range and efficiency are different for the  $zero_{EI}$  and  $max_{STG}$  cases (Fig. 16). For STG plants, η<sub>ch</sub> is in the range of 85.6–91.6%LHV<sub>daf</sub> and η<sub>tot</sub> is in the range of 85.6%–88.2%. For H<sub>2</sub>/biomethane, η<sub>ch</sub> is in the range of 73.5–78%LHV<sub>daf</sub> and η<sub>tot</sub> is stable at around 73.5%. The maximum production

levels of STG are in the range 85.6–91.6 MW<sub>STG</sub>. For the H<sub>2</sub> and biomethane cases, the maximum production levels are 22.5 MW<sub>H<sub>2</sub></sub> and 51 MW<sub>CH<sub>4</sub></sub> in the *zero<sub>El</sub>* case and 42.4 MW<sub>H<sub>2</sub></sub> and 35.6 MW<sub>CH<sub>4</sub></sub>, respectively, using electricity from the grid. It is worth noting that increasing the electricity consumption in the plant to maximise the production of STG or H<sub>2</sub> leads to higher efficiencies than are obtained in the *zero<sub>El</sub>* cases. Therefore, these processes are considered further, as well as highly efficient power-to-gas conversion (with  $\eta_{P2G} \geq 100\%$ ). Overall, due to the high efficiency and the extensive range of convenient locations for *decentralised* and *drop-in* plants, there is no substantial advantage to be gained from *stand-alone* plants producing biomethane, unless methane is the desired final product.

Direct heating and electrolysis power-to-gas technologies are investigated in designs A.5 and A.4, based on design A.3. For both these designs, two cases are investigated: a *zero<sub>El</sub>* case; and a maximum electricity case *max<sub>El</sub>*. In the former case, only the electricity produced from excess heat is converted in the power-to-gas process (zero net production), whereas in the latter case, electricity from the grid up to the maximum amount allowed by technical restrictions is used. The results show that direct heating, in a DFB gasifier, is a much more efficient technology than electrolysis, reaching  $\eta_{P2G}$  of ~115%, as compared with ~63% when using design A.3 as reference. However, the two power-to-gas technologies have different ranges of operation depending on the initial design of the plant. In particular, the application of direct heating is rather limited (4.8 MW<sub>el</sub>) in design A.3, as compared to electrolysis (15 MW<sub>el</sub>). The converse is true when power-to-gas technologies are applied to design A.1 (without drying), where direct heating can convert up to 10.9 MW<sub>el</sub> and electrolysis can convert 3.9 MW<sub>el</sub>. Figure 17 reports the comparison of the different power-to-gas processes applied to designs A.3 and A.1; included in the graph are the STG and H<sub>2</sub>/biomethane production levels.



**Figure 17** – Chemical production versus electricity consumption for plant designs that involve the conversion of electricity to bio-products. The filled symbols indicate net electricity production level equal zero (*zero<sub>El</sub>*).



**Figure 18** - Production ranges of CO<sub>2</sub> as a function of chemical production in the plant.

Figure 18 presents the amounts of CO<sub>2</sub> separated in the investigated plant designs. As expected, design D stands out as having strong potential to separate the used carbon on-site. Designs A.1–A.3 have similar potentials, with CO<sub>2</sub> production increasing linearly towards higher CH<sub>4</sub> yields. The amount of CO<sub>2</sub> produced at the gasification plant (in the range of 57–186 kt/yr; *cf.* Fig. 18) is at the low end of what is generally considered feasible for bio-energy carbon capture and storage (BECCS) Kjärstad, Skagestad [97]. However, the geographical location of the plant, being near a coast-line as well as a harbour, and the fact that it is part of an industrial cluster facilitate the implementation of BECCS at the gasification plant.

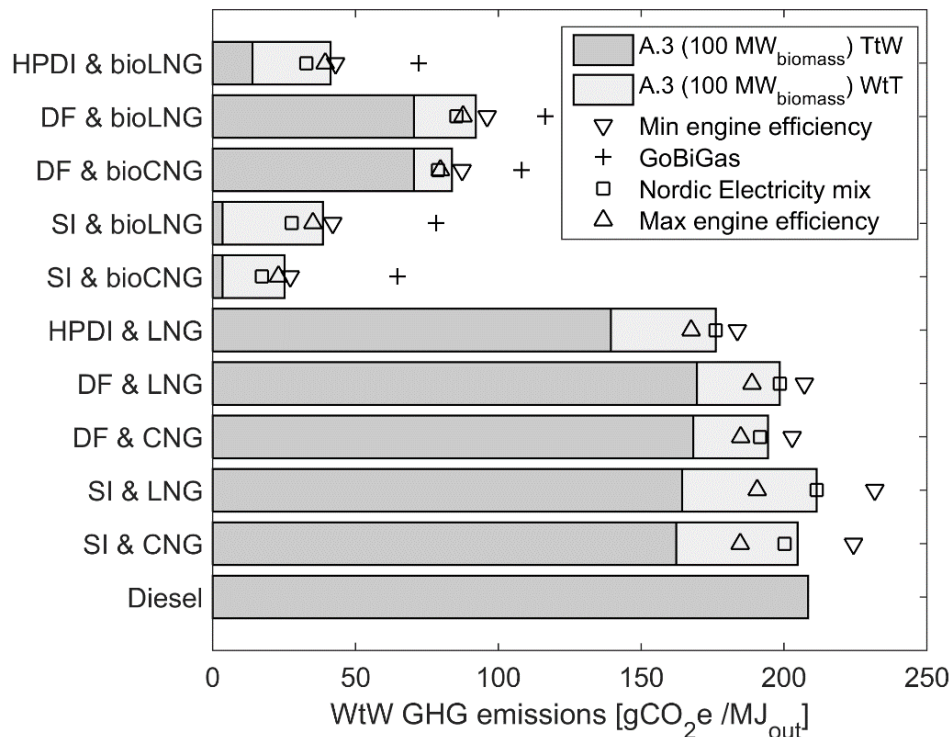
### 3.5 WtW analysis of the biomethane chain

The investigation of the fuel quality of 22 different compositions of NG [98] from a pipeline, LNG, and biogas in the EU market (Paper II) reveals that a purity of >95% for the biomethane product is optimal for combustion in gas engines. Therefore, the composition of biomethane for injection into the NG pipeline is not different from that designated for vehicle fuels. In the WtW analysis, pathways for different engine technologies were compared using fossil CNG and LNG and biomethane (bio-CNG and bio-LNG, respectively). The results in Paper I were here re-calculated for the production of biomethane in a large-scale stand-alone biomethane plant with a double-drying system and local production of electricity (design A.3 in Paper VI) and compared with the results based on the GoBiGas plant (Paper I). This comparison shows substantial improvements in emissions (Fig. 19) and *biomass impact* (Fig. 20) with optimisation of biomethane production. For HPDI engines the GHG WtW emissions with fossil LNG are 176 [gCO<sub>2</sub>e/MJ<sub>out</sub>], and they are reduced to 72 [gCO<sub>2</sub>e/MJ<sub>out</sub>] (a decrease of 59%) with the use of biomethane from the GoBiGas plant. With a large-scale biomethane plant a further reduction to 41 [gCO<sub>2</sub>e/MJ<sub>out</sub>] (a decrease of 77%) is achievable. Similar improvements are

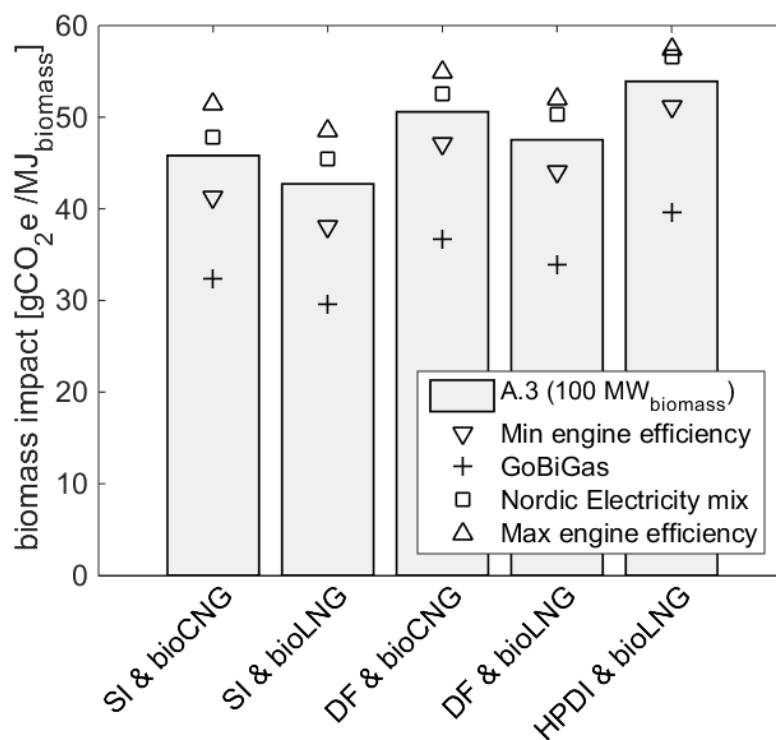
achieved for DF and SI engines, with emissions levels of 198 and 211 [gCO<sub>2</sub>e/MJ<sub>out</sub>] with fossil LNG, 116 and 78 [gCO<sub>2</sub>e/MJ<sub>out</sub>] with bio-LNG from GoBiGas, and 92 and 37 with [gCO<sub>2</sub>e/MJ<sub>out</sub>] large-scale bio-LNG production, respectively.

The effectiveness of the utilisation of the biomass resources is clearly quantified by the *biomass impact*, which expresses the emissions saving (from diesel) specific to the biomass resources used in the process. The pathway that achieves the highest BI is the HPDI engine with bio-LNG, which combines high efficiency and a low fraction of diesel to yield a BI value of 54 gCO<sub>2</sub>e<sub>saved</sub> per MJ<sub>out</sub>. The BI values for the DF cases indicate savings of around 51 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>] for bio-CNG and 47 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>] for bio-LNG, representing the highest BI values among the CNG cases. Despite their low GHG WtW emissions, the SI engines have the lowest BI values due to low engine efficiency, resulting in BI values of around 46 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>] for bio-CNG and 43 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>] for bio-LNG.

The BI enables a comparison of the improvements that could be achieved with a specific engine technology and optimisation of the biomethane process. Considering the current GoBiGas plant that produces bio-CNG for SI engines as the reference WtW chain, the BI can be improved by 7.3 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>] by switching to bio-LNG and HPDI engines and by 13.4 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>] by optimising the biomethane process (design A.3). Thus, the potential benefit of an investment in the production process is higher than that of an investment in the gas engine technology, with the current situation. Overall, it can be concluded that with large-scale production of biomethane the GHG WtW emissions can be lowered to 37–78 [gCO<sub>2</sub>e/MJ<sub>out</sub>] (corresponding to a reduction of 62%–82% from diesel use), generating a BI in the range of 43–54 [gCO<sub>2</sub>e<sub>saved</sub>/MJ<sub>biomass</sub>].



**Figure 19** - WtW, WtT and TtW emissions for different pathways including: fossil fuels, with biomethane from large scale production and biomethane from GoBiGas.



**Figure 20** – Biomass impacts for different combinations pathways, comparing biomethane from large scale production and from GoBiGas.

# 4

## Conclusions

The work carried on in his thesis describes the current status and identifies possible improvements in relation to biomethane production from biomass gasification and the utilisation of biomethane in heavy-duty engines. The focus is on the GoBiGas demonstration plant, which was built and commissioned during the timeframe of this thesis and is the first plant to produce biomethane at commercial scale.

The well-to-wheel analysis of the current production of biomethane in the GoBiGas plant and the utilisation of biomethane in SI heavy-duty engines (the most common technology) reveals GHG emissions for biomethane that are significantly lower than for fossil fuels ( $78 \text{ gCO}_2\text{e/MJ}_{\text{out}}$ ; a decrease of 66% compared to diesel use) and emission savings from diesel case of  $30\text{--}33 \text{ gCO}_2\text{e}_{\text{saved}}/\text{MJ}_{\text{biomass}}$  (*biomass impact*). Nevertheless, the simulations of the biomethane processes combined with the WtW analysis show that there are potentials to reduce the GHG emissions to  $41 \text{ gCO}_2\text{e/MJ}_{\text{out}}$  and increase the *biomass impact* up to  $54 \text{ gCO}_2\text{e}_{\text{saved}}/\text{MJ}_{\text{biomass}}$  through up-scaling and optimisation of the biomass process and the use of advanced engine technologies, such as HPDI. In particular, the potential impact from optimisation and up-scale of the biomethane process ( $+13.4 \text{ gCO}_2\text{e}_{\text{saved}}/\text{MJ}_{\text{biomass}}$ ) is larger than that that can be achieved from the advancement in gas engine technology ( $+7.3 \text{ gCO}_2\text{e}_{\text{saved}}/\text{MJ}_{\text{biomass}}$ ). Therefore, following the demonstration at a commercial scale, biomethane is established as a biofuel with a high environmental impact, although the gap between the current status and its potential application is highlighted.

The comparison of the three gas engine technologies leads us to believe that engines that utilise a small fraction of diesel in the fuel mixture (DF and HPDI) but that reach higher efficiencies can be more beneficial than SI engines that have lower efficiencies but are fuelled solely with biomethane. This conclusion is made under the assumption that biomass is a limited resource (i.e., based on the *biomass impact*) and that the effect of biomass should be maximised. In particular, HPDI fuelling with bio-LNG emerges from the WtW pathways as having the strongest potential.

The performance of the biomethane process is strongly related to the DFB gasifier. The evaluation of the gasification section of the GoBiGas plant reveals a high level of fuel conversion and flexibility in the operation when potassium and sulphur additives are used. In the operation with dried biomass (8% moist w.b.), the char gasification was estimated as  $\sim 54\%$  and the fraction of volatiles directly converted to methane was  $\sim 34\%$ , with a tar content in the range of  $10.0\text{--}20.5 \text{ g/Nm}^3$  depending on the operational conditions. Nevertheless, the DFB system at GoBiGas is not optimised for those thermal measures that reduce the internal heat demand and minimise fuel combustion, such as drying systems,

high-temperature pre-heating, and effective minimisation of convective heat losses from the reactors, thereby affecting the cold gas efficiency, which is calculated as  $71.7\% \text{LHV}_{\text{daf}}$ .

Owing to the high level of fuel conversion and low tar content, the efficiency of the DFB gasifier can be improved at large-scale using existing technologies and by optimising the heat recovery in the plant. The results of the simulation with 40% moist biomass demonstrate that it is possible to increase the cold gas efficiency from  $56.3\% \text{LHV}_{\text{daf}}$  (without drying) to  $\sim 84\% \text{LHV}_{\text{daf}}$ , which corresponds to an increase in the chemical efficiency from  $48\% \text{LHV}_{\text{daf}}$  to  $72\% \text{LHV}_{\text{daf}}$ . These results can be obtained by reducing the heat losses, increasing the pre-heating, and introducing an advanced drying system constituted by a first air dryer (drying to 20% moist) and a second steam dryer with recovery of the moisture as a gasification agent, a concept that was conceived within this work.

Another opportunity offered by gasification processes is the introduction of power-to-gas technologies to convert intermittent renewable energy sources to biofuels. Two power-to-gas technologies were compared: one based on the electrolysis of water and the other involving direct electrical heating of the gasifier. Due to the overall endothermic nature of the gasification process, direct heating is more efficient than electrolysis, reaching a power-to-gas efficiency in the range of 85%–110%. However, the range of applications for direct heating is narrower than that for electrolysis in optimised large-scale plants, and both technologies could be implemented to maximise the conversion capacity.

The introduction of gasification plants in the chemical sector, through the production of intermediate chemicals (STG and hydrogen), was investigated with the focus on the geographical location of the plant (*decentralised* and *local drop-in* strategies), as compared to *stand-alone* plants. Compared to biomethane, the chemical efficiency for STG is substantially higher ( $>85\% \text{LHV}_{\text{daf}}$ ), and for hydrogen/biomethane co-production it is comparable and shifting increasingly towards hydrogen. The results obtained in this thesis highlight that there is no substantial advantage for the chemical sector to use *stand-alone* plants that produce biomethane, unless biomethane is the desired end-product.



# 5

## Consideration for future research

The analysis of large-scale gasification processes, with internally optimised heat recovery and electricity production, highlights several possibilities for integration into local energy systems. This is made possible by the ability of the process to switch between production and conversion (consumption), of both electricity and low-temperature heat (<150°C). Typically, a *stand-alone* plant can balance changes in the energy system by effecting regulation in four directions: 1) increasing electricity production by reducing the chemical efficiency (power boiler); 2) increasing district heating by reducing the electricity production (heat boiler); 3) accumulating low-grade excess heat from other plants *via* drying of feedstock (to 20% w.b.); and 4) converting electricity from intermittent sources *via* power-to-gas technologies. Other possibilities not investigated in the thesis include the use of low-grade heat for the distillation of bio-chemicals (ethanol) and the introduction of heat pumps to convert electricity to and from condensation to heat for drying. Gasification-based plants can have a greater impact than solely biofuel or biochemical production. Therefore, the integration of gasification plants into the local energy system should be complemented with a dynamic analysis that includes seasonal and daily variations.

The successful operation of the GoBiGas plant demonstrates that industrial-scale production of biomethane is feasible, and the results of this work show that scaling-up of the plant is required to maximise the environmental impact. However, the actual establishment of such a WtW chain in the coming years remains in doubt due to the current low prices of oil and natural gas. In the absence of long-term policies for biofuels, it is suggested to investigate gasification plants for the conversion of biomass using a *decentralised* or *local drop-in* strategy, rather than conversion to single biofuels (i.e., the biorefinery concept), and to optimise at the local level their integration with the remainder of the energy system.

Nomenclature and list of abbreviations			
<b>DFB</b>	dual fluidized bed	<b>STG</b>	sustainable town gas
<b>GHG</b>	greenhouse gas	<b>PG</b>	permanent gases
<b>WtW</b>	well-to-wheel	<b>OC</b>	organic compounds (inc. tar)
<b>WtT</b>	well-to-tank	<b>MeO, Me</b>	metal oxide, (reduced form)
<b>TtW</b>	tank-to-wheel	<b>TC</b>	total carbon (measurements)
<b>NG</b>	natural gas	<b>SD</b>	standard deviation
<b>LNG</b>	liquefied natural gas	<b>CFD</b>	computational fluid dynamic
<b>CNG</b>	compressed natural gas	$X_g$ [-]	fraction of char gasified [-]
<b>bio-LNG</b>	liquefied biomethane	$\lambda_{Otr}$ [-]	oxygen transport as equivalence ratio for combustion of the fuel
<b>bio-CNG</b>	compressed biomethane	$Z_i$ [-]	fraction of volatile mass converted in the formation of the <i>i</i> th energy carrying compound
<b>LUC, iLCU</b>	land use change, indirect-	$fCd$ [-]	fraction of carbon in the raw gas detected by the measurements
<b>SI</b>	spark ignited	<b>LHV</b>	lower heating value
<b>DF</b>	dual fuel	<b>LHV<sub>daf</sub></b>	lower heating value of dry-ash-free biomass
<b>HPDI</b>	high pressure direct injection	$El_{in}, El_{out}$	electricity consumed in the plant, produced in the plant
<b>BI</b>	biomass impact	$E_{RG}, E_{CG}, E_{RME}, E_{biomass}, E_{STG}, E_{CH4}$	energy content calculated from the LHV of: raw gas, cold gas, RME, biomass <sub>daf</sub> , STG, biomethane
<b>RME</b>	rapeseed methyl ester	$E_{rec, RG}$	energy in the recirculated raw gas
<b>daf</b>	dry-ash-free	$\eta_{RG}, \eta_{CG}, \eta_{sect}, \eta_{ch}, \eta_{tot}, \eta_{P2G}$	-efficiency, raw gas, cold gas, gasification section, chemical, total, power-to-gas
<b>BTX</b>	benzene-toluene-xylene	$Q_{iHD}$	internal heat demand of the gasifier
<b>WGS</b>	water gas shift	$Q_{tot}$	heat losses in the DFB gasifier

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