Advanced biofuel production via gasification – lessons learned from 200 man-years of research activity with Chalmers' research gasifier and the

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IN THE FIELD

Advanced biofuel production via gasification – lessons learned from 200 man-years of research activity with Chalmers' research gasifier and the GoBiGas demonstration plant

Henrik Thunman[1] (iD), Martin Seemann[1] (iD), Teresa Berdugo Vilches[1] (iD), Jelena Maric[1] (iD), David Pallares[1] (iD), Henrik Ström[2] (iD), Göran Berndes[3] (iD), Pavleta Knutsson[4] (iD), Anton Larsson[5] (iD), Claes Breitholtz[6] & Olga Santos[7]

[1]Division of Energy Technology, Chalmers University of Technology, 412 96 Göteborg, Sweden
[2]Division of Fluid Dynamics, Chalmers University of Technology, 412 96 Göteborg, Sweden
[3]Division of Physical Resource Theory, Chalmers University of Technology, 412 96 Göteborg, Sweden
[4]Division of Energy and Materials, Chalmers University of Technology, 412 96 Göteborg, Sweden
[5]Göteborg Energi AB, Göteborg, Sweden
[6]Valmet Power AB, Göteborg, Sweden
[7]Alfa Laval AB, Lund, Sweden

Abstract

This paper presents the main experiences gained and conclusions drawn from the demonstration of a first-of-its-kind wood-based biomethane production plant (20-MW capacity, 150 dry tonnes of biomass/day) and 10 years of operation of the 2–4-MW (10–20 dry tonnes of biomass/day) research gasifier at Chalmers University of Technology in Sweden. Based on the experience gained, an elaborated outline for commercialization of the technology for a wide spectrum of applications and end products is defined. The main findings are related to the use of biomass ash constituents as a catalyst for the process and the application of coated heat exchangers, such that regular fluidized bed boilers can be retrofitted to become biomass gasifiers. Among the recirculation of the ash streams within the process, presence of the alkali salt in the system is identified as highly important for control of the tar species. Combined with new insights on fuel feeding and reactor design, these two major findings form the basis for a comprehensive process layout that can support a gradual transformation of existing boilers in district heating networks and in pulp, paper and saw mills, and it facilitates the exploitation of existing oil refineries and petrochemical plants for large-scale production of renewable fuels, chemicals, and materials from biomass and wastes. The potential for electrification of those process layouts are also discussed. The commercialization route represents an example of how biomass conversion develops and integrates with existing industrial and energy infrastructures to form highly effective systems that deliver a wide range of end products. Illustrating the potential, the existing fluidized bed boilers in Sweden alone represent a jet fuel production capacity that corresponds to 10% of current global consumption.

Introduction

According to the Intergovernmental Panel on Climate Change (IPCC), scenarios that have a good chance of restricting global warming to less than 2°C involve substantial cuts in anthropogenic greenhouse gas (GHG) emissions, implemented through large-scale changes in energy systems. The use of renewable energy sources and fossil fuels, in combination with carbon capture and storage (CCS), could help to reduce GHG emissions in the

energy sector. Electricity can be produced from noncarbon sources, such as wind, hydro, and solar energy, and from carbon-based feedstocks, which are also needed for the production of fuels, chemicals, and various materials.

There are three main alternatives for producing carbon-based feedstocks: (1) biomass harvesting, that is, relying on photosynthesis as the mechanism for capturing CO_2 from the atmosphere; (2) CO_2 capture via physical or chemical processes from the atmosphere or seawater; and (3) recycling, through the utilization of suitable materials, such as recycled paper and plastics, waste wood, or through CO_2 capture from flue gases. The future potential of the latter option depends on whether burning of hydrocarbons to produce process heat and/or electricity will remain common, which is uncertain.

The future magnitude of biomass resources is currently debated, and estimates of bioenergy potentials vary widely due to differences in the approaches adopted to consider important factors, which in themselves are uncertain [1, 2]. Moreover, biomass supply may be limited by a scarcity of resources, such as land and water, and society may want to avoid over-reliance on biomass harvesting due to concerns regarding negative environmental and socio-economic impacts. Therefore, it is reasonable to expect that future biomass use will be prioritized for applications for which alternatives at similar cost levels are not available.

For example, heat can be produced and stored based on geothermal heat and renewable electricity. As another example, biomass-based electricity may not be needed at locations and during time periods when other renewable or fossil-free alternatives are available. Widespread application of various storage and demand management strategies, together with renewable supply options, such as wind and solar energy, could limit the periods of the year that are suitable for fuel-based thermal electricity generation to hundreds rather than thousands of hours, and might restrict the periods for continuous operation of such plants to days or weeks instead of months.

In such a scenario, it will be advantageous to combine the continuous production of renewable fuels, materials, and chemicals with intermittent generation of heat and electricity. Biorefinery concepts that are based on large-scale gasification represent one such combined production solution. In contrast, large thermal production plants that produce only electricity and/or heat are unlikely to be economically viable. In this context, it will be desirable to introduce novel solutions that exploit the infrastructure that has been built up in recent decades for biomass-based heat and combined heat and power (CHP) production.

The Nordic countries of Sweden, Finland, and Denmark have been forerunners in the development of large thermal production plants using biomass for electricity and/or heat production. In Sweden, more than 60 units, with a thermal capacity of >50 MWth (250 dry tons of biomass/day) biomass or waste (with 40 units of >100 MWth, 500 dry tonnes of biomass/day), have been built at a cost of more than 100 M€ per 100-MWth unit. The main technology used in this sector is bubbling or circulating fluidized bed combustors, and Sweden alone has a total installed thermal capacity of 6400 MWth (1200 MMBtu/hour). If the utilization of those fluidized bed units can be extended, these would represent an asset available for the introduction of a large production capacity for biomass to biofuels.

In this paper, the results of research and development activities conducted in two industrial-scale demonstration units are used to formulate a strategy for how fluidized bed boilers can be converted to gasifiers, in a manner similar to that used in the 1990s to convert coal-fired grate boilers in the Nordic countries to biomass-fired fluidized bed boilers. The conversion offers a low-cost route for the production of alternative fuels, materials, and chemicals (instead of heat or CHP) from carbon-based feedstocks and hydrocarbons based on renewable electricity. It is also described how the gasification concept can be implemented in pulp, paper and saw mills, as well as in oil refineries and the petrochemical industries, as well as the potential of electrification of the processes. The results of 200 man-years of research activity, carried out over the last decade are summarized in this paper.

The research results obtained and experiences gained concerning the gasification process are summarized in *Description of and Results from the Technical Demonstrations*. This section contains brief descriptions of materials that have been published elsewhere, as well as more detailed descriptions of results that have not been published previously. The applications and descriptions of routes for introducing the examined gasification technology into the energy system and industrial systems including the potential for electrification of the processes are presented in *Introduction of the Technology into the Existing Infrastructure*.

Description of and Results from the Technical Demonstrations

With the vision of establishing local production of around 1 TWh/year (86,000 TOE) of advanced biofuels in Gothenburg before Year 2020, the **Go**thenburg **Bi**omass **Gas**ification (GoBiGas) project was initiated in Year 2005. The GoBiGas project currently comprises a 32-MWth dual fluidized bed (DFB) gasifier (150 dry tonnes of biomass/day, 9.5 MMBtu/hour), complemented by state-of-the-art synthetic natural gas (SNG) synthesis,

producing up to 20 MW of biomethane (5.9 MMBtu/hour). It is a first-of-its-kind plant for industrial-scale production of advanced biofuels from woody biomass, whereby methane was identified as the desired end-product due to local conditions. To support the GoBiGas project, a research program with a 2–4-MW (10–20 dry tonnes of biomass/day) DFB gasifier was established in 2007 at Chalmers University of Technology, hereinafter referred to as the Chalmers gasifier. As the research gasifier was constructed by retrofitting an existing boiler, it is an example of the conversion of a boiler to a gasifier. The project has acquired experience from around 10,000 h of operation of the GoBiGas demonstration plant and more than 25,000 h of operation of the research gasifier at Chalmers [3–5].

The gasifier in the GoBiGas demonstration plant is one of two, third-generation dual fluidized bed gasifiers that originated from the 8–MWth (40 dry tonnes of biomass per day) CHP plant that was built in Güssing, Austria, in Year 2000, based on research conducted at the Technical University of Vienna under the supervision of Professor Hermann Hofbauer. This was followed by the construction of a CHP plant of the same size in Oberwart, Austria, which employed integrated fuel drying and an organic cycle to improve the electrical efficiency [6–8]. The other third-generation gasifier is a 16-MWth (80 dry tones of biomass/day) CHP plant in Senden, Germany, which was designed to use forest residues as fuel, that is, similar fuel use but half the capacity of the gasifier in the GoBiGas project. The unique property of the GoBiGas plant is that it is designed for advanced biofuel production via a syngas synthesis process. The Chalmers gasifier builds on the same principles, although the gasifier is built as an add-on to an existing boiler. A similar gasification system of the same size was in the same time period developed and built (under the TIGAR trademark) by IHI Corporation of Yokohama, Japan. This was later scaled up to a 15-MWth (30 dry tonnes of biomass/day) demonstration unit in Kujan, Indonesia and brought into operation in 2015, whereby lignite was used as the fuel and the intended product for a subsequent demonstration plant is ammonia [9, 10].

The research associated with the GoBiGas and Chalmers gasifiers had as its initial goal to demonstrate – on a commercial scale – the feasibility of converting biomass with 50% moisture to methane with a conversion efficiency >75% (higher heating value basis, which is similar to the efficiency on a lower heating value basis when a dry fuel is used). With the successful operation of the GoBiGas plant and the extrapolation of its performance to a commercial unit (5–10-times larger), this goal has been realized [11]. The performance level of the plant was achieved

thanks to an improved understanding of how the process chemistry is affected by the key ash species of potassium, sulfur, and calcium [5]. In particular, research using the Chalmers gasifier has provided essential validation data, which have been complemented by investigations in laboratory-scale reactors of the release of alkali compounds from single biomass particles [12].

The abilities of certain bed materials to capture and release these active ash species, with potassium being identified as the most important, were found to be crucial in limiting the yield of tar from the gasifier [13]. The beneficial properties of potassium are exploited in a way that is similar to that in the process developed by Exxon at the end of the 1970s for the direct production of methane using potassium-impregnated coal [14]. However, the latter process never reached the commercial implementation stage. One issue is the high silicon content of the coal ash consuming the potassium catalyst. Even though similar reactions occur with biomass ash, the consequences are negligible considering the high abundance of potassium in most woody biomasses.

The research findings at Chalmers explain how the biomass ash contributes to a suitable chemistry for the conversion process. This process should be regarded as a steam reforming process rather than a traditional gasifier, as it enables steam reforming processes that are commonly used to produce syngas (H_2, CO, CO_2) for a variety of industries based on lower-value, ash-rich fuels and waste streams. In addition to syngas, the DFB gasification process yields a variety of side products, such as light hydrocarbons and aromatic hydrocarbons, which can be utilized in the production of various chemicals and materials or heat.

In general, the applied process can be divided into three conversion steps (Fig. 1): Heat Generation (1); Gasification (2); and Synthesis (3). In addition, there are two bridging process units for compression (4) and BTX removal (5), which connect the conversion of the solid fuel to an intermediate gas with the upgrading of the gas to the final product. The heat generation and gasification processes are described in *Gasification system*, and optimization of the gasification and heat generation processes are discussed in *Optimization of the gasification process*. The synthesis, bridging process steps, and the material consumption and waste streams are briefly described and discussed in *Synthesis process*, *Bridging processes*, *Material consumption and waste streams during operation of the GoBiGas plant*.

Gasification system

A DFB gasifier of the type applied in the GoBiGas plant is primarily a circulating fluidized bed (CFB) boiler used
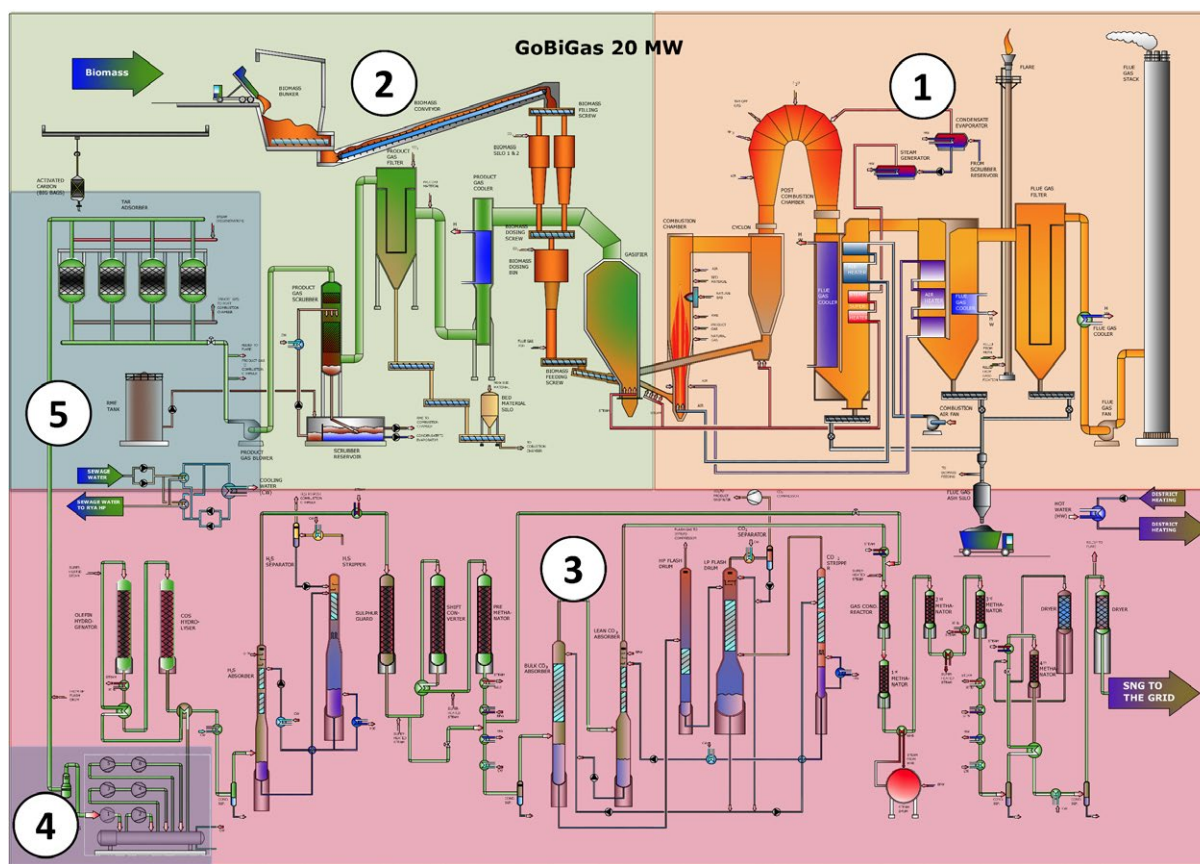
Figure 1. Schematic of the process layout for the GoBiGas demonstration unit, which is a complete system that converts raw biomass to a high-quality biofuel, that is, bio-SNG. In comparison to a commercial plant, the only missing part is the drying of the fuel, which is performed elsewhere. Process layout provided by Göteborg Energi AB.

for heat generation, which is connected to a bubbling fluidized bed (BFB) boiler that is used as the gasifier. For it to be used as a gasifier, the connected boilers have the conventional design of boilers that are intended for fuels with a high moisture content (≥50% moisture in the received fuel). This means that the walls of the combustion and gasification chambers are covered by refractory materials to retain the heat within the process. Even though the GoBiGas plant is large for a biomass gasifier, the small size of the plant is apparent when one compares it to standard CFB and BFB boilers. For example, the CFB reactor (5–10 MWth capacity, 25–50 dry tonnes of biomass/day) is 5–10-times smaller than the smallest commercial CFB boilers on the market. The BFB reactor (10–15 MWth capacity, 50–75 dry tonnes of biomass/day) is within the capacity range of the smallest commercial boilers. The gasification is described in the following section, and the consequences of the small size of the fluidized beds, as well as aberrations that emanate from the standard equipment in the applied design are discussed.

Combustion section

Despite the small size of the GoBiGas plant, the outline of the "CFB boiler" follows that of a commercial system designed for a complex fuel with a high moisture content upon reception; see Figure 2 for a schematic of an 88-MWth (440 dry tonnes of biomass/day) multi-fuel boiler from Valmet, which is comparable to the CFB boiler in the GoBiGas plant.

In contrast to a conventional CFB boiler used in commercial systems, there is no external solid fuel feed to the combustor in the GoBiGas plant. In addition, there is no flue gas condenser, which is installed in systems optimized for district heating, since the moisture content of the flue gas from the gasification system will be very low (a few percent, on a volume basis). Furthermore, the GoBiGas setup includes a large postcombustion section. The process outline of the combustion part of the GoBiGas plant is described in below, treating the convection path (including filtering) and the combustor separately.
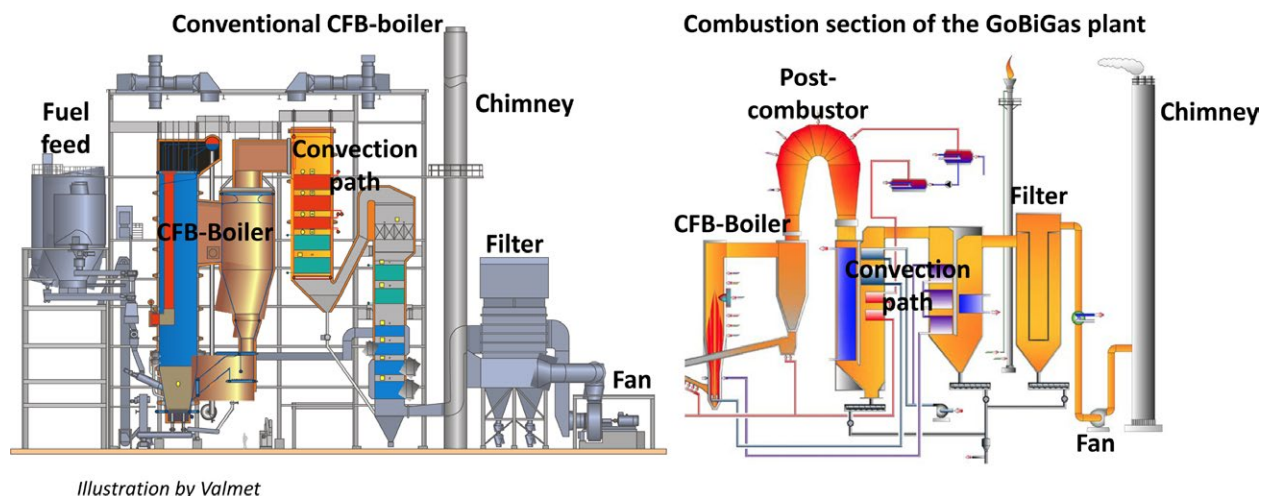
Figure 2. The 88-MW (440 dry tonnes of biomass/day) CFB Multi-Fuel Boiler from Valmet (left panel, illustration published with permission from Valmet), and an outline of the combustion section of the GoBiGas plant (right panel).

The convection path of the GoBiGas process is designed to handle a flow that exhibits an uneven particle load. A significant fraction of the particles in the flue gas flow is alkali salts, which condense during the cooling process inside the convection path. To handle this, the first chamber is an empty downdraft chamber with cooled walls, which gradually reduces the temperature and provides sufficient residence time for the particles to become non-sticky. During the cooling process, alkali can condense onto larger particles. These larger particles, coarse fly ash, consist of small particles of bed material and ash fragments that were generated through attrition, and they are gravity-separated from the flue gas when the flow is redirected to an updraft chamber. Heat transfer surfaces are introduced in the updraft chamber to preheat the combustion air and to produce superheated steam at 320°–350°C (600°–660°F).

Thereafter, the gas is sent to a second downdraft chamber, where the air that enters the combustion chamber is preheated. Here, more particles of the same type as those separated in the bottom of the first chamber are separated from the gas by gravity as the gas is directed to the final updraft chamber, where water is preheated. Downstream of the convection path, the remaining particles, mainly fly ash and free alkali particles, are removed by passage through a filter before the flue gas is finally vented to the atmosphere through the chimney at a temperature of around 140°C (280°F). This stepwise removal of particles enables selective particle recirculation to the gasification process. At GoBiGas, important ash components are thus reintroduced to the process through recirculation of particles from the two downdraft chambers to the process. This setup could be further optimized in

a commercial plant [15]. The design resembles that of a conventional CFB boiler, and it has been operated without major issues since it was first commissioned. The efficiency of energy recovery could be further optimized by introducing a flue gas condenser.

The combustor is optimized for its main purpose of producing high-value heat for the various processes, most notably the gasification. The reactor walls are insulated by refractory materials and the incoming air is preheated. In the present design, the temperature in the outer walls of the reactor is kept above the condensation point (120°–140°C, 250°–280°F), to avoid condensation on the steel sealing, which would otherwise entail corrosion issues. This design was a consequence of the small reactor size, which did not motivate the installation of water-filled panel walls to extract and recycle part of the heat that is transferred through the refractory [4]. In a commercial-sized reactor, the area-to-volume ratio is more favorable, which significantly reduces heat losses through the walls and ensures that the incorporation of water-filled panel walls is reasonable.

Unconverted char from the gasifier is used as the main fuel for the combustor, and recirculated by-products from the downstream process are used as supplementary fuel. In addition, the temperature of the process is regulated by combusting part of the gas produced in the gasification process or, during start-up, by combusting natural gas. While this solution is reasonable for a small pilot-scale unit, it is unlikely to be optimal for the size of the present system and certainly not for a commercial-scale plant. As will be discussed in *Controlling the gas quality using potassium*, this solution creates problems when adjusting the chemistry of the process, especially during

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start-up. For future units of size equivalent to or larger than the GoBiGas plant, it will be convenient to feed solid biomass also to the combustion side. This would simplify the start-up, increase the flexibility of the fuel feed to the gasifier, and reduce the operational costs, since both natural gas and the product gas currently used at GoBiGas have a higher market value than raw biomass. Further, during the start-up phase, and during operation with a fuel that has an unfavorable ash composition, the biomass burned to heat up the system would ensure the necessary ash balance in the system.

To obtain complete combustion of the gases that exit the combustor, a postcombustion chamber needs to be added. The construction of such a chamber depends on the planned mode of operation and the choice of fuel. If the plan is to use waste as fuel and the gasifier is to be operated within the European Union, the afterburner chamber needs to have a residence time of 2 sec at 850°C (1560°F) [16]. In the absence of such design requirements, the experience with the Chalmers DFB system suggests that the size of the afterburner chamber can be reduced significantly while still meeting the requirement to control the potassium-sulfur balance so as to obtain full conversion of the fuel. As will be discussed in *Controlling the gas quality using potassium*, a process that has a potassium-sulfur equilibrium will experience a problem with unconverted CO in the flue gas. This means that in a combustor that is equipped with a potassium-saturated bed, the CO will not be fully converted unless a small amount of sulfur is added [13], for example in the form of elemental sulfur

or ammonium sulfate; the latter can conveniently be added to the afterburner chamber. Furthermore, the costs for construction and maintenance can be reduced by constructing the afterburner chamber with straight panel walls, in line with the design of regular combustors, instead of using the complicated macaroni shape of the GoBiGas system, which has its origin in a small pilot-scale system.

Gasification section

The second reactor in the gasification system is the BFB reactor, which functions as a gasifier in the GoBiGas process, but is comparable to a BFB boiler. The differences between these two systems are mainly evident in the convection pathway (gas cooling and cleaning) and in the fuel feeding (Fig. 3). These differences will be discussed in relation to five areas based on the functionality of the system: (1) the convection path, including filtering and condensation of water and polyaromatic hydrocarbons (PAHs); (2) heat transfer; (3) chemistry; (4) primary fuel conversion; and (5) fuel feeding.

The convection pathway in the gasification part of the GoBiGas process (Repotec design) is very basic compared to the convection pathway on the combustion side (Valmet design). The design of the cooling step applied in GoBiGas, with the gas being passed through hundreds of (externally located) water-cooled tubes, resembles the design of the heat exchangers used for tar and alkaline-poor product gases produced from mainly liquid or gaseous fuels. As
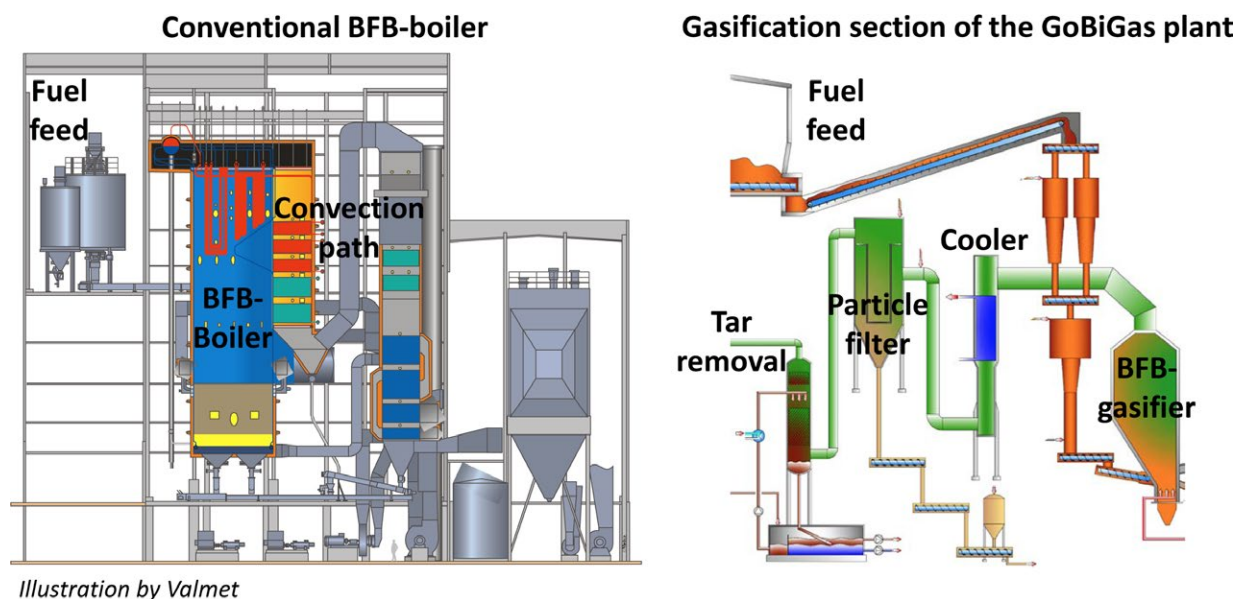

Illustration by Valmet

Figure 3. The second reactor in the gasification system is the BFB reactor, which functions as a gasifier in the GoBiGas process, but is comparable to a BFB boiler. The differences between these two systems are mainly evident in the convection pathway (gas cooling and cleaning) and in the fuel feeding (Fig. 3).

described for the CFB convection path, a more robust design is required for solid, ash-rich fuels. By gradually lowering the temperature, to avoid temperature windows in which various components can condensate (as is done in the CFB and BFB boilers), operational issues related to fouling by tar and alkaline can be handled.

The current GoBiGas design creates a challenge for process start-up, until the activity of the bed material has been reached, as fouling from either alkali salts or large PAH deposits on the surfaces of the heat exchangers occurs readily. The surface temperature of the heat exchangers in the gasification section need to be maintained within a narrow temperature range of 140–200°C (285–390°F), and the cooler will work properly only if the catalytic activity of the bed material is sufficient, since it will otherwise become clogged with tar within a few hours [5]. However, the most common cause of clogging of the cooler is alkaline deposits at the top of the cooler during start-up, when the bed material is saturated to avoid high tar yields, as will be described in *Controlling the gas quality using potassium*.

The experience gained from the research conducted with the Chalmers gasifier suggests that the convection path of the gas exiting the gasifier should follow the same layout principles as those that apply to the combustion side, that is, the employment of an empty downdraft chamber before the heat exchanger, to cope more effectively with the alkaline components of the gas. Furthermore, the outlet temperature from the initial cooling stage should ideally be adjusted from 140 to 200°C (285°–390°F) to around 400°C (750°F) during start-up

and disturbances. It would also be preferable to extract particles at several steps, as is done on the combustion side, to permit more stringent control of the chemical circulation of fines and to reduce the particle loads on the downstream filters.

At GoBiGas, the particles are separated from the product gas using textile bag filters that are coated with limestone to form a filter cake. The added limestone provides the system with additional calcium, which is an important ash component for the process, as described in detail in *Controlling the gas quality using potassium*. With the present design, the filter tolerates temperatures of up to approximately 230°C (450°F), thereby imposing a restriction on the outlet temperature of the gas from the convention pathway. As previously discussed, for a future system, it would be advisable to upgrade the filters so that they can handle temperatures up to 400°C (750°F), at least for short periods of time.

Downstream of the textile bag filters, steam and PAHs are, in the present process, removed by passage through a scrubber. To simplify the separation, rapeseed oil methyl ester (RME) is used as a scrubber agent to absorb the PAHs. However, the composition of the gas at this stage of the process is typically >30% volume steam and <1% PAHs. A typical composition of the PAHs and the separation efficiencies of the different tar components in the RME scrubber are listed in Table 1 (see *Cooling and cleaning of the product gas*). Thus, the scrubber acts mainly as a product gas condenser. The RME used to absorb and separate the PAHs should in future plants be replaced by a less-expensive scrubbing agent or separation method,

Table 1. Typical compositions of the major PAHs that influence the dew-point of the gas entering the RME scrubber in the GoBiGas plant, using wood pellets as the fuel. (n.d.: no significant change determined).

Component	Typical concentration [g/Nm^3]	Average removal, RME-scrubber (%)	Test at Chalmers [g/Nm^3]
Benzene	13.24	n.d.	8.41
Toluene	0.54	n.d	3.00
Xylene	0.00	n.d.	0.00
Styrene	0.13	n.d.	0.95
Indene	0.16	29	0.91
Naphthalene	2.66	72	2.15
2-MethylNaphthalene	0.03	91	0.39
1-MethylNaphthalene	0.02	98	0.27
Biphenyl	0.04	100	0.16
Acenaphthylene	0.52	98	0.60
Acenaphthene	0.02	100	0.57
Dibenzofuran	0.04	98	0.11
Fluorene	0.06	100	0.18
Phenanthrene	0.43	100	0.50
Anthracene	0.04	100	0.08
Fluoranthene	0.13	100	0.10
Pyrene	0.12	100	0.08
Crysene	0.05	100	0.04

as the RME used presently accounts for 5–10% of the operating costs.

Optimization of the gasification process

The experience gained from both the GoBiGas plant and the Chalmers gasifier has led to a comprehensive picture of how best to optimize the DFB gasification technology for the production of advanced biofuels. The optimization measures are founded based on a thorough understanding of the carbon balance of the Chalmers gasifier. This has been possible thanks to the advances in raw gas characterization methods, which include: He tracing to derive the total yield of gas [17]; Gas Chromatography for measurement of the concentrations of permanent gases; and SPA method for characterization of aromatic tar [18]. More recently, the development of a High Temperature Reactor (HTR) provided an independent method to quantify the total carbon in the raw gas [19]. The HTR enabled the closure of the carbon balance of the Chalmers gasifier, as well as the validation of an improved SPA method [20].

The main aspects of the optimization discussed here are related to: the solid circulation of the process; ways to control the gas quality of the process; improvements to the cooling and cleaning of the product gas; ways to

optimize the chemical efficiency of the gasifier; and the design of the fuel feeding.

Solid circulation in a DFB gasifier

The material flows in indirect gasifiers, built according to the example of the Güssing plant, employ two material cycles, as illustrated in Figure 4. In the figure, the material flows are distinguished as the *primary circulation* of bed material between the gasifier and the combustor (blue), and the *secondary circulation* of ash fractions (green and orange).

The primary circulation is the flow of bed material between the gasifier and the combustor through a seal in the bottom of the gasifier and a cyclone at the exit of the boiler with the main function of using the bed material as heat carrier, catalyst and carrier of reactive species. At GoBiGas, the bed material consists of olivine with a particle size distribution between 180 and 500 μm. The circulation flow of bed material depends mainly on the gas velocity through the bed section of the combustor [21]. It therefore depends on the fuel load of the system and the amount of air added to the system. A slight adjustment of the flow can be achieved by adjusting the quotation between primary and secondary air but not
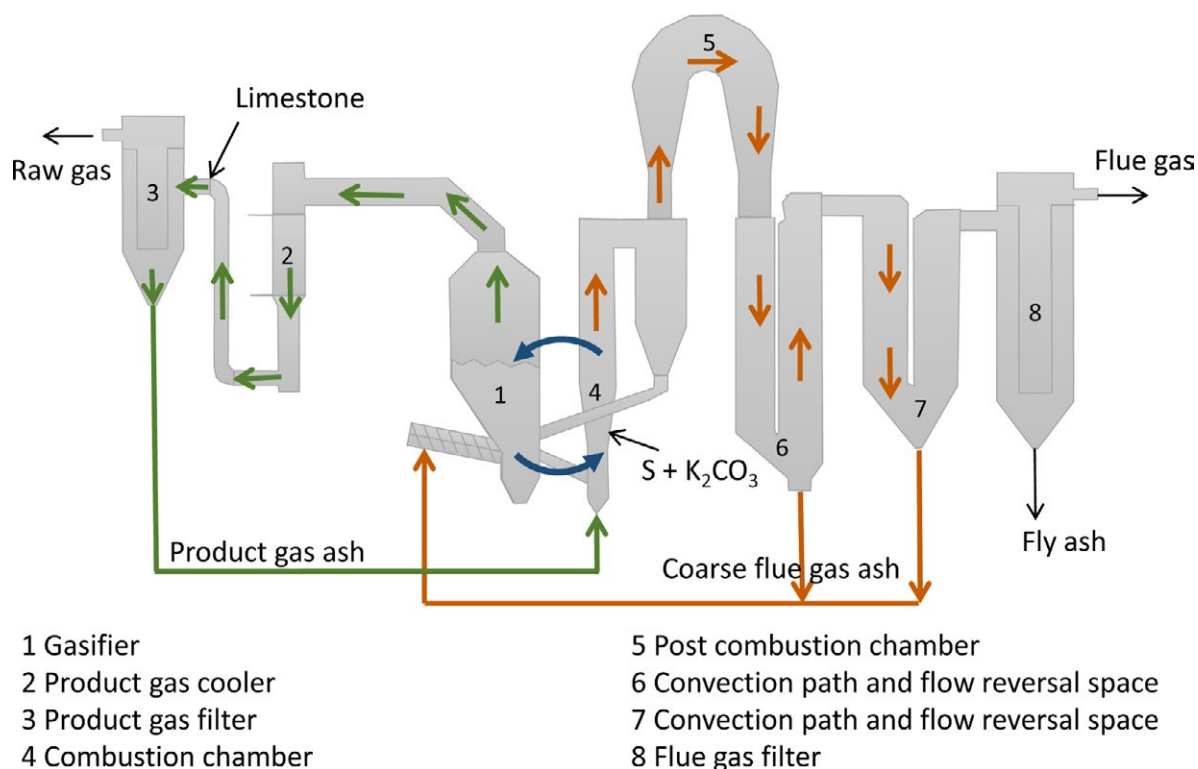

Figure 4. Illustration of the flows in the gasification section of the GoBiGas plant of the bed material (blue), product gas ash (green), and coarse flue gas ash (umber).

enough to keep the circulation flow constant as the load is changed in the system. Thus, both the heat transfer and the transport of active components change with the fuel load and this can limit the operational window of the process. For example, both the excess air ratio and the temperature difference are coupled to the load of the combustor.

To circumvent the limitations cause by changes in the bed material flow and to improve the process control, recirculation of flue gas to the combustion chamber should be included. This possibility is available in the Chalmers system where circulation flow of bed material can be controlled much more specific and the effect of changing the bed material flow has been investigated [17].

The secondary circulation consists of the recirculated material that is trapped in the product gas filter (3) and the coarse ash from the flue gas train. The ash in the product gas is rich in carbon (around 10–15%mass), so it is reintroduced into the combustor for carbon/energy recovery. The other main constituents of the product gas ashes are entrained bed material particles (≤100 μm in diameter), the limestone added as precoat material for the filters, and biomass ash. The ashes from the two flow reversal space (6, 7) are recycled to recover the entrained bed material (mean particle size >100 μm). The main functions are the recovery of entrained bed material, important ash components, and the carbon in the product gas ash (PG-ash). Since the commissioning of the GoBiGas plant, both the bed material and the ash chemistry have been optimized to achieve a high gas quality.

As mentioned above, all the heat needed for fuel conversion in the gasifier is provided by the primary cycle of the bed material, which transports the heat from the combustion side to the gasification side, while at the same time facilitating the return of the unconverted char fraction with the bed material to the combustion side. With conventional circulating bed systems, the experience is that it is favorable to minimize the size of the bed particle so as to avoid erosion and to create a high circulation flux. However, for a combined bubbling and circulating bed system, the gas velocity in the bubbling bed needs to be considered, so as to avoid entrainment of the bed material in the gasifier. As the gas velocity in the combustor is determined by the designated air ratio, a larger particle size distribution of the bed material results in a low circulation flux and, consequently, in a large temperature difference between the two reactors, and *vice versa* for a smaller particle size distribution. This coupling could be reduced by applying flue gas recirculation to the combustion section in a commercial-scale gasifier. The bed material at GoBiGas was changed in Year 2016 from Austrian olivine (pretreated olivine,

100–800 μm; Magnolithe GmbH) to Norwegian olivine (Vanguard 180–500 μm; Sibelco). The main reason for this change is related to reduced transport costs and avoiding energy-intensive pretreatment, which affect both the overall cost and CO_2 emissions related to the bed material. Furthermore, the new olivine had a narrower and on average smaller particle size distribution of 180–500 μm compared with the 100–800 μm of the previous olivine. No significant effect on gas quality was linked to the change of bed material, although the ash components were affected, as will be discussed in *Controlling the gas quality using potassium*.

An unwanted effect of the bed circulation that is often overlooked is that all the bed material used in a DFB system will eventually transfer a certain amount of oxygen from the oxygen-rich combustion reactor to the reducing gasification reactor, either through oxidation/reduction of the bed material itself or ash components attaching to the surface of the bed material [22, 23]. This will result in oxidation of some of the produced gas in the gasifier, and for the process, an increase in the amount of CO_2 that needs to be separated in the synthesis process, most likely via the energy-intensive amine process. Therefore, when considering the whole process from fuel input to synthesized fuel as the output, precautions need to be taken to minimize the oxygen transport.

As expected, when particles of a smaller average size distribution are used there is a noticeable increase in the circulation flux, as was also observed when the olivine was changed from the Austrian to the Norwegian version. For instance, in the Chalmers gasifier, it was shown that this affects the circulation flux of the bed material, which in turn affects both the heat transfer and the oxygen transport [24]. Furthermore, the chemical balance of inorganic species between the gasifier and combustor is affected by the bed material circulation flux, as the rates of uptake and release of species that mediate the catalytic activity will differ at different temperatures. This means that both the overall temperature level in the gasification process and the temperature difference between the combustor and the gasifier are parameters that can be used for optimization of the process. In particular, a lower overall temperature and smaller temperature difference between the combustor and the gasifier will have positive effects on the thermal efficiency of the process, although a small temperature difference between the combustor and the gasifier entails a high circulation flux and increased oxygen transport, which lower the chemical efficiency of the process.

Controlling the gas quality using potassium

A conclusion from the commissioning of the GoBiGas plant is that, for a cooler operating at temperatures <200℃

(390°F), it is crucial to produce a gas of sufficient quality that can be handled by that cooler. Such gas quality can only be achieved if the bed material has a sufficiently high catalytic activity. Complete catalytic conversion of volatiles is, however, unlikely due to mixing limitations.

Previous experiences with smaller demonstration units, for example, the plants in Güssing, Oberwart, and Senden [8, 25], which cumulatively have been in operation for more than 100,000 h, indicate that catalytic activation of the bed is achieved with continuous operation. However, during the commissioning of the GoBiGas plant, which took place at the end of Year 2013 and beginning of Year 2014, it was not feasible to operate the gasifier continuously for a sufficiently long period to obtain a catalytically active bed without clogging the cooler.

This focused the work in the Chalmers research gasifier to finding a solution, whereby the bed could be artificially and sufficiently catalytically activated. From the evaluation of the results obtained from the experimental campaigns conducted between Year 2007 and the beginning of Year 2014, it became clear that it was necessary to saturate the system with potassium. The amount of potassium carbonate (K_2CO_3) needed to saturate the bed material and the refractory material in the demonstration plant was estimated at about 1000 kg, which was the quantity that in April 2014 was dissolved in water and pumped into the combustion side of the GoBiGas gasifier. This resulted in the first successful operation of the demonstration plant with a gas composition and a load of larger polyaromatic hydrocarbons (PAHs), which were handled without clogging the cooler.

Since then, much effort has been expended toward attaining a better understanding of the activation process and ways to control the chemistry in the gasification process [26]. Based on the many experiments performed in the Chalmers gasifier and during the operation of GoBiGas, our current hypothesis is as follows: in the combustor, the bed material becomes saturated with potassium, enabling potassium salt to form on the bed material and the calcium layer formed on the surface of the bed material enhances the storage capacity of such salts. The bed material together with the potassium salts enters the gasifier, where these components are then released through exposure to steam and a reducing atmosphere. As they are released, they transform and interact with the radical pool, providing the necessary catalytic activity. A side-effect of the increased levels of potassium is a dwindling CO-burnout on the combustor side, which has been observed in a context of high catalytic activity [26].The addition of sulfur or silicon to the combustor removes the effect on the burnout. Sulfur also reinforces the catalytic effect on tar formation, in contrast to silicon, which eliminates this effect.

Figure 5 shows a typical example of how the concentration of unconverted CO in the flue gas of the Chalmers boiler increases as the activity of the olivine bed increases. The addition of a continuous flow of 1.0–1.2 kg/h (i.e., 1 mg/kg dry fuel) of elemental sulfur to the bed immediately confers complete combustion of CO, while the CO emissions increase progressively when the sulfur supply is terminated.

The amount of potassium salts on the bed material can be quantified by leaching in water, followed by analysis of the leachate. Figure 5 depicts the relationship between the level of water-soluble potassium in the bed material particles and the catalytic activity of the bed, which can be qualitatively assessed based on the tar concentration of the product gas and the level of CO emissions in the combustor. Here, the drop in the level of tar and the increase of CO emission from the boiler are seen over a very narrow range of concentrations of leachable potassium, leading to the interpretation that the bed material becomes saturated. The active components exit together with the gas from the gasifier and condense as salts in the convection path. The salts are collected in the filter, wherefrom they are returned to the combustor. In the combustor section, alkali components are once again made available for reactions, that is, they are adsorbed or absorbed onto the bed particles to the saturation level, while the remaining excess of alkali is collected in the filter and removed from the system with the fly ash.

As potassium is an essential catalytically active component, the selection of bed material needs to be made with some care. The main criterion is to avoid an excess
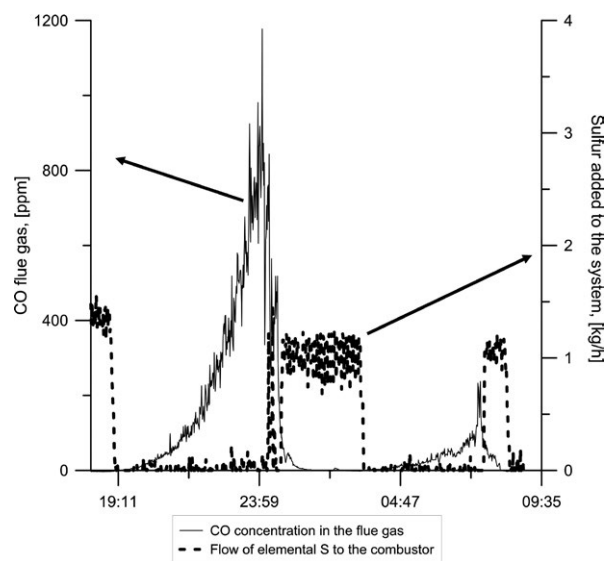

Figure 5. CO emissions from the boiler. Elemental sulfur was added to mitigate the incomplete burning of CO. The test was conducted in the Chalmers gasifier with wood as the fuel on a bed of Norwegian olivine.

of free silica, which will capture the active potassium very efficiently and permanently as silicates, thereby diminishing the activity of the bed material. Figure 6 summarizes the evidence for the loss of activity of a bed of olivine when a silicon-rich material (silica sand) is added to the system. The tar concentration increased 1.7-fold after the addition of 600 kg of silica sand to a bed of 3 tonnes of material in the Chalmers system, as compared to the tar concentration before the addition. Note how the CO burnout in the boiler also improves with the addition of silica sand, as the catalytically active potassium reacts with the silica. When this happens, not only the catalytic activity will be diminished, but also the bed particles will start to agglomerate due to the formation of low-melting-point eutectics of potassium silicates. Therefore, care should be taken to avoid silica sand, either as a bed material or as inert impurities (e.g., soil) in the biomass feed, in cases where the biomass was not handled correctly before being delivered to the plant. Favorable bed materials that can be used in the process are olivine, alkali feldspar, and low-iron-content bauxite [13, 22, 24].

It is crucial to identify the window of operation within which the appropriate proportions of potassium, sulfur, and calcium, together with the aging bed material give the optimal level of catalytic activity. Too little potassium or sulfur leads to too-high levels of large PAHs (*cf.* Fig. 7, *Tar-CO-leachable potassium*), while too much potassium leads to a large fraction of potassium salts in the gas. In both situations, the heat exchanger will become clogged
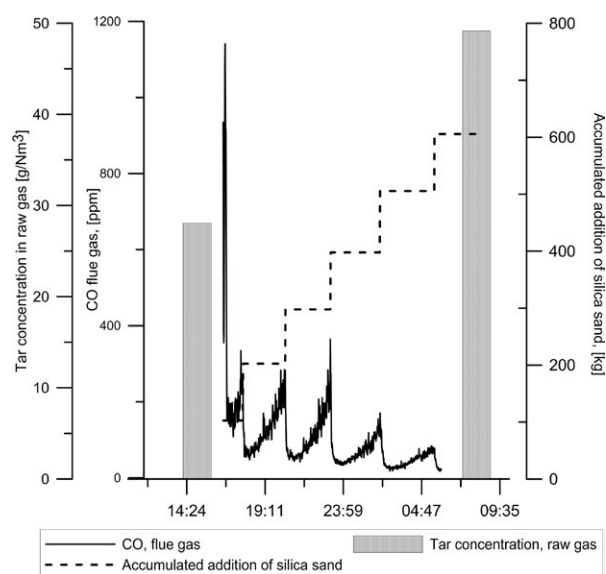
and eventually cause the process to stop. The key to creating a stable process is, therefore, to exploit the flexibility offered by the process design to control the levels of the above-mentioned species.

Data from GoBiGas have shown that there is a correlation between the tar concentration and the methane concentration in the dry gas [5]. Measurements obtained from the system at Chalmers have shown that the fraction of carbon in the fuel that is converted to methane is close to constant, regardless of any operational changes. However, a more active bed increases the conversion of both PAHs and char to mainly H_2 and CO, as well as increasing the shift from CO and H_2O to H_2 and CO_2, thereby reducing the dry methane concentration and providing an online indication of the activity of the bed. The correlation is illustrated in Figure 8, and it is clear that different fuels yield different correlations. Based on operational experience, limitations with regard to the methane concentration are established, as indicated by the dotted lines in Figure 8, where operation with a higher concentration of CH_4 yields tar deposits in the cooler, mainly reducing the heat exchange capacity, whereas a lower concentration of CH_4 yields alkaline deposits at the entrance of the cooler, mainly increasing the pressure drop.

The strategy that is currently applied to control the gas quality is described as follows. When the concentration of methane gradually increases, this indicates that
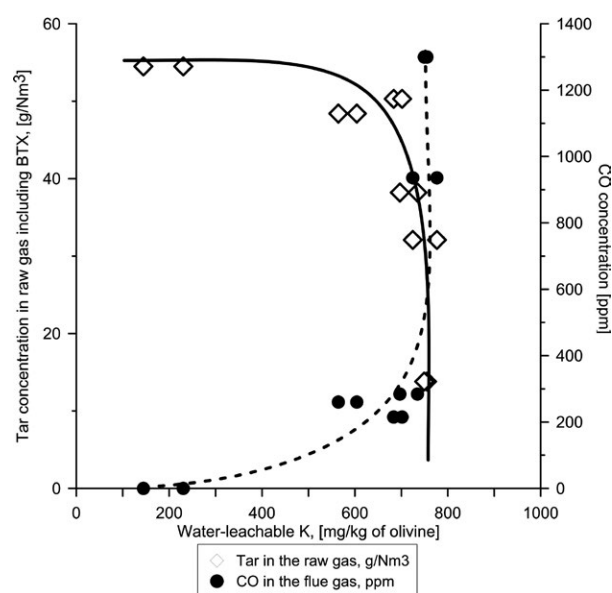

Figure 6. Tar concentrations (including BTX) before and after the addition of 600 kg of silica sand to a bed of 3 tonnes of olivine. The silica sand was added in six steps, and the corresponding CO emissions in the combustor side are shown. The tests were conducted in the Chalmers gasifier with wood as the fuel.

Figure 7. Relationship between the levels of water-leachable potassium in the bed material transferred from the combustor to the gasifier and the signs of catalytic activity (i.e., the levels of CO emissions in the combustor and the tar concentration in the raw gas). The tests were conducted in the Chalmers gasifier with wood as the fuel and Norwegian olivine sand as the bed material.
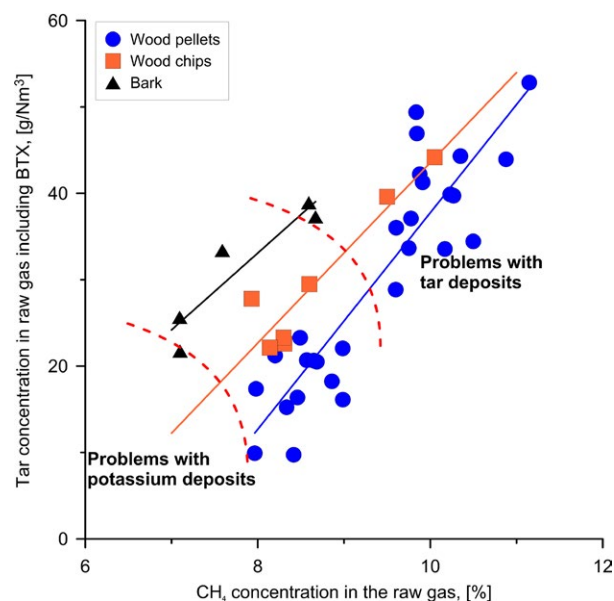
Figure 8. Correlations between the concentrations of methane and tar for various fuels and limits, for which actions related to bed activity are taken. Data from the GoBiGas plant.

the activity of the bed is reduced. In this case, the addition of potassium carbonate to the combustion chamber is increased. If further activation is required, recirculation of particles from the flue gas can be increased, or elemental sulfur can be added to the combustor as well. In a situation in which the dry concentration of methane drops to a level that is too low, the activity of the bed needs to be reduced to prevent the potassium salts falling out as deposits upon the heat exchanger surfaces. In this case, the flow of potassium to the combustor and the recirculation of fines from the flue gas to the gasifier are reduced. If the increased recirculation rate is not sufficient, bed material is removed and replaced with new material. While wood chips and pellets often require addition of alkali, forest residues and bark are self-sustaining, provided a low content of silicon carrying impurities.

Nevertheless, as shown in Figure 8, the limits within which the bed activity needs to be controlled are fuel-dependent. Therefore, the functionality of the strategy will rely on the ingoing fuel having a reasonably stable composition. However, biomass and waste, which are the intended fuels for the technology, are by their nature heterogeneous, so controlling the composition is not a viable option. To overcome this limitation, online measurements of tracer species that indicate changes in the tar concentration/composition and alkali concentration are important, keeping in mind the linkage of low tar levels with high concentrations of alkali. Several developments aimed at achieving this are ongoing and have even been

tested within the context of the project. For online tar monitoring, spectroscopic methods, such as laser-induced fluorescence and UV-vis spectroscopy have demonstrated the ability to give reliable responses for a shift in the tar composition toward larger molecules [27, 28]. Even measurement after a high temperature conversion of a slip stream to assess the overall carbon amount gives information about the yield of condensable hydrocarbons [19]. For alkali species, spectroscopic methods are under development but are not yet ready for application [29, 30]. Surface ionization detectors for alkali species have been developed by different groups [31–33] and shown to be applicable for measurements.

As stated above, the ash in the biomass to some extent compensates for the loss of active components during steady-state operation. However, extra additives might be required during start-up or in the case of a fuel with an unfavorable ash composition. The start-up strategy currently applied in the GoBiGas plant is that the initial activation is carried out by adding calcine and potassium, while heating the system through the combustion of natural gas on the combustion side. Here, the present layout of the process entails a special challenge, as the gasification reactor during the stop phase and initial start phase is fed pure nitrogen, before it is turned over to steam. Operation without adding new ash components to the system leads to a gradual loss of potassium and sulfur from the system and need to be compensated to assure sufficient activity of the bed. The amount of supplementary potassium carbonate and elemental sulfur that have to be added to the bed before start-up has to be estimated based on the interpolation and extrapolation of experiences from previous start-ups. This procedure could be modified by incorporating a recycle stream of preactivated bed material and ash fractions [34].

The combustion side of the process also serves as a regenerator of the catalysts in the system, in which the bed material and ash components are partially oxidized. Furthermore, the bed material takes up the ash components that provide the catalysts for the gasifier in the combustor, which also include supplemented ash components, such as potassium, sulfur, and calcium [35, 36]. For instance, net transport of sulfur from the combustor to the gasifier has been observed in the Chalmers gasifier. Figure 9 shows the levels of sulfur that originated from the H_2S in the raw gas in the Chalmers gasifier during tests conducted with an ash-free fuel, as well as a test conducted with wood pellets when the process was operated with olivine or silica sand as the bed material. For the case in which wood pellets were gasified, the difference in sulfur concentration between the raw gas and the fuel feed proves that the silica sand and olivine bed materials can transport sulfur from the combustor to the gasifier. This was further confirmed
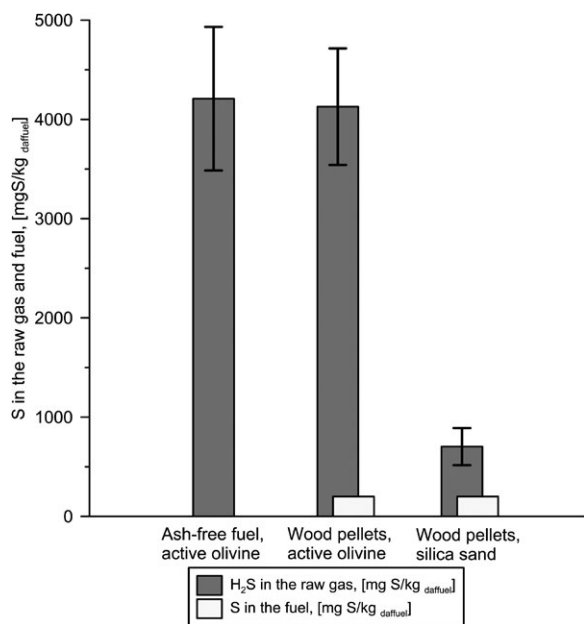
Figure 9. Sulfur transport and release from the bed material (olivine or silica sand) in the gasifier, as well as the levels of sulfur in the fuel (ash-free or wood pellets) fed to the gasifier.

in the test that involved gasification of the ash-free fuel, where the H_2S measured in the raw gas could only originate from the bed material coming from the combustor. Similar transport phenomena for inorganic species, in particular potassium, transferring from the combustor to the gasifier, have been observed in the Güssing plant [35].

In summary, the gas quality is mainly a function of the catalytic activity in the DFB system, which is controlled by:

- the primary circulation of the bed material;
- the secondary circulation of recirculated particles;
- the addition and removal of the bed material;
- the fuel-ash temperature levels of the bed materials in the gasifier and combustor;
- the steam-to-fuel ratio and fuel load; and
- artificial supplementation with potassium, sulfur, and calcium.

Cooling and cleaning of the product gas

Controlling the cooling of the producer gas from a steam-blown gasifier is challenging because of the impending risk of tar fouling on the heat exchanger surfaces. The fouling of heat exchanger surfaces is a well-known industrial problem globally, and it has been estimated to cost industrialized countries approximately 0.25% of their GDP [37]. One approach to mitigating physically the

fouling without changing the overall heat exchanger design or operation is to modify the interactions of the deposit-forming precursors with the heat transfer surface [38]. By taking advantage of recent developments in coating technology, thin and stable coatings that involve ceramics can be produced that outperform the previous generations of pure fluoropolymer coatings [39]. These functional coatings can be made both erosion-resistant and oil- or water-repellant, and they are already used commercially, for example, in the oil and gas industries [40].

A major milestone in progress toward improving product gas cleaning was reached in March 2017 when it was shown an RME scrubber could be replaced by a regular plate heat exchanger with coated surfaces, upon which both steam and aromatic structures with two or more rings could be condensed. Testing was performed using a slipstream of product gas from the Chalmers gasifier, which was cooled in a down-scaled plate heat exchanger. Validating tests was performed in July 2017 at the GoBiGas-plant. This proof-of-concept work, as illustrated in Figure 10, represents a major breakthrough for the technology, as it allows the removal of an otherwise costly and troublesome process unit. The coated heat exchanger plates used in the experimental evaluation in the Chalmers gasifier are inspired by the concept of self-cleaning surfaces [41], in which modifications of the surface chemistry and roughness create a situation in which the liquid water flow that results from the condensing steam effectively keeps the surface clean [42, 43]. The functionality is highly complex, arising from the interplay of the molecular and continuum properties of the gas–liquid mixture and the coated plates. Furthermore, the corrugation of the surfaces of the plates has been shown to affect the flow at both the macro- and microscales [44].
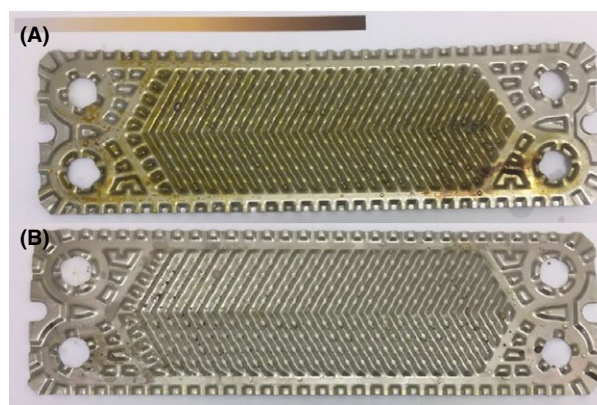

Figure 10. Comparison of a conventional (A) and a novel, coated (B) heat exchanger plate after several hours of exposure to a raw gas side stream in the Chalmers research gasifier. The degradation of the uncoated plate by a yellow-brownish tar residue is clearly visible. In contrast, the coated plate has remained virtually unaffected.

The tar concentrations to which the heat exchanger was subjected during the tests in the Chalmers gasifier are summarized in Table 1, where they are also compared to typical tar concentrations in the gas before the RME scrubber at GoBiGas, and an indication is given as to how well these components are removed in the RME scrubber. The components are presented in order of increasing dew point, starting with benzene, which has the lowest dew-point, and ending with chrysene, which has the highest dew point. The levels of performance of the RME scrubber for the most relevant hydrocarbons are presented in Table 1, which shows that the scrubber efficiently removes components with higher dew points than naphthalene, while a significant fraction of the naphthalene and almost all of the more volatile components remain in the product gas. The remaining components are removed by active carbon beds, see *Bridging processes*. The more components with higher dew points than benzene, such as naphthalene, that enter the carbon beds, the more difficult it becomes to regenerate the beds. Therefore, any new or existing systems need to be considered together to optimize the overall gas cleaning. Improved control of the distribution of tar components via the operation of the gasifier might also offer the possibility to lighten the burden on the carbon beds by producing a tar that is more easily removed in the plate heat exchanger unit.

Optimizing the chemical efficiency of a DFB gasifier

Typical gas compositions and energy fractions from the GoBiGas demonstration operating with wood pellets as fuel are shown in Table 2. Based on the chemically bound energy in the gas, as distinct from the chemically bound energy in the fuel, it is evident that the overall reaction in the gasifier is endothermic, showing an increase in chemical bound energy of 2.3%. In the present GoBiGas system, the heat for the gasification process is provided by the combustion of char, larger polyaromatics, and part of the dry product gas. The BTX (Benzene, Toluene, Xylene) species and the part of the naphthalene that is not captured by the scrubber are at present combusted in the postcombustor at GoBiGas. This represents a loss of more than 3.5% of the chemically bound energy. Thus, to increase the efficiency, these components should be better utilized (see the *Bridging processes* on the removal of BTX). Furthermore, the heat demand of the process should be minimized, so as to optimize the chemical efficiency of the gasifier. This can be achieved by: applying reactor walls that lower the heat losses; preheating the ingoing air and steam streams to higher temperatures; decreasing the overall gasification temperature; lowering the moisture content of the fuel; and preheating the ingoing fuel. Through these measures, the heat demand could be reduced by as much as 20% in a future

Table 2. Wet and dry gas compositions and energy distributions for a gasifier operated with wood pellets and a bed temperature of 870°C. The data shown are extracted from validation experiments at GoBiGas [3].

Component	Wet gas %$_{vol}$	Dry gas %$_{vol}$	Energy %$_{daf}$[1]
H_2	27.7	39.9	30.1
CO	16.6	24.0	21.3
CO_2	13.8	19.9	
CH_4	6.0	8.6	21.4
C_2H_4	1.4	2.0	8.2
C_2H_2, C_2H_6, C_3H_6	0.2	0.3	1.4
Inertization gas (CO_2; during production), N$_2$ (validation experiments))	3.7	5.3	
H_2O	30.2		
BTX[2]	0.3		3.5
Tar[3]	0.1		2.1
Char			14.3
		Sum	102.3
Potential CO and H_2 production from remaining char			19.0
	kg/kg$_{daf}$	mol/mol$_{stoic}$[4]	Loss of chemical bound energy %[5]
Oxygen transport	0.07	0.049	5.3

[1]Dry ash-free fuel (daf).
[2]BTX represented by benzene.
[3]Tar represented by naphthalene.
[4]Stoichiometric combustion (stoic).
[5]Calculated from the average gas composition.

commercial plant, thereby increasing the chemical efficiency of the gasifier to the same extent (for more details see [45]). In addition, by introducing an alternative external heat source, for example, direct heating by electricity, the energy content of the produced gas could be increased even further, up to about 120% compared to the chemically bound energy of the ingoing fuel [11].

Given the relatively narrow time window for the release of volatile matter and the typical rates of lateral mixing of fuels in fluidized beds, the layout of the process provides, at all times, for scales larger than laboratory units and sufficient residence times for the biomass to be pyrolyzed in the gasification chamber. As most of the chemically bound energy in the biomass is stored in the volatile fraction of the fuel, the volatiles will, therefore, represent the major share of the gas released on the gasification side of the process. However, for an optimized biofuel plant, part of the produced char needs to be gasified, so as not to release too much heat on the combustion side of the process.

To control the extent of char conversion in the gasification chamber, several alternative methods are available. The most intuitive option is to increase the overall residence time for the char and the circulated bed material by increasing the volume of bed material in the gasification reactor (given by the cross-sectional area and the bed height). However, the optimal height of a fluidized bed is in the range of 30–60 cm, as the bubbles in taller beds will coalesce to form larger bubbles, which greatly reduce the gas-solids contacts and confer no further benefit in terms of the overall performance of the mass transfer [46]. Furthermore, the pressure drop across beds that are taller than 60 cm requires a specially designed, high-pressure drop gas distributor to ensure good fluidization and to avoid partial defluidization of the emulsion phase located in-between the bubble paths [47]. Similar problems will be experienced following the introduction of tapered walls into the reactor along the height of bubbling bed, which is the case for the present design of the GoBiGas gasifier, as the bed located in the region of the tapered walls will not be properly fluidized or even defluidized.

For the GoBiGas process, if this issue is not addressed sufficiently, it can create a very unfavorable flow profile and suboptimal mixing conditions. An alternative method for increasing the inventory of bed material in the gasifier is to increase the cross-sectional area, which is not an attractive option, as it increases the cost of the plant. A more straightforward strategy to increase the residence time in the gasifier is to increase the temperature difference between the combustor and the gasifier, thereby increasing the heat-carrying capacity and allowing the process to operate with a lower volumetric flow of circulating solids. However, this option also has its limitations, as the temperature difference between the combustion and gasification reactors is restricted to around 100°C, so as to maintain the process below the temperature at which there is a risk of agglomeration.

The remaining options are either to modulate the rate of char gasification or to decouple the residence time of the char from that of the bed material. We start with changing the reaction rate of the char gasification, which is significantly increased by the potassium released from the activated bed material on the gasification side and that diffuses into the char particles [48]. In the demonstration plant, this effect is substantial, where the char that is converted is around 50% and will be sufficient to fulfill the mass balance for most applications [11]. If there is a need to increase the char conversion beyond a level that can be realized by the catalytic activity, the significant differences in density and size between the bed material and the char offer the possibility to decouple the mixing, and thereby the residence time, of the char from that of the bed material. This can be achieved by optimizing the pressure drop over the distributor plate in combination with the fluidization flow and fuel size and shape, such that the char particles float on the bed surface (see Figure 11), and by physically creating an enclosure for the char particles using, for example, baffles [49].

Design of the fuel feeding to the gasification section

The fuel feeding system warrants close attention in the quest to obtain a process with high availability. Here, there is a strong influence of scale, and the experience

Figure 11. Photograph of fuel particles (black dots) floating on the surface of the bubbling fluidized bed in the research gasifier operating at 820°C (1500°F) and using wood pellets as fuel. Source: Erik Sette, Rustan Marberg, Chalmers University of Technology.

from laboratory-scale fluidized bed gasifiers is of a preference for mechanical in-bed feeding over feeding the fuel by gravity fall on-top of the fluidized bed. The same experience was obtained for fluidized bed combustors in the late 1970s and the beginning of the 1980s. To investigate if this also applied to industrial units, the first industrial-sized (16-MWth) bubbling fluidized bed combustion system installed at Chalmers in 1982 was equipped with both in-bed and on-top feeding. The experience from that exercise, which remains unpublished, but has been transferred to and adopted by the major boiler manufacturers [50], was the opposite of the experience with the laboratory units. Here, feeding by dropping the fuel onto the bed surface always gave better results than in-bed feeding.

If there is an ambition to drag the fuel down into the bed in an industrial system, where the cross-sectional area allows fully developed fluidization, it is much more advantageous to increase the fluidization velocity in the area within which the fuel feed is located. Fuel that is fed into the bed via an in-bed feeding system rises very rapidly to the bed surface and remains there, as a consequence of the density difference and the released moisture and volatile matter, which create so-called 'endogenous bubbles' around the fuel particle, helping it to rise towards the bed surface [51]. Despite this knowledge, the technology provider of the demonstration gasifier was not willing to change the design, as the gasifier in the GoBiGas plant was scaled up from the existing unit in Güssing. That decision has created significant problems related to availability for the current unit.

The availability problems related to the in-bed fuel feeding system have to a large extent been related to the transport of heat by the bed material into the fuel feeding system (see Figure 12), which causes the fuel to pyrolyze inside the screw. The tar produced in this region will condense and start to build up inside the screw, which eventually results in a momentum that is too high for the engine driving the feeding screw and eventual blockage of the fuel. This has not been as big a concern in

the smaller units in Güssing and Oberwart in Austria as in the GoBiGas demonstration plant, which to a large extent is attributed to the use of a fuel with a relatively high moisture content (typically around 20%), as compared to the fuel with a moisture content of 8% used in the demonstration plant. The higher moisture content introduces a cooling to the fuel-feeding system, as the heat that is transported into the screw is absorbed by the heat of evaporation of the moisture. In addition, the larger geometrical dimensions of the fuel feeding system exacerbate this problem.

In the research gasifier at Chalmers, the fuel is dropped onto the surface of the bed by gravity, so the problem of heat transfer into the fuel feeding system is not an issue. The main motivation for getting the fuel into the bed is to assure good gas–solids contacts, as the catalytic activity was originally thought to be a heterogeneously catalyzed reaction that involved the volatiles and the coarse bed material. According to specific investigations in the Chalmers gasifier only 48–69% of the volatiles meet the surface of the bed material particles, while the rest of volatiles do not interact with the bed [52]. This means that, if gas–solid contact is required, full catalytic conversion of volatiles to syngas is not achievable, regardless the catalyst applied. Despite the different approaches, both the in bed feeding by screw (GoBiGas) and the gravimetric on-bed feeding (Chalmers) yield the same low levels of tar. As described above, the findings in the present work imply that with a potassium-activated bed material, homogeneous reactions play a significant role, beyond the importance of gas-solid contacts.

This was validated by changing the bed height in the GoBiGas gasifier during operation with a potassium-activated bed material. As shown in Figure 13, there were no significant changes in the gas quality, here indicated by the methane concentration, of the dry product gas. The notion that the fuel feeding depth has a negligible effect is further supported by the finding that an equivalent gas quality can be obtained in the research gasifier, where the fuel is dropped from the top of the gasifier down to

Figure 12. Left photograph: Cold flow model showing how the bed material is pushed into the fuel feeding screw. Source: Claes Breitholtz, Valmet AB. Right photograph: Feeding screw used in the GoBiGas plant, exhibiting mainly graphite-like deposits.

the fluidized bed, as in the demonstration plant, where the fuel is fed into the bed. Nevertheless, if there is a need in future reactor designs to force the volatiles into contact with the bed material to obtain the required gas quality this can be achieved by an integrated feeding chamber [53].

During 2016, a new external fuel feeding system was put into operation at the GoBiGas plant, which allows wood chips or bark as fuel. The experience to date is that after solving the initial mechanical issues, these fuels can be gasified in a satisfactory way if the fuel is predried according to specification. At the moment, the most serious problem connected to changing the fuel type is the variability in moisture content of the batches of fuel delivered to the plant (at present no dryer is installed at the GoBiGas project, as it was planned for the second commercial phase of the project). This emphasizes the importance of incorporating a dryer onsite, instead of relying on external drying capability. A similar experience was gained from the Güssing plant, leading to the incorporation of a dryer in the Oberwart and Senden plants.

Considering the vulnerability of the overall process, where a stop in the fuel feeding will result in several days production stop in downstream synthesis process, in combination with the well-known sensitivity of biomass-feeding systems, at least a second line is recommended for a future plant. This is especially important bearing in mind the general trend toward adopting low-grade fuels. Further, at least two feeding lines allow one to take full advantage of the co-gasification of fuels with chemical

synergy effects, for example, a char-rich fuel plus a fuel with low char content. Even though theoretically these fuels could be blended and fed together through the same port, their different flow properties, arising from shape, surface, and density differences, would create unwanted load fluctuations. Further, additional ports will improve the redundancy of the overall process.

Synthesis process

Methane synthesis is the standard way to produce synthetic natural gas from coal. This process is currently up and running in commercial plants at several locations in China and in the US. The biggest concern for the demonstration unit was its small scale and the risk associated with down-scaling the process units (typically by a factor of 10–100, compared to the scale that is usually applied in today's commercial units). A more detailed description of the synthesis process can be found elsewhere [54].

When pure syngas or hydrogen is the desired product, methane and ethane can be converted in standard operation units. This reaction is quite endothermic and needs a supply of heat at temperatures >800°C (1475°F), which can be provided by catalytic partial oxidation or catalytic steam reforming [55–57]. These units run preferentially at high pressure and using relatively clean gas, meaning that they are placed further downstream in the process chain. However, even the application of catalytic tar cleaning catalysts will decrease the amount of methane in the gas, as has been demonstrated previously [55]. In addition, coking and catalyst poisoning are issues of concern.

Nonetheless, looking at the gasification process as a steam reforming process offers more energy and most likely cost-efficient integration strategies for the production of mixed alkenes, methanol or mixed alcohols. The GoBiGas process is optimized for the production of pure methane, which means that the process was designed with the aim of keeping the methane concentration as high as possible in the gas entering the synthesis step. To achieve this, the char gasification with steam to CO and H_2 needs to be optimized so that the char leaving the gasifier just covers the heat demand for the process, so as to avoid dilution.

The temperature of the process can be controlled through the amount of char that is gasified, although this is impractical, as it is a slow process and the char reactivity of the ingoing fuel can vary over time. Instead, the process is setup to ensure that more char than is necessary is gasified, and the process is controlled by the recirculation of some of the cooled produced gas, as illustrated by Alternative 1 in Figure 14.

In the refinery and petrochemical industries, methane is, however, regarded as the hydrocarbon of least value
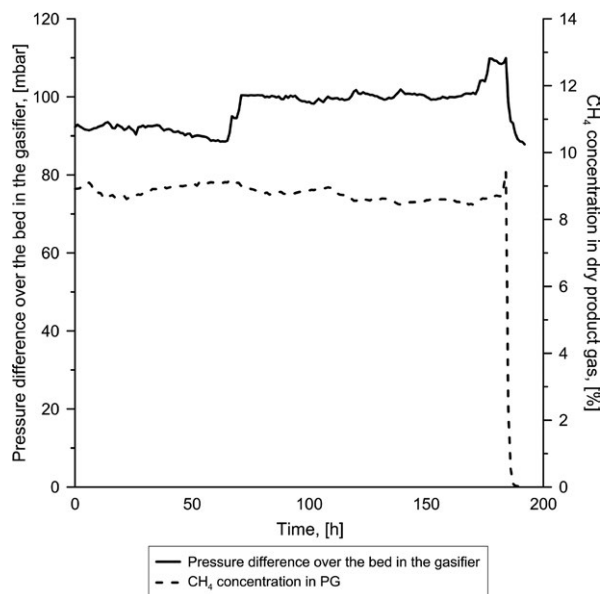

Figure 13. Methane concentrations and the pressure drops over the fluidized bed of the gasifier during 200 h of operation as the bed height was increased in two steps.

and it has a lower value than the syngas from which it is produced. To valorize the CO and H_2 in the produced gas, hydrocarbons of higher value should be produced. Thus, the integration strategy would change, in that as much char as possible should be gasified and as little gas as possible should be internally combusted by the oxygen that is transported by the bed material from the combustion to the gasification side of the gasifier. The latter occurs because the oxygen transported by the bed material to the gasifier will most likely favor the conversion of H_2 and CO over hydrocarbons, as observed previously [24]. In this way, the maximum amounts of CO and H_2 will leave the gasifier for the synthesis process.

In the synthesis processes shown for Alternatives 2 and 3 in Figure 14, the gas can be shifted efficiently to the preferred H_2/CO ratio, dried, and cleaned of excessive CO_2 before the gas proceeds to a second compression step that increases the pressure to the level designated for the intended synthesis process. Downstream of the compressor, it is preferable to have a one-step synthesis process that converts as much as possible of the CO and H_2 to the sought-after hydrocarbon, where methane and shorter alkanes act as inert gases. The gasification process is balanced by part of the off-gas from the synthesis

reactor, and the remaining off-gas is upgraded to methane, as methanation offers close to 100% conversion. The principal integration scheme is illustrated as Alternative 2 in Figure 15.

Examples of suitable one-step synthesis processes are methanol production, as demonstrated for several thousand hours of operation in the DME plant in Piteå using syngas from black liquor gasification [58] or the FT synthesis process developed and demonstrated in connection with the Güssing plant in Austria [59]. The one-step methanol process developed by Haldor Topsøe requires that the pressure in the second step is increased to 130 bar [58], which might offer a possibility to take out the produced methane produced from the off-gas in liquid form. In the FT process developed by Velocys in connection with the Güssing plant, the pressure is slightly raised to around 20 bar, and the synthesis is performed in a slurry reactor, providing good overall conversion of the gas from a DFB gasifier [59]. Both processes are module-based, and the modules operated in both demonstrations are of the same scale as the intended commercial scale.

If there is no market for the methane or there is an excess of electricity available for the process it would be beneficial to carry out steam reformation of the off-gas
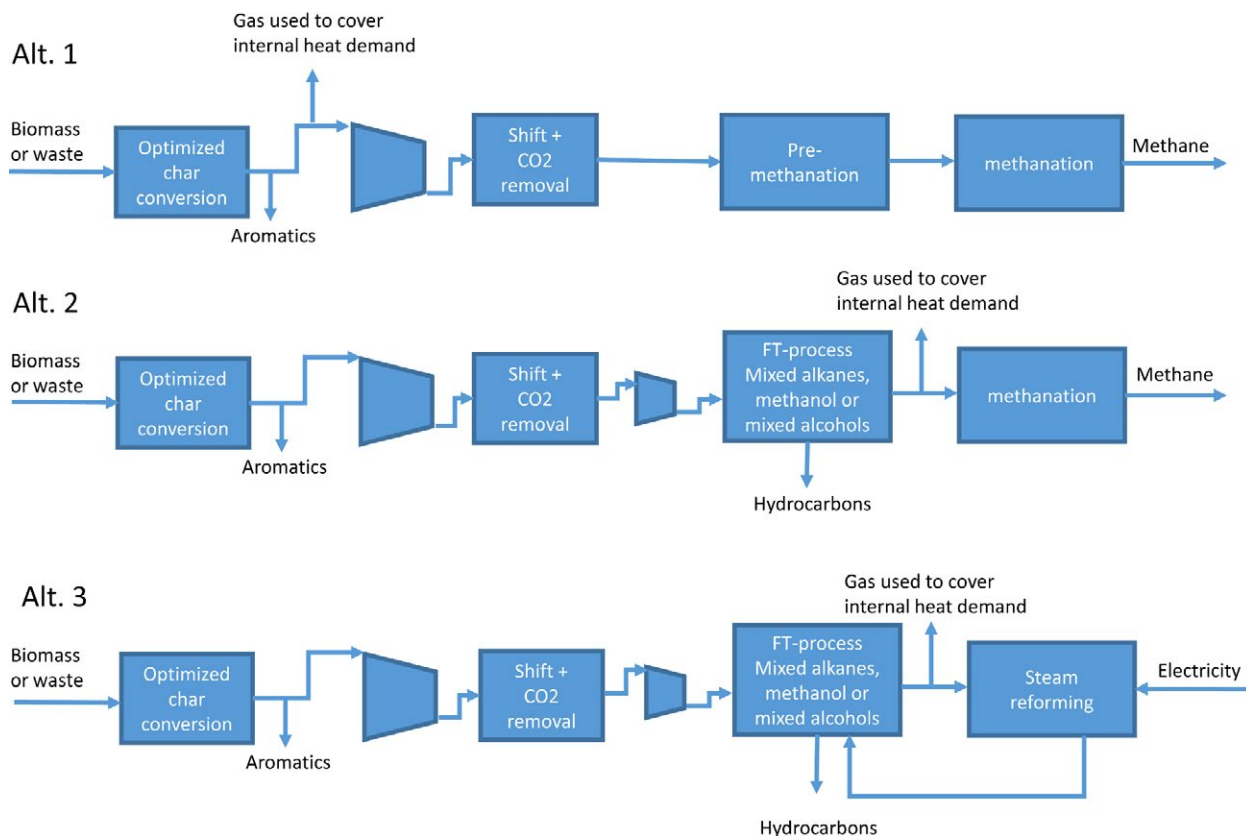

Figure 14. Different integration strategies for different end-products.
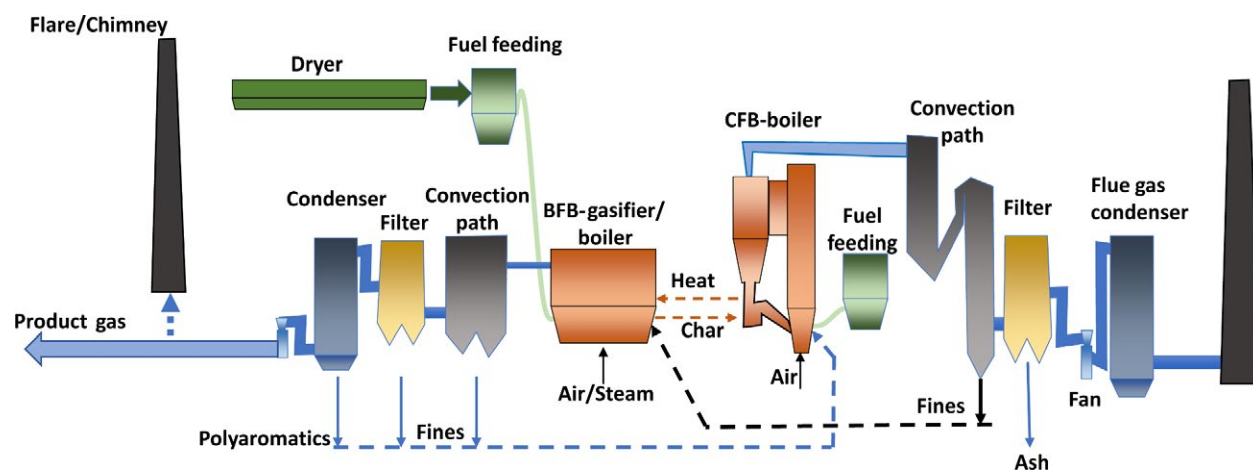
Figure 15. Integration of CFB and BFB boilers to create a dual fluidized bed gasifier.

from the synthesis to CO and H_2, rather than producing pure methane to increase the production of hydrocarbons of higher value, as shown in Alternative 3 in Figure 14. Here, extraction of the gas for heating the gasification process would be beneficial for the overall process, as it would minimize the accumulation of inert gases in the synthesis process, which otherwise would limit the amount of gas that it is possible to synthesize.

Bridging processes

The production of advanced biofuels via gasification merges two industrial branches that traditionally have developed their processes independently with very limited exchange of knowledge. Large-scale biomass gasification has mainly been developed in the energy sector, with the main goal being efficient production of electricity. The focus has been on solid fuel conversion to obtain a gas quality that at atmospheric or moderate pressures can be handled by gas turbines and gas engines, or simply to replace a burner in a regular boiler. In contrast, the synthesis processes are developed for gases that are refined to an extremely high quality and at high pressure. To connect the gasification process with the synthesis process, the "clean" gas from the gasification needs to be compressed and further upgraded to meet the requirements of the synthesis process. This imposes two costly additional steps:

- Compression
- BTX condensation and the removal of other impurities

We start with the compressor, which in the GoBiGas process is an integrally geared centrifugal compressor that takes the gas from atmospheric pressure to 16 bar in six consecutive steps. From the gasification process applied

in the GoBiGas demonstration unit, the produced cold gas contains a substantial amount of BTX and naphthalene, which together with small amounts of HCl, ammonia, and H_2S are preferentially separated from the gas upstream of the compressor. Typical compositions of the BTX for two different gasification temperatures are listed in Tables 1 and 2.

In the GoBiGas demonstration plant, the separation is performed in a configuration that comprises four active carbon beds, which can be operated in any order. Typically, the beds are operated as follows: the gas enters the first bed, which has as its main function the removal of all larger aromatic structures and impurities from the gas leaving the RME scrubber. Thereafter, the gas passes through one additional bed, where it removes the main part of the BTX and impurities, such as H_2S, before the gas leaves for compression. The other two parallel beds cycle between acting as the bulk BTX remover and being regenerated with steam.

The regeneration is performed using steam, and the off-gases are led to the afterburner of the combustor for destruction of the BTX components released during the regeneration. During the regeneration, the steam is condensed in the carbon beds, causing a gradual increase in the temperature of the carbon bed, such that the BTX components are released to the off-gases. This means that most of the BTX is released in a very short time period, as the propagating steam condensation front reaches the opposite side of the bed under regeneration.

As a consequence of the substantial quantity of BTX that is removed and the intermittent release of the individual species, a stability problem arises in the combustor. The released fuel will consume all the available oxygen, creating a large increase of combustion air required, which the operating system attempts to handle. When most of the BTX is released from the bed under regeneration, the

steam tends to cool the postcombustion, which converts unburnt fuel in the flue gas. Currently, this is controlled by the combustion of part of the product gas in the postcombustor during regeneration, which can be dynamically controlled to compensate for the rapid pulse of BTX. However, in this case, there is efficiency penalty in the order of 5–10 percentage points. As the regeneration is intermittent this also cause undesired variations throughout the hole process chain.

To resolve this issue, the steam and BTX components from the regeneration can be condensed instead of the mixture being sent to the postcombustor. By doing so, the BTX can be extracted, separated from the steam, and evenly fed into and combusted in the primary combustor chamber rather than the postcombustor, thereby creating a valuable heat source for the process. Furthermore, the significant cooling effect of the steam feed to the postcombustor can be avoided, so that the combustion of product gas is no longer required in the postcombustor. The separation also opens up the possibility for extraction and utilization of the BTX fractions as green aromatics sold as a product from the plant.

The designed regeneration strategy limits the regeneration temperature of the active carbon to 160°C (320°F), which means that the active carbon will not be fully regenerated, and this reduces the capacity of the carbon beds. To avoid a further reduction of the capacity due to components that are larger than naphthalene entering the bulk adsorbers and not being removed adequately via regeneration, the preadsorber is replaced in intervals. The duration of operation of the preadsorber is based on the product gas flow, the tar levels in the gas (which at GoBiGas depend on the performance of the RME-scrubber), and the temperature and pressure of the product gas.

At present, the preadsorber needs to be replaced every 2.5 months of full operation, and the used carbon is sent as waste for incineration. However, the removed active carbon can more or less be fully regenerated, if it is regenerated at a higher temperature, that is, 400°–500°C (750°–930°F). Therefore, if beds with active carbon are chosen for the removal of BTX in a larger plant it is recommended to include an external high-temperature regeneration step for the recovery of spent active carbon, or to optimize the tar removal prior to the carbon beds, so as to decrease the levels of components that are larger than naphthalene (see *Cooling and cleaning of the product gas*), and, thereby, the need to replace the activated carbon.

The active carbon beds actually exhibit the desired functionality, although due to some design errors, the capacities of the carbon beds in the present installation are too small. As the active coal bed system represents a complex and expensive installation, intensive efforts have been made to optimize the system beyond the design specifications. This has gradually allowed increases in the gas throughput, without compromising the gas quality. Presently, 93% of the design capacity of the plant is reached, and there are further suggestions as to how the capacity can be increased to allow the plant to reach full capacity.

The overall experience with the carbon beds is that it would be beneficial to cool the gas entering the active carbon beds to <30°C (86°F), to condense out more of the water and the remaining hydrocarbons in the product gas. Lower temperature will additionally result in an increase of the loading capacity of the activated carbon. Furthermore, a redesign of the process so as to incorporate a separate unit that would remove the BTX while operating at around 10°C (50°F) should be considered. This would create a smaller active carbon bed system that has the function of a guard bed rather than that of a temperature-swing adsorption system. In this context, the breakthrough of using coated heat exchangers is crucial, as it can reduce the temperature to the desired level in a cost- and energy-efficient way.

Material consumption and waste streams during operation of the GoBiGas plant

The material consumption and waste streams during operation are vital, as they affect the production costs of the plant. The levels of consumption of the different materials used at the GoBiGas plant during start-up and stable operation are summarized in Table 3. It takes about 32 h to heat up the process before the fuel can be fed into the gasifier, and it takes about 24 h for the gasifier to reach stable operation. Once stable operation of the gasifier has been established, methanization can be started, which takes an additional 60–80 h, and during this time the product gas must be flared. To ensure economic viability, the start-up time of the process makes it crucial to avoid starts and stops, meaning that efforts have to be made to attain high availability for the process.

For a new plant or the redesign of an existing plant, several improvements can be made to reduce the level of consumables, with the major improvements (details of which can be found in the sections above) being:

- the condensation of the regeneration steam and BTX components from the carbon beds, which would lower the heat demand of the gasifier and increase the efficiency;
- the feeding of solid fuel to the combustion side of the gasifier, which would reduce the need for natural gas during start-up;

Table 3. Summary of the consumables used during the start-up and during stable operation of the GoBiGas plant when operated at 80% of capacity.

Consumable	Unit	Start-up	Stable operation
Wood pellets	tonnes/h	0	6.2
Natural gas	m_n^3/h	450	100
Nitrogen	m_n^3/h	608	4
Olivine	kg/h	0	65
Rapeseed methyl ester (RME)	kg/h	0	70
Calcine	tonnes/h	1	0.11
Potassium carbonate solution	l/h	5	5
Ash	tonnes/h	0	0.3
Electricity	MW	1	2
Active carbon	kg/h	0	2.7
Waste water	m_n^3/h	0.7	0

- the introduction of an ash-rich fuel, such as bark or forest residues, which would eliminate the need for potassium carbonate supplementation;
- the introduction of coated heat exchangers, which would remove or minimize the need for RME;
- the introduction of a steam cycle, which would allow production of the electricity internally in the process; and
- the on-site, high-temperature regeneration of active carbon, which would minimize the need for replacement by at least a factor of 10.

Introduction of the Technology into the Existing Infrastructure

The GoBiGas demonstration plant and the research gasifier at Chalmers, together with the plants in Güssing, Oberwart, and Senden have contributed to establishing a comprehensive strategy for the design and control of biomass gasification for the production of advanced biofuels based on a variety of biomass sources. Collectively, they show how one can handle these types of processes during start-up and disturbances, which cause low availability and failures in many demonstration and pilot projects around the world. Here, it can be mentioned that the plant in Senden has reached an availability level of more than 6000 h/year, and with recent modifications taken forward in collaboration with the Technical University of Vienna, availability for 8000 h/year seems to be reachable.

A vital insight from the research summarized in this paper is that the ash components of the biomass itself, which are often considered as agents that make biomass problematic as a fuel, are instead the solution to some

of the problems. Furthermore, experience from both the demonstration plant and the research gasifier shows that the basic layout and temperature levels of a combustion reactor (including the convection path and filter) in a conventional fluidized bed setup designed for wet biomass corresponds to the desired design of a biomass gasifier. This means that a DFB gasifier can be constructed from two regular fluidized bed boilers placed next to each other, thereby providing a process that can be operated as a DFB gasifier or two parallel boilers (one CFB and one BFB) for biomass or waste.

The principal integration scheme for this type of system is shown in Figure 15, where the CFB system is a schematic of the Valmet boiler shown in Figure 2, complemented with a flue gas condenser. Starting from the CFB boiler, the recirculation of the bed material separated in the cyclone is directed to the BFB boiler instead of directly back to the combustion chamber (as is the case when the CFB boiler is operated as a boiler rather than as one of the interconnected reactors in a DFB gasifier). This flexibility can be ensured by the introduction of a particle distributor, as demonstrated in the Chalmers research gasifier (item 9 in Figure 16). The functionality is as follows: the pipe to the loop seal connecting the CFB boiler with the BFB boiler is located at a lower position than the pipe that is connected to the combustor. In the absence of fluidization in the loop seal, there is no flow of solids through the seal, which means that they build up the bed level in the distributor until it starts to flow back to the CFB instead. If the seal is fluidized the solid flow will go through the seal, thereby adjusting the bed level in the distributor below the height of the pipe to the CFB and directing the entire solid flow to the BFB reactor. To return the solid material to the CFB reactor from the BFB, there is a second loop seal, through which the solid material enters via a weir, whereby turning on or off the fluidization in the two seals controls the flow between the reactors, as further described elsewhere [60]. With this design, the CFB and the BFB systems can be operated as two separate boilers or as a DFB gasifier.

To operate the BFB reactor as a gasifier, the fuel has to be dried (as discussed above in *Description of and Results from the Technical Demonstrations*) and the combustion air needs to be replaced by steam. Furthermore, the circulation of the coarser fines, which in a regular fluidized bed boiler would be returned to the combustion chamber, should be redirected from the CFB reactor to the BFB reactor. From the BFB reactor, all flows of fines (both the coarser fines that in the boiler mode are returned back to the boiler and the finer fractions that are sent to the ash bin) should in gasification mode be redirected to the CFB reactor. This ensures the recirculation of ash components that control the chemical activity, as
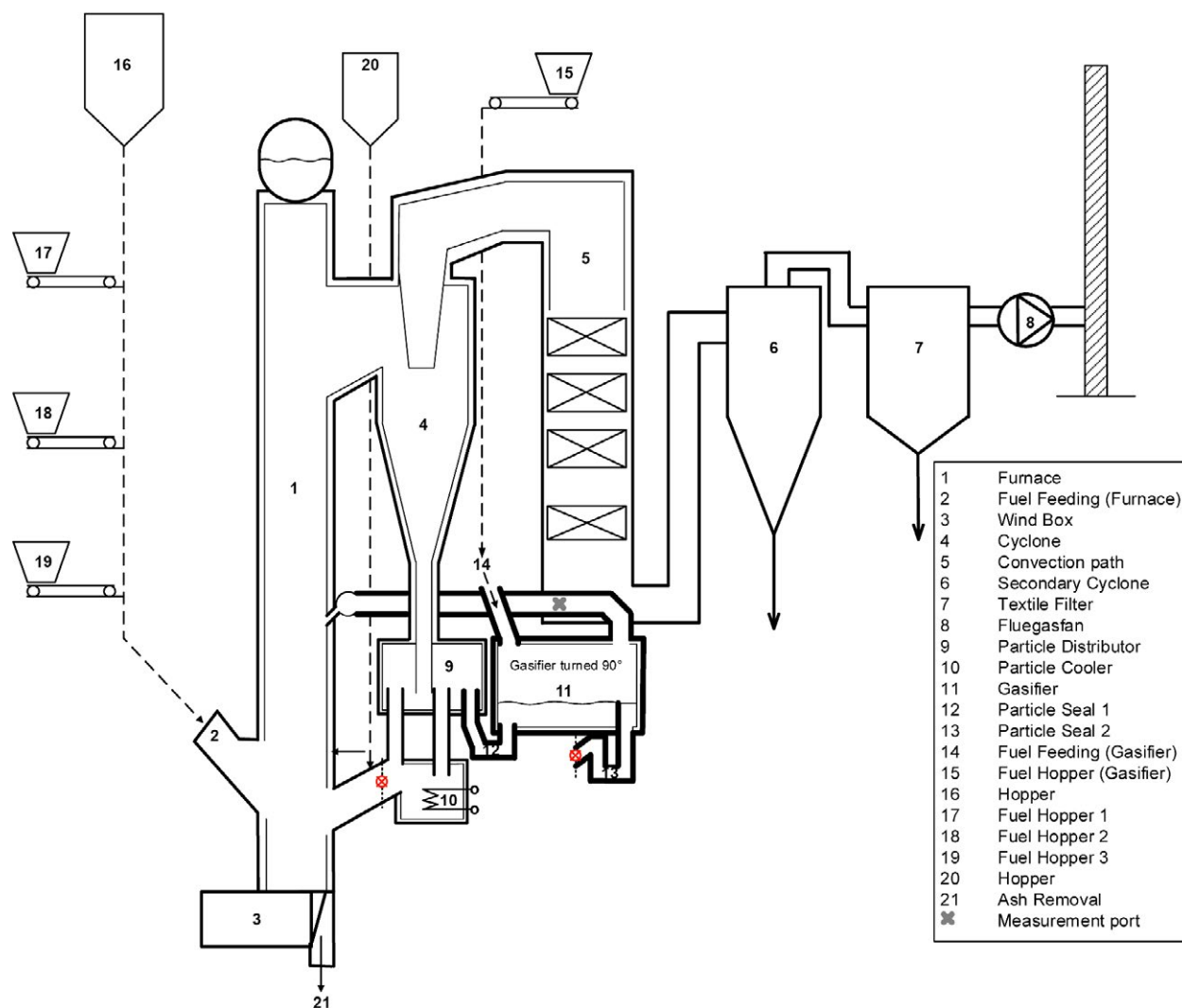
Figure 16. The Chalmers 12-MWth boiler – a 2–4-MWth gasification system.

well as the full conversion of the converted particles of char and soot, as well as tars that are captured in the filter cake. Furthermore, waste streams from the downstream cleaning steps will be directed to the CFB reactor for destruction, which in Figure 15 is depicted as the polyaromatics being separated from the product gas stream in the condenser.

Based on the closure of the heat and mass balance of the GoBiGas demonstration plant [3], it is concluded that for a stand-alone DFB gasifier there is an optimum size relationship between the CFB and the BFB boiler. As role of thumb, a CFB boiler of one energy unit of fuel input and a BFB-boiler of two energy units of fuel input can be combined to a DFB gasifier of up to eight energy units of fuel input. However, this number will be typically between 5 and 8, where the upper number relate to a plant that is fully optimized toward gasification and the lower

number to a plant with, more or less, full flexibility to be used as one gasifier or two parallel boilers.

Fluidized bed boilers in district heating networks or bark boilers in pulp mills are typically operated at an annual rate corresponding to 1500–3000 full-load hours (the annual delivered energy in MWh divided by the full load capacity in MW). This corresponds to load factors of 17– 35%, when operated as a boiler. Considering these boilers potential as DFB gasifiers both the time of operation and ability as fuel converters would increase. The goal for the operation of a gasifier for advances biofuel production will be around 8000 h per year, which also was one of the initial goals of the GoBiGas plant, and be independent of other system needs, for example district heating. The potential as fuel converter will, as explained above, increase with 8/3 when converting from a combustion to a gasification system, which also imply that the
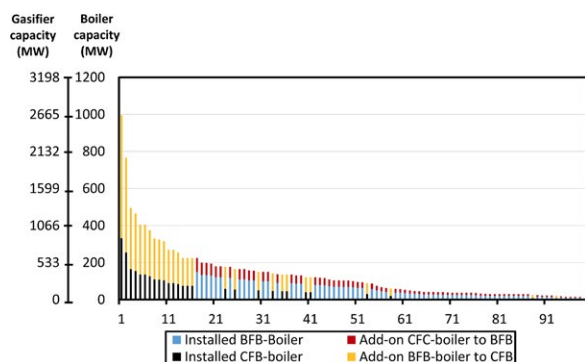
Figure 17. Existing installed capacity of fluidized bed boilers in the Swedish energy system and the corresponding additional boiler sizes needed to realize their conversion to dual fluidized bed gasifiers, Data provided by Christer Gustavsson.

system is complemented with a second boiler. Thus, converting such a boiler to an optimized DFB gasifier can increase the utilization of the plant by 700–1500%.

Thus, the presented process provides a technology that can gradually transform existing infrastructures, such as district heating networks, pulp and paper mills, and saw mills. The production processes in these units can also yield a renewable feedstock for oil refineries and petrochemical industries, as well as enable large quantities of intermittent electricity to be stored as fuel/products (further described in *Electrification and use of intermittent electricity within the process*). Such a transformation of the energy system is in line with the scenario presented in the *Introduction*, in which the most valuable assets are the existing sites and the surrounding infrastructures.

Within these assets, environmental permits are in place and the land is already exploited for this type of activity, which is a value that is hard to assess. Considering that many of the locations are situated at, or close to, the coastline or near water, it is difficult to envisage new claims for virgin land for this type of industrial production. If such new claims were needed, the costs would most likely be significantly higher for the land than for the equipment that is going to be installed, and very long political and legal processes would be required to secure the required permits. The conclusion from this is that the only feasible way to transform the energy system within the timeframe proposed by the Paris agreement is to make the transformation at sites that are currently being utilized by process or energy industries.

Incorporation into the existing district heating infrastructure

In Sweden and Finland, the main application for fluidized bed boilers is for district heating and in the pulp and

paper industry, and in Figure 17, the fluidized boilers currently installed in the Swedish energy system are visualized, together with the types and sizes of boilers with which they need to be combined to realize their full potential as DFB gasifiers. In summarizing the potential, it is clear that to the 6400-MWth (1200 MMBtu/h) installed boilers one needs to add 6800 MWth (1275 MMBtu/h) of boiler capacity to create a gasification potential of 35,000 MW (6550 MMBtu/h). With an assumed annual operation of 8000 h, this correspond to a fuel demand of 280 TWh of biomass (59 million dry tonnes of biomass per year), which can produce between 170 TWh and 200 TWh (14.6–17.2 MTOE) of advanced biofuels or materials. This corresponds to a potential production that is 5-times greater than the Swedish target for biofuel production in a fossil-free nation in Year 2045. However, due to logistic problems, it will for most locations not be feasible to have units with fuel inputs >500 MW (2500 dry tonnes of biomass/day), which reduces the potential by around 30%, decreasing the annual potential fuel demand to around 200 TWh (42 million dry tonnes of biomass). This is, nevertheless, a substantial demand for fuel and corresponds to the total forest growth in Sweden, implying that there will need to import biomass if this is to be realized.

In terms of the required level of investment, retrofitting an existing boiler from district heating or the combined production of electricity and district heating to a gasifier with full downstream synthesis would reduce the cost of the investment by 10–20% compared to a new stand-alone plant. This estimation is based on the projected cost for an nth commercial plant with full synthesis process extrapolated from the costs of the different process units in the GoBiGas plant with available scaling factors [3]. Note that this estimate does not include the potential savings linked to permits and land use.

Incorporation into existing pulp, paper, and saw mills

The integration of biomass/waste gasification with the downstream extraction of hydrogen or synthesis of hydrocarbons for the production of fuels, chemicals, and materials in the forest industry (pulp or saw mill) can be carried out in a stepwise manner using the dual bed gasification technology.

The first step is to change the bark boiler to a fluidized bed of suitable size. Here, a bubbling bed is the most likely system to be used to produce steam for the mill. The second step is to connect the bubbling bed to a circulating bed, transform the BFB boiler to a gasifier, and introduce a primary dryer that reduces the moisture content of the fuel in the process to <30%. The outcome

is a system that provides the mill with steam and a combustible gas with a high heating value, which can be used in, for example, a lime kiln.

The next step is to select the end-products and an appropriate synthesis process. This step also includes a secondary dryer, which decreases the moisture content to well below 10%. This step requires large additional investments, whereas the previous steps in the development can more or less be part of the continuous upgrading of the mill and can be operated independently, regardless of whether the mill decides to produce the refined end-products or not. Therefore, the investment decision for the synthesis or the hydrogen separation process can wait until the dual bed system has proven its performance and the markets for the produced end-products are assured.

Incorporation into existing oil refineries and petrochemical industries

To introduce the production of advanced biofuels, chemicals, and materials into an oil refinery or petrochemical industry that currently lacks both a biomass boiler infrastructure and established logistics for using biomass as fuel, requires a novel strategy for applying the technology. In both these types of industries, there is a large steam demand that is currently covered by combusting the gases that emanate from the internal distillation or conversion process. As the compositions of the gases burned in present processes to cover the heat demand are similar to those of the gases that are produced in a biomass gasifier, they can be upgraded to a syngas and further synthesized to desired hydrocarbons or extracted as hydrogen.

For these processes, it would be a natural first step to incorporate a biomass boiler for part of the steam production. In this way, the synthesis process for the intended system could be put into operation using the excess gases already produced within the industry, where the heat demand is covered by intensified heat integration and the combustion of biomass. This integration also provides the opportunities to increase gradually the demand for biomass and to build up the logistic infrastructure needed to receive biomass at the plant by starting with the installation of a CFB boiler. With a CFB that initially can be operated at 30% of maximum capacity, which correspond to just 4% of the final DFB gasifier capacity, the level of biomass utilization on-site can be gradually increased.

When the supply chain is able to provide biomass/waste corresponding to 80–90% of the capacity of the CFB boiler, the BFB boiler can be constructed, and the biomass could once more be increased at the same time

as more and more process internal gas is made available for the synthesis process. Once 80–90% of the combustion capacity of the two boilers is reached, it is time to connect the boilers to each other and to build a primary biomass dryer, which reduce the moisture content of the ingoing fuel from the initial 50% to 20–30%.

This will increase the conversion capacity of the biomass and, thereby, the production of both steam and gas from the unit. The produced gas can initially be burned in the steam boilers that were originally used to combust the internal gas, bringing the fuel capacity of the DFB gasification system up to 50–60% of the final need. The next steps are as follows: to introduce a second dryer that reduces the moisture content to 3–10%; to clean the gas of BTX; and to compress the gas to the pressure required for the synthesis process. At this stage, all the gas produced from the biomass/waste is going to the production of advanced biofuels, and the refinery or petrochemical industry goes back to using the internally produced gas of fossil origin to produce the steam needed for their processes.

Electrification and use of intermittent electricity within the process

As previously described, in a future energy system, the carbon atoms accumulated through photosynthesis will become a valuable resource, which means that as much carbon as possible needs to be converted to products. In the present process, around 50% of the carbon is released from the process as carbon dioxide. The majority of this comes from the CO_2 separation step in the synthesis process, and this reflects the composition of the biomass, which for woody biomass normalized to the carbon atom typically is $CH_{1.44}O_{0.66}$. This means that there is an excess of oxygen atoms that need to be removed as either carbon dioxide ($0.67 \; CH_{2.15} + 0.33 \; CO_2$) or as water ($CH_{0.12} + 0.66 \; H_2O$).

For most hydrocarbons, such as alkenes and alkanes, the H/C-ratio is around 2. The H/C ratio for aromatics ranges between 0.8 and 1, and for methane it is 4. For the production of methane (as in the GoBiGas demonstration plant), the right balance is achieved by inherent chemical splitting of water by the water gas reaction and, thereby, providing more hydrogen to the hydrocarbon molecule, while at the same time removing some more carbon as CO_2. To be able to convert all the carbon in the biomass to hydrocarbons, there is a need to split water outside the process, through for example, renewable electricity using electrolysis, and to provide pure hydrogen to the gas produced from the biomass in the DFB gasifier. By doing so, the oxygen in the biomass is removed from the downstream synthesis process as water instead

of carbon dioxide, while at the same time there is external production of pure oxygen.

Therefore, this method can be used to store renewable electrical energy in the form of hydrocarbons, whereby all the carbon can be converted into methane if one provides 1.94 H_2 per carbon atom in the biomass (1.94 H_2 + 0.67 $CH_{2.15}$ + 0.33 CO_2 => CH_4 + 0.66 H_2O). When producing methane, the stored energy in the methane can be as high as 173% of the chemically stored energy of the ingoing biomass. For alkene production, the corresponding ratio is 0.94 H_2 per carbon atom in the biomass (0.94 H_2 + 0.67 $CH_{2.15}$ + 0.33 CO_2 => ½ C_2H_4 + 0.66 H_2O), and the energy stored in the alkenes corresponds to 139% of the energy of the ingoing biomass. The energy in the electricity supplied for the water splitting comprises the energy of the vaporization of the water (14%), chemically bound energy (61%), and heat losses (25%). This means that around 60% of the electrical energy is converted to chemical energy in the generated end products.
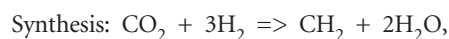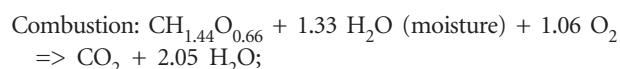
The use of electrolysis to increase the yield from the process entails a significant additional investment and this should be seen as a second step in the electrification of the process. A rough estimate is that large electrolysis processes at the present costs will be of interest if the mean cost for electricity for a major part of the year is half of that of the ingoing fuel, biomass or waste. Given the uncertainties concerning both the future evolution of electricity generation and the cost structure for biomass and waste, investors might be held back due to not seeing a solid business case.

A more straightforward method to introduce electricity into the process is to install electrical heaters to replace the fuel that is burned, so as to produce the heat in the process. This entails a much lower level of investment, and the marginal energy efficiency from electricity to product would be close to 100%, as direct heating with electricity implies that all the introduced energy is converted to heat, while heat generation that originates from combustion will introduce flue gas losses, together with other mechanical losses, example, a primary air fan [45]. Thus, installation of direct heating could increase the production and will be economically beneficial if the cost of the electricity is similar to or lower than the biomass or waste fuel for an extended period of the year. The feasible level of direct heating that could be applied would, however, be lower than that of the theoretical scenario, as combustion also serves other needs for the process, such as achieving 100% fuel conversion, the destruction of slipstreams from the synthesis process, and the regeneration of the catalytic process.

Based on the hydrogen and heat demands of the conversion process [3], the electricity demand in relation to the energy in the biomass used for pure methane production would be in the range of 1.4–1.6, and for alkenes it would be in the range of 0.7–0.9. Building on the example above in regard to the potential for retrofitting fluidized bed boilers in the Swedish energy system (Sweden has 10 million inhabitants, 0.13% of the world population), which has a potential for gasification of 200 TWh biomass annually, an additional 180–300 TWh electricity could be stored through DFB gasification. This corresponds to 120–200% of the present annual electricity production in Sweden.

If the goal is to utilize essentially 100% of the carbon, the introduction of hydrogen and electrical heating will not be sufficient, as there will be a small amount of CO_2 leaving with the flue gases. In such a carbon-optimized case, the CO_2 may need to be separated from the flue gases in an amine scrubber, which could be integrated with the CO_2 cleaning in the synthesis process, and the heat integration could be integrated with the electrolyzer for the reboiling of the amine (see Figure 18). However, this is an extreme scenario which implies that the carbon atoms in the biomass are much more valuable than the energy, and the cost for electricity is in relation to the biomass more or less negligible. During such circumstances, there is no need to optimize the process toward energy and it would most likely be more cost effective to burn the biomass/waste with the pure oxygen from the electrolyser to produce a pure carbon dioxide in a relatively simple combustion unit compared to the complex gasification process described in this paper. The carbon dioxide can thereafter be synthesized together with hydrogen to the desired hydrocarbon. For the production of alkenes from 50% moist biomass, the involved reactions can be summarized as follows:

Combustion: $CH_{1.44}O_{0.66}$ + 1.33 H_2O (moisture) + 1.06 O_2 => CO_2 + 2.05 H_2O;

Electrolysis: 3H_2O + electricity => 3H_2 + 1.5 O_2;

Synthesis: CO_2 + 3H_2 => CH_2 + 2H_2O,

where the combined reaction would be the same independently if applying a combustor or a gasifier for the initial thermal conversion of the biomass and could be written as $CH_{1.44}O_{0.66}$ + 1.33 H_2O (moisture) + electricity => CH_2 + 1.05 H_2O + 0.94 O_2.

However, taking the combustion path would require around twice as much electrical energy.

In summary, advanced biofuels, chemicals, and materials can be produced in large amounts through this process. As an example, a maximum level of production of methane of 346 TWh by retrofitting fluidized bed boilers in Sweden alone, corresponding to 32 bcm of natural gas, could be achieved, and this can be compared with the
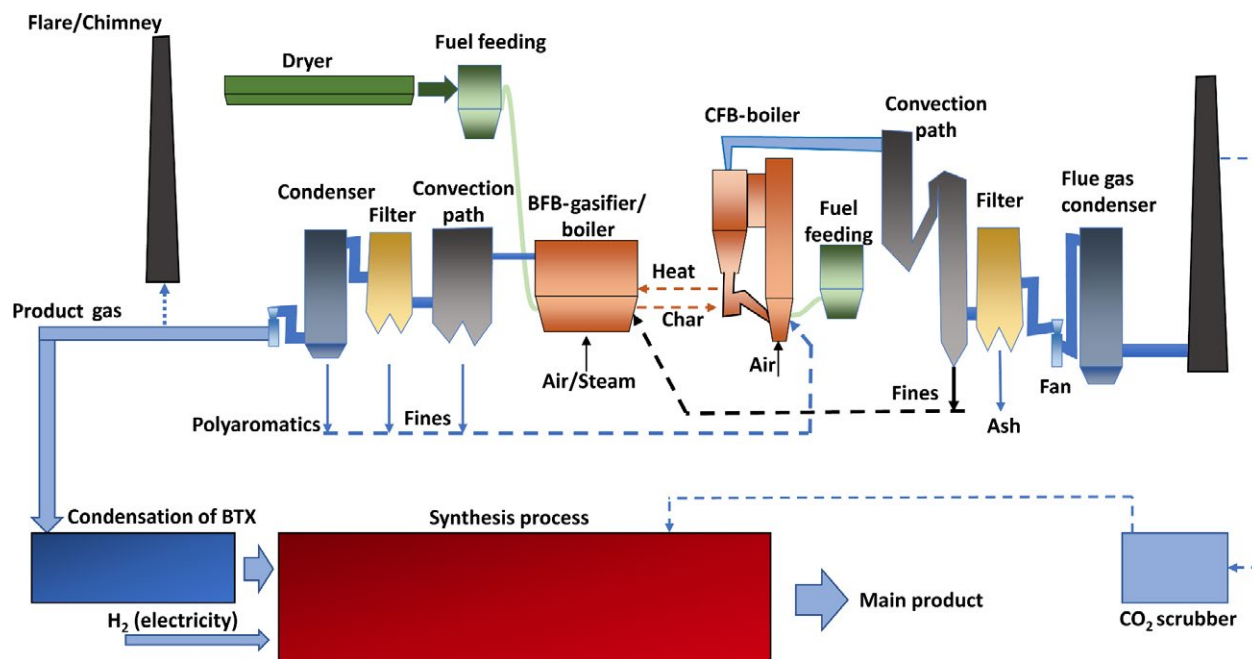
Figure 18. Process layout for a process with close to 100% carbon utilization.

world consumption (2013) of 3350 bcm of natural gas. Alternatively, one could achieve a maximum production level of alkenes of 278 TWh, which corresponds to approximately 23 million tonnes of alkanes, and this can be compared with world consumption (2014) of jet fuel alkanes of 225 million tonnes. These examples show the potential for producing substantial quantities of renewable fuels, chemicals, and materials through application of the technology presented in this paper and within the existing infrastructure.

Conclusions

The experience gathered from the first-of-its-kind demonstration project, GoBiGas, in combination with experimental results obtained from experiments in the Chalmers gasifier have augmented our knowledge on how to control the gas quality and how to design a gasification process such that problems related to fouling on heat exchangers for medium-temperature gasification of biomass can be avoided. The key roles are played by alkali and earth alkali metals (mainly potassium), which come naturally with the biomass ash and previously have been regarded as components that make biomass a problematic fuel in highly efficient thermal processes. Here, we show instead that in combination with specific bed materials and balanced by sulfur and calcium, potassium catalyzes the conversion process to produce a gas from the gasifier that is of sufficient quality to avoid downstream problems.

Furthermore, we show how coated heat exchangers can be used to condense out the steam and hydrocarbons after the gas is cleaned of particles, allowing significant simplification of the gasification process.

Together with new insights on fuel feeding and reactor design, presented in this paper, these solutions form the basis for a comprehensive process layout that can be used to transform fluidized boilers into fluidized bed gasifiers. This route represents an example of how biomass conversion can develop and be integrated with existing industrial and energy infrastructures to form highly effective systems that can deliver a wide range of products. By retrofitting existing district heat, pulp, paper, and saw mills, as well as oil refineries and petrochemical industries, renewable fuels, chemicals and materials can be produced from biomass and waste at increasing scale. To illustrate the potential, transforming existing fluidized bed boilers in the Swedish energy system alone would allow the production of jet fuels corresponding to 10% of the present world consumption.

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Conflict of Interest

None declared.

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