DIPLOMARBEIT

Optimization of a process concept for the industrial production of Bio-SNG from low-grade fuels

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Vorwort

"Es ist nicht genug zu wissen - man muss auch anwenden. Es ist nicht genug zu wollen - man muss auch tun." – Johann Wolfgang von Goethe

Um die lebenswerte Zukunft nachfolgender Generationen sicherstellen zu können, ist nicht nur ein fundamentales Umdenken im Umgang mit unserem Planeten, sondern auch bewusstes und zielgerichtetes Handeln erforderlich. Anstatt unsere rücksichtslose und kurzsichtige Ausbeutung endlicher Ressourcen weiter voranzutreiben, muss die Menschheit neue, nachhaltige Wege zur Sicherung unserer Lebensqualität einschlagen. Es ist hierbei Aufgabe von Wissenschaft und Technik, den politischen Entscheidungsträgern die technologische Grundlage dafür zu bieten. Diese Arbeit soll auch ihren kleinen Teil zu einer durchdachten und wissenschaftlich fundierten Klimastrategie beitragen, fernab von realitätsfremden, emotional geladenen Klimadebatten sowie politisch-populistischen Schlagwörtern.

Das Gelingen der vorliegenden Arbeit wäre in dieser Form ohne das Zutun etlicher Personen nicht möglich gewesen. Zuallererst gilt mein Dank Professor Hermann Hofbauer, der mir ermöglicht hat, nach meiner Bachelorarbeit auch meine Diplomarbeit in seiner Forschungsgruppe zu verfassen. Zudem danke ich Stefan Müller, für das entgegengebrachte Vertrauen und die ausgezeichnete Unterstützung, sowie die aktive Integrierung in den Arbeitsalltag der Forschungsgruppe. All meinen Kollegen der Arbeitsgruppe möchte ich an dieser Stelle auch ganz besonders danken, sowohl in fachlicher als auch in menschlicher Hinsicht - Die durchwegs gute Atmosphäre im Büro und die wertvollen Beiträge eurerseits haben wesentlich zum erfolgreichen Abschluss dieser Arbeit beigetragen. Ich möchte in erster Linie meine zwei Betreuer, Alexander Bartik und Florian Benedikt, herausheben und euch für die zahlreichen Diskussion und eure geopferte Zeit danken. Das ist in meinen Augen überhaupt keine Selbstverständlichkeit und weiß jeder Diplomand sehr zu schätzen.

Der Weg von den Anfängen meines Studiums bis zu diesem Zeitpunkt wäre ohne die zahlreichen unvergesslichen Momente und prägenden Erlebnisse mit meinen Freunden so nicht möglich gewesen. Ich möchte mich dafür herzlich bei euch allen bedanken. Seien es meine langjährigen Freunde aus der Schulzeit, meine Mitstudenten, meine Fußballkollegen, meine Erasmus-Freunde und noch viele, viele mehr – Ihr wart diejenigen, die diese Zeit zu etwas ganz Besonderem gemacht haben.

Zu guter Letzt möchte ich meiner gesamten Familie und dabei insbesondere meinen Eltern danken, auf deren Unterstützung ich in jeder Lebenslage zählen konnte und kann. Ohne euren emotionalen Rückhalt und eure finanzielle Unterstützung wäre ich heute nicht dort, wo ich stehe - Köszönöm nektek.

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Abstract

In the wake of the rising global energy demand and the simultaneous need for the reduction of harmful emissions, the question arises how mankind can ensure a global sustainable development for the (near) future. With a view to the ambitious targets of the Paris Agreement 2015, innovative energy technologies have to be developed to provide green and affordable energy.

In this context synthetic natural gas from gasified biomass (Bio-SNG) represents one of the alternatives among second generation biofuels, which can contribute to defossilization by substituting conventional natural gas. Previous developments have shown that the production of Bio-SNG is technically feasible in combination with the dual fluidized bed steam gasification. However, the technology struggles to compete economically with fossil natural gas, when high-quality woody biomass is utilized for the gasification process.

Within the scope of this work, an optimized concept for the industrial production of Bio-SNG from a low-value fuel (sewage sludge) based on the dual fluidized bed steam gasification is presented. Backed by the current state of knowledge in this area, a process chain was developed and implemented in a simulation model. With the aid of this simulation model, experimental data from the 100 kW_{th} pilot plant at TU Wien and values from literature, a techno-economic evaluation was carried out to assess the performance of the presented process chain.

To ensure a maximum fuel carbon utilization, renewable hydrogen is fed externally to the methanation section of the plant. For sewage sludge a global cold gas efficiency of 59.4 % could be achieved, which is in accordance with typical values for Bio-SNG processes. The economic performance depends on several factors, especially on the hydrogen- and the fuel-purchase price, and was quantified in different scenarios. The comparison within a business case and a subsequent sensitivity analysis showed that with favorable circumstances, such as low hydrogen-prices, investment subsidies, and/or the right process set-up, the technology is more than capable of competing with the energy supply by fossil natural gas.

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Kurzfassung

Im Zuge des steigenden globalen Energiebedarfs und der gleichzeitigen Notwendigkeit der Reduzierung von Schadstoffemissionen stellt sich die Frage, wie die Menschheit eine globale nachhaltige Entwicklung für die (nahe) Zukunft sicherstellen kann. Im Hinblick auf die ehrgeizigen Ziele des Pariser Abkommens 2015 müssen innovative Energietechnologien entwickelt werden, um grüne und bezahlbare Energie bereitstellen zu können.

Synthetisches Erdgas aus vergaster Biomasse (Bio-SNG) ist ein Vertreter von Biokraftstoffen zweiter Generation, das durch die Substitution von fossilem Erdgas zur Defossilisierung beitragen kann. Bisherige Entwicklungen haben gezeigt, dass die Produktion von synthetischem Erdgas in Kombination mit der Zweibettwirbelschicht-Dampfvergasung technisch umsetzbar ist. Allerdings kann das Verfahren aus wirtschaftlicher Sicht nur schwer mit fossilem Erdgas konkurrieren, wenn hochwertige, holzartige Biomasse als Brennstoff für den Vergasungsprozess eingesetzt wird.

Im Rahmen dieser Arbeit soll ein optimiertes Konzept für die industrielle Herstellung von synthetischem Erdgas aus einem minderwertigen Brennstoff (Klärschlamm) ausgehend von der Zweibettwirbelschicht-Dampfvergasung erarbeitet werden. Die Prozesskette wurde basierend auf dem aktuellen Stand des Wissens konzipiert und in ein Simulationsmodell übergeführt. Mithilfe dieses Simulationsmodells, experimenteller Daten aus der 100 kW_{th} Pilotanlage der TU Wien und relevanter Literaturwerten wurde eine techno-ökonomische Analyse zur Beurteilung der Konkurrenzfähigkeit der vorgestellten Anlage durchgeführt.

Um einen maximalen Kohlenstoff-Umsatz und eine hohe SNG-Ausbeute zu gewährleisten, wird direkt vor der Methanierung erneuerbarer Wasserstoff aus einer externen Elektrolyseanlage zudosiert. Für den Prozess konnte so ein globaler Kaltgaswirkungsgrad von 59,4 % erreicht werden, der mit typischen Werten für Verfahren zur synthetischen Erdgaserzeugung übereinstimmt. Das wirtschaftliche Abschneiden des Prozesses hängt wesentlich von einigen Faktoren, wie dem Wasserstoff- und dem Brennstoffpreis, ab und wurde quantitativ für verschiedene Szenarien ermittelt. Der Vergleich innerhalb einer technoökonomischen Fallstudie samt einer anschließenden Sensitivitätsanalyse zeigt, dass die Technologie bei günstigen Rahmenbedingungen, wie beispielweise niedrigen Wasserstoffpreisen, Investitionszuschüssen und/oder der richtigen Prozessgestaltung, mehr als konkurrenzfähig zur Energieversorgung mit fossilem Erdgas sein kann.

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Table of contents

1 INTRODUCTION 1

2 FUNDAMENTALS OF PRODUCT GAS GENERATION 1

2.1 DUAL FLUIDIZED BED TECHNOLOGY 1

2.1.1 Basic Principle 1

2.1.2 Advanced gasification technologies 2

2.2 PRODUCT GAS TREATMENT 2

2.2.1 Gas cleaning units 2

2.2.2 Comparison of gas cleaning technologies and recent developments 8

2.2.3 Product gas quality requirements 10

3 FUNDAMENTALS OF BIO-SNG PRODUCTION 11

3.1 SYNTHESIS OF BIOMASS-DERIVED PRODUCT GAS 11

3.2 BASIC PRINCIPLES OF METHANATION 12

3.3 REACTOR TYPES AND STATE OF THE ART 16

3.3.1 Fixed bed methanation 16

3.3.2 Fluidized bed methanation 18

3.3.3 Alternative methanation concepts and research activities 21

3.3.4 Comparison of methanation concepts 22

3.4 PRODUCT GAS UPGRADING AND QUALITY REQUIREMENTS OF THE NATURAL GAS GRID 22

3.5 R&D CHALLENGES 24

4 APPLIED PRINCIPLES AND METHODS 26

4.1 GASIFICATION EXPERIMENTS WITH SEWAGE SLUDGE 26

4.2 PROCESS SIMULATION WITH IPSEPRO 28

4.3 ECONOMIC EVALUATION TOOLS 29

4.3.1 Plant cost estimation 29

4.3.2 Investment decision - Net Present Value 30

4.3.3 Levelized cost method 33

5 PROCESS DESIGN AND SIMULATION OF THE SEWAGE SLUDGE CONCEPT 35

5.1 PROCESS DESIGN AND OPERATING DATA 38

5.1.1 Fuel supply 38

5.1.2 Gasification 39

5.1.3 Gas cooling and gas cleaning 40

5.1.4 Methanation 43

5.1.5 Gas upgrading 44

5.1.6 Flue gas route 46

5.1.7 Basic Simulation Data 47

5.2 SIMULATION RESULTS 48

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6 ECONOMIC EVALUATION 52

6.1 SEWAGE SLUDGE PLANT COST CALCULATION 52

6.2 TECHNO-ECONOMIC CASE STUDY 55

6.3 COST CALCULATION WITH THE LEVELIZED COST METHOD 60

7 CONCLUSION AND OUTLOOK 62

NOTATION 65

SYMBOLS 65

ABBREVIATIONS 65

LIST OF TABLES 69

LIST OF FIGURES 71

APPENDIX 84

A. IPSEPRO SIMULATION MODELS 85

B. EXTENDED STREAM TABLES 89

C. ADSORBER SIZE ESTIMATION 92

D. EXTENDED EQUIPMENT COST CALCULATION 93

E. CEPCI FACTORS 94

F. CALCULATION OF THE WORKING CAPITAL 95

G. COST RATES 96

H. GOBIGAS PLANT SCALE-DOWN 97

I. ACTIVATED CARBON DATA SHEETS 98

J. PUBLICATION 100

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1 Introduction

The global energy demand has been experiencing a rapid increase during the last decades. According to the BP Statistical Review of World Energy 2019 [1] the primary energy consumption grew at a rate of 2.9 % in 2018, almost doubling its 10-year average of 1.5 % per year. In addition, carbon emissions rose at their highest rate seen for years. These energy data paint a worrying picture considering the Paris climate goals of holding the increase of global average temperature to well below 2 °C compared to pre-industrial levels. Consequently, our society faces great challenges as we try to navigate the transition towards a low-carbon energy system. The development of innovative, green and affordable energy technologies is a key to achieve the goals set [1].

The dual fluidized bed (DFB) steam gasification is a promising energy technology which has been developed and extensively investigated with different generations of pilot plants at TU Wien. By means of this process, solid feedstocks can be converted to a mid-calorific and nitrogen-free product gas with a favorable composition to produce a large variety of end products. Based on this concept, the consideration of holistic process chains is increasingly moving into the focus of scientific studies [2].

One of the promising utilization possibilities of the gasification-derived product gas is the production of synthetic natural gas (SNG) through a methane synthesis step. This conversion process shows the highest efficiency from feedstock to fuel of today's commercially available 2nd generation biofuels. The big advantage of SNG is the ability to substitute natural gas in any of its applications and the possibility to take advantage of the existing natural gas infrastructure, which facilitates a gradual transition towards the renewable form of natural gas [3].

An overview of the entire SNG process chain is given in Figure 1.

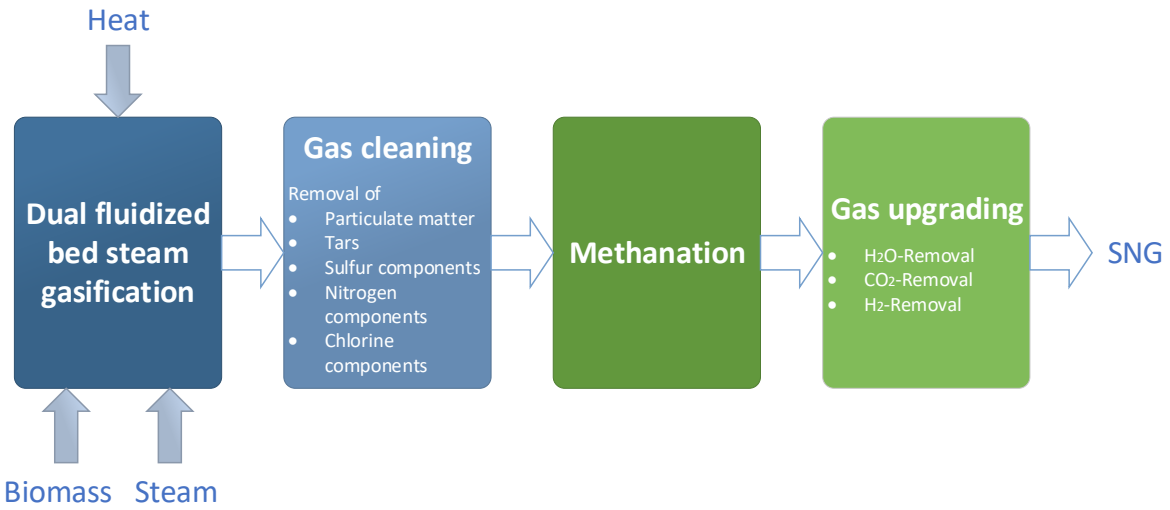

Figure 1: Process chain of synthetic natural gas (SNG) production from biomass, adapted from [4]

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The methanation of coal-derived product gas has been carried out for more than 100 years and is a well-developed process. A lot of the research efforts date back to the early 1970s, when the USA intensified research because of the general oil shortage [5]. Based on the experience gained and the increasing need for a sustainable energy supply, research on the conversion of biomass to SNG has been intensified in the last 15 years. Concomitant with these research efforts two milestones were achieved by the erection of the first Bio-SNG pilot plant in Güssing (2008) and the first large scale demonstration plant in Gothenburg (2012), both based on the DFB gasification [6]. Still, the process has not proven to be economically competitive, if conventional expensive fuels, like wood, are used. Therefore, the focus of research lies on developing ways to successfully convert biogenic residues and waste materials into a high-value product. However, these cheap alternative fuels, usually show rather unfavourable properties for gasification and thus lead to technical challenges and limitations [7].

In the context of the present work, an optimized industrial process concept for the production of Bio-SNG from a low-grade fuel should be developed for a product gas derived from a dual fluidized bed steam gasification. Based on the current state of knowledge in this field this process concept should be implemented in a simulation model for the calculation of mass- and energy balances. With the simulation results, experimental data and literature values a technical and economic evaluation of the process concept should be carried out. The following research questions can be derived from this:

- **What is the best possible process route for the industrial production of Bio-SNG from low-grade fuels based on the dual fluidized bed steam gasification?**
- **What are the mass- and energy balances for this process concept?**
- **To what extent is the proposed process chain technologically mature?**
- **What are the investment costs and how competitive is the proposed process chain?**

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2 Fundamentals of product gas generation

2.1 Dual Fluidized Bed Technology

2.1.1 Basic Principle

The gasification of biomass has proven to be a promising and sustainable method for the production of electricity, heat and in more recent research activities also for the synthesis of high-quality products. In Figure 2 a short overview of the different stages occurring during a gasification process is given.

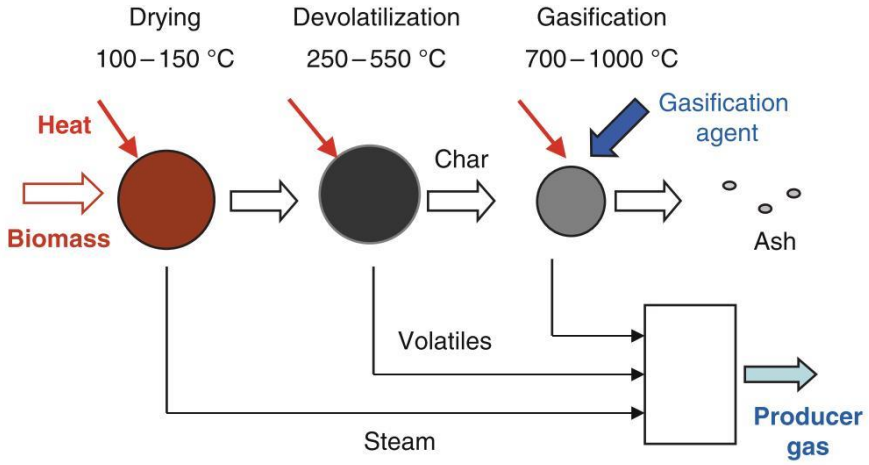

Figure 2: Process during gasification of a single particle [8]

This rising interest in the production of different fuels and energy carriers through gasification puts a stronger focus on fluidized bed gasifiers, which show favourable properties for the synthesis processes. The high mixing rate of the solid fuel, the bed material and the gas phase leads to an increased gas-solid contact, excellent heat transfer and enhanced catalytic effects. These factors all contribute to an optimal conversion of the solid fuel to a mid-calorific product gas with a higher calorific value of about 11-15 MJ/Nm^3 [9,10].

For the generation of a highly-valuable product gas the concept of the DFB gasification offers a well-proven technology, which is on the verge of commercialization at this point. Several demonstration plants in Austria, Germany and Sweden within the thermal fuel power range of 8-32 MW_{th} work according to this concept and delivered promising results for a further implementation of the technology [11].

In Figure 3 the basic principle of the DFB gasification is shown. Because steam is used as gasification agent the heat for the endothermic reaction has to be provided externally. This is realized through the combination of the gasification reactor with a combustion reactor, where a circulating bed material (e.g. olivine, limestone) is heated up and transferred back to the gasifier through loop seals in order to provide the necessary heat. In the gasification section the biogenic feedstock undergoes a thermochemical decomposition through an intense contact

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with the gasification agent and the hot bed material. The bed material and the residual char are then transferred to the combustion reactor, where the char becomes combusted with air and the bed material is heated up simultaneously [8,12].

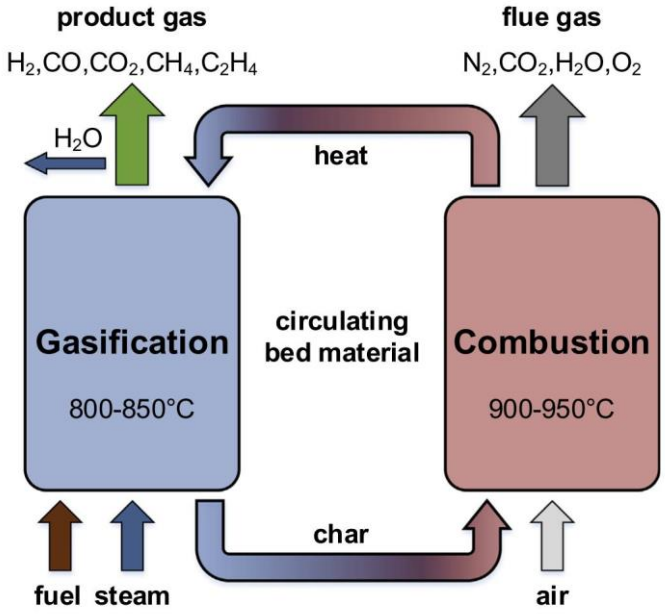

Figure 3: Basic principle of the DFB steam gasification process [7]

With the dual fluidized bed technology, a nitrogen-free product gas can be generated without the application of expensive pure oxygen. Its composition is mainly made up of hydrogen (H_2), carbon monoxide (CO), carbon dioxide (CO_2), methane (CH_4), light hydrocarbons (C_xH_y) and water (H_2O). Disadvantageous compared to other gasifiers types are the increased complexity of the process due to a second fluidized bed and higher dust contents in the product gas stream [13]. Therefore, the higher efforts regarding the plant design are only justified, if the advantages of the higher product gas quality are brought to bear in the utilization of the product gas (e.g. fuel-syntheses, application in a gas turbine) [7,11].

2.1.2 Advanced gasification technologies

In the following chapter two advanced gasification technologies are outlined, which are essential in connection with the topic of this thesis. However, there are several other approaches regarding process improvements and optimizations suggested by different research groups. Although many of these have shown promising early results, further research is needed in this area. For a deeper understanding more extended literature is recommended [12,14,15].

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Sorption Enhanced Reforming (SER)

Based on the DFB principle, the Sorption Enhanced Reforming represents an advanced pathway, where limestone, mainly consisting of calcium carbonate ($CaCO_3$), is used as bed material instead of the conventional bed material olivine. The process arrangement is shown in Figure 4.

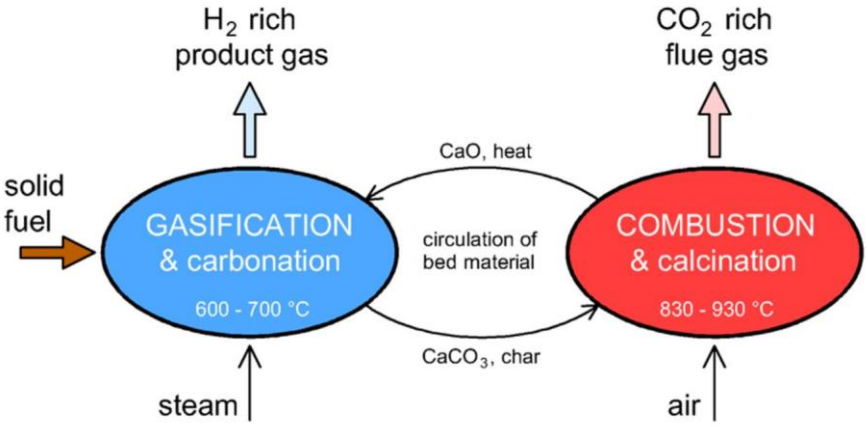

Figure 4: Basic principle of the SER-process with limestone as bed material [2]

By the application of limestone as bed material it is possible to selectively transport CO_2 from the gasification to the combustion reactor. There, the CO_2 is split off from $CaCO_3$ in a calcination reaction at high temperatures. Calcium oxide (CaO) is recirculated to the gasification reactor, where CO_2 is captured to form again $CaCO_3$ [2,8]. The mentioned reactions are given in Table 1.

Due to the reduction of the CO_2 content in the gasifier the product gas composition changes towards higher H_2-contents by pushing the water-gas shift reaction to the product side. As a result, a product gas with high H_2-content (< 70 %) and low CO- and CO_2-contents (< 10 %) can be obtained, which increases the possibilities for product gas utilization [2]. At the same time, Soukup et al. [16] showed that by means of the SER-process a significant reduction of tar formation and by integration of the exothermal reaction heat (water-gas shift, carbonation), a high process efficiency can be reached.

Table 1: Important chemical reactions for the SER-process [2]

Name	Reaction formula	Enthalpy of reaction	Equation
Water-gas shift	$CO + H_2O \leftrightarrow CO_2 + H_2$	exothermic	2.1
Carbonation	$CaO + CO_2 \leftrightarrow CaCO_3$	exothermic	2.2
Calcination	$CaCO_3 \leftrightarrow CaO + CO_2$	endothermic	2.3

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A big advantage of the SER-process is the possibility to adjust the H_2/CO-ratio in the range of 2 to 9 by varying the temperature (Figure 5). Moreover, also ratios < 2 might be possible by changing the bed material circulation rate or by using bed material mixtures (e.g. calcite and silica sand) [17]. Due to this flexibility, it is an outstanding and interesting gasification technology to produce a gas mixture according to the demands of a specific synthesis process.

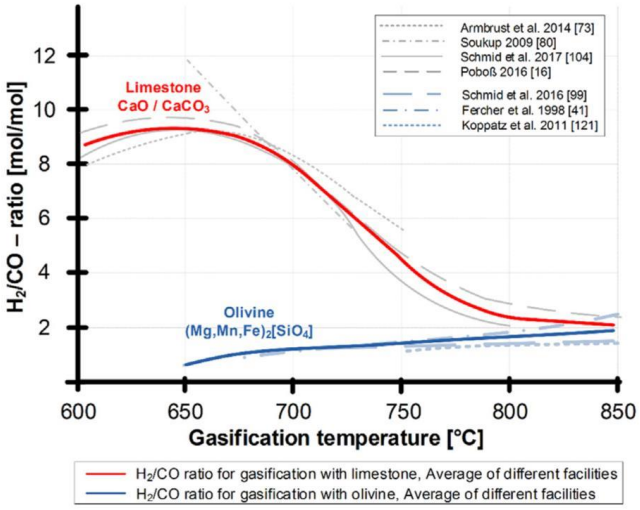

Figure 5: H_2/CO-ratio for gasification with different bed materials [2]

The main downside of the SER-process is the low abrasion resistance of limestone, which results in an increased dust content in the product gas and a loss of bed material in the reactors [9]. However, the higher attrition rates for limestone are more of an issue in terms of sustainability and process design than in terms of economic considerations, given that limestone is a common, cheap material, which is free of heavy metals (in contrast to conventional olivine) [2].

Another disadvantage is the low carbon utilization with regard to synthesis applications. Due to the removal of CO_2 from the product gas route it is not available anymore for the synthesis reaction, which consequently lowers the carbon-based synthesis product output [18,19].

The SER-process was tested at the demonstration plant in Güssing in 2007/2008. Based on these results a 10 MW_{th} plant in Germany was planned to be erected for combined heat and power production in the first place and later for research in the field of biofuel generation. Anyhow, drastic increases in the wood fuel price led to the abandonment of the whole project out of economical inefficiency [15].

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Next-generation dual fluidized bed gasification reactor design

With regard to the challenging properties of a product gas derived from cheap alternative fuels (e.g. high tar contents and dust loads) and the before mentioned low attrition resistance of limestone in the SER-process a novel DFB reactor design was developed at TU Wien [2]. The scheme of the advanced reactor design is shown in Figure 6.

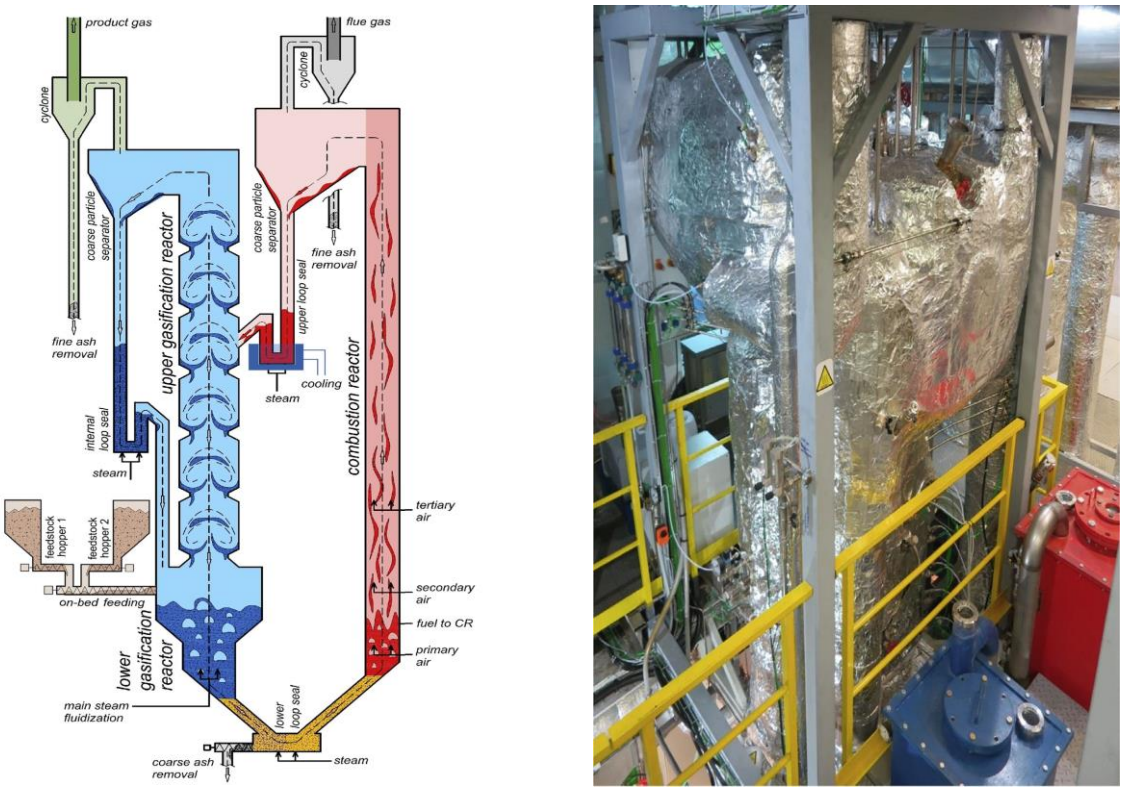

Figure 6: Advanced reactor design at TU Wien [17]

In the upper gasification section, a countercurrent flow of catalytically active bed material and product gas can be observed. Intensive mixing and optimized residence time for the components is achieved by constrictions in the column, which enhance the gas-solid contact and therefore the catalytic effects on the gasification reactions [20,21]. A separation concept comprising a multistage solid separation system is installed on each side for the capture of both coarse and fine particles from the gas stream. Gravity separators inserted between the reactors and the cyclones allow a gentle separation of the solid material. The thereby reduced attrition of the bed material makes the technology attractive for the SER-process [2,21].

Comprehensive experiments showed that a higher gasification efficiency and an increased fuel, water and tar conversion can be obtained with the novel design in comparison with the old plant design. Moreover, a higher operational flexibility could be observed in terms of fuel power and steam to fuel variations [17]. The new design offers the ability to process different alternative solid fuels with widely varying chemical and physical fuel properties [20]. This can

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be considered as a significant progress in gasification technology, because using residual biomass fuels is seen as a major factor in making gasification technologies economically competitive. This economic competitiveness will be discussed in detail in Chapter 6. Further information on the novel reactor design and detailed experimental results of the last years can be found in the literature [9,20–24].

2.2 Product gas treatment

In the course of a recent survey of a panel of experts, it was stated that the control and management of product gas contaminants is still one of the biggest issues preventing a commercial break-through of biomass gasification [25]. The generated product gas from biomass gasification consists mainly of the components H_2, CO, CO_2, H_2O and CH_4. However, impurities are inevitably produced during the gasification process. The main impurities can be listed as:

- particulate matter (PM),
- tar,
- sulfur compounds (Hydrogen sulphide (H_2S), Carbonyl sulphide (COS)),
- nitrogen compounds (Ammonia (NH_3), Hydrogen cyanide (HCN)),
- and chlorine compounds (Hydrogen chloride (HCl)).

These undesirable components can interfere with downstream applications e.g. in form of erosion, corrosion, fouling or catalyst poisoning and therefore have to be reduced to an acceptable level given by the specific downstream application [11,15,26].

Before going into further elaboration on gas cleaning concepts and their advantages and disadvantages, the options of removing each component of the product gas impurities are discussed in the following chapter. This should provide the background knowledge for Chapter 2.2.2.

2.2.1 Gas cleaning units

Particulate matter

Solid particles are always present in gasifier-derived product gas and vary widely in composition and grain size (1-100 µm) depending on the choice of feedstock and on the process. The majority is made up of inorganic compounds (ash) and residual carbon (char, soot). Furthermore, bed material or catalyst particles can be entrained as well, if applied in the gasifier. Particle removal technologies have to be designed in a way that high dust loads and sticky ash particles are of no concern. The amount of particles in the producer gas, the preferred operating temperature and the separation efficiency are the main selection criteria

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for an appropriate technology [15,27]. Particulate matter is classified by an aerodynamic diameter. Common practice is to remove particulates of a certain size below a given concentration level, depending on the product gas application requirements [27]. An overview of the characteristics of particulate matter removal technologies can be found in Table 2. For biomass gasification in particular barrier filters are of interest, because in addition to their simple set-up separation efficiencies up to 99.999 % can be reached [15].

Table 2: Characteristics of particle removal technologies [15]

Parameter	Unit	Cyclones	Electrostatic precipitators	Barrier filters			Wet scrubbers
				Bed filters	Bag filters	Rigid filters	
Operating temp.	°C	<1000	<500	<870	<370	<1150	<100
Pressure drop	mbar	5-30	0.3-4	10-60	6-23	10-100	1-10
Dust conc. raw gas	$g*m^{-3}$	<1000	<50	<100	<100	<100	<10
Dust conc. clean gas	$mg*m^{-3}$	>100	>25	<10	1-10	<1	>10
Filtration grade	µm	5	5	3	1	0.5	1

The basics of most particle separation technologies are well known and applied in various industrial processes. Current research activities focus on the development of efficient high temperature filtration units, as up to now, most of them cannot ensure long-term stability for a biomass-derived product gas at temperatures above 450 °C. The main advantage of hot gas filtration (HGF) solutions is their ability to operate above the condensation temperature of tars and water and therefore prevent tar fouling in plant units and protect heat exchangers of particle induced erosion [15]. However, one of the main obstacles for their assertion has been the design of an effective recleaning system, replacing the conventional jet pulse recleaning method. The Karlsruhe Institute of Technology in cooperation with the Paul Scherrer Institute have been developing an innovative technique called coupled pressure pulse (CPP), which opens new possibilities by enabling higher recleaning intensities at relatively low pressures. A detailed description of the advanced recleaning system can be found in reference [28]. This combination of HGF and CPP has already been successfully implemented at different pilot plant scales for several biomass conversion processes.

Tars

Tars consist of a complex mixture of condensable organic molecules. In general, the term comprises compounds with a molecular weight greater than that of benzene [11]. If the temperature in a given process drops below the tar dew point, they condensate and lead to operational problems like plugging of piping, fouling of equipment, or deactivation of catalysts.

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The fouling of heat exchanger surfaces, for instance, is a common industrial problem and it has been estimated to cost industrialized countries around 0.25 % of their entire GDP [29].

The reduction of the tar content in the product gas can be obtained through primary measures by selecting a suitable gasification technology. Additionally, secondary measures have to be taken for further tar removal and for meeting the product gas criteria depending on the desired application. Physical tar removal methods comprise the adsorptive removal, for example on activated carbon beds, and the absorptive removal within wet scrubbers. The latter technologies are the most commonly applied cleaning process for tar removal [11]. They require a product gas cooling below 450 °C, where tars condensate and form aerosols. In fact, wet scrubbers are able to collect both tar aerosols and soluble tar components. Oil scrubbing liquids, especially rapeseed methyl ester (RME), proved to be superior in the process compared to water due to their lipophilic properties [12,15,30].

A possible process layout was successfully developed by the Energy Research Center of the Netherlands (ECN): The OLGA process (Figure 7) is a multi-stage, oil-scrubbing tar removal concept, which has already been realized in commercial scale. A detailed description of the process can be found in the literature [14,31].

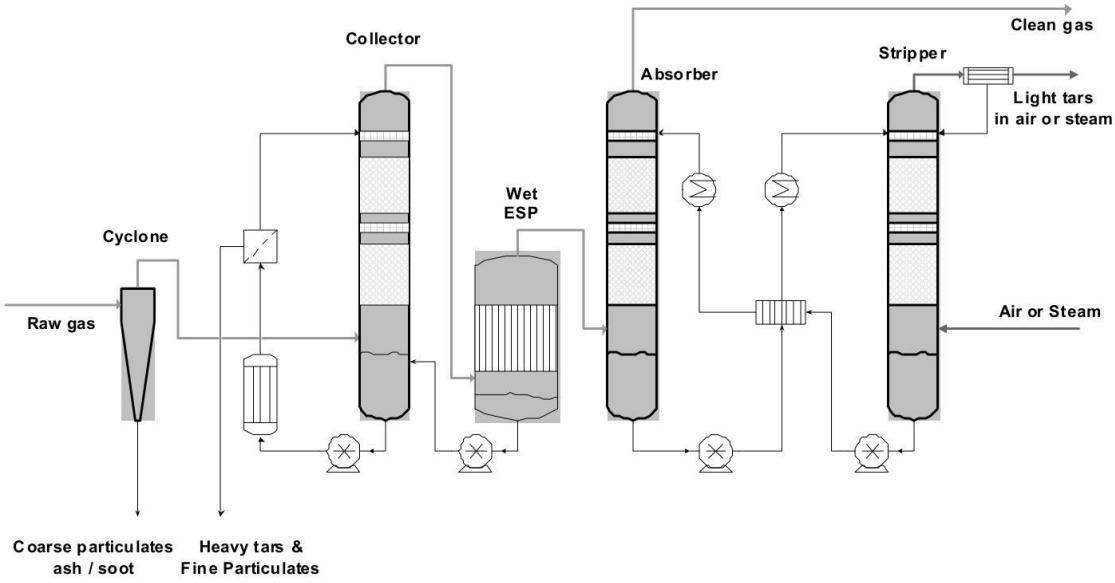

Figure 7: Process scheme of the OLGA tar removal technology [31]

Another cold gas tar removal technology was elaborated at TU Wien and applied downstream of an 8 MW_{th} DFB gasifier located in Güssing. The scrubber in this concept is oil-based as well but is operated with RME instead of the special scrubbing oil used in OLGA. Within the latter technology, the scrubbing liquid is regenerated by means of a stripper. In the Austrian concept a small part of the used RME is burned in the combustion zone of the dual fluidized bed and the rest is used again in the scrubber. However, it has to be considered that the consumption

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of huge volume streams of fresh RME constitutes a considerable contribution to the fuel and energy consumption of the process [15,32]. Apart from these differences, a further significant discrepancy between the two introduced tar removal technologies lies in the fact that the Güssing concept partially condenses water in the scrubber, while the OLGA process keeps the temperature above the water dew point and removes water downstream of the scrubber. A more detailed comparison of the two introduced concepts is covered by reference [33].

Another approach for the reduction of tar concentrations in the product gas happens at temperatures above 1100 °C where tars are cracked to lower molecular hydrocarbons, like CH_4. This temperature needs to be set either in the gasifier itself (e.g. entrained flow gasifier) or downstream of the gasifier either by admission of an air/oxygen stream or by using hot heat exchanger surfaces [12,25,27]. Tar reduction efficiencies of 80-90 % are reported in literature by this technology [11]. Other configurations of thermal cracking methods, e.g. plasma assisted cracking, have not proven to be applicable so far, as a compromise between cost efficiency, safe operational conditions and good tar removal efficiency could not be ensured [11].

Tar cracking may also be obtained at significantly lower temperatures if a catalyst is added in the process. It can be deployed in situ as bed material or as additive either in the gasification reactor or more frequently downstream of the gasifier in separate reactors. The big advantage of this catalytic process is that thermal penalties and additional costs due to higher operating temperatures can be decreased. On the other hand, a big operational challenge is added to the process with the necessity to maintain catalyst activity. Poisoning, fragmentation, and carbon deposition are the main issues regarding catalyst deactivation [27]. Common operating temperatures for catalytic cracking range between 600 °C and 900 °C, whereas certain catalysts maintain high efficiencies also at lower temperatures. Eligible catalysts are either of natural origin, like catalytically active minerals (e.g. olivine, CaO), ferrous metal oxides etc., or of synthetic origin, like char, alkali metal carbonates, or activated alumina [15]. In the course of a literature survey conducted by the National Renewable Laboratory in 2002 different catalysts for catalytic cracking where examined and assessed. The report stated that a tar conversion of > 90 % could be reached between 450-900 °C depending on the catalyst used [12]. For a deeper understanding of this topic a further elaboration of potential catalysts as well as many other references can be found in Schildhauer et al. [15].

Sulfur

The sulfur content in biomass is mainly released as H_2S and COS into the product gas, while smaller sulfur amounts are also present in form of organic sulfur. The ratio between the individual compounds is set mainly by the operating temperature of the gasifier [31]. Sulfur removal is an essential part of gas cleaning sections in order to avoid metal corrosion, downstream catalyst deactivation and the formation of the regulated pollutant sulfur

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dioxide (SO_2). Numerous cleanup technologies have been developed to satisfy downstream sulfur limits. These include both dry and liquid processes, which cover a temperature range from below zero to several hundred degrees Celsius [27]. Basically, state of the art technologies for sulfur removal can be divided into the following three categories:

- wet scrubbing,
- hydrodesulfurization (HDS),
- and adsorption.

For low temperature desulfurization wet scrubbing processes like Rectisol, Purisol, or amine-based scrubbers and adsorption with activated carbon are the most applied cleaning solutions. Due to high operational costs, scrubbers are only favorable in large scale applications, while activated carbons are preferred in small scale plants [15,31]. High temperature sulfur removal can be enabled in situ in the gasifier by the utilization of mainly calcium-based bed materials, such as limestone or dolomite [34]. As a secondary measure sorbent materials, like metal oxides or activated carbon, are applied at temperatures around 300-1000 °C either upstream of a hot gas filter or in fixed beds mostly downstream of an HDS-reactor [15]. The operating range of different metal oxides, which have been attracting a lot of attention in recent research activities, is illustrated in Figure 8. An overview of sorption materials for H_2S-removal can be found in the references [26,27,34].

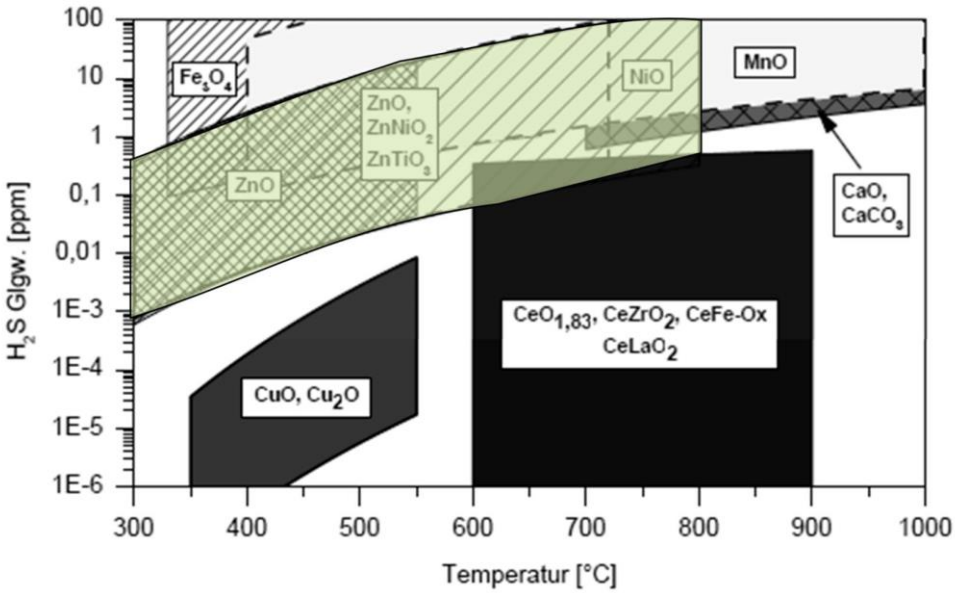

Figure 8: Operating range of metal oxides for desulfurization [4]

Sulfur-containing hydrocarbons are often neglected in gas cleaning studies - partly owed to the lack of appropriate analytical equipment - and are usually not removed by conventional cleaning equipment. The above mentioned hydrodesulfurization is a technique of converting these compounds in the presence of a catalyst and H_2 or H_2O to H_2S. The H_2S can then be

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removed in a second stage by conventional cleaning methods [15,31,35]. For hydrogenolysis higher temperatures around 350 °C are needed and cobalt molybdate ($CoMoO_4$) or nickel molybdate ($NiMoO_4$) catalysts are usually applied [36]. Hydrolysis, on the other hand, only requires moderately enhanced temperatures and catalysts such as titanium dioxide (TiO_2) or aluminium oxide (Al_2O_3) can be used [37,38]. Three exemplary reactions for hydrodesulfurization are shown in Table 3.

Table 3: Exemplary reactions for hydrodesulfurization [35]

Name	Reaction formula	Enthalpy of reaction	Equation
Thiophene hydrogenolysis	$C_4H_4S + 4H_2 \leftrightarrow C_4H_{10} + H_2S$	exothermic	2.4
Ethanethiol hydrogenolysis	$C_2H_5SH + H_2 \leftrightarrow C_2H_6 + H_2S$	exothermic	2.5
Carbonyl sulphide hydrolysis	$COS + H_2O \leftrightarrow H_2S + CO_2$	exothermic	2.6

Nitrogen

Most of the nitrogen contaminants in the product gas occur as NH_3 and less abundantly as HCN. It is reported that enhanced gasification temperatures enable secondary reactions for the increased formation of both HCN and NH_3 [27]. Their removal is required to avert fouling phenomena in the presence of certain compounds (Cl, H_2S), catalyst deactivation (e.g. Fischer-Tropsch (FT) catalyst) and possible formation of the atmospheric nitrogen oxides pollutants (NO_x). The good solubility of NH_3 and HCN in water makes wet scrubbers an efficient solution for low-temperature removal. Reduction of NH_3-concentrations by >99 % could be observed when a spray tower was utilized at ambient temperature. Subsequent solvent recovery is carried out through neutralization with acids to ammonia salts (in case of NH_3), biological conversion to elemental nitrogen (N_2) or stripping from the scrubbing water [31,39]. Several studies state that even condensed vapor in oil based scrubbers or condensation systems can ensure substantial removal of nitrogen compounds [39–41].

Alternatively, the hot gas cleanup of nitrogen compounds focuses on the destruction of NH_3 via catalytic decomposition rather than removing it from gas streams. More precisely, NH_3 can be decomposed by means of catalysts similar to those used for tar cracking or hydrocarbon reforming. Dolomite, nickel and iron-based catalysts are generally used for this purpose, while also more expensive catalysts on noble metals-basis have shown encouraging results in experiments. This combined tar and NH_3 removal seems to be very promising with NH_3-destruction levels of >99 % and has the great advantage of maintaining the heat of the producer gas. Nevertheless, commercial implementations of this technology are still under development [15,31].

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For the specific removal of HCN either hydrolysis/hydrogenation reactors or impregnated activated carbons are used. The catalysts for HCN-hydrolysis are reported to be very similar to those used for COS-hydrolysis, which suggests a joint COS- and HCN-removal [38].

Chlorine

Chlorides are the predominant halides in the product gas, usually present in the form of HCl. Reports indicate that a major part of HCl can already be removed through the filter unit [42]. However, if cleaning steps are necessary, HCl can be removed either by dry or wet processes. For dry removal two types of adsorbents are commercially available: sodium carbonate/trona (Na_2CO_3, $NaHCO_3$) and CaO. Optimal temperatures for the adsorbents were found to be around 400-500 °C [5,31]. Also Al_2O_3 is reported to be able to remove chlorine compounds by adsorption [32]. For wet scrubbing either water or caustic water solutions are used [25].

2.2.2 Comparison of gas cleaning technologies and recent developments

Depending on the operating temperature gas cleaning technologies can be divided into three different categories according to [15]:

- Cold gas cleaning (ambient or lower temperature)
- Warm gas cleaning (100-400 °C)
- Hot gas cleaning (> 400 °C)

To give an overview an exemplary pathway for each of the gas cleaning categories is shown in Figure 9 for the product gas utilization in fuel cells. However, the displayed gas cleaning routes are also valid for catalytically promoted conversion processes, such as methanation or liquid fuel synthesis.

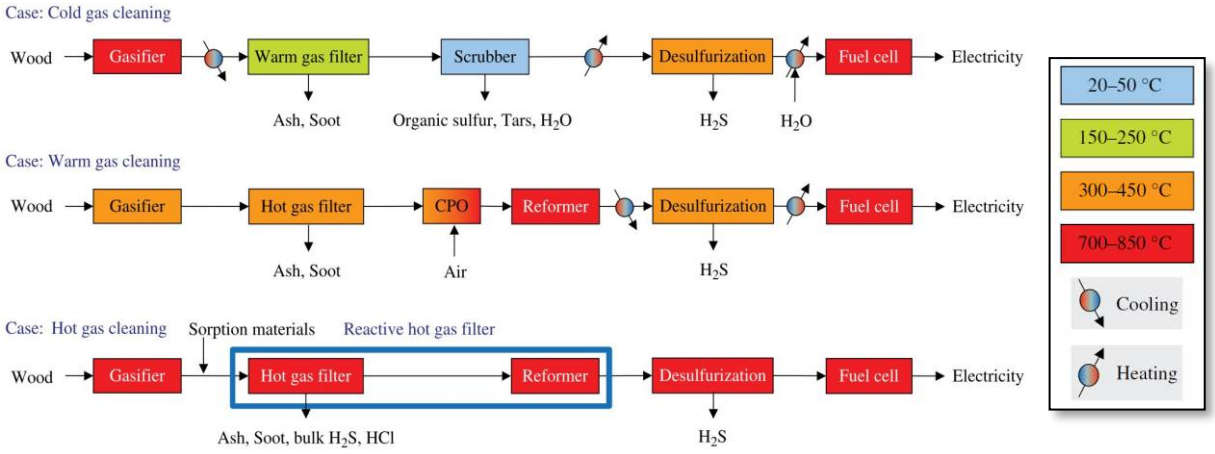

Figure 9: Exemplary cold, warm and hot gas cleaning process chains for fuel cell applications or other synthesis processes, such as methanation [15]

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The hot gas cleaning route is still under research and although promising results for the future could be obtained, there are still barriers that have been preventing the commercial breakthrough of this technology. Rapid catalyst deactivation and difficulties caused by the extreme environments are the main challenges that need to be faced in this context. Warm gas cleaning is a more recent concept, which should comprise the advantages of cold and hot gas cleaning. The temperature lies in between cold and hot gas cleaning but is not clearly defined in literature. Recent research activities with different configurations have shown good results [30], but there is still no commercial deployment available, since the technology is yet to mature. Cold gas cleaning on the other hand is state of the art on a commercial scale and is typically performed as a 'wet' cleanup process, which allows an easy multi-contaminant removal. However, the main disadvantages of this approach are the efficiency penalties to be paid due to the product gas cooling. Furthermore, additional costs arise from the treatment or disposal of the used scrubbing liquid [26,27,43]. A comparison of the different gas cleaning categories is carried out in Table 4.

As an alternative to conventional gas cleaning units different research groups were able to present innovative gas cleaning techniques. Two of those are mentioned here: Thunman et al. [29] carried out experiments with coated heat exchangers, that seem to be able to render expensive wet scrubbers negligible. Their functionality is highly complex and long-term experimental results still have to be collected. Another promising approach was developed by Zhang et al. [44], who combined a mop fan and an electrofilter for the removal of fine particles, tars and chemical contaminants in the product gas.

Table 4: Comparison of cleaning pathways [15,25–27,43]

	Cold gas cleaning	Warm gas cleaning	Hot gas cleaning
Advantages	➢ Well-developed and widely used (especially large scale) ➢ Highly efficient multi-contaminant removal (scrubbers)	➢ Extreme operating conditions are avoided ➢ Water condensation and therefore wastewater treatment is avoided	➢ Destruction of tars ➢ Maintaining the heating value of the gas ➢ High overall efficiency (especially for high-temperature applications, e.g. gas turbines)
Disadvantages	➢ Decrease in overall efficiency ➢ Treatment or regeneration of effluent streams ➢ Increase in cost due to complexity	➢ No commercial use until now	➢ Temperature resistant materials needed ➢ Limited commercial use until now ➢ Rapid catalyst deactivation (for catalytic cracking)

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2.2.3 Product gas quality requirements

The configuration and the extent of product gas cleaning is very much dependent on the quality requirements that have to be ensured for the specific downstream application. For synthesis purposes very low residual impurity levels have to be targeted in order to meet the specified limits of the catalyst. Thus, gas cleaning even plays a greater role if the product gas is used in a synthesis, than it does for the utilization in gas turbines/engine, for instance. An overview of the upper limits of contaminant contents is given in Table 5 for different product gas applications. These values may vary slightly depending on the specific process technology but should give an idea on the relevant magnitudes.

Table 5: Upper limits of product gas contaminants for different process [25–27,44–47]

Contaminant	Unit	Synthesis Application					Gas turbine	Gas engine
		Methanol	Ethanol	FT	Hydrogen	SNG		
PM	*mg/Nm³*	<0.02	0	0	0	0	<20-30	<50
Tars	*mg/Nm³*	<0.01	<0.5[a]	<0.1-1	<1-2	<5	<5-10	<100
Alkali	*ppmv*	<0.005	n.a.	<0.01	n.a.	n.a.	<0.02-0.05	<0.1[b]
N-contaminants	*ppmv*	<0.1-10	<1-10	<0.02-10	<1-10	<30	<50	n.a.
S-contaminants	*ppmv*	<0.5	<1-50	<0.01	<1-50	<0.1[c]	<1-20	<20
Halides	*ppmv*	<0.001	n.a.	<0.01	n.a.	<10	<0.5-1	n.a.

n.a. = not available | a: in ppmv | b: in mg/Nm³ | c: for Ni-catalysts

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3 Fundamentals of Bio-SNG production

3.1 Synthesis of biomass-derived product gas

Biomass-derived product gas can be used for multiple purposes, like the generation of electricity and heat or the synthesis of second-generation biofuels through thermochemical processes. These liquid (e.g. methanol, FT-diesel) and gaseous (e.g. dimethyl ether (DME), H_2 and SNG) biofuels are superior to solid biofuels in several aspects.

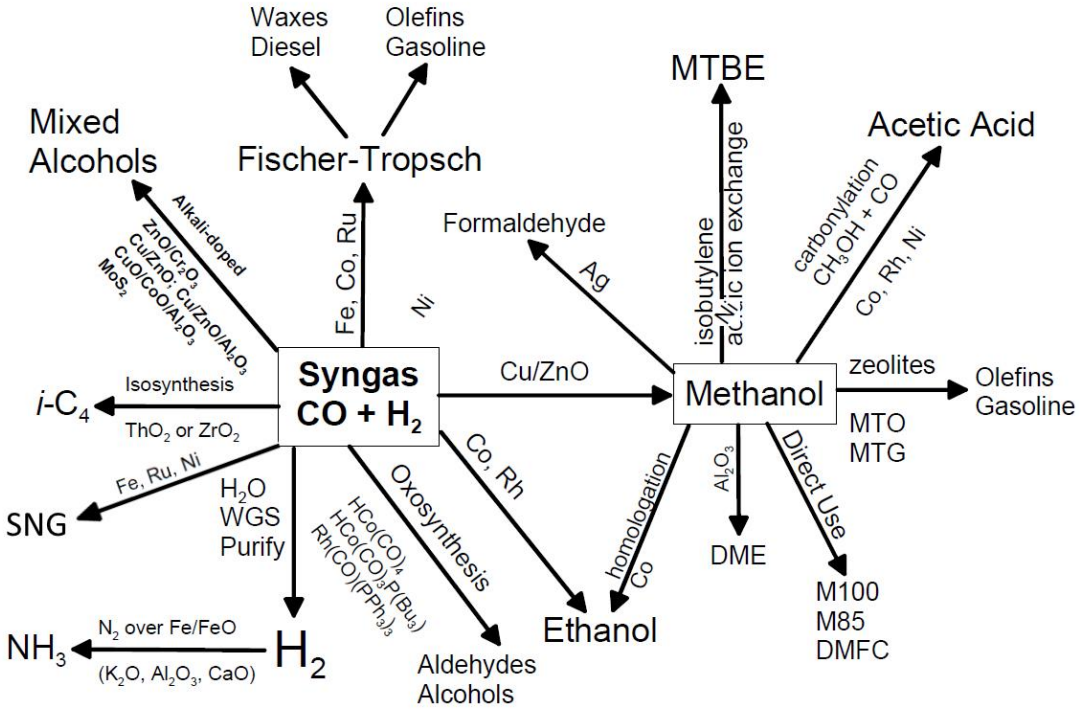

Figure 10: Star diagram showing various high-pressure catalytic processes for converting product gas to fuels and chemicals (adapted from [48])

One big advantage lies in the fact that their market introduction appears to be relatively simple thanks to an existing infrastructure, storage system and transportation system for fossil fuels. Several different synthesis pathways have been subject to research like the FT-synthesis, methanol synthesis, mixed alcohol synthesis (MAS), DME-synthesis and the production of SNG [8,45]. All the different synthesis possibilities and paths based on biomass-derived product gas are illustrated in Figure 10, showing the wide range of possibilities for the chemical conversion of the product gas. Typical reaction conditions (in terms of pressure, temperature and catalyst) as well as the necessary compositions (in terms of CO- and CO_2-ratios) are described in Table 6.

Further details on the synthesis processes and mechanisms can be abundantly found in literature. However, within the scope of this work the focus lies on Bio-SNG production in the following chapters.

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Table 6: Conversion conditions and product gas composition for different biofuels [6,45,49]

Biofuel	Pressure bar	Temperature °C	Catalyst	H_2/CO mol/mol	CO_2 molar unit
Methanol	250-300	350-450	ZnO/Cr_2O_3	3	4-8 %
	50-100	200-300	$Cu/ZnO/Al_2O_3$	2	
Ethanol*	55-65	230-300	Rh catalysts	2	< 1-5 %
	70-105		MoS_2	1-1.2	< 5 %
DME *2-stage synthesis* 1	50-100	200-300	$Cu/ZnO/Al_2O_3$	2	4-8 %
DME *2-stage synthesis* 2	10-20	200-300	γ-Al_2O_3 catalysts;		
DME *1-stage synthesis*	30-70	200-300	Bifunctional catalysts (CuO-ZnO-MnO and zeolite)	1	$CO_2/(CO + CO_2)$ < 0.25
FT	10-40	300-350	Fe catalyst	0.6-1.7; 2**	H_2/CO_2 = 1; 3**
	7-12	200-240	Co catalyst	2.0-2.15	H_2/CO_2 = 3
Hydrogen	1-30	200-1100	Ni, Fe, Mo catalysts	≥ 2***	-
SNG	1-25	200-700	Ni (mainly), Co, Fe, Ru	≥ 3	H_2/CO_2 = 4

* Direct conversion of product gas to ethanol | ** Using potassium as promoter | *** H_2O/CO

3.2 Basic principles of methanation

The methanation reactions are a heterogeneously catalysed synthesis step in the production of SNG, which aim for the delivery of a methane-rich gas. After several upgrading steps this gas can be injected into the existing natural gas grid. Therefore, the intention of the methanation process must be to maximize the CH_4-production through the reaction of H_2 with carbon oxides (CO, CO_2) under the formation of water [15]. The main underlying reactions are called CO- and CO_2-methanation, which are also known as Sabatier reactions. Their reaction equations are shown in Table 7, complemented by all the other reactions occurring in a methanation reactor. Both methanation mechanisms are strongly exothermic, with 2.3 kW (Eq. 3.1) and 1.8 kW (Eq. 3.2) of heat being released for each m^3 of methane produced per hour. Additionally, the reactions result in a strong volume contraction of the gaseous reactants by 50 % (Eq. 3.1) and 40 % (Eq. 3.2) [6].

From Eq. 3.1 it becomes evident that the ratio of H_2/CO in the feed has to be 3:1 for a stoichiometric CO-methanation. However, conventional coal- or biomass-derived product gases usually reach H_2/CO-ratios up to 2:1, which are too low in order to ensure a complete CO-conversion only by Eq. 3.1. In this case the ratio has to be adjusted by enabling the water-gas shift reaction (Eq. 3.3), which converts CO and H_2O to CO_2 and H_2 [50].

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Table 7: Important reactions for methanation [4,15,43]

Name	Reaction formula	Enthalpy of reaction	Equation
Homogeneous reactions (gas-gas)			
Carbon monoxide methanation	$CO + 3\ H_2 \leftrightarrow CH_4 + H_2O$	exothermic	3.1
Carbon dioxide methanation	$CO_2 + 4\ H_2 \leftrightarrow CH_4 + 2\ H_2O$	exothermic	3.2
Homogeneous water-gas shift	$CO + H_2O \leftrightarrow CO_2 + H_2$	exothermic	3.3
Hydrogenation to methane	$C_2H_2 + H_2 \leftrightarrow C_2H_4$	exothermic	3.4
	$C_2H_4 + H_2 \leftrightarrow C_2H_6$	exothermic	3.5
	$C_2H_x + (4\text{-}x/2)\ H_2 \leftrightarrow 2\ CH_4$	exothermic	3.6
Heterogeneous reactions (gas-solid)			
Boudouard reaction	$2\ CO \leftrightarrow C + CO_2$	exothermic	3.7
Carbon methanation	$C + 2\ H_2 \leftrightarrow CH_4$	exothermic	3.8
Heterogenous water-gas shift	$C + H_2O \leftrightarrow CO + H_2$	endothermic	3.9

For the stoichiometric CO_2-methanation, however, a H_2/CO_2-ratio of 4:1 is required. Note that Equation 3.2 can be described as a linear combination of the Equations 3.1 and 3.3. In the thermodynamic equilibrium the CO_2 can be methanated due to the progress of the reverse water-gas shift reaction. From the kinetic point of view, it can be stated that a competition between the CO- and the CO_2-methanation arises. According to [4,6,51] the conversion of CO_2 is inhibited, as long as the CO concentration exceeds a certain threshold, which is close to zero. The reason behind the limited reaction kinetics is assumed to be the favored adsorption ability of CO on the catalyst compared to CO_2. Nevertheless, studies have shown that higher CH_4-yields can be reached, if CO_2 is also present in the reactant product gas [51]. Possible explanations for this phenomenon can be found in [4]. The Equations 3.4-3.6 show the conversion of C_2-species to methane through serial or even direct hydrogenation. Beyond that, the Boudouard reaction (Eq. 3.7) is of great importance, since all hydrocarbons, but especially CO, are known to form carbon on the catalyst surface. This surface carbon can be considered as intermediate in the methanation process (Eq. 3.8, 3.9), but may also lead to catalyst deactivation in form of carbon depositions (fouling) [15,50]. The latter phenomenon is described in detail among other catalyst deactivation mechanisms in [4,52].

The equilibrium of the methanation reactions depends on feed gas composition, pressure and temperature. From the thermodynamic point of view, it can be derived that the methanation is promoted at low temperatures and high pressures [50]. However, at lower temperatures the

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reaction kinetics are rather unfavorable and many catalysts struggle to be still sufficiently active [53,54]. The influence of pressure and temperature on the chemical equilibrium of CO-methanation and the water-gas shift reaction is shown in Figure 11 (a) and analogous to that for CO_2-methanation in Figure 11 (b). Due to the highly exothermic methanation reactions, the heat management turns out to be a major challenge of the process. If the reaction heat is not removed in an efficient way, the temperature in the methanation reactor inevitably rises and the reaction becomes thermodynamically limited. The possibilities of heat removal by the selection of the methanation reactor will be discussed in the following Chapter 3.3 [4].

Although the methanation reactions are favored by elevated pressure, the pressure effect decreases with increasing pressure. An increase from atmospheric pressure to 5 bar has approximately the same positive influence on the methane yield as a further increase to 20 bar would have. It can be concluded that the methanation benefits from slightly increased pressures, but higher pressures are not necessarily required [5].

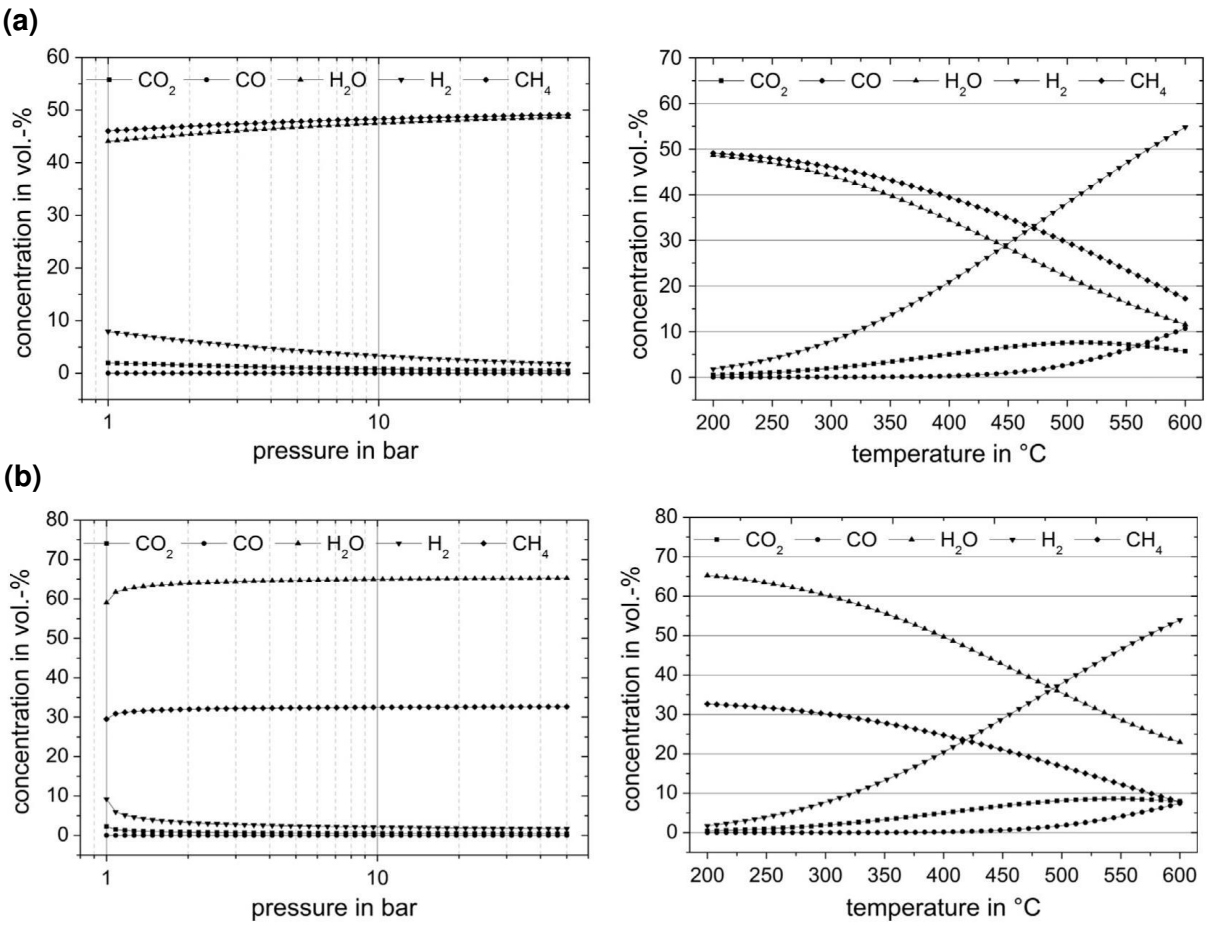

Figure 11: Pressure and temperature influence on the equilibrium composition of (a) CO-methanation (b) CO_2-methanation and water-gas shift reaction for stoichiometric mixtures of H_2 and CO / CO_2 (left: 300 °C, right: 1 bar) [6]

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In 1902 Sabatier and Senderens discovered that transition metals, such as nickel, iron or ruthenium, catalyze the methanation reaction. Table 8 shows these catalytic metals ranked by their activity and selectivity [32]. The activity of a catalyst describes its ability to increase the rate of a specific reaction, while the selectivity indicates its ability to direct a reaction to yield a particular product. The more important methanation catalysts according to Mills and Steffgen [55] are highlighted. A detailed comparison of all the relevant methanation catalysts can be found in [32,43].

Table 8: Catalysts ordered by activity and selectivity for methanation [4,32]

Activity	***Ru***	***Fe***	***Ni***	***Co***	*Rh*	*Pd*	*Pt*	*Ir*
Selectivity	*Pd*	*Pt*	*Ir*	***Ni***	*Rh*	***Co***	***Fe***	***Ru***
	→ *Decrease*							

Nickel was and still is the material of choice due to its advantageous catalytic properties and its relatively cheap price. However, Ni proved to be very sensitive towards product gas contaminants, especially towards sulfur species (e.g. H_2S, COS, organic sulfur) [50]. The tolerable sulfur level for Ni-based catalysts is dependent on several factors, such as the H_2-content and the temperature. Long-term exposal of the catalyst to sulfur enables the formation of a highly stable and also practically irreversible coating on the catalyst surface. This effect leads to a blockade of the active surface and prevents H_2 and CO from accessing the catalyst [43,52]. A summary of regeneration techniques and experiments can be found in reference [15]. The deactivation of methanation catalysts in the presence of sulfur impurities is actually common to all methanation catalysts. The only exception is molybdenum sulfide, which is not mentioned in Table 8 and which in fact even needs a certain amount of sulfur to maintain sulfidity. However, it suffers from rather low activity and requires relatively high conversion temperatures [43,45].

As a matter of fact, the activity of methanation catalysts depends not only on the choice of the active metal itself but also on supports, promotors and preparation conditions. Commonly used supports are metal oxides of large surface area, e.g. Al_2O_3, SiO_2, or TiO_2 [4,6].

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3.3 Reactor types and state of the art

Several methanation concepts have been developed over the last 50 years, of which only a few could reach commercial deployment [6]. Figure 12 gives an overview and indicates the state of development of the so-far available concepts. Based on this categorization, the methanation solutions are subdivided in this chapter into the two more established concepts fixed bed reactors and fluidized bed reactors. Approaches, which are still under research, are briefly discussed under the subitem 'alternative methanation concepts and research activities' [32].

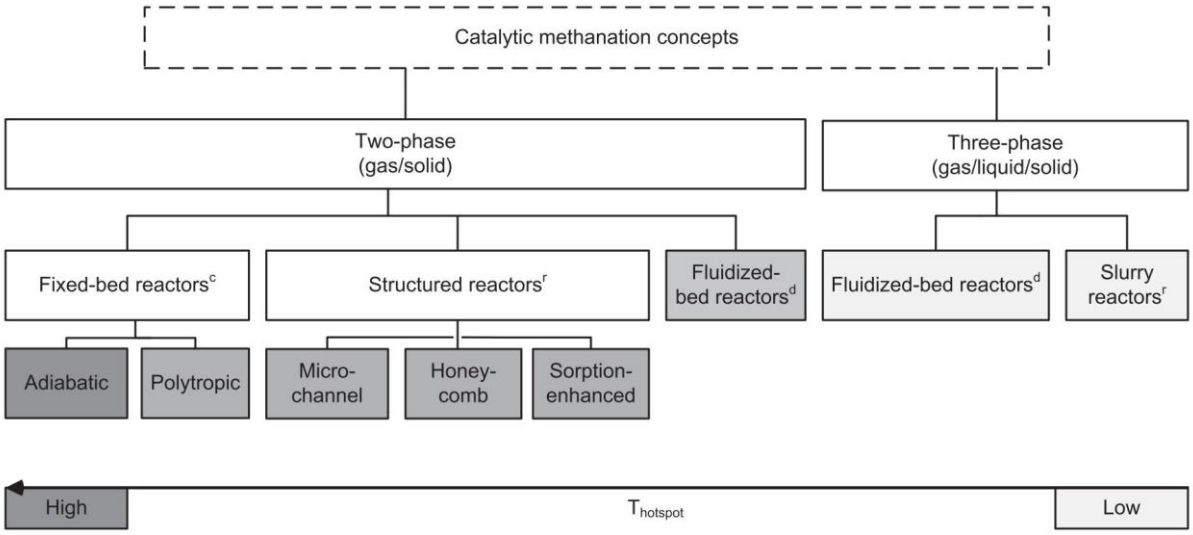

Figure 12: Overview of different methanation concepts; State of development: c – commercial, d – demonstration scale, r – research [6]

3.3.1 Fixed bed methanation

Fixed bed methanation concepts are the best developed and most frequently applied methanation systems. This is partly related to the fact, that in the past they were already used and tested extensively for the removal of CO in the ammonia synthesis and in large scale coal-to-SNG plants [32]. Furthermore, adiabatic fixed bed methanation processes are relatively simple in terms of construction and gas flow control. In general, they are realized as multi-stage processes, where in each of the reactors temperature and conversion are driven to the thermodynamic equilibrium by the exothermic reactions. To overcome this limitation, the exiting gas must be cooled down after each reactor to maintain the driving force for the methanation reaction. By means of an intermittent cooling and gas recycling an almost full conversion can be reached, as shown in Figure 13 (left) in the example of a 5-stage methanation. While the low mechanical stress on the catalyst can be named as one of the main advantages of fixed bed methanation, the challenge of heat dissipation, the limited gas-solid interaction and occurring carbon depositions constitute massive drawbacks [15].

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The production of SNG gained importance during the oil crisis in the late 1970s, which led to the development of several fixed bed methanation solutions for coal-to-SNG processes by companies, like Lurgi, Haldor Topsøe and the Ralph M. Parsons Company [32]. The commercially available technologies are briefly described in Table 9. The Lurgi fixed bed approach, for instance, was demonstrated in pilot plant scale in the project SASOL in Sasolburg (South Africa) and in Schwechat (Austria). From the experience gained in these pilot plants, the first commercial large scale lignite-to-SNG plant was erected in Great Plains, North Dakota, in 1984 [6,15,32,50]. Another state-of-the-art process was developed by the company Haldor Topsøe and is called TREMP (Topsøe Recycle Energy-efficient Methanation Process). The process is depicted in Figure 13 (right) and usually consists of three adiabatic fixed bed reactors in series. It is reported that approximately 80 % of the total heating value of the methanation feed gas is converted into methane. The rest of the energy is released in form of heat and is recovered as high-pressure steam, ensuring a high overall efficiency of the process [56,57]. With the development of a highly temperature stable Ni-based catalyst (between 250 °C and 700 °C) an energy efficient process could be designed with significantly decreased recycle gas rates and optimized heat integration in comparison to other reactor concepts [15,56].

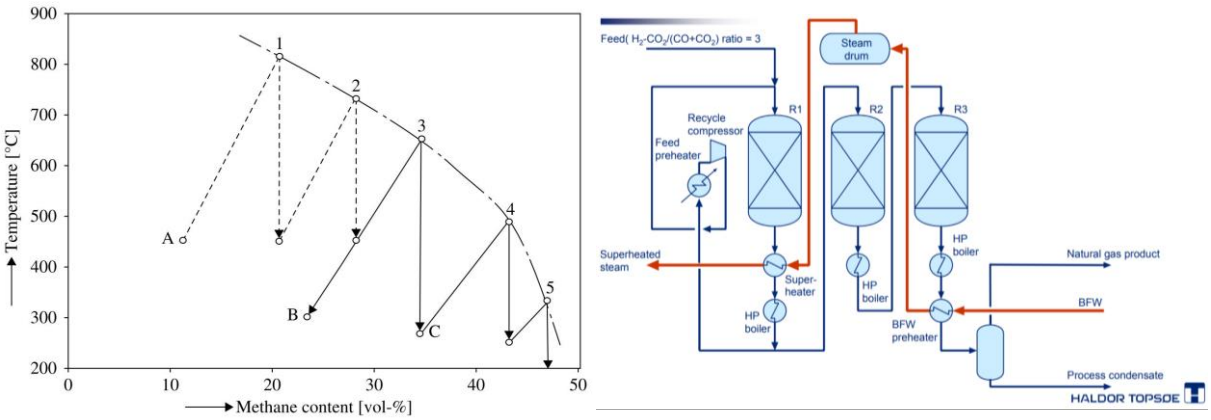

Figure 13: Left: Temperature profiles in series of adiabatic fixed bed reactors with (full lines) and without (dashed lines) product gas recycle [15] Right: Scheme of the TREMP-process [57]

In contrast to that, Foster Wheeler and Clariant developed the so-called VESTA process that does not require a product gas recycle. Instead, in-process CO_2 is used as ballast gas and water vapor is added to dampen the temperature increase [15]. Linde disengaged from the idea of adiabatic fixed beds and developed an isothermal fixed bed reactor. However, this concept has never been brought into commercial application for SNG-production, but is already successfully in operation for the methanol synthesis [6,32].

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Table 9: CO-methanation technologies available on the market [6]

Supplier	Technology name	Concept
Air Liquide (formerly Lurgi)	Lurgi methanation	2 adiabatic fixed bed reactors with gas recycling and intermediate cooling
Haldor Topsøe	TREMP	3–4 adiabatic fixed bed reactors with gas recycling and intermediate cooling
Foster Wheeler and Clariant	Vesta	3 fixed bed reactors with steam addition and without gas recycle
Johnson Matthey Davy Technologies	HICOM	3 adiabatic fixed bed reactors with gas recycling and intermediate cooling
Linde	Linde isothermal reactor	1 isothermal fixed bed reactor with internal contorted heat exchanger

In the last few years, three coal-to-SNG plants have been commissioned in China, the only emerging market for coal-to-SNG plants right now, while further projects are pending. Among these three erected plants, two are based on the TREMP process and one on the process developed by Davy Technologies. Foster Wheeler and Clariant introduced a slightly modified coal-to-SNG process as well, for which a demonstration plant in China is currently under construction. Recent reports indicate that Lurgi (as part of Air Liquid) and Haldor-Topsøe have been conducting further investigations in order to adapt their technologies to the specific conditions arising in biomass-to-SNG conversion processes [15]. In fact, the TREMP process was already applied in a biomass-to-SNG project, namely the GoBiGas biomass gasification demonstration plant in Gothenburg (Sweden), where the production of 20 MW_{SNG} was realized. Further augmentation of the plant capacity to 100 MW_{SNG} was initially planned but the plant was shut down in April 2018 due to economic reasons. For detailed information on the GoBiGas project reference [58] is recommended.

3.3.2 Fluidized bed methanation

Another concept of methanation reactors are fluidized bed reactors, which are known to be suitable for large scale operation of heterogeneously catalyzed reactions with high exothermicity. Nearly isothermal conditions are achieved by an effective heat removal through the fluidization and mixing of the heated particles. Furthermore, smaller catalyst particles compared to fixed beds lead to an increase of the specific surface. Consequently, heat and mass transfer are significantly higher than in fixed bed reactors [15,50]. As a result, it is sufficient to use only one single reactor instead of a reactor cascade. Also the downstream water-gas shift unit can be neglected because the fluidized bed allows for a simultaneous methanation and water-gas shift reaction [59]. On the one hand, the enhanced mixing and collision of the catalyst particles reduce carbon depositions on the catalyst surface. This constitutes a big advantage of fluidized beds not only in connection with catalyst deactivation, but also in terms of process simplicity. Unlike for fixed bed methanation reactors, it is not

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necessary to remove or convert the unsaturated hydrocarbons upstream of the methanation unit anymore. On the other hand, the intense particle contact also causes severe attrition of the catalyst [4,60]. This issue is tried to be solved amongst others by the Chinese Academy of Sciences, which is aiming to develop catalysts specially designed for fluidized bed reactor conditions [61,62]. Apart from catalyst attrition, the entrainment of particles and the limited operating range can be named as the major disadvantages of the fluidized bed concept [4,63].

Kopyscinski et al. [50] describe the history of fluidized bed methanation in detail, including the multiple-feed fluidized bed reactor developed by the Bureau of Mines (United States Department of the Interior) in 1952, the Bi-Gas project initiated in 1963 by Bituminous Coal Research Inc. focusing on SNG production from coal and finally the Comflux process in Germany with the successful realization of a simultaneous water-gas shift and methanation reaction in the same reactor. The Comflux process was developed by Thyssengas GmbH and the University of Karlsruhe and led to the erection of the biggest fluidized bed methanation reactor so far (20 MW_{SNG}). The concept is shown in Figure 15. However, all three of the above mentioned fluidized methanation projects were terminated due to economic considerations or fluidization problems [62,63].

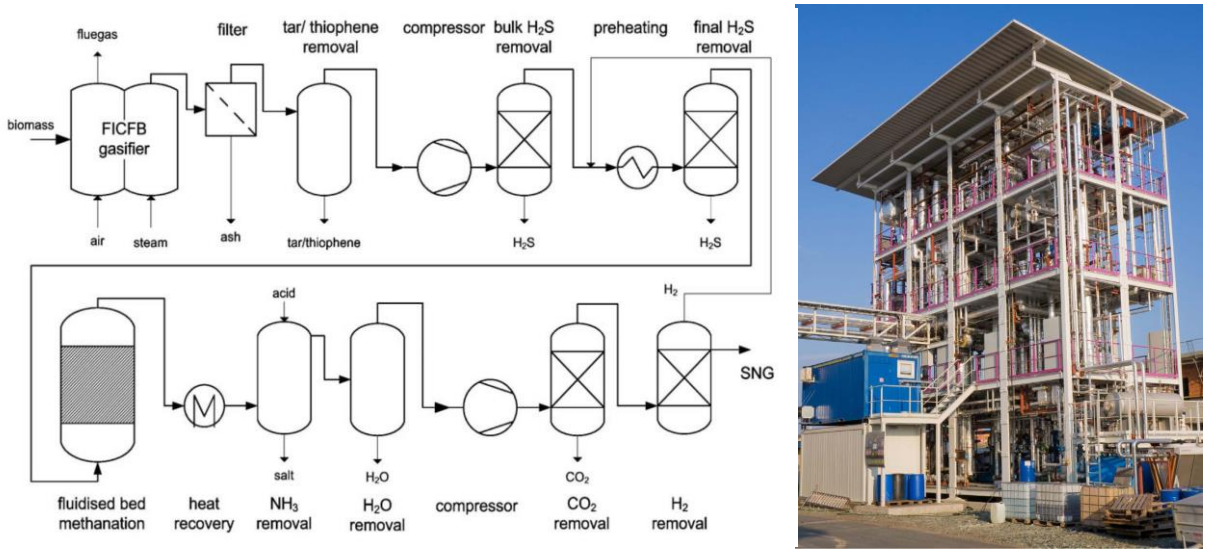

Figure 14: Block flow diagram (left) and picture (right) of the 1 MW_{SNG} process development unit (PDU) in Güssing, Austria [50]

Around the turn of the millennium the Comflux process was revitalized and adapted by the Paul Scherrer Institute (PSI, Switzerland). During their research, it was demonstrated that besides the simplicity of the process, this reactor type has various advantages for the conversion of gasification-derived product gas to SNG. These results were also supported by several other research groups, which have drawn similar conclusions in comparative studies with fixed bed reactors. Under the same conditions higher CH_4-yields, higher CO-conversion, decreased carbon deposition, lower bed temperature and avoidance of hot spots were

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observed with fluidized beds [61,64,65]. The initial results of the research conducted by PSI led to the assessment that the DFB gasification process developed at TU Wien (Austria) seems to be the most promising technology for Bio-SNG production. This process was implemented and commercialized by Repotec in Güssing (Austria) in 2002. Until the end of 2004 comprehensive investigations were conducted on a fully automated 10 kW_{SNG} reactor system named 'Cosyma' with a slip stream from the Güssing plant. It was observed that the organic sulfur species were the primary cause for rapid catalyst deactivation and that improved sulfur removal in the gas cleaning stage increased catalyst lifetime significantly [32,50,63]. In view of this, catalyst stability of more than 1000 h could be reached on the same bench scale test rig through process adaptations [4,32,50]. Based on this success, in 2009 a 1 MW_{SNG} Process Development Unit (PDU, Figure 14) was erected by Conzepte Technik Umwelt AG (CTU, Switzerland) and Repotec Umwelttechnik GmbH within the European Union funded project 'Bio-SNG' and commissioned with the support of the TU Wien, PSI and Biomassekraftwerk Güssing. The gasification plant was operated with a load range from 30 to 100 % and converted up to 20 % of its product gas to high-quality SNG with a cold gas efficiency of 61 % [15,50,57,60]. The methanation section was operated at temperatures around 350 °C and at H_2/CO-ratios in the range of 1 to 5 [12,57]. Additionally, the suitability of the produced SNG as car fuel was shown by feeding a compressed natural gas fueling station [15,50]. Through the successful operation of the PDU in Güssing the demonstration of the complete process chain from wood-to-SNG comprising the gasification, gas cleaning, methanation and gas purification could be carried out in a half-commercial scale.

For further optimization of the process a pilot plant with the power production capacity of 160 kW_{SNG} is planned to be constructed in Villingen (Switzerland) by PSI [6]. The combination of a dual fluidized bed gasifier with fluidized bed methanation is also subject of research within the French project GAYA, which aims at the SNG production from lignocellulosic biomass in the scale of 20-80 MW_{SNG} [15].

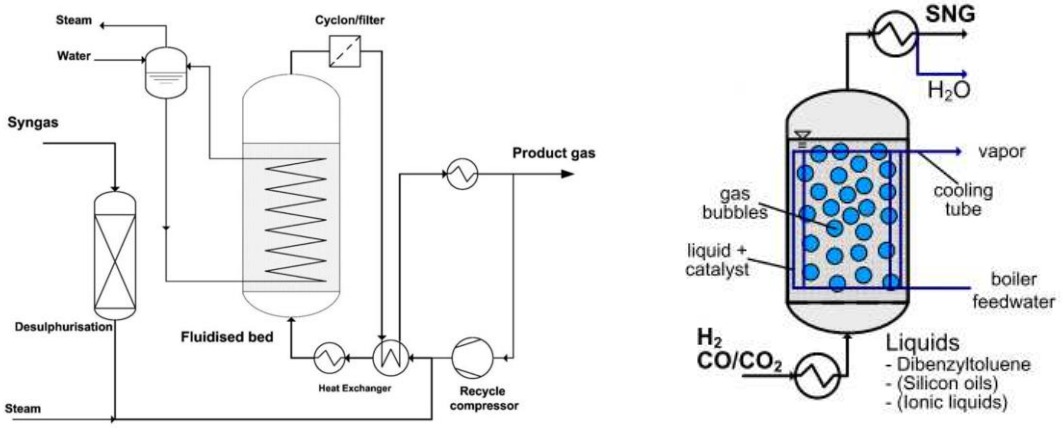

Figure 15: Comflux (Thyssengas) process flow diagram (left) [50,63] and schematic of a three-phase methanation reactor (right) [66]

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3.3.3 Alternative methanation concepts and research activities

Addressing the challenges and drawbacks of state of the art methanation concepts, several research groups have been developing and investigating different innovative methanation approaches. In Figure 12 these technologies are marked with an ‘r’, indicating that they are still in a research stadium and their feasibility has not been tested in larger scales yet.

The slurry reactor or bubble column (Figure 15) is a three-phase methanation reactor and represents one of these concepts. By introducing a liquid phase into the fluidizing reactor, the formation of hot spots can be avoided through the buffering ability of the liquid phase. Additionally, it also serves as a heat reservoir if frequent start-ups and shut-downs are necessary, e.g. in power- to-gas applications [15]. The disadvantages in connection with the liquid phase are the additional gas-liquid mass transfer resistance as well as the possible decomposition and evaporation of the heat transfer fluid [66]. This Liquid-Phase Methanation (LPM) concept was developed by Chem Systems Inc. in the 1970s and consisted of only one three-phase reactor operating at pressures up to 70 bar [6]. On basis of that, a pilot plant was implemented, which showed low gas conversion and high catalyst loss from the bubble column. As a consequence, the project was terminated in 1981, although some research groups still conduct investigations in this field up to today [6,50].

Other process optimization approaches try to modify fixed beds in order to overcome the heat dissipation difficulties and to lower pressure drops. Thus, investigations in structured reactors have been conducted, which should have the ability to tackle both above mentioned challenges. Reports indicate that the structured catalyst improves radial heat transport by two or three orders of magnitude. Microchannel reactors and especially honeycomb catalysts with their high surface to volume ratio seem to be both promising optimized variants of fixed bed methanation [6,67]. The concept of sorption enhanced methanation constitutes another development of structured fixed bed reactors. The special feature of this approach is the removal of water produced during the methanation reaction by means of an adsorption on a catalyst support. By removing a product, the equilibrium can be pushed further to the product side and the methane yield can be enhanced. The method of choice for the sorbent regeneration depends on its material properties, but mainly comprises temperature and/or pressure swing technologies [6,68]. Im et. al [53] also conducted research on sorption enhanced methanation, but instead were focusing on the removal of CO_2 from the methanation reactor, promoting the water-gas shift reaction in this way. Liu et al. [64] on the other hand worked on a two-stage methanation variant based on a combination of a fast fluidized bed main reactor and a fixed bed clean-up reactor to ensure a complete conversion of CO.

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3.3.4 Comparison of methanation concepts

To provide a summary of this essential chapter, a comparison of three different methanation reactor types is carried out in Table 10. Besides fluidized and fixed bed reactors, the bubbling column was selected as third reactor type, because it differs more from the first two concepts than other optimized variants. More detailed comparisons including other reactor types can be found in the references [6,15].

Table 10: Comparison of Different Methanation Reactor Concepts [6,15,69,70]

Parameter	Adiabatic fixed bed	Fluidized bed	Bubbling column
Operation mode	Adiabatic	~ Isothermal	Isothermal
Reactor stages	2-7	1-2	1-2
Gas recycling	Usually	Sometimes	No
Temperature range (°C)	250-700	300-400	300-350
Particle size (µm)	>1000	100-500	<100
Limitation	Heat transfer; Thermal stability of catalyst	Catalyst attrition	Gas-solid mass transfer; Thermal stability of liquid
Mechanical stress of catalyst	Low	High	Moderate
Thermal stress of catalyst	High	Low	Low
Complexity of reactor	Low	Medium-high	Medium-high
Complexity of process set-up	High	Low	Low
Process costs	High	Low	Low-medium
Gas hourly space velocity (GHSV)	Medium-high	Medium	Low-medium
Temperature level for heat recovery	High	Low	Low
Heat control	Difficult	Medium	Easy
Mass transfer	Medium	High	Low-medium
Tolerance of unsaturated hydrocarbons	No	Yes	?
Technical readiness level	9	7	4-5

3.4 Product gas upgrading and quality requirements of the natural gas grid

After methanation, the gas stream leaving the reactor does not fulfill the quality specifications of the natural gas grid and its quality needs to be improved by gas upgrading steps. In detail, this means that a removal of CO_2, H_2O, H_2, and possibly also N_2 needs to be carried out in order to be able to inject it [5,15]. If a stoichiometric or overstoichiometric gas mixture is fed to the methanation reactor, ideally only a separation of H_2O and perhaps H_2 is necessary (e.g. TREMP process). H_2O-removal is achieved similarly to natural gas drying by condensation, adsorption, absorption, or membrane separation. Absorption is usually carried out with calcium chloride ($CaCl_2$), lithium chloride (LiCl), and glycol solutions, with the latter being the most commonly used absorbent. Solid materials, like aluminium-based adsorbents, activated carbon, molecular sieves, and silica-based adsorbents are capable of reducing the moisture content of gases through a pressure swing adsorption (PSA) or temperature swing adsorption (TSA) [15]. For a small scale process Tremel et al. [5] propose the combination of a

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condensation unit operating at ambient temperature and a following adsorption step to be an economically reasonable solution.

A possible way for the separation of H_2/CH_4 gas mixtures is the adsorption within a PSA process, for example on activated carbons. The deployment of membrane techniques for the separation of H_2/CH_4- and N_2/CH_4-mixtures was also investigated by a few studies [15].

Furthermore, a significant amount of CO_2 can be present in the raw SNG. For instance, in the fluidized bed methanation process developed by the PSI the water-gas shift reaction is used in situ in the methanation reactor to adjust the H_2/CO-ratio by pushing the equilibrium to the CO_2-side [15]. Other processes leave a surplus of CO_2 in the methanation feed gas to serve as a diluting component, which can dissipate some of the reaction heat (e.g. VESTA process). However, in the latter case it has to be taken into account that a diluted product gas stream is also related to a higher energy effort if an upstream gas compression is performed [6]. In these cases an additional CO_2-separation step has to be considered. CO_2-removal techniques include PSA, pressurized scrubbers (e.g. amine or water scrubbers), membrane systems, or cryogen units. For small scale plants the PSA is used predominantly, while scrubber systems are becoming economically attractive for higher gas volume flows [5]. An overview of the most common technologies and an assessment of their energy demand, methane recovery and investment costs is given in [71].

Table 11: Requirements for feeding into the natural gas grid in Austria [72]

ÖVGW-DIRECTIVE G31		
Combustion characteristics		
Wobbe-index	*kWh/Nm³*	13.3 – 15.7
Higher heating value	*kWh/Nm³*	10.7 – 12.8
Relative density	-	0.55 – 0.65
Accompanying substances		
Hydrocarbons dew point	-	max. 0 °C at operating pressure
Water vapour dew point	-	max. -8 °C at 40 bar
Oxygen (O_2)	*vol. %*	≤ 0.5
Carbon dioxide (CO_2)	*vol. %*	≤ 2
Nitrogen (N_2)	*vol. %*	≤ 5
Hydrogen (H_2)	*vol. %*	≤ 4
Total sulfur	*mg/Nm³*	≤ 10 (on long-term)
Mercaptan sulfur	*mg/Nm³*	≤ 6
Hydrogen sulfide (H_2S)	*mg/Nm³*	≤ 5
Carbonyl sulfide (COS)	*mg/Nm³*	≤ 5
Halogen compound	*mg/Nm³*	0
Ammonia (NH_3)	-	technically free*
Solids and liquids	-	technically free*
ÖVGW-DIRECTIVE G33		
Methane (CH_4)	*mol %*	≥ 96
Total silicium (Siloxane, Silane)	*mg/Nm³*	≤ 10

* 'technically free' ensures the continuous operation of gas appliances and equipment with standard or customary design

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For more extensive information on the current state of the art of SNG upgrading Schildhauer et al. [15] is recommended. In the stated reference a detailed description and subsequent comparison of the upgrading technologies by a techno-economic assessment can be found.

If the upgraded gas stream complies with all the necessary quality requirements, the SNG is compressed to grid pressure and fed to the natural gas grid as last step. The level of compression is dependent on the location of the production plant and the grid into which the SNG is to be fed. While local distribution networks e.g. in Sweden reportedly have a pressure of up to 10 bar, large transmission pipelines are even pressurized up to 80 bar [71]. The natural gas quality requirements differ between countries but plans for a standardization of gas quality within the European Union members are in progress [71]. A comparison of quality criteria of some European countries can be found in [72]. For Austria the quality requirements for natural gas are specified in the ÖVGW-Directive G31. Additionally, the criteria set by the ÖVGW-Directive G33 have to be satisfied, if upgraded biogas is to be injected to the grid. A summary of the quality requirements of these two directives are displayed in Table 11.

3.5 R&D challenges

In the last 15 years, research on the efficient use of biomass has been attracting a lot of attention throughout Europe, triggering a deep investigation of biomass-to-SNG processes [15]. New combinations of gas cleaning and methanation units are being developed with focus on the reduction of product gas cleanup complexity and the integrated conversion of light hydrocarbons (e.g. benzene and toluene) to minimize substance losses and to increase the fuel-to-SNG efficiency. The majority of the lab-scale research projects in this field still work on the optimization of fixed bed methanation reactors. However, a growing part of the community puts effort in the development of fluidized bed methanation, three-phase methanation or structured methanation reactors [73]. While in the field of coal-to-SNG there are only very few open question marks in research, it is different in the case of biomass-to-SNG processes and power-to-gas applications: There are still key questions which demand further investigation by pilot/demo scale experimental projects and concomitant reactor simulation and modeling. The scale-up and simulation of adiabatic fixed bed reactors is generally solved, however for other reactor types the lack of results still complicates scale-up and the prediction of the reactor behavior [15].

The improvement of methanation catalyst lifetime is certainly one of the main topics which occupies research groups. This is closely related to the issue of optimizing gas cleaning technology solutions, which need to protect the catalyst from deactivating substances (e.g sulfur compounds, carbon depositions). For future power-to-gas applications, frequent start-up and shut-down cycles as well as efficient part-load operation may be a necessary feature

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of methanation units [15].

Another key problem of methanation has been the effective removal of the reaction heat released in the methanation process to avoid sintering of the catalyst and to cope with thermodynamic limitations at higher temperatures. Thus, some research groups try to design methanation reactors, where a heat removal without costly equipment or high gas recycle rates is ensured (e.g. fluidized bed methanation) [43,62].

For the future it is not expected that one reactor concept will outperform the others for all types of applications. Instead, there will be various efficient methanation technologies, out of which it will be possible to choose the most suitable one depending on the application. In this selection of methanation solutions at least adiabatic and cooled fixed beds as well as fluidized bed methanation reactors are expected to play a major role [15].

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4 Applied principles and methods

Within the scope of this work lies not only the development of a process chain for Bio-SNG production from low-grade biogenic fuels but also the subsequent evaluation of the concept. For this purpose, the applied approaches and methods are discussed in the following chapter.

4.1 Gasification experiments with sewage sludge

As representative for low-grade fuels sewage sludge was identified as a suitable fuel. The decision was made in favour of sewage sludge due to its economic advantages (see Chapter 6) on the one hand and the experience gained by two different relevant gasification experiments on the other hand. The second experiment was carried out during the period of this thesis and served together with the previous experiment as basis for the simulation part of this work. In Figure 16 the process of fuel pelletizing and the fuel pellets are shown. This fuel treatment needed to be performed before the gasification experiment but is not ultimately necessary for an industrial scaled gasification processes.

Figure 16: Pelletizing of the sewage sludge for the gasification experiment (2019)

An overview of both sewage sludge experiments, a reference experiment with wood pellets and the chosen data for the simulation are displayed in Table 12. More detailed information on the experimental set-up and the operational parameters can be found in [74]. As can be seen, the majority of the simulation input data was taken from the older gasification experiment, even though it was run with the old gasifier design. This is due to the greater accordance with conventional product gas compositions that are expected from biomass gasification. However,

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considering the experience gained from biomass gasification experiments the product gas content of NH_3 seemed too low. Therefore, the data from the more recent experiment were used in the case of NH_3, which matched a lot better with the expected values [75]. All the components that were not explicitly measured during the test runs (HCl, COS, HCN, N_2) were inserted according to typical literature values, which relate to the respective element content in the fuel and its subsequent conversion behavior in the gasifier [14,27,76]. For COS this calculated value was added up to the H_2S-content as the simulation software does not support other sulfur gas compounds than H_2S. In the end, the product gas components were normalized to a sum of 100 %, which explains the slight deviations from the base values.

Table 12: Product gas compositions from different gasification experiments

	Parameter	Unit	Classic 100kW$_{th}$ gasifier (2013)	Classic 100kW$_{th}$ gasifier (2012)	Advanced 100kW$_{th}$ gasifier (2019)	Reference data for simulation
Gasification parameters	Fuel	-	Wood pellets [77]	Sewage sludge pellets [78]	Sewage sludge pellets [79]	Sewage sludge pellets
	$T_{Bubbling\ bed}$	°C	849	810	815	810
	T_{Column}	°C	-	-	931	-
	Steam to fuel ratio	kg/kg	0.9	0.9	1.63	0.9
	Gasification agent	-	steam	steam	steam	steam
	Bed material	-	olivine	olivine	80% olivine / 20% limestone	80% olivine / 20% limestone
Main components	H_2O	vol%	34.8	48	62	55
	H_2	$vol\%_{db}$	43.3	39	30.3	39.72
	CO	$vol\%_{db}$	26.6	16.5	11.7	16.8
	CO_2	$vol\%_{db}$	17.2	25.5	31.1	25.97
	CH_4	$vol\%_{db}$	8.2	7.8	10.0	7.94
Minor components & impurities	C_2H_4	$vol\%_{db}$	1.45	2.4	2.15	2.44
	C_2H_6	$vol\%_{db}$	0.1	0.1	0.1	0.1
	C_3H_8	$vol\%_{db}$	0.11	0.6	0.0	0.61
	N_2	$vol\%_{db}$	0.5	-	9.34	1.01
	H_2S	$ppmv_{db}$	16	5300	13200	6000
	COS	$ppmv_{db}$	-	-	-	
	NH_3	$ppmv_{db}$	500	22700	42800	43600
	HCN	$ppmv_{db}$	-	-	-	4400
	HCl	$ppmv_{db}$	-	-	-	70
	Fly dust	g/Nm^3_{db}	4.1	68.4	8.05	68.4
	Fly char	g/Nm^3_{db}	15.7	3.2	1.14	3.2
	Tar grav.	g/Nm^3_{db}	2.4	3.9	4.75	10
	Tar GCMS	g/Nm^3_{db}	3.5	8.2	16.4	

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4.2 Process simulation with IPSEpro

Process simulation is a very useful tool for the prediction of plant and process behavior by means of mathematical models. In the context of this work the equation-oriented steady state simulation environment IPSEpro was used, which was originally developed for the simulation of power plants. The advantages and features of IPSEpro in comparison with other process simulation software are discussed in [80].

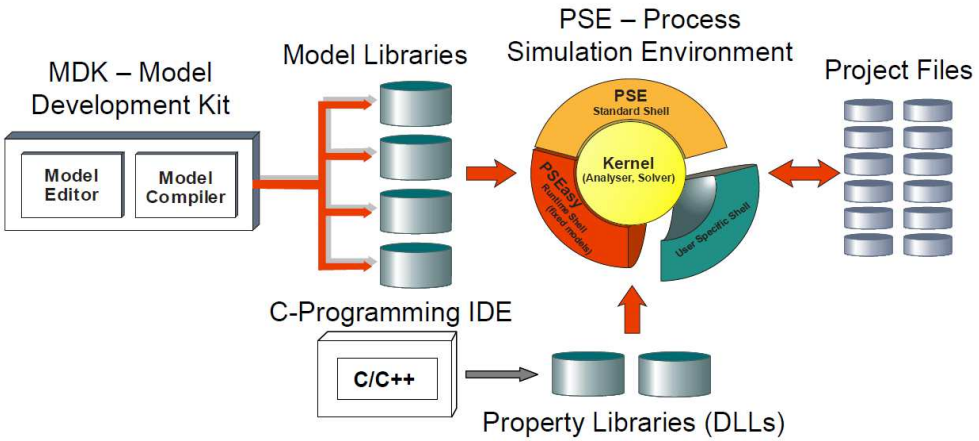

Figure 17: Structure of the simulation software IPSEpro [80]

The modular structure of IPSEpro is shown in Figure 17. The Process Simulation Environment (PSE) module serves as the graphical interface, where the simulation flowsheet is developed. It calls the equation solver, which in turn calculates the result data of a simulation flowsheet. The model equations of the process components are lodged in the Model Development Kit (MDK) and can be edited in this area [80]. Those single-unit models are grouped in model libraries and contain all the process components which are provided to the PSE for further calculations. The required substance data are taken from dynamic link libraries (DLL), which IPSEpro accesses during the calculation [13]. Initial values, process parameters and the set-up of a flowsheet can be saved and called up through project files [80].

Apart from the utilized initial data set, the significance and trustworthiness of the simulation results depend heavily on the accuracy of the applied model units. All models are based on mass- and energy balances, which together form an equation system. For biomass gasification a detailed biomass gasification library (BG_Lib) together with a DLL file (SimProp) was developed by [80]. Throughout the years the library has been extended and optimized in the context of several dissertations and master theses [32,81,82]. Also in the course of this work several new units have been introduced to the library which are listed in the Appendix A.

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4.3 Economic evaluation tools

4.3.1 Plant cost estimation

By the time an industrial plant is ready to go into operation, already a large amount of money will have been invested for the erection of the plant. This capital investment is made up of the following two components [83–85]:

$$Total\ capital\ investment = Fixed\ capital + Working\ capital \qquad \text{Eq. 4.1}$$

While the fixed capital covers all the costs associated with building the plant (land, design and engineering, equipment, freight, etc.), the working capital is the amount of capital which is required to start up the plant and finance the first period of operation before revenues from the process are generated [84,85].

Numerous techniques are available for estimating the fixed capital investment of a plant. However, they can be classified in five generally accepted classes that are most likely to be encountered in the process industries. Table 13 sums up the most important features of these methods.

Table 13: Classification and quality of estimates [83,86–88]

Estimate type	Information basis	Accuracy range (%)	
		Lower limit	Upper limit
Order of magnitude estimate	Cost data of previously built similar plants	-30	+50
Study estimate	Major items of equipment	-25	+30
Preliminary estimate	More detailed equipment and process data than study estimate	-20	+25
Definitive estimate	Considerable data prior to completion of drawings and specifications	-10	+20
Detailed estimate	Complete drawings, specifications, and site surveys	-5	+10

In this work both order of magnitude and study estimate methods were deployed. In particular the latter one is well suited for feasibility studies according to [85,88]. In order to obtain the capital cost of a chemical plant through a study estimate, the costs associated with the major plant equipment have to be known. A common and convenient method for equipment cost estimation is the so-called six-tenths rule, which relies on a logarithmic relationship between the equipment costs (C_2, C_1) and characteristic attributes of two similar equipment items [85]. It is shown in Equation 4.2.

$$C_2 = C_1 \cdot \left(\frac{S_2}{S_1}\right)^n \cdot \frac{I_2}{I_1} \qquad \text{Eq. 4.2}$$

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With this equation it is possible to calculate the cost of an equipment item of a different size (S_2) when its cost is known for a given size (S_1). Depending on the equipment type, the exponent n varies usually between 0.4 and 0.8 with an average value of about 0.6. If the exponent is unknown, this average value can be used [86]. The basis of this rule is a concept referred to as the economy of scale. It predicts a decrease of the cost per unit of capacity with an increase in equipment size, which is reflected in the exponent n<1. As the exponent approaches the value of 1, the effect of the economy of scale shrinks [85,86]. In general, an estimation is always based on cost data of previous years. Thus, the inflation affecting the chemical process industries has to be taken into account through cost indices (I_2, I_1), which update the cost level to the point of estimation. The most frequently used ones are the Chemical Engineering Plant Cost Index (CEPCI) and the Marshall and Swift Equipment Cost Index (M&S) [88–90].

To derive the fixed capital cost of the plant from the sum of equipment cost the so-called Lang method was applied. It was developed by Lang [91] in 1948, who proposed a factor based upon the type of processing plant. This factor is used for the multiplication of the delivered equipment cost to determine all the direct and indirect costs in connection with the equipment (Eq. 4.3 [92]). Over time, this Lang factor was updated and optimized by different authors [83,92].

$$Fixed\ capital = Lang\ factor \cdot \sum (Delivered\ equipment\ cost) \qquad \text{Eq. 4.3}$$

For study and preliminary methods the working capital is usually calculated as a percentage rate of the fixed capital investment. Ranges between 10 and 25 % are reported in literature [83,85–87].

4.3.2 Investment decision - Net Present Value

The construction of a new plant entails on the one hand high initial investment costs but on the other hand the expectation for maximal profits through high operational hours over a long period of time. Therefore, the decision about the final realization of such a project is inevitably connected to economic risks and uncertainties [13]. Various techniques of economic analysis exist to assess the profitability and risk of projects involving both capital expenditures and yearly operating costs [83–85,87].

The most commonly used investment analysis tool is the net present value (NPV) method - also known as net present worth (NPW) method which, unlike other methods, has few disadvantages and treats the time-related value of money and its effect on project profitability properly [86,93]. The net present value stands for the cumulative discounted cash position at the end of the project and indicates if a pending investment is favourable or not. In this method

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all the future cash flows need to be discounted back to the beginning of a project (time = 0) with a chosen discount rate. Thereby, each annual cash flow (revenues (R) minus expenses (E)) is discounted one by one with the discount rate i. If the simplifying assumption is made that the annual cash flow remains unchanged in the considered period, also a cumulative discount factor (CDF) can be calculated (Table 14, Eq. 4.4). Under these conditions the NPV can be obtained with Equation 4.5, where the initial investment volume (I_0) is compared with the cumulative discounted cash flow [85,94,95].

Table 14: Calculation formula for the net present value method [95,96]

$$CDF = \frac{(1+i)^n - 1}{i \cdot (1+i)^n} \quad \text{Eq. 4.4}$$

$$NPV = -I_0 + (R - E) \cdot CDF = -I_0 + (E - C) \cdot \left(\frac{(1+i)^n - 1}{i \cdot (1+i)^n}\right) \quad \text{Eq. 4.5}$$

NPV	net present value in €
I_0	capital expenditure in €
i	discount rate in %/100
n	useful lifetime in a
R	annual revenues in €/a
E	annual expenditures in €/a
CDF	cumulative discount factor

If the NPV for an individual project at a fixed discount rate is greater than or equal to 0, the investment seems to be economically favourable. However, it is also possible to compare several investment variants [95]. For this purpose the set-up of a business case is expedient, as it allows a clear comparison between different scenarios for achieving a specific goal. The basis is a so-called reference case, which serves as a "benchmark" for the alternative options.

A useful modification of the so-far presented absolute NPV method constitutes the relative NPV method. Here the reference case is set as a zero variant with an NPV of 0. The NPV of each alternative scenario is placed in relation to the reference case to have a direct comparison between the scenarios [94]. The equations connected to the relative NPV method are listed below in Table 15 (Equations 4.6-4.8).

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Table 15: Calculation formula for the relative net present value method [95,96]

$$\Delta I_0 = I_{0,var,0} - I_{0,var,x} \quad \text{Eq. 4.6}$$

$$\Delta CF = \left(R_{var,0} - E_{var,0}\right) - \left(R_{var,x} - E_{var,x}\right) \quad \text{Eq. 4.7}$$

$$NPV_{rel} = -\Delta I_0 + \Delta CF \cdot CDF \quad \text{Eq. 4.8}$$

NPV_{rel}	relative net present value in €
ΔI_0	difference in capital expenditure in €
ΔCF	difference in cash flow in €/a
$I_{0,var,0}$	capital expenditure for the zero variant (reference case) in €
$I_{0,var,x}$	capital expenditure for the alternative scenario ‘x’ in €
$R_{var,0}$	annual revenues of the zero variant (reference case) in €/a
$R_{var,x}$	annual revenues of the alternative scenario ‘x’ in €/a
$E_{var,0}$	annual expenditures of the zero variant (reference case) in €/a
$E_{var,x}$	annual expenditures of the alternative scenario ‘x’ in €/a
CDF	cumulative discount factor

In Figure 18 the relative net present value (NPV_{rel}) is plotted in a diagram depending on the discount rate (i). It can be seen that the zero variant (reference case) always exactly corresponds to the zero line of NPV_{rel} while all alternative scenarios vary in relation to the reference case with a changing discount rate. The exemplary diagram in Figure 18 shows that the first scenario is always more advantageous than the zero variant in the considered discount rate interval. The second scenario, however, would only be profitable compared to the reference case if a discount rate below 5 % could be guaranteed. The direct comparison of the two scenarios leads to the conclusion that the second scenario is only preferable over the first scenario with discount rates below 2 % [94,95].

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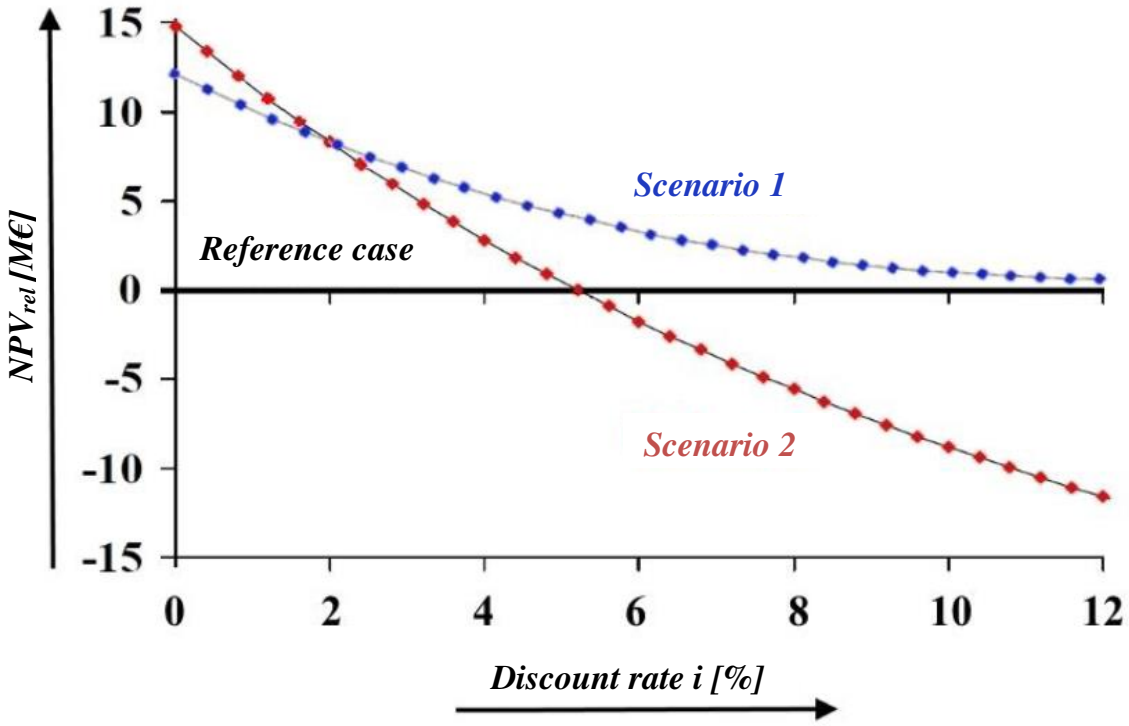

Figure 18: Comparison of two investment scenarios with a zero variant using the relative net present value in dependency of the discount rate [95]

4.3.3 Levelized cost method

A comparison of net present values is often not the desired outcome of investment calculations for projects in the energy sector. Instead, the actual profitability is measured by means of specific costs per product unit [96]. The energy production costs are calculated by putting all incurring costs in connection with the construction and operation over the useful life of the plant in relation with the total amount of energy generated in that period of time [97]. While in literature different terms exist to name this value, in the course of this work the terms Levelized Cost of Energy (LCOE) and Levelized Cost of Product (LCOP) are used. If the assumption from the previous chapter is made again that the annual expenditures and the annual energy output remain constant in the considered lifetime period, the LCOE can be calculated by Equation 4.9 from Table 16. The LCOP is defined in a similar way, the only difference being that all revenues generated by other income sources than the main product are included in the calculation as well. This allows the determination of the specific production costs of a certain product. The formula for the LCOP calculation is given in Equation 4.10.

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Table 16: Calculation formula for the levelized cost method [97]

$$LCOE = \frac{\left(\frac{I_0}{CDF}\right) + E}{AP} \quad \text{Eq. 4.9}$$

$$LCOP = \frac{\left(\frac{I_0}{CDF}\right) + E - R_{emp}}{AP} \quad \text{Eq. 4.10}$$

LCOE	Levelized Cost of Energy in €/MWh
LCOP	Levelized Cost of Product in €/MWh
I_0	capital expenditure in €
E	annual expenditures in €
R_{emp}	annual revenues which derive from other sources than selling the main product in €
AP	annual energy production in MWh
CDF	cumulative discount factor

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5 Process design and simulation of the sewage sludge concept

To find an answer to the research question how an optimized process concept for the production of Bio-SNG from residual fuels should look like, a plant concept was developed based on the DFB gasification. The first part of the concept is set up very similar to conventional DFB co-generation plants which are mentioned in [13]. However, instead of feeding the product gas into a gas engine, it is converted in the second part of the plant concept to Bio-SNG, which can be fed into the natural gas grid. The concept consists of the following major sections:

- Fuel supply
- Gasification
- Gas cooling & Gas cleaning (Coarse and fine gas cleaning)
- Methanation
- Gas upgrading and utilization
- Flue gas route

The detailed process flow diagram of the Bio-SNG concept is shown in Figure 19. The plant can be divided into two parts: The first part of the plant, comprising all the plant equipment up to the product gas blower including the entire flue gas route, is a state-of-the-art technology, which has proven its reliability in various industrial realizations. The second part of the plant is still in the stage of demonstration. This is due to the facts that not many industrial Bio-SNG routes have been realized so far and some reactor units originate from other industrial applications. The glycol drying is a good example for this, which is widely used in natural gas processing, but has not been established in Bio-SNG processes. However, the chosen process design relies on the technical expertise of several authors and manufacturers and thus seems to be a promising technology.

Based on the selected process design, a simulation model was created in IPSEpro. This allows a calculation of the mass- and energy balances of the process and an evaluation of the plant performance. In the first part of this chapter the plant design and all the relevant operating data are discussed in detail, which constitute the basis of the simulation and have a big influence on the final results. In the second part of this chapter the simulation results are displayed and key figures are presented, which are derived from the obtained data. In Appendix B the extended simulation results with a detailed breakdown of the gas streams can be found. For all the presented values an indication is provided, whether the value was set by the user in the simulation (s for set), calculated numerically by IPSEpro (r for result), or calculated externally (c for calculation). The simulation flowsheet is shown in Figure 20 and basically represents the implementation of the process flow diagram from Figure 19 in IPSEpro.

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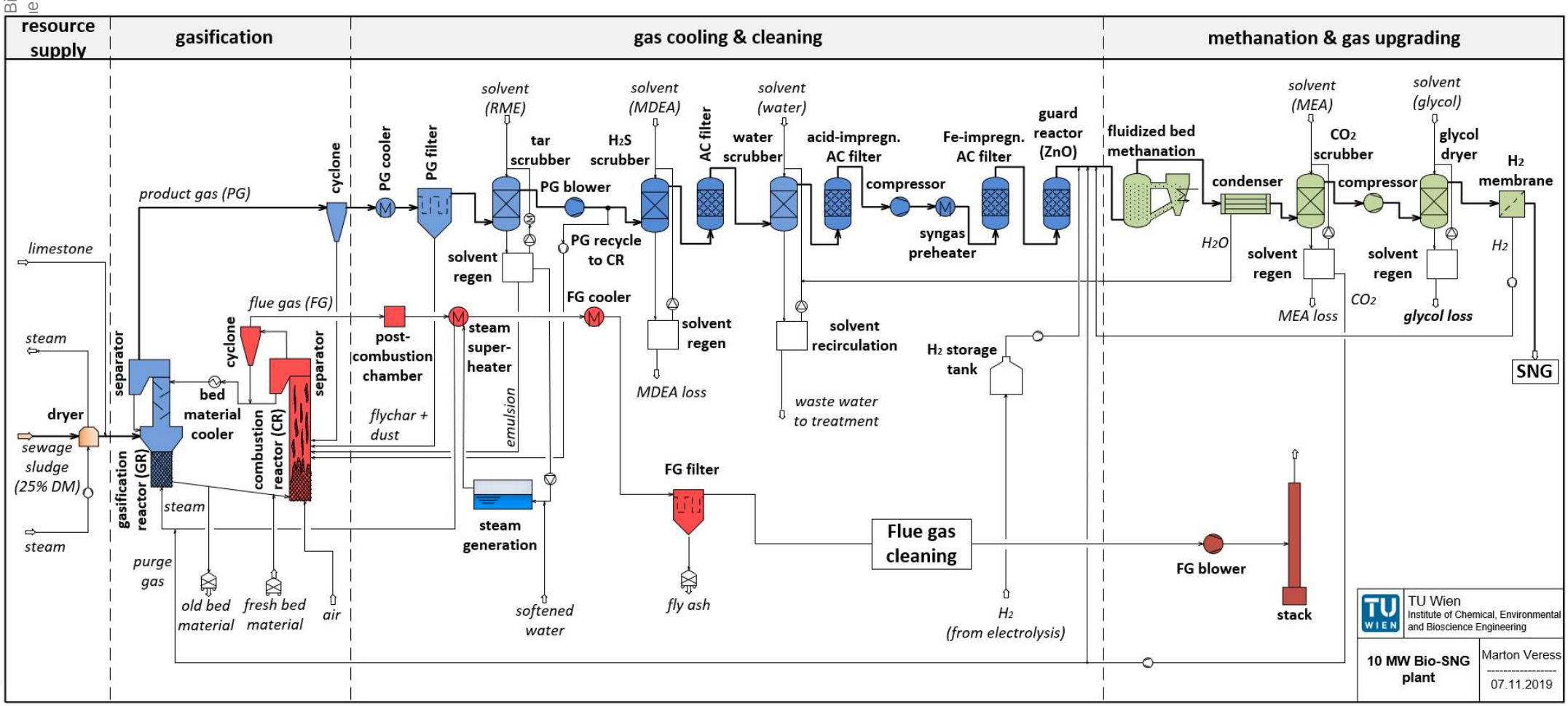

Figure 19: Process flow diagram

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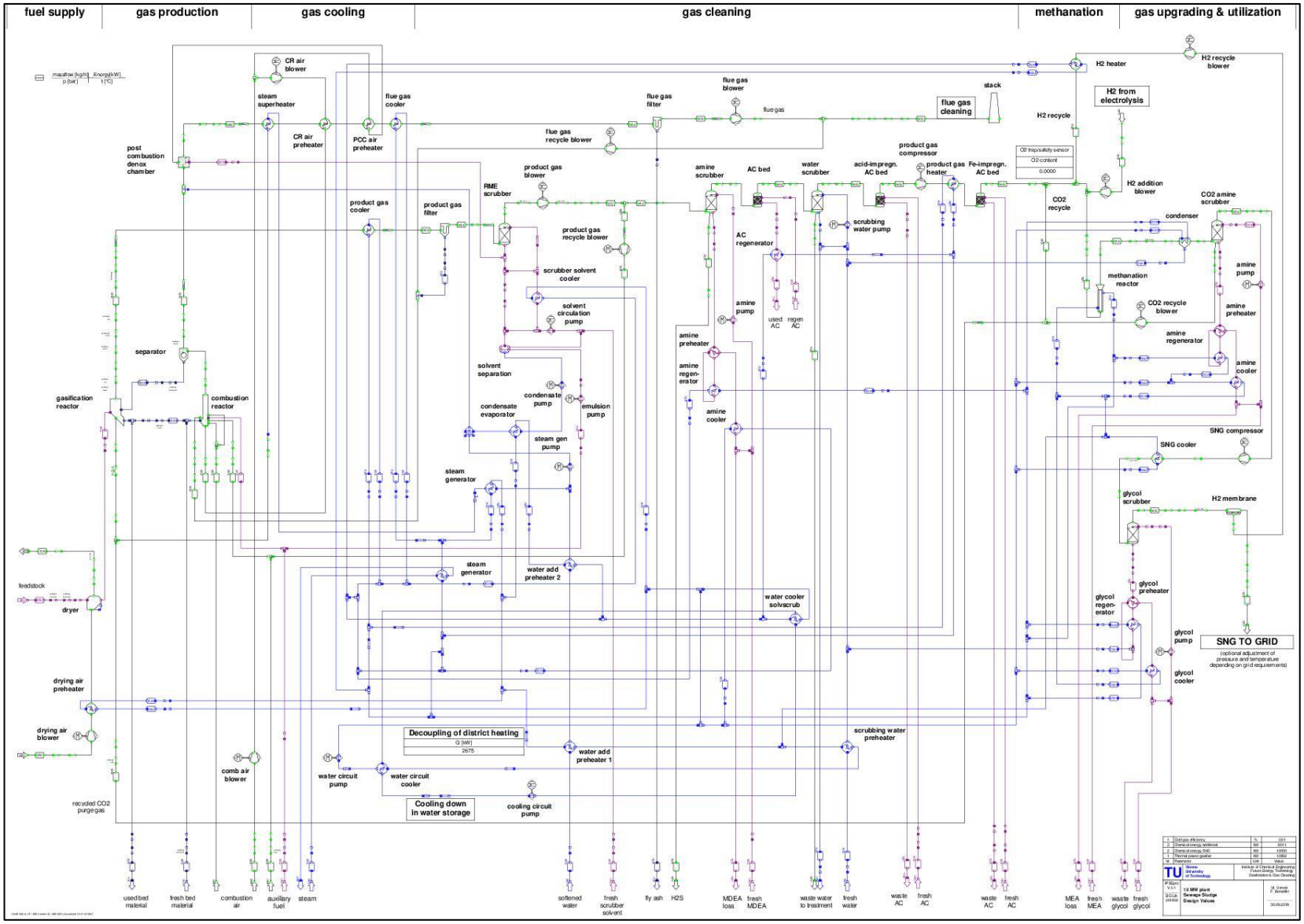

Figure 20: IPSEpro simulation flowsheet

5.1 Process design and operating data

5.1.1 Fuel supply

As discussed in Chapter 4.1, sewage sludge was chosen as representative low-grade feedstock for the process. In order to increase the efficiency of the gasification the fuel needs to be dried to a reasonable extent. When sewage sludge is withdrawn from a sewage plant, it is digested and mechanically dewatered to a water-content of 70-80 %. These treatment steps are usually performed right on the sewage plant site [98]. However, this water-content is still way too high for thermal utilization, so a further reduction of the water-content has to be achieved by a thermal dryer.

In the simulation the sewage sludge was dried from an initial 75 wt% to 11 wt% H_2O before entering the gasifier. This can be done for example by a steam-heated disc dryer which is able to decrease the water-content to values below 10 wt% [98]. In the simulation air was used for the drying process instead of steam because the dryer model in the gasification library does not accept water as heat carrier medium. However, the electricity consumption of the dryer was adapted to typical literature values of steam-heated disc dryers [98]. The necessary heat for the air preheating is provided by the water cycle of the process. Table 17 summarizes the results of the fuel analysis at TU Wien previous to the drying step [78].

Table 17: Sewage sludge analysis

Biomass inlet		
Fuel characteristics		
Net calorific value (LHV)	kJ/kg	1201
Proximate composition		
Organics	$wt\%_{db}$	58.5
Ash	$wt\%_{db}$	41.5
Water	wt%	75
Elementar analysis		
C	$wt\%_{db}$	30.0
H	$wt\%_{db}$	3.7
O	$wt\%_{db}$	19.8
N	$wt\%_{db}$	3.9
S	$wt\%_{db}$	1.0
Cl	$wt\%_{db}$	0.1

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5.1.2 Gasification

The gasification step is based on a conventional DFB steam gasification (see Chapter 2.1) using an 80-20 wt% mixture of olivine-limestone as bed material. The simulation data for the product gas composition was already presented in Table 12 and derives mainly from a sewage sludge gasification experiment carried out at the 100 kW_{th} pilot plant of TU Wien in 2012 [78]. However, since then the gasification technology has evolved and some features have been optimized. These recent scientific findings comprise for example the addition of limestone to the bed material or the application of the advanced reactor design (Chapter 2.1.2). Although in the simulation they are assumed to be valid, it has to be clear that the majority of the data derives from an experiment, where these aspects where not known and applied yet. In a more recent sewage sludge experiment at TU Wien [79] these changes were already included but the data was not applied in the simulation due to reasons mentioned in Chapter 4.1. Another change compared to the experimental test runs was the switch of the gasification purge gas from N_2 to CO_2. The advantage lies on the one hand in a decreased N_2-product gas content and on the other hand in an efficient in-process recycling of the removed CO_2 further downstream in the gas upgrading section.

The operating data and some key figures for both the gasification and the combustion reactor are displayed in Table 18. The equations for the calculation of gasification key figures are taken from reference [13] and comply with the common assessment methods of gasification processes. For the cold gas efficiency of the gasifier two different values are calculated. The only difference is that for the net cold gas efficiency the product gas recycling was taken into account when calculating the energy content of the product gas. This product gas recycle is needed to promote the reaction in the combustion reaction.

Table 18: Simulation data of the DFB gasification

Gasification reactor				Combustion reactor			
Bed material: olivine (80 wt%) + limestone (20 wt%)							
Operating temperature	°C	810	S	Operating temperature	°C	895	R
Operating pressure	bar	1	S	Operating pressure	bar	0.98	S
Thermal fuel input	MW_{th}	10.8	R	Air ratio (λ)	-	1.25	S
Steam to carbon ratio	mol_{H2O}/mol_{C}	2	R	Char combusted	$kg_{char}/kg_{fuel,waf}$	0.14	C
Steam to fuel ratio	$kg_{H2O}/kg_{fuel,waf}$	0.9	S	Heat loss	kW	54	R
Fuel related water conversion rate	kg_{H2O}/kg_{db}	0.17	R				
Product gas yield	Nm^3_{db}/kg_{waf}	1.27	C				
Cold gas efficiency	$kW_{tar\&char\ free}/kW$	0.76	C				
Net cold gas efficiency	$kW_{tar\&char\ free}/kW$	0.58	C				
Heat loss	kW	108	R				

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5.1.3 Gas cooling and gas cleaning

Gas cooling and coarse gas cleaning

The generated hot and raw product gas is cooled by a heat exchanger to 180 °C with the transferred heat being recovered in the process. The gas cleaning section can be divided into a coarse and a fine cleaning part. The coarse gas cleaning consists of the necessary treatment steps for an application of the product gas in a gas engine. This includes the removal of dust and some heavier tars in a bag filter at 180 °C to protect downstream process equipment. Table 19 is in accordance with Table 2 and shows the simulation data of the bag filter.

Table 19: Simulation data of the product gas filter

Parameter		Product gas filter	
Operating temperature	°C	180	R
Operating pressure	bar	0.96	R
Pressure drop gas	bar	0.03	S
Heat loss	kW	1	R
Separation efficiency	wt%	99.9 - ash	S
		99.9 - dust	S
		30 - tars	S

After the removal of particulates the product gas is led into the RME scrubber where it is further cooled to 40 °C by the scrubbing liquid. Thereby water vapour condenses from the gas stream and entrains a significant part of the NH_3 (30 wt% [40]) which is highly soluble in water. Furthermore, most of the tars are absorbed by the RME solvent. The used scrubbing liquid is fed into a phase separator, from where the clear water phase is recycled as gasification agent via a steam generator and superheater. The NH_3 is evaporated during the steam generation process and is injected directly in the post combustion chamber of the flue gas route in order to reduce the NO_x-emissions [42]. While the clear RME phase is reused in the scrubbing process the emulsion phase, which is a mix of RME, absorbed tars and water, is pumped to the combustion reactor and serves there as an auxiliary fuel. The amount of solvent and the tar separation efficiency in the RME scrubber was set according to the experience gained in industrial co-generation plants [99]. After those two vital cleaning steps a significant part of the product gas is also recycled to the combustion reactor to promote the combustion reaction. Hence, there is no need for additional external auxiliary fuels to operate the process.

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Table 20: Simulation data of the gas cleaning scrubbers

		RME scrubber		Amine scrubber		Water scrubber	
Solvent		Rapeseed methyl ester		Methyl diethanolamine		Water	
Operating temperature	°C	40	S	40	S	40	S
Operating pressure	bar	0.95	R	1.1	R	1	R
Pressure drop gas	bar	0.01	S	0.01	S	0.01	S
Pressure drop solvent	bar	1.2	R	1	R	1	R
Solvent to gas ratio	kg/kg	22.8	R	0.3	R	15	S
Heat loss	kW	0	S	18.86	R	0.64	R
Separation efficiency	wt%	89.7 - tar 30 - NH_3	R S	82 - H_2S 2.3 - CO_2	R R	98.5 - NH_3 99.9 - HCN 97.3 - HCl	R R R
Capacity	g/g_{solv}	-		0.0286 - H_2S 0.0443 - CO_2	S S	-	

Fine gas cleaning

The fine gas cleaning section is a more sophisticated system that should be capable of removing all the undesired components for the synthesis. Reaching this goal by a series of fixed activated carbon bed adsorbers turned out to be infeasible, since the measured gas impurity volume contents were very high, partly in the percentage range. In a discussion with an employee of the activated carbon manufacturer Donau Carbon GmbH it was pointed out that an economically and technically reasonable deployment of activated carbons makes sense below impurity levels of around 500 ppmv [100]. For this reason the contents of the main impurities needed to be reduced by scrubber units beforehand.

In the first step after the coarse gas cleaning an amine scrubber is placed to decrease the H_2S-content in particular. A 40 wt% solution [101] of methyl diethanolamine (MDEA) was chosen as scrubbing liquid, because it is the most selective towards H_2S-absorption among the commonly used amines. Nevertheless, it is inevitable that CO_2 also becomes absorbed to a certain extent. The amine consumption was calculated according to absorption capacities reported in [102]. The solvent is regenerated in a stripper column and can be reused again in the scrubber. However, minor vaporization and degradation losses need to be compensated by adding fresh amine to the process. The values for the fresh amine demand derive also from the tables presented in [102]. In the simulation model the amine regeneration was represented in a simplified form by means of a set of heat exchangers.

An activated carbon filter filled with the commercial activated carbon Desorex K47 is able to remove the remaining light tars (BTX) and further reduce the H_2S-content of the gas. The adsorption capacities of the activated carbon were taken from experiments conducted by [103] and from the expert interview with the representant of the company Donau

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Carbon GmbH [100]. The activated carbon filter is operated as a temperature swing adsorption where two beds are alternately either loaded with impurities or regenerated with steam at 180 °C [103]. The activated carbon is deliberately put at this position to avoid the contamination of water with tar in the following water scrubber [31].

The task of the water scrubber is to remove all the water-soluble gas components, such as NH_3, HCN and HCl. The wastewater needs to be discharged in a sewage plant for water treatment. Due to the lack of operating data in literature assumptions had to be made for this absorber. A solvent to gas ratio of 15 was set in the simulation, which is based on operating data of the RME scrubber. Around a fifth of the circulating water amount is assumed to be replaced each hour. Almost a complete removal of HCN and HCl is presumed, while the thorough removal of NH_3 is done in a further purification step. However, it has to be added that the water scrubber certainly needs further investigation and a collection of more accurate data. This can either be done by solubility calculations or by contacting manufacturers. A list of manufacturers can be found for instance in reference [31].

As mentioned before, the thorough removal of NH_3 is carried out in a further purification step by means of an acid-impregnated activated carbon called Desorex K43 P10. However, this activated carbon cannot be regenerated due to the chemical bonding of NH_3 with the impregnated acid and needs to be replaced after every adsorption cycle. The adsorption capacity is derived from information obtained from the manufacturer [100]. The data sheets of both yet presented activated carbons are presented in Appendix I.

After gas compression and heating to around 5 bar and 350 °C a final gas cleaning step for the removal of the sulfur components H_2S and COS is provided. This is accomplished by an iron-impregnated activated carbon which is not commercially available, but has proven its ability to remove both components simultaneously in two different studies [104,105]. Just as in the case of the acid-impregnated activated carbon a regeneration is not possible due to the chemical adsorption of the impurities. As last step of the gas cleaning section a ZnO guard filter with a layer of aluminium oxide on top is installed. This reactor serves the purpose of ensuring that the gas sulfur content is well below the allowed threshold of the methanation catalyst. The purpose of the aluminium oxide is to capture possibly remaining halogen compounds in order to protect the ZnO from a conversion to zinc chloride [32]. However, the ZnO filter is not included in the simulation flowsheet because it serves only as guard reactor, if any sulfur components unexpectedly manage to break through the gas cleaning section.

After this thorough gas cleaning section, the product gas meets the requirements of methanation catalyst (see Table 5) and should not cause any operational problems.

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Table 21: Simulation data of the activated carbon filters

		Activated carbon bed		Acid-impregnated activated carbon bed		Fe-impregnated activated carbon bed	
Adsorbent		Desorex K 47		Desorex K43 P10		Activated carbon with $FeCl_3$ impregnation	
Operating temperature	°C	40	R	39	R	350	R
Operating pressure	bar	1	R	0.98	R	5	R
Pressure drop gas	bar	0.1	S	0.1	S	0.1	S
Pressure drop adsorbent	bar	0	S	0	S	0	S
Heat loss	kW	0	S	0	S	0	S
Separation efficiency	wt%	36 - H_2S 100 - tars	R R	99 - NH_3	R	99.3 - H_2S & COS	R
Capacity	g/g_{ads}	0.1 - H_2S 0.2 - tars	S S	0.15 - NH_3	S	2.4 - H_2S & COS	S

All the simulation data of the scrubbers and activated carbon filters are shown in Table 20 and Table 21. For all the scrubbers a gas pressure drop of 0.01 bar was set in accordance with the experience from Güssing [99] and with the data from Table 2. Furthermore, the simplification was made that the regenerated lean solvent is always entirely pure with no residual absorbate left. These assumptions have been applied downstream for the scrubbers in the gas upgrading section as well. The upper value suggested by Table 2 for the pressure drop of bed filters is 0.06 bar. For all the activated carbon filters that value was rounded up to 0.1 bar to be on the safe side.

5.1.4 Methanation

Before the methanation is carried out the stoichiometric ratios of H_2/CO and H_2/CO_2 (see Table 7) need to be adjusted to ensure a high carbon conversion. Therefore, renewable H_2 from an external electrolysis unit is buffered in a storage and added to the product gas stream. The methanation reaction is carried out over an aluminium-supported nickel catalyst [62] in a single-stage fluidized bed methanation reactor. At the chosen reaction conditions of 5 bar and 350 °C [106] the reaction is assumed to reach thermodynamic equilibrium [5,15]. In IPSEpro the equilibrium reactions (Equation 3.1-3.3) are calculated through a chemical equation unit, which in turn retrieves the calculation data from the property library SimProp. For C_2H_4, C_2H_6 and C_3H_8 a complete conversion was assumed in the reactor which is in accordance with equilibrium calculations and also reference [59], where a full conversion of ethylene (C_2H_4) was observed. In terms of temperature an increase of 4 °C fits well with the nearly isothermal character of this reactor type. The 150 mbar, which were set as pressure drop for the methanation reactor, derive from reference [107]. All the relevant simulation data for the methanation reactor can be seen in Table 22.

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Table 22: Simulation data of the methanation reactor

Methanation reactor			
Type		Fluidized bed	
Catalyst		Al_2O_3-supported nickel	
Operating temperature	°C	350	S
Operating pressure	bar	5	R
Pressure drop gas	bar	0.15	S
Temperature increase	°C	4	R
Heat removal	kW	1918	R
Heat loss	kW	0	R
Conversion	% C_2H_4	100	S
	% C_2H_6	100	S
	% C_3H_8	100	S
	% CO	99.9	R
	% CO_2	87	R
	% H_2	95	R

5.1.5 Gas upgrading

In order to obtain a gas that fulfills the feed-in criteria of the natural gas grid (see Table 11) some gas upgrading steps have to be performed. First, the raw SNG is led into a condenser where the gas is cooled down to 40 °C and condensing water is removed. An amine scrubber subsequently separates the residual CO_2, which is not converted in the methanation step. The set-up of the amine scrubber including the regeneration system is identical to the amine scrubber in the gas cleaning section. However, instead of MDEA it is operated with a 20 wt% aqueous solution of monoethanolamine (MEA) as a result of its high CO_2-absorption capacity [102]. The CO_2 released within the regeneration step is partly recycled back to the gasifier as purge gas, while the rest is added to the product gas stream right before the methanation reactor. The data of the scrubbers in the gas upgrading section are summarized in Table 23.

Table 23: Simulation data of the gas upgrading scrubbers

		CO_2 amine scrubber		Glycol scrubber	
Solvent		Monoethanolamine		Triethylene glycol	
Operating temperature	°C	40	S	40	S
Operating pressure	bar	4.84	R	39.99	R
Pressure drop gas	bar	0.01	S	0.01	S
Pressure drop solvent	bar	1.15	S	1	S
Solvent to gas ratio	kg/kg	8.4	R	0.7	R
Heat loss	kW	0.18	R	10.47	R
Separation efficiency	wt%	99.9 - CO_2	S	99.6 - H_2O	R
Capacity		0.08 g/g_{solv}	S	0.025 m^3/kg_{solv}	S

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After the MEA scrubber the pressure of the gas stream is raised to 40 bar through a multi-stage compression. The compressors are followed by a cooler decreasing the gas temperature again to 40 °C. In order to meet the low water-content required for natural gas substitution a glycol scrubber removes most of the water from the raw SNG. The separation efficiency of 99.6 in the absorber is determined by the saturation temperature that is specified in Table 11. Triethylene glycol (TEG) is used as solvent for the dewatering and is regenerated thermally at 180 °C [108]. The simulation of the regeneration was thereby simplified in the same way as it was done for the amine scrubbers. A detailed flowsheet of the regeneration concept together with the absorption capacity of TEG can be found in [109]. The last step of the gas upgrading deals with the separation of residual H_2 from the gas stream by a membrane. According to reference [110] polyimide membranes are well suited for separating H_2 and CH_4. The membrane simulation model was simplified in a way that the module is only permeable to different degrees for H_2, CH_4 and N_2, while other components remain entirely in the retentate. The permeability values for each of these gas species and the H_2-separation efficiency are taken from [110]. The H_2-rich permeate stream is recycled back to support the adjustment of the stoichiometric ratio before the methanation reactor. For the calculation of the membrane area the thickness of the membrane must be specified, which was assumed to be 1000 Å analogously to Gassner et al. [107]. The simulation data of the membrane are displayed in Table 24.

Table 24: Simulation data of the H_2 membrane

H_2 membrane			
Membrane material		Polyimide	
Membrane thickness	m	$1 \cdot 10^{-7}$	S
Membrane area	m^2	42	R
Operating temperature	°C	40	R
Operating pressure	bar	40	R
Pressure drop permeate	bar	34.99	R
Pressure drop retentate	bar	0.29	R
Heat loss	kW	0	R
Separation efficiency	wt% H_2	90	C
Permeability H_2	$m^2/(s \cdot Pa)$	$2.1075 \cdot 10^{-16}$	S
Permeability CH_4	$m^2/(s \cdot Pa)$	$1.875 \cdot 10^{-18}$	S
Permeability N_2	$m^2/(s \cdot Pa)$	$2.4 \cdot 10^{-18}$	S

In the medium term a raise of the tolerated H_2-level in natural gas can be expected. A H_2-share of up to 10 % depending on the application of the natural gas is already common in Germany. Discussions regarding a further raise of the H_2-tolerance are currently led and corresponding investigations are performed. According to the German Association of Gas and Water (DVGW), increasing hydrogen contents require also adjustments in the grid and some equipment [111–113]. However, also in Austria a loosening of the H_2-limit values can be

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expected in the future. In such a case the H_2-membrane can be neglected, which improves the SNG process in terms of economics and simplicity.

The design of the gas upgrading section is all in all similar to the gas upgrading route proposed by Heyne et al. [114]. The choice of reactors and the operating conditions of 40 °C and 40 bar are all based on this reference. After this process route the SNG is ready to be fed into the natural gas grid. However, depending on where the SNG should be injected, the gas stream has to be adapted to the necessary pressure level of the local grid.

5.1.6 Flue gas route

The other gas route of the process is of the flue gas, which is released from the combustion reactor. A cyclone separates the hot bed material from the gas stream in order to transport it back to the gasifier. The flue gas is led to a post combustion chamber where the residual CO is burnt at 850 °C. This temperature is the minimum temperature to incinerate waste materials according to the regulation [115]. Furthermore, the NO_x-content is reduced by injecting NH_3 coming from the RME scrubber of the product gas route. Values for the NO_x-reduction potential can be found in reference [116]. Afterwards the gas passes through a cooling section, where the heat is utilized to produce steam and preheat the combustion air. The entrained fly ash is then separated in a bag filter and disposed.

Table 25: Simulation data for the mechanical separators of the flue gas route

		Cyclone DFB		Flue gas filter	
Operating temperature	°C	895	S	150	R
Operating pressure	bar	0.95	R	0.81	R
Pressure drop gas	bar	0.03	S	0.025	S
Heat loss	kW	0	S	0	S
Separation efficiency	%	97.3	R	100	R

The data for the mechanical separators are shown in Table 25. This flue gas route is sufficient for so-called easy fuels, such as woody biomass, but is not adequate for the gas cleaning challenges arising in connection with sewage sludge gasification. However, the extension of the flue gas route was not part of this work. Thus, the development and simulation of a more complex flue gas cleaning system is neglected but is nevertheless roughly considered in the cost calculation.

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5.1.7 Basic Simulation Data

The basic simulation data comprises all the settings and data which are not only valid for a specific section but for the whole process. In Table 26 the ambient conditions assumed for the plant are displayed. For auxiliary equipment, such as pumps or heat exchangers, default values were applied throughout the simulation flowsheet, which are listed in Table 27 to Table 29.

Table 26: Ambient conditions [13]

Ambient conditions		
Altitude above sea level (for Vienna)	m	150
Temperature (mean annual temperature)	°C	10
Pressure	bar	1
Relative air humidity	%	80

Table 27: Efficiencies of used blower and compressor units [13]

Efficiency factors		Product gas blower	Flue gas blower	Air blower	Product gas compressor	SNG compressor
Isentropic efficiency	%	50	65	65	75	75
Mechanical efficiency	%	98	98	98	98	98
Drive system						
Electrical efficiency	%	90	90	90	90	90
Mechanical efficiency	%	100	100	100	100	100

Table 28: Efficiencies of used pumps [13]

Efficiency factors		Pumps used in main process chain	Pumps used for heat utiziliation
Isentropic efficiency	%	50	70
Mechanical efficiency	%	98	98
Drive system			
Electrical efficiency	%	80	90
Mechanical efficiency	%	100	100

Table 29: Simulation data for used heat exchangers

Parameter		Liquid-liquid	Gas-liquid
Type	-	counter-current	counter-current
Pressure drop	bar	0.25	0
Minimum temperature difference	°C	5	10

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5.2 Simulation results

By means of the simulation model mass- and energy balances were calculated for the described sewage sludge concept based on the values presented in Chapter 4.1 and Chapter 5. The most important simulation results are summarized in Table 30 showing the plant input and output streams. According to the results, 10 MW of SNG can be produced from an input of 4.4 MW wet sewage sludge and 6 MW of hydrogen. Additionally, also around 2 MW of superheated steam needs to be provided for the process, which was assumed to be available on-site. The electricity consumption of the process amounts to 0.9 MW. In addition to the SNG also 2.7 MW of district heating can be decoupled from the process. The achieved SNG purity meets the requirements of the natural gas grid and can be directly injected.

Table 30: Simulation results

Plant input			Plant output		
Term	**Unit**	**Value**	**Term**	**Unit**	**Value**
Biomass (sewage sludge)	kg/h	13,149	SNG	Nm^3/h	1035
Biomass (chemical energy - before drying)	kW	4,387	SNG (chemical energy)	kW	10,000
Added hydrogen	Nm^3/h	2,016	District heating	kW	2,675
Added hydrogen (chemical energy)	kW	6,041			
Electricity consumption	kW	906			
Combustion air	Nm^3/h	6,242	Exhaust gas	Nm^3/h	8,395
Fresh bed material (olivine - limestone)	kg/h	25	Ash and dust from exhaust gas filter	kg/h	1,389
Auxiliary fuel	kg/h	0			
Fresh water	kg/h	1,935	Removed water in dryer	kg/h	9,456
Fresh scrubber solvent (RME)	kg/h	22	Wastewater	kg/h	1,552
Fresh amine (MDEA)	kg/h	$3 \cdot 10^{-5}$	MDEA loss	kg/h	$3 \cdot 10^{-5}$
Fresh amine (MEA)	kg/h	$4 \cdot 10^{-4}$	MEA loss	kg/h	$4 \cdot 10^{-4}$
Fresh glycol	kg/h	$5 \cdot 10^{-3}$	Waste glycol	kg/h	$5 \cdot 10^{-3}$
Acid impregnated activated carbon	kg/h	4.3	Waste acid impregnated activated carbon	kg/h	5.0
Fe impregnated activated carbon	kg/h	0.8	Waste Fe-impregnated activated carbon	kg/h	2.7
Steam (420°C)	kg/h	2,676	Steam (143.5°C)	kg/h	2,676
Steam heat input (420°C)	kW	2,433	Steam heat output (143.5°C)	kW	371
			Removed H_2S & tar in AC bed	kg/h	4.0
			Removed H_2S in amine scrubber	Nm^3/h	42

The chemical energy flow within the process boundaries is represented in Figure 21 by means of a Sankey diagram. For the sake of clarity only streams with a chemical energy content of ≥ 0.3 MW were considered for this schema. It becomes evident that through the intense gas cleaning also a huge amount of chemical energy is removed by separating the impurities.

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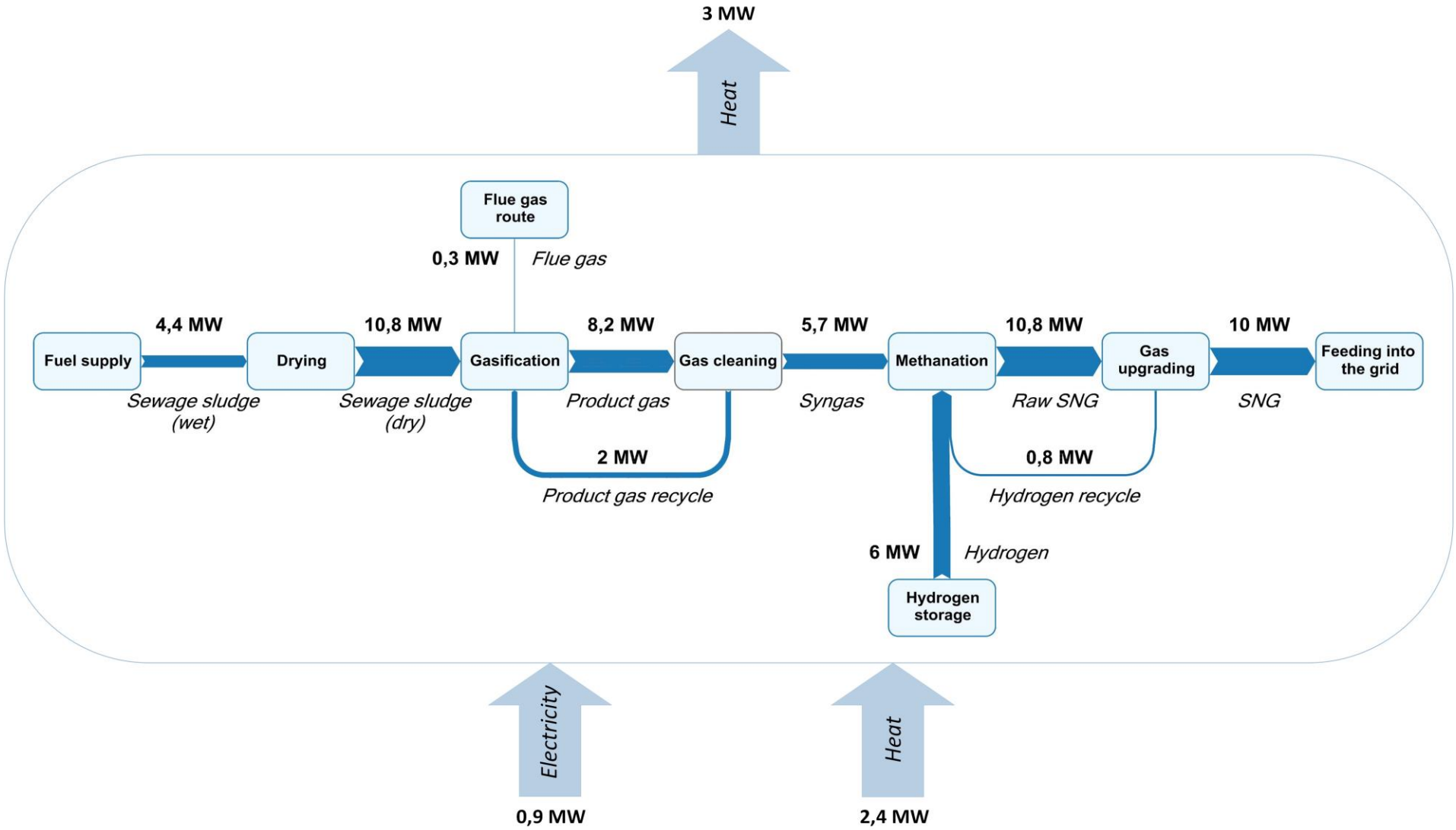

Figure 21: Representation of the essential chemical energy flows (≥0.3 MW) and the overall energy balance in a sankey diagram

In order to be able to assess the entire process with regard to its efficiency, a meaningful definition of key figures is necessary. Therefore, the so-called cold gas efficiency is determined. This key figure relies on the comparison of the chemical energy input and output within defined system boundaries. Analogously to the cold gas efficiency used for the evaluation of the gasifier in Chapter 5.1.2 different cold gas efficiencies can be defined for the entire process according to Table 31.

Table 31: Calculation of the cold gas efficiency

$$\eta_{net,BM\rightarrow PG} = \frac{\dot{m}_{PG} \cdot LHV_{PG} - \dot{m}_{PG,rec} \cdot LHV_{PG,rec}}{\dot{m}_{BM,dried} \cdot LHV_{BM,dried}} \quad \text{Eq. 5.1}$$

$$\eta_{net,PG\rightarrow SNG} = \frac{\dot{m}_{SNG} \cdot LHV_{SNG}}{\dot{m}_{PG} \cdot LHV_{PG} - \dot{m}_{PG,rec} \cdot LHV_{PG,rec} + \dot{m}_{H2,add} \cdot LHV_{H2,add}} \quad \text{Eq. 5.2}$$

$$\eta_{global,BM\rightarrow SNG} = \frac{\dot{m}_{SNG} \cdot LHV_{SNG}}{\dot{m}_{BM,dried} \cdot LHV_{BM,dried} + \dot{m}_{H2,add} \cdot LHV_{H2,add}} \quad \text{Eq. 5.3}$$

$$\eta_{global,BM\rightarrow SNG} = \frac{\dot{m}_{SNG} \cdot LHV_{SNG}}{\dot{m}_{BM,wet} \cdot LHV_{BM,wet} + \dot{m}_{H2,add} \cdot LHV_{H2,add}} \quad \text{Eq. 5.4}$$

η	cold gas efficiency
ṁ	mass flow
LHV	lower heating value
BM	biomass
rec	recycled

Table 32 shows the values of the calculated efficiencies. The higher cold gas efficiency value obtained with Eq. 5.3 than with Eq. 5.1 results from the addition of H_2 to the product gas before the methanation step. It becomes also apparent that the water-content of the biomass plays a major role for the value of the cold gas efficiency. Because of this, a comparison with other processes is only valid if the water-content of the feedstock is clearly stated. For the GoBiGas project a global cold gas efficiency of 61.8 % was reported for dry wood chips [117]. If a water-content of zero is assumed for the sewage sludge, the global cold gas efficiency amounts to 58.4 %. For Güssing a global cold gas efficiency of 61 % can be found in literature [15] using wood chips with a water-content between 25-40 wt%. If the gasifier of the sewage sludge concept was fed with sewage sludge containing 25 wt% water, the global cold gas efficiency would increase to 61 % as well. For a higher water-content the global cold gas efficiency would obviously exceed the values reported from Güssing.

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Table 32: Plant efficiencies

Plant efficiency factors (LHV based)		
Net cold gas efficiency (Dried Biomass to PG)	*Eq. 5.1*	57.6 %
Net cold gas efficiency (PG to SNG)	*Eq. 5.2*	81.6 %
Global cold gas efficiency (Dried biomass to SNG)	*Eq. 5.3*	59.4 %
Global cold gas efficiency (Wet biomass to SNG)	*Eq. 5.4*	95.9 %

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6 Economic evaluation

In this chapter an economic evaluation based on the mass- and energy balances from the previous chapter is performed. First of all, the plant costs are calculated with the methods presented in Chapter 4.3.1. For the analysis of the economic plant performance a techno-economic case study with different scenarios is set up based on Chapter 4.3.2. With the tools presented in Chapter 4.3.3 the results are shown also from a different angle, which together allow the assessment of the economic potential of the presented sewage sludge concept. The procedure in this chapter is represented in Figure 22.

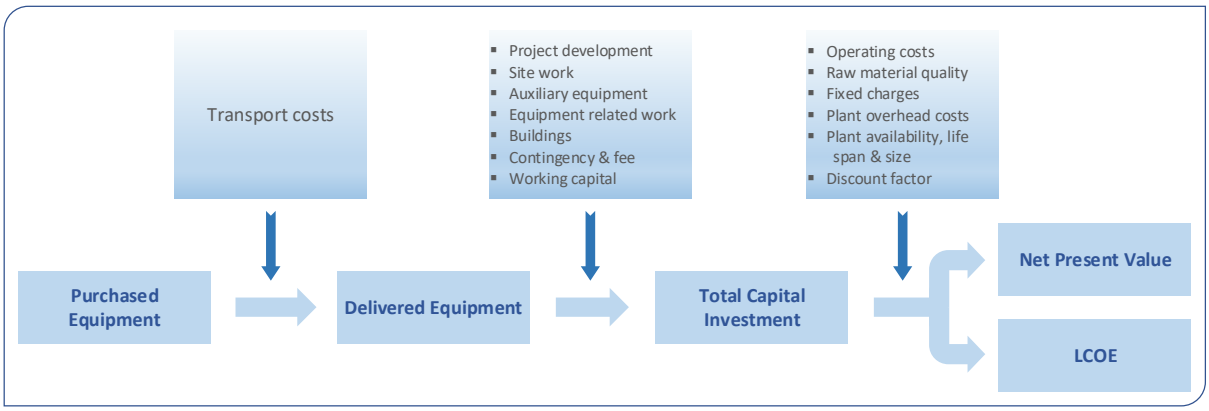

Figure 22: Procedure for the economic plant evaluation (adapted from [118])

6.1 Sewage sludge plant cost calculation

The plant cost calculation was carried out by the application of the methods described in Chapter 4.3.1. For the first part of the plant (fuel supply, gasification, coarse gas cleaning, flue gas route) an order of magnitude estimate method was applied, while for the second part of the plant (fine gas cleaning, methanation, gas upgrading) the Lang study estimate method was applied. This is due to the fact, that the cost of the first part of the presented plant could be estimated quite accurately according to literature data deriving from four different, already realized DFB co-generation plants in Austria and Germany. The investment costs for each of the plants in the thermal fuel power range of 8-15 MW_{th} are mentioned in Müller [13]. Schmid [95] built the mean average of these four projects to estimate a specific capital cost of 1.7 $M€/MW_{th,}$ which was used in this work as well. However, the costly gas engine that is included in each of the costs of these four plants was not deducted from the final result for the sewage sludge concept. This is done deliberately to economically compensate for the need of an advanced flue gas cleaning system, which was not considered in this work from a technical point of view.

For the second part of the plant, which was added for the actual SNG synthesis, the cost was surveyed by means of the Lang study estimate method. Therefore, the determination of the

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total sum of delivered equipment cost was necessary, which served as the basis for the Lang method. The results are presented in Table 33. As can be seen, most of the equipment items were calculated based on the sixth-tenth rule. For those equipment items, where no useful data could be found, the cost was calculated according to size-specific cost rates found in literature (membrane, hydrogen storage tank) or according to a method presented in reference [119] (activated carbon beds). For the latter method a quick equipment sizing needed to be performed, which is shown in Appendix C. In Appendix D also the cost for each auxiliary equipment is listed in detail, as they are only presented in a summarized form in Table 33. If the base equipment cost was only available without a delivery charge (free on board - f.o.b.), the purchased equipment cost was increased by 5 % of its own value to account for the delivery of the equipment to the plant site [86,87]. The cost update of the equipment items to the year 2018 was performed with the corresponding CEPCI factors found in [88–90] (see Appendix E).

Table 33: Calculation of the total delivered equipment cost

Equipment	Base scale	Base cost	Scale factor	Scale in the present study	Equipment cost (€, 2018)	Source
Main equipment						
MDEA scrubber	14,040 Nm³/h	5,190,000 €, 2010	0.6	1,955 Nm³/h	1,827,389	[114]
Activated carbon*					303,088	[119]
Water scrubber	43,560 m³/h	3,000,000 €, 2002	0.7	2,137 m³/h	581,394	[120]
Acid impregnated AC*					145,247	[119]
Fe impregnated AC*					146,337	[119]
ZnO guard reactor	81,360 kg/h	49,000 €, 2009	0.56	2,842 kg/h	11,892	[121]
Methanation reactor	43,560 Nm³/h	2,860,000 €, 2010	0.6	4,055 Nm³/h	836,467	[114]
Condenser	448 MW	4,700,000 $, 2007	0.7	1.06 MW	74,504	[122]
MEA scrubber	14,040 Nm³/h	5,190,000 €, 2010	0.6	1,286 Nm³/h	1,421,736	[114]
Glycol scrubber	41,667 m³/h	316,386 $, 2006	0.6	1,223 Nm³/h	43,707	[123]
Membrane	1 m²	3,531 $, 2006	1	42 m²	169,003	[114]
Hydrogen storage	1 kg	470 €, 2017	1	1,450 kg	760,660	[124]
Auxiliary equipment						
Blowers / Compressors**					405,811	[88]
Pumps**					23,244	[88]
Heat exchangers**					283,368	[88]
Total cost of delivered equipment					**7,033,848**	

*according to Appendix C | **according to Appendix D

With the calculation of the total cost of delivered equipment the Lang factor can be applied to determine the capital cost of the plant. As mentioned in Chapter 4.3.1 different Lang factors are suggested in the literature, depending also on the basis of the calculation (purchased or delivered equipment costs) and the process circumstances. For this work the adapted Lang factor from [83] for solid-fluid processing plant additions to an existing site was used. Its value

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for the just described circumstances amounts to 4.1. A detailed breakdown of the factor components can be found there as well.

Subsequently, an estimation of the working capital was carried out. The usual proceeding consists of assuming a percentage of the fixed capital as working capital. However, within this work its value has been reduced strongly because in the considered business case (Chapter 6.2) there are no revenues and therefore no delays of payment expected. Thus, the working capital is basically only made up of a raw material inventory for 1 month of production [83,92] resulting from the mass balances from the simulation and the initial raw material fill of the reactors. The only exception to this is the renewable hydrogen, which originates from the external electrolysis and is buffered in a hydrogen storage. With respect to a reasonable storage size a hydrogen inventory necessary for 8 h production was assumed. For the methanation reactor an initial catalyst fill of 400 kg was estimated based on the data reported for the fluidized bed methanation reactor within the Comflux project [50]. In order to determine the working capital, these raw material amounts are multiplied with cost rates found in the literature. These cost rates are listed in detail in Appendix G.

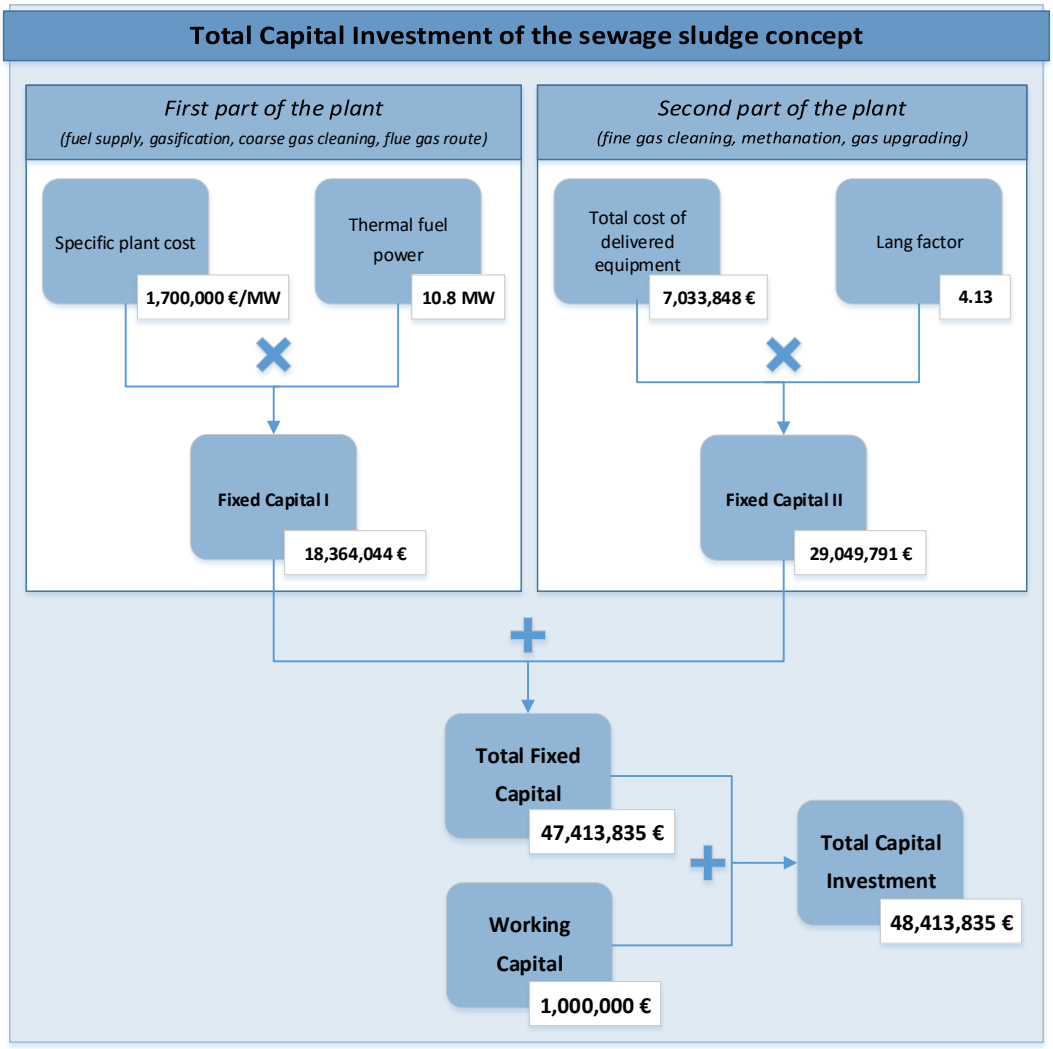

Figure 23: Calculation procedure for the total capital investment

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In the end the sum the working capital (0.9 M€) was rounded up to 1 M€ to have a little buffer. The entire procedure for the calculation of the total capital investment for the sewage sludge concept is illustrated in Figure 23. It is evident that the total fixed capital cost is made up of the two fixed capitals calculated for the two parts of the plant. This sum amounts to 47.4 M€, which needs to be invested to erect the presented sewage sludge plant. The total capital investment takes also the working capital into account and is therefore slightly higher (48.4 M€).

6.2 Techno-economic case study

The evaluation of the plant competitiveness in terms of economic performance is carried out with the aid of a techno-economic case study. Hereby different scenarios are compared to achieve one and the same goal. For this purpose, it makes sense to fall back on the relative net present value (Chapter 4.3.2), where each alternative scenario is compared to a reference case. The set-up of the business case is described in detail in Table 34.

Table 34: Set-up of the business case

Business Case
Case description
A fictive Austrian cement producer with a natural gas demand of 10 MW is striving to phase out the fossil natural gas energy consumption. With its new strategy the company seeks to cover its own energy needs through the production of Bio-SNG instead of buying natural gas **(Scenario 0)**. The person responsible for this transformation identified the following options as a potential solution for this challenging task:
Alternative scenarios
➢ **Scenario 1** Production of SNG with a down-scaled GoBiGas plant ➢ **Scenario 2** Production of SNG with the presented sewage sludge concept ➢ **Scenario 3** Production of SNG with the presented sewage sludge concept and an investment subsidy (IS) of 84 % ➢ **Scenario 4** Production of SNG from sewage sludge using an SER-gasification to circumvent additional H_2-feeding

Scenario 1 was included in the business case to present the data of an already realized industrial Bio-SNG plant. This can be seen as an “additional reference case” for the sewage sludge concept to show its economic performance in direct comparison with a similar technology. As the GoBiGas plant was designed for an SNG output of 20 MW_{SNG} the plant

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needed to be scaled down to a plant size of 10 MW_{SNG}. This was performed according to the GoBiGas cost publication and scaling suggestions of Thunman et al. [125] and is shown in Appendix H. It has to be considered though that the goal of the GoBiGas plant in this stage was not to operate as a commercial plant but as a demonstration plant. Thus, the overall cost reductions potential through plant optimizations were included in the calculations, as it was done for the optimized reference plant in [125]. Nevertheless, a poor economic performance could be expected from this scenario.

Scenario 2 and **Scenario 3** are basically identical with the only difference of having secured an investment subsidy of 84 % in the latter case. This amount of investment subsidy marks the lowest limit to obtain a positive relative NPV in the business case. While the calculation of the total investment cost was presented in the previous chapter, the operating costs were calculated based on the mass- and energy balances determined by the simulation and cost rates for each raw material listed in the Appendix G.

For the calculation of the operating utilities the replacement of the methanation catalyst was considered as well. The assumption was made that the catalyst needs to be replaced after every 1500 h of operation. This value is based roughly on the operational experience of the Güssing plant, where up to 1000 h stability of a fluidized bed catalyst could be demonstrated [15].

In Table 36 the economic comparison of the scenarios is shown. The discount rate was set according to the data provided by Damodaran [126], who is an internationally renowned valuation expert. For the year 2018 he suggested a discount rate of almost 6 % for green energy technology projects in Europe. Falling back on Equation 4.4 this corresponds with a cumulative discount factor of 11.47. The economic assumptions made for the relative net present value method are summed up in Table 35.

Table 35: Economic assumptions for the relative net present value calculation

Parameter	Unit	Value	Relevance for scenario	Source
Location of plant	-	Austria	All	Set
Reference year	-	2018	All	Set
Plant lifetime	a	20	All	[127]
Number of employees	1/a	3	2, 3, 4	[128]
Operating hours plant	h/a	7500	All	[95]
Operating hours district heating	h/a	5800	2, 3, 4	[95]
CO_2-emissions of natural gas	t_{CO2}/MWh	0.20	0	[13]
Currency conversion 1 $	€	1.104	2, 3, 4	[129]
Currency conversion 1 SEK	€	10.93	1	[130]
Location index Austria-Sweden	-	0.96	1	[131]
Discount rate	%	6	All	[126]

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Table 36: Business Case - Relative net present value calculation

Techno-economic Case Study						
Parameter	**Unit**	**Natural gas**	**GoBiGas**	**10 MW_{SNG} sewage sludge**	**10 MW_{SNG} sewage sludge with IS**	**10 MW_{SNG} sewage sludge with SER**
		Scenario 0	**Scenario 1**	**Scenario 2**	**Scenario 3**	**Scenario 4**
Circumstances						
Thermal fuel power	MW_{th}	-	16	10.8	10.8	17
SNG output	MW	-	10	10	10	10
Operating hours plant	h/a	-	7,500	7,500	7,500	7,500
Natural gas demand	MW	10	-	-	-	-
Natural gas demand	MWh/a	75,000	-	-	-	-
Forest residues demand	MWh/a	-	120,000	-	-	-
Sewage sludge demand	MWh/a	-	-	81,018	81,018	127,500
Electricity consumption	MWh/a	-	11,250	6,794	6,794	6,794
Heat production	MWh/a	-	25,000	15,514	15,514	15,514
Investment cost (I_0) (incl. interests)	€	0	74,877,794	48,413,835	7,746,214	58,189,131
Investment subsidy (84 % I_0)	€	-	-	-	40,667,621	-
Yearly expenses						
Fuel costs (natural gas)	€/a	2,700,000	-	-	-	-
Fuel costs (wood chips)	€/a	-	2,700,000	-	-	-
Maintenance, insurance, admin., tax	€/a	-	956,388	1,694,484	1,694,484	2,036,620
Labor costs	€/a	-	2,224,938	187,500	187,500	187,500
Operating utilities	€/a	-	362,964	5,190,603	5,190,603	434,635
Electricity consumption	€/a	-	0	706,598	706,598	706,598
CO_2-emission certificates	€/a	373,500	-	-	-	-
Wastewater disposal costs	€/a	-	-	29,092	29,092	29,092
Ash disposal costs	€/a	-	-	937,749	937,749	1,475,762
Yearly earnings						
Fuel earnings (sewage sludge)	€/a	-	-	5,917,264	5,917,264	9,312,161
District heating	€/a	-	750,000	465,429	465,429	465,429
SNG earnings	€/a	-	-	-	-	-
Costs						
Sum (Earnings-Expenses)	€/a	-3,073,500	-5,494,290	-2,363,333	-2,363,333	4,907,383
Net present value calculation						
Cumulative present value factor	-	11.47	11.47	11.47	11.47	11.47
Additional investment cost	€	0	74,877,794	48,413,835	7,746,214	58,189,131
Operating cost savings	€/a	0	-2,420,790	710,167	710,167	7,980,883
Net present value	**€**	**0**	**-102,644,261**	**-40,268,221**	**399,401**	**33,351,601**

It can be noticed in Table 36 that for the chosen parameters and cost rates the presented process chain is not able to compete with the reference case without the aid of an investment subsidy of minimum 84 %. With a closer look at the operating utilities it becomes apparent that the main cost driver of the process is the addition of the costly hydrogen. This is due to the fact, that renewable hydrogen from electrolysis is still very expensive and that only a limited price cut is expected for the future [132]. Originally the hydrogen addition was added to maximize the carbon conversion of the process and develop an optimal process from the

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technical point of view.

In the end, the business case was extended by **Scenario 4** which uses a SER-gasification. In this way the stoichiometric ratio required for methanation is set already in the gasifier by enabling the water-gas shift reaction through the transportation of CO_2 to the combustion reactor. This allows a circumvention of the additional H_2-feeding upstream of the methanation reactor. Based on the lessons learned so far, a significantly better economic performance can be expected from this scenario. The additional H_2-feeding can also be circumvented if the product gas from a conventional gasification is methanated non-stoichiometrically. Hereby, a large quantity of non-converted CO_2 needs to be removed downstream in the MEA scrubber. Scenario 4 represents both cases, as similar results can be expected. For the sake of completeness, it has to be said that for Scenario 4 the input data for the cost calculation was not obtained by a simulation model but through rough estimations on the basis of empirical values [133]. In Table 36 the values which were changed due to the modification of the plant operation are underlined. The other values were assumed to remain constant. It can be seen that for this process configuration huge revenues can be expected in comparison with the reference scenario. This is not only related to the waiver of the costly H_2-addition. With the SER-gasification also a higher amount of fuel is needed to produce the same amount of SNG like in Scenario 2 and 3. Because currently sewage sludge has a negative market price, the feeding of more fuel has paradoxically a positive effect on the plant economics. These two effects together not only compensate for the increased expenses due to the higher investment, maintenance and ash disposal costs but even manage to reach a remarkable positive relative net present value of 33 M€.

In any case, Scenario 4 constitutes an inferior option from the technical point of view as the carbon utilization is significantly lower due to the removal of CO_2 in the gasification reactor. Also, the SER-gasification has mostly been tested with woody biomass fuels so far. Only few pilot-plant experiments have been reported using fuels that cannot be classified as high-grade fuels [2]. In effect, the stable operation with very challenging fuels, like sewage sludge, still needs to be proven in many test runs on different scales. Nevertheless, in terms of economics it appears to be superior because the costly H_2-addition can be neglected.

It has to be added, that according to the principle of the economy of scale a significant cost reduction is also possible through a scale-up process. A Bio-SNG plant with 10 MW_{SNG} output is industrial/demonstration size but a commercial plant would be expected to have around 100 MW_{th}, as it was initially planned in the GoBiGas project. Through this scale-up the specific investment costs are expected to decrease making the process economically more competitive [63,125]. A reduction of investment and operating costs can also be expected through the learning curve by the construction of multiple commercial plants. Improved

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operations, optimized processes, higher plant utilization factors and higher biomass-to-SNG efficiencies are expected to be the driving factors of the cost reductions [134].

Based on the relative net present value calculation of the sewage sludge concept (Scenario 2) from Table 36 a sensitivity analysis was carried out. With this method an input parameter of a model is systematically varied to investigate its effect on a model output parameter. Thereby, parameters that have a major influence on the economic plant performance can be identified and the impact of the deviation of the parameter value can be quantified.

The following parameters were independently varied in the sensitivity analysis:

- Fuel price
- Hydrogen price
- Discount rate
- Feed-in tariff for district heating
- Natural gas price
- CO_2-emission certificates

The effect of the parameters on the relative net present value of the sewage sludge concept is illustrated in Figure 24. In brackets the initial value of each parameter is presented. It is evident that the plant investment cost, the fuel price and the hydrogen price have the biggest impact on plant economics. For a fuel price of around -95 €/t for sewage sludge with 25 wt% dry matter content the process can be economically viable without any subsidies. A hydrogen price of approximately 0.9 €/kg would mark the break-even point of the process in comparison with the reference case. However, a hydrogen price of 0.9 €/kg is unrealistically low even for fossil-derived hydrogen. Renewable H_2-prices are only expected to decrease moderately in the future, so this parameter variation can only render the relative net present value positive in combination with other favorable circumstances. Besides, it can be seen that the natural gas price also has a significant impact. Although it does not have any absolute effect on the economics of the sewage sludge process itself, it makes the reference case more expensive and through that improves the relative economic performance of Scenario 2. The same applies for the CO_2-emission certificates which, however, do not have a major impact on the value of the relative net present value. Similarly, the variation of the discount rate and the feed-in tariff for district heating both display very little influence.

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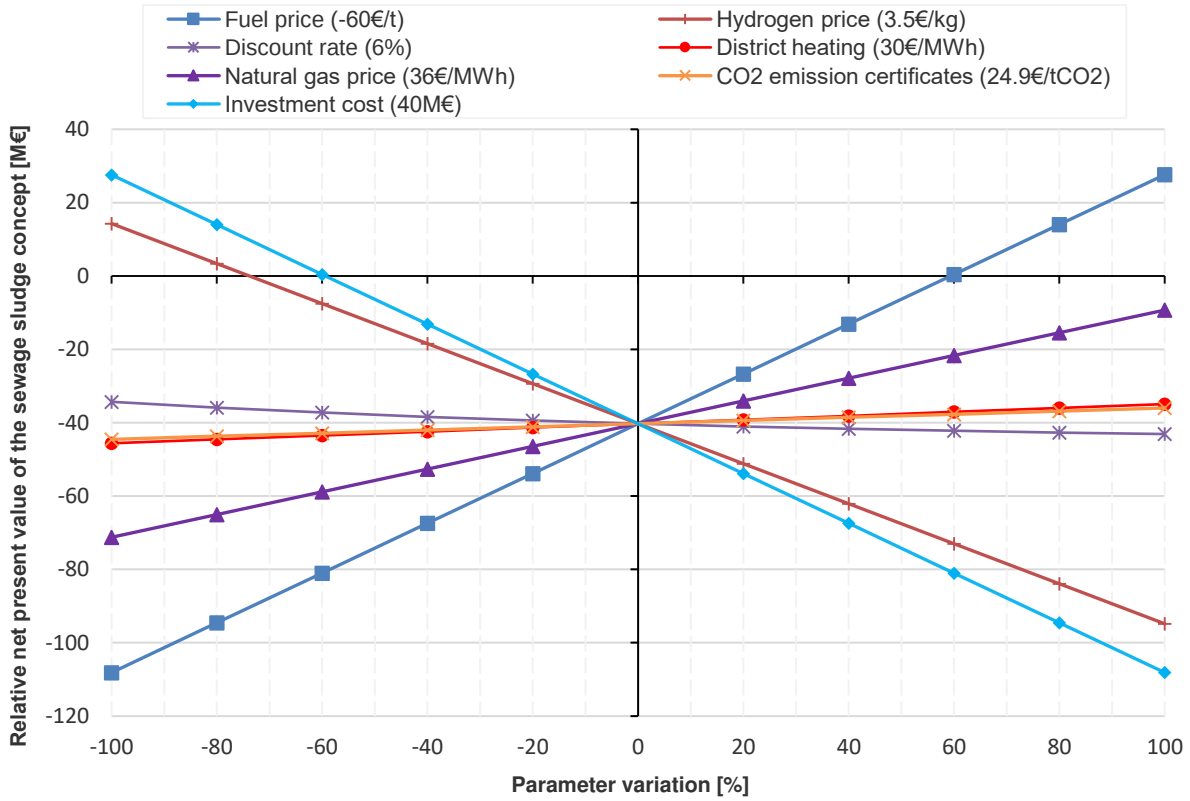

Figure 24: Sensitivity analysis for the sewage sludge concept (Scenario 2)

6.3 Cost calculation with the levelized cost method

In Figure 25 the economic performance of the different scenarios from Table 34 is represented with the LCOE method. By means of this illustration a huge gap between the market price of fossil natural gas and the production cost of Bio-SNG can be identified.

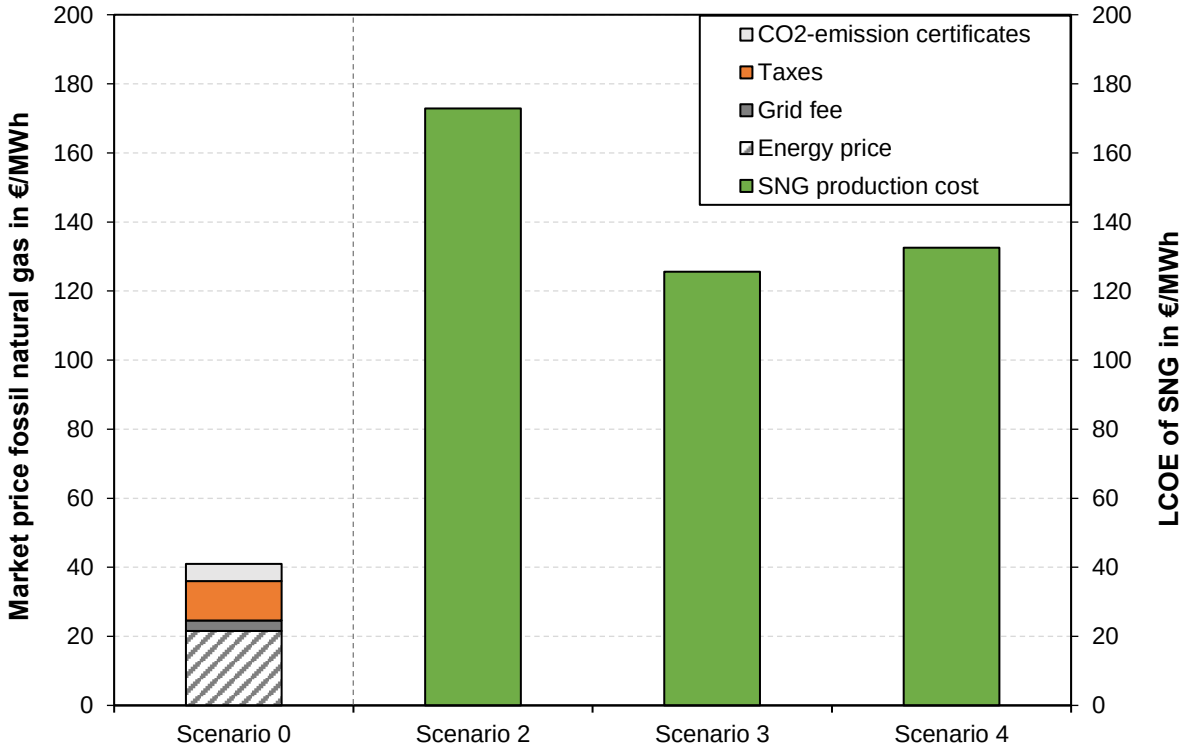

Figure 25: Comparison of the Levelized Cost of Energy with the fossil natural gas market price

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However, through this method additional revenues in the alternative scenarios deriving from the decoupling of district heating and the processing of sewage sludge are not considered. Thus, the LCOP calculation method is more adequate for a fair comparison of all the scenarios. By deducting the revenues generated by other income sources than SNG from the overall production costs, a very illustrative comparison can be drawn. The results of the LCOP calculation are shown in Figure 26. It is affirmed that Scenario 2 struggles in terms of economics and requires more than double the costs of the fossil reference case for producing the same amount of gas. This huge cost discrepancy can be tackled by two remedies: The first one was presented in the previous chapter with Scenario 3, where the technology is made economically viable through an investment subsidy. With an investment subsidy of 84 % the levelized SNG production costs approximately settle down to the same value as determined in Scenario 0 (40.5 €/MWh). The second solution would be a price increase of CO_2-emission certificates, which is expected to happen anyway in the upcoming years due to global climate goals. However, if the necessary price increase of CO_2-emission certificates is considered for Scenario 0 the total price of emission certificates corresponds to 259 €/t_{CO2}. This is more than the tenfold of the actual price level and is therefore assessed as unrealistically high. Thus, this measure would not be enough on its own but could be conceivable in a combination with an investment subsidy. Finally, it becomes evident that Scenario 4 holds a great potential with levelized SNG production costs of 2.2 €/MWh. As discussed before this is mainly attributable to the increased amount of fuel with a negative price and the no longer required addition of the costly H_2.

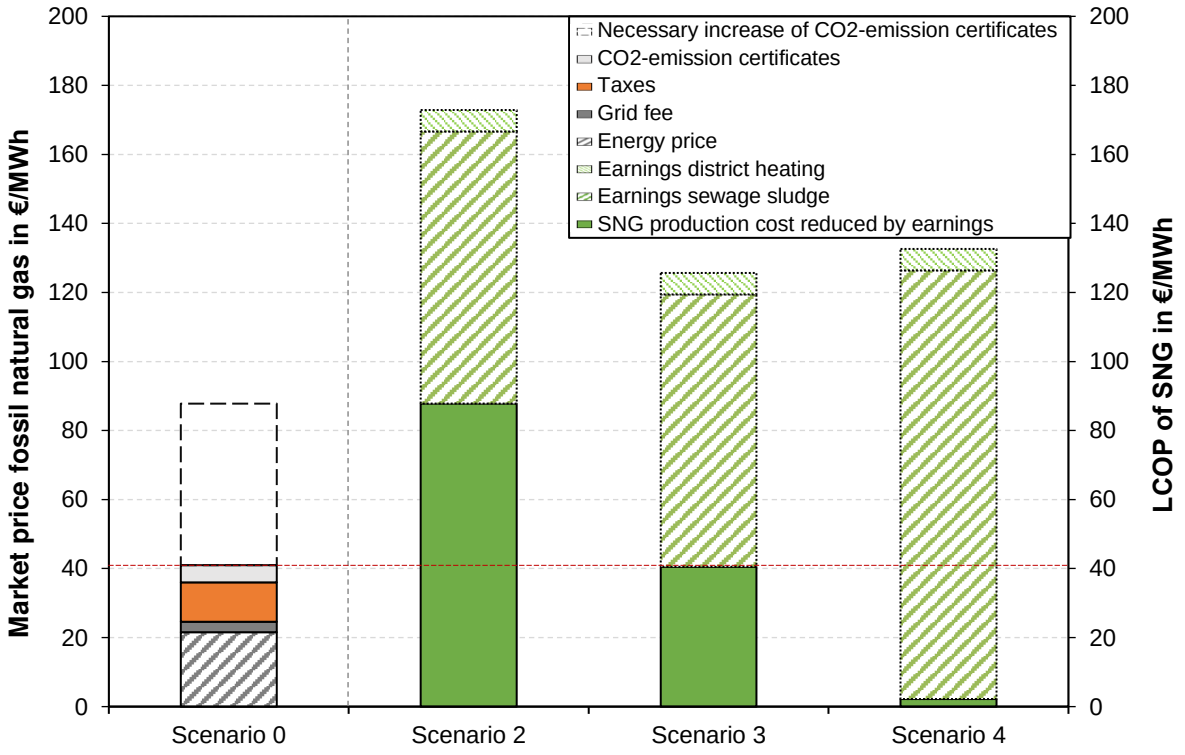

Figure 26: Comparison of the Levelized Cost of Product with the fossil natural gas market price

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7 Conclusion and outlook

The background and motivation for the global climate efforts are the necessity to reduce fossil carbon emissions by accomplishing a gradual transition to a low-fossil carbon energy system. Within the scope of the present work the development, simulation and techno-economic evaluation of a process concept for the production of 10 MW synthetic natural gas from sewage sludge is presented. For this purpose, a detailed process flow sheet was elaborated in the first place. This served as the basis for the simulation model which was subsequently implemented in the simulation program IPSEpro. By the simulation mass- and energy balances were obtained which allow a further techno-economic evaluation of the entire process.

This approach proved suitable to answer the following research questions:

- What is the best possible process route for the industrial production of Bio-SNG from low-grade fuels based on the dual fluidized bed steam gasification?
- What are the mass and energy balances for this process concept?
- To what extent is the proposed process chain technologically mature?
- What are the investment costs and how competitive is the proposed process chain?

The simulation results show that the production of 10 MW SNG requires around 4.4 MW of dried sewage sludge (11 wt% moisture content) and 6 MW additional H_2 from an external electrolysis are needed. From that a global cold gas efficiency of 59.4 % can be calculated, which proves to be very efficient with regard to the utilization of a low-grade fuel.

The total capital investment for the plant was obtained by two different methods. The cost of the first part of the plant was estimated based on several already realized plants with a similar set-up. For the second part of the plant (fine gas cleaning, methanation, gas upgrading) a study cost estimate method was chosen due to the lack of industrial scale Bio-SNG plants. A total capital investment of around 48 M€ was calculated for the sewage sludge process, which seems realistic compared to other realized DFB gasification-based plants.

The cost analysis within a business case showed that the presented concept is struggling to compete economically with fossil natural gas due to the high purchase prices of renewable H_2. However, economic competitiveness can be reached either by securing an investment subsidy or by waiving the costly H_2-addition. The latter scenario can be realized, for instance, through the SER-gasification process, where the stoichiometric ratio can be adjusted through the removal of CO_2 in the gasifier. Alternatively, the product gas can be fed in to the methanation reactor without any H_2-addition. However, after this non-stoichiometric methanation a separation of a huge amount of non-converted CO_2 is necessary. From a technical point of view this scenario is of course less appealing as the removed CO_2 is not converted in the

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methanation unit and thereby lowers the carbon utilization of the plant significantly. Besides, most of the SER-investigations until now have been carried out in pilot plant scale. The scale-up to demonstration scale is still pending and is inevitable to show the principle working in the long-term and in MW-scale. Nevertheless, a rough estimation of the SER-gasification showed very promising results in terms of economics and should be subject to further investigations. With the levelized cost method the specific production costs are calculated with 2.2 €/MWh for this scenario. Similar specific production costs can be assumed for a non-stoichiometric methanation with subsequent CO_2-separation. In comparison the sewage sludge process concept with H_2-addition reaches 87.8 €/MWh, while the market price of fossil natural gas including CO_2-emission certificates lies around 40 €/MWh. However, substantial cost reductions for Bio-SNG processes are expected in the next decades, taking advantage of the economy of scale. Besides, the realization of multiple plants is expected to result in a learning curve for an optimized process control. On the other side a price increase of fossil natural gas can be assumed in the next decades. All these future trends will contribute to making Bio-SNG processes economically more attractive.

Although the present work is based on a lot of experience and expertise in the field of biomass gasification, it nevertheless has to be stated that the results are obtained through a theoretical work. The gasification of difficult low-grade fuels inevitably leads to (long-term) operational challenges which are not taken into consideration by the simulation model. Difficulties experienced in connection with the gasification of sewage sludge comprise for example:

- Development of an effective ash handling system to remove the high amount of ash
- Development of a flue gas cleaning system to reduce NO_x- and other emissions
- Unfavorable influence of certain ash components on the product gas composition

Thus, it is necessary to perform further gasification test runs with low-grade fuels in different scale variations to generate a reliable knowledge base. The same holds true for the gas cleaning, methanation and gas upgrading. These sections still need to mature and lack commercialization until now. The goal needs to be the demonstration of the entire process chain, as it was successfully implemented in the Güssing and GoBiGas projects. Concomitant lab-scale research is required as well in order to investigate the mechanisms and details of the process units.

The focus of research should lie on the investigation of the following essential factors:

- Improvement and demonstration of gas cleaning methods
- Improvement of the methanation catalyst robustness
- Improvement of the overall plant integration and efficiency

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Summing up, the obtained simulation data for the conversion of sewage sludge to SNG provides valuable data for the design of the proposed concept and a gradual scale-up. The technology has a great potential from the ecologic, the economic and technical point of view. Hence, a demonstration at a reasonable scale is highly recommended.

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Notation

Symbols

Δ	Difference	-
η	Cold gas efficiency	-
Å	Angstrom	10^{-10}m
C_1	Base equipment cost	€
C_2	Design equipment cost	€
E	Annual expenditures	€/a
i	Discount rate	%
I_0	Capital expenditure	€
I_1	Base equipment cost index	-
I_2	Design equipment cost index	-
$\dot{m}$	Mass flow	kg/h
n	Plant lifetime	a
p	Pressure	bar
R	Annual revenues	€/a
S_1	Base equipment size	-
S_2	Design equipment size	-
T	Temperature	°C

Abbreviations

AC	Activated carbon
Al_2O_3	Alumina
Ar	Argon
ads	Adsorbent
AP	Annual energy production
BG_Lib	Biomass gasification library
BM	Biomass
BTX	Benzene, toluene and the three xylene isomers
c	Calculation
C	Carbon
$CaCl_2$	Calcium chloride
$CaCO_3$	Calcium carbonate
CaO	Calcium oxide
CDF	Cumulative discount factor
CEPCI	Chemical Engineering Plant Cost Index
CF	Cash flow
CH_4	Methane
conc.	Concentration
CO	Carbon monoxide
CO_2	Carbon dioxide

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Co	Cobalt
$CoMoO_4$	Cobalt molybdate
CPP	Coupled pressure pulse
CR	Combustion reactor
CTU	Conzepte Technik Umwelt AG
C_xH_y	Hydrocarbon, general
C_2H_2	Acetylene
C_2H_4	Ethylene
C_2H_5SH	Ethanethiol
C_2H_6	Ethan
C_3H_8	Propane
C_4H_4S	Tiophene
C_4H_{10}	Butane
db	Dry basis
DLL	Dynamic link library
DFB	Dual fluidized bed
DM	Dry matter
DME	Dimethyl ether
DVGW	German Association of Gas and Water
ECN	Energy Research Center of the Netherlands
emp	Excluding main product
Fe	Iron
Fe_3O_4	Iron (III) oxide
FG	Flue gas
FICFB	Fast internally circulating fluidized bed
FT	Fischer-Tropsch
GCMS	Gas chromatography coupled with mass spectrometry
GDP	Gross domestic product
GHSV	Gas hourly space velocity
GoBiGas	Gothenburg Biomass Gasification demonstration project
GR	Gasification reactor
grav	Gravimetric
h	Hours
HCl	Hydrogen chloride
HCN	Hydrogen cyanide
HDS	Hydrodesulfurization
HGF	Hot gas filtration
HHV	Higher heating value (kJ/kg)
H_2	Hydrogen
H_2O	Water
H_2S	Hydrogen sulphide
IDE	Integrated development environment
IPSEpro	Process simulation software manufactured by Simtech

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IS	Investment subsidy
Ir	Iridium
LiCl	Lithium chloride
LCOE	Levelized Cost of Energy
LCOP	Levelized Cost of Product
LHV	Lower heating value (kJ/kg)
LPM	Liquid-Phase Methanation
M	Million
MAS	Mixed alcohol synthesis
MDEA	Methyl diethanolamine
MDK	Model Development Kit
MEA	Monoethanolamine
MnO	Manganese (II) oxide
M&S	Marshall and Swift Equipment Cost Index
$NaHCO_3$	Trona
Na_2CO_3	Sodium carbonate
Ni	Nickel
$NiMoO_4$	Nickel molybdate
NiO	Nickel oxide
NO_x	Nitrogen oxides
Nm^3	Gas cubic meter according to standard conditions (0 °C, 1.013 bar_{abs})
NPV	Net present value
NPW	Net present worth
n.a.	Not available
N_2	Nitrogen
OLGA	Oil-based gas scrubber by ECN
ÖVGW	Austrian Association for Gas and Water
O_2	Oxygen
Pd	Palladium
PDU	Process Development Unit
PG	Product gas
PM	Particulate matter
ppmv	Parts per million by volume
PSA	Pressure swing adsorption
PSE	Process simulation environment
PSI	Paul Scherrer Institute
Pt	Platinum
r	Result
rec	Recycled
rel	Relative
Rh	Rhodium
RME	Rapeseed methyl ester
Ru	Ruthenium

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s	Set
SASOL	South African Coal, Oil and Gas
SER	Sorption enhanced reforming
SiO_2	Silica
SNG	Synthetic natural gas
solv	Solvent
SO_2	Sulfur dioxide
TEG	Triethylene glycol
temp.	Temperature
th	Thermal fuel power
TiO_2	titania
TREMP	Topsøe Recycle Energy-efficient Methanation Process
TSA	Temperature swing adsorption
var	Variant
vol%	Percentage by volume
waf	Dry and ash free
wt%	Percentage by weight
ZnO	Zinc oxide

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List of tables

Table 1: Important chemical reactions for the SER-process 1

Table 2: Characteristics of particle removal technologies 3

Table 3: Exemplary reactions for hydrodesulfurization..... 7

Table 4: Comparison of cleaning pathways 9

Table 5: Upper limits of product gas contaminants for different process 10

Table 6: Conversion conditions and product gas composition for different biofuels 12

Table 7: Important reactions for methanation 13

Table 8: Catalysts ordered by activity and selectivity for methanation 15

Table 9: CO-methanation technologies available on the market 18

Table 10: Comparison of Different Methanation Reactor Concepts 22

Table 11: Requirements for feeding into the natural gas grid in Austria 23

Table 12: Product gas compositions from different gasification experiments 27

Table 13: Classification and quality of estimates 29

Table 14: Calculation formula for the net present value method 31

Table 15: Calculation formula for the relative net present value method 32

Table 16: Calculation formula for the levelized cost method 34

Table 17: Sewage sludge analysis 38

Table 18: Simulation data of the DFB gasification 39

Table 19: Simulation data of the product gas filter 40

Table 20: Simulation data of the gas cleaning scrubbers.....41

Table 21: Simulation data of the activated carbon filters.....43

Table 22: Simulation data of the methanation reactor.....44

Table 23: Simulation data of the gas upgrading scrubbers 44

Table 24: Simulation data of the H_2 membrane 45

Table 25: Simulation data for the mechanical separators of the flue gas route 46

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Table 26: Ambient conditions 47

Table 27: Efficiencies of used blower and compressor units....47

Table 28: Efficiencies of used pumps 47

Table 29: Simulation data for used heat exchangers 47

Table 30: Simulation results 48

Table 31: Calculation of the cold gas efficiency 50

Table 32: Plant efficiencies....51

Table 33: Calculation of the total delivered equipment cost 53

Table 34: Set-up of the business case....55

Table 35: Economic assumptions for the relative net present value calculation 56

Table 36: Business Case - Relative net present value calculation 57

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List of figures

Figure 1: Process chain of synthetic natural gas (SNG) production from biomass 1

Figure 2: Process during gasification of a single particle 1

Figure 3: Basic principle of the DFB steam gasification process 2

Figure 4: Basic principle of the SER-process with limestone as bed material 1

Figure 5: H_2/CO-ratio for gasification with different bed materials 2

Figure 6: Advanced reactor design at TU Wien 1

Figure 7: Process scheme of the OLGA tar removal technology 4

Figure 8: Operating range of metal oxides for desulfurization 6

Figure 9: Exemplary cold, warm and hot gas cleaning process chains for fuel cell applications or other synthesis processes, such as methanation 8

Figure 10: Star diagram showing various high-pressure catalytic processes for converting product gas to fuels and chemicals 11

Figure 11: Pressure and temperature influence on the equilibrium composition of (a) CO-methanation (b) CO_2-methanation and water-gas shift reaction for stoichiometric mixtures of H_2 and CO / CO_2 (left: 300 °C, right: 1 bar) 14

Figure 12: Overview of different methanation concepts; State of development: c – commercial, d – demonstration scale, r – research 16

Figure 13: Left: Temperature profiles in series of adiabatic fixed bed reactors with (full lines) and without (dashed lines) product gas recycle [15] Right: Scheme of the TREMP-process 17

Figure 14: Block flow diagram (left) and picture (right) of the 1 MW_{SNG} process development unit (PDU) in Güssing, Austria 19

Figure 15: Comflux (Thyssengas) process flow diagram (left) [50,63] and schematic of a three-phase methanation reactor (right) 20

Figure 16: Pelletizing of the sewage sludge for the gasification experiment (2019) 26

Figure 17: Structure of the simulation software IPSEpro 28

Figure 18: Comparison of two investment scenarios with a zero variant using the relative net present value in dependency of the discount rate 33

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The approved original version of this thesis is available in print at TU Wien Bibliothek.

List of figures

Figure 19: Process flow diagram 36

Figure 20: IPSEpro simulation flowsheet 37

Figure 21: Representation of the essential chemical energy flows (≥0.3 MW) and the overall energy balance in a sankey diagram 49

Figure 22: Procedure for the economic plant evaluation 52

Figure 23: Calculation procedure for the total capital investment 54

Figure 24: Sensitivity analysis for the sewage sludge concept (Scenario 2) 60

Figure 25: Comparison of the Levelized Cost of Energy with the fossil natural gas market price 60

Figure 26: Comparison of the Levelized Cost of Product with the fossil natural gas market price 61

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Appendix

- **A.** IPSEpro simulation models
- **B.** Extended stream tables
- **C.** Adsorber size estimation
- **D.** Extended equipment cost calculation
- **E.** CEPCI factors
- **F.** Calculation of the working capital
- **G.** Cost rates
- **H.** GoBiGas Plant scale-down
- **I.** Activated carbon data sheets
- **J.** Publication

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A. IPSEpro simulation models

In the course of the master thesis several equipment items needed to be introduced to the Model Development Kit of IPSEpro in order to obtain meaningful simulation results. The detailed codes of the following simulation unit models can be found in the BG_Lib of TU Wien:

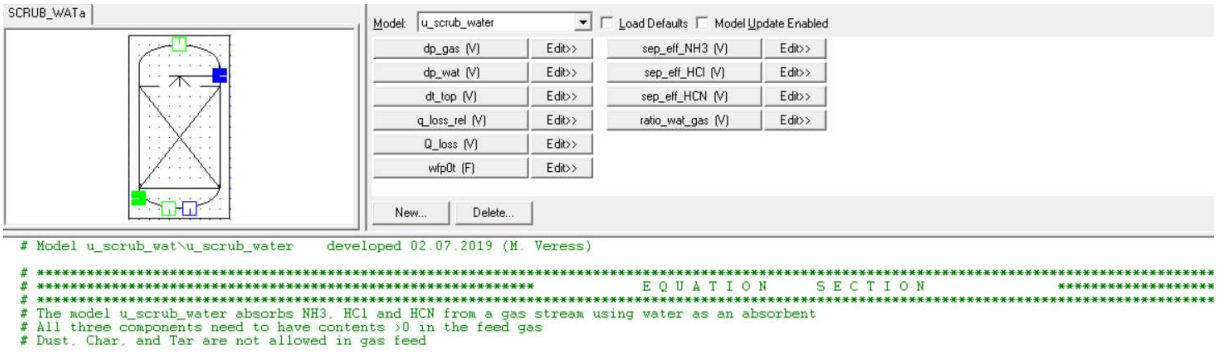

Figure A1: Simulation model of a water scrubber

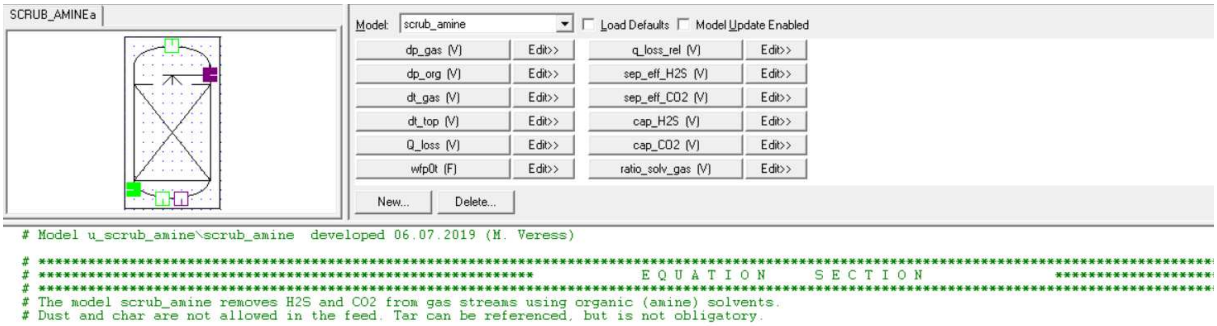

Figure A2: Simulation model of an amine scrubber

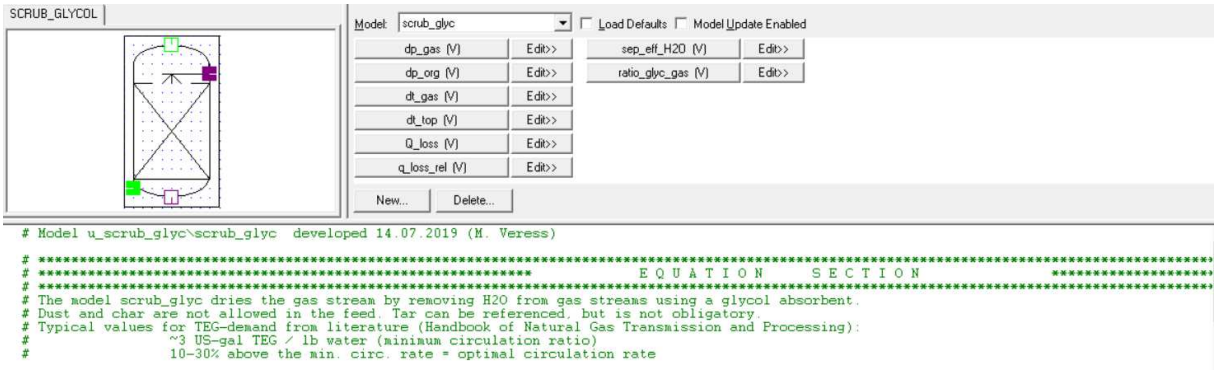

Figure A3: Simulation model of a glycol scrubber

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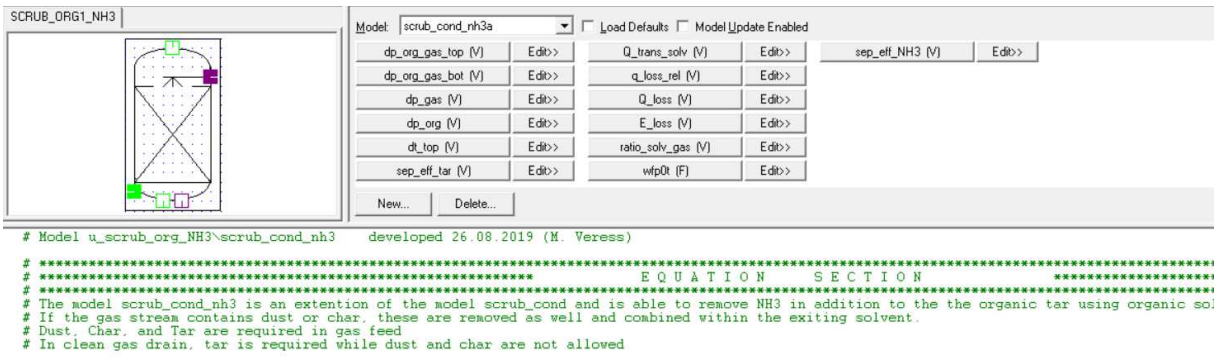

Figure A4: Simulation model of an RME scrubber with NH_3-removal

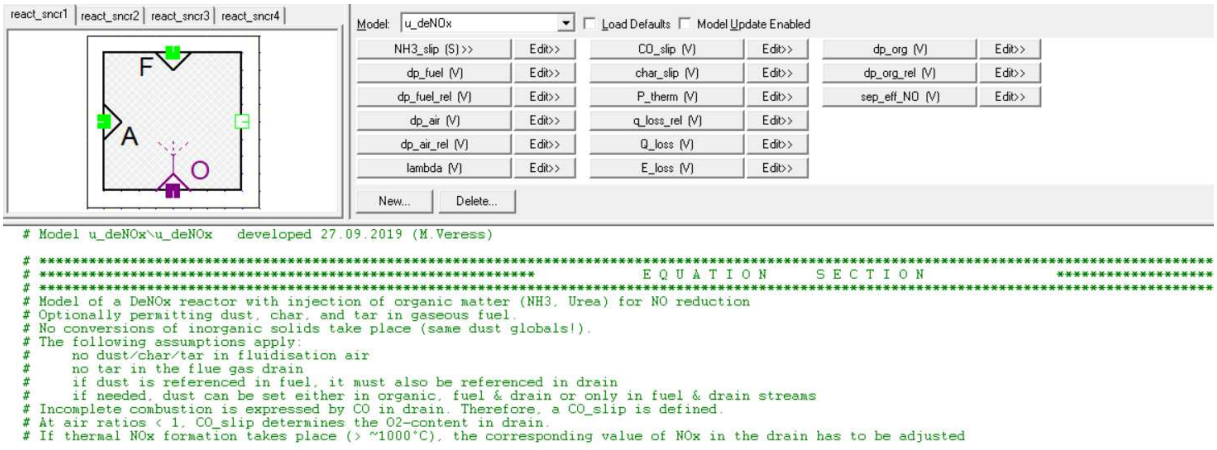

Figure A5: Simulation model of a combustion chamber with NO_x-removal

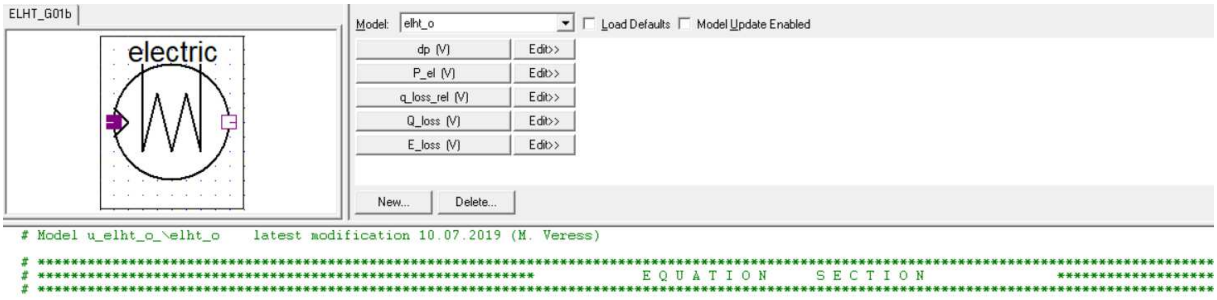

Figure A6: Simulation model of an electric heater for organic streams

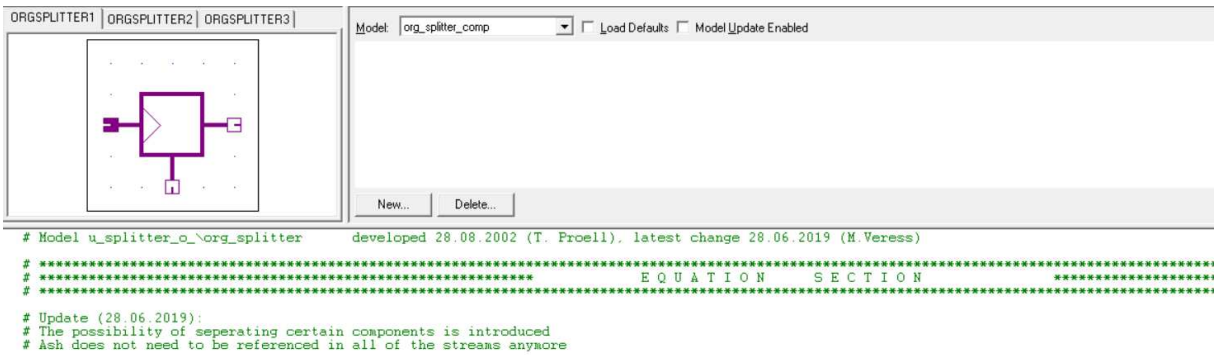

Figure A7: Simulation model of a component splitter for organic streams

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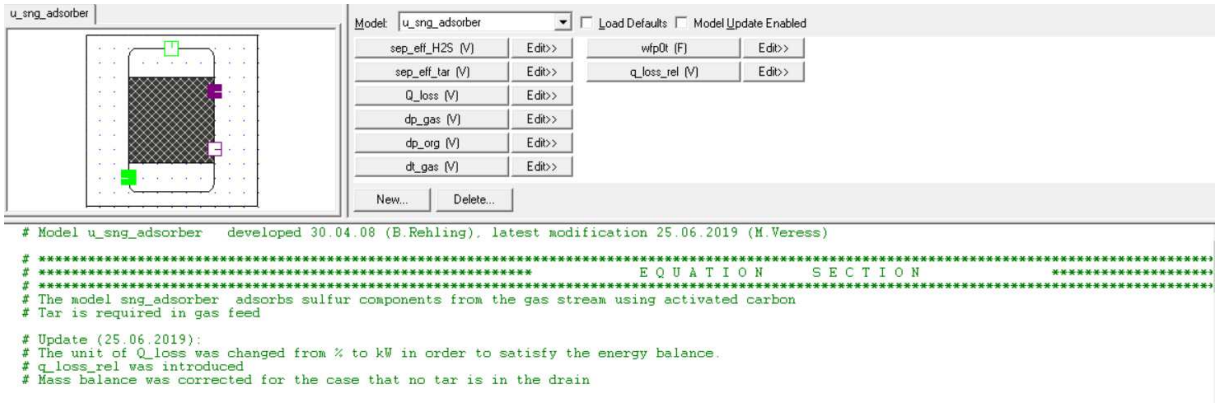

Figure A8: Simulation model of an activated carbon adsorber

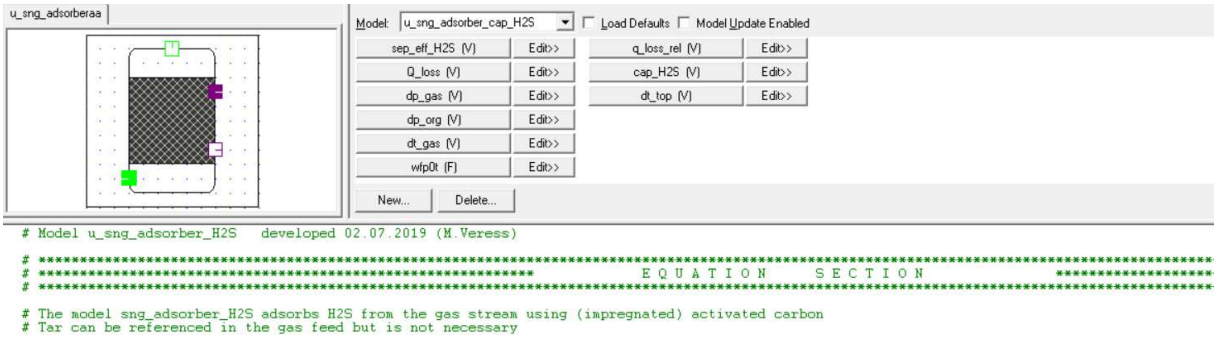

Figure A9: Simulation model of an Fe-impregnated activated carbon adsorber

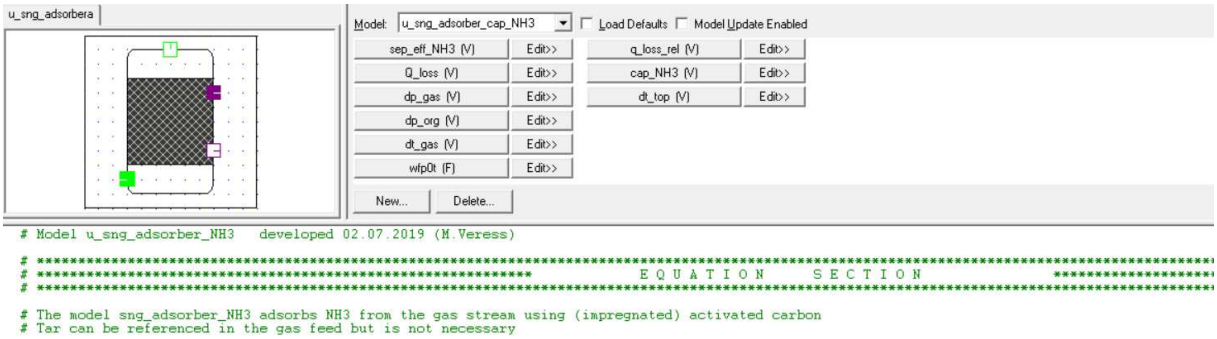

Figure A10: Simulation model of an acid-impregnated activated carbon adsorber

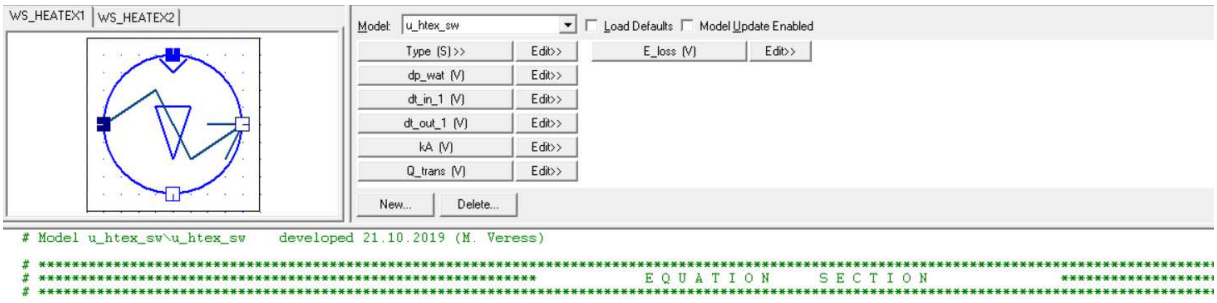

Figure A11: Simulation model of a water-solid heat exchanger

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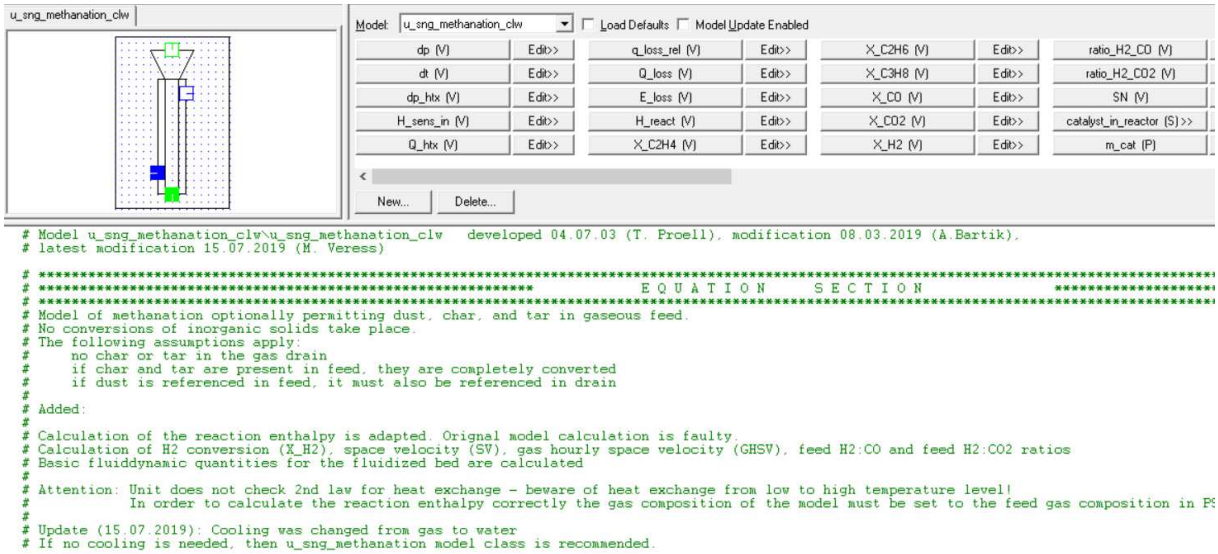

Figure A12: Simulation model of a fluidized bed methanation reactor with water cooling

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B. Extended stream tables

The target values in Table B1 are taken from internal research data of TU Wien for methanation catalysts, which are reported in reference [43].

Table B1: Gas stream properties from the gasification to the methanation reactor

Gas Stream Property	Unit	Gas production	Gas cooling	Gas cleaning										Target	
		Exit gasifier	Exit PG cooler	Exit PG filter	Exit RME scrubber	Exit PG blower	Exit amine scrubber	Exit AC bed	Exit water scrubber	Exit acid AC	Exit PG compressor	Exit PG heater	Exit Fe AC	Inlet methanation	
Intensive properties															
Pressure	bar	1	1	0.96	0.95	1.1	1.09	1	1	0.98	5.1	5.1	5	5	~ 5 ✓
Temperature	°C	810	180	180	40	63	40	40	40	39	242	350	350	354	~ 350 ✓
Density	kg/m³	0.21	0.51	0.49	0.74	0.80	0.85	0.81	0.80	0.76	2.41	2.00	1.96	1.03	- ✓
Flow properties															
Mass flow (gas)	kg_{gas}/h	4625	4625	4625	2371	2371	1726	1724	1673	1672	1672	1672	1670	1939	- ✓
Standard volume flow (gas)	Nm³/h	5412	5412	5412	2606	2606	1913	1912	1849	1848	1848	1848	1847	4055	- ✓
Operation volume flow (gas)	m³/h	21856	9144	9477	3187	2958	2038	2137	2086	2186	693	838	854	1886	- ✓
Combustion characteristics															
Net calorific value (LHV)	MJ/Nm^3_{db}	12.2	12.2	12.2	12.2	12.2	12.2	12.2	12.0	12.0	12.0	12.0	12.0	11.5	- ✓
Gross calorific value (HHV)	MJ/Nm^3_{db}	13.6	13.6	13.6	13.5	13.5	13.6	13.5	13.4	13.4	13.4	13.4	13.4	13.2	- ✓
Chemical composition															
H_2O	vol%	55.0	55.0	55.0	7.8	7.8	6.8	6.8	7.0	7.0	7.0	7.0	7.0	3.2	≤10 ✓
C_2H_4	$vol\%_{db}$	2.4	2.4	2.4	2.5	2.5	2.5	2.5	2.6	2.6	2.6	2.6	2.6	1.1	- ✓
C_2H_6	$vol\%_{db}$	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.0	≤0.2 ✓
C_3H_8	$vol\%_{db}$	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.3	≤0.4 ✓
CH_4	$vol\%_{db}$	7.9	7.9	7.9	8.0	8.0	8.1	8.1	8.4	8.4	8.4	8.4	8.4	4.7	- ✓
CO	$vol\%_{db}$	16.8	16.8	16.8	17.0	17.0	17.2	17.2	17.8	17.9	17.9	17.9	17.9	7.8	- ✓
CO_2	$vol\%_{db}$	26	26	26	26.3	26.3	26	26	27	27	27	27	27	12.4	- ✓
H_2	$vol\%_{db}$	39.7	39.7	39.7	40.2	40.2	40.7	40.7	42.2	42.2	42.2	42.2	42.3	73.1	$H_2 = 3 \cdot CO + 4 \cdot CO_2$ ✓
N_2	$vol\%_{db}$	1	1	1	1	1	1	1	1.1	1.1	1.1	1.1	1.1	0.5	≤0.8 ✓
H_2S + COS	$ppmv_{db}$	6000	6000	6000	6080	6080	1100	700	726	726	726	726	5	2	≤10.8 ✓
HCl	$ppmv_{db}$	70	70	70	71	71	72	72	2	2	2	2	2	0.9	≤2 ✓
HCN	$ppmv_{db}$	4400	4400	4400	4458	4458	4508	4510	0.8	0.8	0.8	0.8	0.8	0.4	≤0.8 ✓
NH_3	$ppmv_{db}$	43600	43600	43600	30924	30924	31268	31280	500	5	5	5	5	2	≤5 ✓
O_2	$ppmv_{db}$	0	0	0	0	0	0	0	0	0	0	0	0	0	0 ✓
Fly dust content	g/Nm³	252.06	252.06	0.3	0	0	0	0	0	0	0	0	0	0	- ✓
Fly char content	g/Nm³	3.2	3.2	0	0	0	0	0	0	0	0	0	0	0	- ✓
Tar content	g/Nm³	10	10	7	1.5	1.5	1.53	0	0	0	0	0	0	0	≤3 ✓

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The target values in Table B2 are taken from the ÖVGW G31 and G33 (Table 11).

Table B2: Gas stream properties from the methanation reactor to the final product

Gas Stream Property	Unit	Methanation	Gas upgrading and utilization						Target	
		Exit methanation reactor	Exit condenser	Exit CO_2 amine scrubber	Exit SNG compressor	Exit SNG cooler	Exit glycol scrubber	Exit H_2 membrane / Upgraded SNG		
Intensive properties										
Pressure	bar	5	5	4.8	40	40	40	39.7	-	✓
Temperature	°C	350	40	40	270	40	40	40	-	✓
Density	kg/m^3	1.60	3.00	2.72	12.96	22.50	22.41	24.50	-	✓
Flow properties										
Mass flow (gas)	kg_{gas}/h	1939	924	799	799	799	783	742	-	✓
Standard volume flow (gas)	Nm^3/h	2550	1286	1223	1223	1223	1204	1035	-	✓
Operation volume flow (gas)	m^3/h	1215	308	294	62	36	35	30	-	✓
Combustion characteristics										
Net calorific value (LHV)	MJ/Nm^3_{db}	30.6	30.6	32.3	32.3	32.3	32.3	34.8	-	✓
Gross calorific value (HHV)	MJ/Nm^3_{db}	34.1	34.1	35.9	35.9	35.9	35.9	38.6	38.5 - 46.1	✓
Wobbe index	MJ/Nm^3_{db}	not calculated						51.8	47.9 - 56.5	✓
Relative density	-							0.56	0.55 - 0.65	✓
Chemical composition										
H_2O	vol%	50.3	1.5	1.6	1.6	1.6	0.01	0.01	T_{sat} = max -8°C at 40 bar	✓
C_2H_4	$vol\%_{db}$	0	0	0	0	0	0	0	-	✓
C_2H_6	$vol\%_{db}$	0	0	0	0	0	0	0	-	✓
C_3H_8	$vol\%_{db}$	0	0	0	0	0	0	0	-	✓
CH_4	$vol\%_{db}$	82.2	82.2	86.6	86.6	86.6	86.6	96.8	≥ 96	✓
CO	$vol\%_{db}$	0	0	0	0	0	0	0	-	✓
CO_2	$vol\%_{db}$	5.0	5.0	0	0	0	0	0	≤ 2	✓
H_2	$vol\%_{db}$	11.2	11.2	11.8	11.8	11.8	11.8	1.4	≤ 4	✓
N_2	$vol\%_{db}$	1.5	1.5	1.6	1.6	1.6	1.6	1.8	≤ 5	✓
H_2S + COS	$ppmv_{db}$	7	7	7	7	7	7	8	≤ ~ 10	✓
HCl	$ppmv_{db}$	3	3	3	3	3	3	3	0	(✓)
HCN	$ppmv_{db}$	1	1	1	1	1	1	1	-	✓
NH_3	$ppmv_{db}$	7	7	7	7	7	7	8	technically free	✓
O_2	$ppmv_{db}$	0	0	0	0	0	0	0	≤ 0.5	✓
It can be assumed that all the other (here not explicitly mentioned) criteria from ÖVGW 31 and 33 are met										

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Table B3: Gas stream properties of the flue gas stream

Gas Stream Property	Unit	Exit CR	Exit seperator	Exit combustion chamber	Exit FG filter	Inlet FG cleaning
Intensive properties						
Pressure	bar	0.98	0.95	0.93	0.81	1
Temperature	°C	895	895	850	150	187
Density	kg/m^3	0.29	0.28	0.27	0.62	0.71
Flow properties						
Mass flow (gas)	kg_{gas}/h	11261	11261	12536	12536	10150
Standard volume flow (gas)	Nm^3/h	8875	8875	10368	10368	8395
Operation volume flow (gas)	m^3/h	39245	40484	46448	20217	14386
Chemical composition						
H_2O	vol%	16.9	16.9	27.9	27.9	27.9
Ar	$vol\%_{db}$	0.94	0.94	0.94	0.94	0.94
O_2	$vol\%_{db}$	3.9	3.9	3.7	3.7	3.7
N_2	$vol\%_{db}$	79.6	79.6	80.2	80.2	80.2
CH_4	$vol\%_{db}$	0	0	0	0	0
CO	$vol\%_{db}$	0.7	0.7	0.0	0.0	0.0
CO_2	$vol\%_{db}$	13.6	13.6	14.1	14.1	14.1
H_2	$vol\%_{db}$	0	0	0	0	0
H_2S	$ppmv_{db}$	0	0	0	0	0
HCl	$ppmv_{db}$	153	153	151	151	151
HCN	$ppmv_{db}$	0	0	0	0	0
NO	$ppmv_{db}$	9796	9796	7744	7744	7744
NH_3	$ppmv_{db}$	0	0	682	682	682
SO_2	$ppmv_{db}$	2051	2051	2023	2023	2023
Fly dust content	g/Nm^3	5728	157	157	0	0
Fly char content	g/Nm^3	0	0	0	0	0
Tar content	g/Nm^3	0	0	0	0	0

C. Adsorber size estimation

The best possible cost data found for adsorbers is dependent on the adsorber size. Therefore, a quick reactor sizing had to be performed in order to be able to calculate the costs connected to them. The adsorber size estimation was carried out according to the mass balance based short-cut-method from [135]. The procedure is presented by the example of the activated carbon filter. Similar calculations were carried out for the acid-impregnated activated carbon bed and the Fe-impregnated activated carbon bed.

Table C1: Calculation data for the activated carbon bed filter

Data for calculation			
Superficial gas velocity	0.4	m/s	[135]
Bulk density	2200	kg/m³	[136]
Pore volume	0.3	-	[136]
Adsorption time	120	h	Set
Adsorbent capacity	0.2	g_{tar}/g_{AC}	[103]
Tar concentration in gas	1.5	g/Nm³	Simulation
Gas volume flow	1913	Nm³/h	Simulation
Safety factor	1.2	-	[135]

The adsorber cross-section $A_{Adsorber}$ and diameter $d_{Adsorber}$ can be calculated as follows:

$$A_{Adsorber} = \frac{1912.59\,\frac{Nm^3}{h}}{3600\,\frac{s}{h} \cdot 0.4\,\frac{m}{s}} = 1.33m^2$$

$$d_{Adsorber} = \sqrt{\frac{4 \cdot 1.33m^2}{\pi}} = 1.3m$$

This permits the calculation of the adsorbed tar mass $M_{Adsorbent}$ within an adsorption cycle:

$$M_{Adsorbent} = \frac{1.5\,\frac{g}{Nm^3} \cdot \frac{kg}{1000g} \cdot 1912.59\,\frac{Nm^3}{h} \cdot 120h}{0.2\,\frac{g}{g}} = 1721kg$$

The height of the adsorber $h_{Adsorber}$ results in:

$$h_{Adsorber} = \frac{M_{Adsorbent}}{400\,\frac{kg}{m^3} \cdot (1 - 0.3) \cdot A_{Adsorber}} = 4.63m$$

With the application of a safety factor the real adsorber height $h_{Adsorber,real}$ can be obtained. If the ratio between $h_{Adsorber,real}$ and the $d_{Adsorber}$ lies within a technically reasonable range of 3-5, the chosen adsorption time is realistic. If the adsorber is too large/small, the adsorption time must be reduced/increased and the calculation has to be restarted.

$$h_{Adsorber,real} = 4.63m \cdot 1.2 = 5.6m$$

$$\frac{h_{Adsorber,real}}{d_{Adsorber}} = 4.3$$

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D. Extended equipment cost calculation

Table D1 lists the equipment cost calculation for all the major equipments of the process. For the heat exchangers a calculation of each of the respective surface needed to be performed. This was done according to the procedure and material data used by [94].

Table D1: Equipment cost calculation

Equipment	Base scale	Base cost	Scale factor	Scale in the present study	Equipment cost (€, 2018)	Source
Main equipment						
MDEA scrubber	14,040 Nm^3/h	5,190,000 €, 2010	0.6	1,955 Nm^3/h	1,827,389	[114]
Activated carbon					303,088	[119]
Water scrubber	43,560 m^3/h	3,000,000 €, 2002	0.7	2,137 m^3/h	581,394	[120]
Acid impregnated AC					145,247	[119]
Fe impregnated AC					146,337	[119]
ZnO guard reactor	81,360 kg/h	49,000 €, 2009	0.56	2,842 kg/h	11,892	[121]
Methanation reactor	43,560 Nm^3/h	2,860,000 €, 2010	0.6	4,055 Nm^3/h	836,467	[114]
Condenser	448 MW	4,700,000 $, 2007	0.7	1.06 MW	74,504	[122]
MEA scrubber	14,040 Nm^3/h	5,190,000 €, 2010	0.6	1,286 Nm^3/h	1,421,736	[114]
Glycol scrubber	41,667 m^3/h	316,386 $, 2006	0.6	1,223 Nm^3/h	43,707	[123]
Membrane	1 m^2	3,531 $, 2006	1	42 m^2	169,003	[114]
Hydrogen storage	1 kg	470 €, 2017	1	1,450 kg	760,660	[124]
Blowers / Compressors						
PG Compressor	224 kW	133,000 $, 1994	0.84	183 kW	174,977	[88]
SNG compressor	224 kW	133,000 $, 1994	0.84	160 kW	156,627	[88]
H_2 addition blower	16,992 Nm^3/h	67,000 $, 1994	0.6	2,016 Nm^3/h	27,679	[88]
H_2 addition blower motor	7.5 kW	12,300 $, 1994	0.56	11.6 kW	24,446	[88]
H_2 recycle blower	16,992 Nm^3/h	67,000 $, 1994	0.6	169 Nm^3/h	6,256	[88]
H_2 recycle motor	7.5 kW	12,300 $, 1994	0.56	2.3 kW	9,917	[88]
CO_2 recycle blower	16,992 Nm^3/h	67,000 $, 1994	0.6	63 Nm^3/h	3,474	[88]
CO_2 recycle motor	7.5 kW	12,300 $, 1994	0.56	0.2 kW	2,436	[88]
Pumps						
Water circuit pump	7.5 kW	1,600 $, 1994	0.3	1 kW	1,365	[88]
Water circuit pump motor	7.5 kW	12,300 $, 1994	0.56	1.0 kW	6,221	[88]
Cooling circuit pump	7.5 kW	1,600 $, 1994	0.3	4 kW	2,074	[88]
Cooling circuit pump motor	7.5 kW	12,300 $, 1994	0.56	4.1 kW	13,585	[88]
Heat exchangers						
PG Heater	93 m^2	21,700 $, 1994	0.59	170 m^2	45,980	[88]
SNG cooler	93 m^2	21,700 $, 1994	0.59	50 m^2	22,336	[88]
H_2 heater	93 m^2	21,700 $, 1994	0.59	40 m^2	19,580	[88]
Water circuit cooler	93 m^2	21,700 $, 1994	0.59	90 m^2	31,594	[88]
Water add preheater 1	93 m^2	21,700 $, 1994	0.59	5 m^2	5,741	[88]
Water add preheater 2	93 m^2	21,700 $, 1994	0.59	20 m^2	13,008	[88]
Scrubbing water preheater	93 m^2	21,700 $, 1994	0.59	5 m^2	5,741	[88]
Steam generator	93 m^2	21,700 $, 1994	0.59	450 m^2	81,659	[88]
Water cooler solvscrub	93 m^2	21,700 $, 1994	0.59	250 m^2	57,729	[88]
Total cost of delivered equipment					**7,033,848**	

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E. CEPCI Factors

Table E1: CEPCI factors for equipment cost updating [88–90]

Year	Chemical Engineering Plant Cost Index (CEPCI)
1987	320
1988	343
1989	355
1990	358
1991	361
1992	358
1993	359
1994	368
1995	381
1996	382
1997	387
1998	390
1999	391
2000	394
2001	394
2002	396
2003	402
2004	444
2005	468
2006	500
2007	525
2008	575
2009	521
2010	551
2011	586
2012	585
2013	567
2014	576
2015	557
2016	542
2017	568
2018	603

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F. Calculation of the working capital

Table F1: Calculation of the working capital

Working Capital					
Bed material	49,452	kg	7,665	€	Investment in initial fill
	18,250	kg	131	€	Inventary for 1 month production
RME	105,366	kg	101,151	€	Investment in initial fill
	16,060	kg	15,141	€	Inventary for 1 month production
MDEA	478	kg	828	€	Investment in initial fill
	0,022	kg	0.04	€	Inventary for 1 month production
AC	1,721	kg	3,804	€	Investment in initial fill
Acid-impregnated AC	1,090	kg	3,434	€	Investment in initial fill
	2,920	kg	8,717	€	Inventary 1 month production
Fe-impregnated AC	1,204	kg	3,852	€	Investment in initial fill
	584	kg	1,857	€	Inventary 1 month production
Water (scrubber)	25,867	kg	52	€	Investment in initial fill
H_2	132,130	kg	463,213	€	Inventary 1 month production
MEA	7,785	kg	10,898	€	Investment in initial fill
	0.3	kg	0.4	€	Inventary 1 month production
TEG	528	kg	1,055	€	Investment in initial fill
	3.7	kg	7	€	Inventary 1 month production
Methanation catalyst	400	kg	20,000	€	Investment in initial fill
Water circuit	11,045	kg	22	€	Investment in initial fill
Cooling water circuit	89,738	kg	179	€	Investment in initial fill
Sum			**642,009**	**€**	
Rounded sum			**1,000,000**	**€**	

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G. Cost rates

Table G1: Cost rates for the NPV and levelized cost method

Parameters for Net Present Value Calculation			
Parameter	**Unit**	**Value**	**Source**
Plant parameters			
Location of Plant	-	Austria	Set
Reference year	-	2018	Set
Plant lifetime	a	20	[127]
Number of employees	1/a	3	[128]
Operating hours plant	h/a	7,500	[95]
Operating hours district heating	h/a	5,800	[95]
Cost rates for expenses			
Consumption related costs			
Natural gas	€/MWh	36	[137]
Hard wood chips	€/MWh	22.5	[13]
Added hydrogen	€/kg	3.5	[138]
Fresh water	€/t	2	[13,127,139]
Fresh scrubber solvent (RME)	€/kg	0.96	[13]
Fresh amine (MDEA)	€/kg	1.73	[140]
Fresh amine (MEA)	€/kg	1.4	[141]
Fresh glycol (TEG)	€/kg	2	[142]
Activated carbon	€/kg	2.21	[100]
Acid-impregnated activated carbon	€/kg	3.15	[100]
Fe-impregnated activated carbon	€/kg	3.2	Assumption
Zinc oxide	€/kg	20	[127]
Olivine	€/t	185	[13]
Limestone	€/t	35	[128]
Nickel catalyst	€/kg	50	[142]
Disposal costs ash	€/t	90	[13]
Disposal costs wastewater	€/m^3	2.5	[139]
CO_2-emission certificates	€/t_{CO2}	24.9	[143]
Electricity costs	€/kWh_{el}	0.104	[144]
Operation related costs			
Maintenance costs	%$_{Io}$/a	2.0	[13]
Insurance, administration and other costs	%$_{Io}$/a	1.50	[145,146]
Personnel costs per employee	€/a	62,500	[145,146]
Cost rates for incomes			
Biomass (sewage sludge, 25% DM)	€/t	60	[147]
Feed-in tariff district heating	€/MWh	30	[94]

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H. GoBiGas plant scale-down

The plant costs of the GoBiGas plant were scaled down from an SNG output of 20 MW to 10 MW to have a basis of comparison for the presented sewage sludge. The cost data of Table H2 and H3 derive from reference [125], where the cost data of an optimized GoBiGas reference plant was taken for the scale down. The plant parameters of the GoBiGas plant are shown in Table H1. To have a fair comparison within the business case of Chapter 6.2 the following adaptations of the cost calculation were made:

- Operating hours were reduced to 7500 h
- The wood chip price was set to 22.5 €/MWh [13]
- The district heating output was included with a scale factor of 1
- The plant costs were multiplied with a location index of 0.96 [131] to account for the price level differences due to the shift in location to Austria.

Table H1: GoBiGas plant parameters

GoBiGas plant parameters			
Parameter	**Unit**	**Value**	**Source**
Thermal fuel power	MW_{th}	32	[125]
SNG output	MW	20	[125]
District heating	GWh/year	50	[15]
Electricity consumption	MW	3	[15]
Operating hours	h/a	8,000	[125]
Number of employees	1/a	28	[125]

Table H2: Calculation of the total investment cost of a down-scaled GoBiGas plant

Total Investment Cost				
Cost parameter	**20 MW (SEK)**	**20 MW (€)**	**Scale factor**	**10 MW (€)**
Reactor systems	238,000,000	21,774,931	0.68	13,591,156
Auxiliary equipment and project	955,000,000	87,374,199	0.44	64,406,546
Steam cycle, external fuel handling	182,000,000	16,651,418	0.67	10,465,528
Total	**1,375,000,000**	**125,800,549**		**77,997,702**

Table H3: Calculation of the operating costs of a down-scaled GoBiGas plant

Operating Costs					
Cost parameter	**20 MW (SEK/MWh)**	**20 MW (€/MWh)**	**20 MW (€/a)**	**Scale factor**	**10 MW (€/a)**
Personnel	181	16.6	2,483,989	0.1	2,317,644
Maintenance	89	8.1	1,221,409	0.67	767,664
Electricity	0	0.0	0	1	0
RME	0	0.0	0	1	0
Activated carbon	10	0.9	137,237	1	68,618
Other	45.1	4.1	618,939	1	309,469
Other costs	26.5	2.4	363,678	0.67	228,574
Total	**351.6**	**32**	**4,825,252**		**3,691,969**

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I. Activated carbon data sheets

KENNDATENBLATT / TECHNICAL DATA SHEET

Desorex K 47

Zylindrisch geformte Aktivkohle

®Desorex K 47 ist eine wasserdampfaktivierte Aktivkohle auf der Basis von Steinkohle mit ausgezeichneter Härte und mechanischer Abriebfestigkeit.
®Desorex K 47 wird im Bereich Abluftreinigung für die Abscheidung von organischen Stoffen und Lösemittel eingesetzt.

Spezifikation:

Rütteldichte (kg/m³)		500 ± 30
Wassergehalt (Gew.-%) (beim Abpacken)		< 8
Benzoladsorption (Gew.-%) aus Luft bei 20°C		
p/p_S^*	0,9	> 31
	0,1	> 26
	0,01	> 21
	0,001	> 13

Typische Eigenschaften:

Innere Oberfläche (m²/g) (BET-Methode)	> 900
Korndurchmesser (mm)	ca. 4
CCl4-Adsorption (Gew.-%)	> 50

Bemerkungen:

Die genannten Daten basieren auf Untersuchungen der Aktivkohle im Anlieferungszustand und Analysenmethoden gemäß ASTM und von Donau Carbon. Diese sind auf Anfrage erhältlich.

*p/p_S = relative Sättigung (Sättigungskonzentration bei 20°C, 320 g/m³)

d1307/07

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Figure I1: Data sheet of the commercial activated carbon Desorex K 47

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KENNDATENBLATT / TECHNICAL DATA SHEET

Desorex K 43 P10

Zylindrisch geformte Aktivkohle / imprägniert

Typische Eigenschaften:

Imprägnierung (Gew.-%)	9,5 - 9,9
Rütteldichte (kg/m³)	550 ± 30
Wassergehalt (Gew.-%) (beim Abpacken)	< 20
pH-Wert	ca. 1,5
Korndurchmesser (mm)	ca. 4

Produkt-Kenndaten vor der Imprägnierung:

Innere Oberfläche (m²/g) (BET-Methode)	1000
CCl_4-Adsorption (Gew.-%)	> 60

Bemerkungen:

Säure-imprägnierte Aktivkohle für die Abscheidung von basischen Schadstoffen.

Die genannten Daten basieren auf Analysenmethoden der Donau Carbon.
Diese sind auf Anfrage erhältlich.

Desorex K 43P10 ist schwach ätzend. Kontakt mit Haut und Augen vermeiden; betroffene Stellen mit reichlich Wasser spülen, ggf. Arzt aufsuchen!

d1301/02

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Figure I2: Data sheet of the commercial activated carbon Desorex K 43 P10

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J. Publication

Poster

Veress M., Bartik A., Benedikt F., Müller S., Hofbauer H.

"Optimization of industrial Bio-SNG production from low-grade fuels"

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Optimization of industrial Bio-SNG production from low-grade fuels

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Motivation

Previous developments have shown that the production of synthetic natural gas (SNG) is technically feasible in combination with the dual fluidized bed (DFB) steam gasification. However, the technology struggles to compete economically with fossil natural gas, when high-quality woody biomass is utilized for the gasification process. Hence the aim of the presented study is:

- the development of an industrial Bio-SNG concept using a low-grade fuel (sewage sludge),
- an implementation of a simulation model,
- a techno-economic comparison with the GoBiGas demonstration plant.

Methodology & Concept

The investigations in this work are based on a Bio-SNG concept with an output of 10 MW_{SNG}. The process chain in Fig. 1 is designed for the conversion of sewage sludge to SNG and was elaborated with the aid of an extensive literature study.

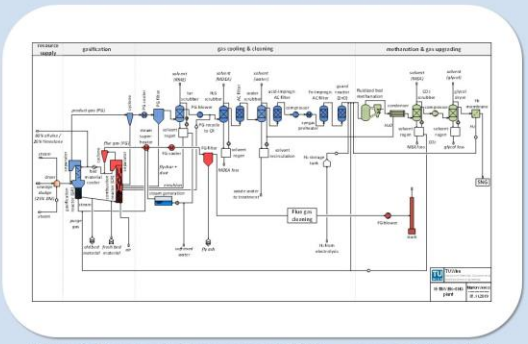

Figure 1: Flowsheet of the proposed 10MW_{SNG} sewage sludge plant

For the calculation of the mass- and energy balances and the evaluation of the plant parameters the simulation software IPSEpro was used.

Results

Table 1 shows relevant mass and energy flows of the simulation model. A biomass-to-SNG efficiency of **58,5%** could be calculated from these results, which corresponds with typical literature values for Bio-SNG production.

Table 1: Important mass and energy flows of the simulation

Plant input			Plant output		
Biomass (sewage sludge, 25 wt% dry matter)	kg/h	13532	SNG	Nm^3/h	1034
Biomass (chemical energy - before drying)	kW	4514	SNG (chemical energy)	kW	10000
Added hydrogen	Nm^3/h	1896	District heating	kW	2675
Added hydrogen (chemical energy)	kW	5682	Exhaust gas	Nm^3/h	8220
Electricity consumption	kW	858	Fly ash and dust	kg/h	1429

In Fig. 2 a comparison was carried out between:

- the GoBiGas concept (down-scaled to 10 MW_{SNG}),
- the proposed 10 MW_{SNG} sewage sludge concept.

As illustrated, a significant cost reduction in investment and operating costs can be achieved by the suggested process compared to the GoBiGas demonstration plant.

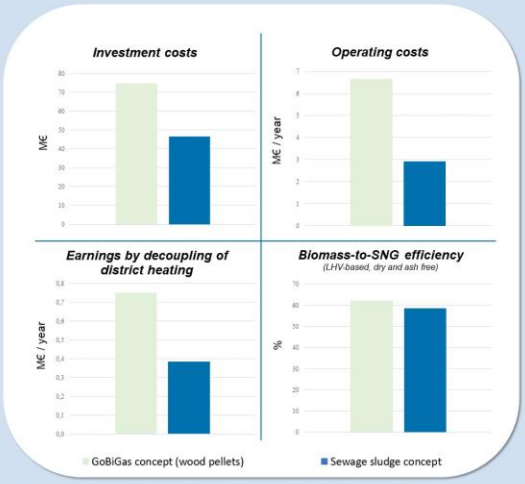

Figure 2: Comparison of the proposed concept and the GoBiGas set-up

Conclusion

The results indicate a great potential of the technology from both the economic and technical point of view. This study makes a further step towards economic competitiveness of Bio-SNG with fossil natural gas. However, in the future further optimizations of the process and a demonstration in reasonable scale are necessary.

Literature:
Thunman H, Gustavsson C, Larsson A, Gunnarsson I, Tengberg F. Economic assessment of advanced biofuel production via gasification using cost data from the GoBiGas plant. Energy Sci Eng 2019;7:217-229

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