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NATURE

ENERGY TECHNOLOGY DEVELOPMENT FOR INDUSTRIAL APPLICATION

MODELLING-BASED DEVELOPMENT OF PROCESSES ENABLING REDUCED FOSSIL CARBON DIOXIDE EMISSIONS BY ADVANCED DIGITAL METHODS

For the fulfillment of the requirements
for the *venia docendi* in the fields of
Energy Process Technology & Industrial Plant Design

Approved by the
Faculty of Technical Chemistry and the
Institute of Chemical, Environmental and
Bioscience Engineering

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PREFACE

The upcoming challenges for the transformation of our current energy system suggests a situation where well-thought-out, effective results are needed urgently. At the same time, the quick acceleration of digitalization activities inhibits the danger of wide scale system failures as well as significant security risks. The technological progress within the last century has led to the development of prospering societies in large parts of our world. Therefore, well-thought-out technological solutions form an important basis for the development of a peaceful and prosperous society in the future.

With the present work, I would like to make a contribution to enable a reliable and target-oriented development of energy technology by smart application of advanced digital methods. The presented approach is based on results and practical experiences collected during my work in the process development environment at the Institute of Chemical, Environmental and Bioscience Engineering at TU Wien between 2010 and 2022. Reliability is an important value for the implementation of advanced digital methods. Therefore, the present work aims to present a systematic approach to ensure reliability of energy technology development by advanced digital methods as a basis for industrial plant design. An accompanying [modelling readiness level](#) is introduced as follows:

1. definition of the main development goals;
2. basic standards for model development;
3. guidelines for quality management of integrated model libraries;
4. boundary conditions used to prepare test plant models;
5. validation of experimental test plants following the idea of a digital twin;
6. evaluation of results following the idea of ecologic value;
7. transformation of validated process models into industrial plant models;
8. validation of industrial plant information models by successful demonstration projects; and
9. a collaborative strategy via platform-based implementation, monitoring and sharing.

The presented approach is explained via practical results achieved in the past and summarized by an endeavor aiming to describe a well-thought-out methodology for stronger integration of advanced digital methods during technology development.

The scientific basis for this summary is described by the included publications in the annex of the present work. [Paper I](#) summarizes the achievements made in the field of the experimental development of sorption enhanced reforming by using an advanced gasification test plant. [Paper II](#) demonstrates promising results when utilizing CO₂ as a gasification agent. [Paper III](#) illustrates the progress made to enable the future application of fuel flexible gasification systems. [Paper IV](#) illustrates the large-scale production of hydrogen (H₂). [Paper V](#) prepares the experimental investigation of synthetic natural gas production from biogenic residues by thermodynamic investigations. [Paper VI](#) presents experimental results of SNG production. [Paper VII](#) investigates the production of diesel from biomass and wind power to enable energy storage by the use of the Fischer–Tropsch process. [Paper VIII](#) describes a future-oriented evaluation of biomass gasification to enable a below zero emission supply of reducing gas for the iron and steel industry. Finally, [Paper IX](#) investigates an overall concept for the reduction of fossil CO₂ emission in the context of industrial hot metal production.

The results have been achieved by a highly motivated interdisciplinary team. My personal contribution can be summarized as follows. During my scientific career, the perspective of modelling-based process development has dominated my approach to the execution of experimental development. Therefore, this perspective has also dominated my personal contribution to the executed research work: First, as the creator of process simulation models. Later, as the provider of a database for the erection of test plants as well as an engineer executing the validation. Finally, as an industrial engineer enabling the interpretation of results with respect to large-scale implementation.

It must be mentioned once more that the described work has depended on a highly motivated interdisciplinary team each in different phases of their scientific career. The present work represents an important contribution of my scientific career so far as an industrial engineer, data scientist, model developer, chemical engineer, certified project manager and the head of a research group. At this stage, I would like to thank my exceptional colleagues. I would like to thank the testing engineers from the laboratory for combustion systems as well as all members of the research team who have contributed important perspectives to realize the present work. Thank you all for your kind support during the demanding, hard work as well as many the inspiring moments. Your support has enabled the creation of engineering solutions. A special thank you is given to: Hermann Hofbauer, Michael Fuchs, Tobias Pröll, Reinhard Rauch, Johannes Schmid, Roland Diem, Maximilian Kolbitsch, Martin Hammerschmid, Constantin Walcher, Josef Fuchs, Florian Benedikt, Anna Mauerhofer, Alexander Bartik, Benjamin Fleiß, Tom Popov, Veronica Gubin, Ferdinand Thelen, Sebastian Diem, Jürgen Loipersböck, Katharina Fürsatz, Gerald Weber, Matthias Kuba, Harald Kohl, Christine Wukovits, Julia Hofbauer, Edith Partl, Thomas Weinberger, Lisa Adam, Andreas Lunzer, Stefan Jankovic, Herbert Weissenbaeck, Anton Friedl and Robert Mach.



Don't be afraid, the present work is not the end of the road. It is the beginning for hard work enabling target-oriented implementation in near future. So stay ready and prepare yourself for tough projects to come.

Ing. Dipl.-Ing. Dr. Stefan Müller

PHILOSOPHY

Statement of Mohandas Karamchand Gandhi:

An etwas zu glauben und es nicht zu leben, ist unehrlich. Ziel im Leben ist es, richtig zu leben, zu denken und zu handeln. Die Welt ist ausreichend für die Bedürfnisse des Menschen, aber nicht für seine Habgier. Die Zukunft hängt davon ab, was du heute tust.

Statement of the First Female President of the European Commission:

Our goal is to be the first climate neutral continent by 2050. If we want to achieve that goal, we have to act now, we have to implement our policies now. Because we know that this transition needs a generational change. The European Green Deal is Europe's new growth strategy. It will cut emissions while also creating jobs and improving our quality of life. For that we need investment! Investment in research, innovation, green technologies. To pull this off, we will deliver a Sustainable Europe Investment Plan – which will support one trillion euros of investment over the next decade. Therefore, in March next year, we will propose the first-ever European Climate Law to make the transition to climate neutrality irreversible. This will include extending emission trading to all relevant sectors, clean, affordable and secure energy, the boosting of the circular economy as well as a biodiversity strategy. Brussels, December 2nd, 2019

ACKNOWLEDGMENT

The present work summarizes the outcome of several research projects conducted at the Institute of Chemical, Environmental and Bioscience Engineering at TU Wien. A special thank is given to the related funding agencies and project partners of following projects:

ERBA II

Optimization of "sorption enhanced reforming" for an improved CO₂-balance of a raw iron production by the use of biomass (515766)

RENEWABLE STEEL GASES

Integration of renewable energy in the steel production in order to increase energy efficiency and to reduce CO₂ emissions (1377908)

BIOH₂ 4REFINERIES

Production of hydrogen for refineries via biomass gasification (187952)

REGAS 4 INDUSTRY

Gases from renewable waste sources for industrial use (1658177)

WINDDIESEL

Investigation of the "Load-Change-Behavior" of a slurry reactor for the integration of H₂ and production of FT-diesel (453423)

HEAT TO FUEL

Upgrading of alternative, residual biomass feedstock and conversion of excess heat to liquid fuels in a combined gasification, Fischer-Tropsch and Aqueous Phase Reforming plant (722385)

This is just a few of the projects. The latest results can be found at <https://tiss.tuwien.ac.at/pdb/projects/> (as of September 1, 2021)

ABSTRACT

The present work provides a proposal for a methodology for the development of energy technology supported by advanced digital methods. The proposed methodology was found as a result of the gained experience during many different research and development projects aiming at a reduction of fossil carbon dioxide (CO₂) emissions via the utilization of biogenic raw materials. The investigated processes included the production of a hydrogen-rich gas from biomass, the production of synthetic natural gas as well as the production of liquid biofuels.

The transformation of experimental results into industrial scale supported by modern software tools highlighted important aspects of the interaction between development goals, experimental campaigns, measurements and modern software tools. As a result, the introduction of a methodology based on modelling readiness levels can be name as important measure to accompany process development from the process idea until the erection of an industrial plant.

The proposed methodology ensures a well-balance monitoring of set development goals during the entire development process and aims at the generation of digital role models preparing a best possible implementation of novel energy technology. The target-oriented implementation of advanced digital methods could enable an acceleration of the transition of scientific and experimental results into real industrial processes, if the implementation is executed in a smart manner.

KURZFASSUNG

Die vorliegende Arbeit erläutert den Vorschlag einer Methode für die Entwicklung von Energietechnologien unterstützt durch moderne digitale Methoden. Die vorgestellte Methode wurde auf Basis langjähriger Erfahrung im Rahmen verschiedener Forschungs- und Entwicklungsprojekte mit der Zielsetzung der Reduktion fossiler Kohlenstoffdioxid (CO₂) Emissionen durch den Einsatz biogener Rohstoffe entwickelt. Die untersuchten Verfahren beinhalteten dabei die Produktion von wasserstoffreichem Gas aus Biomasse, die Produktion von synthetischem Erdgas sowie die Produktion von flüssigen Biokraftstoffen.

Die Übersetzung von experimentellen Ergebnissen in den industriellen Maßstab unterstützt durch moderne Software veranschaulicht das Zusammenspiel zwischen Entwicklungszielen, Experimenten, Messungen und modernen IT Systemen. Als Ergebnis der Arbeit kann die Empfehlung der Einführung von „Modelling Readiness Levels“ hervorgehoben werden, um Prozessentwicklung von der Prozessidee bis zur Errichtung einer industriellen Anlage bestmöglich zu begleiten.

Die vorgeschlagene Methode stellt eine ausbalancierte Überwachung der Entwicklungsziele über den gesamten Entwicklungsprozess sicher und verfolgt das Ziel der Bereitstellung digitaler Vorzeigemodelle um die Technologieimplementierung bestmöglich vorzubereiten. Der zielgerichtete Einsatz von digitalen Modellen ermöglicht an dieser Stelle die Konservierung und rasche Überführung wissenschaftlicher bzw. experimenteller Ergebnisse in reale industrielle Prozesse.

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1. INTRODUCTION

The observed global temperature increase has raised major concerns regarding future development of the global climate. In 2017, Johan Rockström published the results of an analysis with respect to the necessary speed for the transition of our energy system. His work indicates that rapid implementation of effective measures is needed urgently to limit the increase in the global mean temperature in the year 2100 to below 1.5°C compared with the year 1850. From a scientific point of view:

- 50% reduction in fossil carbon dioxide (CO₂) emissions within the next 10 years,
- 100% increase in fossil CO₂ free energy supply within the next 5 years and
- the active removal of CO₂ from the atmosphere.

would be necessary to reach the defined temperature goals.¹ As a result, in 2019 the European Commission formulated the goal to become the first climate-neutral continent in the world by the year 2050. The so-called “Green Deal” and the accompanying programs from the European Commission indicate a major change in the European Energy Policy.^{2,3} This would mean far-reaching changes in the existing energy system in Europe as well as rapid changes in the applied fuel conversion and energy technology. Therefore, following question needs to be answered satisfying in near future: How can novel processes be applied most suitably to reach a 100% reduction in fossil CO₂ emissions by 2050 within our existing system? Only focusing on fossil CO₂ emissions would be not sufficient to cover all important aspects for the development of a sustainable energy system. Therefore, the Sustainable Development Goals of the United Nations contain further important aspects for a implementation. Especially climate action, clean energy supply, industry, innovation and infrastructure as well as access to clean water and sanitation need to be acknowledged.



Figure 1: The United Nations Sustainable Development Goals⁴

1 Rockström J., 2017

2 Energy roadmap 2050, 2012 (accessed on March 27, 2020)

3 European climate law, 2020 (accessed on March 27, 2020); 4 <https://www.un.org/en/> (accessed on August 17, 2021)

Past experiences indicate that a holistic perspective is necessary for a target-oriented implementation of new industrial plants.¹ Modelling-based approaches offer beneficial tools if all important factors are considered and assessed to an adequate extent before the implementation phase. Negative examples from the past indicate that the following important factors to protect the surrounding ecosystems had not been considered to a sufficient extent:

- additional greenhouse gas emissions,
- additional resource consumption,
- expected impact on ground water and surface water quality,
- the implied change in land use,
- how the biomass is utilized,
- the implied impact on the ecology in the surrounding areas,
- the expected impact on soil and ground conditions,
- additional production of waste materials,
- the influence of stakeholders in the surrounding areas,
- economic operation and the loss of biodiversity.^{1,2}

Therefore, an early-phase stakeholder dialog is recommended to manage the complex relations with a holistic approach. Advanced digital methods could be a supportive methodology if applied in a smart manner. So far, digital methods were not able to follow the desired holistic approach. In the past, different software environments have allowed investigation of single aspects mainly focused on physical models of energy process technology. Hence, the following examples from TU Wien indicate the used methodology that has been applied.

Pröll T. developed a simulation tool to describe biomass gasification–based processes mainly focused on chemical and thermodynamic aspects of the described processes.³ Kotik J.⁴ and Stidl M.⁵ applied the developed methodology on large-scale biomass gasification plants focused on optimizing the investigated plants. Wukovits W. used different simulation environments for a detailed description of different process units.⁶ Harasek M. was very active with respect to computational fluid dynamics, gas separation technology as well as advanced digital methods enabling important biomedical applications.⁷ Benedikt F. focused his work on up-scaling biomass gasification–based processes.⁸ Industrial plant manufactures demonstrated the capability of different software tools.⁹

Grafinger M. focused his work on virtual product development.¹⁰ Goger G. investigated the application of building information models (BIM).¹¹ Bednar T. investigated the optimization of a building with respect to energy efficiency.¹² Kranzl L. carried out his work with respect to modelling energy regions.¹³ Hammerschmid M. investigated the development of a virtual engineering space.¹⁴ Cherubini F. and Jungmeier G.¹⁵ carried out extensive work with respect to life cycle assessment (LCA). Hofmann R. proposed a systematic approach for main definitions accompanying the design and operation of industrial energy systems for a future optimized implementation.^{16,17}

1 Müller-Grabherr D. (expert interview October 28, 2020); 2 CL:AIRE, 2020; 3 Pröll T., 2008
4 Kotik J., 2010; 5 Stidl M., 2012; 6 Wukovits W., 2003 7 www.cfd.at (accessed on November 4, 2020); 8 Benedikt, F., 2020
9 <https://www.autodesk.de> (accessed on May 5, 2021); 10 Grafinger M., 2010; 11 Goger G., 2017; 12 Bednar, T. 2010
13 Kranzl L., 2005; 14 Hammerschmid M., 2019; 15 Cherubini F., 2010; 16 Hofmann R., 2020; 17 Hofmann R., 2019

Pröll T. indicated interaction of modelling and experimental facilities to generate design proposals.¹ And finally, Fei T. reviewed the state of the art of digital twins in industry. Despite extensive growth, digital twins remain a rapidly evolving concept. Many pressing issues should enhance its viability in practice and a unified digital twinning method is needed.² Overall, there is a rapid growth in interest regarding the described topic. At the same time, a major drawback of the results has been a missing unified methodology and interlink between different software tools.

1.1. Aim and Scope

The aim of the present work is to evaluate a methodology for the development of energy technology supported by advanced digital methods. The evaluation of different examples should find answers to:

- How can advanced digital methods be applied to support energy technology development?
- Which approach leads to the best preparation of novel technology before industrial application?
- How can novel processes be applied most suitably to reach the desired development goals and thereby reduce fossil CO₂ emissions?

Utilizing biomass represents an alternative to utilizing fossil carbon and follows the goals of the European Commission^{3,4} as well as the Austrian national goals^{5,6}. Figure 2 shows an overview of the investigated examples for the potential utilization of solid fuels for the production of clean energy carriers within the present work. Following examples were investigated in detail to answer the research questions above:

- Experimental development of sorption enhance reforming and CO₂ gasification.
- Industrial plant model development for the production of hydrogen (H₂) production, synthetic natural gas production, liquid biofuel production as well as an integrated concept for a hot metal plant.



Figure 2: Considered development topics for novel energy technology⁷

1 Pröll T., 2011; 2 F. Tao, 2019; 3 Energy roadmap 2050, 2012 (accessed on March 27th 2020)
 4 European climate law, 2020 (accessed on March 27, 2020)
 5 "Regierungsprogramm" Austria, 2020 (accessed on March 27, 2020)
 6 www.icps-conference.org, 2019 (accessed on March 27, 2021); 7 Müller S., 2017

1.2. Methodology

Figure 3 shows the approach used to apply accompanying digital methods at different stages of energy technology development from the process idea until the realization of a commercial plant. As can be seen, the different phases of technology development are accompanied by modelling important relations. Past examples have shown that implementing a novel process can take between 5 and 10 years. Hofbauer H. developed a dual fluidized bed gasification system.² Pröll T., Schmid J. and Hofbauer H. improved the chosen reactor design by using cold flow model investigations as well as further experimental development activities.^{3,4} Aichernig C. realized a demonstration plant at an industrial scale.⁵ Hammerschmid M. and Müller S. created digital models of performed work.^{6,7} Different plant manufacturers realized large-scale commercial plants.⁸ Increasing fuel prices and changes of the related funding framework have led to the shutdown of commercial plants and therefore the desired ecologic value could not reach full potential as well as rising expectations.

The application of a reasonable development strategy is essential with respect to successful technology implementation. Analysis of the general legal and economic framework highlights the opportunities for process development. The applied strategy should acknowledge interrelated development goals and define the final utility value of the proposed energy technology. Process development via advanced digital methods enables a consistent summary of the experimental and operational experiences. Finally, process development should lead to the expected technological, economic and ecologic results. Many examples have shown dissatisfactory results with respect to the final goal to reduce fossil CO₂ emissions. For this reason, the present work investigates how digital methods could be applied most suitably to support the desired reduction in fossil CO₂ emissions. Figure 4 illustrates the introduction of MRL to ensure target-oriented implementation of novel energy technology.

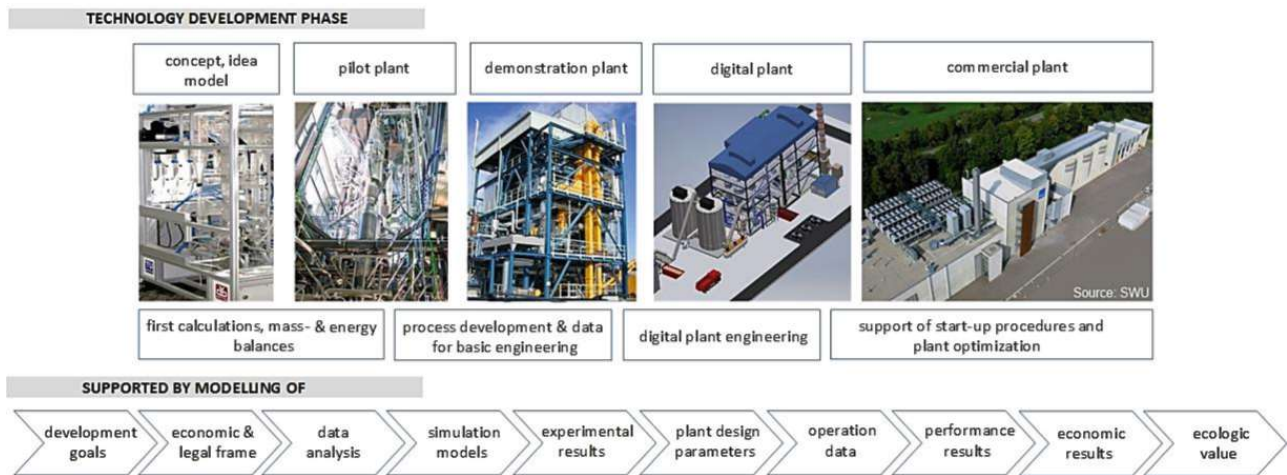


Figure 3: Methodology for the development of energy technology supported by advanced digital methods¹

1 cf. Müller S., 2013; 2 Hofbauer H., 1993; 3 Pröll T. 2004; 4 Schmid J.C., 2014; 5 Hofbauer H., 2020; 5 www.repotec.at (accessed on March 27, 2021); 6 Hammerschmid M., 2019; 7 Müller S., 2013; 8 <https://de.wikipedia.org/wiki/Zweibettwirbelschicht-Gaserzeugung> (accessed on March 27, 2021)

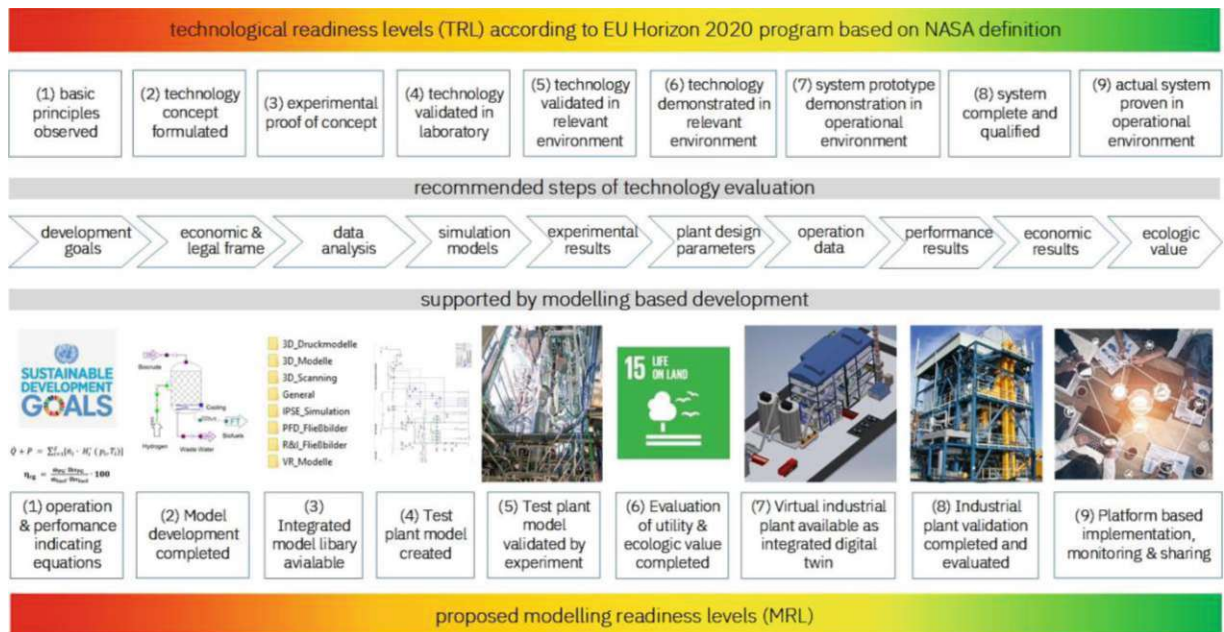


Figure 4: Introduction to modelling readiness levels ¹

As can be seen, modelling-based development by advanced digital methods should include following:

1. basic equations determining the relations of key figures with respect to defined goals;
2. the creation of a process simulation model;
3. the creation of a model library enabling integrated investigations;
4. the creation of a test plant model;
5. the validation of created models by the use of experimental test plants;
6. a holistic evaluation of the results achieved by the aid of an ecologic value;
7. the creation of an industrial plant model following a scale-up perspective;
8. the validation and evaluation of the industrial plant model; and
9. finally platform-based implementation, monitoring and sharing of the results.

It has to be mentioned that planning and development processes are based on iterative loops enabling continuous learning and improving each component at each stage. Furthermore, the proposed methodology contains several evaluation steps to ensure that the expected utility and ecologic values are reached. Within the present work, this perspective is used to analyze the modelling-based development of energy technology for the supply of clean energy carriers such as hydrogen, synthetic natural gas and liquid biofuels.

For this reason, the present work has been structured as follows. The present work starts with a general discussion of the existing background with respect to the engineering of energy technology. Then, the applied principles and methods are discussed with respect to their suitability for the development of novel energy technology. Subsequently, different case studies for the reduction of fossil carbon utilization are described. Investigated cases for the application of industrial plant information models (IPIM) are calculated and interpreted. Finally, the results are discussed to make recommendations for the implementation of the proposed methodology.

1.3. Preliminary Definitions

“Sustainable development is considered as development that meets the needs of the present without compromising the ability of future generations to meet their own needs.”¹ A sustainable energy system provides an energy supply to society without impairing possibilities for the energy supply of future generations.

Industry 5.0 is defined as a guiding concept reframing industrial revolutions from the past, including the currently postulated Industry 4.0, and follows the idea of a circular economy according to sustainable systems found in nature. Instead of increased industrial production and economic growth based on increased efficiency—for example, via digitalization, causing increased resource consumption—Industry 5.0 follows the fundamental principles of nature and therefore sustainable systems. This concept, so called “imagineering nature,” was defined as guiding principle of the Institute of Chemical, Environmental and Bioscience Engineering in 2016. The present work is inspired by this idea and represents a small piece of work supporting the described concept.²

Polygeneration aims at the production of many (poly) equally important products.³ The thermochemical conversion of solid fuels into product gas enables the production of many different products such as electricity and heat, synthetic natural gas (SNG), liquid fuels, hydrogen and different platform chemicals. The production of different products in a single plant is named polygeneration.

Digital twins are used to connect the physical and the virtual space. Digital twins include data acquisition, data communication, simulation and further advanced technologies. Therefore, digital twins are defined here as cyber-physical integration of physical and virtual data throughout a product lifecycle. The available data volume is analyzed by advanced analytics to improve the performance of a product or a process.^{4,5}

The technological readiness level (TRL) is a method for estimating the maturity of technologies during the acquisition phase, developed at the National Aeronautics and Space Administration (NASA) around 1970. This use enables consistent, uniform discussions of technical maturity across different types of technology. The European Union Horizon 2020 program adapted preliminary versions of the TRL with respect to the classification of the development status of novel technologies.⁶

The modelling readiness level (MRL) is introduced within the present work to describe the development status of virtual model in accordance with the definition of a digital twin as well as the concept of TRL.

The utility value of a solution is defined within the present work as the score reached with respect to the defined development targets. This value summarizes key performance indicators as well as other factors such as economic goals from a holistic perspective as a part of a competitive comparison with best available practice.⁷

1 cf. Brundtland G., 1987, p54; 2 www.i5-0.com, 2020 (accessed on March 27, 2021)

3 Fürnsinn S., 2007, p32; 4 Tao F., 2019; 5 cf. Hofmann R., 2020; 6 Mihaly H., 2017; 7 Haas G., 2005

The **ecologic value** of a technology, an economic sector or even an industrial plant is defined in the present work as the ability of a unit or system to protect the surrounding ecology or even support the recreation of lost ecosystems in accordance with the idea of a circular economy and sustainability.

The **validation** of data is defined within the present work as affirmation of measurement results collected during experimental or operational campaigns. The validation of a model is an affirmation of modelling assumptions.

Sorption enhanced reforming (SER) is defined as a product gas reforming process supported by absorption. Sorption enhanced reforming is applied for the reformation of a product gas from biomass gasification. Calcium based bed materials enable the selective transport of CO₂ from the product gas stream to an exhaust gas stream. Sorption enhanced reforming leads to a hydrogen-rich product gas stream and a reduced CO₂ content in the product gas stream.^{1,2}

Oxyfuel combustion uses pure oxygen mixed with exhaust gas instead of air to operate a combustion process. This way, the combustion process leads to high CO₂ contents in the exhaust gas stream and offers good characteristics for an efficient disposal. Oxyfuel combustion is currently considered as one of the most economic carbon capture processes.^{3,4}

Oxyfuel sorption enhanced reforming (OxySER) represents a combined process of sorption enhanced reforming and oxyfuel combustion. The combined process enables the production of a hydrogen-rich product gas and an exhaust gas stream mainly consisting of carbon dioxide. Hereby, the produced carbon dioxide can be described as ready for sequestration or further utilization.^{4,5}

Carbon dioxide gasification is defined in the present work as the thermos-chemical conversion of solid fuels by the action of heat and the usage of carbon dioxide as gasification agent. Hereby, the gasification agent can consist out of 100% carbon dioxide or a mixture of carbon dioxide and steam. As a result, carbon dioxide is consumed by primary and secondary gasification reactions and the received product gas contains an increase share of carbon monoxide.^{6,7}

Industrial plant information model (IPIM) is defined in the present work as an integrated model of all relevant perspectives with respect to the planning, the erection and the operation of an industrial plant. This includes a technological description of the operated process streams, the used equipment, related control and measurement data as well as economical figures and indicators with respect to environmental targets.

Power-to-X is defined in the present work as many optional pathways for the conversion, the storage and reconversion of electricity from surplus power, typically during periods where fluctuating renewable energy generation exceeds load. Hereby, the primary focus lies on the conversion in gaseous or liquid energy carriers.^{8,9}

1 cf. Soukup G., 2009; 2 Müller S., 2017b

3 cf. Höttl W., 2010; 4 Hammerschmid M., 2021; 5 Müller S., 2021,

6 cf. Mauerhofer A., 2020; 7 Mauerhofer A., 2021; 8 Drünert S., 2019; 9 Müller S., 2017a

2. ENERGY SYSTEM ANALYSIS

Large amounts of easily accessible primary energy resources have enabled the development of wealth in many parts of our world. The low-cost availability of energy has provided the foundation for economic growth, wealth and power to shape our modern society.¹ The marked increase in energy consumption has already caused negative ecologic impacts on the environment, including the ongoing climate change as well as the rapidly ongoing reduction in biodiversity. Hence, there has been a strong wish for a major change in our energy system.² In response, researchers have aimed to transform our current energy system into a system based on sustainable utilization of available resources following the idea of a circular economy.³ Prior investigations have already indicated promising concepts for the future implementation of a sustainable energy system.⁴ At the same time, the progress we have made with respect to implementing a sustainable energy system has not been able to prevent an intensification of the ongoing climate change. In 2019, the European Commission published an advanced strategic long-term vision for a climate-neutral economy in Europe.³ It consists of seven pathways aiming at a climate-neutral economy:

1. energy efficiency;
2. deployment of renewables;
3. clean, safe and connected mobility;
4. competitive industry and a circular economy;
5. infrastructure and interconnections;
6. bioeconomy and natural carbon sinks; and
7. tackling emissions with carbon capture and storage (CCS).

These pathways should lead to a reduction in fossil CO₂ emissions as shown in Figure 5.

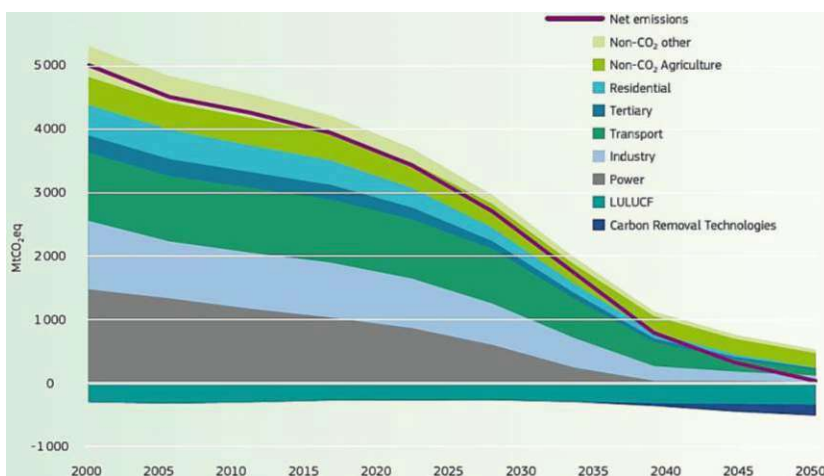


Figure 5: The trajectory of greenhouse gas emissions in the 1.5°C scenario⁴

1 cf. Nakicenovic N., 2010; 2 www.wwf.de (accessed on August 8, 2021)

3 European Commission, 2019; 4 cf. European Commission, 2003

The overall greenhouse gas emissions should be reduced significantly to limit the increase in the global mean temperature to 1.5°C compared with pre-industrial levels. The foreseen measures include climate-friendly housing, eco-design, digitalization and home automation. Furthermore, decentralized electricity production from renewables and smart energy grids should contribute to reduce fossil fuel utilization. At this point, hydrogen and power-to-x technologies are interesting concepts to be investigated. Within the mobility sector, electric and hydrogen-based zero-emission vehicles as well as biofuels are foreseen as important building blocks. A competitive industry is expected to follow the idea of a circular economy by replacing energy-intensive materials; producing new products and materials; recovering raw materials; and supplying energy based on hydrogen, biomass, SNG and electricity. The infrastructure should be based on smart transportation and energy networks. A sustainable way to use land, to protect existing ecosystems, is seen as another important building block. Finally, carbon capture and utilization (CCU) should ensure the set objectives are achieved. In the past, the gross domestic product of a country has shown a strong correlation with its CO₂ emissions. The latest results indicate that a decoupling is already taking place in Europe, China and the United States of America.¹

Whereas the European way is strongly on reducing fossil fuel consumption, the United States of America is still focusing on further fossil fuel utilization. Besides, an increased utilization of renewables is projected.² The energy supply in China has historically been based on coal. At the same time, China's energy policy is willing to support a reduction in greenhouse gas emissions, and the Chinese government announced the aim for carbon neutrality by 2060.³ As a result of the strategy in Europe, a major shift in the gross inland energy consumption is expected. Figure 6 shows the result of different scenarios until 2050. Renewables are foreseen to replace fossil gas and fossil liquids. This would mean an intensive transition phase for fuel-based energy technologies and related energy systems.

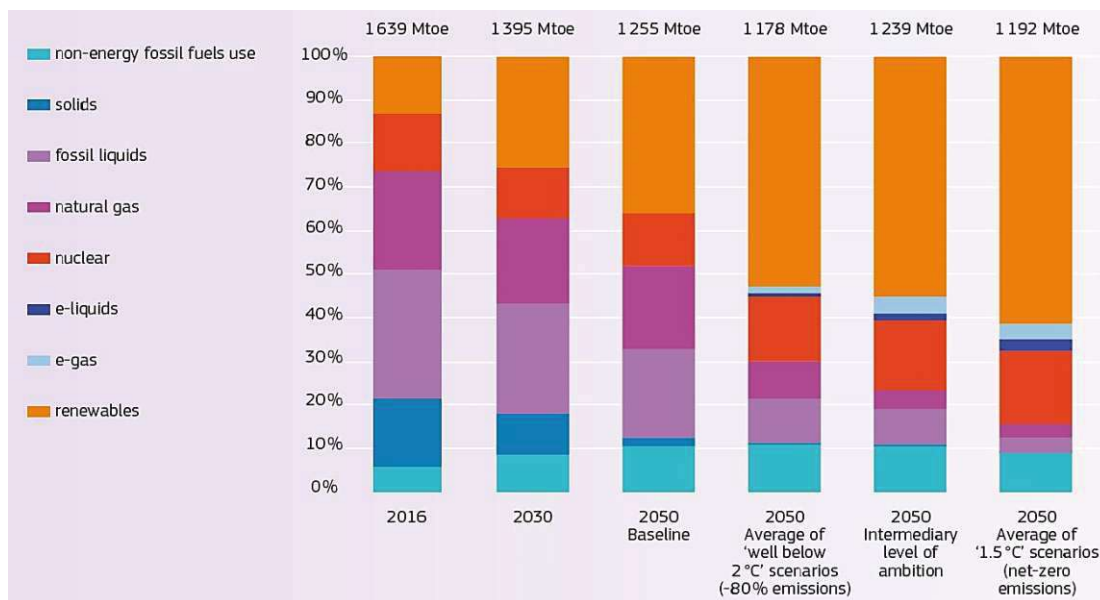


Figure 6: Gross inland consumption of energy trajectories³

1 European Commission, 2019;

2 www.eia.gov (accessed on March 27, 2021); 3 www.iea.org (accessed on March 27, 2021)

2.1. State of the Art of Process Engineering

In the past, the main focus of process engineering had been to develop efficient processes for the creation of a process or a product. The applied tools and methodology were focused on mathematical, mechanical, chemical and thermodynamic descriptions of the behavior of the process itself as well as the best possible surrounding conditions. Figure 7 shows an overview about important steps of process engineering.

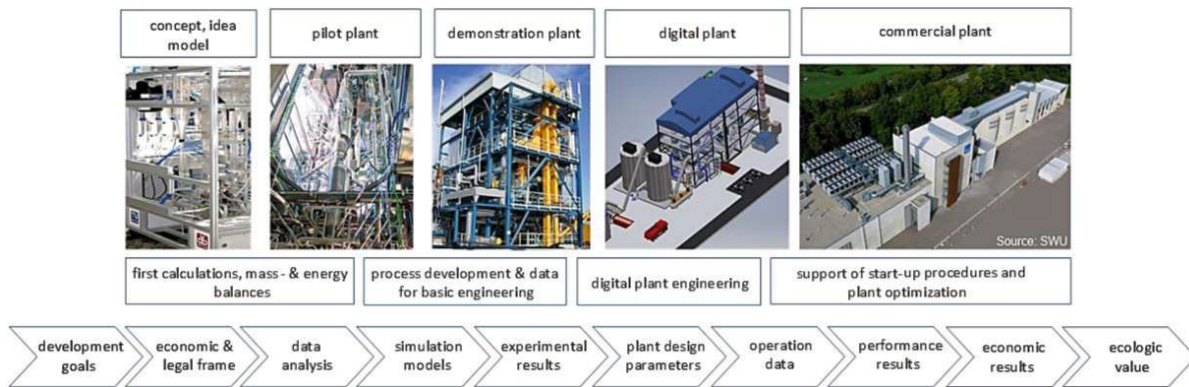


Figure 7: Important steps of process engineering and technology evaluation¹

The successful development of society based on economic growth has shifted the related targets of process development into new dimensions. Several research activities aim to improve the state of the art of process engineering in the field of energy technology. From a scientific point of view at TU Wien, Werner A. and Ponweiser K. focused their work on optimization of thermodynamic aspects of energy storage.^{2,3} Walter H. described important building blocks for the realization of an improved future energy system.⁴ Haider M. contributed impressive results with respect to optimization of conventional large-scale power plants.⁵ Hofbauer H. developed fluidized bed systems enabling efficient biogenic fuel conversion.⁶ Pröll T. developed fluidized bed systems enabling CCU during combustion processes.⁷ Wukovits W. and Harasek M. focused their work on optimizing process technology and gas separation by the used of digital methods.^{8,9} And, Grafinger M. improved the applied methodology of virtual product development.¹⁰

As a result, the development of novel processes has been based mainly on adequate preliminary modelling. This includes the formulation of a balanced target system for the development of an industrial plant. The formulation of basic equations is used to develop a representative model supported by an adequate graphic illustration. The mathematical model is calibrated by related property data to summarize all the relevant building blocks of an adequate physical model. In the past, phase diagrams and process characteristic diagrams have been used to describe the operational behavior of a system or single units. Today, advanced digital models enable dynamic and multidimensional illustration of the behavior of a system.

1 cf. Müller S., 2013, p122; 2 Werner A., 2018; 3 Ponweiser K., 1997; 4 Walter H., 2006; 5 Haider M., 2017
6 Hofbauer H., 1993; 7 Pröll T., 2011; 8 Wukovits W., 2018; 9 Harasek M., 2015; 10 Grafinger M., 2010

2.2. European Innovation Strategy

Historically, the design of our present energy system has been initiated by the representatives of our society and the resulting politics. National states have implemented public bodies and related companies that have been responsible for the erection of power plants and energy grids. The implementation of a modern energy system in Europe has enabled a reliable supply of electricity, heat, natural gas and liquid fuels. The European integration process has led to the development of an European Energy policy. Latest developments aim at reducing fossil carbon utilization to diminish greenhouse gas emissions. The accompanying legislation aims at implementing European guidelines to enable an effective implementation by the national states. At the same time, our energy system can look back at a long-lasting history of more than 100 years. This has to be acknowledged if we aim for a rapid transformation. The transition of our actual energy system will be executed by private and public bodies accompanied by European and national energy policies. Figure 8 (left) shows a vision of the future energy system by the European Commission from 2003.¹ Improving the energy infrastructure represents a critical measure. At the same time, demonstration projects should be funded to enable validation of novel energy technology. Within the NER 300 program, the European Union has funded such projects since 2012 with revenues from the emission trading system (EU ETS). NER 300 is a funding program pooling together about 2 billion euros for innovative low-carbon technology, focusing on the demonstration of environmentally safe CCS and innovative renewable energy technology on a commercial scale within the European Union. Since 2012, around 40 projects have been funded in the fields of wind power, ocean energy, sun energy as well in the field of energy management and bioenergy.² Figure 9 shows the locations of the described projects. In 2020, an innovation fund took over the responsibility of demonstration projects.³ The total budget was increased to 10 billion euros for large- and small-scale projects focusing on innovative low-carbon technologies, CCU, CCS, renewable energy generation as well as energy storage. (cf. Figure 8, right)

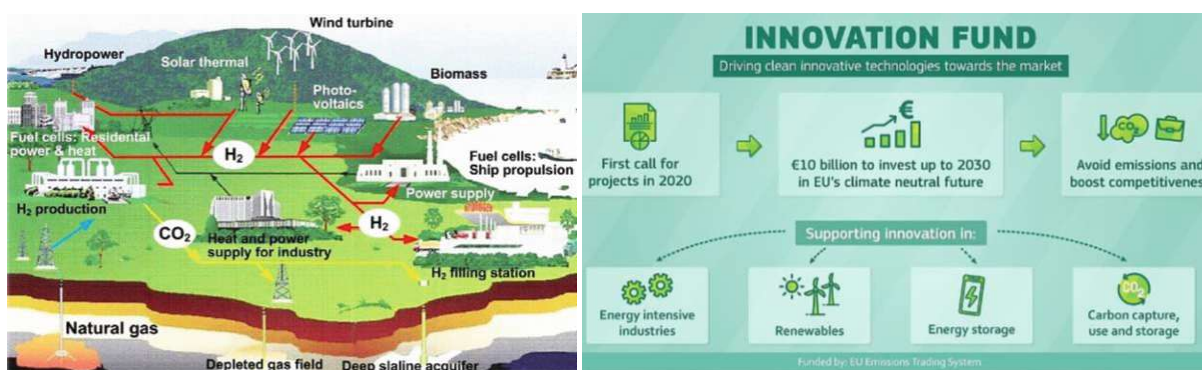


Figure 8: Vision for the future energy system based on hydrogen and fuel cells (left)¹ and the innovation fund of the European Commission supporting demonstration projects (right)³

1 European Commission, 2003, p4

2 https://ec.europa.eu/clima/policies/innovation-fund/ner300_en (accessed on March 27, 2021)

3 https://ec.europa.eu/clima/policies/innovation-fund_en#tab-0-0 (accessed on March 27, 2021)

2.3. Advanced Digital Methods

The implementation of computer systems with high processing power, high data transfer rates and related storage capacity has led to a disruptive change in our working environments. Implementation pathways have been discussed by many researches for a long time. In 1987, Scheer A.-W. described computer-integrated manufacturing (CIM), a smart interaction between physical manufacturing and the related information flow hosted by computer systems.¹ Retrospectively, he acknowledged missing performance of the implemented systems at that time. At the same time, the number of available software toolkits has increased rapidly, although in many cases interaction of single software tools had not been foreseen. [Table 1](#) gives a short overview of the available software tools for the development and engineering of industrial plants in the context of a virtual planning space (summarized by Hammerschmid M. in 2019).

[Table 1](#): Software tools for the realization of a virtual planning space²

| Product | Supplier | Application | Product | Supplier | Application |
|-------------------|---------------------|-------------------------|-----------------|---------------|-----------------|
| Dropbox | Dropbox Inc. | Cloud system | AutoCAD | Autodesk | Virtual Reality |
| Microsoft Teams | Microsoft | Communication platform | Autodesk ReCap | Autodesk | Virtual Reality |
| Microsoft Word | Microsoft | Word processor | Revit | Autodesk | Virtual Reality |
| Microsoft Excel | Microsoft | Spreadsheet calculation | FARO SCENE | FARO | Virtual Reality |
| Microsoft Visio | Microsoft | Diagramming application | As-Built | FARO | Virtual Reality |
| Acrobat Reader DC | Adobe Systems | Document management | 3ds Max | Autodesk | Virtual Reality |
| Autodesk Inventor | Autodesk | Computer added design | Corona Renderer | Render Legion | Virtual Reality |
| HSC Chemistry | Outotec | Property database | Stingray | Autodesk | Virtual Reality |
| FactSage | GTT-Technologies | Property database | V-Ray | Chaos Group | Virtual Reality |
| IPSEpro | SimTech GmbH | Mass & energy balances | VRED Pro | Autodesk | Virtual reality |
| SysCAD | KWA Kenwalt | Mass & energy balances | Autodesk Netfab | Autodesk | 3D printing |
| Barracuda | CPFD Software LLC | CFD simulation tool | Cura | Ultimaker | 3D printing |
| e!Sankey4 | ifu Hamburg | Sankey diagram editor | Azure | Microsoft | Cloud Computer |
| Python | Software Foundation | Programming language | | | |

There are myriad software tools available to enable a virtual planning space. At this point, each software tool depends on a suitable hardware and software environment. Of particular importance, the application of virtual reality depends on high-performance graphic boards. Therefore, the chosen system architecture needs to fulfill the expectations of the software tools as well as the foreseen user group. Hereby, high-performance computer systems enable novel approaches for the implementation of advanced digital methods. [Figure 10](#) shows a simplified concept for a system architecture enabling the development of novel energy technologies. As can be seen, a central development space provides important information about existing plants as well as the latest measurement data from energy technology development.

¹ Scheer A.-W., 1987

² Hammerschmid M., 2019

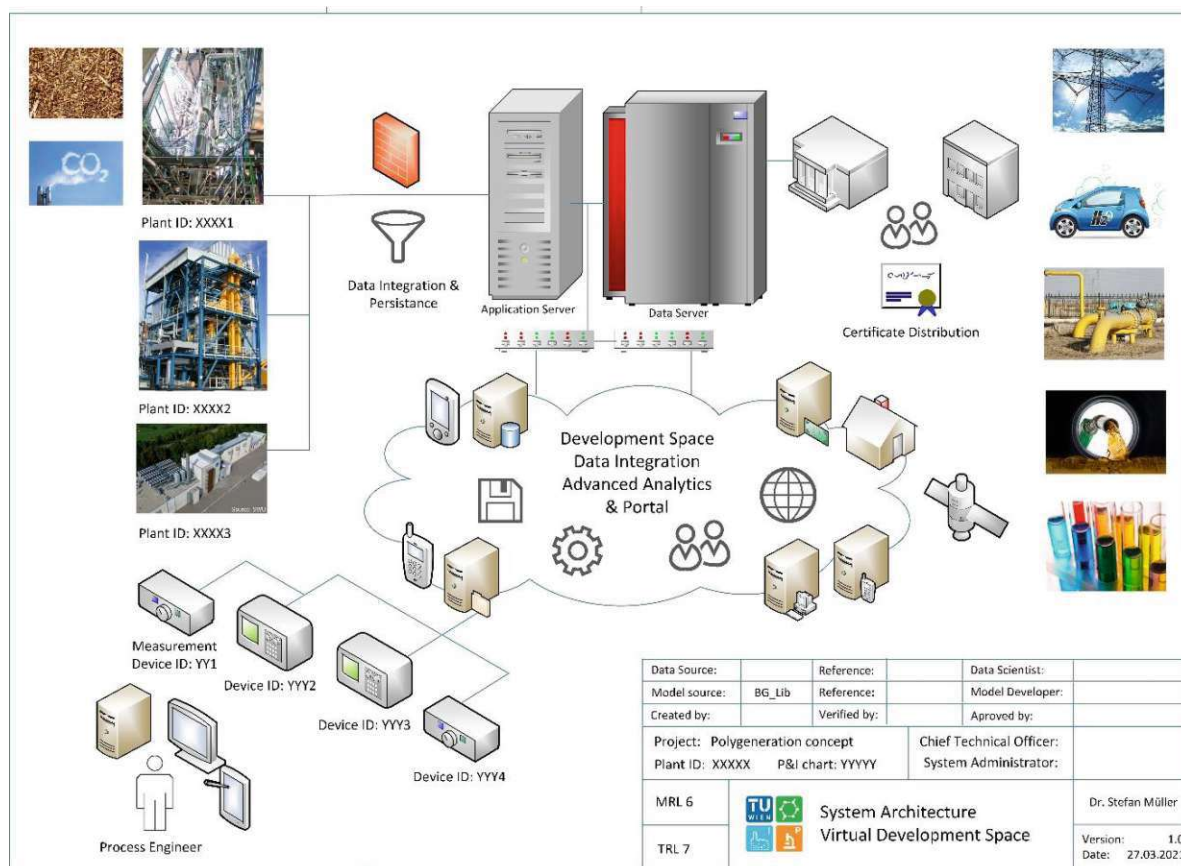


Figure 10: Concept for the implementation of advanced digital methods in the context of energy technology development

The shown concept has been implemented within the research area fuel and energy system engineering (166-7) and should enable the best possible pre-condition for the development of digital role models to implement best practice energy technology. The development space provides ideal conditions for smart, target-oriented development of building blocks for the transition of our existing energy system. Hereby, smart means the supply of correct information at the right time and the right location. Finally, the described environment and related software tools should enable ideal conditions for computer integrated manufacturing (CIM) of system models to reduce fossil CO₂ emissions.

The presented concept is only a small building block for the operation of a small-scale laboratory. In the future, high-level implementation of the next generation of data infrastructure in Europe is foreseen under GAIA-X,¹ a project for the development of an efficient and competitive, secure and trustworthy federation of data infrastructure and service providers for Europe. It is supported by representatives of business, science and administration from Germany and France, together with other European partners.² Therefore, it represents an important European initiative for the creation of a reliable data infrastructure.

1 <https://www.data-infrastructure.eu/GAIA-X/Navigation/EN/Home/home.html> (accessed on March 27, 2021)

2 <https://www.bmwi.de/Redaktion/EN/Dossier/gaia-x.html> (accessed on August 18, 2021)

2.4. Summary

The present work investigates the best possible solutions for modelling-based development of novel energy technology enabling the reduction of fossil CO₂ emissions. Therefore, [Chapter 2](#) has provided a concise overview of the major trends aiming at the transformation of the current energy system in Europe. The overview identified important aspects with respect to the design of novel processes enabling the desired transformation of our existing energy systems. The most important aspects are summarized below.

- The current energy policy in Europe is aiming at a full replacement of fossil carbon utilization mainly based on natural gas and crude oil.
- In the past, process engineering had been focused on optimizing the efficiency of single devices and did not acknowledge a holistic perspective following integrated perspectives such as the idea of the United Nations Sustainable Development Goals. At this point, an integrated perspective should acknowledge technological, economic and ecologic aspects of implemented solutions. Therefore, within the present work the [ecologic](#) and [utility values](#) are introduced.
- An integrated European energy policy has not yet resulted in implementation of a sustainable energy system. There has been increased effort to reach a climate-neutral energy system in Europe. The funding of demonstration project from revenues of the European Emission Trading system has also increased. Overall, the actions that have been taken seem to be insufficient to reach the ambitious goals. Adequate modelling before implementation of novel energy technology could improve the impact of actions taken. Therefore, a proposal for an improved methodology is introduced within the present work. The proposed methodology recommends a clear definition of development targets at the beginning as well iterative assessment of development results by the use of an [MRL](#).
- Advanced digital methods offer a powerful tool to support the described actions. Advanced digital methods include the interaction of physical models with databases and an effective data infrastructure. Digital twins could be a powerful tool to accelerate the implementation of successful demonstration projects if reliable results are shared in professional manner. Therefore, a concept for the [system architecture](#) of a [virtual development space](#) has been presented.

Overall, advanced digital methods could be a powerful solution for effective transformation of the energy system in Europe if implemented in a smart manner. [Integrated development targets need to be defined at the beginning of a development process. The introduced ecologic value offers great potential to moderate target conflicts during the development process and could ensure the best possible utilization of deployed resources.](#) The following chapter contains a proposal for a unified method to support an interactive development process across the borders of single organization units.

3. PROPOSAL FOR IMPLEMENTING MODELLING READINESS LEVELS

Based on preliminary work, the interest in advanced digital methods has grown rapidly. At the same time, a missing unified methodology has hindered the potential impact of advanced digital methods.¹ Smart industrial energy systems hereby include data handling treatment, design optimization, operational optimization as well as sector coupling.² Within the present work, different practical examples are investigated to optimize the applied principles and methods during the technology development phase. This chapter describes important aspects during the different stages of model development. Figure 11 presents the approach used by introducing MRL. The modelling procedure starts by defining development goals summarized by important equations defining the foreseen operation as well as the expected performance of a novel industrial plant. Well-defined development goals are critical before beginning the development process. Otherwise, the development involves the risk of final implementation failure. The following sections describe each level of development, highlighting important aspects of each phase.

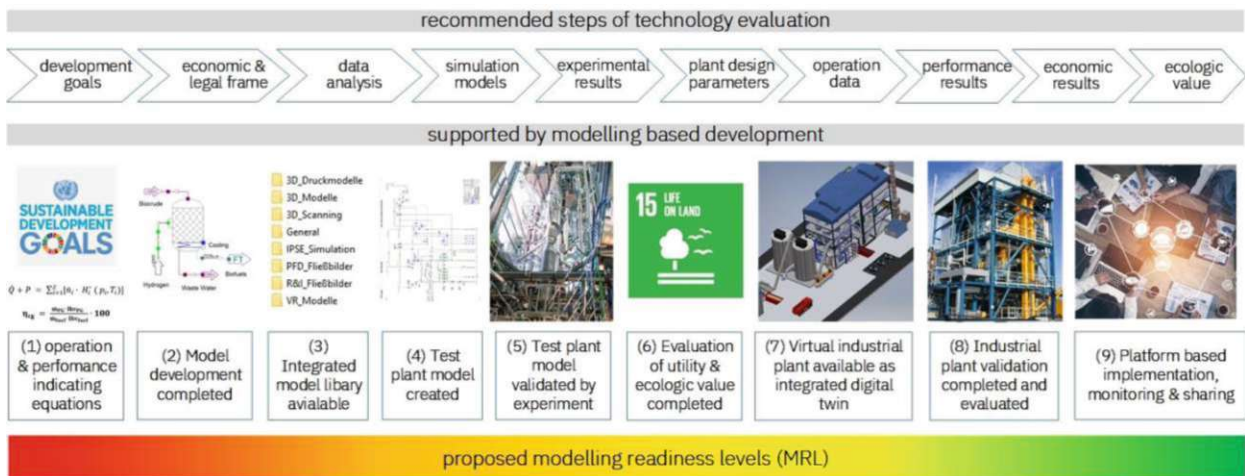


Figure 11: Advanced digital methods application concept by the use of modelling readiness levels

3.1. Development Goals and Targets

The United Nation Sustainable Development Goals defines important pre-conditions for the realization of future projects. These goals can be summarized as a collection of 17 interlinked goals designed to be a “blueprint to achieve a better and more sustainable future for all.”³

1 Tao F., 2019

2 Hofmann R., 2019; 3 United Nations, 2017

In the context of this project, goal number 7 aims at affordable and clean energy supply whereas goal number 13 relates to climate action activities. At the beginning of project development, it is essential to define specific targets that should be reached during the project lifetime. Besides, it is important to acknowledge the holistic perspective of the investigated project as well as local circumstances and interests of relevant local stakeholders. The execution of a utility analysis (German: Nutzwertanalyse) or the application of quality function deployment (QFD) represent an important step to define specific project targets and to moderate conflicts of targets at the beginning of a development phase.

A utility analysis is a valuable methodology for systematic decision making process. Different outcomes and solutions are evaluated by quantified decision and development criteria. Using a utility analysis enables a holistic perspective during a decision-making process.¹ Table 2 shows an example for the execution of a utility analysis. First, the decision criteria are defined. Second, the defined criteria are weighted following the importance with respect to the final utility value. Different decision criteria can be weighted—for example, by a conjoint-analysis.² Subsequently, each dimension is valued based on available data or by a moderated expert discussion. At this point, although most decision-making processes face missing information basis, information-based decision-making should be preferred. Therefore, enough time should be spent on preliminary data assessment and market research. At the end, a final utility score of each solution can be calculated on a valid basis. This score represents the expected benefit. This example shows that erection of a pipeline reaches a higher score due to the high weighting of the TRL and the reached security of the supply. A major shift in the energy policy within Europe has led to greater weighting of CO₂ emissions and environmental aspects for the financing of projects in future. This factor has impaired important pre-conditions for the investigated topic within the present work.

Table 2: Example of a utility analysis¹

Which energy technology supply solution option best fulfills the expectation?

| Decision criteria weighted between 0 and 20, valued between 0 and 10 | | Option A: Natural gas via the Nord Stream pipeline | | Option B: Erection of an SNG production plant | |
|----------------------------------------------------------------------|---------------|----------------------------------------------------|---------------|-----------------------------------------------|---------------|
| Decision criteria | Weighting (w) | Value (v) | Score (w × v) | Value (v) | Score (w × v) |
| Technological readiness level | 20 | 10 | 200 | 7 | 140 |
| Security of supply | 20 | 10 | 200 | 7 | 140 |
| Economic competitiveness | 15 | 9 | 135 | 7 | 105 |
| Efficiency | 15 | 8 | 120 | 8 | 120 |
| Land use/footprint | 10 | 5 | 50 | 10 | 100 |
| Water consumption | 10 | 8 | 80 | 5 | 50 |
| Carbon utilization | 10 | 5 | 50 | 9 | 90 |
| CO ₂ emission factor | 10 | 5 | 50 | 8 | 80 |
| Modelling readiness level | 10 | 10 | 100 | 8 | 80 |
| Sum weighting | 120 | Evaluation result | 985 | Utility value | 905 |
| Maximum score | 1 200 | | 82.1% | Utility rate | 75.4% |

Option B has a higher utility score and therefore better contributes to the development targets!

1 Haas G., 2005

2 Pataky, 2007

QFD is another powerful tool to define a suitable development strategy. The aim is to understand the needs and desires of the final customer. In the present case, general society can be seen as the final customer if the idea of the United Nations Sustainable Development Goals is followed. Therefore, the primary goals of this method are a specified concept, creation and sale of a product and its related services to meet the society's needs. Hereby, QFD involves all important dimensions according to responsibility for quality.¹ The assessment is carried out in a suitable matrix by an expert team. Furthermore, an evaluation and ranking according to most important aspects and facts is carried out. The described methodology has been used by many entities—for example, by Toyota to improve the results of product development.^{1,2}

Figure 12 shows the results of a QFD analysis of a car rear-view mirror. First, customer needs are evaluated and rated. Next, existing products are analyzed with respect to the satisfaction of customer needs to determine the position of relevant competitors. At this point, extensive market and literature research—part of a technical due diligence—is recommended to determine the position of the best available technology. Then, technical measures are suggested in the roof section to improve the analyzed situation. The interaction of different technical measures is reflected and weighted with respect to customer needs and satisfaction. As a result, specific targets for the subsequent development steps are defined. In the presented case, optimization of the design, stiffness, water resistance, deicing properties and adjustment speed of the car rear-view mirror have been recommended.

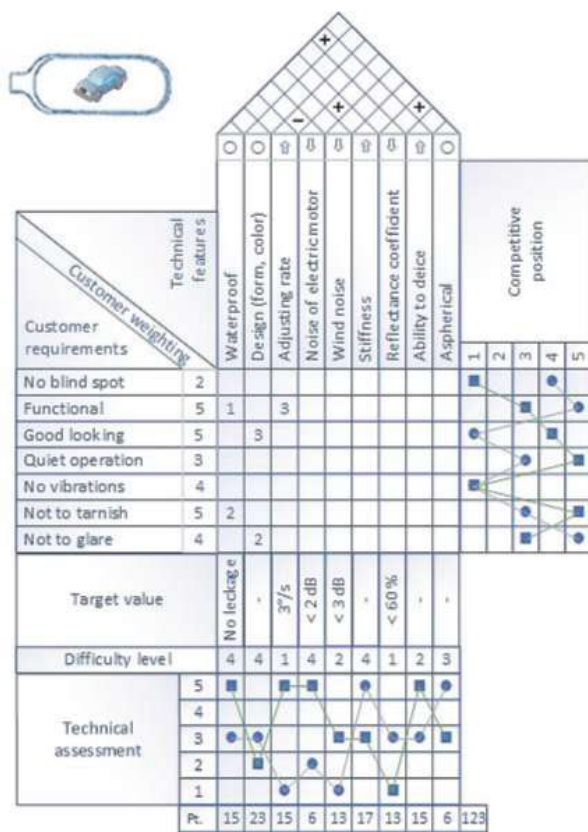


Figure 12: An example of quality function deployment of a car rear-view mirror¹

1 Pfeiffer T., 1993

2 Schmidt R., 2002

QFD could be an important tool for the definition of technology development targets regarding energy technology. Recently, within the present research topic it has been applied to develop a fuel flexible gasification unit¹ and to develop a methanation unit for the production of SNG.^{2,3} The main conclusions were a strategic focus on fuel flexibility, measurement methods, gas cleaning equipment as well as the used catalysts. Figure 13 shows the results of QFD analysis aiming at an optimized biomass gasification system for the supply of product gas for SNG production. As can be seen, an optimization strategy has been found, and the following development targets were identified:

- improve control and automation principles,
- utilize cost-effective fuels,
- improve gas cleaning strategy,
- improve operational management and
- and fuel pre-treatment.

The QFD results enable the definition of necessary research activities. The research project ReGas 4Industry investigates the utilization of cost-effective fuels and the improvement of fuel pre-treatment and gas cleaning strategy.⁴ The research project ADOReE-SNG encompasses improvement of control and automation principles as well as the operational management.⁵ The QFD results provide confidence that the defined project targets represent well-chosen development steps.

The illustrated examples indicate the importance of defining adequate targets before the development of an industrial plant. High costs of modifications and changes at later stages could be eliminated and novel solutions could be implemented smartly. The latest policies of the United Nations and the European Commission have led to an initial situation where the following targets are rated with higher priority than in the past:

- reduce fossil carbon utilization,
- ensure economic competitiveness and
- protect important environmental indicators.

Within the present work, the achieved results are evaluated according to the operation performance, the economic results as well as environmental aspects summarized as the ecologic value.

1 Schmid J.C., 2014

2 Walcher C., 2018

3 Bartik A., 2020; 4 Bartik A., 2021a

5 <https://projekte.ffg.at/projekt/3862075> (accessed on August 18, 2021)

| Technical characteristics | | | | | | | | | | | | | | | | | | | |
|----------------------------------------------------------------------------------------------------|----|--------|------|---------------------------|----------------------------------------|---------------------------------------------------|---------------------|----------------------------------------------------|---------------------|-----------------------|------------------------|-------------------------------------------|------------------|-----------------|-----------------------------------------|-----------------------------------------------|---|---------------------|--|
| Safety technology | | | | | | | | | | | | | | | | | | | |
| Measuring points, measuring technology | | + | | | | | | | | | | | | | | | | | |
| Control room, controlling, regulation, automation | | + | + | | | | | | | | | | | | | | | | |
| Cost-effective fuel | | | | + | + | | | | | | | | | | | | | | |
| Product gas cleaning and preparation for synthesis | | | | + | + | + | - | | | | | | | | | | | | |
| Maintenance options | | + | + | + | - | + | | | | | | | | | | | | | |
| Design and appearance | | | | | | | | - | | | | | | | | | | | |
| Operational management | | | | + | + | - | | | | | | | | | | | | | |
| Equipment specification by target costing | | - | - | - | - | - | - | - | | | | | | | | | | | |
| Legal frame work | | | | | | | | | | | | | | | | | | | |
| Public relation | | + | | | + | + | + | + | | | | | | | | | | | |
| Simple plant concept, conceptual design | | - | - | + | - | - | + | - | + | + | + | + | | | | | | | |
| Fuel pretreatment controlled by fuel analysis | | | | + | + | - | + | + | - | + | + | + | | | | | | | |
| Requirements Nr.: | | Rating | | Technical characteristics | | | | | | | | | | | | Rating of existing plants | | Competitors ability | |
| | | | | Safety technology | Measuring points, measuring technology | Control room, controlling, regulation, automation | Cost-effective fuel | Product gas cleaning and preparation for synthesis | Maintenance options | Design and appearance | Operational management | Equipment specification by target costing | Legal frame work | Public relation | Simple plant concept, conceptual design | Fuel pretreatment controlled by fuel analysis | | | |
| Suitability for SNG production | 1 | 5 | | 2 | 3 | 3 | 3 | 3 | | 3 | 1 | 3 | | 1 | 2 | 3 | 2 | | |
| Economic operation | 2 | 5 | | 2 | 2 | 3 | 2 | 3 | | 3 | 3 | 3 | | 3 | 3 | 1 | 4 | | |
| Comply with official directives | 3 | 5 | | 2 | 3 | 3 | 3 | 3 | 1 | 2 | 1 | 3 | | | 1 | 1 | 4 | | |
| Operational safety | 4 | 5 | 3 | 3 | 3 | | | 3 | | 3 | 2 | | | 2 | 1 | 3 | 2 | | |
| Positive image | 5 | 5 | 1 | 1 | 2 | 3 | 3 | | 3 | | 1 | | 3 | 1 | | 1 | 4 | | |
| Fuel flexibility | 6 | 5 | | 1 | 2 | 3 | 3 | | | | | 3 | 2 | 1 | 3 | 1 | 4 | | |
| High uptime | 7 | 4 | | 2 | 3 | 1 | 3 | 3 | 1 | 3 | 3 | 2 | | 2 | 1 | 2 | 2 | | |
| High efficiency | 8 | 4 | | 1 | 1 | 3 | 2 | | | 3 | | | | 1 | 3 | 3 | 1 | | |
| Long life span | 9 | 3 | 1 | 2 | 3 | 3 | 2 | 3 | 1 | 1 | 3 | | | 2 | 1 | 3 | 0 | | |
| Easy operation | 10 | 4 | | 3 | 3 | | | 2 | | 3 | | | | 1 | 3 | 2 | 2 | | |
| Low maintenance costs | 11 | 3 | 1 | 2 | 2 | 2 | 2 | 3 | 3 | | 2 | | | 2 | 3 | 1 | 2 | | |
| Product protection | 12 | 3 | | | | | | | 1 | | | | 3 | 3 | 1 | | 2 | | |
| Targets | | | | | | | | | | | | | | | | | | | |
| Industry standard, compliance with legal requirements | | | | | | | | | | | | | | | | | | | |
| Monitoring of significant operating variables to ensure efficiency and operating hours | | | | | | | | | | | | | | | | | | | |
| Minus 50% operating costs | | | | | | | | | | | | | | | | | | | |
| Minus 50% fuel costs | | | | | | | | | | | | | | | | | | | |
| Grid feeding guidelines, requirements foster wheeler | | | | | | | | | | | | | | | | | | | |
| Low maintenance needs, reduction of operational media | | | | | | | | | | | | | | | | | | | |
| Simple constructions, acceptance of the population, increase of the market value of the technology | | | | | | | | | | | | | | | | | | | |
| Efficiency increase, operational media reduction | | | | | | | | | | | | | | | | | | | |
| Minus 50% investment costs | | | | | | | | | | | | | | | | | | | |
| Total protection of company-internal data | | | | | | | | | | | | | | | | | | | |
| Acceptance of local residents and authorities | | | | | | | | | | | | | | | | | | | |
| Avoidance of feedback effects, complexity reduction | | | | | | | | | | | | | | | | | | | |
| Increasing fuel flexibility | | | | | | | | | | | | | | | | | | | |
| Difficulty | | 3 | 3 | 5 | 4 | 2 | 3 | 5 | 3 | 5 | 4 | 3 | 3 | 3 | 4 | | | | |
| Technical evaluation of existing plants | | 2 | 2 | 2 | 1 | 3 | 2 | 3 | 2 | 1 | 1 | 1 | 1 | 2 | 3 | | | | |
| Technical evaluation of future plants | | 3 | 3 | 4 | 4 | 5 | 3 | 5 | 3 | 4 | 4 | 4 | 4 | 4 | 4 | Sum | | | |
| Evaluation points | | 26 | 81 | 113 | 106 | 102 | 68 | 39 | 94 | 67 | 77 | 34 | 71 | 90 | 968 | | | | |
| % | | 2,7% | 8,4% | 11,7% | 11,0% | 10,5% | 7,0% | 4,0% | 9,7% | 6,9% | 8,0% | 3,5% | 7,3% | 9,3% | 100% | | | | |
| | | | | Safety technology | Measuring points, measuring technology | Control room, controlling, regulation, automation | Cost-effective fuel | Product gas cleaning and preparation for synthesis | Maintenance options | Design and appearance | Operational management | Equipment specification by target costing | Legal frame work | Public relation | Simple plant concept, conceptual design | Fuel pretreatment controlled by fuel analysis | | | |

Figure 13: An example of quality function deployment aiming at an optimized biomass gasification system¹

¹ Walcher C., 2018

3.2. Model Development

The model development of a novel process or a novel plant is based on the definition of specific targets. The modelling steps can be focused on defined target dimensions. A model can be defined as a simplified reproduction of reality focused on investigating important existing relations and observed behavior of experimental facilities or commercial plants. Modelling important relations by mathematical equations enables a predictive calculation of future behavior with respect to existing boundary conditions. Modelling can be described as the creation of a model based on an existing system of important observed relations.¹ Process simulation predicts the behavior of a process, its basic process chemistry and relevant thermodynamic relations, by calculating the results of a mathematical model describing the process by the use of a computer. Therefore, necessary simulation model units need to be implemented in simulation software. Simulation of processes in the field of energy technologies are typically based on a description of physical or chemical interactions of modelled mass- and energy flows.² Energy balance calculations are based on total enthalpy as shown by Equation 1.² Figures 14 and 15 illustrate an example for the creation of a simulation model unit. The calculation of mass and energy balances for different operation points enables validation of measurement data and the calculation of key performance indicators (KPI). The efficiency, the hydrogen output due to water conversion based on the dominating reactions as well as the CO₂ transport within the investigated system have received emphasis in the present work. Equations 2–4 show how the definitions are used to describe these target dimensions to discuss the experimental results. A detailed description of each term can be found in the literature.^{3,4} The overall cold gas efficiency (η_g) is a key figure for the energetic efficiency of the investigated gasifier system. The steam-related water conversion rate (X_{H_2O}) describes the share of water that is consumed by the gasification reactions. The CO₂ load (X_{CaO}) describes the specific transport of CO₂ from the gasification reactor to the combustion reactor by circulating bed material. The chosen mathematical model is based on a classical perspective for the evaluation of thermal power plants. Equations 5 and 6 describe important relations within a thermal power plant.²

$$\text{Eq. 1} \quad \dot{Q} + P = \sum_{i=1}^Z [\dot{n}_i \cdot H_i^*(p_i, T_i)] \quad [\text{W}]$$

$$\text{Eq. 2} \quad \eta_g = \frac{\dot{m}_{PG} \cdot \text{lhv}_{PG}}{\dot{m}_{fuel} \cdot \text{lhv}_{fuel} + \dot{m}_{add fuel} \cdot \text{lhv}_{add fuel}} \cdot 100 \quad [-]$$

$$\text{Eq. 3} \quad X_{H_2O} = \frac{\dot{m}_{steam} + \dot{m}_{fuel} \cdot w_{H_2O, fuel} - \dot{m}_{PG} \cdot w_{H_2O, PG}}{\dot{m}_{fluid} \cdot w_{H_2O, fluid} + \dot{m}_{fuel} \cdot w_{H_2O, fuel}} \quad \left[\frac{\text{kg}_{H_2O}}{\text{kg}_{H_2O}} \right]$$

$$\text{Eq. 4} \quad X_{CaO} = \frac{\frac{\dot{m}_{CO_2, capt}}{M_{CO_2}}}{\dot{m}_{bed, G, in} \cdot \left(\frac{w_{CaO, bed, G, in}}{M_{CaO}} + \frac{w_{CaCO_3, bed, G, in}}{M_{CaCO_3}} \right)} \quad \left[\frac{\text{mol}_{CO_2}}{\text{mol}_{CaO}} \right]$$

$$\text{Eq. 5} \quad P_{th} = \text{lhv}_{fuel} \cdot \dot{m}_{fuel} \quad [\text{W}]$$

$$\text{Eq. 6} \quad P_{th} = P_{SNG} + P_{FT} + P_{H_2} + P_{el} + \dot{Q} + \dot{Q}_{loss} \quad [\text{W}]$$

1 VDI 1993; 2 Pröll T., 2008; 3 Müller S., 2013; 4 Jentsch R., 2015

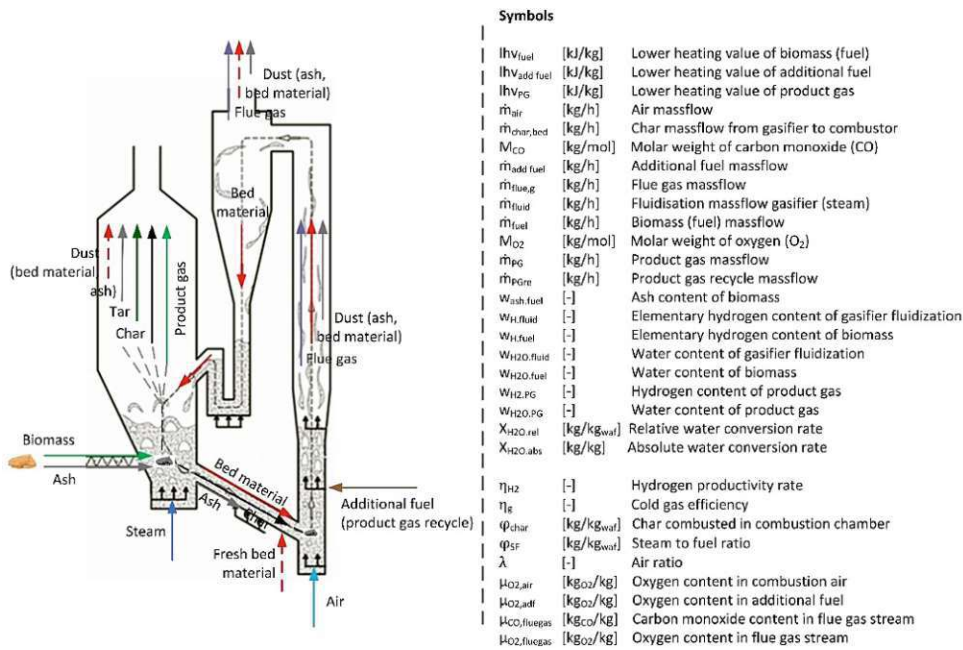


Figure 14: Modelling concept for the calculation of the mass and energy balances within a dual fluidized bed system^{1,2}

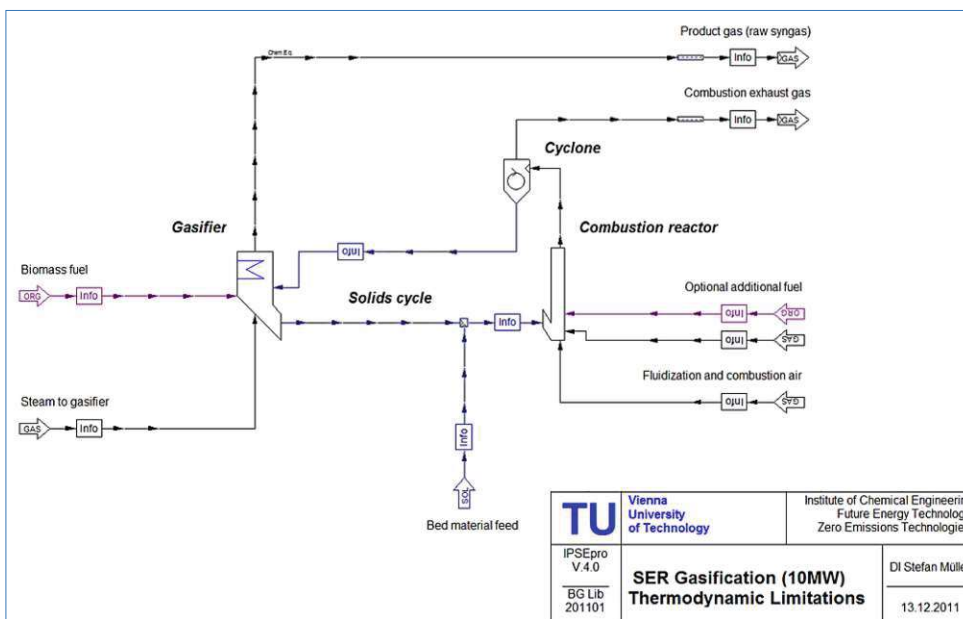


Figure 15: Completed simulation model unit of a dual fluidized bed gasification system³

The created simulation model serves as the basis for additional process development as well as plant realization. Other examples and detailed descriptions of the development of simulation models can be found in the literature.⁴ Each simulation model forms an important building block for all subsequent steps of process development. Therefore, a strategy for a reflected structure and management of the quality of all gained simulation models is necessary.

1 Müller S., 2016; 2 Müller S., 2013; 3 Müller S., 2012; 4 Ebner J., 2018

3.3. Management of Integrated Model Libraries

Past work has led to development of several models, and effective monitoring of the quality of the generated is necessary to ensure the reliability of the generated virtual development space. Modern approaches to quality management systems are used to ensure the quality of the final product, service as well as the quality of the providing organization.¹ The approach used in the present work can be summarized as follows. The development of model libraries within different software environments at the application server is continuously improved by a quality management system. This includes:

1. definition of development goals,
2. documentation of the model development process,
3. model testing and approval,
4. model structuring by the aid of model libraries and
5. continuous improvement of model libraries,

as well as related model developer, model and application user, data scientist, data citizen and system administrator following the idea of a learning system. This approach has led to the development of several model libraries providing a powerful virtual development space. Figure 16 is an overview of the current model library at the data server as well as the related applications of the application server. In the past, IPSEpro has acted as central software tool to create physical models of novel processes. Different models within the biomass gasification library (BG_Lib) and the Fischer–Tropsch library (FT_Lib) have been validated through experimental results and operating data from industrial plants like Güssing,² Oberwart,^{3,4} Senden and Göteborg.⁵ This approach has improved the quality of predictive calculations of new concepts at an early stage of development. Related examples can be found in the literature. In 2004, Pröll T. calculated several different concepts to improve electricity and heat generation.⁴ In 2012, Stidl M. modelled several concepts to supply energy from biomass gasification for the pulp and paper industry.⁵ In 2013, Müller S. provided a precise description for the documentation and management of industrial plant models.⁶ These endeavors have been accompanied by scientific documentation in the form of different types of final theses. Furthermore, the results serve as a basis for a comprehensive database.

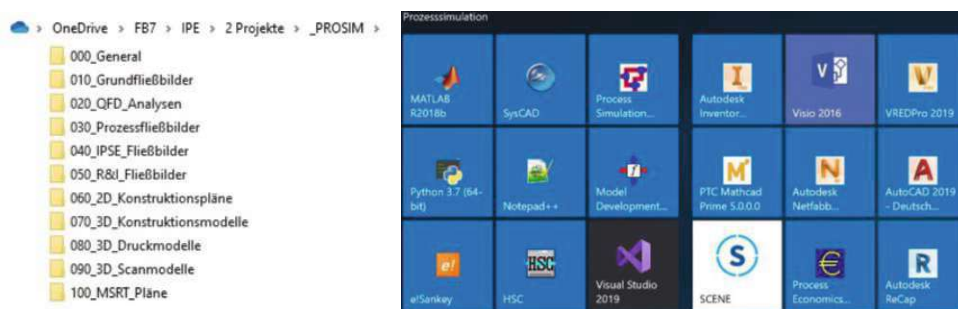


Figure 16: Integrated model library of the data server (left) and main applications of the application server (right)

1 ISO 9001, 2015; 2 Pröll T., 2004; 3 Stidl M., 2012; 4 Kotik J., 2010; 5 Veress M., 2020; 6 Müller S., 2013

3.4. Creation of a Test Plant Model

The aim of creating a test plant model is to provide preliminary calculations of expected results as well as analysis of necessary measurement data. The creation of a test plant model itself enables effective discussion of a conceptual process at an early stage of development. It allows analysis of whether expected development targets could be reached and allows a target-oriented calculation of design values for the realization of a test plant. Therefore, this development step offers a valuable contribution before incurring the high costs of test plant realization and operation. A central role for the test plant model lies within a defined research question: Which foreseen improvements lead to a best possible result for the operation of a novel energy cycle technology with respect to operation efficiency, economic circumstances as well as its contribution to the ecologic value for the surrounding?

In 2012, the existing knowledge with respect to dual fluidized bed steam gasification was summarized as the basis to design an optimized approach aiming at increased fuel flexibility and improvements for the application of sorption enhanced reforming (SER). The state of the knowledge was used to design a pilot plant to collect experimental data. Thereby, the test plant design should provide valid information enabling further realization steps at an industrial scale. Figures 17 and 18 show the resulting process simulation models for a 100 kW gasification test plant at TU Wien. It was created in 2012 during the design phase of the pilot plant and allowed preliminary analysis of necessary measurement data to achieve the set goals. Furthermore, the model was used to provide detailed calculations of design values before the engineering, procurement and construction of the test plant.^{1,2} The resulting model provided precise data with respect to the expected behavior of the test plant and formed the basis for a validation of results achieved.

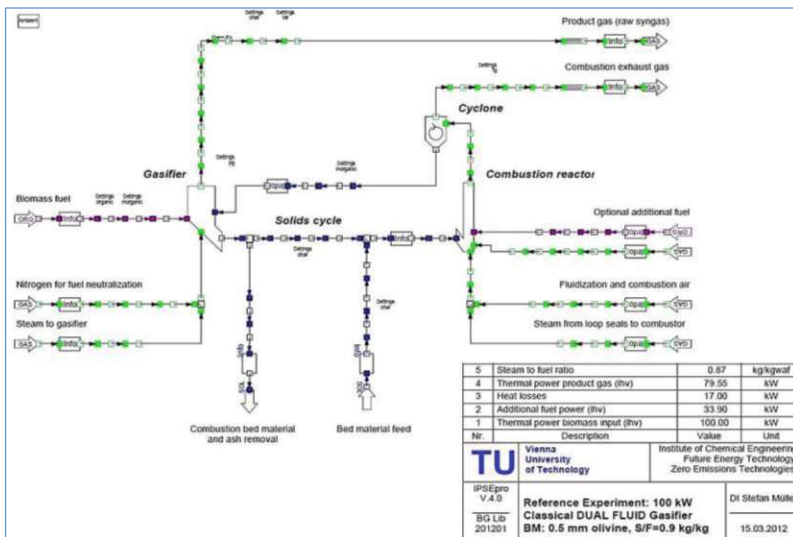


Figure 17: Process simulation model of the 100 kW gasification unit at TU Wien^{3,4}

1 Müller S., 2012a; 2 Schmid J.C., 2012a; 3 cf. Müller S., 2016; 4 Müller S., 2017b

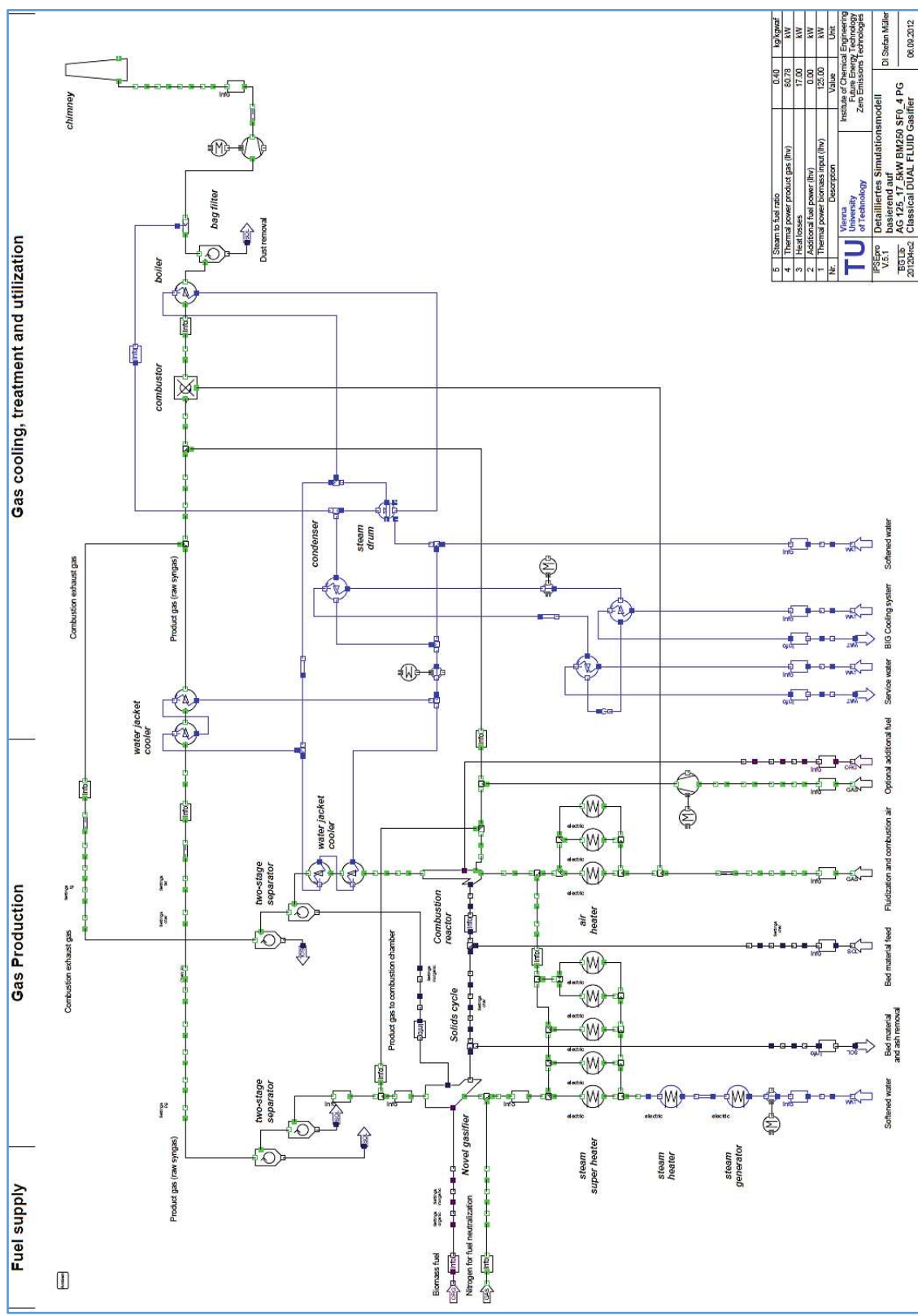


Figure 18: Process simulation model of the 100 kW gasification test plant at TU Wien ^{1,2}

1 cf. Müller S., 2016; 2 Müller S., 2017b

3.5. Experimental Validation by Test Plants

The experimental validation of a novel process idea is essential to implement a novel process concept. Therefore, an experimental facility is necessary to demonstrate the basic chemical reaction, the related thermodynamics and the operation efficiency under real conditions. The experimental facility requires the appropriate equipment to ensure precise measurement results. Figure 19 is a picture of a dual fluidized bed gasification test plant at TU Wien, which was built between 2012 and 2015 to prove the concept of a fuel flexible gasification system. Furthermore, during the design phase important aspects of SER were considered. A detailed description of the preliminary thoughts of this experimental facility can be found in the literature.¹ Overall, the used procedure of experimental validation includes:

1. installing the test plant and experimental setup,
2. executing an experimental campaign,
3. collecting experimental data,
4. validating measurement data by the use of process simulation software and
5. finalizing a validated process model for a central database.

as basis for up-scaling the investigated process to an industrial scale. The following example is used to describe important steps as a reference for the validation procedure. A new 100 kW_{th} dual fluidized bed gasification test plant with an advanced reactor design, shown in Figure 20, has been installed for an experimental campaign aimed at optimizing the process performance. The results are compared with the findings from a previous generation with a classic reactor design, by using the process simulation model shown in Figure 18. The results have allowed a comparison with best practice following the idea of QFD. Figure 20 shows pictures and simplified sketches of both mentioned test plants. A detailed description of both plants and the experimental setup can be found in the literature.²⁻⁶



Figure 19: Gasification test plant used for the experimental validation of fuel flexible gasification³

1 Schmid J.C., 2014; 2 Müller S., 2017b; 3 Müller S., 2016; 4 Schmid J.C., 2014; 5 Diem R., 2015; 6 Kolbitsch M., 2016

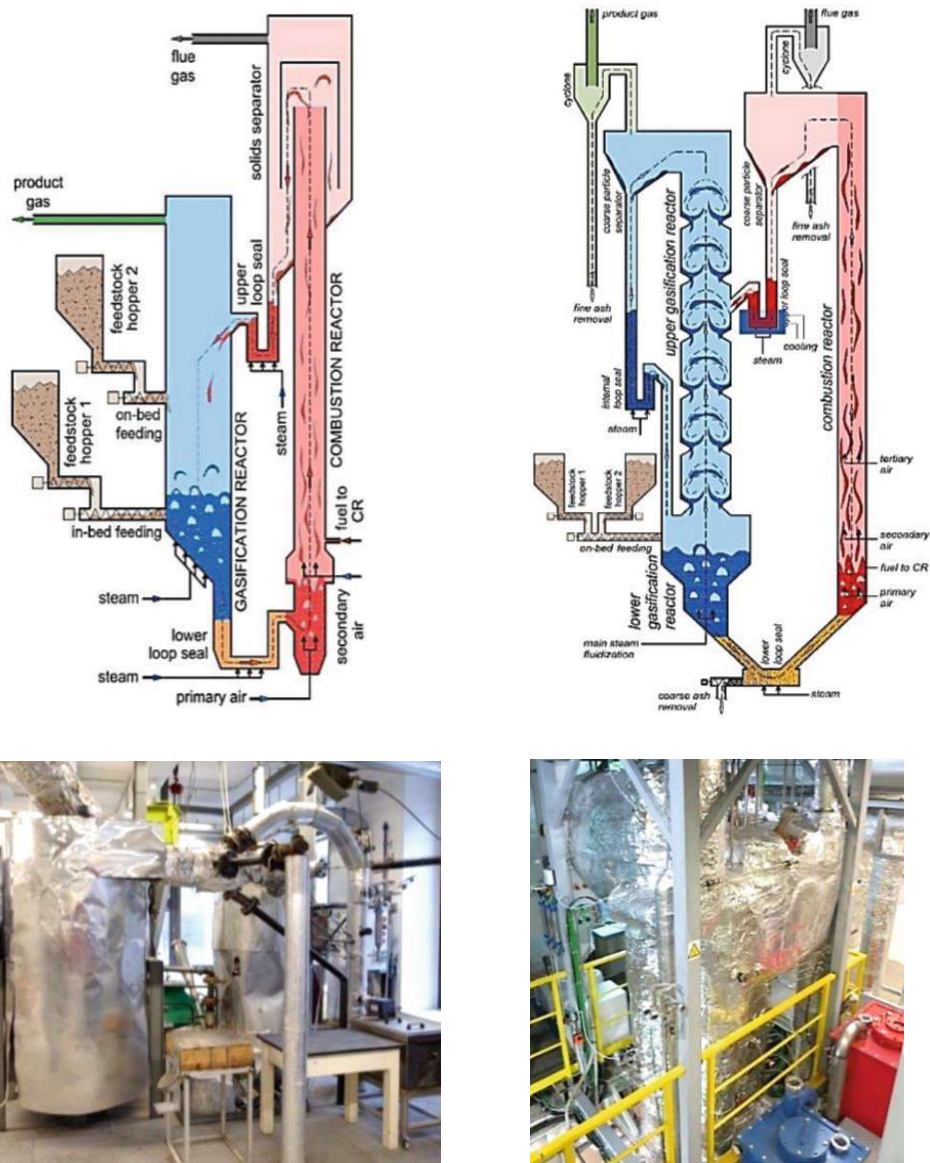


Figure 20: Schematic sketch (top) and pictures of the gasification test plants at TU Wien: classic design (bottom left) and advanced design (bottom right)¹⁻³

Both test plants were built with a similar size and a reactor height of about 7 m. The test plants have been used to execute experimental campaigns with different fuel types and varying process parameters. The aim of both test plants was to collect experimental data. The available measurement points and the used measurement equipment defined the precision of the investigation. In the present case, the main product gas components were analyzed by Rosemount NGA2000 measurement equipment in parallel with a Perkin Elmer ARNEL – Clarus 500 gas chromatograph.¹⁻³ The measurements were supported by the Accredited and Notified Testing Laboratory for Combustion Systems at TU Wien.⁴

1 Müller S., 2017b; 2 Müller S., 2016; 3 Schmid J.C., 2019

4 https://www.vt.tuwien.ac.at/test_laboratory_for_combustion_systems/EN/ (accessed on August 18, 2021)

Over 100 temperatures are measured with thermocouples manufactured by Kalinsky. Process media flows like air and steam are detected by flowmeters. The input of fuel is measured by scales combined with calibrated dosing screws. The product gas and flue gas output are determined by orifice plates. Seventy pressure sensors from Kalinsky (DS2) are used as an effective process control for operation of the fluidized bed. Tar is sampled isokinetically with impinger bottles. Thus, the gravimetric tar content as well as tar components detected by gas chromatography coupled with mass spectrometry (GCMS) are determined. The realized measurement method is based on CEN/TS 15439.^{1,2} The implementation of an adequate measurement strategy requires significant effort. Table 3 shows a comparison of important process parameters of both plants and Table 4 shows the number of increased measurement points as a part of the new test plant. Improved measurement methods have been established by a team of experts enabling advanced analytics for research projects.^{3,4} The advanced test plant has been designed to handle smaller particle sizes and offers more flexibility with respect to its operation with different fuel types as well as different operation modes. As a part of the executed experimental campaigns, the aim of the experiment setup was to validate the advanced fuel flexible gasification test plant model. Besides fuel flexibility, a special focus was placed on operation of SER as well as CO₂ gasification. Hereby, the ability of the used bed material with respect to heat transfer, catalytic activity and attrition resistance has been set as important design criteria. Figures 21 and 22 show additional drawings and pictures of the installed test plant. Table 5 gives an overview of the main design parameters. The example described here indicates the advantage of research and development work based on best practice solutions. The improvements were achieved based on the classic design representing best available technology for an experimental plant at that time.

Table 3: Important general operation parameters of both test plants⁵

| Parameter | Unit | Classical | Advanced | Classical | Advanced |
|------------------------|----------------------|--------------|--------------|--------------|--------------|
| Fuel type | - | Wood pellets | Wood pellets | Wood pellets | Wood pellets |
| Bed material type | - | Olivine | Olivine | Calcite | Calcite |
| Bed material inventory | kg | 100 | 80 | 100 | 80 |
| Particle size | µm | 300–600 | 100–400 | 500–1000 | 250–800 |
| Gasification temp. | °C | 800–900 | 650–860 | 630–700 | 590–730 |
| Combustion temp. | °C | 840–940 | 850–1 050 | 780–860 | 800–1 000 |
| Fuel power to GR | kW | 75–100 | 50–100 | 75–80 | 85–115 |
| Steam-to-fuel ratio | kg/kg _{daf} | 0.6–1.2 | 0.5–1.7 | 1.3–1.8 | 0.5–1.7 |
| Fuel mass flow GR | kg/h | 15–20 | 10–20 | 15–16 | 17–23 |

Table 4: Number of increased measurement points at the test plant⁵

| Parameter | Temperature | Pressure | Volume flow | Gas analytic | Weight | Motor speed | Level |
|------------------|-------------|----------|-------------|--------------|--------|-------------|-------|
| Supply units | 31 | 4 | 11 | 0 | 2 | 1 | 0 |
| Gas production | 50 | 48 | 0 | 0 | 0 | 3 | 0 |
| Downstream units | 24 | 18 | 2 | 22 | 0 | 1 | 4 |
| Sum | 105 (27) | 70 (16) | 13 (8) | 22 (22) | 2 (2) | 5 (2) | 4 (4) |

The values in brackets illustrate the number of measurements at the classical DFB pilot plant

1 Wolfesberger U., 2013; 2 Aigner I., 2011;

3 www.icblab.at; 4 Benedikt F., 2020; 5 Müller S., 2017b

Table 5: Main design parameters^{1,2,3,4}

| Parameter | Unit | Lower gasification reactor | Upper gasification reactor | Combustion reactor |
|--------------------------------------------|---------------|---------------------------------|----------------------------|--------------------|
| Range of temperature | °C | 700–850 | 800–950 | 830–980 |
| Fuel power | kW | 40–110 | | 30–57 |
| Pressure | - | Close to atmospheric conditions | | |
| Fluidization regime | - | Bubbling bed | Turbulent zones | Fast fluidized |
| Fluidization agent | - | Steam | Steam | Air |
| Amount of bed material (olivine) | kg | 75–110 | | |
| Superficial gas velocity (U) | m/s | 0.47–0.93 | 1.7–2.1 | 6.3–7.6 |
| Minimal fluidization velocity (U_{mf}) | m/s | 0.037 | 0.037 | 0.028 |
| Fluidization ratio (U/U_{mf}) | - | 13–25 | 40–55 | 220–270 |
| Terminal velocity (U_t) | - | 2.06 | 2.06 | 1.61 |
| Fluidization ratio (U/U_t) | - | 0.23–0.45 | 0.8–1.1 | 3.8–4.8 |
| Mean bed material diameter | μm | 250 | 250 | 250 |
| Fluidization media for fluid dynamic | - | Product gas | Product gas | Flue gas |
| Relevant cross section, fluid dynamic | mm | 68 × 490 | 128 × 128 | Ø 125 |
| Height of reactor part | m | 1.03 | 3.33 | 4.73 |
| Geometry | - | Conical bottom section | Square | Cylindrical |
| Inner dimensions of reactor | mm | 560 × 490; 68 × 490 | 128 × 128 | Ø 125 |

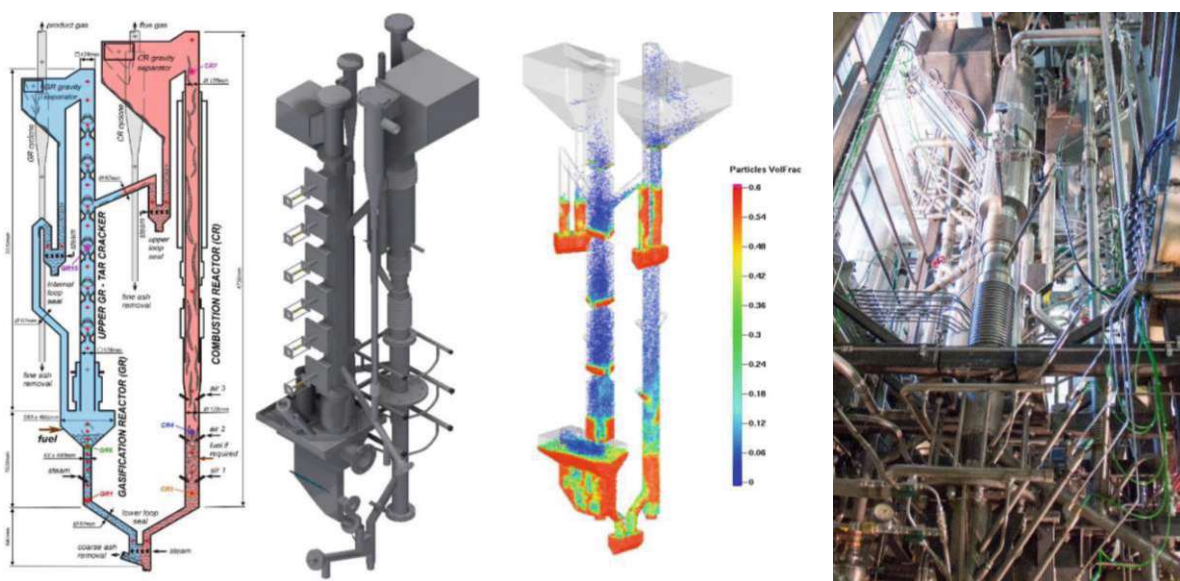


Figure 21: Basic design idea (left),² three-dimensional model,³ CFD model⁵ and a picture of the pilot plant excluding thermal insulation (right)^{6–9}

1 Müller S., 2013; 2 Schmid J., 2014
 3 Diem R., 2015 4 Kolbitsch M., 2016
 5 Lunzer A., 2021; 6 Müller S., 2016
 7 Schmid J.C., 2019; 8 Pasteiner H. A., 2015; 9 Müller S., 2016



Figure 22: Pictures of the pilot plant including thermal insulation¹

Additional measurement points are important to enable better understanding of the investigated processes by validated process simulation models based on overdetermined equation systems. The built test plant allows comparison between the results and previous findings, including improvements in methodology and measurement equipment. The installed test plant has been used for a series of experimental campaigns. Kolbitsch M.² provided analysis about the first gasification experiments and Schmid J.C.³ provided a summary about main experimental results. Furthermore, Benedikt F.,⁴ Fuchs J.⁵ and Mauerhofer A.⁶ carried out specific investigations with respect to fuel flexibility, SER and CO₂ gasification. As a result, extensive experimental data have been collected. Each experimental campaign was validated by the use of already described test plant model. A detailed description about the validation of experimental data can be found in the literature.⁷ Table 6 shows a short summary of the main relevant dimensions of a validation procedure. After importing gained measurement data into the simulation model, the used software tool minimizes small errors in measurement and calculates additional key performance indicators. As a result, a validated model of each experiment has been added to a central database and is available for further interpretation. Gubin V. used this database to carry out an investigation with respect to important correlations.⁸ Overall, the experimental demonstration of a novel process concept is essential for a successful execution of process development. Furthermore, the experimental demonstration represents a solid foundation for further large-scale realization if implemented in a smart manner. Hereby, a validated test plant model can be mentioned as important step of process development to enable a holistic interpretation. Within the present work, experimental data from the following experimental test plants is used as the basis of validated test plant models.

Table 6: Table template for reporting the validated experimental results⁷

| Parameter | Unit | Typical value | Measured | Validated result |
|--------------------------------|------------------------------------------------------------|---------------|----------|------------------|
| Gasification temperature | °C | 800–860 | 674 | 679 |
| Product gas volume flow | Nm ³ /h | 14–22 | - | 12 |
| Absolute water conversion rate | kg _{H₂O} /kg _{H₂O} | 0.09–0.16 | - | 0.25 |

1 Müller S., 2016; 2 Kolbitsch M., 2016; 3 Schmid J.C., 2019; 4 Benedikt F., 2020
 5 Fuchs J., 2020; 6 Mauerhofer A., 2020; 7 Müller S., 2013; 8 Gubin V., 2020

Figure 23 shows an experimental facility for the production of hydrogen (left) and SNG (right). The hydrogen production test plant was installed beside the biomass gasification plant in Güssing. The SNG test plant is attached to the advanced gasification test plant at TU Wien shown in Figure 22 and allows the execution of experimental campaigns of the whole process chain. The experimental validation is carried out due to missing data sets as well as identified potential with respect to process optimization.¹ Furthermore, Figure 24 (right) shows a picture of an experimental test plant for the investigation of the Fischer–Tropsch process. The plant was attached to biomass gasification plant in Güssing shown in Figure 24 (left). The experimental setup allowed the execution of experimental campaigns enabling the production of Fischer–Tropsch fuels such as gasoline, diesel, wax and naphtha. The collected data also allowed a deep insight into important aspects of process operation and allows the creation of validated test plant models. Table 7 shows an example for a comparison of the actual TRL compared with the current MRL. Preliminary modelling is recommended to enable the best experimental development. The available experimental facilities are crucial for analysis of a potential future operation at an industrial scale. An evaluation of the validated models before implementation is recommended.

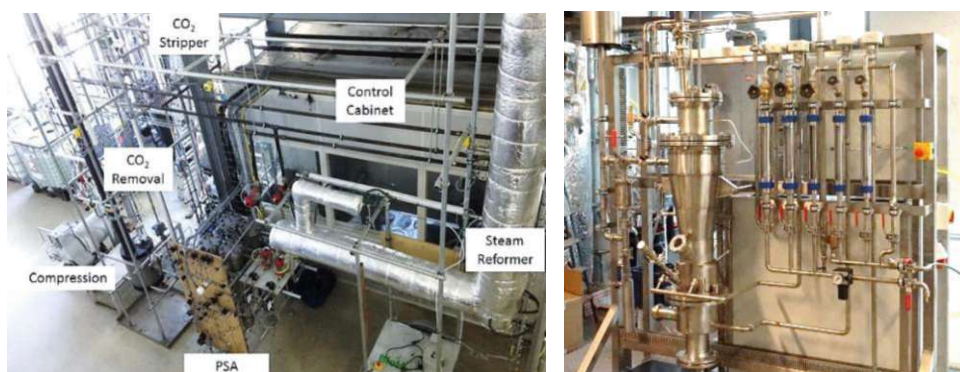


Figure 23: Experimental facility for hydrogen production (left) and synthetic natural gas production (right)^{2,3}



Figure 24: Combined heat and power plant Güssing (left) and laboratory-scale Fischer–Tropsch plant (right)⁴

Table 7: Rating the modelling readiness level and technological readiness level of the presented research facilities

| Parameter | Heat and power | Liquid biofuels | SNG | Hydrogen |
|-------------------------------|----------------|-----------------|-----|----------|
| Modelling readiness level | 9 | 5 | 5 | 4 |
| Technological readiness level | 9 | 6 | 7 | 7 |

1 Bartik A., 2021; 2 Loipersböck J., 2018; 3 Bartik A., 2020; 4 Müller S., 2017a

3.6. Evaluation of Validated Plant Models

The experimental results achieved allow an evaluation with respect to the consequences of further realization step. This includes the expected process performance. Furthermore, the model development stage allows forecasting the economic side of operation over the plant lifetime. This represents an important step during the development process because there are high costs associated with further implementation steps. Therefore, an evaluation before the industrial implementation is recommended. In addition, the present work includes an evaluation of the ecologic value. This endeavor is recommended at this stage of plant development to ensure the subsequent implementation steps will be effective. The **ecologic value** of a technology, an economic sector or even an industrial plant is defined in the present work as the ability of a unit or system to protect the surrounding ecology or even support the recreation of lost ecosystems in the past in accordance with the idea of a circular economy. [Table 7](#) shows a simplified illustration of an evaluation of the reached ecologic value of a novel solution based on experimental results and development data. The investigated solution is compared with the best available technology, determined as described within [Chapter 3.1](#) as the result of QFD analysis. The evaluation indicates strengths and weaknesses of a novel approach. An interdisciplinary team of experts weights different criteria. The reached value of a solution should be based on technical data and translated into the specific evaluation value scale. LCA methods can be used to reassess the reached ecologic score of different solution options.

Table 8: Example for an evaluation of a validated plant model

Which ecologic value can be reached by large-scale implementation of an SNG production?

| Decision criteria weighted between 0 and 20, valued between 0 and 10 | | Option A: Natural gas via the Nord Stream pipeline | | Option B: Erection of an SNG production plant | |
|----------------------------------------------------------------------|---------------|----------------------------------------------------|---------------|-----------------------------------------------|---------------|
| Decision criteria | Weighting (w) | Value (v) | Score (w × v) | Value (v) | Score (w × v) |
| Operation efficiency | 10 | 10 | 100 | 7 | 70 |
| Economic competitiveness | 10 | 10 | 100 | 5 | 50 |
| Ecologic Value | | | | | |
| Air quality | 10 | 10 | 100 | 5 | 50 |
| Greenhouse gas/CO ₂ emission factor | 10 | 6 | 60 | 9 | 90 |
| Land use/footprint | 10 | 6 | 60 | 8 | 80 |
| Carbon utilization | 10 | 0 | 0 | 8 | 80 |
| Water consumption | 10 | 10 | 100 | 5 | 50 |
| Impact on local flora and fauna | 10 | 9 | 90 | 5 | 50 |
| Generated waste | 10 | 10 | 100 | 5 | 50 |
| Resource recycling rate | 10 | 0 | 0 | 4 | 40 |
| Social impact | 10 | 5 | 50 | 7 | 70 |
| Contribution to culture | 10 | 5 | 50 | 8 | 80 |
| Sum weighting | 120 | Evaluation result | 610 | Ecologic value | 640 |
| Maximum score | 1 200 | | 61.0% | Ecologic rating | 64.0% |

Option B shows a higher ecologic value and therefore better contributes to the ecologic goals. Industrial demonstration should be foreseen. Strengths and weaknesses should be monitored during the implementation phase!

Besides this simplified evaluation, a broad stakeholder dialog is recommended to ensure a well-balanced rating of a novel solution accompanied by a precise calculation of all relevant dimensions. Further description of the latest state of the methodology can be found in the literature.^{1–3,9} The Eco-Management and Audit Scheme (EMAS) is a voluntary environmental management instrument that was developed by the European Commission. ISO 14000 is a family of standards related to the environmental management to help organizations to minimize negative affects to environment. And furthermore, in many cases an environmental impact assessment, like the Umweltverträglichkeitsprüfung (UVP) in Austria, is compulsory due to public authorities in accordance with the Austrian constitution.

3.7. Industrial Plant Information Model

An IPIM includes all relevant information to operate a process at the industrial scale. This includes the technical side of an operation as well as important economic parameters and legal aspects. The creation of an IPIM also prepares a framework for the documentation of a future operation of an investigated plant. Walcher C. and Hammerschmid M.^{4,5} executed detailed analyses regarding related documents and the necessary information flow. The creation of an IPIM accompanies the plant engineering process. The engineering process is based on a basic flow chart, a process flow chart, a detailed simulation model and a pipe and instrumentation flow chart as central documents guiding the overall process. As a result, detailed drawings of a plant can be produced. An engineering process has to be understood as an iterative process comprising several engineering phases, including conceptual design, basic engineering, detail engineering, manufacturing, commissioning and operation. Virtual development environments allow acceleration of the development time and cost reduction if applied in a smart manner, because costs of the installation and operation are determined at an early stage of development. It is recommended to monitor all design changes in a central database by using a central planning model.⁶ Computer-aided automation of important planning steps has been investigated by Bakosch C.⁷ The development results of main stages for the realization of an IPIM are illustrated in the following figures. [Figure 25](#) shows an example for a process flow diagram of an industrial plant. [Figure 26](#) shows a three-dimensional (3D) drawing of an industrial plant. [Figure 27](#) shows a detailed pipe and instrumentation flow diagram. A central planning model also allows analyzing the investment and operational costs over the plant lifetime, also known as the total cost of ownership (TCO).⁸ [Table 9](#) shows an example of an analysis of relevant costs with respect to the investment decision. [Equations 7–10](#) show the main important relations that have been used to model the investment decision for a plant. Heat and power supply by utilizing biogas offers moderate investment costs as well as a positive operation result from a financial point of view due to high subsidies for electricity generation. A precise description of the shown example can be found in the literature.⁵ The proposed methodology recommends the calculation of investment costs (I_0), the expected operating costs (A) and the expected income (E) before a scheduled installation. This endeavor allow for the best decision to be made. The financial model also requires validation with real operational data after realizing the investigated plant.

1 EMAS, 2009; 2 DIN EN ISO 14001, 2015; 3 Umweltverträglichkeitsprüfungsgesetz, 2000; 4 Walcher C., 2018; 5 Hammerschmid M., 2019; 6 Grafinger M., 2020; 7 Bakosch C., 2021; 8 Wildemann H., 2008; 9 Heimerl P., 1990

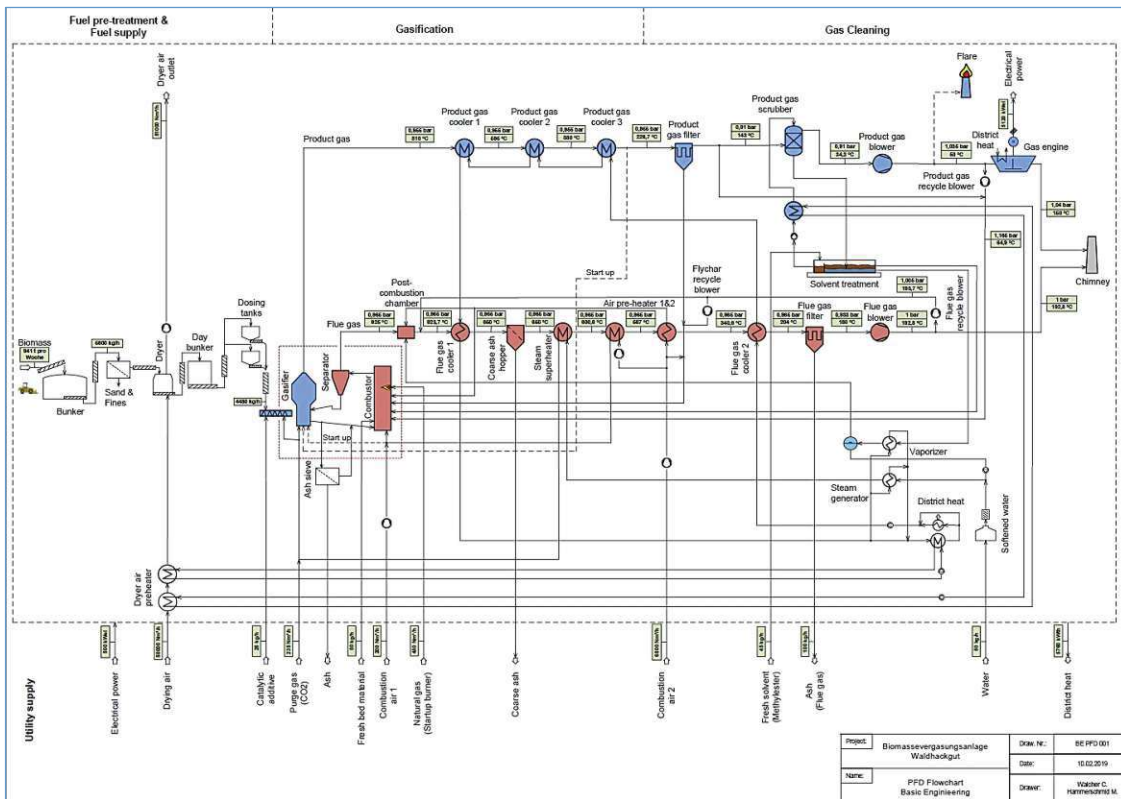


Figure 25: Example of a process flow diagram for the production of power and heat via biomass steam gasification^{1,2}

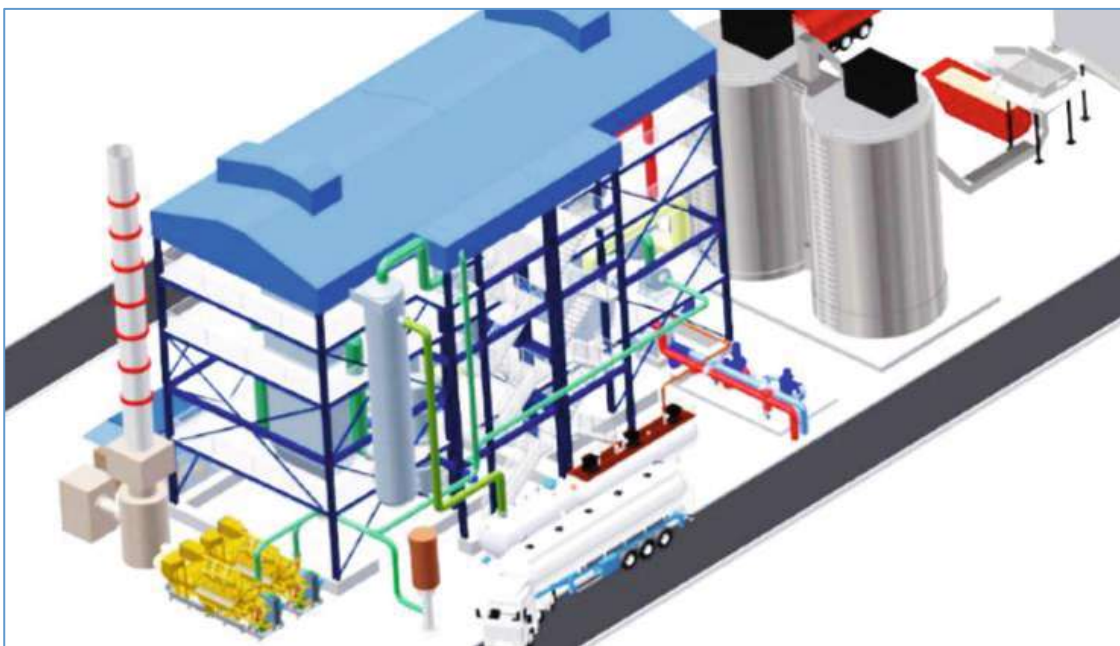


Figure 26: Example of a 3D model of an industrial plant for the production of power and heat via biomass steam gasification^{2,3}

1 Walcher C., 2018; 2 Hammerschmid M., 2019; 3 Bakosch C., 2021

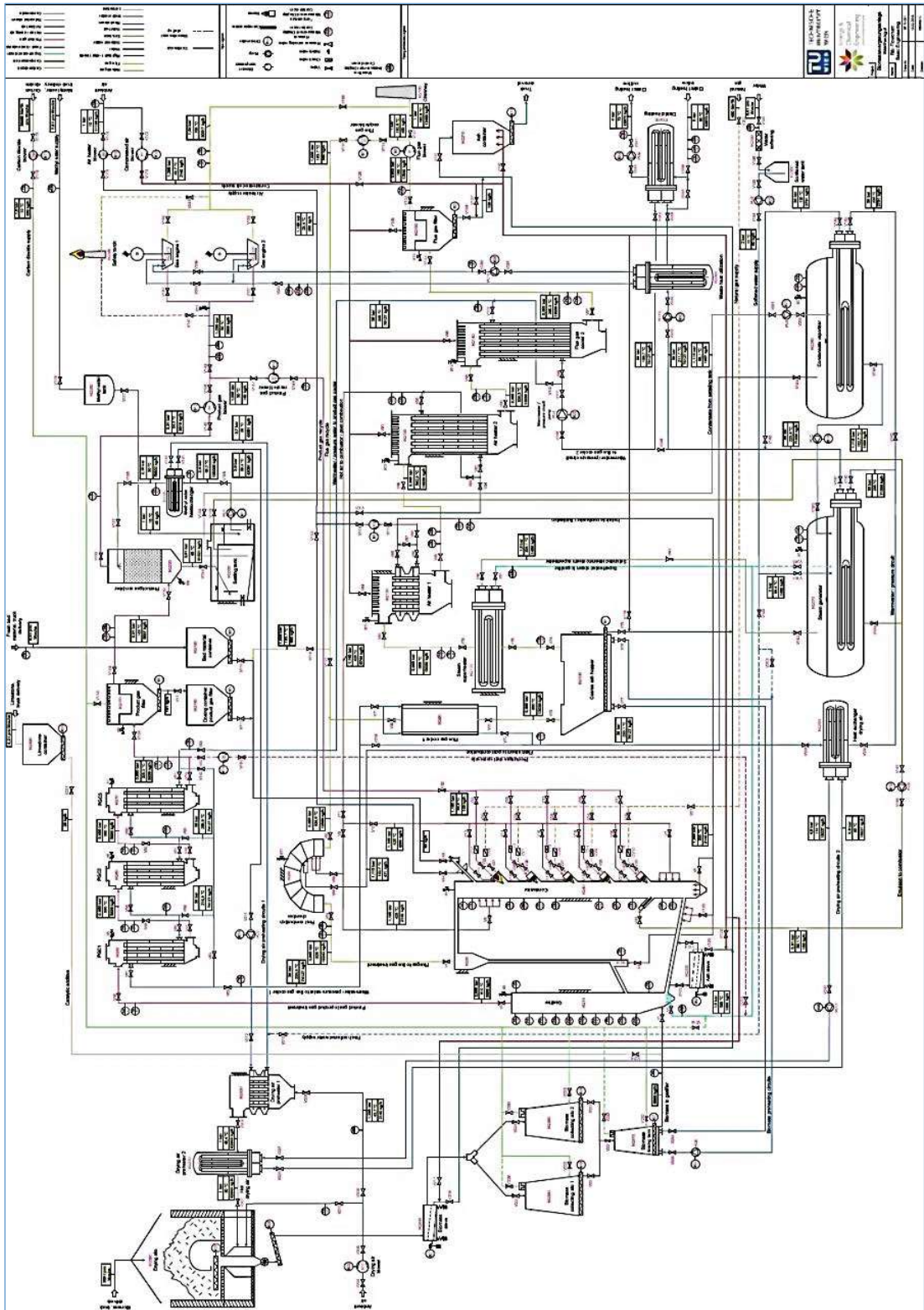


Figure 27: Schematic illustration of a pipe and instrumentation flow sheet to produce power and heat via biomass steam gasification^{1,2}

1 Walcher C., 2018; 2 Hammerschmid M., 2019;

Table 9: Financial model to evaluate the investment decision¹

| Parameter | Unit | Natural gas CHP | Biomass CHP | Biogas CHP | Wood chips DFB gasifier | Bark DFB gasifier | SLF DFB gasifier |
|--------------------------------------------------|-------|--------------------|----------------|---------------|----------------------------|----------------------|---------------------|
| Boundary conditions | | | | | | | |
| Thermal fuel power | MW | 16 | 16 | 16 | 16 | 16 | 16 |
| Operating hours | h/a | 3 500 | 6 500 | 7 000 | 7 500 | 7 500 | 7 500 |
| Natural gas consumpt. | MWh/a | 56 000 | | | | | |
| Biomass consumption | MWh/a | | 104 000 | 112 000 | 120 000 | 120 000 | |
| Consumpt. of residuals | MWh/a | | | | | | 120 000 |
| Electricity production | MWh/a | 22 000 | 41 000 | 45 000 | 48 000 | 48 000 | 48 000 |
| Heat production | MWh/a | 20 000 | 28 000 | 28 000 | 28 000 | 28 000 | 28 000 |
| Investment costs (I ₀) | € | 5 100 000 | 24 100 000 | 16 300 000 | 22 500 000 | 22 500 000 | 27 000 000 |
| Expenses (A) | | | | | | | |
| Fuel costs | €/a | 1 848 000 | 2 094 000 | 1 876 000 | 2 700 000 | 1 548 000 | 720 000 |
| CO ₂ certificates | €/a | 258 000 | | | | | 718 000 |
| Maintenance, insurance & administration costs | €/a | 242 000 | 1 145 000 | 774 000 | 1 069 000 | 1 069 000 | 1 283 000 |
| Costs of utilities | €/a | 102 000 | 756 000 | 593 000 | 951 000 | 951 000 | 951 000 |
| Labor costs | €/a | 29 000 | 399 000 | 399 000 | 399 000 | 399 000 | 399 000 |
| Ash disposal | €/a | | 110 000 | | 147 000 | 147 000 | 147 000 |
| Sum | €/a | 2 479 000 | 4 504 000 | 3 642 000 | 5 266 000 | 4 114 000 | 4 218 000 |
| Earnings (E) | | | | | | | |
| Heat production | €/a | 600 000 | 840 000 | 840 000 | 840 000 | 840 000 | 840 000 |
| Electricity production | €/a | 1 056 000 | 4 367 000 | 6 334 000 | 5 112 000 | 5 112 000 | 1 632 000 |
| Sum | €/a | 1 656 000 | 5 207 000 | 7 174 000 | 5 952 000 | 5 952 000 | 2 472 000 |
| Operation result (P = E - A) | | | | | | | |
| Net income | €/a | -823 000 | 703 000 | 3 532 000 | 686 000 | 1 838 000 | -1 746 000 |
| Investment evaluation (i = 6%, k = 20) | | | | | | | |
| Net present value (NPV) | € | -14 539 810 | -16 036 000 | 24 212 040 | -14 631 5800 | -1 418 140 | -47 026 620 |

$$\text{Eq. 7} \quad P_{\epsilon} = E - A \quad \left[\frac{\epsilon}{a} \right] \quad \text{Eq. 8} \quad \text{BWSF} = \frac{(1+i)^k - 1}{(1+i)^k \cdot i} \quad [-]$$

$$\text{Eq. 9} \quad \text{NPV} = -I_0 + \text{BWSF} \cdot P_{\epsilon} \quad [€] \quad \text{Eq. 10} \quad C_{\text{opt},X} = \sum A_i - E_i \quad \left[\frac{\epsilon}{a} \right]$$

Besides, the produced model includes important aspects with respect to the final ecologic value, namely an estimation of the expected emissions² and further data required by the responsible authorities. The described computer-integrated plant engineering process enables the creation of a digital twin of the foreseen industrial plant. The produced model needs to be updated throughout the entire engineering process and serves as central documentation over the plant lifetime. Measurement data from real plant operation is needed to validate the created IPIM.

1 Hammerschmid M., 2019; 2 GEMIS V 5.0 Database

3.8. Validation of Industrial Plant Models

An industrial plant model is validated after the start-up and commissioning phase of an industrial-scale. The gained measurement data are used to affirm the preliminary assumptions. After the plant is completed, it is handed over to the plant operator. The plant operator aims to improve and optimize the commercial performance of the plant. Besides, environmental goals need to be monitored to satisfy the legal framework for the operation of the plant.

Figure 28 shows a picture of the biomass power plant Güssing after the plant commissioning phase. The plant was installed as demonstration project of dual fluidized bed steam gasification for the production of electricity and heat. Extensive measurement data and operation experience were gained from 100 000 hours of operation. The results enabled the validation of an industrial plant model.¹ Figure 29 shows a picture of a process simulation model to calculate related KPIs. A first digital role model for further implementation projects was created and served as basis for the creation of many more industrial plant models. A summary regarding the available models and their usage to investigate additional large-scale concepts can be found in the literature.^{2,3}

Table 10 shows a simplified example of how the results could be evaluated. This endeavor provides quantified data describing the capability of the investigated technology with respect to the set development targets. In summary, industrial demonstration projects allow the IPIM to be validated with respect to KPIs, economic results as well as the ecologic value. At this stage, quantified ecologic measurements as well as validated financial data with respect to the continuous operation are available.



Figure 28: Drawing and Picture of industrial plant⁴

1 Pröll T., 2004

2 Müller S., 2013

3 Hofbauer H., 2020; 4 Müller S., 2017a

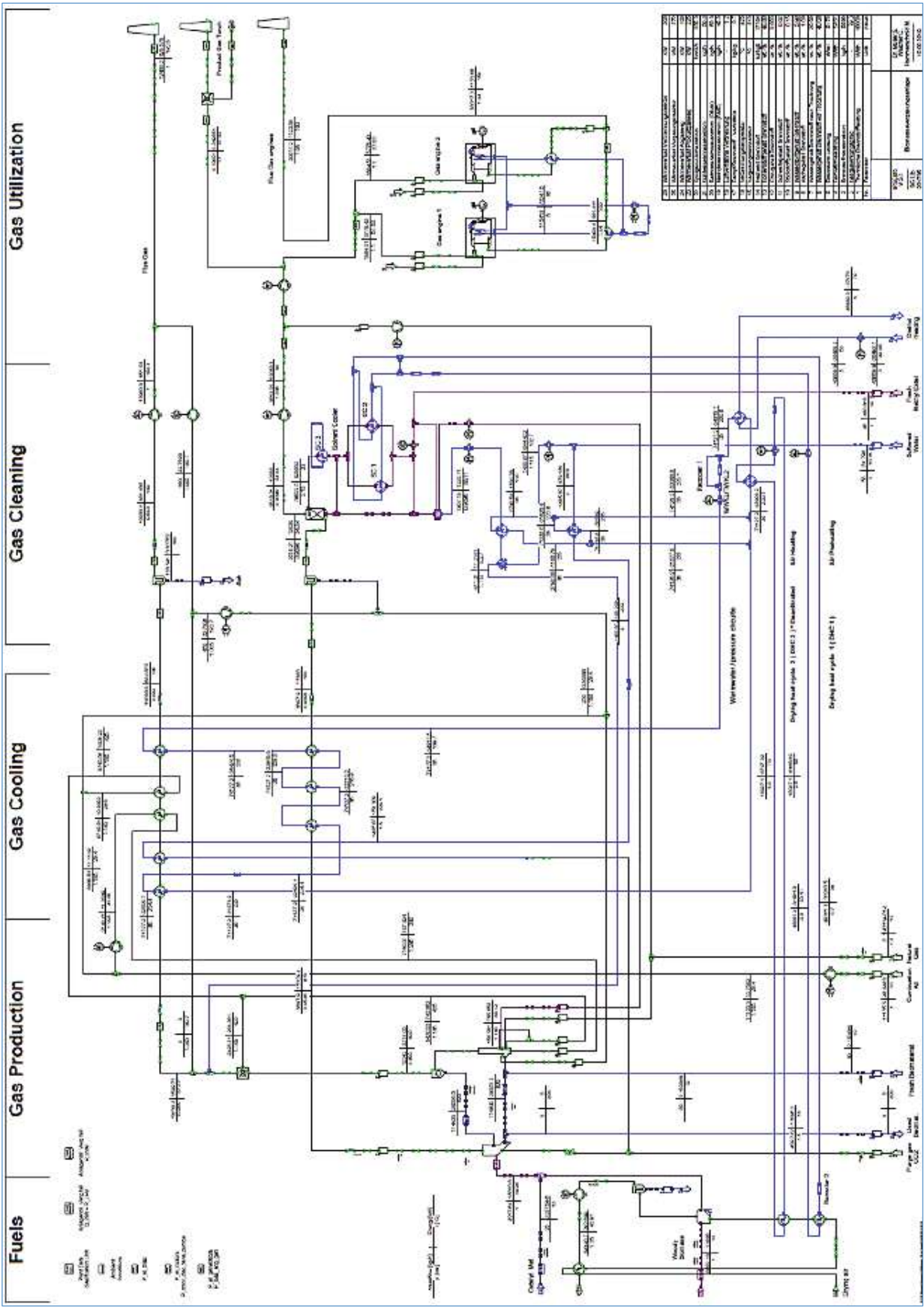


Figure 29: Schematic illustration of the validated mass and energy balances of an industrial plant

Table 10: Simplified evaluation of the outcome of a large-scale demonstration project^{1,2}

Did the demonstration project reach the expected outcome?

| Target dimension | Unit | Development target expected value | Demonstrated validated value |
|----------------------------|------------------------|-----------------------------------|------------------------------|
| Key performance indicators | | | |
| Operation hours | h | 113 880 | ~ 100 000 |
| Electric efficiency | % | 30 | 31 |
| District heating output | MW | 4 | 4.2 |
| Ecologic value | | | |
| Particle matter | mg/Nm ³ | < 5 | 3 |
| Greenhouse gas emissions | tCO _{2 eq} /a | < 50 000 | 43 200 |
| Generated jobs | - | 7 | 9 |
| Economic results | | | |
| Income return | % | 10 | 5 |

The demonstration met the technological and environmental targets.

Following this approach, a decision regarding the demonstration outcome can finally be made. If the demonstration project has reached the set target values, the investigated system can be described as complete and qualified for further implementation. Hence, the industrial plant model is validated, evaluated and completed as a role model for further platform-based implementation, monitoring and sharing.

1 cf. Müller S., 2013

2 cf. Wulf C., 2018

3.9. Implementation, Monitoring and Sharing

As a result of the modelling process, validated models enable target-oriented implementation of novel technology as well as monitoring and sharing valuable solutions. Figure 30 illustrates a future-oriented concept for a collaboration platform to implement, monitor and share validated industrial information models. A central collaboration platform is an important pre-condition to solve complex interdisciplinary development projects. Hofmann R. reported the importance of adequate modelling as important pre-condition to optimize our existing energy system.¹ Killian M. described the importance of models for the predictive control of energy efficiency.² Grafinger M. described best practice solutions for virtual development environments.³ Biffl S. indicated a valuable methodology to accompany the related software engineering methods.⁴ Within the present work, these approaches are acknowledged as an important outlook for the future development of the proposed methodology. Therefore, future work should concentrate on the development of a suitable digital framework to support collaboration platforms for the development of industrial plants enabling reduced fossil CO₂ emissions.

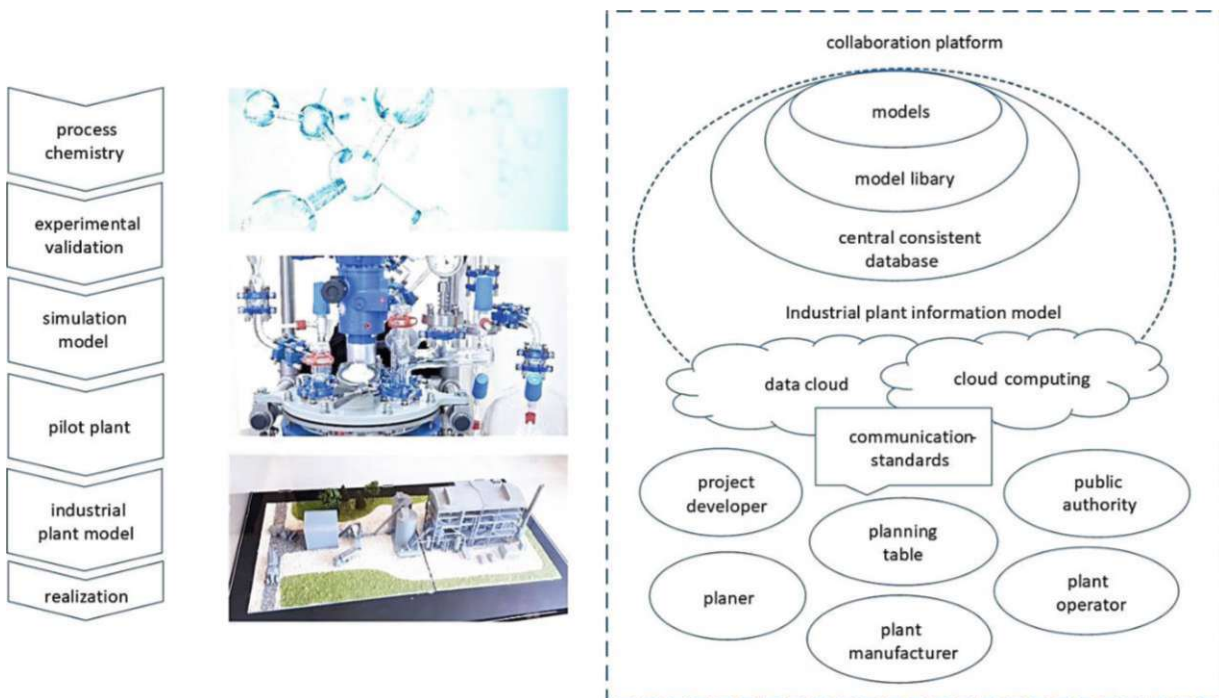


Figure 30: Concept for the implementation of a collaboration platform

1 Hofmann R., 2019

2 Killian M., 2016

3 Grafinger M., 2020; 4 Biffl S., 2006

3.10. Summary

Chapter 3 has provided a concise overview of the relevant steps from defining a development target up to sharing a digital role model for its implementation within an improved future energy system. The important aspects are summarized below.

- The definition of [development goals](#) at the beginning of the development process from a holistic point of view is essential to enable a target-oriented implementation. The interdependence of different targets needs to be acknowledged. QFD can offer a suitable method to find a consistent development strategy (cf. [Chapter 3.1](#)).
- During the model development, all relevant relations need to be considered by the formulation of adequate equations reflecting the identified target dimensions. Model development enables the preliminary calculation of the [expected benefits](#) of a novel process concept. Quality management of all developed models in a [central consistent database](#) ensures a powerful and reliable development space enabling a continuous improvement process (cf. [Figure 16](#)).
- The creation of a test plant model enables a preliminary analysis of the purpose of an experimental test plant and indicates important fields before further steps of cost-intensive experimental investigations. A detailed analysis which experimental data is missing and desired is recommended before the realization of a test plant. At this stage, this includes also reflecting foreseen [measurement methods](#) and their reliability (cf. [Figure 18](#)).
- The [installation of a test plant aims to generate data](#) and involves a great commitment of resources. Experimental campaigns provide the necessary data to validate mathematical models. [Validated models](#) provide sufficient data to evaluate the impact of a large-scale realization before incurring high implementation costs. At this stage, a deep analysis of the proposed concept is recommended before additional realization steps (cf. [Chapters 3.5](#) and [3.6](#)).
- The creation of an IPIM enables a preliminary calculation following a holistic perspective and serves as central documentation framework over the plant lifetime. The [validation of an IPIM](#) affirms preliminary assumptions as well as collected measurement data during experimental and operational campaigns. Evaluation of the results allows the production of a high-value digital role model for an optimized implementation. As a result of the modelling procedure, digital twins can be provided and [shared as role models](#) to improve applied energy systems.

Overall, the [implementation of MRL](#) would enable introducing a unified methodology for the implementation of digital twins as a part of a development process. It simultaneously ensures practical value if applied in an adequate manner. The following chapters contain representative examples for process and energy technology development. [Chapter 4](#) describes the advances in processes enabling a reduction in fossil CO₂ emissions. Then, [Chapter 5](#) discusses the translation into large-scale operation using industrial plant models based on practical examples from the last 10 years.

4. ADVANCED PROCESSES FOR REDUCED FOSSIL CARBON DIOXIDE EMISSIONS

Energy-intensive industries, mobility and transportation make up the largest share of energy consumption within the European Union. Historically, the energy supply has been based on fossil carbon sources such as coal, crude oil and natural gas imported from outside the European Union. This chapter illustrates improvements made to reduce fossil CO₂ release with novel approaches in thermochemical conversion. The proposed concepts are based on the utilization of biogenic fuel types fulfilling important requirements with respect to sustainable utilization. At this point, the reduction in fossil carbon utilization from coal, crude oil and synthetic gas is seen as the primary aim. Therefore, existing processes have had to be improved to meet future expectations. Biomass offers promising potential as sustainable energy resource, if applied in a smart manner. As a starting point, the basic principles of thermal fuel conversion of each specific process need to be considered. The process chemistry as well as the related thermodynamics represent important pre-conditions for the implementation of novel processes enabling the reduction of fossil CO₂ emission. Therefore, Chapter 4 describes the fundamentals of the generation of synthesis gas (syngas) from biogenic feedstock via the advanced processes SER and CO₂ gasification.

These processes are explained in detail to illustrate their potential to contribute to a reduction of fossil CO₂ emissions. The described fundamentals have been used within the present work in the context of advanced digital methods to prepare large-scale implementation. All mentioned processes are based on the state of the knowledge of biomass steam gasification. A detailed description of biomass steam gasification can be found in the literature.^{1,2} Figure 31 illustrates main important aspects of thermal fuel conversion as a part of a gasification process. These aspects of biomass steam gasification have been described in the fundamental literature.¹ In recent years, biomass utilization has been identified as one possibility to avoid fossil CO₂ emissions. Volatile feedstock prices have led to increased attention on flexible utilization of alternative biogenic fuel types. This includes the possibility to reduce financial risks of negative consequences of feedstock price development.

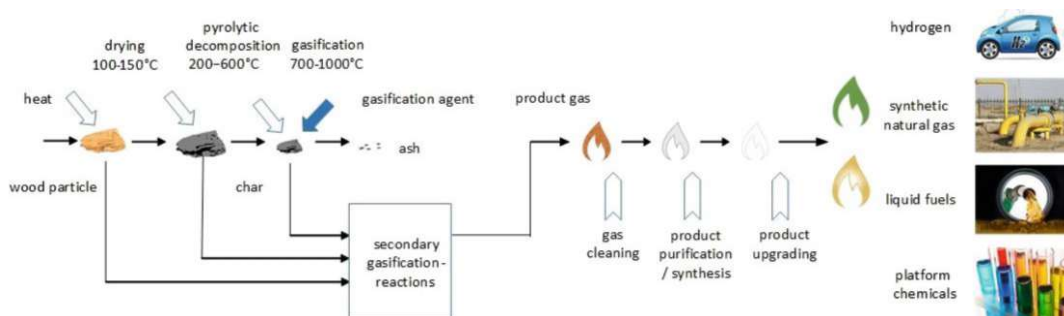


Figure 31: Basic principles of thermal fuel conversion²

1 Hofbauer H., 1993; 2 cf. Kaltschmitt M., 2009

Gasification is the thermochemical conversion of solid or liquid fuel into gaseous fuel. The main driving force for the conversion process is the action of heat in the presence of a gasification agent. Complete combustion—full oxidation—of the fuel particle is prevented because of a lack of available oxygen.¹ The produced gas is called product gas. All types of solid fuels, which are applicable for combustion, are technically also fuel for gasification. In the present work, the described process steps have been applied with the aim of preventing fossil CO₂ emissions. In recent years, a wide range of different solid fuel types have been analyzed as feedstock with respect to their suitability to reduce fossil CO₂ emissions. Table 11 presents an overview of the main fuel parameters of eight solid fuel types. Figure 32 shows pictures of used fuel samples. Whereas biogenic feedstock have few impurities, different waste materials are more impure, a factor leading to relevant challenges with respect to gas cleaning before subsequent synthesis. Utilization of biogenic feedstock prevents fossil CO₂ emissions. There is currently increased research attention to analyze the consequences of utilizing different fuel types with respect to greenhouse gas emissions and the related eco balance. Therefore, Table 11 includes emission factors for each fuel type from a global emission model database of integrated systems (GEMIS). Hereby, carbon capture and utilization (CCU) aims at utilizing processed carbon instead of disposal as CO₂ emission. Besides, all fuel types include a small share of hydrogen. The addition of steam (H₂O) enables the production of H₂-rich product gas if the production of H₂-rich energy carriers is the aim of the investigated process.

Table 11: Proximate and ultimate analysis of different solid fuel types^{2–6}

| Parameter | Unit | SW | SCB | EOP ^b | BA | RH | SLF | MWF | LIG | Observed values |
|------------------------------|------------------------|-------|-------|------------------|-----------------|----------------|-------|-------|-----------------|------------------|
| Water content | mass.-% | 7.2 | 7.7 | 11.8 | 7.6 | 7.5 | 7.1 | 1.6 | 13.0 | 1.5–15.0 |
| Ash content | mass.-% | 0.2 | 2.3 | 11.0 | 7.0 | 15.2 | 12.2 | 7.8 | 4.2 | 0.1–16.0 |
| Carbon (C) | mass.-% _{daf} | 50.8 | 48.9 | 52.4 | 52.3 | 51.1 | 80.5 | 76.0 | 68.4 | 48.0–81.0 |
| Hydrogen (H) | mass.-% _{daf} | 5.9 | 5.9 | 6.2 | 6.0 | 6.1 | 12.1 | 11.3 | 3.9 | 3.5–12.5 |
| Nitrogen (N) | mass.-% _{daf} | 0.2 | 0.4 | 1.1 | 0.3 | 0.6 | 0.5 | 0.6 | 0.9 | 0.1–1.1 |
| Sulfur (S) | mass.-% _{daf} | 0.005 | 0.05 | 0.1 | 0.05 | 0.07 | 0.3 | 0.1 | 0.4 | 0.005–0.5 |
| Chlorine (Cl) | mass.-% _{daf} | 0.005 | 0.06 | 0.1 | 0.05 | 0.1 | 2.2 | 1.2 | 0.05 | 0.005–2.5 |
| Oxygen (O) ^a | mass.-% _{daf} | 43.1 | 44.7 | 40.1 | 41.3 | 42.0 | 4.4 | 10.8 | 26.3 | 4.5–45.0 |
| Volatile matter | mass.-% _{daf} | 85.6 | 85.7 | 85.2 | 77.7 | 80.7 | 91.8 | 93.3 | 54.1 | 54.0–93.0 |
| LHV, dry | MJ/kg _{db} | 18.9 | 17.8 | 17.6 | 18.2 | 15.9 | 31.1 | 31.5 | 24.3 | 15.0–32.0 |
| Ash defo. temp. (A) | °C | 1 330 | 1 180 | 750–850 | na ^c | >1 350 | 1 210 | 1 180 | na ^c | 750 to > 1 350 |
| Ash flow temp. (D) | °C | 1 440 | 1 330 | >1 440 | na ^c | - ^d | 1 320 | 1 290 | na ^c | 1 290 to > 1 440 |
| Emission factor ^d | t _{CO2} /MWh | 0 | 0 | 0 | 0 | 0 | 0.24 | 0.36 | 0.34 | 0–0.36 |
| Fuel price | €/MWh | 30 | 20 | 20 | 20 | 20 | 1 | 1 | 1 | 1–30 |

a: calculated by difference to average 100 mass-%_{daf}

b: consists of 4.4 mass-% initial ash and 6.6 mass-% CaCO₃ addition before pelletization

c: not analyzed or did not occur

d: fossil carbon, value according to GEMIS V.5.0 database; biogenic raw material assumed as carbon neutral

1 Hofbauer H., 2009; 2 Benedikt F., 2018; 3 Benedikt F., 2020; 4 Pribyl M., 2020; 5 Friedl A., 2005
6 <https://www.umweltbundesamt.at/angebot/leistungen/angebot-cfp/gemis> (accessed on March 27, 2021)

Reducing fossil CO₂ emissions by applying dual fluidized bed steam gasification of woody biomass has been investigated on different scales over several years. This endeavor includes the results achieved with the advanced gasification test plant at TU Wien described in [Chapter 3.5](#).¹ This work has demonstrated the capability of the gasification process with respect to the utilization of different types of feedstock. [Figures 33 and 34](#) show the main results of experimental campaigns aimed at optimizing process conditions for the gasification of shown fuel types in [Table 11](#). [Figure 33](#) shows the product gas composition during gasification experiments and [Figure 34](#) illustrates crucial results of the tar measurements, a critical aspect with respect to the necessary gas cleaning strategy.



Figure 32: Different solid fuel types¹

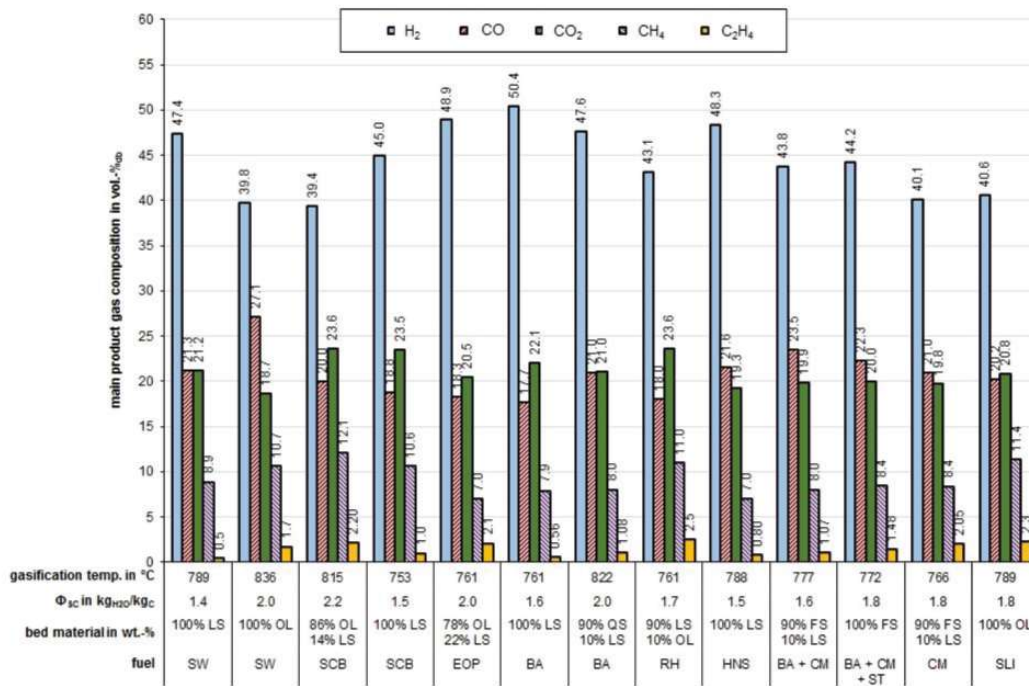


Figure 33: Results of experimental gasification of different fuel types—product gas—with an advanced test plant^{2,3}

1 Benedikt F., 2018; 2 Benedikt F., 2020; 3 cf. Chapter 3.5

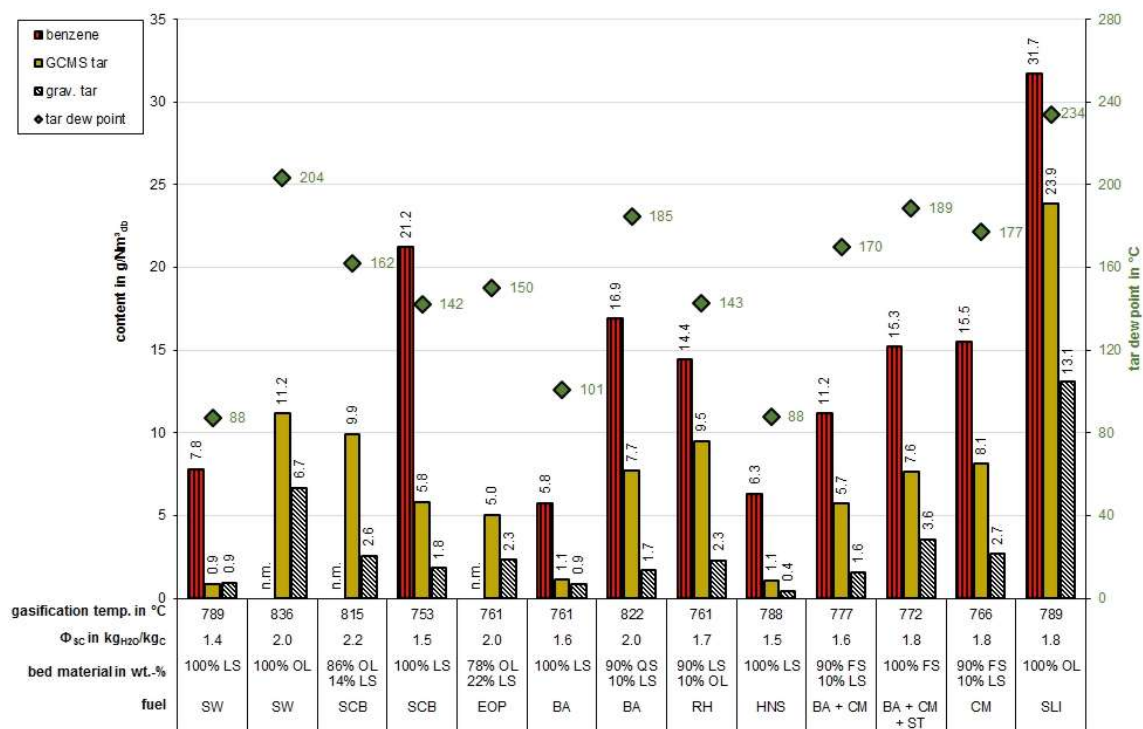


Figure 34: Results of experimental gasification of different fuel types—tar content—with an advanced test plant^{1,2}

These results have been validated by using a simulation model. The executed measurements have formed the basis for a solution of an overdetermined equation system. Furthermore, the precise measurement of the product gas components have generated a solid database for the development of a suitable gas cleaning strategy. The experiments have proved the fuel flexibility of the used gasification system with respect to fuel quality. Consequently, a greater variety of different solid fuel types could be applied to replace fossil carbon sources. The results have led to progress with respect to validating experimental data of the advanced gasification test plant at TU Wien described in Chapter 3.5. Besides, the latest research activities aim at improving the basic process with respect to the reduction of fossil CO₂ emissions by improving the applied process conditions. Recycling CO₂ as gasification agent as well as the selective transport of CO₂ via SER is introduced within the present work as a novel approach to biomass steam gasification to decrease fossil CO₂ emissions.

4.1. Sorption Enhanced Reforming

SER aims at producing a gas with high hydrogen content via application of limestone as bed material. A description of the state of the knowledge with respect to SER can be found in the literature.³⁻⁵ A high hydrogen content in the product gas is reached by reducing the CO₂ content in the produced gas due to a selective CO₂ transport enabled by the operated bed material.

1 Benedikt F., 2018; 2 cf. Chapter 3.5

3 Müller S., 2017b; 4 Fuchs J., 2019a; 5 Hafner S., 2021

The carbonation reaction and the calcination reaction enable the selective transport of CO₂ from the gasification reactor to the combustion reactor. The decay in the CO₂ load after many carbonation cycles causes a high demand of bed material renewal. So far, experiments have indicated further potential for process optimization. At this stage, the process optimization has aimed:

- to lower attrition causing bed material loss,¹
- to increase CO₂ transport and
- to increase the hydrogen production rate.^{2,3}

Figure 35 shows the basic principles of SER. The bed material plays a critical role in the process. The limestone transports CO₂ from the gasification reactor to the combustion reactor, decreasing tar and catalyzing the water–gas shift reaction. Therefore, the bed material particles are very relevant with respect to a favorable process operation. During the process operation, there is a decay in the CO₂ load of the circulating particles. Whereas particles from the gasification reactor transported via lower loop seal⁴ to the combustion reactor mainly contain calcium carbonate (CaCO₃) and char, particles from the combustion reactor transported via the upper loop seal⁴ to the gasification reactor are mainly based on calcium oxide (CaO) and ash. Figure 35 also includes pictures from bed material samples from the lower as well as from the upper loop seal. The samples were taken during an experiment and analyzed by using a scanning electron microscope as well as a thermo-gravimetric analysis (TGA). The results showed that only a low share of the CO₂ load capacity of circulating particles is used during the process operation. Furthermore, the used bed material is deactivated. The scanning electron micrographs show that the particle structure changed significantly after many calcination and carbonation cycles. Figure 36 shows the declining ability of a calcium particle to take up a CO₂ load.

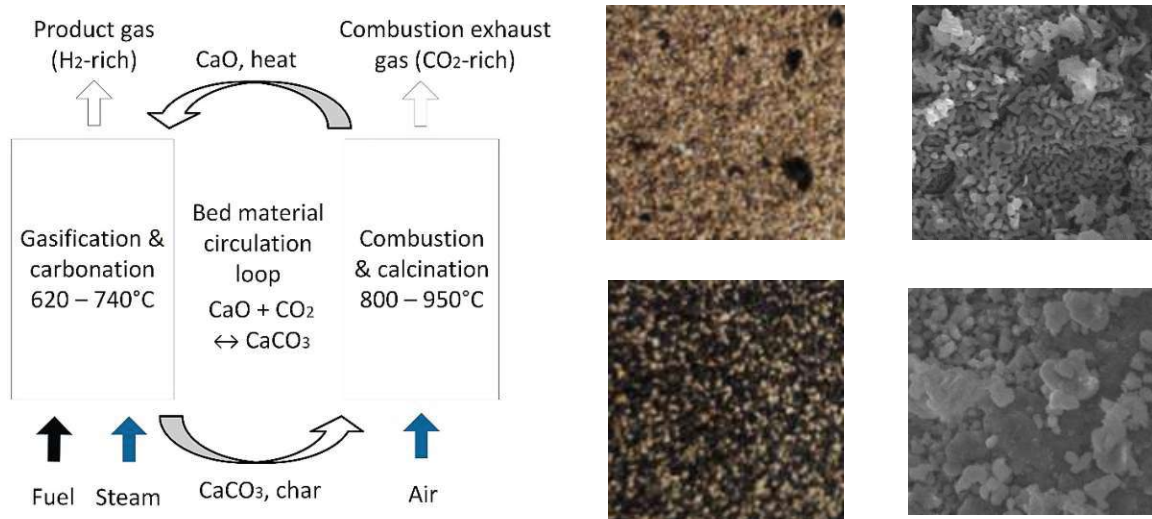


Figure 35: The basic principle of sorption enhanced reforming (left); bed material samples upper & lower loop seal (middle)^{4,5} and scanning electron micrographs after one cycle and 22 hours in a dual fluidized bed gasifier (right)⁶

1 Scala F., 2000; 2 Müller S., 2013; 3 Müller S., 2012
4 cf. Figure 20; 5 Fuchs J., 2019; 6 Fuchs J., 2019a

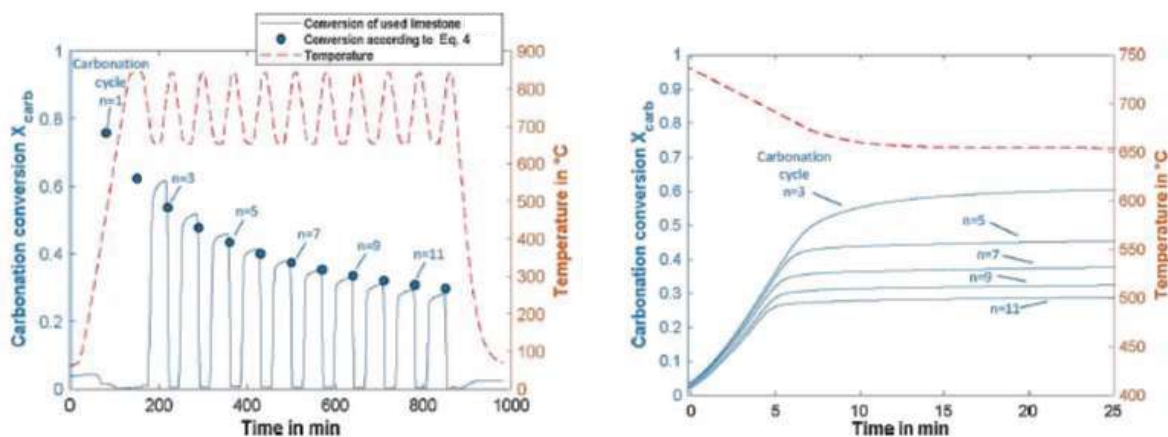


Figure 36: Decreasing carbonation conversion (X_{carb}) during alternating carbonation and calcination¹

After 12 carbonation cycles, only about 35% of the initial CO_2 load can be taken up on the operated calcium particle. This represents a relevant technical boundary for the removal of CO_2 from the product gas stream. Furthermore, it leads to the conclusion that the achievable product gas composition depends on the maintained reaction kinetics as well as the operated circulation rate² and the maintained CO_2 load. Figure 37 shows how the product gas composition changes as well as the decline in cold gas efficiency. The operated bed material circulating between the two reactors strongly influences the product gas composition as well as the efficiency of the overall process. Therefore, these relations have been investigated by several experimental campaigns. Table 12 shows a comparison of the main experimental results achieved with different designs of 100 kW gasification test plants at TU Wien. A description of the used test plants can be found in Chapter 3.5. The hydrogen content increases significantly due to the advanced reactor design as well as optimization of the used bed material and the operated residence time. Furthermore, the CO_2 content in the produced gas is very low.

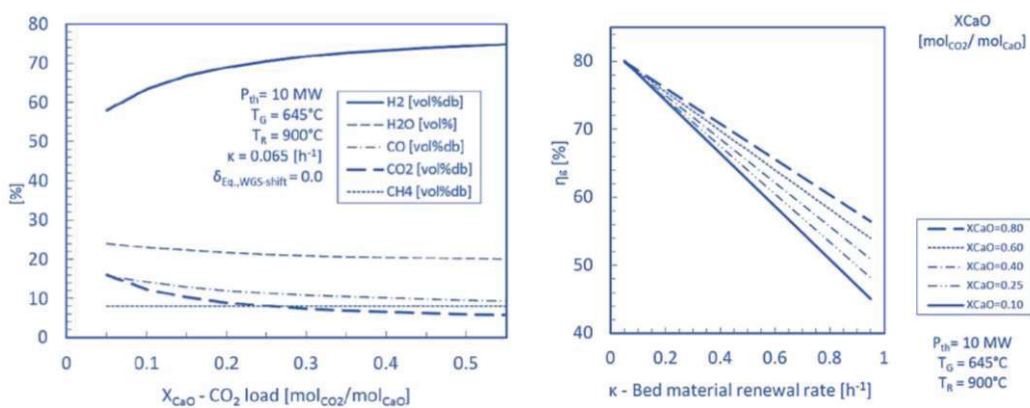


Figure 37: The dependence of product gas composition on the circulation rate (X_{CaO} , CO_2 load) and the decreasing cold gas efficiency based on increased demand for bed material renewal^{3,4}

1 Fuchs J., 2019b

2 Fuchs J., 2020

3 Fuchs J., 2017; 4 Müller S., 2013

Table 12: Comparison of experimental results from different experimental campaigns¹

| Parameter | Unit | 2012 ² | 2015 | 2013 ³ | 2016 | Observed values |
|------------------------------------------|----------------------|---------------------|-----------------------|-------------------|------------------|---------------------|
| Reactor design | - | Classical | Advanced | Classical | Advanced | Varying |
| Bed material type | - | Olivine | Olivine & 10% calcite | Calcite | Calcite | Olivine and calcite |
| Fuel type | - | Wood | Wood | Wood | Wood | Wood |
| Fuel power to GR | kW | 97 | 95 | 73 | 110 | 70–120 |
| Add. fuel power to CR | kW | 23 | 51 | 7 | 4 | 0–50 |
| Steam-to-fuel ratio | kg/kg _{daf} | 0.8 | 0.9 | 0.9 | 0.8 | 0.7–1.0 |
| Gasification temperature | °C | 802 | 830 | 679 | 630 | 620–840 |
| Product gas | | | | | | |
| Hydrogen (H ₂) | vol.-% _{db} | 35.3 | 43.8 | 63.9 | 67.5 | 35–75 |
| Carbon monoxide (CO) | vol.-% _{db} | 29.1 | 22.3 | 8.9 | 8.4 | 5–30 |
| Carbon dioxide (CO ₂) | vol.-% _{db} | 17.3 | 20.2 | 11.0 | 5.4 | 4–25 |
| Methane (CH ₄) | vol.-% _{db} | 11.1 | 9.7 | 11.8 | 13.6 | 6–16 |
| Ethene (C ₂ H ₄) | vol.-% _{db} | 1.5–3.5 | 0.9 | 1.3 | 1.1 | 0.5–4 |
| Ethane (C ₂ H ₆) | vol.-% _{db} | 0.5–1.5 | 0.06 | 0.9 | 1.1 | 0.05–1.5 |
| Propane (C ₃ H ₈) | vol.-% _{db} | | 0 | 0.05 | 0.13 | 0.05–0.5 |
| Water (H ₂ O) | vol.-% | 40–55 | 36 | 50 | 41 | 30–50 |
| GCMS tar | g/Nm ³ | 11.7 | 4.5 | 5.3 | 4.8 ^x | 3–12 |
| Grav. tar | g/Nm ³ | 6.0 | 1.5 | 2.3 | 0.9 ^x | 2–6 |
| Lower heating value | MJ/Nm ³ | 14.0 | 11.6 | 13.6 | 14.7 | 10–15 |
| Product gas power | kW | 61 | 87 | 46 | 81 | 45–100 |
| Key figures | | | | | | |
| Cold gas efficiency | % | 63 | 92 | 63 | 74 | 60–95 |
| Overall cold gas effic.* | % | 51 | 60 | 58 | 71 | 50–80 |
| Water conversion rate | - | ~ 0.15 ^x | 0.28 | 0.25 | 0.36 | 0.1–0.4 |
| CO ₂ load | mol/mol | 0 | 0 | 0.06 | 0.13 | 0–0.2 |

* existing industrial plants currently ~ 70%

^x supplemented measurement

These experimental campaigns have identified a strong influence of gasification temperature and operated bed material circulation rate, enabling a significant change in the product gas composition. Figure 38 illustrates the described influence as a function of the operated gasification temperature with the gasification test plant at TU Wien. A significant variation in the hydrogen (H₂) to carbon monoxide (CO) ratio can be reached by modifying the gasification temperature as well as the operated bed material circulation rate. The results have allowed defining different operation modes:

- carbonation-dominated SER,
- water–gas shift–dominated SER and
- conventional dual fluidized bed steam gasification.

Equations 11 and 12 describe important relations to model SER with respect to the operated selective CO₂ transport as well as the pre-dominant water–gas shift reaction.³ At lower temperatures—between 575 and 700°C—the gas composition shows a relevant deviation from the thermodynamic equilibrium of the water–gas shift reaction.

1 Müller S., 2017; 2 Schmid J., 2012; 3 Müller S., 2013

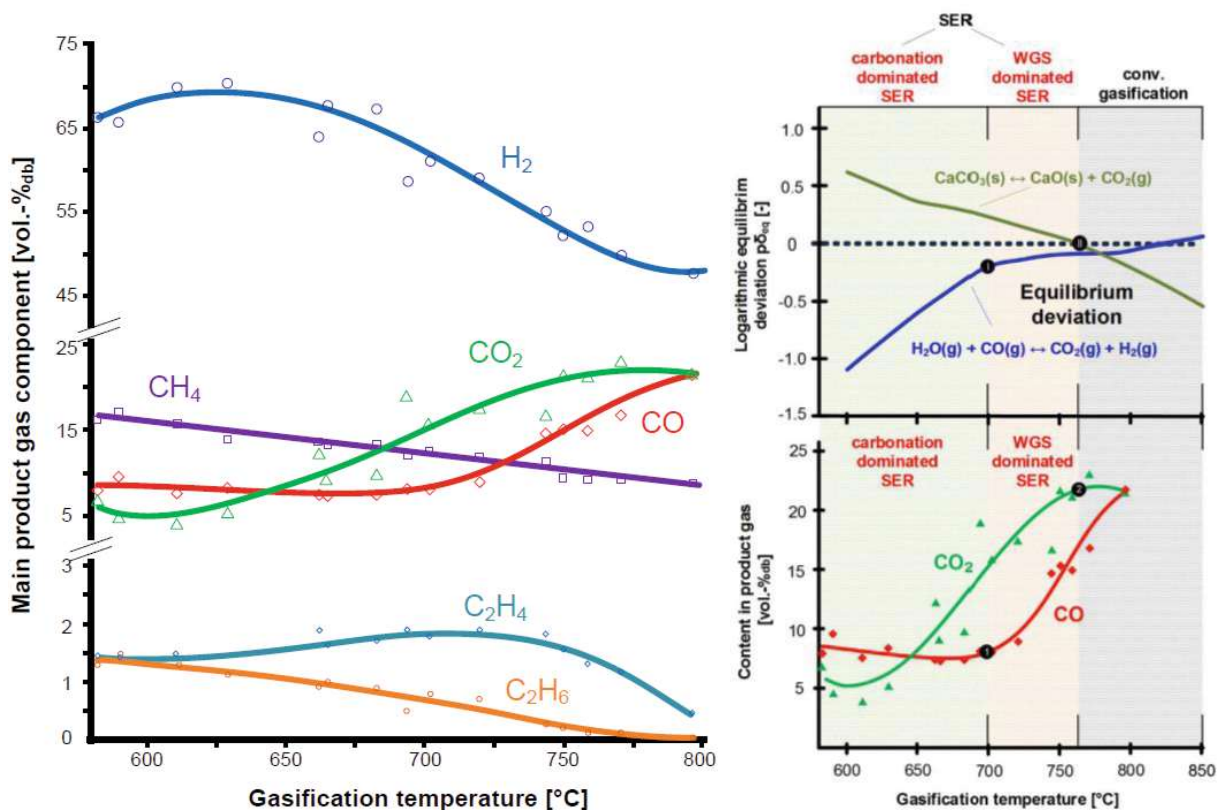


Figure 38: Variation in the gasification temperature during sorption enhanced reforming (left) and definition of different process operation modes (right)^{1,2}

$$\text{Eq. 11 } X_{\text{CaO}} = \frac{\dot{n}_{\text{CO}_2}}{\dot{n}_{\text{CaO}} + \dot{n}_{\text{CaCO}_3}}$$

$$\text{Eq. 12 } 10^{\delta_{\text{Eq.WGS-shift}}} \cdot K_p(T) = \frac{p_{\text{CO}_2} \cdot p_{\text{H}_2}}{p_{\text{CO}} \cdot p_{\text{H}_2\text{O}}} \quad \delta_{\text{Eq.WGS-shift}} = 0$$

Therefore, this sector has been named carbonation-dominated SER. Above 700°C, the gas composition shows good agreement with the thermodynamic equilibrium of the water-gas shift reaction. Above 760°C, the temperature is too high to favor the necessary process conditions for the carbonation reaction.

Finally, adaption of the gasification temperature as well as the operated circulation rate allows significant variation in the produced gas composition. This flexibility can be beneficial if different gas compositions are preferred for the operation of a subsequent synthesis process such as the production of liquid biofuels or SNG. Figure 39 shows the expected water conversion for large-scale operation of conventional hydrogen production compared with SER. The hydrogen mainly originates from fuel conversion. Besides, the conversion of water could be used to increase the extent of gaseous hydrogen that is received. SER offers the possibility for increased hydrogen production due to an increased water conversion rate. Therefore, this process could be a suitable solution for the reduction of fossil CO₂ emissions.

1 Fuchs J., 2018

2 Fuchs J., 2019

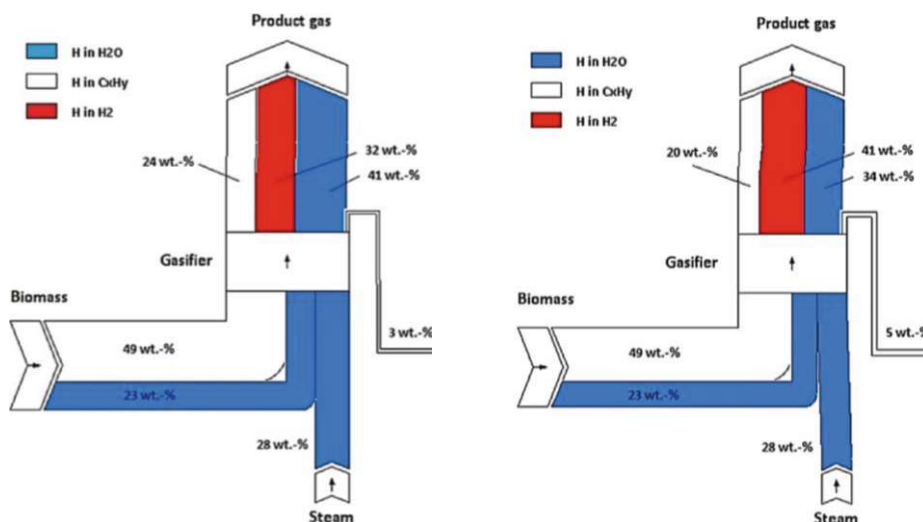


Figure 39: Expected water conversion for large-scale operation of conventional hydrogen production (left, $P_{th} = 50$ MW) compared to the SER (right, $P_{th} = 100$ MW)¹

A further modification of the process by applying oxyfuel combustion would allow the production of a sequestration-ready CO_2 stream. Oxyfuel combustion is carried out by using oxygen (O_2) instead of air as a combustion agent. Figure 40² shows the basic principle of oxy-SER. CCS technologies are used to dispose of fossil CO_2 emissions safely. Therefore, carbon capture within a dual fluidized bed gasification plant could be used to enable additional reduction in CO_2 emissions. This would lead to additional improvement of plant emissions in terms of the European emission allowances trading scheme. The oxyfuel combustion process is currently considered one of the most economic carbon capture processes. For the combustion process, pure oxygen mixed with exhaust gas is used instead of air. In this way, the combustion process leads to a high CO_2 content in the exhaust gas stream and offers good characteristics for efficient disposal or further utilization.^{4,3} The process configurations offer an interesting potential for large-scale application. Within the present work, the idea of CCU is used to enable a reuse of carbon for the production of clean energy carriers. The presented experimental results have been validated by using simulation models and are employed within Chapter 5 for the calculation of large-scale application.

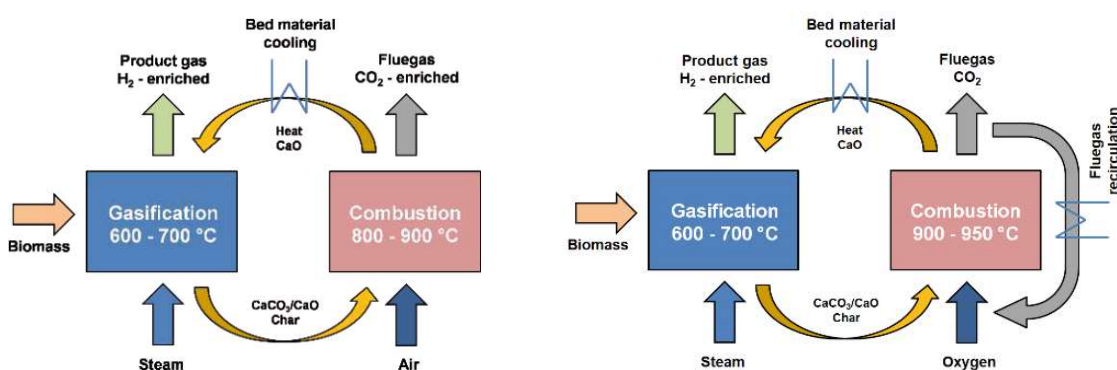


Figure 40: Basic principle of sorption enhanced reforming compared with oxyfuel sorption enhanced reforming.⁴

1 Müller S., 2013; 2 Hammerschmid H., 2016; 3 Scheffknecht G., 2011; 4 Hammerschmid M., 2019a

4.2. Carbon Dioxide Gasification

CO₂ gasification is another approach to reduce fossil CO₂ emissions. Figure 41 (left) shows the basic principle of dual fluidized bed CO₂ gasification.^{1,2} CO₂ is used in gasification to replace a relevant share of steam. CO₂ is converted into CO to allow further carbon utilization within subsequent synthesis processes. CO₂ gasification represents an option to apply CCU strategies. As a result, CO₂ is not seen as leftover emission; rather, it is seen as raw material—feedstock—for the production of valuable products. CO₂ gasification thus represents an important first building block to provide gas for subsequent utilization steps. The reverse water–gas shift reaction as well as the Boudouard reaction enable CO₂ reduction.³ Figure 41 (right) indicates the expected product gas composition if thermodynamic equilibrium of the water–gas shift reaction is reached. As can be seen, a significant decrease of the hydrogen (H₂) content and an increase in the CO content is expected. The existing advanced gasification test plant at TU Wien has been used to execute the first dual fluidized bed CO₂ gasification experiments. A detailed description of the gasification test plant can be found in Chapter 3.5. As a result, a significant change in the product gas composition has been observed. Table 13⁴ shows the results of five experimental campaigns using the fuel type softwood and olivine as bed material. As expected, the H₂ content within the product gas decreased, whereas the CO content increased significantly. Besides, there was a high share of CO₂ in the product gas if it was not converted into CO. Figure 42 shows the observed change in the product gas composition during the experimental campaign due to an increased use of CO₂ as gasification agent as well as increasing temperature. The collected data show that CO₂ gasification enables the production of an increased share of CO and serves as the basis for modelling a large-scale application. At the same time, the results also indicate that further optimization of the operated process is possible.

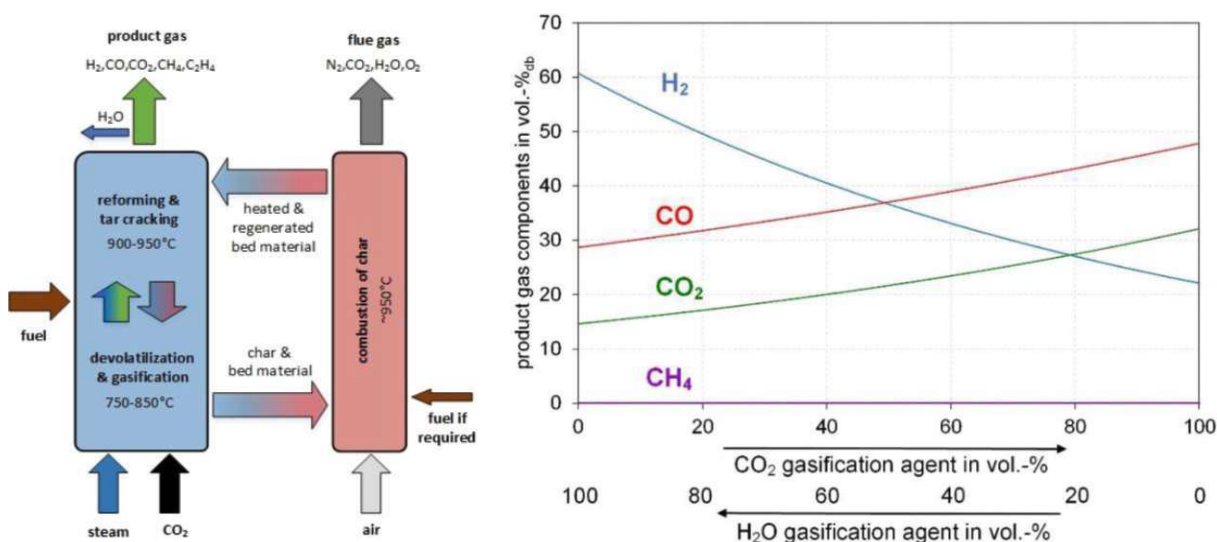


Figure 41: Basic principle of CO₂ gasification (left)¹ and product composition in equilibrium of the water–gas shift reaction (right)^{3–5}

1 Mauerhofer A.M., 2019; 2 Mauerhofer A.M., 2019a; 3 Mader R., 2018; 4 Mauerhofer A.M., 2019b; 5 Mauerhofer A.M., 2020

Table 13: Comparison of experimental results from different experimental campaigns

| Parameter | Unit | Exp. 1 | Exp. 2 | Exp. 3 | Exp. 4 | Exp. 5 | Observed values |
|------------------------------------------------------------------|----------------------------------------|-------------------|-------------------|-------------------|-------------------|--------|-----------------|
| CO ₂ /H ₂ O fluidization (ϕ_{CO_2}) | vol.-% | 0/100 | 32/68 | 45/55 | 68/32 | 100/0 | 0/100–100/0 |
| Fuel to GR | kW | 95 | 92 | 86 | 87 | 83 | 80–100 |
| Fuel to CR | kW | 68 | 59 | 53 | 53 | 56 | 50–70 |
| Steam-to-fuel ratio | kgH ₂ O/kg _{fuel} | 0.8 | 0.6 | 0.5 | 0.3 | 0.1 | 0.1–0.8 |
| CO ₂ -to-fuel ratio | kgCO ₂ /kg _{fuel} | 0.0 | 0.5 | 0.7 | 1.1 | 2.3 | 0–2.3 |
| T GR _{lower} | °C | 827 | 833 | 838 | 838 | 837 | 820–840 |
| T GR _{upper} | °C | 935 | 936 | 938 | 934 | 947 | 925–950 |
| T CR _{outlet} | °C | 947 | 944 | 944 | 941 | 964 | 940–965 |
| Hydrogen (H ₂) | vol.-% _{db} | 38.8 | 29.1 | 25.8 | 19.4 | 13.9 | 13–39 |
| Carbon monoxide (CO) | vol.-% _{db} | 25.9 | 27.9 | 29.9 | 31.1 | 38.3 | 25–39 |
| Carbon dioxide (CO ₂) | vol.-% _{db} | 18.6 | 25.9 | 30.9 | 35.6 | 38.9 | 18–39 |
| Methane (CH ₄) | vol.-% _{db} | 10.9 | 9.9 | 9.0 | 7.1 | 6.6 | 6–11 |
| Ethylene (C ₂ H ₄) | vol.-% _{db} | 2.1 | 1.6 | 1.27 | 1.11 | 1.06 | 1–2 |
| Ethane (C ₂ H ₆) | vol.-% _{db} | 0.15 | 0.10 | 0.08 | 0.06 | 0.02 | 0.01–0.15 |
| Water (H ₂ O) | vol.-% _{db} | 34.6 | 28.4 ^d | 29.7 ^d | 23.9 ^d | 7.9 | 7–35 |
| GCMS tar content ^b | g/Nm ³ | 11.2 ^c | nm | nm | 8.9 | 12.4 | 9–12 |
| Grav. Tar content ^b | g/Nm ³ | 6.7 ^c | nm | nm | 4.1 | 6.2 | 4–7 |
| Lower heating value ^a | MJ/Nm ³ | 12.7 | 11.2 | 10.6 | 9.2 | 8.7 | 8–13 |
| Product gas power | kW | 91 | 80 | 70 | 66 | 84 | 70–91 |
| Cold gas efficiency | % | 96 | 87 | 80 | 76 | 100 | 76–100 |
| Overall cold gas efficiency | % | 72 | 70 | 67 | 66 | 72 | 66–72 |
| CO ₂ conversion rate | kg _{CO2} /kg _{CO2} | 0 | -0.25 | -0.05 | 0.09 | 0.29 | -0.25–0.3 |
| H ₂ O conversion rate | kg _{H2O} /kg _{steam} | 0.28 | 0.18 | 0.06 | -0.16 | -0.54 | -0.55–0.3 |
| H ₂ /CO ratio | - | 1.49 | 1.04 | 0.86 | 0.63 | 0.39 | 0.4–1.5 |

^a free of tar and char; ^b measured by the Accredited and Notified Testing Laboratory for Combustion Systems at TU Wien

^c values from another comparable test run with soft wood as fuel and olivine as bed material

^d calculated based on mass and energy balances; nm not measured

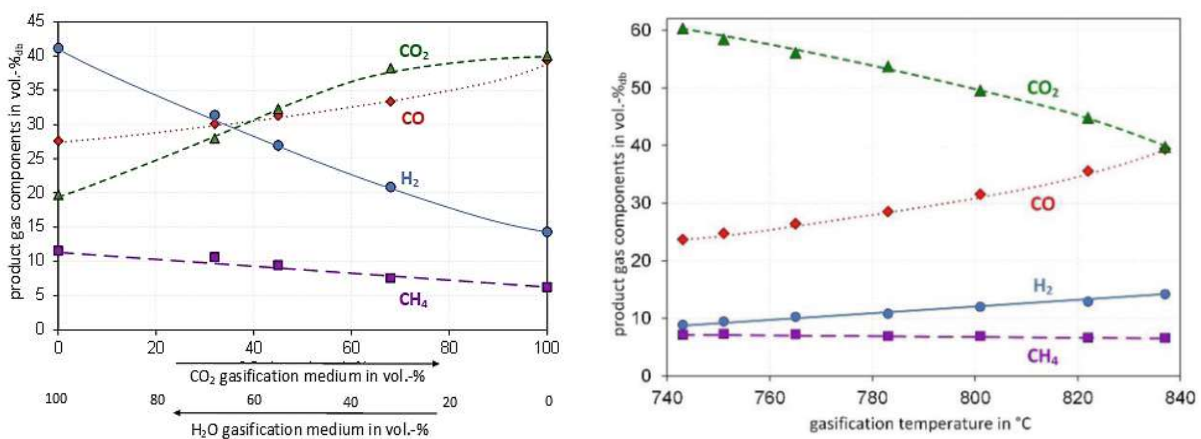


Figure 42: Change in product composition using CO₂ as a gasification agent (left) and increasing the gasification temperature (right)^{1,2}

1 Mauerhofer A.M., 2019b

2 Mauerhofer A.M., 2020

The operation of the used test plant (cf. Chapter 3.5) has been limited to temperatures of about 840°C, which is not sufficient to promote CO₂ reduction by using the Boudouard reaction. At the same time, the water–gas shift reaction enables the reduction of CO₂ to CO. Figure 43 illustrates the observed CO₂ reduction during the experimental campaign. Equations 13–14 show the definitions used to discuss the experimental results. The share of CO₂ in the gasification agent as well as the gasification temperature are the main influences on the observed carbon utilization. Optimizing the reactor system could improve the process operation. Higher temperatures—around 950°C—would be necessary to promote the Boudouard reaction and reach a CO content up to 80 vol.-%. Therefore, further improvements of the applied reactor concept are recommended. Overall, the applied process offers an interesting potential to enable a further reduction in fossil CO₂ emissions by utilizing it as syngas. This gas would allow the production of energy carriers such as SNG or Fischer–Tropsch liquids, as transportation fuels for ships or aviation. At the same time, it has to be acknowledged that additional hydrogen is necessary to ensure a sufficient H₂-to-CO ratio for most synthesis processes. The executed experimental campaign with respect to CO₂ gasification have been used to create validated test plant models. The collected data should allow analyzing the potential of the proposed concept at an industrial scale. Hence, the presented results represent crucial preliminary investigations for the realization of a CO₂ refinery for the production of different types of refinery products.⁴

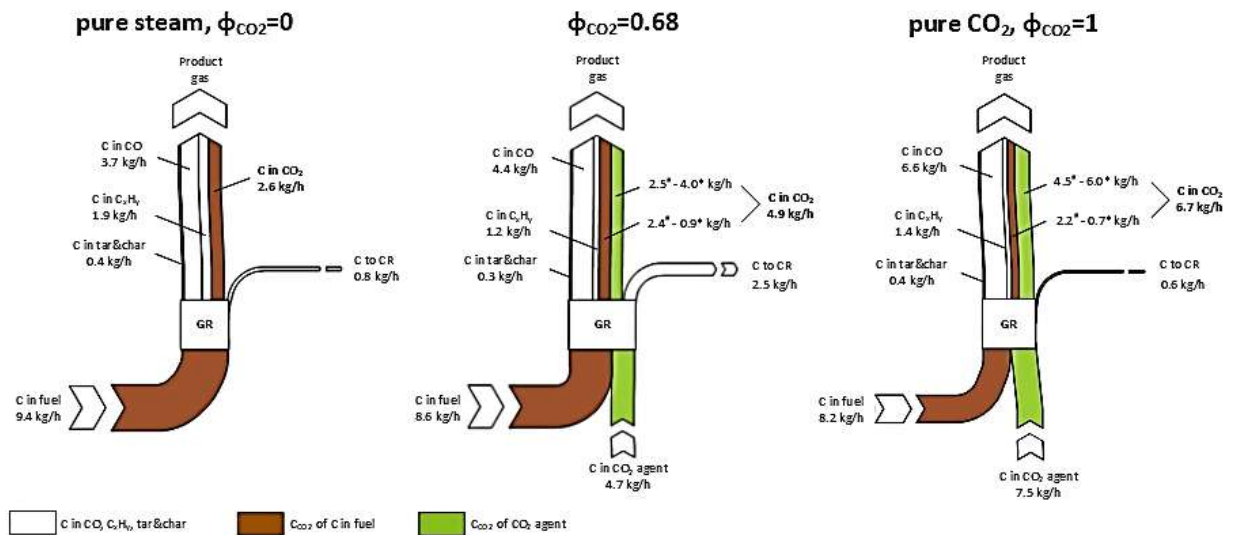


Figure 43: Experimental results with respect to CO₂ conversion Experiment 1, Experiment 3 and Experiment 5¹⁻³

$$\text{Eq. 13} \quad \phi_{CO_2} = \frac{X_{CO_2,fluid}}{X_{CO_2,fluid} + X_{H_2O,fluid}} \quad [\text{kg/kg}]$$

$$\text{Eq. 14} \quad X_C = \frac{X_{C,PG} \cdot \dot{m}_{PG}}{X_{C,fuel} \cdot \dot{m}_{fuel,db} + X_{C,CO_2 fluid} \cdot \dot{m}_{CO_2,fluid}} \quad [-]$$

$$\text{Eq. 15} \quad X_{C,FT} = \frac{X_{C,FT liquid} \cdot \dot{m}_{FT liquid}}{X_{C,fuel} \cdot \dot{m}_{fuel,db} + X_{C,CO_2 fluid} \cdot \dot{m}_{CO_2,fluid}} \times 100 \quad [\%]$$

1 Mauerhofer A.M., 2019b; 2 Mauerhofer A.M., 2020; 3 Mauerhofer A.M., 2021
4 <https://www.vt.tuwien.ac.at/co2refinery/> (accessed on March 27, 2021)

4.3. Summary

Chapter 4 has presented the advancement with respect to the development of processes to reduce fossil CO₂ emissions. The results have been reached by executing experimental campaigns accompanied by different levels of modelling. Consequently, there have been deep insights into important aspects of SER as well as CO₂ gasification. The following conclusions can be drawn.

- **SER** allows the production of a H₂-rich gas including selective transport of CO₂ to a separate flue gas stream. Based on the executed campaigns, the produced gas shows favorable characteristics for utilization in the context of the steel industry as **reduction agent** or for the **production of SNG**. The gas composition could be changed by altering the operated bed material circulation rate and the gasification temperature. The results achieved represent a sufficient basis to prepare an industrial-scale.
- **Oxyfuel combustion** represents an interesting option to generate a CO₂-rich flue gas stream with an **adequate gas quality** to utilize CO₂ as feedstock for the production of different types of products. Hence, it is also a valid option for the supply of CO₂ as feedstock for a **CO₂ refinery**.
- **CO₂ gasification** enables utilizing CO₂ to produce CO. This gas can be employed as feedstock for subsequent synthesis. The initial experimental campaigns have indicated that **CO₂ gasification** could be a valid technological option for syngas preconditioning. During the experimental campaigns, conversion of CO₂ into CO has been observed. The observed **consumption of CO₂** could be suitable to recycle CO₂ as CO for subsequent synthesis steps.

Overall, the investigated processes represent valid options for further implementation steps. The implementation should be accompanied by adequate modelling steps following the idea of MRL to ensure an overall successful implementation at an industrial scale.

5. INDUSTRIAL PLANT MODEL DEVELOPMENT

The ambitious aims of the current energy policy in Europe have led to a high demand for a methodology ensuring target-oriented implementation of existing solutions. Negative examples from the past have shown insufficient use of financial and natural resources with unsatisfactory outcomes. Therefore, the present work contains a proposal for the implementation of a unified methodology to support the implementation of novel energy technology into our existing energy system. Figure 44 shows once more the proposed concept, including technology evaluation supported by modelling-based development. The definition of adequate development goals at the beginning represents a critical moment. Chapter 3.1 describes a solution pathway to define well-balanced development goals. After a successful demonstration within an experimental environment, moving to an industrial scale represents a further critical phase of technology implementation. The present work recommends the introduction of IPIM to enable evaluation and prediction of large-scale operation before the erection of a demonstration project. Chapter 5 summarizes practical examples from modelling-based development at TU Wien aiming at the best-possible preparation of novel processes for future implementation. The presented examples are in line with the Austrian energy policy aiming at replacement of fossil carbon for the production of electricity and heat, the implementation of advanced biofuels as well as increased utilization of “green gas.” Furthermore, they follow the aim to reduce fossil CO₂ emissions supported by advanced digital methods. The illustrated examples contain process development work for the production of gaseous as well as liquid energy carriers such as SNG or liquid biofuels.

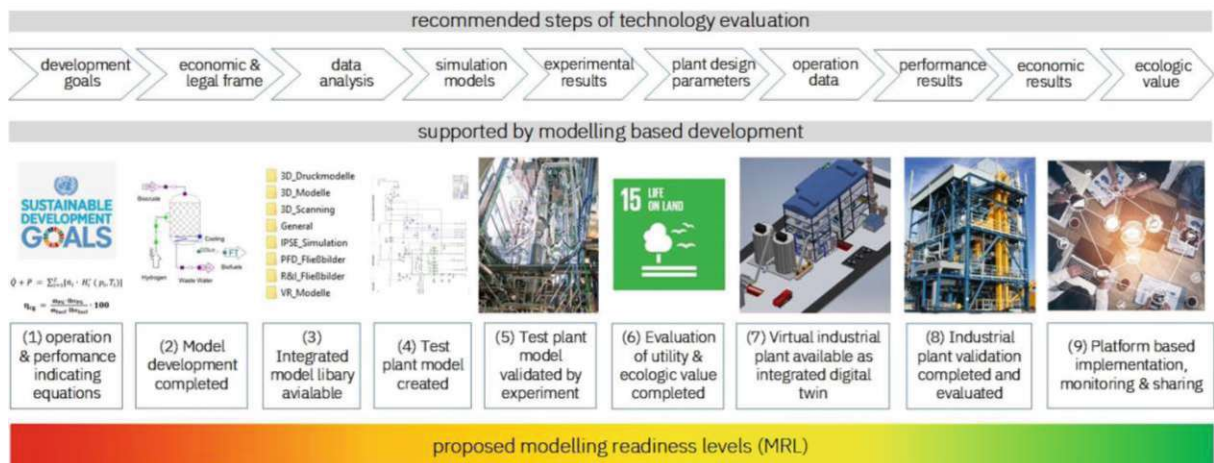


Figure 44: Application of the advanced digital methods concept by using modelling readiness levels¹

1 cf. Chapter 3

The mentioned energy carriers form the basis of our current energy system and therefore represent important standards regarding the development of a climate-neutral energy system. In addition, hydrogen is being discussed as an important energy carrier for the future. The generation of IPIM based on experimental results and related measurement data is used in this chapter to analyze the expected impact of a large-scale implementation. The approach to realize low-emission energy technology is presented based on the progress that has been reached in the field of thermochemical conversion of solid fuels such as biogenic feedstock as well as achieved advances in the field of process development described in [Chapter 4](#). This approach is used to investigate four application cases:

- the production of hydrogen from biomass,
- the production of SNG from biogenic feedstock,
- the production of liquid biofuels from biogenic feedstock and CO₂ and
- an analysis of an integrated case study in the context of an energy-intensive industry.

The illustrated cases acknowledge the achieved advancements:

- fuel flexibility in the field of gas production,
- sorption enhanced reforming,
- CO₂ gasification and
- the ongoing developed work for CCU.

Each investigation includes:

- a description of the initial situation,
- a description of the chosen process design,
- simulation models of large-scale plants and
- an evaluation of the results achieved.

As a result, the analyzed examples provide information to a following evaluation with respect to:

- the predicted reduction in fossil CO₂ emissions,
- their contribution to increase the ecologic value,
- important economic aspects and
- their readiness with respect to an industrial demonstration.

5.1. Industrial Hydrogen Production

Initial Situation

Hydrogen is often discussed as a potential future energy carrier for several applications.¹ Today, hydrogen is mainly produced from natural gas, which leads to relevant fossil CO₂ emissions. These fossil CO₂ emissions could be reduced if renewable energy resources could be used as feedstock.² Possible hydrogen production processes from renewable feedstock are still uncertain and need to be investigated to find applicable process routes.³ By using biogenic feedstock, an environmentally friendly hydrogen production route could be established. Figure 45 (left) shows a process design for the production of pure hydrogen from biomass via biomass steam gasification.

Process Design

The developed process design consists of a biomass gasification system, subsequent gas cleaning and hydrogen purification steps. The shown process design was evaluated by using process simulation software and these findings informed the erection of an experimental test plant. Figure 45 (right) shows the realized experimental facility. The created test plant layout consists of a dual fluidized bed gasifier including a gas cooler and a dust filter. Subsequently, a water–gas shift unit was installed to enhance the hydrogen yield and a biodiesel scrubber was used to remove tar and water from the syngas. CO₂ was removed and the gas was compressed to separate hydrogen via pressure swing adsorption. A steam reformer was used to reform the hydrocarbon-rich tail gas of the pressure swing adsorption and increase the hydrogen yield. Based on this work, a research facility was erected and the experimental results achieved were validated by using simulation software. Table 14 shows the reached hydrogen purity. This experiment has proved that a biomass-based hydrogen production process could be realized.

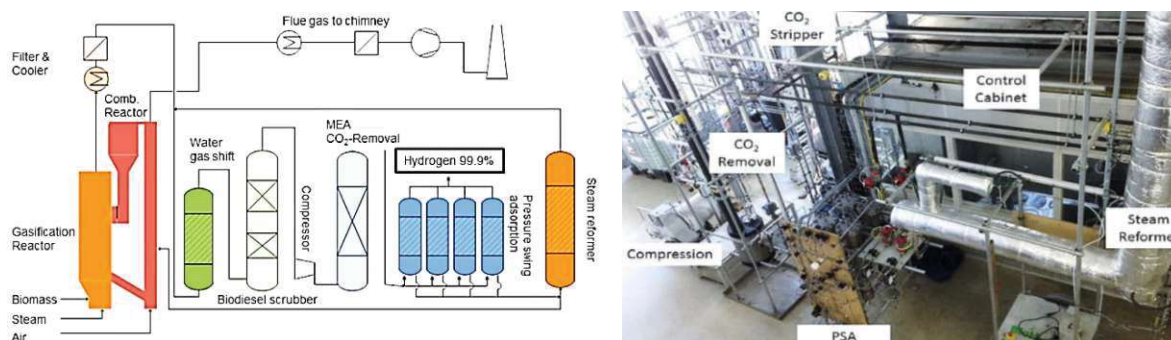


Figure 45: Hydrogen production via biomass gasification (left)^{2,4} & test plant for experimental validation [TRL 5, MRL 5] (right)⁴

1 European Commission, 2003

2 Müller S., 2013

3 <https://www.h2future-project.eu/> (accessed on March 27, 2021)

4 Loipersböck J., 2018

Table 14: Experimental results achieved¹

| Parameter | Unit | Raw gas from gasifier | Tail gas PSA | Hydrogen |
|-----------------------------------------------|----------------------|-----------------------|--------------|----------|
| Hydrogen (H ₂) | vol.-% _{db} | 39.7 | 28.9 | 99.99 |
| Carbon monoxide (CO) | vol.-% _{db} | 19.1 | 25.5 | 0.00 |
| Carbon dioxide (CO ₂) | vol.-% _{db} | 26.4 | 2.0 | 0.00 |
| Hydrocarbons (C _x H _y) | vol.-% _{db} | 14.3 | 39.5 | 0.00 |
| Nitrogen (N ₂) | vol.-% _{db} | 0.5 | 4.1 | 0.01 |

Industrial Plant Model

The experimental results were used to upscale the research plant to a 50 MW fuel feed scale aiming at MRL 6–7. Figures 46 and 47¹ show a simplified process flow diagram as well as the used simulation model. The maturity of the used models can be described as follows:

- the biomass gasification process including gas cooling and gas cleaning via a dust filter is based on validated commercial industrial plant models [MRL 8, TRL 9];
- calculation of the water–gas shift process, the scrubber, the pressure swing adsorption as well as the steam reformer was executed based on experimental data [MRL 5, TRL 5];
- calculation of the CO₂ removal unit was executed based on data from the literature and experiments [MRL 5, TRL 7];
- and further components have been modelled according to the manufacturers' specifications [MRL 9, TRL 9].

Validated models from commercial plants were available for the biomass gasification process itself. Most of the models used for the subsequent steps of hydrogen purification have been based on experimental results provided by the shown test plant. Moreover, the experimental validation of the investigated system indicated that a chemical efficiency of 60% could be reached.

Evaluation of Plant Operation Impact

Table 15 shows the simulation results achieved using the illustrated model shown in Figure 47. The industrial plant model enabled calculating a potential future large-scale operation. The mass and energy balance showed the predicted efficiency of a large-scale plant. Furthermore, important aspects regarding the realization of a large-scale demonstration plant have been identified. Energy and **material intensive gas cleaning methods need to be evaluated** by experimental tests. Moreover, CO₂ removal is an energy-intensive process, and the additional energy it consumes could be used for district heat instead. Therefore, **additional experimental investigations are recommended**, as an intermediate step, before building up a large-scale demonstration plant.^{2–4}

1 Müller S., 2013

2 Fail S., 2013

3 Loipersböck J., 2018

4 Fail S., 2014

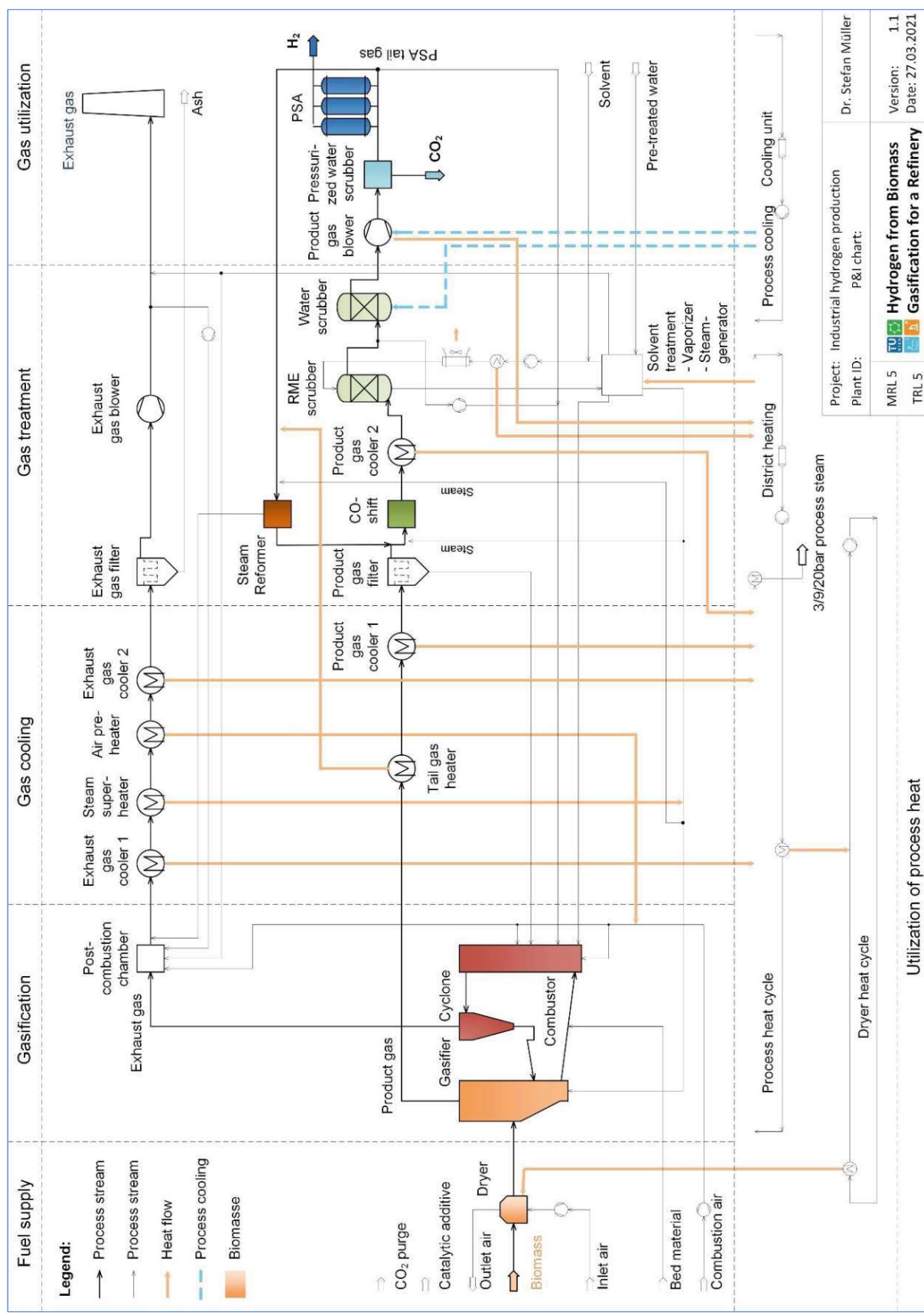


Figure 46: Process flow diagram of a large-scale hydrogen production based on experimental plant [MRL5, TRL 5]

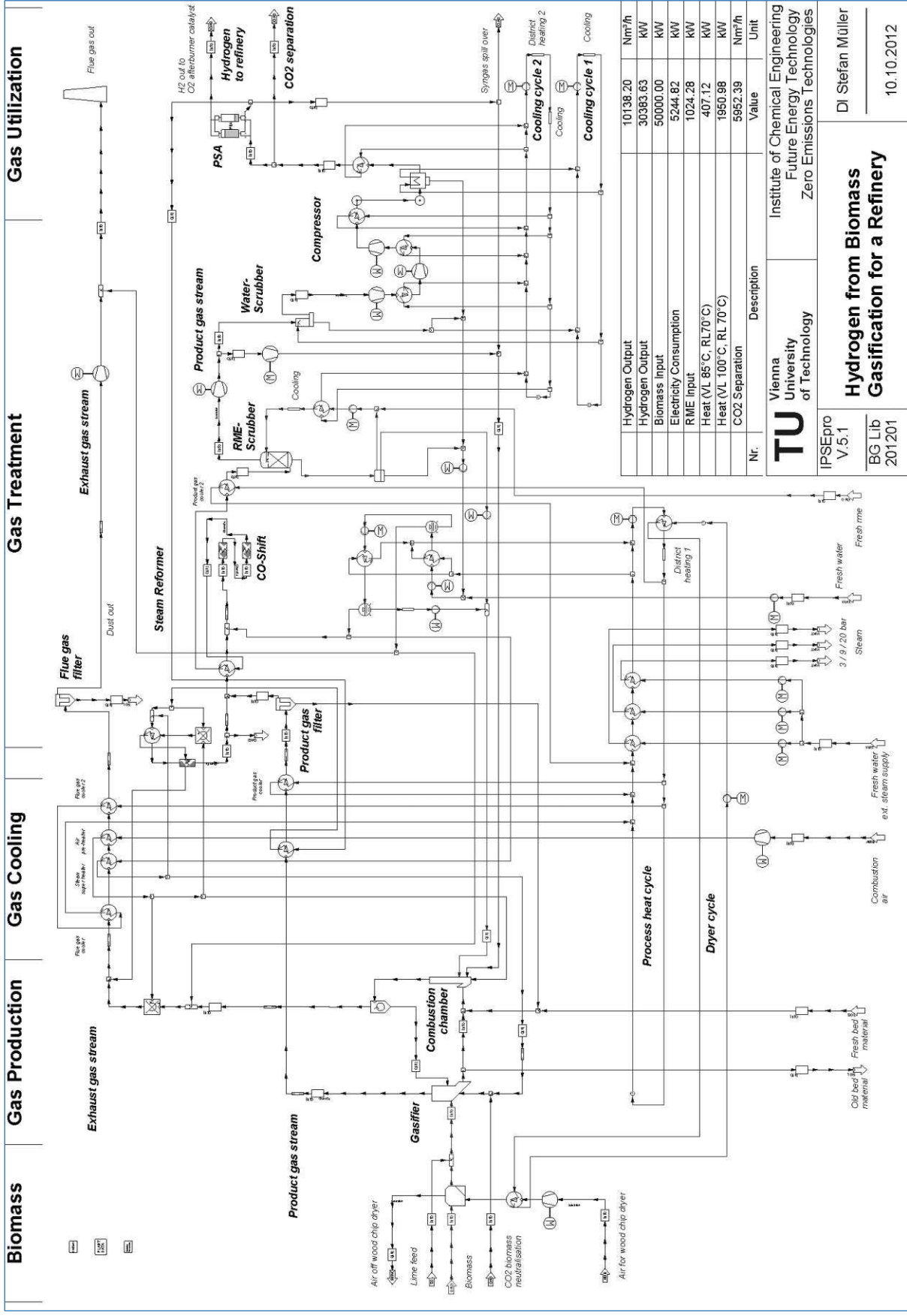


Figure 47: Industrial plant model of a large-scale hydrogen production plant aiming at MRL 6–7

Table 15: Simulation results⁴

| Plant input | | | Plant output | | |
|------------------------------------------|------|--------|---------------------------------------------------------------------------------------------------|--------------------|--------|
| Term | Unit | Value | Term | Unit | Value |
| Biomass (dry wood chips) | kg/h | 18 898 | Hydrogen | Nm ³ /h | 10 138 |
| Fresh water | kg/h | 2 965 | Steam | kg/h | 4 328 |
| and others | ... | ... | and others | - | ... |
| Biomass fuel power (P_{th}) | MW | 50 | Hydrogen (P_{H_2}) | MW | 30.4 |
| Electricity (P_{el}) | MW | 5.2 | District heating (\dot{Q}_{DH}) | MW | 2.3 |
| Key performance indicator | | | Development status: [MRL 5, TRL 5] | | |
| Chemical efficiency (P_{H_2}/P_{th}) | % | 60.8 | Development targets: fossil CO ₂ emissions: 0 t/h; and production costs: 60 EUR/MWh | | |
| Carbon utilization (X_c) | % | 0 | | | |

Besides, important aspects for a decision about further steps are still missing. An economic evaluation of the proposed concept showed that decentral production could be a feasible solution if hydrogen demand and storage ability exist at the desired location.^{1–3} At the same time, the lowest costs could be achieved by producing hydrogen from coal or natural gas. Ecologically, the investigated production route shows reasonable results. Of note, wind power electrolysis and production via steam reforming of biomethane achieve better results.^{2,3} Hence, [further modelling aims at identifying suitable application cases](#) for the best possible economic and ecologic implementation. Moreover, experimental development aims at continuous operation at a larger scale, ensuring reasonable resource consumption and reasonable operation costs. The described aspects are currently being investigated. Within the research project FCTrac, the production of pure hydrogen from biomass is realized. Therefore, the construction of the BioH₂ module is foreseen to provide pure hydrogen to a fuel cell–powered tractor via a simplified decentral biomass gasification concept.⁴ The foreseen demonstration is accompanied by related modelling steps aiming at MRL 8.

1 Müller S., 2013

2 Yao J., 2017

3 Wulf C., 2018

4 <https://projekte.ffg.at/projekt/3719251> (accessed on March 27, 2021)

5.2. Industrial Production of Synthetic Natural Gas

Initial Situation

Today, natural gas is one of the main energy carriers used to operate industrial processes in Europe. Natural gas imports are mainly from Norway, Russia and Algeria. The production of synthetic natural gas (SNG) from biogenic feedstock offers the possibility to reduce fossil CO₂ emissions. Thereby, SNG from biogenic residues offers a promising alternative to the utilization of fossil fuels and represents a novel concept to support the current energy strategy of the European Union.¹ The production of SNG from product gas of dual fluid gasification has been demonstrated at the SNG-plant in Güssing in 2009. To feed the generated gas into the Austrian and international gas grids, the feed-in regulations must be satisfied. The produced SNG met the specification of the Austrian natural gas grid and the operation of a compressed natural gas (CNG) car has been demonstrated.² These findings have encouraged the construction of a 32 MW gasifier for the production of 20 MW SNG in Goteborg, Sweden.³ Economic challenges have led to the stoppage of the plant operation. Therefore, many ongoing research activities aim at an optimized implementation of different types of renewable gas technology.⁴

Process Design

The reported results so far indicate that utilizing low-cost feedstock could support economic operation. Besides, fluidized bed methanation could reduce the complexity of the overall process chain. Therefore, the chosen process route consists of a fuel flexible gasifier to utilize biogenic residues or waste materials. Application of SER enables the production of a nitrogen-free product gas with an adjustable H₂-to-CO ratio. Before the product gas can be fed to a methanation unit, rigorous gas cleaning is required to protect the downstream equipment and the methanation catalyst. The applied gas cleaning strategy is essential if low-cost alternative feedstock is used. Figure 48 shows a basic flow chart of the proposed process chain⁵ and a picture of the installed test plant.⁶

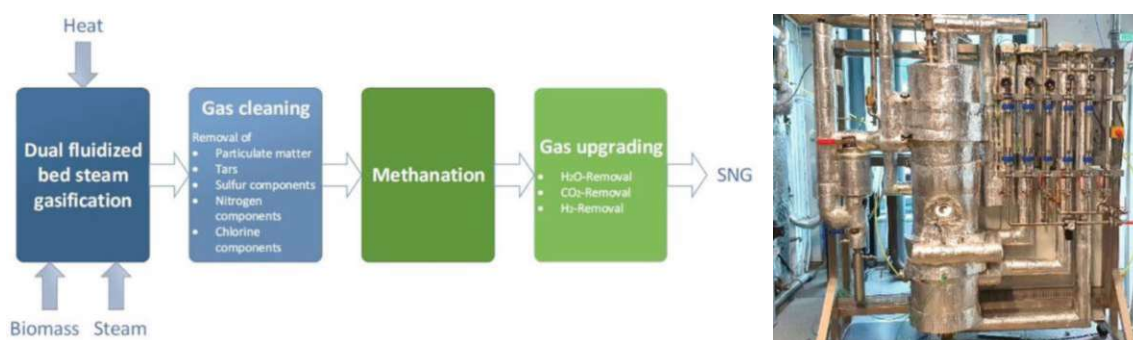


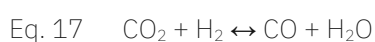
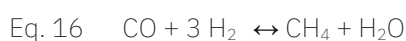
Figure 48: Basic flow chart of investigated synthetic natural gas (SNG) production route (left)⁶; a picture of the 10 kW methanation test plant [TRL 4] (right)⁷

1 Müller S., 2013; 2 Rehling B., 2011; 3 Thunman H., 2018

4 Kolb S., 2021; 5 Veress M., 2020; 6 Bartik A., 2021a

The operated gas cleaning strategy protects downstream equipment as well as the catalyst and ensures that the produced gas meets the required specifications of the operated gas grid. Consequently, the illustrated test plant was used to validate the process chain. The results achieved allowed prediction of how the operated plant would behave at a large scale. From a thermodynamic point of view, methanation is favored at low temperatures and high pressures. The main chemical species involved in the methanation reaction system are CH₄, H₂, CO, CO₂ and H₂O. The corresponding reaction equations are CO-methanation, the reverse water-gas shift reaction, and CO₂-methanation as shown in Equations 16, 17 and 18, respectively.^{1,2}

Additional experimental campaigns were executed at TU Wien to investigate ideal process conditions with respect to efficiency and carbon utilization. Besides, there was a special focus on the gas cleaning steps with respect to the utilization of cheap alternative feedstock. This endeavor has produced valuable additional data important to optimize the large-scale plant operation. Figure 49 gives insight into the results. Product gas from conventional gasification (DFB) and SER was converted in raw SNG with a high share of methane (CH₄). Of note, the applied gas cleaning chain showed satisfactory results.



Eq. 19 $\eta_{\text{overall}} = \frac{P_{\text{SNG}}}{P_{\text{th}}} \times 100$

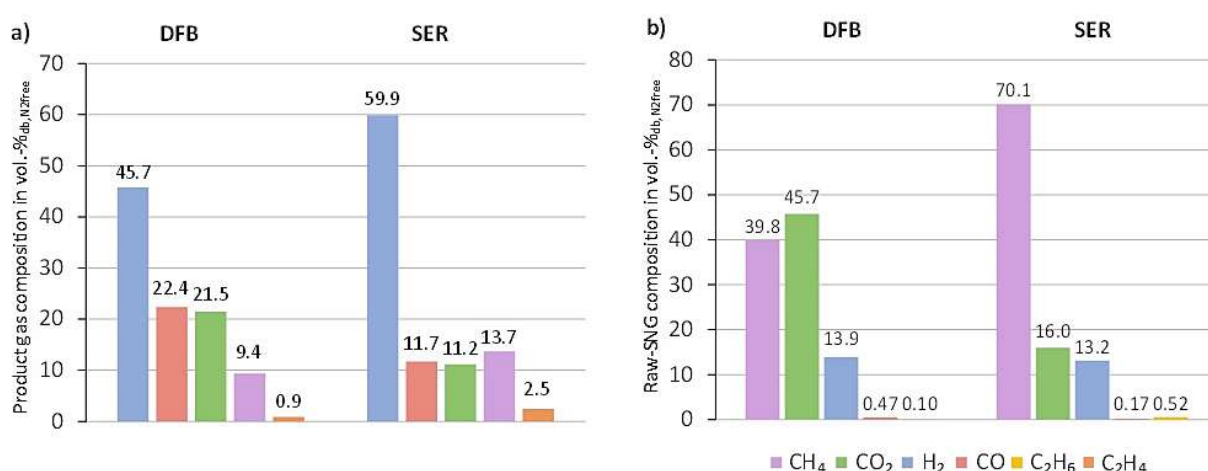


Figure 49: Product gas composition at the exit of the gasifier (left) and raw synthetic natural gas (SNG) composition at the exit of methanation unit (right)³

Experiences from the past encouraged the construction of a 32 MW gasifier for the production of 20 MW BioSNG in Goteborg, Sweden. Figure 50 shows pictures^{4,6} of the mentioned plants in Güssing and Goteborg. The experiences have enabled the creation of a process simulation model enabling optimization of an industrial plant concept.⁵

1 Bartik A., 2020

2 Bartik A., 2021;

3 Bartik A., 2021a; 4 Rehling B., 2011; 5 Teles M., 2017

6 <https://www.goteborgenergi.se/.../The GoBiGas Project>, technical report by Larson A. (accessed on January 7, 2022)

Industrial Plant Model

Figure 51 shows a process flow diagram of an optimized concept based on the reported experiences.¹ The shown concept includes:

- utilization of low-cost feedstock,
- a dual fluidized bed steam gasification unit operating SER,
- an advanced gas cleaning concept² and
- fluidized bed methanation enabling improved process heat management.

Figure 52 shows the created process simulation flow chart. The maturity of used models can be described as follows:

- the biomass gasification process of conventional wood including gas cooling and primary gas cleaning via dust filter is based on a validated commercial plants [MRL 9, TRL 9],
- calculation of gas cleaning steps if alternative biogenic feedstock is used was executed based on experimental data as well as data from different manufacturers [MRL 4, TRL 4],
- calculation of the methanation unit and gas upgrading was executed based on data from the literature and experiments [MRL 4, TRL 5],
- and additional components have been modelled according to manufacturers' specifications [MRL 9, TRL 9].

Overall, most of the used models have been based on reported experiences from different plants. At the same time, relevant measurement data of key components of the proposed concept are still missing; hence, improvements of the applied measurement methodology as well as additional experimental investigations are necessary.³ The reliability of the created industrial plant model could be enhanced in the future if validated data from additional experimental investigations are used. Indeed, within the research project ReGas4Industry additional experimental campaigns are being executed.⁴



Figure 50: A 1 MW demonstration plant, Güssing [TRL 5] (left); a 32 MW Plant, Goteborg [TRL 7] (right)

1 Veress M., 2019

2 Teles, M., 2017

3 Fuchs M., 2010

4 <https://energieforschung.at/projekt/gase-aus-regenerativen-reststoffquellen-fuer-die-industrie/> (accessed on March 27, 2021)

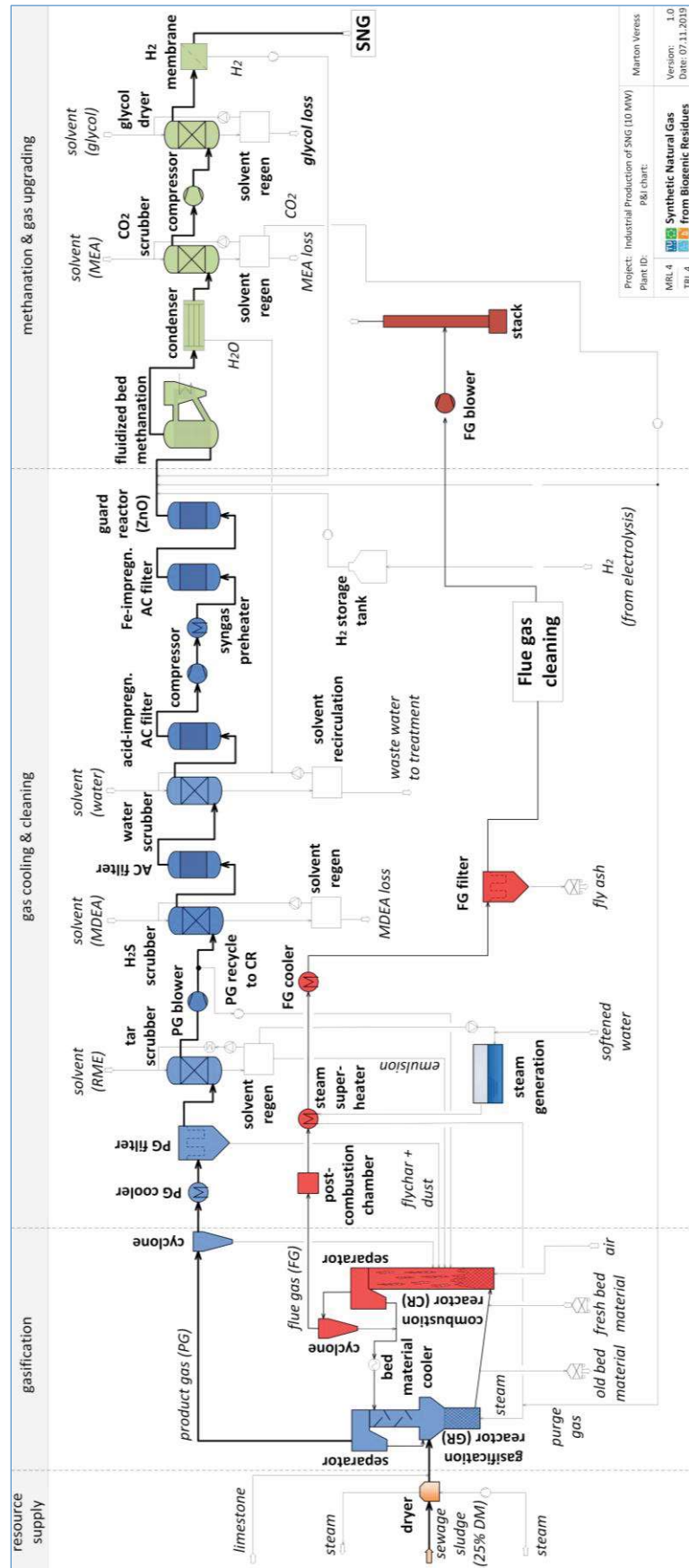


Figure 51: Process flow diagram of a 10 MW SNG plant [MRL 4, TRL 4] (Malicha M. 2018; Veress M. 2019)

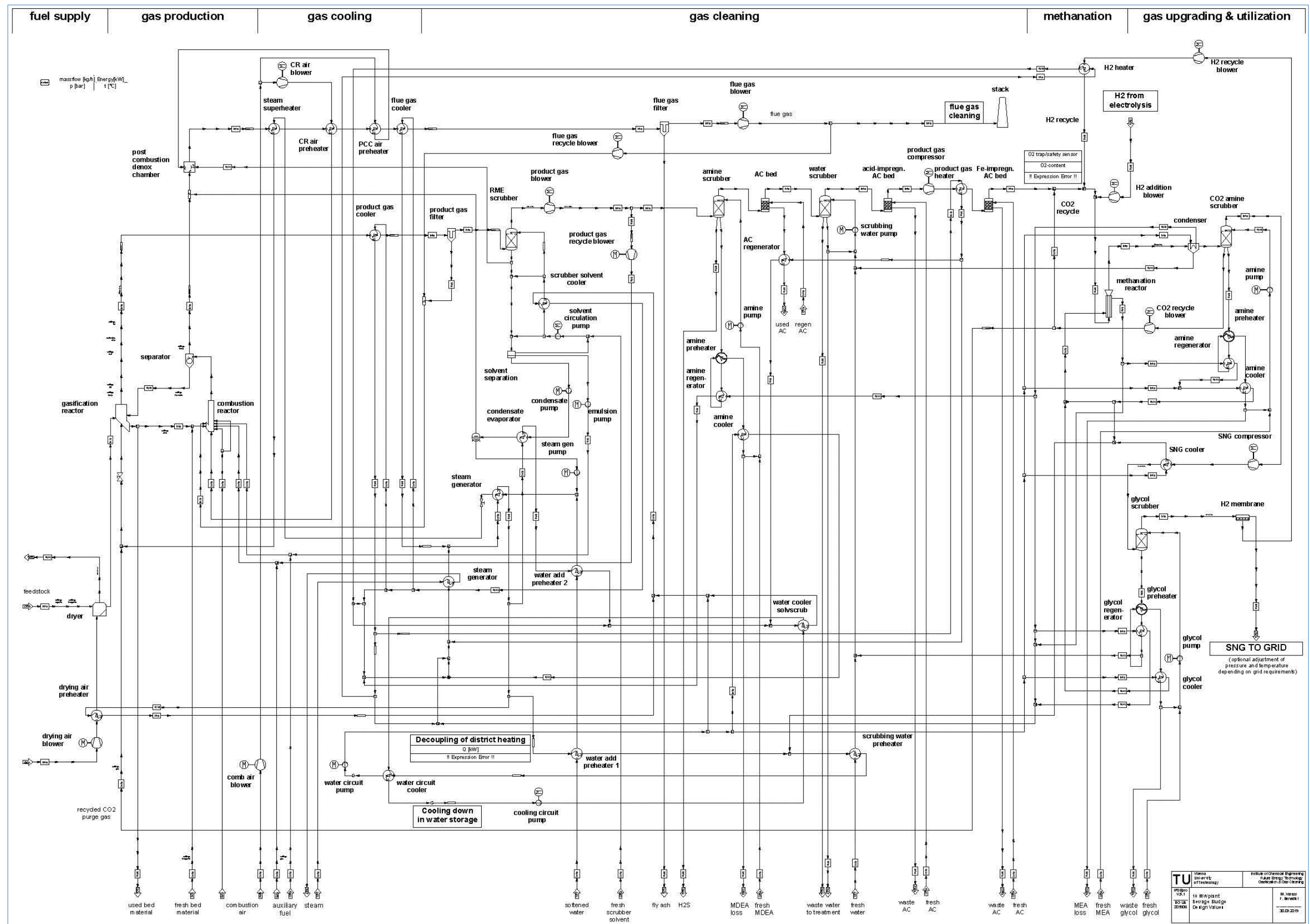


Figure 52: Industrial plant model of a 10 MW SNG plant aiming at MRL 6–7 (Veress M., 2020)

Evaluation of Plant Operation Impact

Table 16 summarizes important simulation results of the executed calculations using bark as feedstock. As can be seen, 10 MW of SNG can be produced from 9.3 MW biomass and 5.5 MW hydrogen. Besides, 0.7 MW electricity are needed mainly for the compression of the gas before the methanation step. In addition, 2.4 MW of sensible heat are available to cover the eventual nearby district heating demand.

Table 16: Simulation results^{1,2}

| Plant input | | | Plant output | | |
|--------------------------------------------------|------|-------|---------------------------------------------------------------|--------------------|-------|
| Term | Unit | Value | Term | Unit | Value |
| Biomass (sewage sludge) | kg/h | 2 570 | SNG product | Nm ³ /h | 1020 |
| Biomass fuel power (P_{th}) | MW | 9.3 | SNG product (P_{SNG}) | kW | 10 |
| Additional hydrogen (P_{H_2}) | MW | 5.5 | District heating (\dot{Q}_{DH}) | kW | 2.4 |
| and others | - | ... | and others | - | ... |
| Key performance indicator | | | Development status: [MRL 5, TRL 7] | | |
| Chemical efficiency ($P_{SNG}/P_{th}+P_{H_2}$) | % | 67.5 | Development targets: fossil CO ₂ emissions: 0 t/h; | | |
| Carbon utilization ($X_{C,SNG}$) | % | 40 | and production costs: 50 EUR/MWh | | |

The utilization of sewage sludge as feedstock would offer economic benefits. Successful experimental campaigns have proved that [utilizing sewage sludge is possible](#). At the same time, the [high share of ash leads to operational challenges](#). The large-scale demonstration of SNG production from wood pellets in Goteborg, Sweden, was impressive.³ At the same time, [important economic as well as technological challenges were not considered](#). Adequate modelling before implementation would be beneficial to the overall outcome of the executed development work. As a lesson learned, it can be mentioned that development work is an iterative process and it is recommended that modelling is always one step ahead of the next development step. Within the present work, additional experimental work as well as modelling work is described to improve the MRL of the proposed concept. Many questions remain and need to be clarified before further implementation steps. Therefore, the long term operation behavior of gas cleaning equipment and catalyst⁵ needs to be investigated. Furthermore, an optimization of the operational behavior of the overall process chain can be recommended. Modelling work should concentrate on open questions for the preparation of further demonstration steps such as long term behavior of sorption enhanced reforming, scale-up, ecologic evaluation of reduced CO₂ emissions and economic operation.² The research project Adore SNG includes further optimization of the overall process, aiming at adequate modelling of the process operation before further implementation steps. The mentioned project includes [experimental](#) as well as an [economic optimization](#). The experimental investigation combined with advanced digital methods should ensure a feasible improvement of the overall concept.^{2,4}

1 Veress M., 2020; 2 Bartik A., 2021b;

3 Thunman H., 2018; 4 <https://projekte.ffg.at/projekt/3862075> (accessed on September 27, 2021)

5 cf. Grundner S., 2015

5.3. Industrial Production of Biofuels

Initial Situation

Mobility and transportation make up the largest share of energy consumption within the European Union. The required crude oil for the production of motor fuels is mainly imported from Russia, Norway, Libya, Iran and Saudi Arabia.¹ Related production processes in refineries cause additional fossil CO₂ emissions. European emission reduction targets put pressure on refinery operators to decrease the emissions of the refining process. The Fischer–Tropsch process is one option to produce liquid biofuels. The mobility sector—heavy load transportation, shipping and aviation—has had a long-standing demand for liquid fuels. Since 2005, the Fischer–Tropsch process has been investigated by TU Wien by using laboratory-scale plants. The research activities have focused on the production of liquid biofuels from woody biomass, and the results of several experimental campaigns have been reported.^{2–5} The results have validated the used test plant models and have formed the basis for analyzing industrial-scale operation.⁶ The results have proved that Fischer–Tropsch fuel can be produced from wood chips and used as fuel for regular cars. Preliminary studies have already indicated that up to 0.55 MW of Fischer–Tropsch liquids can be produced from 1 MW of dry biomass depending on the applied production concept.^{2,5}

Process Design

Fischer–Tropsch synthesis has been well known for many years as a way to produce hydrocarbons from biomass or coal as feedstock. [Figure 54](#) shows a simplified process flow diagram of the experimental Fischer–Tropsch facility used for the present investigation. The Fischer–Tropsch reaction is a heterogeneously catalyzed polymerization reaction in which CO and H₂ are converted to hydrocarbons. The main products are *n*-paraffins and *n*-olefins. Furthermore, alcohols, ketones, aldehydes and *i*-paraffins are formed in small amounts. After synthesis, the products are separated into gasoline/naphtha (C₅–C₉), diesel (C₁₀–C₁₉), wax (C₂₀–C₆₀) and water fractions through distillation. After further purification and upgrading steps, high-quality naphtha, diesel, kerosene and biowaxes can be obtained. Fischer–Tropsch diesel contains very low amounts of sulfur, nitrogen and aromatics, and the CO₂, CO and soot emissions related to combustion are reduced significantly compared with conventional liquid fuels. Moreover, Fischer–Tropsch biofuel is regarded as an advanced second-generation biofuel, which uses non-food feedstocks and therefore is not involved with the fuel-versus-food debate. After an upgrading or hydrotreating step of the Fischer–Tropsch wax, naphtha, kerosene or diesel can be produced. Other application of Fischer–Tropsch waxes can be found in the pharmacy and personal care product sectors.^{4,7} [Figure 53](#) shows a picture of the Fischer–Tropsch-pilot plant operated in Güssing. [Figure 54](#) and [55](#) show an illustration of the created test plant model for the evaluation of experimental results.

1 Eurostat, 2011; 2 Sauciuc A., 2011; 3 Rauch R., 2013; 4 Maier L., 2014

5 Fürnsinn S., 2007; 6 Müller S., 2017; 7 Gruber H., 2020



Figure 53: Combined heat and power plant Güssing [TRL9, MRL9] (left); Fischer-Tropsch test plant [TRL 6, MRL 5]¹

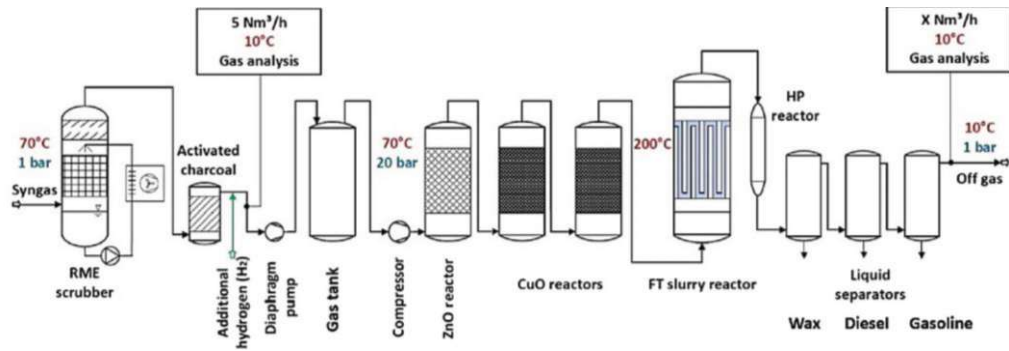


Figure 54: Simplified process flow chart of a laboratory-scale Fischer-Tropsch plant²

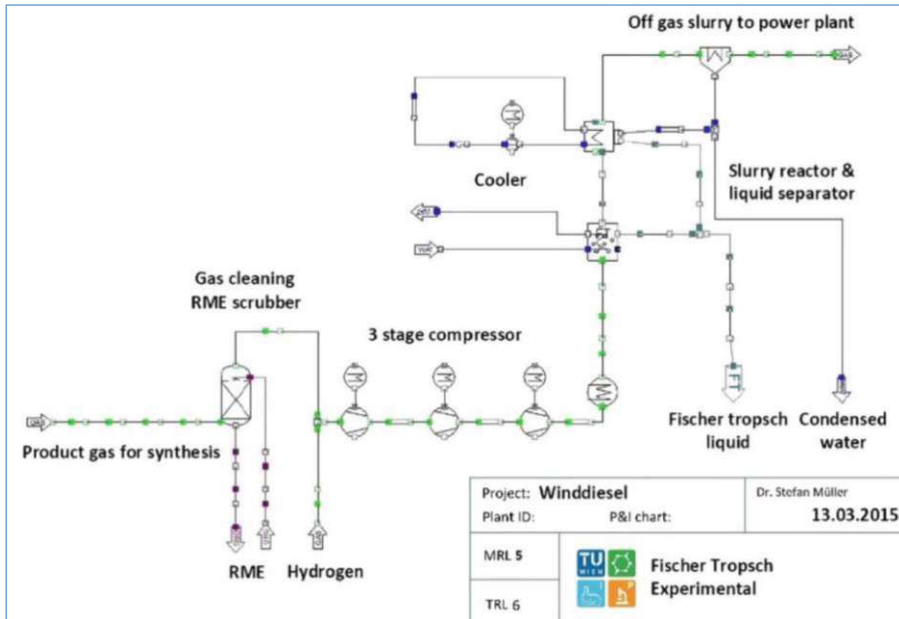
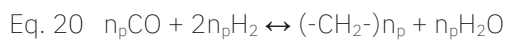


Figure 55: Created test plant model [MRL 5]²

1 Maier L., 2014
2 Müller S., 2017a

Equations 20 and 21 show the main relevant chemical reaction for the production of biofuel from CO and H₂ as well as the Anderson–Schulz–Flory distribution describing the received products.¹ Table 17 gives a short overview about relevant operation parameters and Table 18 about experimental data achieved.



$$\text{Eq. 21 } \log\left(\frac{W_m}{m}\right) = m \cdot \log \alpha + \log \frac{(1-\alpha)^2}{\alpha}$$

Table 17: Important operation parameters for the calculation of the Fischer–Tropsch process²

| Term | Unit | Value | Data source |
|-----------------------------|------|-------|-------------|
| Catalyst load | kg | 2 | Parameter |
| Temperature | °C | 200 | Parameter |
| Pressure | bar | 20 | Parameter |
| H ₂ -to-CO ratio | - | ~2:1 | Measurement |

Table 18: Experimental data²

| Term | Unit | Typical values | Validated measurement |
|----------------------------------------------------|--------------------|----------------|-----------------------|
| Mean gas volume flow | Nm ³ /h | 4–6 | 4.6 |
| Gasoline/naphtha (C ₅ –C ₉) | kg/h | 0.02–0.04 | 0.036 |
| Diesel (C ₁₀ –C ₁₉) | kg/h | 0.04–0.08 | 0.07 |
| Wax (C ₂₀ –C ₆₀) | kg/h | 0.04–0.15 | 0.13 |
| Condensed water | kg/h | 0.2–0.35 | 0.3 |
| Alpha (α) | - | 0.89–0.93 | 0.93 |
| Gas flow off gas | Nm ³ /h | 3.5–4.6 | 3.5 |
| CO conversion | % | 10–40 | 40 |

The executed experimental campaigns have enabled the creation of a validated test plant model. Validated measurements of important process data and resulting efficiencies represent an important basis for the creation of an industrial plant model aiming at MRL 6–7. Therefore, Equations 15 and 22 describe important performance indicating relations such as CO conversion as well as the chemical efficiency and the realized carbon utilization.^{2,3}

$$\text{Eq. 22 } X_{\text{CO}} = 1 - \frac{n_{\text{CO.out}}}{n_{\text{CO.in}}}$$

1 Patzlaff J., 1999

2 Müller S., 2017a;

3 Mauerhofer A., 2020

Industrial Plant Model

The above-mentioned results were used to calculate mass and energy balances of a large-scale plant. [Figure 56](#)¹ shows a process flow diagram of the investigated process route. The process route consists of a biomass gasification system operating CO₂ gasification attached to a Fischer–Tropsch synthesis and an electrolysis unit to provide sufficient amounts of hydrogen to the overall process. The created simulation model at an industrial scale as shown in [Figure 57](#).²

The maturity of used models can be described as follows:

- the biomass gasification process of biogenic residues by using CO₂ gasification has been calculated based on validated test plants [MRL 5, TRL 4],
- calculation of gas cleaning steps if alternative biogenic feedstock is used was executed based on experimental data [MRL 4, TRL 4],
- calculation of the Fischer–Tropsch unit was based on validated test plant data [MRL 5, TRL 6],
- calculation of the electrolysis unit was based on data from the literature [MRL 4, TRL 6]
- and additional components have been modelled according to the manufacturer's data [MRL 9, TRL 9].

Most of the used models have been based on validated test plant data from different experimental plants. At the same time, relevant measurement data of key components of the proposed concept are still missing and make an improvement of applied measurement methodology as well as additional experimental investigations necessary. Hence, analytical tools need to be applied to monitor the quality of different process stages. Besides, especially the final fuel upgrading represents an open issue that has to be clarified before further implementation steps. Currently, fuel upgrading is executed in existing refineries. No validated data was available at the time to model the electrolysis unit.

1 Mauerhofer A., 2020
2 Ebner J., 2018

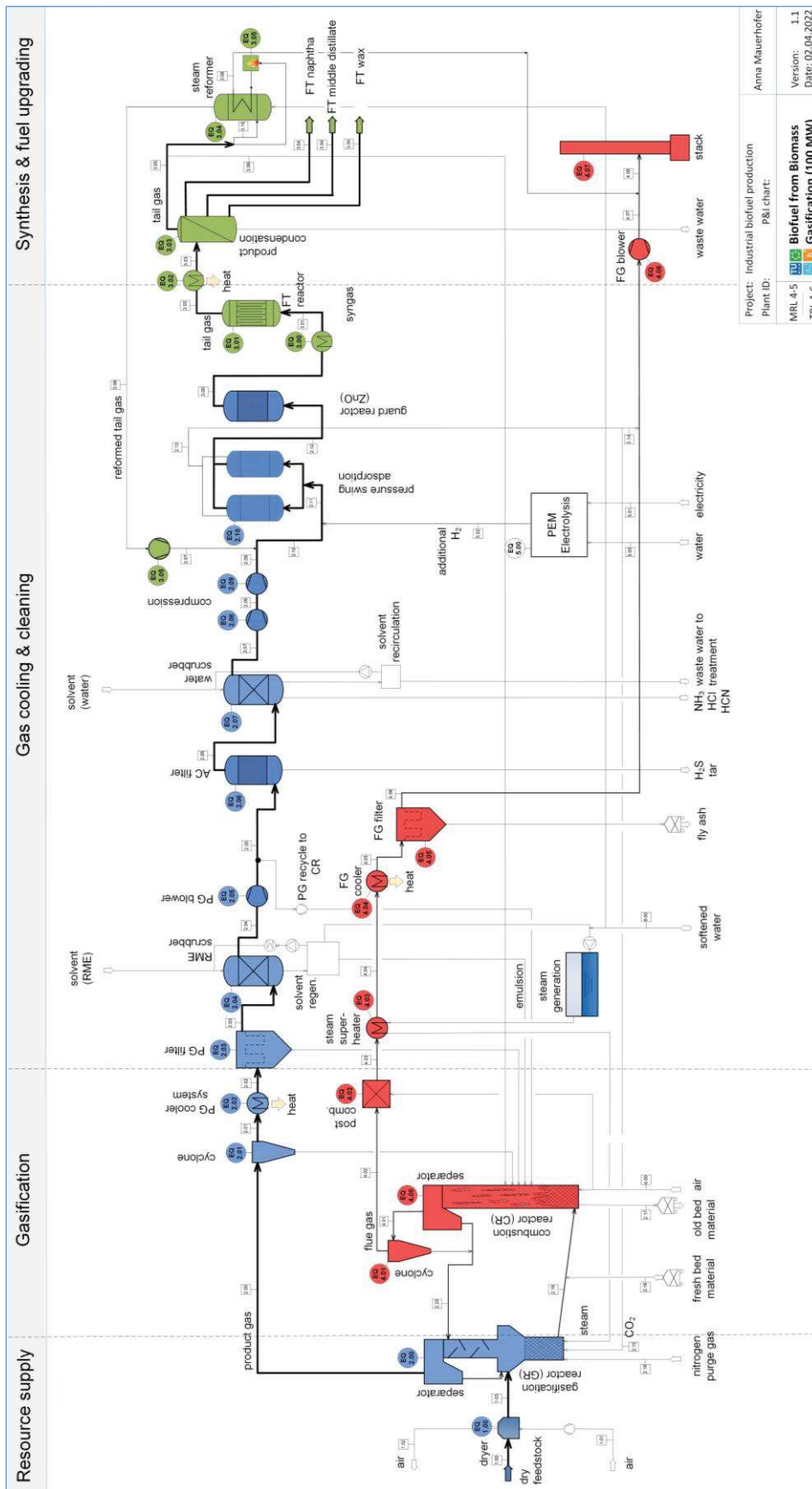


Figure 56: Process flow diagram of a biofuel production plant based on validated test plant data [MRL 4–5, TRL 4–6]

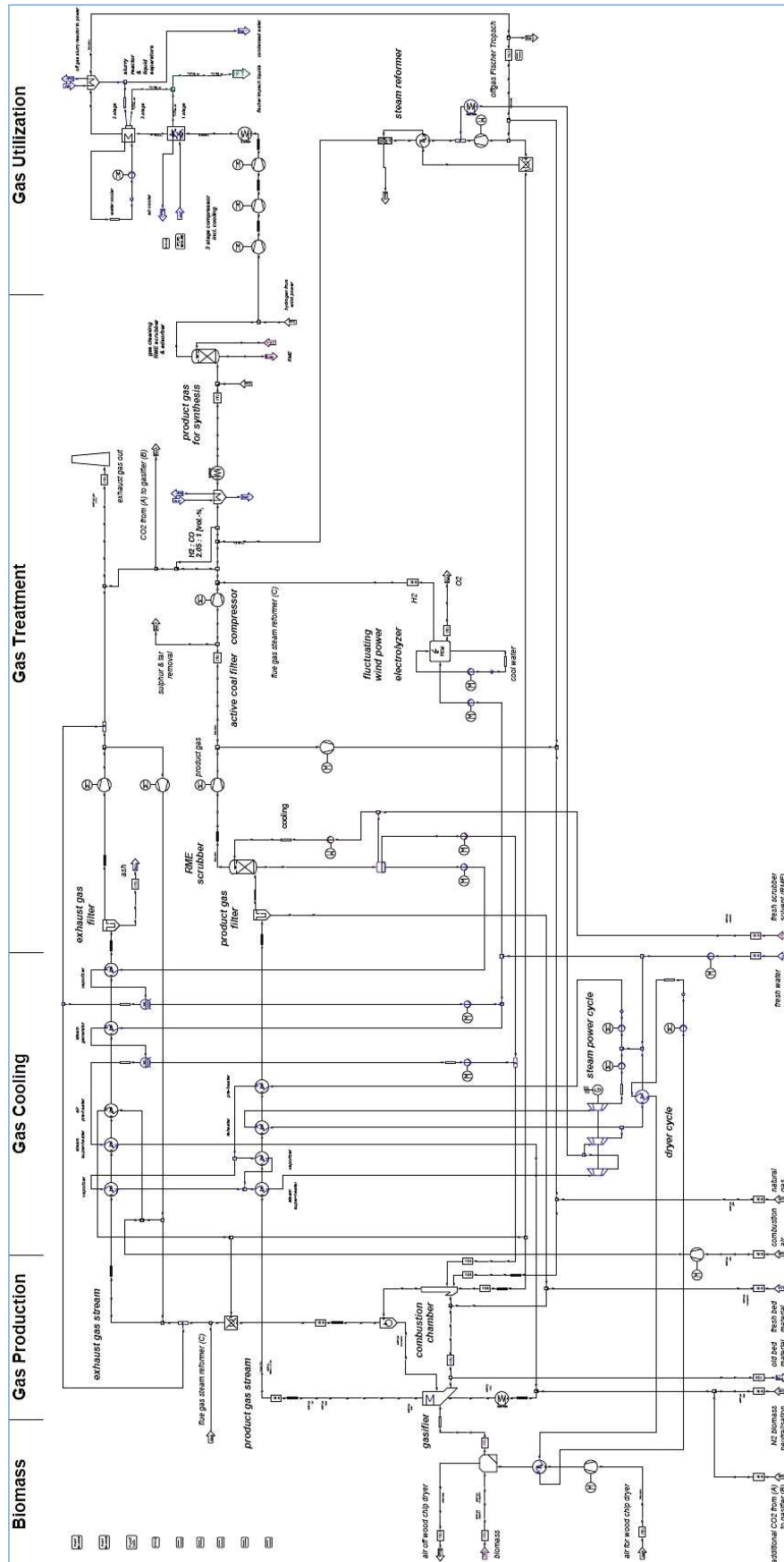


Figure 57: Simulation model of an industrial biofuel production plant aiming at MRL 6–7

Evaluation of Plant Operation Impact

Table 19 gives a short summary of the main calculation results, showing the expected performance of the investigated biofuel production as a basis for the comparison with other biofuel production routes. As can be seen, 51.8 MW of Fischer–Tropsch product can be produced from 100 MW Biomass. In addition, 22.4 MW of sensible heat are available to cover the eventual nearby district heating demand.

Table 19: Simulation results^{1,2}

| Plant input | | | Plant output | | |
|-----------------------------------------|------|--------|---------------------------------------------------------------------------------------------------|------|-------|
| Term | Unit | Value | Term | Unit | Value |
| Biomass | kg/h | 36 096 | Fischer–Tropsch product | kg/h | 4 235 |
| Biomass fuel power (P_{th}) | MW | 100 | Fischer–Tropsch product (P_{FT}) | MW | 51.8 |
| Electricity for electrolysis | MW | 0 | Diesel | kg/h | 1 538 |
| and others | - | ... | Gasoline | kg/h | 957 |
| Key performance indicator | | | Wax | kg/h | 1 740 |
| CO conversion (X_{CO}) | % | 80 | District heating (\dot{Q}_{DH}) | MW | 22.4 |
| Chemical efficiency (P_{FT}/P_{th}) | % | 51.8 | Development status: [MRL 5, TRL 5] | | |
| Carbon utilization ($X_{C,FT}$) | % | 50 | Development targets: fossil CO ₂ emissions: 0 t/h; and production costs: 70 EUR/MWh | | |

Overall, producing biofuels from biogenic feedstock offers the greatest potential to reduce fossil CO₂ emissions due to its importance for the transportation sector. Furthermore, the proposed concepts offer the potential to realize CO₂ recycling. Therefore, additional activities are being carried out. Within the research project Heat-to-Fuel, the described process route is benchmarked with an alternative process enabling biofuel production. The investigated process show a relatively high maturity for a second-generation biofuel production process.^{3–5} Subsequently, a techno-economic assessment by comparing fuel market prices and expected production costs of the different biofuel concepts revealed that reasonable production costs could be reached if the existing tax system would be modified. Besides, an increasing share of power production from sun and wind energy in Europe has led to increased interest in novel energy storage technologies. The production of hydrogen from electricity via electrolysis enables the conversion of electrical energy into chemical energy, which could be stored with a high energy density if further process steps are applied.⁶ Additional investigations have highlighted that a substantial part of liquid fuels used in agriculture could be substituted by liquid biofuels produced from wood.⁷ Furthermore, the development of sustainable aviation fuels represents an important development target in the field of biofuel production.^{8,9} This endeavor includes establishing analytical labs enabling the assessment of products achieved ensuring safe operation of aircraft turbines.

1 Ebner J., 2018; 2 Müller S., 2017a

3 Chiamonti D., 2007; 4 <https://www.heattofuel.eu> (accessed on September 27, 2021)

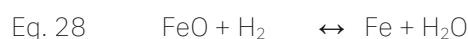
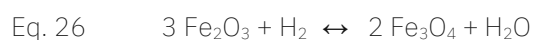
5 Mauerhofer A., 2020; 6 <http://www.winddiesel.at> (accessed on September 27, 2021)

7 Hofbauer H., 2020; 8 Chiamonti D., 2019; 9 Drünert S., 2020

5.4. Integrated Model Enabling Reduction of Carbon Dioxide Emissions of Hot Metal Production

Initial Situation

Iron and steel are important base materials for various industrial applications. Moreover, iron and steel production is one of the main industrial causes of fossil CO₂ emissions. A main share of these CO₂ emissions arises due to the energy demand and the chemical reduction of iron oxide (Fe_xO_y) to raw iron in a blast furnace at up to 2 000°C. Major reaction partners for the reduction of iron oxide (Fe_xO_y) are CO and H₂ provided by fuels like coal, natural gas, oil and plastic waste. Equations 23-28 illustrate the iron reduction process by using CO and H₂.^{1,2} Existing emission-reduction targets have forced the iron and steel industry in Europe to find new approaches to reduce their fossil CO₂ emissions. Therefore, several solution pathways are being investigated to reduce the related resource consumption. The replacement of fossil energy carriers should not impair the quality of the final products or lead to reduced availability of the production process. At worst, failing to implement solutions could lead to a transfer of iron production to regions outside of the European Union, where environmental regulations are not considered in an equal way. Therefore, numerous researchers investigate the replacement of fossil energy for iron production.²



Process Design

As described before, SER offers great potential for the production of a H₂-rich gas, which has excellent preconditions for use as a reduction agent. Furthermore, the produced gas contains a good H₂-to-CO ratio for the production of SNG, enabling its substitution for fossil natural gas.^{3,4} SER could be used to supply H₂-rich gas as well as an important process step for the substitution of SNG. The chosen process design aims at reasonable changes to reduce fossil carbon usage of a hot metal production process. Therefore, an electrolysis unit enables the production of additional hydrogen, and a CO₂ removal unit enables the recycling of CO₂ within the process. A further modification of the process by applying oxyfuel combustion allows the production of a sequestration-ready CO₂ stream.⁵ Oxy-SER is a relatively new, complex and sensitive process. The process has been proven at the lab and pilot scales.

1 Hubacek H., 1994;

2 <https://www.k1-met.com> (accessed on September 27, 2021)

3 Müller S., 2015; 4 Fuchs J., 2017; 5 Lehner M., 2020

Inappropriate operation could cause inhomogeneous combustion with negative effects on reactor materials. Based on existing experimental results, enrichment of the oxygen-exhaust gas mixture has been limited to the maximum oxygen content of 30 vol.-% and is preheated to a temperature of 350°C. A detailed description of oxyfuel combustion can be found in the literature.¹⁻⁴

Industrial Plant Model

Figure 58 shows the used concept for the creation of an industrial plant model enabling full replacement of the natural gas supply of a hot metal production. The proposed concept comprises a biomass gasification system, an electrolysis unit, a CO₂ removal unit and a methanation unit. The biomass gasification system is operated as a dual fluidized bed gasification system that converts woody biomass into a H₂-rich gas. Besides, the operation mode enables the production of a CO₂-rich gas by applying oxyfuel combustion to follow a carbon capture perspective and enable further utilization; this approach has been investigated intensively in recent years.⁵⁻⁷ The electrolysis unit is used to provide hydrogen to the overall process,⁸ allowing methanation according to stoichiometric ideal conditions accounting for the present biogenic carbon as well as recycled CO₂ from a blast furnace. A CO₂ removal unit is used to recycle CO₂ from the CO₂-rich blast furnace gas as feedstock for the methanation process. A detailed description of the process units can be found elsewhere.⁹⁻¹¹ Figure 59 shows the created industrial plant model used to calculate mass and energy balances of the proposed concept. The creation of the overall plant model was supported by an existing model library. The maturity of the used models can be described as follows:

- simulation of SER is based on validated test plant models [MRL 5, TRL 5],
- calculation of the oxyfuel combustion process was executed based on data from the literature [MRL 4, TRL 4],
- calculation of the electrolysis unit was based on data from the literature [MRL 4, TRL 6],
- calculation of the CO₂ removal unit was executed based on literature data [MRL 5, TRL 7],
- calculation of the methanation unit was executed based on experimental results as well as data from the literature [MRL 4, TRL 5]
- and additional components have been modelled according to the specification of different manufacturers [MRL 9, TRL 9].

Figure 59 shows the created plant model based on the illustrated concept shown in Figure 58.^{12,13} Overall, most of the models have been based on experimental results reported from different laboratories.¹⁴ The reliability of the created industrial plant model could be increased if validated data from demonstration projects in operational environments are used to improve the available database. The calculations are presented in the following section.

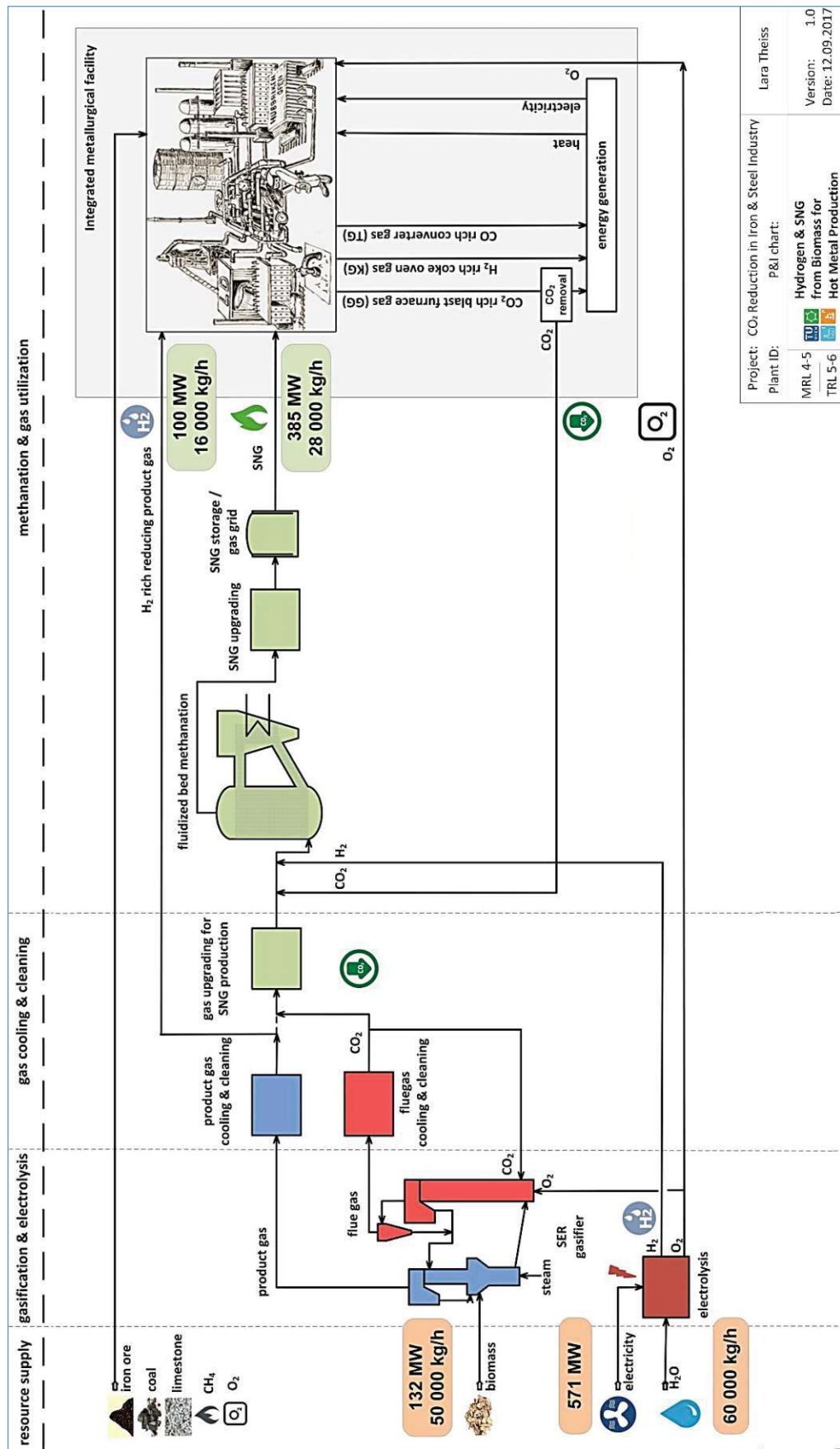
1 Hammerschmid H., 2016; 2 Hammerschmid M., 2019a; 3 Müller S., 2013

4 Scheffknecht G., 2011; 5 Fuchs J., 2019; 6 Fuchs J., 2018a

7 Fuchs J., 2019b; 8 <https://www.h2future-project.eu/> (accessed on March 27, 2021)

9 Götz M., 2017; 10 Schildhauer T.J.; 2016; 11 Schöny G., 2015

12 Theiss L., 2019; 13 Müller S., 2021; 14 Rosenfeld C.D., 2020



Project: CO₂ Reduction in Iron & Steel Industry
 P&I chart:
 Hydrogen & SNG
 From Biomass for
 Hot Metal Production
 Lara Theiss
 Version: 1.0
 Date: 12.09.2017

Figure 58: Hot metal production with reduced fossil carbon usage [MRL 4–5, TRL 5–6]

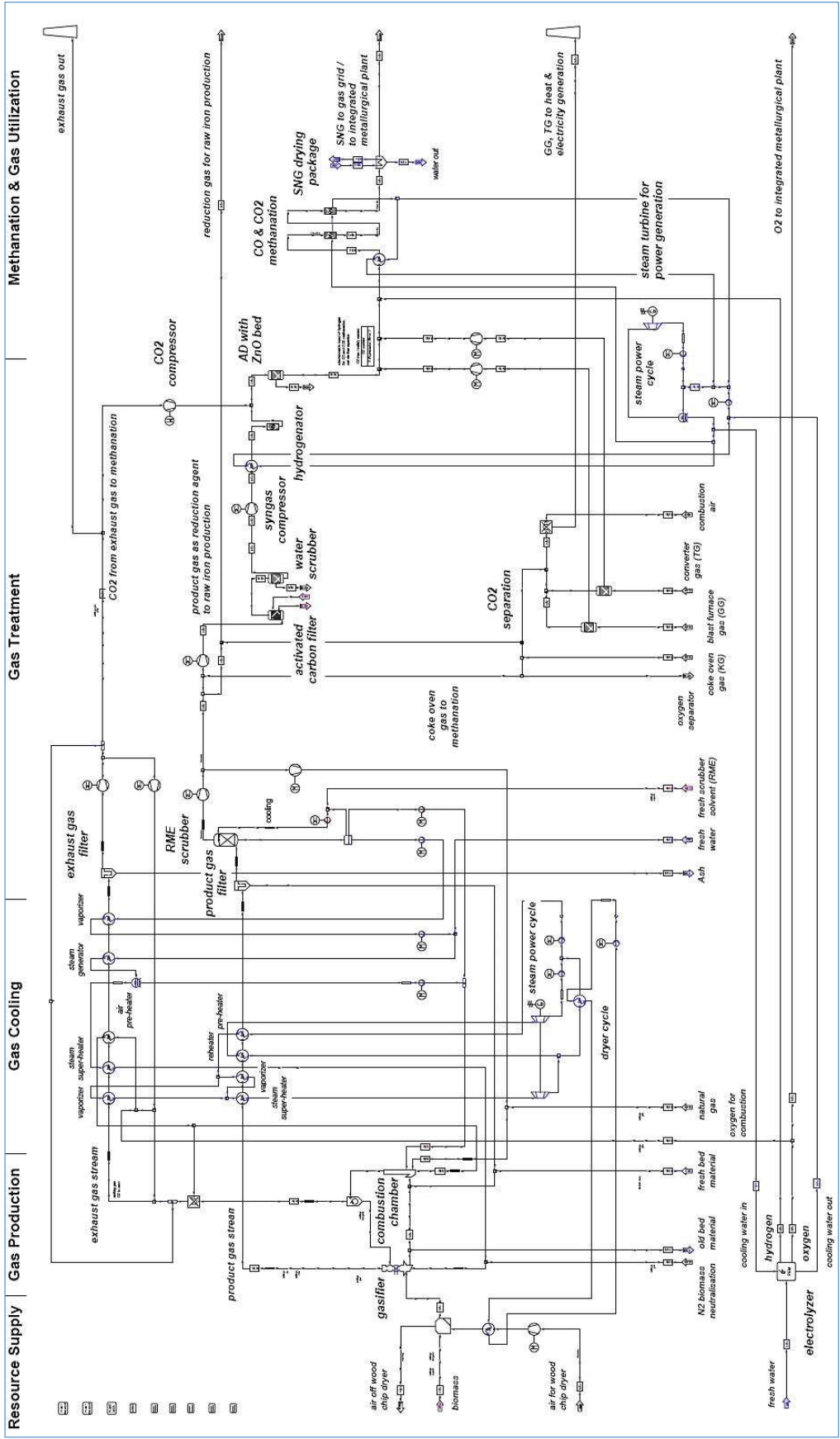


Figure 59: Industrial plant model of a hot metal production with reduced fossil carbon usage aiming at MRL 6–7

Evaluation of Plant Operation Impact

Table 20 shows the main results achieved. The calculations showed that

- 50 t/h of woody biomass,
- 60 t/h of fresh water and
- 571 MW of electricity

would be necessary to replace 385 MW of fossil natural gas of the analyzed hot metal production plant. Moreover, 50 t/h of woody biomass would represent about 1% of the primary energy usage of biomass in the energy sector in Austria.^{1,2}

Table 20: Simulation results²

| Plant input | | | Plant output | | |
|---------------------------------------------|------|--------|---------------------------------------------------------------|------|--------|
| Term | Unit | Value | Term | Unit | Value |
| Biomass (dry wood chips) | kg/h | 37 468 | H ₂ -rich product gas | kg/h | 15 671 |
| CO ₂ | kg/h | 40 247 | Synthetic natural gas | kg/h | 28 206 |
| Fresh water | kg/h | 60 365 | and others | - | ... |
| and others | ... | ... | H ₂ -rich product gas (P _{H2}) | MW | 100 |
| Biomass fuel power (P _{th}) | MW | 132 | Synthetic natural gas (P _{SNG}) | MW | 385 |
| Electricity electrolysis (P _{el}) | MW | 571 | Development status: [MRL 5, TRL 5] | | |
| Key performance indicator | | | Development targets: fossil CO ₂ emissions: 0 t/h; | | |
| Efficiency | % | 69 | | | |

Besides, recycling 20 000 Nm³/h of CO₂ is required to operate the proposed process configuration. The simulation model has provided valuable data for the design of proposed modifications. Upscaling oxy-SER to 150 MW, upscaling electrolysis to 600 MW and a long-term methanation test with real gas from hot metal production are recommended before larger scale implementation.² Table 20 includes important aspects for evaluation of the ecologic value of the proposed concept. The proposed concept enables a reduction in fossil carbon utilization. The necessary water and biomass consumption are a potential drawback of the presented solution, if these resources are not used in a sustainable way. At the same time, a realization of the proposed concept would enable implementation of a novel innovative energy technology developed within Europe. This would follow up a long tradition of innovation leadership in the field of iron and steel making in Austria and offers great potential for economic advantages in this field. As a result of the presented evaluation, it is recommended to follow up the proposed concept with respect to implementation.

1 Brauner, G., 2019;

2 Müller, S., 2021

5.5. Summary

Chapter 5 has presented the current state of industrial plant model development aiming at reduced fossil carbon utilization. Available data has been used to create [industrial plant models](#) to predict the potential future impact of investigated processes. The findings can be summarized as follows.

- An industrial plant model for the [production hydrogen with high-purity](#) has been established. The results achieved indicate that 30 MW of hydrogen can be produced from 50 MW of biomass. Energy- and material-intensive gas cleaning methods need to be evaluated by performing more experiments. Therefore, additional investigations are recommended, as an intermediate step, before building a large-scale demonstration plant.
- The [production of SNG](#) from dual fluidized bed steam gasification would enable the production of an energy carrier according to existing standards and infrastructure. The calculation of an industrial plant model indicated that 10 MW of SNG can be produced from 8.8 MW biomass and 5.5 MW of hydrogen. Applying sorption enhanced reforming (SER) could significantly reduce additional hydrogen demand. Important economic and technological challenges have not yet been considered adequately. The utilization of low-cost feedstock would be beneficial from an economic point of view. Additional research aims at analyzing practical large-scale operation as well as the expected economic and ecologic impact.
- The [production of liquid biofuels](#) via a Fischer–Tropsch process represents a promising option to provide a carbon-neutral solution for the mobility sector via second-generation biofuels. The simulation of a large-scale plant indicates that 51.8 MW of Fischer–Tropsch products can be produced from 100 MW of biomass. At the same time, questions remain with respect to the implementation of the investigated concept. Further improvement of the efficiencies and purities that have been reached is recommended.
- Within the present work, an integrated concept for the reduction of fossil CO₂ emissions of a [hot metal production](#) has been investigated. SER combined with SNG production and electrolysis would be capable of significantly reducing fossil CO₂ emissions of hot metal production.

Overall, all investigated plants offer large potential to contribute a reduction of fossil CO₂ emissions. At the same time, all investigated plant concepts need further investigation to reach the aim of MRL 7. Especially the [analysis of the ecologic and economic impacts](#) of the proposed concepts is an open issue which needs to be clarified in near future before large-scale application. The created plant models provide a basis for further evaluation by important evaluation tools such as life cycle analysis (LCA), the evaluation of the ecologic value or other evaluation tools.

6. DISCUSSION OF RESULTS

The present work has compared industrial plant models to enable a target-oriented implementation of innovative process ideas as valid technology in industry. The creation of industrial plant models has been used to determine the expected impact of the proposed concepts. The experimental results have been translated into industrial plant models for hydrogen production, SNG production, the production of liquid biofuels and an integrated solution for hot metal production.

The experimental results as well as the simulation models indicate the capability of the investigated processes by describing the technological as well as the modelling readiness level. Figure 60 summarizes the predicted efficiency of different investigated process routes based on validated experimental results. All investigated process routes offer the potential to reduce fossil CO₂ emissions. At the same time, the analysis of the ecologic and economic impacts of the proposed concepts is an open issue which needs to be clarified in near future before large-scale application.

The economics lead to the conclusion that it is preferable to use low-cost feedstock for the generation of clean energy carriers or platform chemicals. At the same time, low-cost feedstock inhibit remaining technological challenges as well as potential negative effects of local pollution. According to the latest state of methodology, software tools for life cycle assessment such as “ganzheitliche Bilanzierung” (GaBi) or databases like GEMIS allow an evaluation of the expected savings of greenhouse gas emissions.¹⁻⁴ Within the present work, an estimation of the ecologic value is carried out. (cf. Chapter 3.6, Table 8)

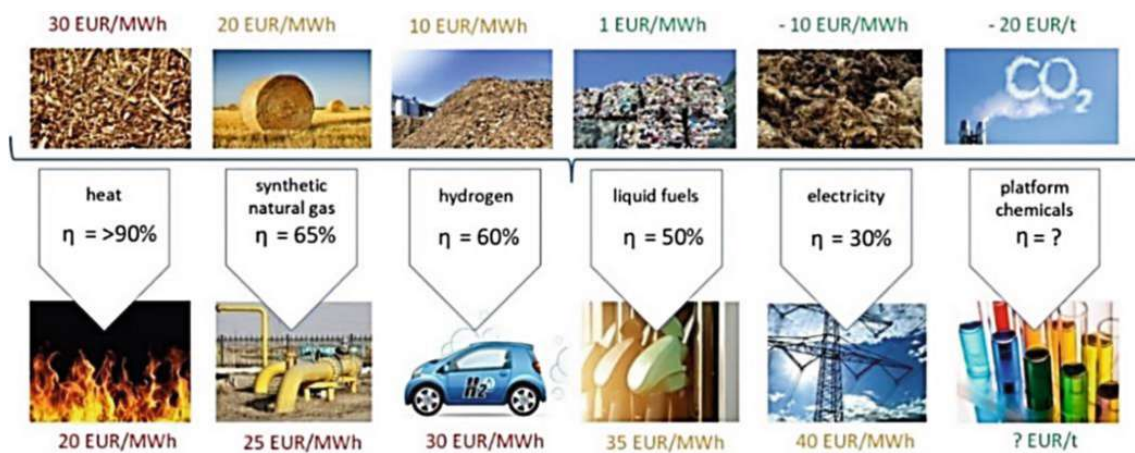


Figure 60: The expected efficiencies of the investigated process routes⁵⁻¹⁰

1 <https://gabi.sphera.com/software/gabi-software/> (accessed on March 27, 2021)

2 <http://iinas.org/gemis-de.html> (accessed on March 27, 2021); 3 ISO 14040

4 ISO 14044; 5 Müller S., 2017; 6 https://www.vt.tuwien.ac.at/prueflabor_fuer_feuerungsanlagen/ (accessed on March 27th, 2021)

7 Veress M., 2020; 8 Müller S., 2013; 9 Ebner J., 2018; 10 Stidl M., 2012

Figure 61 shows a Sankey-diagram identifying fossil carbon utilization in an Austrian model region.⁴ As a result of investigating different scenarios aiming at a reduction of fossil carbon utilization by using the shown energy flow chart, thermal heat collectors and heat pumps are recommended for space heating applications besides implementation of biomass-based heating and district heating concepts. Furthermore, central production of SNG is recommended. A further increase in electricity production from regenerative energy sources is foreseen. A reduction in fossil CO₂ emission of the mobility sector could be reached by using electrified vehicles as well as by utilizing liquid biofuels. The production of SNG or liquid biofuels offers the potential for carbon recycling following the idea of a carbon circular economy.

The evaluation of the ecologic value allows a well-balanced ranking of development results compared with the best available technology. An evaluation of the expected ecologic value of investigated processes within the present work lead to following conclusions. Table 21 shows the ranking of the used target criteria as a result of conjoint analysis. As usual, supply security can be named as the most important criterion. Besides, social aspects, the impact on local flora and fauna, air quality, water consumption and land used are weighted higher than operation efficiency, economic aspects or greenhouse gas emissions. Now, Table 22 summarizes a first estimation of the ecologic value of the evaluated technological pathways within the present work. As can be seen, all process routes utilize biogenic feedstock as an alternative to fossil carbon sources. High ecologic rating is achieved by SNG, heat from biomass as well as electricity from the Austrian power grid. Hydrogen production and electricity from biomass also achieve high score. Liquid biofuels achieve a lower score and conventional refineries reached the lowest rating.

Table 21: Results of conjoint analysis of used evaluation criteria by the author

Which target criteria contribute most to high ecologic value & beneficial plant operation?

| Target criteria | Ranking | Score | Weighting (w) |
|------------------------------------------------|---------|-------|---------------|
| Security of supply | 1 | 89 | 8.48% |
| Impact on local flora and fauna | 2 | 84 | 8.00% |
| Social impact | 3 | 83 | 7.90% |
| Air quality | 4 | 82 | 7.81% |
| Water consumption | 5 | 76 | 7.24% |
| Resource recycling rate | 5 | 76 | 7.24% |
| Land use/footprint | 7 | 74 | 7.05% |
| Generated waste | 8 | 69 | 6.57% |
| Contribution to culture | 8 | 69 | 6.57% |
| Carbon utilization | 10 | 66 | 6.29% |
| Modelling readiness level | 11 | 65 | 6.19% |
| Economic competitiveness | 11 | 65 | 6.19% |
| Greenhouse gas/CO ₂ emission factor | 13 | 56 | 5.33% |
| Technological readiness level | 14 | 54 | 5.14% |
| Operation efficiency | 15 | 42 | 4.00% |

The found weighting is used to evaluate different energy supply options.

1 Wulf C., 2018; 2 cf. CL:AIRE, 2020; 4 Hammerschmid M., 2022

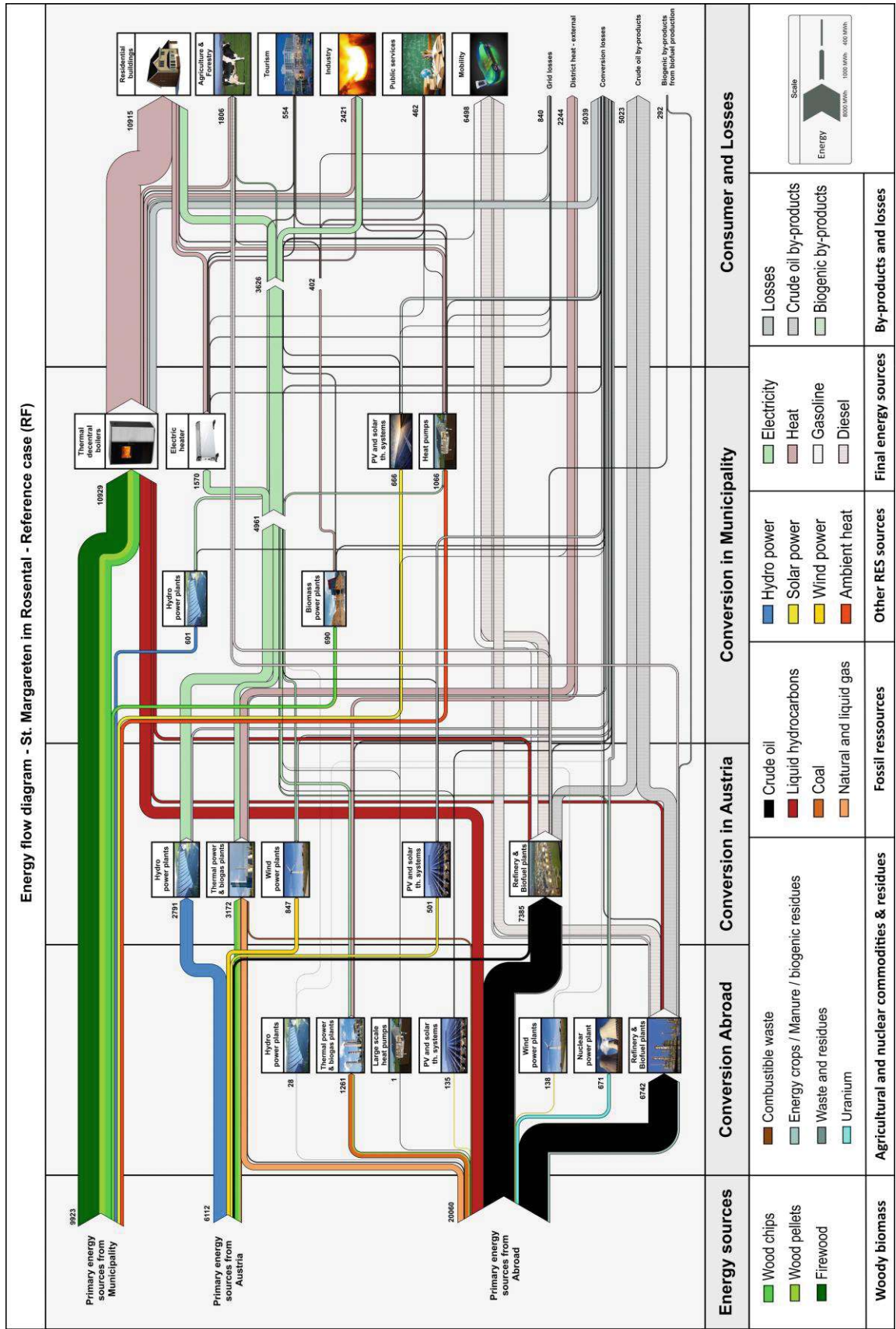


Figure 61: Energy flow diagram presenting the annual energy demand of St. Margareten im Rosental from reference case 2020

Table 22: Evaluation of the ecologic value of different process routes

Which ecologic value can be reached by large-scale implementation?

| Parameter ^{1,2} | Weighting | Unit | Natural gas combustion | Heat from Biomass | Austrian powergrid | Electricity from biomass | H2 via natural gas reforming | Hydrogen from biomass | Austrian natural gas grid | Synthetic natural gas | Refinery | Biofuels from FT | Value range (min./max.) | Comment | |
|----------------------------------|---------------|--------------------------|------------------------|-------------------|--------------------|--------------------------|------------------------------|-----------------------|---------------------------|-----------------------|--------------|------------------|-------------------------|---------|----------------------|
| Reference | Haas G., 2005 | SI, CGPM | DIN EN 15502-1 | Pröll, 2004 | www.apg.at | Müller, 2013 | Wawrzinek K., 2010 | Müller, 2013 | www.enisog.eu | Veress, 2018 | www.omv.at | Ebner J., 2018 | Wulf C., 2018 | | |
| TRL | 5,1 | - | 9 | 9 | 9 | 9 | 9 | 5 | 9 | 7 | 9 | 5 | 0 | 9 | cf. Chapter 5 |
| MRL | 6,2 | - | 5 | 5 | 4 | 5 | 5 | 5 | 4 | 4 | 5 | 5 | 0 | 9 | cf. Chapter 5 |
| Efficiency | 4 | % | 92,0 | 80,0 | 96,0 | 35,0 | 85,0 | 60,0 | 95,0 | 64,1 | 99,0 | 51,8 | 0 | 100 | Data sheets |
| Security of Supply | 8,5 | % | 99,0 | 86,8 | 99,999 | 86,8 | 97,0 | 86,8 | 100,0 | 86,8 | 96,0 | 83,3 | 0 | 100 | E-Control |
| Economic competitiveness | 6,2 | €/MWh | 30 | 30 | 40 | 90 | 30 | 60 | 25 | 50 | 35 | 70 | 100 | 0 | E-Control |
| Air quality | 7,8 | - | 9 | 6 | 9 | 6 | 9 | 6 | 9 | 6 | 8 | 6 | 0 | 10 | Estimation |
| CO2 emission factor ^a | 5,3 | t _{CO2-eq} /MWh | 0,2 | 0 | 0,14 | 0 | 0,25 | 0 | 0,2 | 0 | 0,25 | 0 | 0,25 | 0 | GEMIS |
| Land use / Foot print | 7,1 | m ² /MW | 250 | 250 | 250 | 250 | 80 | 160 | 313 | 313 | 200 | 200 | 313 | 0 | Estimation |
| Carbon utilization | 6,3 | % | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 40 | 0 | 50 | 0 | 100 | Chapter 4.2, Eq. 1.4 |
| Water consumption | 7,2 | - | 1 | 2 | 3 | 2 | 3 | 3 | 1 | 2 | 3 | 3 | 5 | 1 | Estimation |
| Impact flora & fauna | 8 | - | 2 | 3 | 3 | 3 | 2 | 3 | 2 | 3 | 3 | 4 | 5 | 1 | ISO 14042 |
| Generated waste | 6,6 | - | 1 | 2 | 1 | 2 | 1 | 2 | 1 | 2 | 3 | 2 | 5 | 1 | Estimation |
| Resource recycling rate | 7,2 | % | 0,0 | 100,0 | 80,0 | 100,0 | 0,0 | 100,0 | 0,0 | 100,0 | 0,0 | 100,0 | 0 | 100 | Biogenic share |
| Social Impact | 7,9 | - | 3 | 1 | 1 | 1 | 3 | 1 | 3 | 1 | 5 | 2 | 5 | 1 | HDI |
| Contribution to culture | 6,6 | - | 3 | 2 | 1 | 1 | 3 | 1 | 3 | 1 | 5 | 2 | 5 | 1 | UNESCO Criteria |
| Sum weighting | 100 | Evaluation | 615 | 693 | 692 | 654 | 603 | 662 | 599 | 683 | 444 | 616 | Ecologic value | | |
| Maximum score | 1000 | result | 61,5% | 69,3% | 69,2% | 65,4% | 60,3% | 66,2% | 59,9% | 68,3% | 44,4% | 61,6% | Ecologic rating | | |

a: value according to GEMIS V.5.0 database; biogenic raw materials assumed as carbon neutral; 1 cf. Wulf C., 2018; 2 CL-AIRE, 2020

CGPM (Central conference on weights and measures); HDI (Human Development Index); MFRL (Modelling readiness level); SI (International system of units) TRL (Technological readiness level);

As can be seen, an open discussion of data and best practice is necessary to promote the implementation of best possible solution. An open-minded exchange of a data between researchers, plant manufacturers and future plant operators is necessary to provide sufficient pre-conditions for the creation of digital industrial plant models as integrated digital twins in the future. Collaboration platforms enable open sharing of data and results achieved as well as interorganizational collaboration to accelerate effective development. Figure 62 indicates an initial proposal to realize a collaboration platform in the near future. A central planning table is coupled to a central database containing the latest state of knowledge. The MRL is used to describe the certainty of available models to calculate future operation at an industrial scale. During the iterative development process, modelling steps predict the results of the next step before cost-intensive demonstration steps. Furthermore, many examples from the past have shown that the final implementation of an industrial plant did not reach the desired outcome. Therefore, comparison of results achieved with the best available technology from a holistic point of view is essential.^{1,2} In this endeavor, the environmental impact of implemented solutions are more important than in the past and need to be monitored during the entire plant realization process. A lack of common definitions and standards leads to significant problems during comparison of different options. The present work contributes first important aspects for the recommended methodology. The definition of an ecologic value as well as the MRL can be highlighted as important perspectives for future.

Further integration of CO₂ recycling processes is being executed via a doctoral school a TU Wien with the title CO₂ Refinery.³ The described perspective forms the basis for the ongoing research project “Comprehensive Automation, Digitalisation & Optimization of Renewable & Sustainable SNG-production,” with the short title Adore SNG.⁴

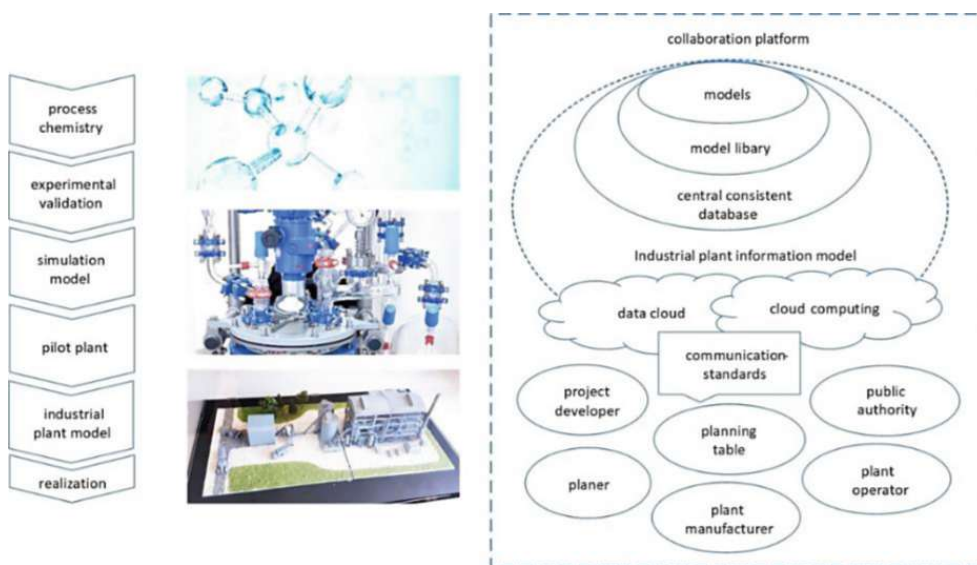


Figure 62: Concept for the future digital plant engineering via a digital collaboration platform

1 Wulf C., 2018; 2 CL:AIRE 2020

3 <https://www.tuwien.at/co2refinery> (accessed on November 16, 2021)

4 <https://projekte.ffg.at/projekt/3862075> (accessed on November 16, 2021)

7. SUMMARY & OUTLOOK

The present work has investigated a modelling-based approach for target-oriented development of energy technology enabling reduced fossil carbon dioxide emissions.

- How can advanced digital methods be applied to support energy technology development?
- Which approach leads to the best preparation of novel technology before industrial application?
- How can novel processes be applied most suitably to reach the desired development goals and thereby reduce fossil CO₂ emissions?

As a result, the present work recommends the introduction of a unified methodology. The proposed methodology suggests the introduction of a [modelling readiness level](#) (MRL) and the evaluation of found solutions by utility analysis with respect to the reached ecologic value. The following examples were investigated to analyze the proposed methodology:

- Experimental development of advanced processes for reduced fossil CO₂ emissions:
 - sorption enhanced reforming
 - and CO₂ gasification.
- Industrial plant model development for the application of:
 - hydrogen production,
 - SNG production,
 - biofuel production and
 - an integrated concept for the operation of a hot metal plant.

As a result, the following conclusions can be drawn. Analysis of the current energy system in Europe shows that the substitution of coal, oil and natural gas reduces the utilization of fossil carbon. Energy-intensive processes, which are based on these energy carriers, need to be adapted to [sustainable energy carriers](#). Processes providing liquid biofuels, SNG, hydrogen, electricity and heat from sustainable energy sources could enable the desired transformation if they are applied in a smart manner. This means a target-oriented development and implementation of novel [energy technology](#) following a holistic perspective proposed in the present work as [ecologic value](#).

[Advanced digital methods](#) support and accelerate [implementation](#) as shown in [Figures 63](#) and [64](#). A common language with accepted standards needs to be found as a part of the implementation of the proposed methodology in near future. Modern digital methods contain great potential for open collaboration of all acting parties. Therefore, the establishment of an open collaboration platform ensuring quality protected data and models is necessary.

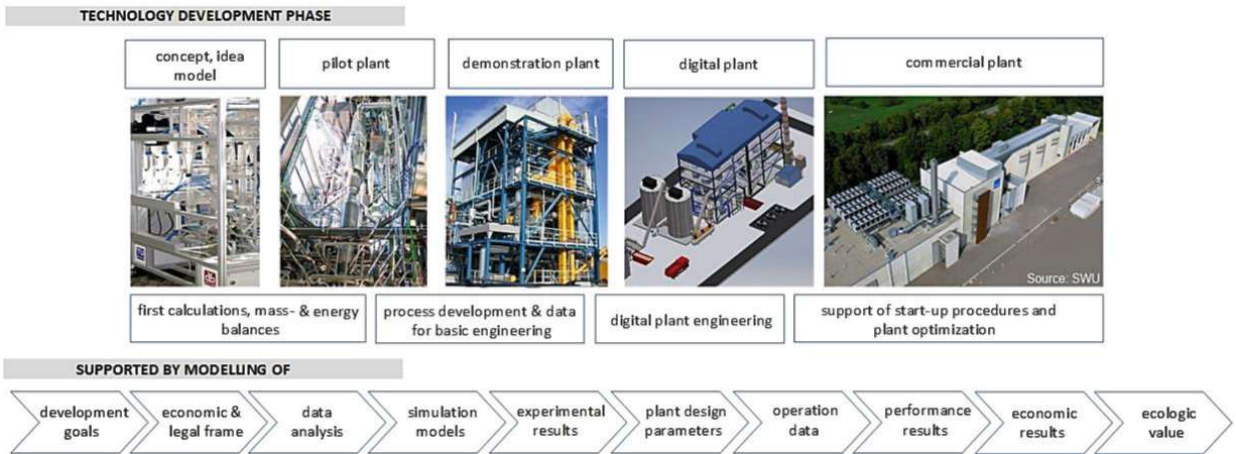


Figure 63: Methodology for the development of energy technology supported by advanced digital methods

The experimental demonstration of the investigated processes has shown that many options exist to reduce fossil carbon utilization from a technological point of view. Executed CO_2 gasification experiments indicated that this process could be valid technological option for syngas preconditioning and for recycling of CO_2 as CO . The experimental results of sorption enhanced reforming provide a sufficient basis to prepare an industrial demonstration. The produced hydrogen, SNG as well as liquid biofuels met the relevant specifications for its utilization within our existing energy system. Although the technological maturity has not reached commercial application, the experimental results indicate the suitability of the investigated process for a relevant reduction in fossil carbon utilization. Table 22 provided a valuable summary into expected advantages and disadvantages with respect to the impact of energy technology development aimed at reducing fossil CO_2 emissions.

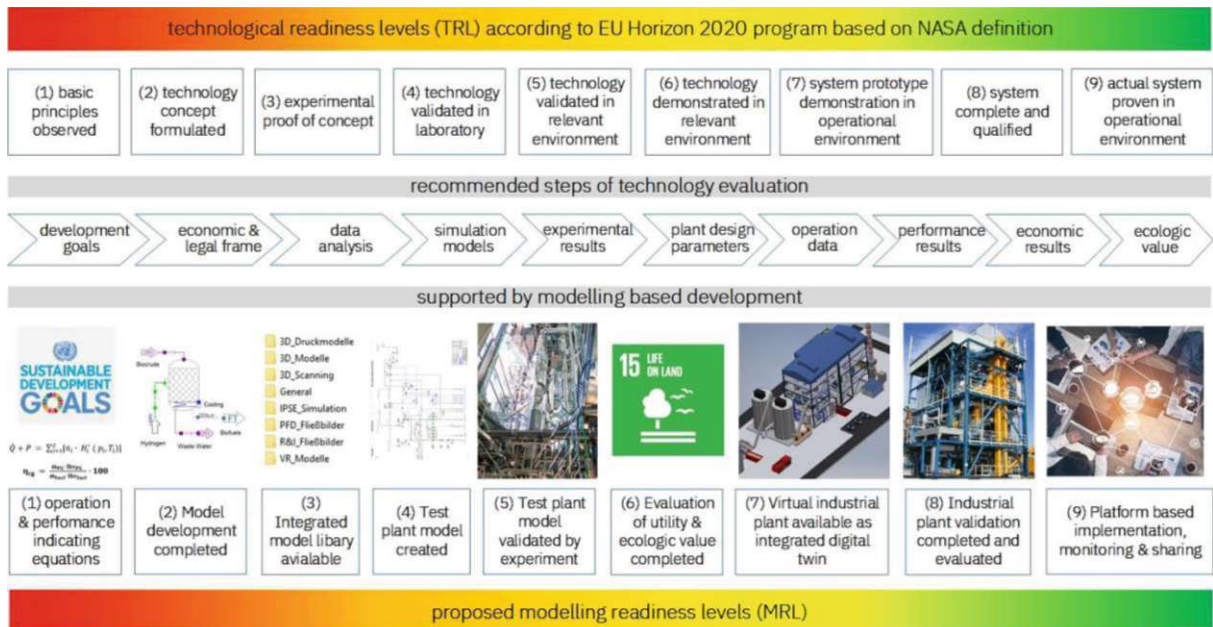


Figure 64: Definition of modelling readiness levels

As can be seen in Table 22, the production of [high-purity hydrogen](#) via biomass gasification has been rated at 0 t_{CO2}/MWh with a price of 60 €/MWh. At this point, the utilization of biogenic raw materials was assumed as carbon neutral. This has to be mentioned as an important assumption for the executed development work in the past as well as the present work. Investigation of the production of [SNG](#) has been rated at 0.0 t_{CO2}/MWh with a price of 50 €/MWh. Both production pathways allow avoiding fossil carbon emissions of energy-intensive industries such as hot metal production and could be applied in the context of existing industrial plants. Furthermore, the analysis within the present work indicates that [liquid biofuels](#) would be capable of covering the mobility sector demand for high energy density fuels. At the same time, the used production path has been rated with 0 t_{CO2}/MWh and a price of 70 €/MWh. Herein, the existing taxation system needs to promote eco-friendly utilization of provided energy carriers. Further development steps should be executed according to the proposed methodology aiming at a [reasonable utilization of existing biogenic raw materials](#) as well as modelling readiness level 8.

The developed approach forms the basis for the future work of the research group “[Industrial Plant Engineering & Application of Digital Methods](#)“ (E166-7-2) within the research area “[Fuel- and Energy System Engineering](#)” (E166-7) at TU Wien. The chosen approach includes testing, modelling, analysis, validation and scale-up of novel energy technology into industrial scale by advanced digital methods. Several projects will provide additional experimental data as well as validated simulation models preparing industrial demonstrations. The research project “FCTRAC” will enable demonstration of hydrogen production within a representative operational environment. The research project “ReGas 4Industry” contributes important experimental data to validate process simulation models for the production of SNG. The research project “Adore SNG” will contribute to improve markedly the used methodology for the creation of digital twins in preparation of an industrial demonstration within a relevant operational environment. Overall, the mentioned projects will contribute important aspects for platform-based sharing of [digital role models](#). Implementation of advanced digital tools provides great potential for tackling the huge challenges we face. The goal of a climate-neutral Europe by 2050 requires implementation of existing solutions. Therefore, the accompanying political measures have to be formulated to provide a sufficient legal and economic framework for technology implementation. [Advanced digital methods](#) enable an open approach for collective learning and cross organizational exchange. In the end, [Figures 65–68](#) indicate how advanced digital tools can be used to accompany the implementation of a novel industrial plant as an important building block of our future energy system.



Figure 65: Pictures of a 3D scan of the gasification test plant at TU Wien



Figure 66: A 3D-printed industrial plant model

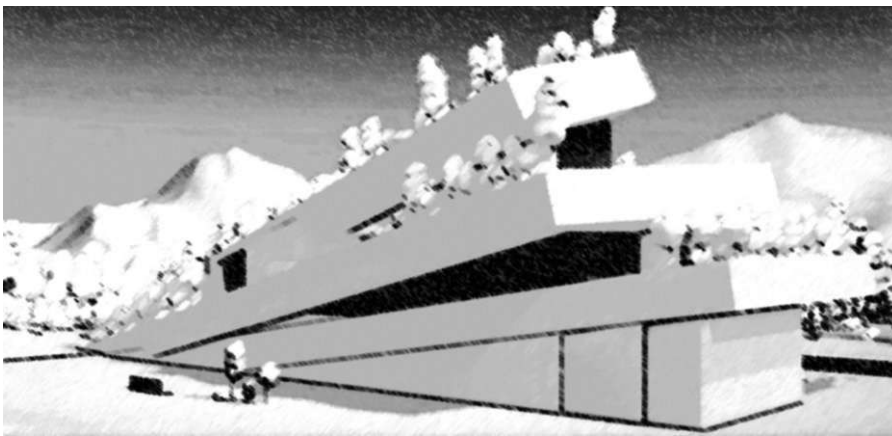


Figure 67: Industrial plant design as a contribution to culture with respect to further implementation

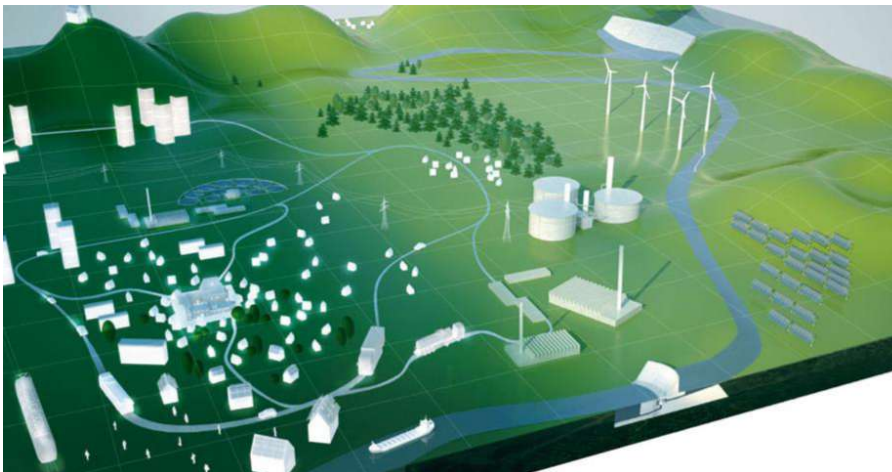


Figure 68: Virtual energy system engineering¹

1 <https://energiewelten.tuwien.ac.at> (accessed on March 27, 2021)

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ABBREVIATIONS

| | | | |
|------------|--------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------|---------------------------------------------------------------------------------------------|
| 3D | Three-dimensional | Heat-to-Fuel | Biorefinery combining HTL and FT to convert wet and solid organic, industrial wastes |
| ADOREe-SNG | Comprehensive Automation, Digitalisation & Optimization of Renewable & Sustainable SNG-production | | into 2nd generation biofuels with highest efficiency |
| APG | Austrian Power Grid | HNS | Hazelnut shells |
| BA | Bark | HP | Heat pipe reactor |
| BAT | Best available technology | HTL | Hydrothermal liquefaction |
| CCS | Carbon capture and storage | IPIM | Industrial plant information model |
| CCU | Carbon capture and utilization | ISO | International Organization for Standardization |
| CEN | French: Commitee Europeen de Normalisation, English: European Committee for Standardization | KPI | Key performance indicator |
| cf. | confer | LCA | Life cycle assessment |
| CFD | Computational fluid dynamics | LHV | Lower heating value |
| CGPM | General Conference on Weights and Measures | LIG | Lignite |
| CHP | Combined heat and power | LS | Lime stone |
| CIM | Computer-integrated manufacturing | MRL | Modelling readiness level |
| CM | Chicken manure | MWF | Municipal solid waste fraction |
| CNG | Compressed natural gas | NASA | National Aeronautics and Space Administration |
| CL:AIRE | Contaminated land: applications in real environments | NER | Funding programme pooling together about 2 billion EUR for innovative low-carbon technology |
| CR | Combustion reactor | Nm ³ | Gas cubic meters according to standard conditions (1.0 bar, 0.0 °C) |
| daf | dry and ash free | OL | Olivine |
| db | dry basis | PSA | Pressure swing adsorption |
| DFB | Dual fluidized bed | PtX | Electricity conversion, energy storage and reconversion pathways |
| DIN | German: Institute for Standardization (Deutsches Institut für Normung) | QFD | Quality function deployment |
| DS | German: Drucksensor, English: pressure sensor | QS | Quartz sand |
| EMAS | Eco management and audit scheme | ReGas 4Industry | Gas from regenerative waste for industry |
| EN | European standards | RH | Rice husk |
| EOP | Exhauste olive pomace | RME | Rapeseed methyl ester |
| Eq. | Equation | SER | Sorption enhanced reforming |
| EU-ETS | European Union emission trading system | SI | International system of units |
| EU Horizon | Funding programme for research and innovation | SCB | Sugar cane bagasse |
| FS | Feldspar | SW | Soft wood |
| FT | Fischer-Tropsch | SLF | Shredder light fraction |
| GaBI | German: holistic balancing (Ganzheitliche Bilanzierung) | SLI | Straw derived lignin |
| GAIA-X | Project for the development of an efficient and competitive, secure and trustworthy federation of data infrastructure and service providers for Europe | SNG | Synthetic natural gas |
| GCMS | Gas chromatography-mass spectrometry | ST | Straw |
| GEMIS | Global emission model of integrated systems | TCO | Total cost of ownership |
| GR | Gasification reactor | TGA | Thermogravimetric analysis |
| HDI | Human Development Index | TRL | Technological readiness level |
| | | TS | Technical specification |
| | | UNESCO | United Nations Educational, Scientific and Cultural Organization |
| | | UVP | Environmental impact assessment act (Umweltverträglichkeits-prüfungsgesetz) |
| | | VR | Virtual reality |
| | | WGS | Water-gas shift reaction |

SYMBOLS

| | | |
|----------------------------------|-------------------------------------------------------------------|----------|
| A | Expenses | [€/a] |
| BWSF | Cumulative present value factor | [-] |
| $C_{opt,X}$ | Costs option X | [€/a] |
| CO | Carbon monoxide | |
| CO ₂ | Carbon dioxide | |
| CH ₄ | Methane | |
| C ₂ H ₄ | Ethylene | |
| C ₅ -C ₉ | Gasoline/naphta fraction | [kg/h] |
| C ₁₀ -C ₁₉ | Diesel fraction | [kg/h] |
| C ₂₀ -C ₆₀ | Wax fraction | [kg/h] |
| E | Earnings | [€/a] |
| Fe | Iron | |
| FeO | Iron(II) oxide | |
| Fe ₂ O ₃ | Iron(III) oxide | |
| Fe ₃ O ₄ | Iron(II,III) oxide | |
| H ₂ | Hydrogen | |
| H ₂ /CO ratio | Ratio between hydrogen and carbon monoxide | [-] |
| H ₂ O | Water, Steam | |
| H* | Total enthalpy | [kJ/mol] |
| i | Interest rate | [%] |
| I ₀ | Investment costs | [€] |
| k | Lifetime | [a] |
| K _p (T) | Equilibrium constant | [-] |
| lhv _{add fuel} | Lower heating value of additional fuel | [kJ/kg] |
| lhv _{fuel} | Lower heating value of fuel (biomass) | [kJ/kg] |
| lhv _{PG} | Lower heating value of product gas | [kJ/kg] |
| m | Chain length number | [-] |
| M _{CaO} | Molar weight of CaO | [g/mol] |
| M _{CaCO₃} | Molar weight of CaCO ₃ | [g/mol] |
| M _{CO₂} | Molar weight of CO ₂ | [g/mol] |
| m | Mass | [kg] |
| $\dot{m}_{add fuel}$ | Additional fuel mass flow | [kg/h] |
| $\dot{m}_{bed,G,in}$ | Mass flow of bed material entering the gasifier from combustor | [kg/h] |
| $\dot{m}_{CO_2,capt}$ | Mass flow of CO ₂ absorbed by bed material in gasifier | [kg/h] |
| $\dot{m}_{CO_2,fluid}$ | Mass flow of CO ₂ used as fluidization agent | [kg/h] |
| \dot{m}_{fluid} | Fluidization mass flow gasifier | [kg/h] |
| $\dot{m}_{FT liquid}$ | Mass flow of Fischer-Tropsch liquids | [kg/h] |
| \dot{m}_{fuel} | Fuel mass flow (e.g. biomass to gasifier) | [kg/h] |
| $\dot{m}_{fuel,db}$ | Fuel mass flow dry basis (e.g. biomass to gasifier) | [kg/h] |
| \dot{m}_{PG} | Product gas mass flow | [kg/h] |
| \dot{m}_{steam} | Steam mass flow gasifier | [kg/h] |
| n | Cycle number | [-] |
| n _p | Variable number for polymerization reaction | [-] |
| \dot{n} | Molar flow rate | [mol/s] |
| \dot{n}_{CaCO_3} | Present CaCO ₃ | [mol] |
| \dot{n}_{CaO} | Present CaO | [mol] |
| \dot{n}_{CO_2} | Absorbed CO ₂ | [mol] |
| n _{CO,in} | Carbon monoxide exiting slurry reactor | [mol/h] |

| | | |
|-------------------------|-----------------------------------------------------------------------------|-------------------------------------------|
| $n_{CO,out}$ | Carbon monoxide entering slurry reactor | [mol/h] |
| NPV | Net present value | [€] |
| p | Pressure | [bar] |
| P | Mechanical power | [W] |
| p_{CO} | Partial pressure of CO in product gas | [Pa] |
| p_{CO_2} | Partial pressure of CO ₂ in product gas | [Pa] |
| p_{H_2} | Partial pressure of H ₂ in product gas | [Pa] |
| p_{H_2O} | Partial pressure of H ₂ O in product gas | [Pa] |
| P_{ϵ} | Profit | [€/a] |
| P_{el} | Electricity output | [W] |
| P_{FT} | Output of Fischer-Tropsch products | [W] |
| P_{H_2} | Output of hydrogen | [W] |
| P_{SNG} | Output of synthetic natural gas | [W] |
| P_{th} | Thermal power based on fuel power | [W] |
| \dot{Q} | Heat output, heat flow | [W] |
| \dot{Q}_{DH} | Heat flow district heating | [W] |
| T | Temperature | [°C] |
| T_G | Gasification temperature | [°C] |
| T_R | Combustion temperature | [°C] |
| U | Superficial gas velocity | [m/s] |
| U_{mf} | Minimum fluidization gas velocity | [m/s] |
| U_t | Terminal velocity | [m/s] |
| v | Value | [-] |
| w | Weighting | [-] |
| $w_{CaCO_3,bed,G,in}$ | CaCO ₃ in bed material to gasifier from combustor | [kg _{CaCO3} /kg _{bed}] |
| $w_{CaO,bed,G,in}$ | CaO in bed material to gasifier from combustor | [kg _{CaO} /kg _{bed}] |
| $w_{H_2O,fluid}$ | Water content of gasifier fluidization | [-] |
| $w_{H_2O,fuel}$ | Water content of biomass | [-] |
| $w_{H_2O,PG}$ | Water content of product gas | [-] |
| w_m | Fraction share with specific chain length | [kg] |
| X_C | Carbon utilization efficiency | [-] |
| X_{carb} | Carbonation conversion | [mol _{CO2} /mol _{CaO}] |
| X_{CaO} | CO ₂ load on CaO particle | [mol _{CO2} /mol _{CaO}] |
| $X_{C,FT}$ | Carbon utilization to Fischer-Tropsch products | [kg/kg] |
| X_{CO} | Carbon monoxide conversion | [%] |
| $X_{C,SNG}$ | Carbon utilization to SNG | [kg/kg] |
| X_{H_2O} | Water conversion rate | [kg _{H2O} /kg _{H2O}] |
| Z | Number of connected streams | [-] |
| α | Anderson-Schulz-Flory product distribution | [-] |
| $\delta_{Eq,WGS-shift}$ | Logarithmic distance to equilibrium (water-gas-shift reaction, gasifier) | [-] |
| K | Bed material renewal rate | [h ⁻¹] |
| Φ_{CO_2} | Share of CO ₂ used as gasification agent | [-] |
| Φ_{SC} | Steam to carbon ratio | [kg/kg] |
| η_g | Cold gas efficiency | [-] |
| $\eta_{overall}$ | Overall efficiency | [-] |

ANNEX

- I. EXPERIMENTAL DEVELOPMENT OF SORPTION ENHANCED REFORMING BY THE USE OF AN ADVANCED GASIFICATION TEST PLANT
(14 pages)
- II. CONVERSION OF CO₂ DURING THE DFB BIOMASS GASIFICATION PROCESS
(13 pages)
- III. FUEL FLEXIBLE GASIFICATION WITH AN ADVANCED 100 KW DUAL FLUIDIZED BED STEAM GASIFICATION PILOT PLANT
(15 pages)
- IV. HYDROGEN FROM BIOMASS - LARGE SCALE HYDROGEN PRODUCTION BASED ON A DUAL FLUIDIZED BED STEAM GASIFICATION SYSTEM
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- V. THERMODYNAMIC INVESTIGATION OF SNG PRODUCTION BASED ON DUAL FLUIDIZED BED GASIFICATION OF BIOGENIC RESIDUES
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(2 pages)

PAPER I

Müller S., Fuchs J., Schmid J.C., Benedikt F., Hofbauer H.

EXPERIMENTAL DEVELOPMENT OF SORPTION ENHANCED REFORMING BY THE USE OF AN ADVANCED GASIFICATION TEST PLANT

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Responsibility of the author:

- Designed the experimental campaign
- Created the simulation model used for the validation of the received experimental data
- Interpreted the results
- Independent of mentoring support

PAPER II

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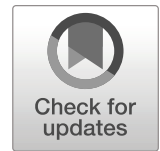
CONVERSION OF CO₂ DURING THE DFB BIOMASS GASIFICATION PROCESS

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Responsibility of the author:

- Designed the experimental campaign and key performance indicators
- Created the simulation model used for the calculations
- Interpreted and visualized the results
- Independent of mentoring support



Conversion of CO₂ during the DFB biomass gasification process

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Abstract

In many industrial processes, the climate-damaging gas CO₂ is produced as undesired by-product. The dual fluidized bed biomass gasification technology offers the opportunity to tackle this problem by using the produced CO₂ within the process as gasification agent. Therefore, a 100 kW_{th} pilot plant at TU Wien was used to investigate the use of CO₂ as gasification agent by converting softwood as fuel and olivine as bed material into high-valuable product gas. A parameter variation was conducted, where the typically used gasification agent steam was substituted stepwise by CO₂. Thereby, the amount of CO and CO₂ increased and the content of H₂ decreased in the product gas. These trends resulted in a declining H₂/CO ratio and a decreasing lower heating value when CO₂ was increased as gasification agent. In contrast to these declining trends, the carbon utilization efficiency showed an increasing course. As second part of this work, a temperature variation from 740 to 840 °C was conducted to investigate the change of the main product gas components. With increasing temperature, CO and H₂ increased and CO₂ decreased. To determine the degree of conversion of CO₂ in the DFB reactor system, two approaches were selected: (1) a carbon balance and (2) a hydrogen balance. This way, it was found out that a certain amount of CO₂ was indeed converted at the investigated process conditions. Furthermore, under certain assumptions, the reverse water-gas shift reaction was identified to be the predominant reaction during CO₂ gasification.

Keywords Pure CO₂ gasification · Biomass · 100 kW_{th} pilot plant · Carbon utilization efficiency · Reverse water-gas shift

1 Introduction

Starting from the Kyoto protocol [1], which was published in 1998, followed by the Renewable Energy Directive (RED) established in 2009 by the European Union up to the Paris Agreement from the United Nations Framework Convention on Climate Change in 2015 [2], several approaches for CO₂ mitigation were established in the past. It went further with a recast of the Renewable Energy Directive—Recast to 2030 (RED II) in December 2018 to strengthen the awareness of climate change and its possible effects on the environment and humanity [3]. To sum up, all these protocols and agreements urgently appeal to reduce CO₂ emissions and to mitigate the negative effects of climate change worldwide.

Furthermore, the predicted increase of CO₂ emissions up to 60% in 2050 compared with that in 2011 presents a driving force for the development and realization of renewable energy technologies [4]. Additionally, the reutilization of unavoidably produced CO₂ and in parallel the conversion of CO₂ into valuable products is urgent. A possible technology to tackle these problems could be the thermochemical conversion process of biomass through gasification. In this way, fossil energy sources like crude oil or lignite can be substituted by renewable, alternative feedstocks and CO₂ used within the process as gasification agent. In this way, a high-valuable product gas can be generated, which can be further upgraded in different chemical synthesis steps to produce advanced biofuels [5, 6] or other chemicals [7]. For this purpose, the dual fluidized bed (DFB) biomass gasification process, which was developed at TU Wien, could serve as a key technology. Successful test runs with steam as gasification agent have been carried out for more than 20 years [8]. However, the use of CO₂ as gasification agent presents a novel research topic. First experimental test runs using mixtures of steam

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and CO₂ were already carried out, starting in 2018 [9, 10]. The main findings of these test runs were:

- In contrast to pure steam biomass gasification, where a hydrogen (H₂)-rich product gas is generated, a carbon monoxide (CO)-rich product gas is created, when CO₂ is used as gasification agent.
- The H₂/CO ratio, which presents an important factor for different downstream synthesis processes, decreased, when a higher content of CO₂ was used in the gasification agent mixture.
- The utilization of CO₂ as gasification agent showed an increase in the carbon utilization efficiency.
- Through the supplementing properties of steam and CO₂ as gasification agent, lower tar contents were generated compared with pure steam gasification.

Other research groups like CEA in France [11], Jeremias et al. [12–14] in the Czech Republic, Stec et al. [15] in Poland, Cheng et al. [16] in Singapore, and Szul et al. [17] from the Institute of Chemical Processing of Coal (IChPW) in Poland also already examined the use of CO₂ as gasification in fluidized bed reactor systems. The main outcomes of their works comply with the findings of the first experimental test runs in the DFB reactor system. The H₂/CO ratio was reduced [11], mixtures of steam and CO₂ had a positive effect on tar reduction [12], the CO₂/C ratio influenced the CO yield [15], and the cold gas efficiency increased, when CO₂ was used as gasification agent [14].

The use of pure CO₂ as gasification was not investigated in the DFB reactor system during the first experimental test runs so far. Therefore, this missing building block was investigated within the scope of this publication. The influence of the step-wise substitution of steam by CO₂ as gasification up to 100 vol.-% on the H₂/CO ratio, the CO₂ conversion, the carbon utilization efficiency, and the cold gas efficiency was examined. Furthermore, a temperature variation from about 740 to about 840 °C was carried out under pure CO₂ atmosphere to determine the influence on the product gas quality. As a concluding chapter, investigations regarding the determination of the conversion of CO₂ within the DFB reactor system are presented. For this purpose, carbon and hydrogen balances were set up around the gasification reactor for pure CO₂ gasification and compared with a pure steam gasification test run.

2 Materials and methods

For the experimental test runs, a 100 kW_{th} DFB pilot plant, which was built at TU Wien, was used. The principle of the DFB gasification pilot plant is shown in Fig. 1. The pilot plant is composed of two reactors: a gasification reactor (GR, blue rectangle) and a combustion reactor (CR, red rectangle),

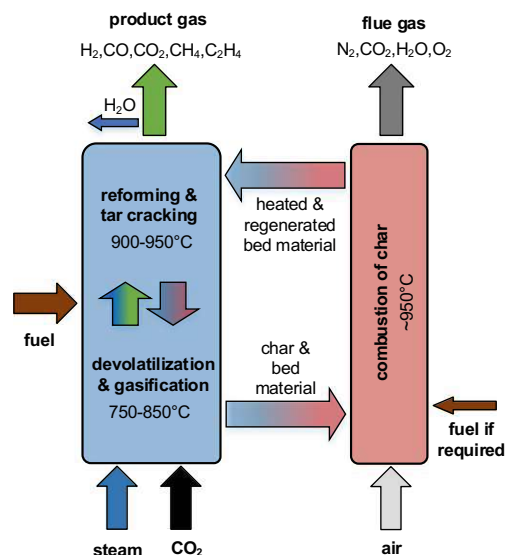


Fig. 1 Principle of the DFB biomass gasification

which are connected by loop seals (horizontal arrows). The GR is divided into a lower part, where the devolatilization and gasification reactions take place and an upper part, where reforming and tar cracking reactions occur. The GR can be fluidized with CO₂ and/or steam and mixtures thereof and the CR is fluidized with air. Biomass is introduced into the lower part of the GR. In the GR, a product gas, which is composed of carbon monoxide (CO), hydrogen (H₂), carbon dioxide (CO₂), methane (CH₄), ethylene (C₂H₄), water (H₂O), and other minor components, is generated. In the CR, a flue gas, which mainly contains CO₂, H₂O, nitrogen (N₂), and oxygen (O₂), is produced.

The 100 kW_{th} DFB biomass gasification pilot plant went into operation in 2014 at TU Wien [18]. Fig. 2 shows the upper part of the pilot plant with three fuel hoppers and the lower part of the reactor system with some ash removal containers. The GR of the pilot plant is operated as a bubbling fluidized bed in the lower part and as a counter-current column with turbulent fluidized bed zones in the upper part. In the upper part of the gasification reactor, also constrictions are installed. These constrictions enable an increased interaction of downward flowing hot bed material particles with upward streaming product gas. In this way, the contact time as well as the conversion efficiency can be increased [19, 20]. A more detailed description of the pilot plant and the corresponding measurement equipment can be found in literature [18, 21].

2.1 Relevant chemical reactions during biomass gasification

In Table 1, a selection of important heterogeneous gas-solid and homogeneous gas-gas reactions, which can occur during the DFB biomass gasification process, is presented. The gas-solid reactions are displayed in Eqs. 1–3 and the gas-gas



Fig. 2 Upper part and lower part of the DFB biomass gasification system

reactions are stated in Eqs. 4–7. Gas-gas reactions are secondary gasification reactions, which occur between the gasification agent, the gaseous products of char gasification, and the gaseous products of pyrolysis.

2.2 Investigated materials

For the presented test runs, softwood (SW) pellets were used as fuel and olivine as bed material. The proximate and ultimate analyses of SW are shown in Table 2 and the composition of olivine is presented in Table 3. Olivine, which shows catalytically active behavior [23, 24], was used because it is known as state-of-the-art bed material and typically used in industrial-sized biomass gasification plants [25, 26].

Table 1 Relevant gasification reactions [22]

| Reaction name | Heterogeneous reactions (gas-solid) | Enthalpy | |
|-----------------------------------------|------------------------------------------------------------|---------------------|-------|
| Water-gas reaction | $C + H_2O \rightarrow CO + H_2$ | Endothermic | Eq. 1 |
| Boudouard reaction | $C + CO_2 \rightarrow 2 CO$ | Endothermic | Eq. 2 |
| Hydrogenated gasification | $C + 2 H_2 \rightarrow CH_4$ | Slightly exothermic | Eq. 3 |
| | Homogeneous reactions (gas-gas) | | |
| Reverse water-gas shift reaction (RWGS) | $CO_2 + H_2 \leftrightarrow CO + H_2O$ | Endothermic | Eq. 4 |
| Methanation | $CO + 3 H_2 \leftrightarrow CH_4 + H_2O$ | Exothermic | Eq. 5 |
| Steam reforming | $C_xH_y + x H_2O \rightarrow x CO + (x + \frac{y}{2}) H_2$ | Endothermic | Eq. 6 |
| Dry reforming | $C_xH_y + x CO_2 \rightarrow 2x CO + \frac{y}{2} H_2$ | Endothermic | Eq. 7 |

Table 2 Proximate and ultimate analysis of softwood pellets

| Parameter | Unit | Value |
|-----------------------------|---------------------|-------|
| Proximate analysis | | |
| Water content | wt.-% | 7.2 |
| Volatiles | wt.-% _{db} | 85.4 |
| Fixed C | wt.-% _{db} | 14.6 |
| LHV (dry) | MJ/kg _{db} | 18.9 |
| LHV (moist) | MJ/kg | 17.4 |
| Ultimate analysis | | |
| Ash content | wt.-% _{db} | 0.2 |
| Carbon (C) | wt.-% _{db} | 50.7 |
| Hydrogen (H) | wt.-% _{db} | 5.9 |
| Oxygen (O) | wt.-% _{db} | 43.0 |
| Nitrogen (N) | wt.-% _{db} | 0.2 |
| Sulfur (S) | wt.-% _{db} | 0.005 |
| Chloride (Cl) | wt.-% _{db} | 0.005 |
| Ash content | wt.-% _{db} | 0.2 |
| Ash melting behavior | | |
| Deformation temperature (A) | °C | 1335 |

2.3 Validation of process data with IPSE

The validation of the process data was carried out by the calculation of mass and energy balances with the software tool IPSEpro. In this way, data, which cannot be measured directly during experimental test runs, can be determined. For the simulation with IPSEpro, a detailed model library, which was developed at TU Wien over many years, was used [27, 28]. All experimental results presented within this publication were validated with IPSEpro. Based on the validated data, the following key figures were selected to describe the performance and efficiency of the presented test runs in detail. All input and output streams, which were used for the calculation of the performance indicating key figures, are presented in Fig. 3.

Table 3 Composition of bed material olivine

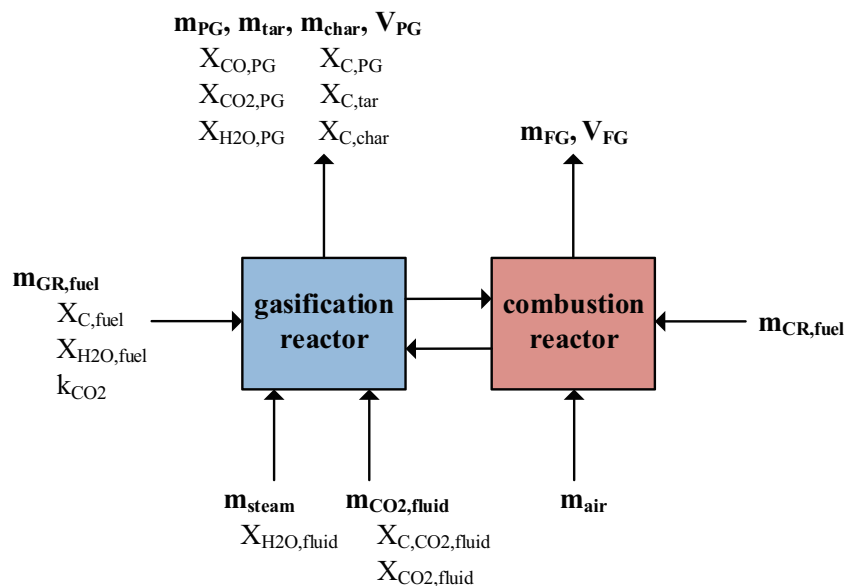
| Parameter | Unit | Value |
|----------------------------------------------|-------------------|----------|
| Iron oxide (Fe ₂ O ₃) | wt.-% | 8.0–10.5 |
| Magnesium oxide (MgO) | wt.-% | 48–50 |
| Silicon oxide (SiO ₂) | wt.-% | 39–42 |
| Calcium oxide (CaO) | wt.-% | ≤ 0.4 |
| Trace elements (< 0.4 per element) | wt.-% | ≤ 5 |
| Hardness | Mohs | 6–7 |
| Sauter mean diameter | mm | 0.243 |
| Particle density | kg/m ³ | 2850 |

The CO₂ to carbon ratio, $\varphi_{\text{CO}_2\text{C}}$, presented in Eq. 8, is defined as the introduced CO₂ as gasification agent to C in the dry and ash-free fuel. The product gas yield PGY describes the ratio between the volume flow of dry product gas to the mass flow of dry and ash-free fuel introduced into the GR (see Eq. 9). The carbon to CO conversion $X_{\text{C} \rightarrow \text{CO}}$ describes the amount of CO in the product gas to the total amount of introduced C as fuel and gasification agent (see Eq. 10). C in CO₂ as gasification agent is calculated through the share named $X_{\text{C,CO}_2,\text{fluid}}$ and the mass flow of CO₂ as gasification agent. Eq. 11 shows the CO₂ conversion rate X_{CO_2} , which gives the ratio of consumed CO₂ during gasification to the amount of CO₂ introduced into the GR via CO₂ as gasification agent and CO₂ produced from the pyrolysis of the fuel. Detailed information about the calculation of X_{CO_2} can be found in [10]. $X_{\text{H}_2\text{O}}$ is defined as the steam-related water conversion. It presents the water consumed for e.g. CO and H₂ production in relation to the sum of water, which is fed to the GR as gasification agent and fuel water (see Eq. 12). The overall cold gas

efficiency $\eta_{\text{CG},o}$ is presented in Eq. 13. It describes the amount of chemical energy in the product gas in relation to the chemical energy of the fuel introduced into the gasification and combustion reactor minus appearing heat losses. Due to the fact that the GR was fluidized with different ratios of CO₂, the value ϕ_{CO_2} was introduced, which describes the share of CO₂ used as gasification agent. The calculation of ϕ_{CO_2} is shown in Eq. 14. The carbon utilization efficiency (X_{C}) (see Eq. 15) gives the ratio of the amount of carbon leaving the GR via the product gas minus the content of carbon in char and tar to the amount of carbon introduced into the GR with the fuel and CO₂ as gasification agent.

The equation for calculating the deviation from the reverse water-gas shift reaction (RWGS), which is displayed in Eq. 4, is given in Eq. 16. The equilibrium constant $K_{\text{p, RWGS}}(T)$ was calculated using the software tool HSC Chemistry [29]. If the deviation is zero, it means that the equilibrium state of the equation is reached. A negative value would indicate that the gas composition is on the side of the reactants, which would mean that a further reaction is thermodynamically possible. A positive sign would imply that the actual state is on the side of the products. However, this state cannot be reached thermodynamically through the RWGS reaction alone. Additional reactions are required as stated in [30]. In Eq. 17, the logarithmic deviation from the Boudouard (BOU) reaction (see Eq. 2) $p\delta_{\text{eq, BOU}}$ is shown. The equilibrium constant ($K_{\text{p, BOU}}(T)$) was calculated by the use of the software tool HSC [29] as well. When $p\delta_{\text{eq, BOU}}$ is 0, the Boudouard reaction is in equilibrium. When $p\delta_{\text{eq, BOU}} > 0$, the state of equilibrium lies on the product side, whereas when $p\delta_{\text{eq, BOU}} < 0$, the equilibrium is located on the reactants side.

In Eq. 18, the ratio between water in the fuel and steam introduced into the GR as gasification agent to the amount of

Fig. 3 Input and output streams for the calculation of the key figures

C in the fuel and CO₂ introduced into the GR as gasification agent is shown.

$$\varphi_{\text{CO}_2\text{C}} = \frac{\dot{m}_{\text{CO}_2,\text{fluid}}}{x_{\text{C},\text{fuel}} \times \dot{m}_{\text{GR},\text{fuel},\text{db}}} \quad (8)$$

$$\text{PGY} = \frac{\dot{V}_{\text{PG}}}{\dot{m}_{\text{GR},\text{fuel},\text{daf}}} \quad (9)$$

$$X_{\text{C} \rightarrow \text{CO}} = \frac{x_{\text{CO},\text{PG}} \times \dot{m}_{\text{PG}}}{x_{\text{C},\text{fuel}} \times \dot{m}_{\text{GR},\text{fuel},\text{db}} + x_{\text{C},\text{CO}_2,\text{fluid}} \times \dot{m}_{\text{CO}_2,\text{fluid}}} \quad (10)$$

$$X_{\text{CO}_2} = \frac{\dot{m}_{\text{CO}_2,\text{fluid}} + k_{\text{CO}_2} \times \dot{m}_{\text{fuel},\text{daf}} - x_{\text{CO}_2,\text{PG}} \times \dot{m}_{\text{PG}}}{\dot{m}_{\text{CO}_2,\text{fluid}} + \dot{m}_{\text{GR},\text{fuel},\text{daf}} \times k_{\text{CO}_2}} \quad (11)$$

$$X_{\text{H}_2\text{O}} = \frac{\dot{m}_{\text{steam}} + x_{\text{H}_2\text{O},\text{fuel}} \times \dot{m}_{\text{fuel}} - x_{\text{H}_2\text{O},\text{PG}} \times \dot{m}_{\text{PG}}}{\dot{m}_{\text{steam}} + x_{\text{H}_2\text{O},\text{fuel}} \times \dot{m}_{\text{GR},\text{fuel}}} \quad (12)$$

$$\eta_{\text{CG},\text{o}} = \frac{\dot{V}_{\text{PG}} \times \text{LHV}_{\text{PG}}}{\dot{m}_{\text{GR},\text{fuel}} \times \text{LHV}_{\text{GR},\text{fuel}} + \dot{m}_{\text{CR},\text{fuel}} \times \text{LHV}_{\text{CR},\text{fuel}} - \dot{Q}_{\text{loss}}} \cdot 100 \quad (13)$$

$$\phi_{\text{CO}_2} = \frac{x_{\text{CO}_2,\text{fluid}}}{x_{\text{CO}_2,\text{fluid}} + x_{\text{H}_2\text{O},\text{fluid}}} \quad (14)$$

$$X_{\text{C}} = \frac{x_{\text{C},\text{PG}} \times \dot{m}_{\text{PG}} - x_{\text{C},\text{tar}} \times \dot{m}_{\text{tar}} - x_{\text{C},\text{char}} \times \dot{m}_{\text{char}}}{\dot{m}_{\text{fuel},\text{db}} \times x_{\text{C},\text{fuel}} + \dot{m}_{\text{CO}_2,\text{fluid}} \times x_{\text{C},\text{CO}_2,\text{fluid}}} \quad (15)$$

$$p_{\delta_{\text{eq}},\text{RWGS}} = \log_{10} \left[\frac{\prod_i p_i^{\nu_i}}{K_{\text{p},\text{RWGS}}(T)} \right] \quad (16)$$

$$p_{\delta_{\text{eq}},\text{BOU}} = \log_{10} \left[\frac{\prod_i p_i^{\nu_i}}{K_{\text{p},\text{BOU}}(T)} \right] \quad (17)$$

$$\frac{\text{H}_2\text{O}}{(\text{C} + \text{CO}_2)} = \frac{\dot{m}_{\text{steam}} + \dot{m}_{\text{GR},\text{fuel}} \times x_{\text{H}_2\text{O},\text{fuel}}}{x_{\text{C},\text{fuel}} \times \dot{m}_{\text{GR},\text{fuel},\text{db}} + \dot{m}_{\text{CO}_2,\text{fluid}}} \quad (18)$$

2.4 Thermodynamic calculations

To develop efficient biomass conversion technologies, which can also compete with fossil energy technologies, it is required to determine their energy efficiency. For the determination of the energy efficiency of different processes, various performance indicators, mostly based on thermodynamics, are used [31]. Due to that fact, thermodynamic calculations were carried out for the test runs carried out within the scope of this work as well and compared with the experimental results. In this way, a better understanding of the ongoing chemical reactions in the DFB reactor system could be gained and the energy efficiency of the process evaluated.

For the thermodynamic calculations, the product gas compositions at different shares of ϕ_{CO_2} and at different temperatures were calculated assuming thermodynamic equilibrium

with the software tool HSC Chemistry [29]. HSC Chemistry uses the Gibbs free energy minimization method. In the equilibrium state, the Gibbs free energy is minimized. Detailed descriptions of this approach can be found in literature [32, 33].

As already mentioned beforehand, the product gas of the DFB reactor system is mainly composed of CO, H₂, CO₂, CH₄, H₂O, and higher hydrocarbons. Higher hydrocarbons with the formula C_xH_y were summarized by the compound C₂H₄. Based on these chemical compounds, the following simultaneous chemical gas-gas reactions were taken into account to take place in the DFB reactor system during the presented gasification test run:

- the water-gas shift reaction (Eq. 4),
- the methanation reaction (Eq. 5),
- the steam reforming reaction (Eq. 6),
- and the dry reforming reaction (Eq. 7).

The results of the thermodynamic calculations are presented in the results section and compared with the experimental results of the test runs.

3 Results and discussion

In this chapter, the main findings of experimental test runs are presented. The results of the stepwise substitution of steam by CO₂, the temperature variation under pure CO₂ atmosphere as well as the carbon and hydrogen balances are shown.

3.1 From pure steam to pure CO₂ as gasification agent

In Table 4, the main operational parameters from five test runs for investigating the stepwise substitution of steam by CO₂ are shown. Softwood pellets were used as fuel and olivine as a bed material for all test runs. ϕ_{CO_2} was changed from 0 to 1. The fuel power introduced into the GR (P_{GR}) was in a range of 83 to 95 kW. The amount of additional fuel, which was introduced into the CR (P_{CR}) to control the gasification temperature and to compensate for the relatively high heat losses of the pilot plant, was between 59 and 68 kW. To enable a comparison of these test runs with test runs, where pure steam was used as gasification agent, a ratio between P_{CR} and P_{GR} was calculated. For pure steam gasification test runs, a $P_{\text{CR}}/P_{\text{GR}}$ ratio of around 0.5 is a typical value, but it depends on the type of fuel introduced into the GR as well as the operating parameters [24, 34]. Test run 1 (pure steam) showed a quite high $P_{\text{CR}}/P_{\text{GR}}$ compared with other pure steam gasification test runs in literature. However, this outlier can be explained by the relatively high heat losses for this test run. Taking into account a typical $P_{\text{CR}}/P_{\text{GR}}$ ratio for pure steam gasification of around 0.5, it can be seen that adding CO₂ to the

Table 4 Main operational parameters

| Parameter | Unit | Test run | | | | |
|-----------------------------------------------|------------------------------------------------------|----------|---------|---------|---------|---------|
| | | 1 | 2 | 3 | 4 | 5 |
| Fuel | - | SW | SW | SW | SW | SW |
| Bed material | - | Olivine | Olivine | Olivine | Olivine | Olivine |
| ϕ_{CO_2} | - | 0 | 0.32 | 0.45 | 0.68 | 1 |
| $\text{H}_2\text{O}/(\text{C} + \text{CO}_2)$ | - | 1.61 | 0.59 | 0.43 | 0.22 | 0.04 |
| Fuel to GR | kW | 95 | 92 | 86 | 87 | 83 |
| Fuel to CR | kW | 68 | 59 | 53 | 53 | 59 |
| $P_{\text{CR}}/P_{\text{GR}}$ ratio | - | 0.72 | 0.64 | 0.62 | 0.61 | 0.71 |
| $\varphi_{\text{CO}_2\text{C}}$ | $\text{kg}_{\text{CO}_2}/\text{kg}_{\text{C, fuel}}$ | - | 0.8 | 1.3 | 2.0 | 4.5 |
| $T_{\text{GR, lower}}$ | $^{\circ}\text{C}$ | 827 | 833 | 838 | 838 | 837 |
| $T_{\text{GR, upper}}$ | $^{\circ}\text{C}$ | 935 | 936 | 938 | 934 | 947 |
| $T_{\text{CR, outlet}}$ | $^{\circ}\text{C}$ | 947 | 944 | 944 | 941 | 964 |

gasification agent resulted in a higher $P_{\text{CR}}/P_{\text{GR}}$ ratio. This phenomenon can be declared by the fact that CO_2 gasification, where the RWGS and the Boudouard reaction are predominated to take place, required more heat and therefore a higher input of additional fuel into the CR was required. Similar findings can be found in literature [15, 35].

The CO_2 to carbon ratio increased with an increasing value of ϕ_{CO_2} and therefore an increasing amount of CO_2 introduced into the GR as gasification agent. The temperatures in the gasification and the combustion reactors were in the same range for all test runs (830–840 $^{\circ}\text{C}$). In the following, the experimental results are presented. To compare the experimental results with theory, the thermodynamic calculations explained above were used.

Figure 4 shows the course of the main product gas components based on the data of Fig. 5 in the thermodynamic equilibrium depending on the gasification agent. In the thermodynamic equilibrium, the H_2 content decreased and the CO content increased. The CO_2 content showed an increasing trend as well. The water content was quite stable between ϕ_{CO_2} of 0 and 0.68 but decreased for ϕ_{CO_2} of 1.

Figure 5 presents the experimental results of the 5 test runs with increasing ϕ_{CO_2} . CO_2 and CO showed an increasing trend with increasing ϕ_{CO_2} . The opposite phenomenon was seen for H_2 , which was decreasing with increasing CO_2 input. CH_4 slightly declined but remained relatively stable. However, this declining trend could also be an effect of dilution by CO_2 . The water content showed a decreasing trend as well, which can also be seen for the thermodynamic calculations. The trends of the experimental results were in accordance with the trends of the thermodynamic calculations, however, there are high deviations in the amounts of the product gas components. This indicates that it was experimentally not possible to produce this thermodynamically possible

product gas composition in the DFB reactor system. Nevertheless, the thermodynamic calculations provide a good insight into the theoretically possible limits.

Figure 6 shows the deviation from the equilibrium of the RWGS reaction with increasing ϕ_{CO_2} . Findings in literature showed that the deviation of the equilibrium of the RWGS lies on the side of the products between 827 and 838 $^{\circ}\text{C}$ in the thermodynamic equilibrium [37]. This was also the case for pure steam as gasification agent and when CO_2 was added as gasification agent. When ϕ_{CO_2} approaches 1 (100 vol% CO_2), the gas composition was completely on the side of the educts, which was explained by the high amount of CO_2 in the product gas for pure CO_2 gasification. A certain amount of CO_2 was not converted during the gasification process, which diluted the product gas.

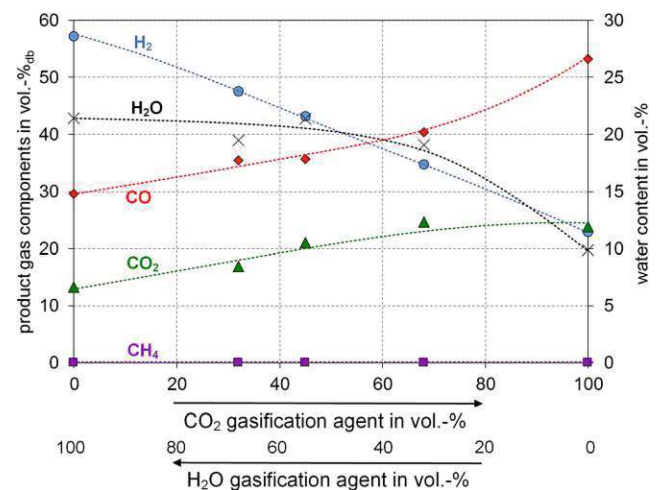


Fig. 4 Change of the product gas composition over increasing ϕ_{CO_2} in the thermodynamic equilibrium at 835 $^{\circ}\text{C}$

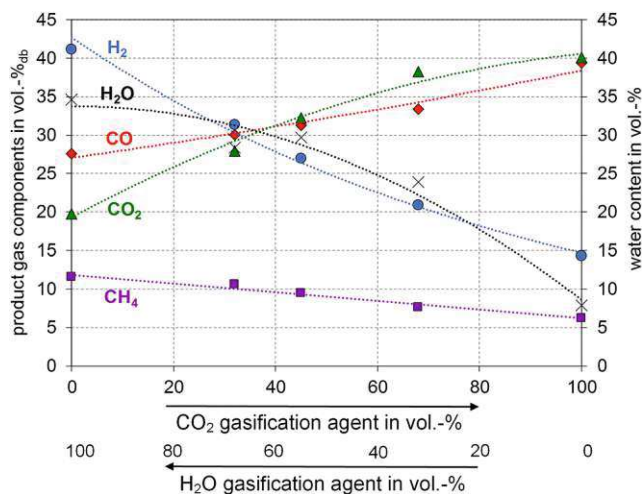


Fig. 5 Change of the product gas composition over increasing ϕ_{CO_2} ; experimental results

To sum up, kinetic effects like a too low contact time between gas and particles could explain the huge deviation from the thermodynamic equilibrium and the high content of CO_2 in the product gas when using pure CO_2 as gasification. It is well known that the reaction rate of the Boudouard reaction is much slower than the reaction rate of the RWGS reaction [38]. Longer contact times between gas and particles would improve the conversion efficiency as stated in literature [39]. Additionally, higher temperatures, especially in the lower gasification reactor ($T_{GR_{lower}}$) would have also been favorable for the progress of the mentioned chemical reactions and thus the conversion efficiency. This assumption was also proven in literature by Sadhwani et al. [36]. If higher temperatures would be reached in the gasification reactor, the conversion efficiency of CO_2 via the RWGS and Boudouard reactions could be enhanced and the deviation from the RWGS equilibrium reduced. In contrast to that, when steam and CO_2 were used as gasification agents (test runs 2, 3, and 4), the applied

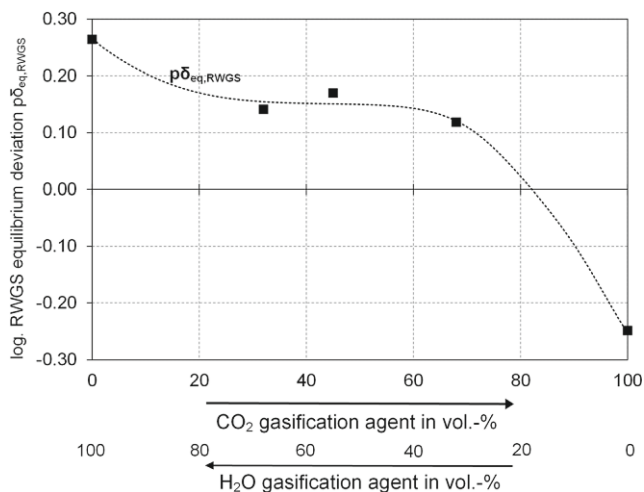


Fig. 6 Change of the deviation from the RWGS equilibrium over CO_2 input as gasification agent

temperatures were sufficient and the deviations from the chemical equilibrium were close to zero.

Table 5 shows the performance indicating key figures of validated data with IPSEpro. The CO_2 conversion rate is at maximum for the pure CO_2 gasification test run. The water conversion decreased. This could be explained by the RWGS reaction, where H_2O was formed (see Eq. 4 in the opposite direction) at temperatures over $800\text{ }^\circ C$. The carbon to CO conversion $X_{C \rightarrow CO}$ is at maximum, when the GR was fluidized with pure CO_2 . An increase in the carbon utilization efficiency X_C with increasing CO_2 as gasification agent was visible. Overall, cold gas efficiencies around 70% were reached for all test runs. The H_2/CO ratio was lowered from 1.49 for $\phi_{CO_2} = 0$ to 0.36 for $\phi_{CO_2} = 1$. The same declining trend was seen for the lower heating value (LHV), which could be explained by the increasing amount of CO_2 in the product gas. The gravimetric tar content of pure steam and pure CO_2 gasification was higher than the one, which was produced when a value of ϕ_{CO_2} of 0.68 was applied as gasification agent. This could be explained by the combined effect of steam and dry reforming reactions [9, 12]. The dust contents were in the range of 0.3 to 1.0 g/m^3_{stp} and are typical values for the gasification with olivine as bed material [21, 24]. The char contents were lower, when CO_2 was present as gasification agent and higher when only steam was used as gasification agent. This could be explained by a higher amount of fuel, which was introduced into the CR for test run 1.

Table 5 Performance indicating key parameters

| Key figure | Unit | Test run | | | | |
|------------------------|--------------------------------|------------------|-------|-------|-------|------------------|
| | | 1 | 2 | 3 | 4 | 5 |
| PGY | $m^3_{stp,db}/kg_{fuel,daf}$ | 1.39 | 1.42 | 1.39 | 1.81 | 2.06 |
| X_{CO_2} | kg_{CO_2}/kg_{CO_2} | - | -0.25 | -0.05 | 0.09 | 0.35 |
| X_{H_2O} | kg_{H_2O}/kg_{steam} | 0.28 | 0.18 | 0.06 | -0.16 | -0.30 |
| $X_{C \rightarrow CO}$ | $kg_{C,CO}/kg_{C,fuel\&fluid}$ | 0.38 | 0.34 | 0.33 | 0.32 | 0.42 |
| X_C | % | 88 | 82 | 79 | 79 | 94 |
| $\eta_{CG,o}$ | % | 72 | 70 | 67 | 66 | 73 |
| H_2/CO | - | 1.49 | 1.04 | 0.86 | 0.63 | 0.36 |
| LHV ^a | MJ/m^3_{stp} | 12.7 | 11.2 | 10.6 | 9.2 | 9.4 |
| Grav. tar ^b | g/m^3_{stp} | 6.7 ^c | n.m. | n.m. | 4.1 | 6.2 ^d |
| Dust ^b | g/m^3_{stp} | 0.3 ^c | n.m. | n.m. | 1.0 | 0.6 ^d |
| Char ^b | g/m^3_{stp} | 2.4 ^c | n.m. | n.m. | 1.5 | 0.5 ^d |

^a Free of tar and char;

^b Measured by the test laboratory for combustion plants a TU Wien;

^c Values from another comparable test run with SW as fuel and olivine as bed material;

^d Values from another comparable operating point with SW as fuel and olivine as bed material;

n.m. not measured;

3.2 Temperature variation under pure CO₂ atmosphere

A temperature variation from 740 to 840 °C with pure CO₂ as gasification agent was conducted. Additionally, the main product gas components based on data of Fig. 8 in the thermodynamic equilibrium depending on the gasification temperature are displayed in Fig. 7. In the thermodynamic equilibrium, CO contents between 39 and 53 vol.-%_{db} were possible, while the amount of CO₂ ranged between 24 and 38 vol.-%_{db}. The H₂ content was around 22 vol.-%_{db} and the CH₄ content was practically zero. The water content decreased from about 14 to 10 vol.-%.

Figure 8 shows the experimental results of the temperature variation when a value of ϕ_{CO_2} of 1 was used as gasification agent. The trends of CO₂ and CO of the thermodynamic calculations were equal to that of the experimental results; however, the amounts showed quite high deviations. The CO content showed an increase from 23 to 38 vol%_{db} and the CO₂ content a decrease from 58 to 39 vol%_{db} in the experimental investigations. In contrast to the quite constant trend of H₂ in the thermodynamic calculations for an increasing gasification temperature, the experimental results showed an increasing course of H₂. CH₄ remained relatively stable with increasing temperature but was almost completely converted in the thermodynamic calculations. The water content showed a decreasing trend for the experimental results and the thermodynamic calculations. In general, there are deviations in the amounts of the product gas components between the thermodynamic calculations and the experimental results, but the trends of CO, CO₂, H₂O, and CH₄ of the thermodynamic calculations corresponded to the trends of the experimental investigations.

Based on the trends of CO and CO₂ in Fig. 8, one can conclude that higher temperatures, over 840 °C, would be favorable for using pure CO₂ as gasification agent. At higher temperatures, the RWGS reaction as well as the Boudouard

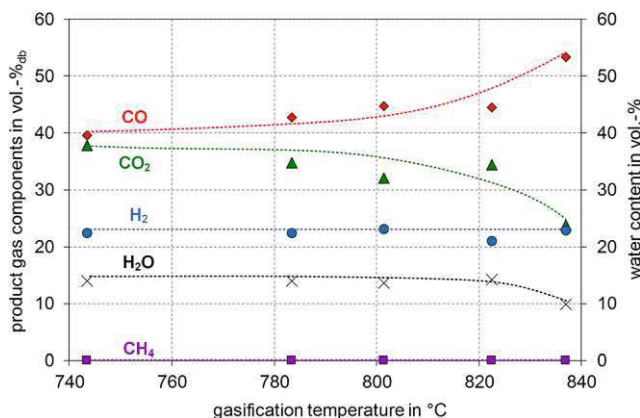


Fig. 7 Change of the product gas composition over gasification temperature in the thermodynamic equilibrium

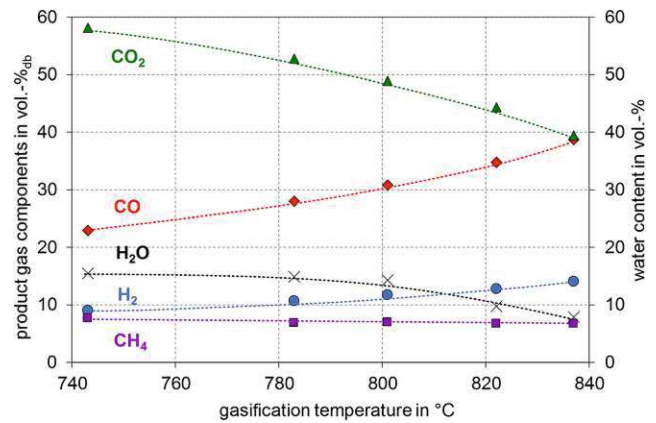


Fig. 8 Change of the product gas composition over gasification temperature; experimental results

reaction, which both favor the production of CO, would take place to a higher extent (see [37, 39]).

Figure 9 depicts the deviation from the RWGS and the Boudouard reaction equilibrium calculated with Eqs. 16 and 17 of the different operating points of the temperature variation displayed in Fig. 8. It is obvious that the deviation from the Boudouard equilibrium was much higher than the deviation from the RWGS equilibrium over the whole temperature range. This points out that the RWGS reaction could be the predominant reaction during the temperature variation. However, further experiments at higher gasification temperatures are recommended to investigate this assumption in more detail.

In Fig. 10, the correlations between the CO₂ conversion, the carbon utilization efficiency, and the overall cold gas efficiency over the increasing gasification temperature during pure CO₂ gasification are shown. With increasing gasification temperature, the CO₂ conversion, the carbon utilization efficiency, and the overall cold gas efficiency increased. This indicated again that higher gasification temperatures would be favorable for utilizing and in parallel converting CO₂ within the DFB reactor system, because an increasing trend of these key figures can be foreseen.

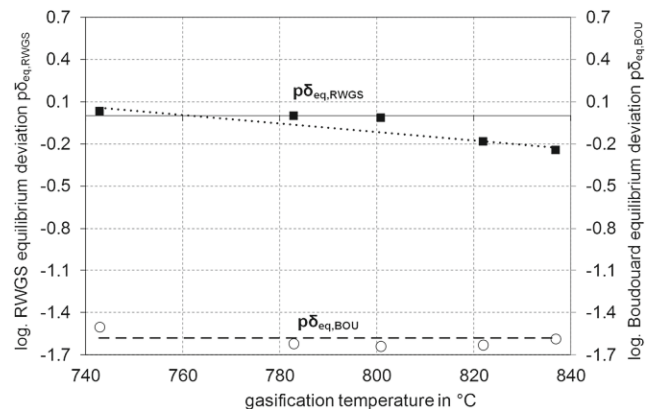


Fig. 9 Change of the deviation from the RWGS and the Boudouard reaction equilibrium over the gasification temperature

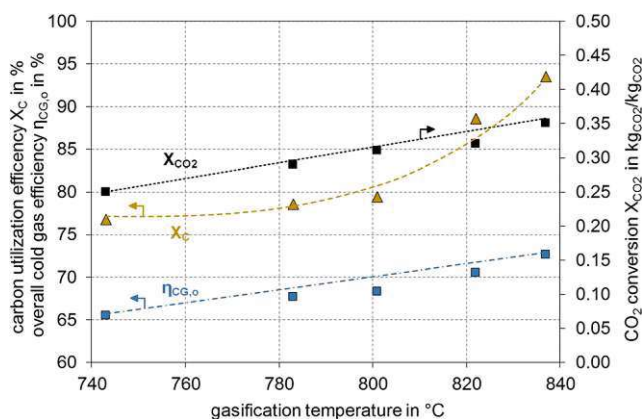


Fig. 10 Correlations between key figures and gasification temperature during pure CO₂ gasification

3.3 Two approaches to determine the conversion of CO₂ during biomass gasification

Due to the reason that it is very difficult to measure the exact conversion of CO₂ during the gasification process in the DFB reactor system, two approaches were investigated and established:

- a carbon balance around the gasification reactor
- and a hydrogen balance around the gasification reactor.

The main material streams around the GR for the carbon and the hydrogen balances are shown in Fig. 11.

3.3.1 Carbon balance

The first approach to investigate the CO₂ conversion during the DFB biomass gasification process was carried out through setting up a carbon balance around the GR. This was carried out for pure steam gasification with $\phi_{CO_2} = 0$ and values of ϕ_{CO_2} of 0.68 and 1, which means pure CO₂ gasification. The carbon balances are shown in Fig. 12. Softwood was used as fuel and olivine as a bed material for all three cases. For the test run with $\phi_{CO_2} = 0$ (pure steam gasification), it was assumed that the whole amount of CO₂ in the product gas was produced from C in the fuel (biomass). This resulted in a value of about 2.6 kg/h C in CO₂ of the product gas, which was formed from 9.4 kg/h of C in the fuel. However, for the runs with $\phi_{CO_2} = 0.68$ and 1, two sources of CO₂ in the product gas were possible: (1) carbon in the fuel (C in fuel) and (2) carbon in CO₂ as gasification agent (C in CO₂ agent) (see Fig. 12).

Therefore, this stream was calculated (a) based on data with $\phi_{CO_2} = 0$ (labeled with a number sign) and (b) based on data of pyrolysis experiments from Neves et al. [40] (labeled with an asterisk). They investigated the production of the pyrolysis gas based on more than 60 different types of biomasses regarding the amount and the composition of the pyrolysis gas depending on the temperature. For the calculation based on

data with $\phi_{CO_2} = 0$, about 2.4 kg/h “C_{CO₂} of C in fuel” and 2.5 kg/h “C_{CO₂} of CO₂ agent” for the test run with ϕ_{CO_2} of 0.68 were generated. For the other case, experimental data of pyrolysis were used for the calculation. This resulted in an amount of 0.9 kg/h “C_{CO₂} of C in fuel”. Through the subtraction of 0.9 kg/h “C_{CO₂} of C in fuel” from the total amount of 4.9 kg/h “C in CO₂” in the product gas, a value of 4.0 kg/h “C_{CO₂} of CO₂ agent” was obtained. The amount of “C_{CO₂} of C in fuel” ranged between 0.9 and 2.4 kg/h and the amount of “C_{CO₂} of CO₂ agent” laid in a range of 2.5–4.0 kg/h.

For the gasification test run with $\phi_{CO_2} = 1$, about 2.4 kg/h “C_{CO₂} of C in fuel” and 4.4 kg/h “C_{CO₂} of CO₂ agent” were produced, calculated based on the reference steam gasification test run. The calculation based on pyrolysis data showed that about 0.9 kg/h “C_{CO₂} of C in fuel” from 6.8 kg/h “C in CO₂” of the PG was generated for the gasification with ϕ_{CO_2} of 1. To sum up, the carbon balances around the GR present the first approach to determine the amount, of how much C of CO₂ in the PG originates from C of CO₂ as gasification agent and how much originates from C in the fuel.

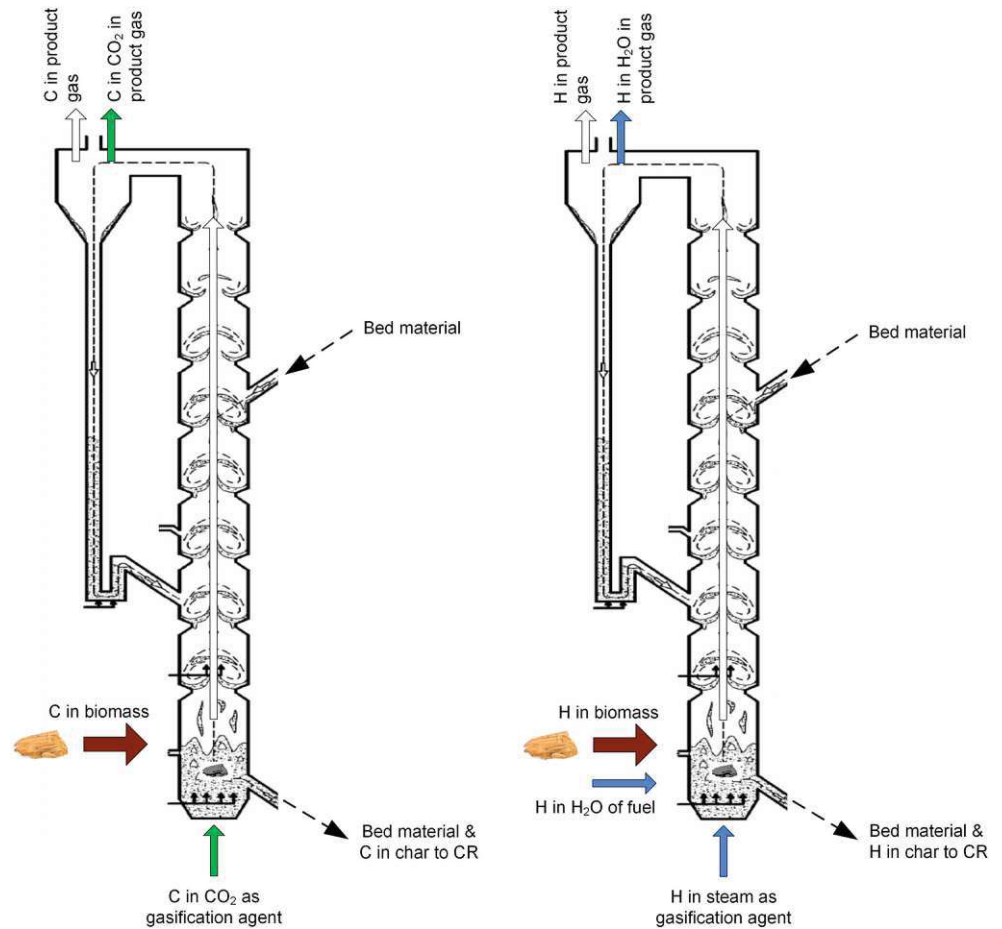
3.3.2 Hydrogen balance

The second approach to investigate the CO₂ conversion during the gasification process was conducted by establishing hydrogen balances around the GR. Based on the experimental results presented above, it can be concluded that the RWGS plays a crucial role during CO₂ gasification. The same is also stated in literature, that the WGS or RWGS reaction acts as a central part during CO₂ gasification [11, 13, 41]. To examine this topic in more detail, hydrogen balances were set up around the GR for a pure steam gasification test run as a reference case and for CO₂ gasification test runs with ϕ_{CO_2} of 0.68 and 1 (see Fig. 13).

H in the fuel (H in fuel), H in H₂O in the fuel (H_{H₂O} in fuel), and H in steam as gasification agent (H in steam) were regarded as input streams. H in H₂O in the product gas (H in H₂O), H in H₂ in the product gas (H in H₂), H in higher hydrocarbons in the product gas (H in C_xH_y), H in tar and char in the product gas (H in tar and char), and H transported to the CR via char together with the bed material (H to CR) were considered as output streams. For the interpretation of the H balances, only the WGS reaction was taken into account. It was assumed that when H in H₂O in the product gas was lower than the sum of H_{H₂O} in fuel and H_{H₂O} in steam, the introduced water into the GR was consumed to produce H₂. This would indicate that the WGS reaction took place. For the reference case with $\phi_{CO_2} = 0$ displayed in Fig. 13, the sum of H in steam and H_{H₂O} in fuel was higher than the amount of H in H₂O in the PG. Thus, the WGS reaction took place.

For the test run with ϕ_{CO_2} of 0.68, H in H₂O was higher than the sum of H_{H₂O} in fuel and H_{H₂O} in steam. This means

Fig. 11 Material streams around the gasification reactor of the DFB gasification system for the carbon balance (left) and the hydrogen balance (right)



that water was produced during the gasification process. Hence, the RWGS reaction was the predominant reaction for this case. The same result was found for $\phi_{CO_2} = 1$. H in H₂O

was higher than the sum of H_{H₂O} in fuel and H_{H₂O} in steam, which also points out that the RWGS reaction proceeded during the gasification process predominantly.

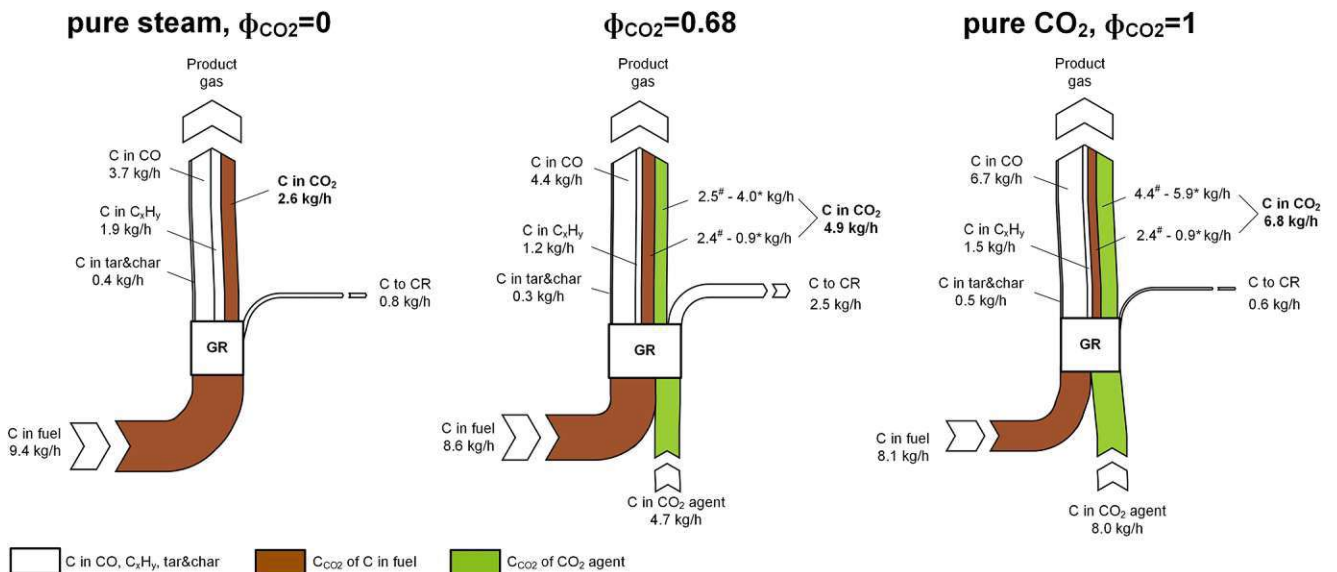


Fig. 12 Carbon balance around the GR for the test runs with ϕ_{CO_2} of 0, 0.68 and 1; * pyrolysis data; # reference steam gasification test run

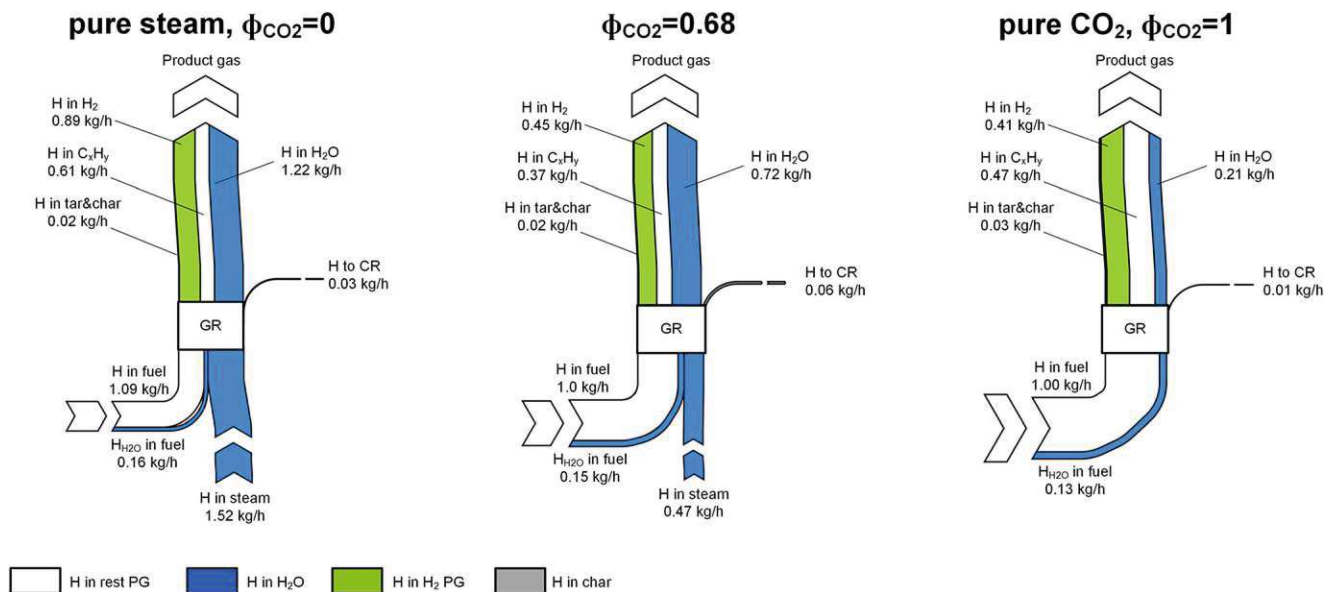


Fig. 13 Hydrogen balances around the GR for test runs with ϕ_{CO_2} of 0, 0.68, and 1

In summary, carbon balances around the GR were established as a first approach to determine the amount of CO₂, which is converted during the gasification process. Based on the results shown in Fig. 12, it can be concluded that a certain amount of CO₂ was converted. Based on the data of the reference test run with pure steam ($\phi_{CO_2} = 0$), it was possible to convert about 26% of C in the fuel to C in CO₂ in the product gas. The rest, 72% of C in the fuel, was converted to other products like CO, C_xH_y, tar, and char. For the test runs with ϕ_{CO_2} of 0.68 and 1, C in CO₂ in the product gas also originated to a certain part from C in CO₂ as gasification agent. For ϕ_{CO_2} of 0.68, between 15% (asterisk sign means pyrolysis data) and 47% (number sign means reference steam gasification test run) of C in CO₂ as gasification was converted to other product gas components, except CO₂. For the test run with ϕ_{CO_2} of 1, which means pure CO₂ gasification, it was possible to convert between 26% (asterisk sign means pyrolysis data) and 45% (number sign means reference steam gasification test run) of C in CO₂ as gasification agent to other product gas components like CO, C_xH_y, tar, or char.

To sum up, these two approaches present a first way to investigate the conversion efficiency as well as the predominant reaction during CO₂ gasification under past assumptions. It was found out that CO₂ is indeed converted in the DFB reactor system to a certain extent and that the RWGS seems to be the predominant reaction, which occurs when using CO₂ as gasification agent.

4 Conclusions and outlook

In the scope of this publication, the influence of the stepwise substitution of steam by CO₂ as gasification was investigated.

Additionally, a temperature variation from about 740 to about 840 °C was carried out under a pure CO₂ atmosphere. To give an overview of the main findings of the performed test runs and investigations, the obtained results can be summarized as follows.

- By substituting steam by CO₂, the product gas was shifted towards higher CO and lower H₂ contents. Using pure CO₂ as gasification agent shows already promising results between 827 and 838 °C. With an increase in CO₂ as gasification agent, an increase in the production of CO, an increase of the carbon utilization efficiency, and an increase of the overall cold gas efficiency was observed.
- The temperature variation indicated that higher temperatures, over 840 °C, would be favorable for pure CO₂ gasification. At higher temperatures, the RWGS reaction as well as the Boudouard reaction could take place to a higher extent. With an increase of the gasification temperature, the CO₂ conversion, the carbon utilization efficiency, and the overall cold gas efficiency could be improved. Thus, an increase of the gasification temperature over 840 °C presents a promising approach to convert a higher amount of CO₂ in the DFB reactor system.
- The carbon balances revealed that between 26% (asterisk sign means pyrolysis data) and 45% (number sign means reference steam gasification test run) of C in CO₂ as gasification agent was converted to other product gas components except CO₂. This implies that CO₂ was indeed utilized in the DFB reactor system at the investigated process conditions. Additionally, the results of the hydrogen balances and that of Fig. 9 indicated that the RWGS reaction might be the predominant reaction during CO₂ gasification at the investigated operation conditions. In general, it must be

noted that the findings of the carbon and hydrogen balances are based on certain assumptions. Therefore, further research regarding these investigations is recommended.

Concluding from all these results, the gasification temperature seems to be the crucial parameter during CO₂ gasification in the DFB reactor system. Future research should focus on investigations at higher temperatures, over 840 °C, in the DFB reactor system to strengthen the outcomes of the carried out investigations.

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Abbreviations CBP, Carbon boundary point; CR, Combustion reactor; DFB, Dual fluidized bed; GC/MS, Gas chromatography coupled with mass spectrometry; GR, Gasification reactor; Grav. tar, Gravimetric tar; IChPW, Institute of Chemical Processing of Coal; LHV, Lower heating value; PGY, Product gas yield; RED, Renewable energy directive; RWGS, Reverse water-gas shift; SW, Softwood; Vol.-%, Volumetric percent; WGS, Water-gas shift; Wt.-%, Weight percent

Subscripts C, Carbon; CR, Combustion reactor; Daf, Dry and ash-free; Db, Dry basis; Fuel, Fuel to gasification reactor; GR, Gasification reactor; H₂O, Water; PG, Product gas; Stp, Standard temperature and pressure; Th, Thermal

Symbols x, y , Stoichiometric factors (-); \dot{m} , Mass flow (kg/s); X , Mass fraction, volume fraction (-); k_{CO_2} , Conversion factor of C to CO₂ of fuel introduced into GR (-); \dot{V}_{PG} , Dry volumetric product gas flow (m³/s); X_{CO_2} , CO₂ conversion (kg_{CO₂}/kg_{CO₂}); X_{C} , Carbon utilization efficiency (%); $X_{\text{C} \rightarrow \text{CO}}$, C to CO conversion (kg_{C,CO}/kg_{C,fuel&fluid}); $\varphi_{\text{CO}_2\text{C}}$, CO₂ to carbon ratio (kg_{CO₂}/kg_C); $\eta_{\text{CG,o}}$, Overall cold gas efficiency (%); \dot{Q}_{loss} , Heat loss (kW); LHV, Lower heating value (MJ/kg); PGY, Product gas yield (m³-_{stp,db}/kg_{fuel,daf}); $P_{\text{CR}}/P_{\text{GR}}$ ratio, Ratio of power introduced into CR to power introduced into GR (-); H₂/CO ratio, Ratio of H₂ to CO of product gas (-); ϕ_{CO_2} , Share of CO₂ used as gasification agent; $p\delta_{\text{eq, WGS}}$, Deviation from water-gas shift equilibrium (-); $K_p(T)$, Equilibrium constant of specific chemical reaction depending on temperature; p_i , Partial pressure of component i ; ν_i , Stoichiometric factor of component i ; $p\delta_{\text{eq, BOU}}$, Deviation from Boudouard equilibrium (-)

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PAPER III

Benedikt F., Schmid J.C., Fuchs J., Mauerhofer A., Müller S., Hofbauer H.

FUEL FLEXIBLE GASIFICATION WITH AN ADVANCED 100 KW DUAL FLUIDIZED BED STEAM GASIFICATION PILOT PLANT

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Responsibility of the author:

- Erected the experimental test plant and design of the experimental campaign
- Created the simulation model used for the validation of the received experimental data
- Interpreted the results

PAPER IV

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HYDROGEN FROM BIOMASS – LARGE SCALE HYDROGEN PRODUCTION BASED ON A DUAL FLUIDIZED
BED STEAM GASIFICATION SYSTEM

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Responsibility of the author:

- Formulated and investigated the determined production concepts
- Created the simulation model used for the calculations
- Interpreted the results
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PAPER V

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THERMODYNAMIC INVESTIGATION OF SNG PRODUCTION BASED ON DUAL FLUIDIZED BED GASIFICATION FOR BIOGENIC RESIDUES

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Responsibility of the author:

- Designed the theoretical investigation
- Supported the interpretation of the results
- Scientific mentoring



Thermodynamic investigation of SNG production based on dual fluidized bed gasification of biogenic residues

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Abstract

Natural gas is an important commodity in the European energy market. The gasification of biogenic residues and the further reaction to a methane-rich gas represent a promising concept for the production of synthetic natural gas on a fossil-free basis. This paper investigates the thermodynamics of methanation in a fluidized bed reactor for different product gas compositions of the dual fluidized bed gasification technology. The investigated product gases range from conventional steam gasification, over CO₂ gasification, to product gases from the sorption enhanced reforming process. All investigated product gases from conventional steam gasification show an understoichiometric composition and therefore require a proper handling of carbon depositions and a CO₂ separation unit downstream of the methanation reactor. The product gas from CO₂ gasification is considered disadvantageous for the investigated process, because it only exhibits a carbon utilization efficiency of 23%. Due to the high flexibility of the sorption enhanced reforming process, a nearly complete methanation of the carbonaceous species is possible without the need for a CO₂ separation step or the addition of steam upstream of the methanation reactor. Furthermore, the carbon utilization efficiency is found to be between 36 and 38%, similar to the results for conventional steam gasification. Temperature and pressure variations allow a thermodynamically optimized operation, which can increase the performance of the methanation and lower the extent of gas upgrading for grid feed-in. Additionally, if a higher hydrogen content in the natural gas grid would be allowed, the overall process chain could be further optimized and simplified.

Keywords Thermodynamics · Fluidized bed methanation · Synthetic natural gas · Dual fluidized bed gasification · Biogenic residues

1 Introduction

Increasing greenhouse gas emissions and the limited availability of primary energy carriers directed the energy policy of the European Union towards sustainable and innovative energy technologies [1]. Natural gas is one of the most important primary energy carriers in Europe, but its availability is heavily dependent on the non-European market. The production of synthetic natural gas (SNG) from biogenic residues offers a promising alternative to the utilization of fossil fuels and

represents a novel concept to support the current energy strategy of the European Union [1, 2].

One possible process route is the dual fluidized bed (DFB) gasification, which allows the utilization of locally available residual biogenic or waste resources and offers possibilities for the production of highly valuable secondary energy carriers on a fossil-free basis. Wilk [3] and Benedikt et al. [4], for example, increased the fuel flexibility of the DFB process towards residues and waste for two generations of a 100 kW_{th} DFB gasifier at TU Wien, while Schweitzer [5] and Schmid et al. [6, 7] further extended the feedstock towards sewage sludge and manure. In addition, the combination of the DFB technology with sorption enhanced reforming (SER) enables the production of a nitrogen-free product gas with adjustable hydrogen to carbon monoxide or hydrogen to carbon dioxide contents [8]. Before the product gas from the DFB gasification process can be fed to the methanation unit, rigorous gas cleaning is required in order to protect the

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downstream equipment and the methanation catalyst. Dust, tar, as well as sulfur and nitrogen containing compounds need to be removed. Gas cleaning is not further elaborated here, but in [9] a comprehensive overview over different gas cleaning strategies is provided. The exothermic methanation itself has been carried out in adiabatic or cooled fixed bed reactors, fluidized bed reactors, three-phase reactors, and structured reactors. The only commercially available reactor types thereof are adiabatic fixed bed reactors [10]. For this reactor type, many similar process concepts were developed mainly between the 1960s and the 1980s. All concepts consist of 2–7 adiabatic reactors with or without intermediate gas cooling and/or gas recycling. Two prominent representatives thereof are the TREMP and HICOM processes. Both utilize three adiabatic reactors with intermediate cooling and gas recycling. They are applied in various coal-to-SNG projects in China, whereas an adapted TREMP process is also installed in the biomass-to-SNG project GoBiGas in Sweden [11]. In general, this reactor type shows disadvantages in terms of heat management and resistance against carbon depositions on the catalyst. Especially, the heat evolution and therefore the temperature peaks in the adiabatic reactors necessitate a reactor cascade and increase the complexity of the process setup [11, 12]. Simultaneously to fixed beds, research activities concerning the development of fluidized beds as methanation reactors started [13]. One of the most prominent fluidized bed concepts is the COMFLUX process, which successfully demonstrated the production of 20 MW_{SNG} from coal. The 1 MW_{SNG} fluidized bed methanation unit connected to the DFB gasifier in Güssing on the other hand was developed by the Paul Scherrer Institut (PSI) and was the first demonstration of a biomass-to-SNG process on a large scale [10]. Fluidized beds can overcome the limitations imposed to fixed beds by their inherently good heat and mass transfer. This results in nearly isothermal operation conditions and an intrinsic catalyst regeneration [14]. However, high particle forces and therefore high attrition rates have prevented the commercialization of fluidized beds in catalytic methanation processes so far. Continued research work is thus put into the development of appropriate catalysts as reported in [15–17]. Other research groups focus on the development of structured reactors. The catalyst is dispersed on thermally highly conducting structures, thus reducing temperature hotspots. This concept, for example, was applied by the Engler-Bunte-Institut for the load-flexible methanation of gasifier product gas with additional hydrogen from electrolysis [12] or by Biegger et al. [18] for a power-to-gas (PtG) concept with a honeycomb methanation catalyst. The variety of reactor types also explains the wide range of operation conditions in the methanation reactor. Temperatures from 250 to 700 °C and pressures from 1 to 87 bar_a have been applied. From a thermodynamic point of view, the methanation is favored at low temperatures and high pressures. A more comprehensive comparison of different reactor concepts can be found in literature [10–13].

Depending on the composition of the raw-SNG after methanation, different gas upgrading steps might be necessary before the gas can be fed to the gas grid. In the case of DFB gasification and the consecutive catalytic methanation, the upgrading steps can include drying, CO₂ separation, and H₂ separation. Various kinds of CO₂ separation technologies have been proposed for this task. Heyne and Harvey [19] compared membranes, pressure swing adsorption (PSA), and chemical absorption with monoethanolamine and concluded that chemical absorption results in the highest cold gas efficiencies. Physical absorption is another method for the removal of CO₂. However, high pressures are usually required for these processes and Gassner and Maréchal [20] showed that it is the least favorable option for allothermal gasification processes compared with PSA and membrane technologies. For the separation of H₂, mainly membrane technologies are proposed [19–21]. However, to the best of our knowledge, no comparative study on H₂ separation technologies for the investigated process has been carried out so far.

In order to feed the generated gas into the Austrian gas grid, the feed-in regulations must be satisfied. In Austria, the limits for the most important accompanying substances are defined at 4 vol.-% for H₂ and 2 vol.-% for CO₂. Limitations for other trace substances and calorific properties are defined as well but are not relevant to this investigation. The values are standardized in [22, 23]. Interestingly, there is no specification mentioned for CO. This is due to the fact that the guidelines were developed for natural gas and later extended to biogas from biological methanation. Both sources do not contain CO and therefore this issue has not arisen. However, for the SNG production via the thermochemical pathway, a limit for the CO content would be necessary to ensure a high quality gas. This is an issue not only in Austria but also all around Europe, since no threshold levels are defined as summarized in [24]. Currently, the discussion focuses on an increased H₂ content in the natural gas grids all around Europe [25]. Studies have shown that up to 10 vol.-% of H₂ in the natural gas grid has no adverse effects on the grid and most applications [26, 27]. However, as long as this is not transferred to national or European law, the strict limits—as defined before—must be fulfilled. Therefore, an alternative is the generation of a CH₄/H₂ mixture, also referred to as hythane, which can be used as a substitute for natural gas directly in industrial applications without the need to feed it into the gas grid first [28].

In Güssing (Austria) and Gothenburg (Sweden), two plants for the conversion of woody biomass to SNG were operated on a large scale. Both concepts utilized a DFB gasification process but applied different gas cleaning and synthesis steps. In Gothenburg, an adapted four-step adiabatic fixed bed methanation process with intermediate cooling was used (TREMP process). Additionally, a water-gas shift reactor, a pre-methanation reactor, and an amine-based CO₂ separation

unit were installed upstream of the methanation reactors. The gasifier system was operated with a thermal fuel power of 32 MW_{th} and therefore was the largest DFB gasifier built so far. The DFB section was operated in total for 12,000 h with wood pellets and later with wood chips and forest residues as feedstock. During the operation, they identified some issues regarding the fuel feeding, the tar formation, and the product gas cooling [29]. Because of these problems, the SNG production periods were quite limited but nevertheless about 67 GWh of SNG was produced in total. From December 2017 to February 2018, they achieved the design goal and the installed capacity of 20 MW_{SNG} was reached. Chemical efficiencies for the production of SNG from 50 to 63% with wood pellets were reported. The carbon utilization efficiency was about 30%, which means that 30% of the carbon in the biomass is transferred to the SNG while the rest is exhausted mainly as CO₂ [30].

In contrast to this concept, the Güssing plant utilized a single fluidized bed methanation reactor and the amine-based CO₂ separation was performed downstream of the methanation reactor. Unlike the GoBiGas plant, a membrane for the separation of excess H₂ was required as the final gas-upgrading step. The 1 MW_{SNG} methanation section was mainly operated in 2009 and was the first plant to produce SNG from woody biomass on a demonstration scale. The gas was not injected into the gas grid but was stored in a compressed natural gas (CNG) tank. Nevertheless, the Austrian gas grid specifications were reached and SNG with about 95 vol.-% CH₄ and 3.8 vol.-% of N₂ in minor amounts of H₂, CO₂, CO, and C₂H₆ was produced. Additionally, a cold gas efficiency of 62% is reported for this process [31]. Because of the application of a fluidized bed methanation reactor the Güssing concept allowed a simpler process setup compared to GoBiGas. However, the Güssing setup was the first of its kind and was not optimized technically. The methanation section applied in Gothenburg on the other hand is commercially available and technically optimized to the specific requirements of the plant [10, 12].

Several other concepts follow the same goal to convert biogenic feedstock to SNG. Anaerobic digestion allows bacteria to convert non-woody biomass to biogas with approximately 60 vol.-% CH₄ and 40 vol.-% CO₂. This biogas can then be upgraded to SNG quality by removing the CO₂ and other minor impurities [32]. The same concept is applied to biogas from landfills or wastewater treatment plants where the biogas is produced naturally without the additional supply of feedstock [33].

Besides biological approaches, a significant amount of research is put into PtG concepts. The hydrogen produced via electrolysis can be utilized to methanate various kinds of carbon resources as the comprehensive review by Götz et al. [34]

shows. One of these sources is the separated CO₂ from biogas plants, which can be upgraded to CH₄ by catalytic methanation instead of the simple exhaustion. One of the most prominent representatives of this technology is the Audi e-gas plant in Germany, which uses a molten salt cooled tube bundle reactor [10]. Besides the classical PtG concepts, also hybrid processes have been developed. For example, Witte et al. [35] directly upgraded the biogas to biomethane on a smaller scale in Switzerland by feeding it together with hydrogen to a bubbling fluidized bed reactor. Instead of the downstream catalytic methanation, Bensmann et al. [36] on the other hand proposed a direct introduction of the hydrogen into the biogas reactor which induced a biological methanation process. Other hybrid concepts add hydrogen to the product gas of a biomass gasification process in order to increase the hydrogen to carbon ratio and therefore increase the overall carbon utilization efficiency of the biomass-to-SNG process. Here, the DemoSNG project is mentioned, where this combination was experimentally tested with a honeycomb-type methanation reactor. It was shown that despite the fluctuating availability of the hydrogen, a continuous production of SNG was possible [37].

From a thermodynamic point of view, the main chemical species which are involved in the methanation reaction system are CH₄, H₂, CO, CO₂, and H₂O. The corresponding reaction equations are the CO methanation (Eq. 1),



the reverse water-gas shift reaction (Eq. 2), and



the CO₂ methanation (Eq. 3) which is a combination of Eq. 1 and Eq. 2.

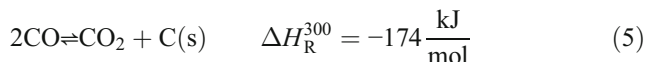


Additionally, the reaction enthalpies at 300 °C ($\Delta H_{\text{R}}^{300}$) are given. Besides these species, the product gas of the DFB gasifier also contains hydrocarbons. As one of the main components, ethylene (C₂H₄) is identified and is thus included here [38]. The hydrogenation to methane is given in Eq. 4.

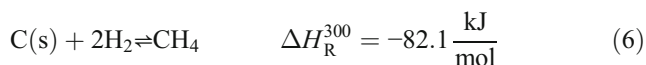


A deactivation mechanism of the catalyst, which cannot be prevented by gas cleaning steps, is the formation of solid carbon on the catalyst. While adsorbed carbon on the catalyst surface is a necessary reaction intermediate during

methanation, the formation of stable deposits leads to catalyst fouling [39]. Thermodynamically, this deposition can be accounted for by the Boudouard reaction in Eq. 5.



The deposited surface carbon can also be hydrogenated to methane according to Eq. 6,



or undergo gasification with steam as shown in Eq. 7 [40].



These reactions show that increased amounts of H_2 , H_2O , or CO_2 in the gasifier product gas might prevent the carbon deposition.

A different form of deposition occurs through the adsorption of hydrocarbons like C_2H_4 on the catalyst surface. Between 500 and 800 °C, the adsorption can lead to coke deposits [40]. In general, there is a large number of different forms and structural types of carbon or coke deposits which can occur at different temperature intervals in methanation processes [41].

If kinetic models are considered, all of the abovementioned reaction pathways have to be taken into consideration. The catalytic methanation of syngas is, however, mostly limited by heat transfer and not by kinetics under typical operating conditions. This limitation mostly applies for fixed bed reactors and thus multiple reactors with intermediate cooling are necessary in order to manage the heat released by the exothermic reactions [10]. Fluidized beds were shown to overcome this limitation and allow a low-temperature methanation in a single reactor step. The process was mainly found to be limited by the mass transfer between the bubble and the dense phase of the fluidized bed. Nevertheless, the gas composition is close to the thermodynamic equilibrium for temperatures down to 320 °C and kinetic limitations apply for lower temperatures as some studies confirm [17, 42, 43]. Additionally, the adjustment of the H_2/CO ratio of the feed gas to the required level of three can be directly carried out in the fluidized bed methanation reactor. Fixed bed applications usually require a separate water-gas shift reactor upstream of the methanation for this task [44, 45]. A thermodynamic calculation including the water-gas shift reaction thus provides a good estimation of the expected gas composition. Because of the broad variety of possible carbon species, deviations from the thermodynamic equilibrium for carbon depositions have to be expected [10]. Nevertheless, graphitic carbon has previously been used to elucidate this issue since kinetic models are often only valid for specific reaction conditions and catalysts [46].

Extensive studies have been performed on the thermodynamics of methanation. Bia et al. [39] used ternary diagrams to visualize the calculated boundaries of carbon formation under methanation conditions. Frick et al. [46] applied the same method but extended the investigation to different feed gas mixtures. They concluded that ternary diagrams are an appropriate tool for the design of methanation processes. Gao et al. [47] performed a systematic thermodynamic investigation on the methanation of CO and CO_2 under varying parameters like pressure, temperature, or the H_2/CO ratio. As a result, they give general indications on the effects of the parameter variations. Other research groups extended the modelling to a larger part of the process setup and used different modelling approaches. For example, Witte et al. [48] used rate-based modelling and investigated different combinations of methanation reactors and hydrogen membranes to upgrade biogas from biological digestion to biomethane. In order to upgrade the biogas, they proposed a PtG concept with renewable hydrogen via electrolysis. They concluded that, in order to reach the gas grid requirements, a combination of a bubbling fluidized bed reactor with a second-stage fixed bed methanation unit or a gas separation membrane are the technically and economically favorable options [49]. Neubert [50] proposed a similar two-stage methanation setup within the PtG concept. The first stage consists of a structured methanation reactor followed by an intermediate water condensation and a second-stage fixed bed reactor. Within his work, he elaborately used thermodynamic models and ternary diagrams to define the optimal CO_2 removal as well as steam and hydrogen addition in general. For the production of SNG from coal, Liu et al. [51] used thermodynamic calculations in Aspen Plus to find the most suitable process setup. They concluded that a circulating fluidized bed followed by a second-stage fixed bed methanation reactor poses the most promising concept. For small-scale air blown biomass gasifiers Vakalis et al. [52] thermodynamically modelled the methanation with additional hydrogen. They reached CH_4 concentrations of only 40 mol.-% because of the high N_2 concentrations inherent to the product gas of air-blown gasifiers. The modelling of a combination of the SER process with a TREMP methanation process was carried out in [53]. They reached cold gas efficiencies of 62% with this setup and about 60% when additional hydrogen from an electrolyzer was added. In [54], three different gasifier types were compared for the production of SNG with the conclusion that allothermal gasification systems, like the DFB system, result in the highest overall efficiencies. Rönsch et al. [11] give a comprehensive overview over many different modelling approaches for methanation reactors and SNG production plants. Depending on the scope of the study, the investigations range from detailed one-, two-, or three-dimensional methanation reactor models to flow sheet simulations of entire SNG process chains with zero-dimensional equilibrium models. However, no evaluation of

the results from the latest DFB gasifier design in terms of SNG production has been carried out. Furthermore, no detailed thermodynamic analysis of the SNG production from biogenic residues exists and no evaluation of the process in terms of the carbon utilization efficiency is reported.

In this paper, a thermodynamic model of a fluidized bed methanation reactor is developed and applied to specific feed gas mixtures, which have been obtained by experimental gasification test runs of different biogenic residues with a new generation of a 100 kW_{th} DFB gasifier at TU Wien. The chosen feed gas compositions for the methanation aim at covering the broad range of product gas compositions which can be produced by the DFB gasifier. The results show a detailed thermodynamic analysis of the raw-SNG gas compositions and key values for different feed gas mixtures and varying operation conditions like temperature and pressure. These results are discussed and evaluated in terms of their suitability for a feed-in into the natural gas grid. Because of the different process setups regarding the CO₂ separation unit in Güssing and Gothenburg, the placement of the CO₂ separation unit upstream or downstream of the methanation reactor is discussed as well.

2 Concept and methodology

In order to calculate the thermodynamic equilibrium, only four of the seven reaction equations (Eq. 1 to Eq. 7) need to be considered. Otherwise, the system would be overdetermined, because only four equations are linearly independent of each other. For example, the CO₂ methanation reaction can be seen as the reversed water-gas shift reaction followed by the CO methanation.

Thermodynamic calculations were performed with HSC Chemistry 6 and MATLAB. HSC Chemistry is a commercially available software tool for thermodynamic calculations and contains a database with thermodynamic property data. It calculates the thermodynamic equilibrium concentrations with the Gibbs free energy minimization method. For the purpose of this work, a MATLAB-based program for the thermodynamic equilibrium calculations was developed. This program calculates the thermodynamic equilibrium based on the temperature dependent thermodynamic property data from HSC Chemistry. The solution was obtained by numerically solving the equilibrium constant expressions for each reaction equation. The equilibrium concentrations were then automatically plotted over temperature and pressure. The model was validated by comparing the calculated results on a random basis to results obtained with HSC Chemistry. This comparison showed that the model is highly accurate.

Figure 1 visualizes the modelling approach with a basic flowsheet. In the DFB gasification process, the feedstock is

converted to the gasifier product gas. The validated results for a multitude of experimental test runs in a 100 kW_{th} DFB gasifier at TU Wien have already been published elsewhere (see Sect. 3) and are used as a basis for the modelling of the methanation in this study. In the gas cleaning section, impurities like dust, tar, as well as sulfur and nitrogen containing contaminants are removed. The gas cleaning is not included in the model because it does not influence the thermodynamic calculations of the methanation. Therefore, the gas cleaning is treated as a black box which removes all impurities except ethylene. Ethylene was found to be the main hydrocarbon in the gasifier product gas besides CH₄ which is not removed by conventional gas cleaning steps like scrubbers or activated carbon filters. Besides ethylene, also hydrocarbons like benzene, toluene, xylene, or naphthalene are often not completely removed [55–57]. In this investigation, they are neglected because the concentrations are comparably low. After the gas cleaning, the gasifier product gas is fed to the methanation unit. Here, the thermodynamic model is applied and the conversion of the feed gas to raw-SNG is calculated. Since the raw-SNG does not fulfill the requirements of the gas grid, the necessary gas upgrading steps are also discussed but not modelled. Optionally, the CO₂ separation can be carried out as shown in Fig. 1 or as part of the raw-SNG upgrading after the methanation reactor. The standard setup in this investigation is the downstream CO₂ separation as part of the raw-SNG upgrading. However, also the upstream CO₂ separation as indicated in Fig. 1 is discussed.

The main focus of this investigation is a low-temperature methanation (300 °C) at ambient pressure. These parameter settings result from the current efforts on the scientific investigation of a novel bench-scale fluidized bed methanation setup for the given parameters. As the DFB gasification process also operates at ambient pressure an additional energy input for compression is avoided. This bench-scale methanation setup has been designed and built at TU Wien and is currently in the commissioning phase. Nevertheless, also a temperature variation from 200 to 500 °C and a pressure range from 1 to 10 bar_a are investigated. While thermodynamic calculations are in general independent of the reactor design, the validity of the underlying assumptions is nevertheless defined by the process-related circumstances. In this study, this translates to the following assumptions: (i) the water-gas shift reaction takes place simultaneously to the methanation reactions in one reactor without a need for a prior adjustment of the H₂/CO ratio, (ii) C₂H₄ is hydrogenated to methane, and (iii) despite the high exothermicity of the reactions, a low-temperature methanation (e.g. 300 °C) is possible in one reactor. These assumptions are only valid for fluidized bed methanation but would not be valid for fixed bed methanation as reported in literature [10, 44, 45, 58]. Graphite is chosen as the prevailing carbon species, since Frick et al. [46] found that

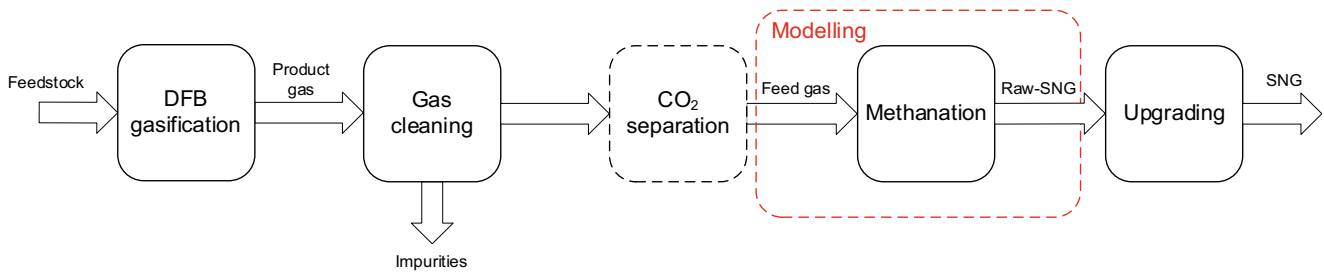


Fig. 1 SNG production flowsheet via the DFB gasification route; the highlighted area defines the modelled part of the process in this study

the Gibbs free energy is lower than for amorphous carbon and is thus preferentially formed.

In order to classify the feed gas composition, the stoichiometric number (SN) is defined in Eq. 8.

$$SN = \frac{y_{H_2}}{3y_{CO} + 4y_{CO_2} + 2y_{C_2H_4}} \quad (8)$$

SN gives the ratio between the molar fraction of H_2 (y_{H_2}) to the molar fractions of the carbonaceous species in the feed gas which react to CH_4 . If SN is equal to 1, there is a stoichiometric amount of H_2 available according to Eqs. 1, 3, and 4. Because the regarded pressures in this study are relatively low, an ideal gas behavior is assumed and molar fractions are thus equal to volume fractions. The definition of SN is not unambiguous, because the chemical equilibrium is influenced by all available species and therefore also by CH_4 and H_2O . Nevertheless, it allows an approximate classification of the feed gas mixture. Typical product gases from the DFB gasification of biogenic feedstock show similar CH_4 concentrations; moreover H_2O concentrations in the feed gases are assumed 0. The latter is attributed to the required gas cleaning which is conventionally carried out at low temperatures [59]. If similar CH_4 concentrations and a water-free feed gas are assumed, the implementation of SN is justified.

Additionally, the CH_4 yield (Y_{CH_4}) is defined in Eq. 9. It describes how much of the carbon in the feed gas is converted to CH_4 .

$$Y_{CH_4} = \frac{\dot{n}_{CH_4,eq}}{\sum_i N_i \dot{n}_{i,feed}} \times 100 \quad (9)$$

The carbon yield (Y_C) in Eq. 10 is a measure for carbon deposition.

$$Y_C = \frac{\dot{n}_{C,eq}}{\sum_i N_i \dot{n}_{i,feed}} \times 100 \quad (10)$$

Index i refers to the carbonaceous species in the feed ($i = CH_4, CO, CO_2, C_2H_4$), and N_i is the number of carbon atoms in species i .

The CO conversion (X_{CO}) in Eq. 11 gives the amount of CO which is converted during the reaction.

$$X_{CO} = \frac{\dot{n}_{CO,feed} - \dot{n}_{CO,eq}}{\dot{n}_{CO,feed}} \times 100 \quad (11)$$

Analogously to Eq. 11, the CO_2 conversion (X_{CO_2}) is defined in Eq. 12.

$$X_{CO_2} = \frac{\dot{n}_{CO_2,feed} - \dot{n}_{CO_2,eq}}{\dot{n}_{CO_2,feed}} \times 100 \quad (12)$$

In order to assess the performance of the overall process, the carbon utilization efficiency (η_C) is introduced (Eq. 13). It sets the amount of carbon in the methane of the raw-SNG ($\dot{n}_{CH_4,eq}$) in relation to the amount of carbon which is introduced to the process via the feedstock ($\dot{n}_{C,feedstock}$). If CO_2 is used as gasification agent, the amount of carbon in the gasification agent must be considered as well ($\dot{n}_{C,gasif}$). The carbon utilization efficiency illustrates how much of the carbon is valorized as CH_4 in the SNG and how much is “lost” mainly as CO_2 .

$$\eta_C = \frac{\dot{n}_{CH_4,eq}}{\dot{n}_{C,feedstock} + \dot{n}_{C,gasif}} = \eta_{C,DFB} \times Y_{CH_4} \quad (13)$$

An analogous way to calculate the carbon utilization efficiency is by the multiplication of the carbon utilization efficiency over the DFB gasifier ($\eta_{C,DFB}$) and the methane yield in the methanation section (Y_{CH_4}). In this paper, $\eta_{C,DFB}$ is calculated from the validated results of test runs with the 100 kW_{th} DFB gasifier at TU Wien. This value is therefore only valid for this gasifier. An extrapolation of $\eta_{C,DFB}$ to large-scale gasifiers is not recommended since the internal energy and mass balances might differ. In this small-scale gasifier, the high heat losses are balanced by the addition of heating oil in the combustion section of the DFB process which is not the case for large-scale plants. Large-scale gasifiers exhibit much lower heat losses, but, depending on the feedstock, a partial recycling of product gas to the combustion section might still

be necessary. The recycled amount of product gas is not available for methanation. This factor cannot be considered in the calculation, and the shown results therefore need to be seen as a maximum.

Additionally, the minimum amount of steam (H_2O_{feed}), which needs to be added upstream of the methanation reactor to prevent carbon formation, is introduced. In order to calculate H_2O_{feed} , every investigated reaction condition with each feed gas is checked for the possibility of carbon formation. If carbon formation is possible, the water content in the feed gas is incrementally increased until the thermodynamic possibility for carbon formation yields 0. At this point, H_2O_{feed} can be obtained. Furthermore, gas cleaning is not within the scope of this study and the feed gas mixtures for the methanation are assumed free of impurities and other minor components. Besides, kinetics or heat and mass transfer phenomena are not considered.

3 Results and discussion

Table 1 shows the investigated feed gas compositions for the methanation derived from DFB gasification. In the upper part of the table, the operational parameters of the DFB gasification process are shown. All displayed feed gas compositions are obtained with a new generation 100 kW_{th} DFB gasifier at TU Wien. The DFB process is not elaborated in this study and further information can be found in literature [4, 7, 8, 56, 60, 61]. The lower part of Table 1 depicts the gas compositions which are derived from the DFB gasification process and are in further consequence used as the feed gas compositions for the methanation process. All feed gases are assumed to be free of

H_2O . Feed gas no. 1 shows a typical SER product gas with a high hydrogen content. Limestone (L) is used as bed material, and bark (BA) is chosen as feedstock. Feed gas no. 2–no. 4 present product gases from conventional gasification. With feed gas no. 2, the same fuel and bed material as with feed gas no. 1 is used but the gasification temperature is higher which results in lower H_2 and higher CO and CO_2 contents. For feed gas no. 3, lignin (LI) is used as fuel and olivine (O) as bed material. Sewage sludge (SS) and an olivine/limestone mixture (O/L) are the basis for feed gas no. 4, which results in low H_2 and high CO_2 contents. For feed gas no. 5, a CO_2/H_2O mixture is used as gasification agent and rapeseed cake (RSC) and O as fuel and bed material, respectively. This results in even lower H_2 and high CO and CO_2 concentrations. Feed gas no. 6 shows a temperature variation for SER gasification. This is included to demonstrate the adaptability of the DFB gasification process to the requirements of the methanation process (cf. Fig. 6). Data for this variation is only available for softwood (SW) as feedstock.

In Fig. 2, the results of the chemical equilibrium calculations at 300 °C and 1 bar_a are shown for feed gas nos. 1–5. The volume fractions of the dry gas components after the methanation (referred to as raw-SNG) and the water content of the raw-SNG ($H_2O_{\text{raw-SNG}}$) as well as the minimum required water content in the feed gas in order to prevent carbon deposition (H_2O_{feed}) are depicted.

Additionally, Table 2 lists some key figures as defined in Eqs. 8–12 complementary to the results in Fig. 2. In Fig. 2a and the left part of Table 2 (without H_2O_{feed}), the results for a water-free feed gas are displayed. Figure 2b and the right part of Table 2 (with H_2O_{feed}) display the results with steam addition to the feed gases in order to prevent carbon formation. C_2H_4 is not depicted in any of the figures, because it is

Table 1 Investigated feed gases

| DFB parameters | Unit | Feed gas number | | | | | |
|-------------------------------------------------------|--------|-----------------|--------|--------|------------------|---------------|-----------|
| | | 1 | 2 | 3 | 4 | 5 | 6 |
| Source | – | [8] | [4] | [56] | [7] | [60] | [61] |
| Gasification agent | – | H_2O | H_2O | H_2O | H_2O | CO_2/H_2O^a | H_2O |
| Feedstock | – | BA | BA | LI | SS | RSC | SW |
| Bed material | – | L | L | O | O/L ^b | O | L |
| Gasification temperature | °C | 625 | 761 | 789 | 800 | 840 | 582–797 |
| Combustion temperature | °C | 820 | 998 | 945 | 945 | 938 | 830–1041 |
| Feed gas composition to methanation (water-free feed) | | | | | | | |
| H_2 | vol.-% | 68.3 | 51.1 | 42.6 | 35.6 | 25.8 | 71.1–47.6 |
| CO | vol.-% | 6.5 | 17.9 | 21.2 | 13.7 | 32.1 | 7.3–21.6 |
| CO_2 | vol.-% | 8.9 | 22.4 | 21.8 | 36.5 | 33.7 | 4.1–23 |
| CH_4 | vol.-% | 14.5 | 8.0 | 12.0 | 11.7 | 7.3 | 17.4–8.8 |
| C_2H_4 | vol.-% | 1.9 | 0.6 | 2.4 | 2.5 | 1.1 | 1.9–0.5 |

^a $CO_2/H_2O = 68/32$ vol.-%

^b O/L = 80/20 wt.-%

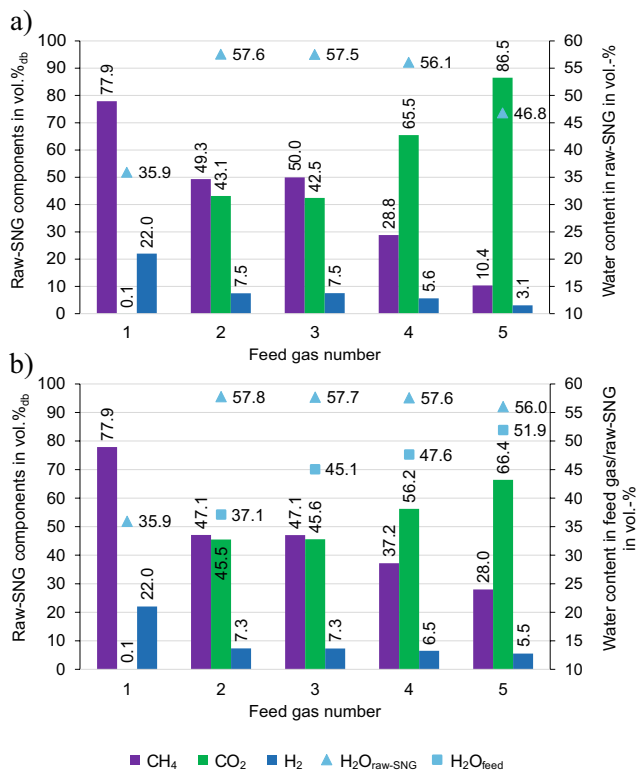


Fig. 2 Raw-SNG gas composition for feed gas nos. 1–5 at 1 bar_a and 300 °C. **a** Water-free feed gas. **b** Feed gas with steam addition to prevent carbon deposition

completely converted under all investigated conditions. CO is also not shown in Fig. 2 because it is almost entirely converted (see Table 2) and only trace amounts remain in the raw-SNG. The feed gases are displayed in descending order for *SN* in Fig. 2 as well as in Table 2. This results in a decreasing trend for CH₄ and H₂ and an increasing trend for CO₂ in the raw-SNG. Analogously, the methane yield and the CO₂ conversion drop significantly with understoichiometric feed gases.

A closer look at the results for the water-free feed gases reveal that the SER feed gas (feed gas no. 1) allows an almost complete conversion of CO and CO₂ to CH₄. Thus, no CO₂ separation is necessary. In addition, no carbon formation is thermodynamically expected. However, 22 vol.-%_{db} of H₂ is still in the raw-SNG and needs to be separated below 4 vol.-%

before grid feed-in according to the Austrian regulations [22, 23]. Feed gas nos. 2–5 result in a lower CH₄ content and a higher CO₂ content. The CO conversion is almost complete even though *SN* is well below one for feed gas nos. 2–5. This is possible because thermodynamically the feed-CO is rather converted to solid carbon than left unreacted in the raw-SNG. This results in severe carbon depositions with a carbon yield as high as 54.5%. More than half of the carbon in the feed would be deposited on the catalyst. This deposition would result in a high loss of carbon and deactivate the catalyst. Therefore, feed gas nos. 2–5 should not be introduced into the methanation reactor without a previous steam addition. Thus, in Fig. 2b and the right part of Table 2, the results with the addition of steam to the feed gas are depicted. The amount of steam added corresponds to the minimum amount needed to prevent carbon formation. For feed gas no. 1, no steam addition is necessary and therefore the results are the same as in Fig. 2a. All other feed gases require steam addition in a range of 37 to 52 vol.-%. The raw-SNG for these feed gases therefore shows a different composition compared with the water-free feed gases. For feed gas nos. 2 and 3, about half the raw-SNG consists of CH₄, the rest is CO₂ and H₂. For feed gas nos. 4 and 5, CO₂ constitutes the main component in the raw-SNG with a CH₄ yield of approximately 40% and 30%, respectively. All four gas compositions require the separation of both CO₂ and H₂ before grid feed-in, even if the less stringent limitation of 10 vol.-% H₂ is applied. Compared with the results of the dry feed gases, the CH₄ yield is slightly increased but the CO₂ conversion is significantly lowered. All four gases show a negative CO₂ conversion, which implies that more moles of CO₂ are produced than consumed during the reaction. The influence of the steam addition on the reactions can be pictured as follows: The water-gas shift reaction (Eq. 2) proceeds towards CO₂ and H₂. This way, more H₂ is available for the methanation of CO and less CO needs to be methanated because it is shifted towards CO₂. The additional H₂ is used to hydrogenate the solid carbon. From this point of view, it also becomes apparent that the CO₂ conversion is less compared with the results of the water-free feed or even negative. There are of course many ways to illustrate this effect. The reaction pathway is only important for the consideration

Table 2 Key figure results of the equilibrium calculations

| Parameter | Unit | Feed gas number (without H ₂ O _{feed}) | | | | | Feed gas number (with H ₂ O _{feed}) | | | | |
|------------------------------------|------|-------------------------------------------------------------|------|------|------|------|----------------------------------------------------------|------|-------|-------|-------|
| | | 1 | 2 | 3 | 4 | 5 | 1 | 2 | 3 | 4 | 5 |
| <i>SN</i> | - | 1.16 | 0.35 | 0.27 | 0.19 | 0.11 | 1.16 | 0.35 | 0.27 | 0.19 | 0.11 |
| <i>Y</i> _{CH₄} | % | 99.9 | 28.2 | 24.6 | 14.4 | 5.2 | 99.9 | 50.8 | 49.2 | 39.8 | 29.6 |
| <i>Y</i> _C | % | 0 | 47.2 | 54.5 | 52.8 | 50.9 | 0 | 0 | 0 | 0 | 0 |
| <i>X</i> _{CO} | % | 100 | 99.9 | 99.9 | 99.8 | 99.9 | 100 | 99.8 | 99.8 | 99.6 | 99.8 |
| <i>X</i> _{CO₂} | % | 99.7 | 45.6 | 42.7 | 39.9 | 2.2 | 99.7 | -8.5 | -34.9 | -10.3 | -57.1 |

of kinetic effects and does not influence the thermodynamic equilibrium. Table 2 shows that the CO conversion remains almost complete for all feed gases. Nevertheless, the CO₂ methanation is found to be kinetically inhibited even for very low CO concentrations [62]. For feed gas no. 1, only 7 ppm_{v,db} of CO remain in the raw-SNG in the thermodynamic equilibrium. At least 600–700 ppm_{v,db} need to be expected for feed gas nos. 2–5. As long as there are no regulations on the allowed CO content, no statement about the grid feed-in can be made. The authors recommend a threshold value for CO if the production of SNG via the thermochemical pathway is further pursued at industrial scale.

3.1 Investigation of the sewage sludge product gas

In the following section, a more in-depth discussion of the feed gas derived from SS gasification follows (feed gas no. 4). Because of the expected carbon deposition for this feed gas composition, H₂O should be added if a long catalyst lifetime and a high conversion efficiency are aimed at. This was already discussed in the previous section. Hence, Fig. 3 depicts the raw-SNG gas composition after the addition of steam for a temperature variation from 200 to 500 °C and pressures of 1, 5, and 10 bar_a (Fig. 3b). The amount of steam added corresponds to the minimum amount needed to prevent carbon

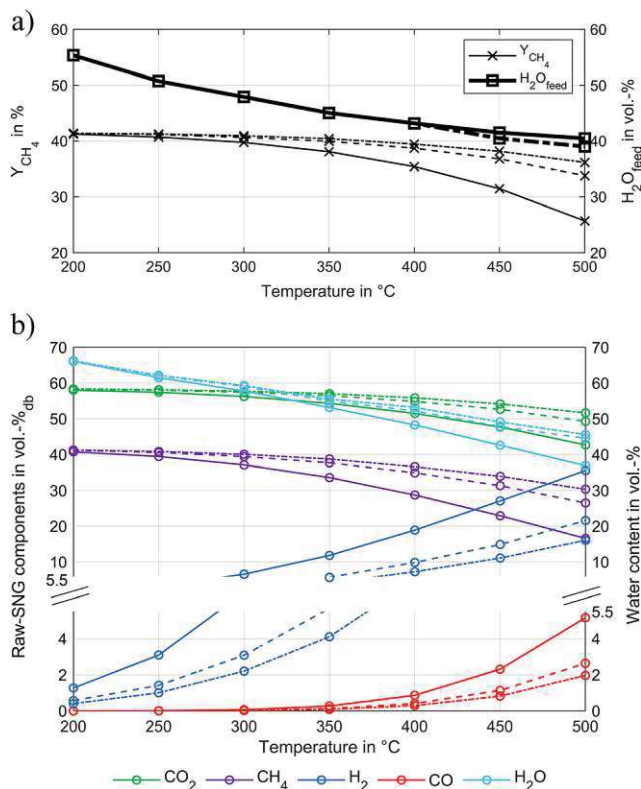


Fig. 3 Temperature and pressure variation for feed gas no. 4 in the thermodynamic equilibrium: 1 bar_a (full line), 5 bar_a (dashed line), and 10 bar_a (dash-dotted line). **a** CH₄ yield and feed water content. **b** Raw-SNG gas composition

deposition. This minimum volume fraction of H₂O in the feed gas (H_2O_{feed}) as well as Y_{CH_4} is also displayed (Fig. 3a). With increasing temperature, less CH₄ and CO₂ and more CO and H₂ are present. Accordingly, the CH₄ yield decreases from 41 to 26% with increasing temperature at 1 bar_a. H_2O_{feed} decreases from 55 to 40 vol.-% within the displayed temperature range. Nevertheless, the methanation is preferred at low temperatures from a thermodynamic point of view if the additionally required steam is not seen as the decisive factor. Especially, the low methane yield and the strongly rising CO content at higher temperatures make low-temperature methanation attractive. Pressure only has a significant influence on the gas composition at higher temperatures. At 500 °C, Y_{CH_4} can be substantially elevated and the H₂ content significantly lowered if the pressure is increased to 5 bar_a. A further pressurization only allows a minor improvement of Y_{CH_4} but still reduces the H₂ content by 5 percentage points. At 200 °C, Y_{CH_4} is almost constant for all pressures. For H_2O_{feed} , hardly any influence of pressure can be observed.

In general, this feed gas shows a rather unfavorable composition for methanation. The stoichiometric number is far below 1, and the CO₂ content in the feed gas is even higher than the H₂ content. For grid feed-in, the CO₂ needs to be separated from the raw-SNG. A maximum of only 2 vol.-% is allowed. A H₂ content below the allowed threshold level of 4 vol.-% after CO₂ separation and without an additional H₂ separation unit could be achieved by increasing the pressure at 260 °C to 5 bar_a or at 280 °C to 10 bar_a. If the stringent feed-in specification of the natural gas grid is loosened and 10 vol.-% H₂ is allowed in the future, the methanation can be performed at 350 °C at 10 bar_a, 320 °C at 5 bar_a, or 270 °C at 1 bar_a. Even though there is only a slight influence of pressure on the gas composition at these temperatures, a small increase can nevertheless enable the grid feed-in without an H₂ separation unit. This is especially interesting if 10 vol.-% of H₂ would be allowed in the gas grid because the reaction temperature would be in a range where catalysts were found to be kinetically active. If the desired commodity is hythane, only CO₂ separation is necessary and the discussion concerning the H₂ content and the pressurization can be neglected.

3.2 Investigation of the feed gases with upstream CO₂ separation

Firstly, the upstream CO₂ separation is discussed with the sewage sludge product gas (feed gas no. 4) in detail before the discussion is extended to all other investigated feed gas compositions. In Fig. 4, the equilibrium calculations for feed gas no. 4 in a temperature range from 200 to 500 °C and pressures of 1, 5, and 10 bar_a are shown. In contrast to Fig. 3, the CO₂ separation is done upstream of the methanation reactor as demonstrated in the GoBiGas project in

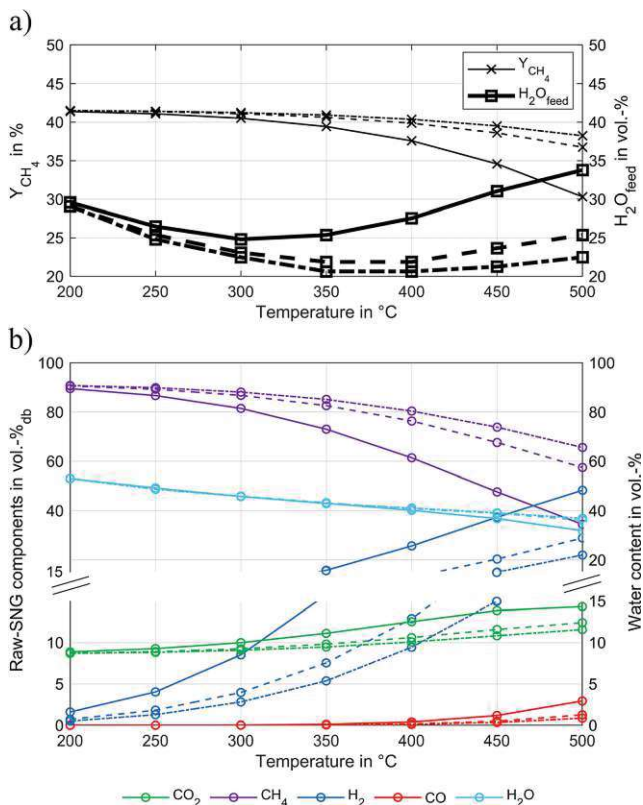


Fig. 4 Temperature and pressure variation for feed gas no. 4 in the thermodynamic equilibrium with CO_2 separation upstream of the methanation: 1 bar_a (full line), 5 bar_a (dashed line), and 10 bar_a (dash-dotted line). **a** CH_4 yield and feed water content. **b** Raw-SNG gas composition

Gothenburg. The feed gas to the methanation is therefore free of CO_2 . In order to enable a fair comparison to Fig. 3, the calculation of the methane yield includes the CO_2 separation step in this case. A comparison between Figs. 3 and 4 reveals that Y_{CH_4} is slightly increased, whereas the required amount of steam in the feed is substantially lowered. The lower amount of steam in the feed could lead to a more energy-efficient process because less steam needs to be provided to the methanation reactor. Interestingly, at higher temperatures, H_2O_{feed} increases again and the pressure sensitivity is much more pronounced in comparison. The H_2 content is a little higher and the CO content slightly lower (e.g., 250 ppm_{v,db} compared with 667 ppm_{v,db} at 300 °C and 1 bar_a) comparing CO_2 separation upstream and downstream of the methanation reactor. The CO_2 content is in a range of 9 to 15 vol.-%_{db}, which implies that CO_2 is formed during the reaction. The CO_2 content as well as the higher H_2 and lower CO content in the raw-SNG can be explained by the water-gas shift reaction (Eq. 2) which is shifted towards CO_2 and H_2 due to the missing CO_2 and the understoichiometric H_2/CO ratio in the feed. In this case, the CO_2 needs to be separated again, which requires a second CO_2 separation unit. The same applies for feed gas nos. 2, 3, and 5. These feed gases also have a H_2/CO ratio

below 3, and therefore CO_2 is formed during the reaction in an order that it exceeds the limit of 2 vol.-% for all investigated operation conditions. Hence, a simple process setup with a single CO_2 separation step upstream of the methanation reactor does not suffice for a single stage methanation when understoichiometric feed gases, like feed gas nos. 2–5, are introduced to the methanation reactor. Two possible arrangement results are as follows: (i) The CO_2 separation unit is placed downstream of the methanation reactor. The resulting disadvantage is the slightly lower methane yield, as shown above, and a higher gas volume flow through the methanation reactor because of the surplus CO_2 . The latter increases the capital expenditures (CAPEX) of the methanation reactor. On the other hand, the strong volume contraction during methanation reduces the gas flow through the CO_2 separation unit which in turn reduces the CAPEX. (ii) A CO_2 separation unit is placed upstream and downstream of the methanation reactor. The methane yield is slightly higher and the gas flow through the methanation is lower. The disadvantages in this case are the increased CAPEX for the second CO_2 separation step and the increased heat flux in the methanation reactor due to the missing ballast gas. Hence, the second option does not seem to be favorable because of the additionally required process unit in the case of a single stage fluidized bed methanation with the investigated understoichiometric feed gases (feed gas nos. 2–5). For the SER feed gases (feed gas no. 1 and no. 6), the CO_2 separation can be neglected completely if the right operating conditions are chosen as is explained below. For a multistage process, like GoBiGas, the upstream CO_2 removal is nevertheless justified. The water-gas shift reaction is carried out in a separate reactor followed by the CO_2 separation unit, both upstream of the methanation reactors. This way, the production of CO_2 and surplus H_2 in the methanation section can be suppressed and no further gas upgrading besides drying is necessary.

3.3 Investigation of the SER product gas

Feed gas no. 1 is a typical SER product gas with a high H_2 content. The SN is greater than 1, which allows a practically complete methanation of the carbonaceous species ($CO + CO_2 + C_2H_4$) at temperatures up to 300 °C with a CH_4 yield of nearly 100% (Fig. 5).

Pressure only has a significant influence on the gas composition at higher temperatures. With pressurization, the decreasing trend of CH_4 and the increasing trends of H_2 , CO , and CO_2 at higher temperatures can be counteracted. In addition, above 440 °C at 1 bar_a carbon formation is thermodynamically possible. As is shown in Fig. 5a, H_2O needs to be added in this small operating window. At higher pressures, the steam addition can be prevented. Below 300 °C, there is practically no influence of pressure or temperature on the gas composition. In this case, methanation around 300 °C and

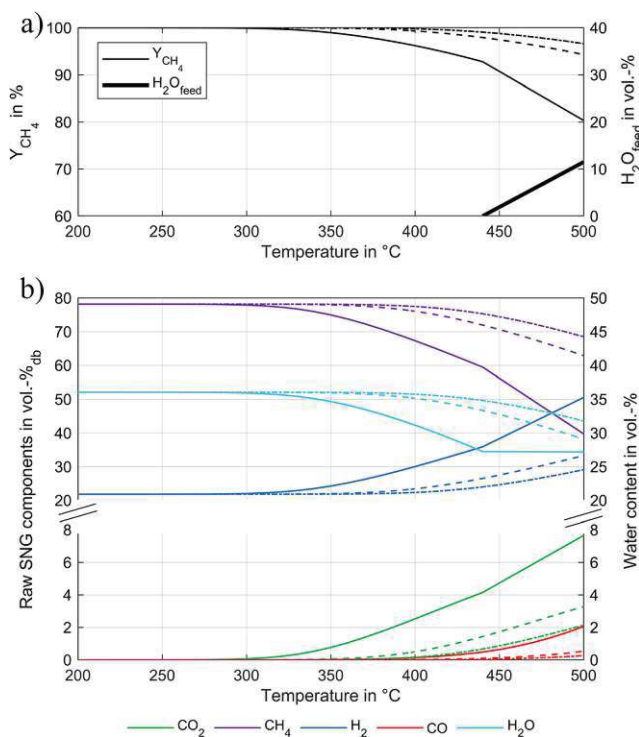


Fig. 5 Temperature and pressure variation for feed gas no. 1 in the thermodynamic equilibrium: 1 bar_a (full line), 5 bar_a (dashed line), and 10 bar_a (dash-dotted line). **a** CH₄ yield and feed water content. **b** Raw-SNG gas composition

1 bar_a shows a favorable raw-SNG composition without the need of compression. Lower temperatures would not improve the gas composition but increase the challenge of employing an active catalyst. For grid feed-in, only H₂ would need to be separated from the raw-SNG. For the application as hythane on the other hand, no further upgrading step is necessary except water condensation.

3.4 Investigation of variable product gas compositions of the SER process

Fuchs et al. [61] already described the adaptability of the SER process with regard to the product gas composition. In Fig. 6, the evolution of the product gas components over the gasification temperature of the 100 kW_{th} DFB gasifier at TU Wien is depicted. The product gas can be adjusted to the required feed gas for methanation by varying the gasification temperature. However, this also adds an additional parameter to the modelling of the methanation reactions. The range for the gas components, the temperatures, the used bed material, and the fuel is already listed in Table 1 (feed gas no. 6).

Figure 7 displays the composition of the raw-SNG in the thermodynamic equilibrium for all data points of Fig. 6 over *SN*. Temperature and pressure are again set to 300 °C and 1 bar_a respectively, for the methanation process. In order to assess the carbon formation, *Y_C* is given. There is a decreasing

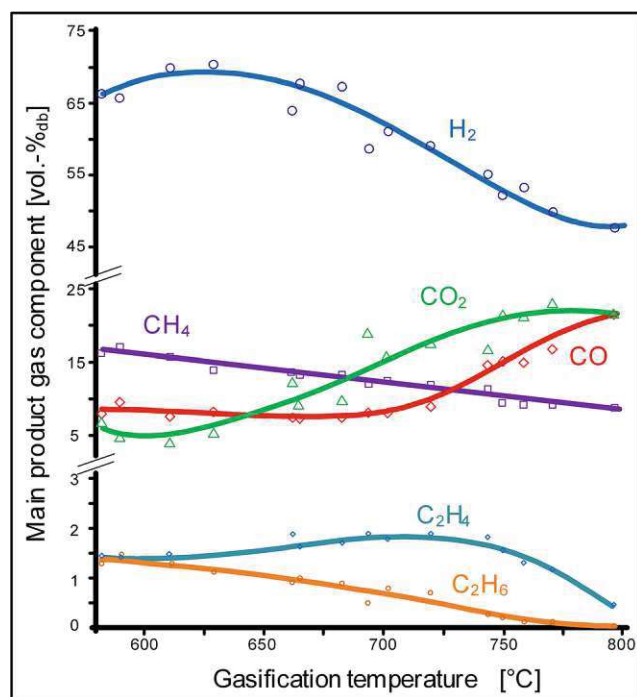
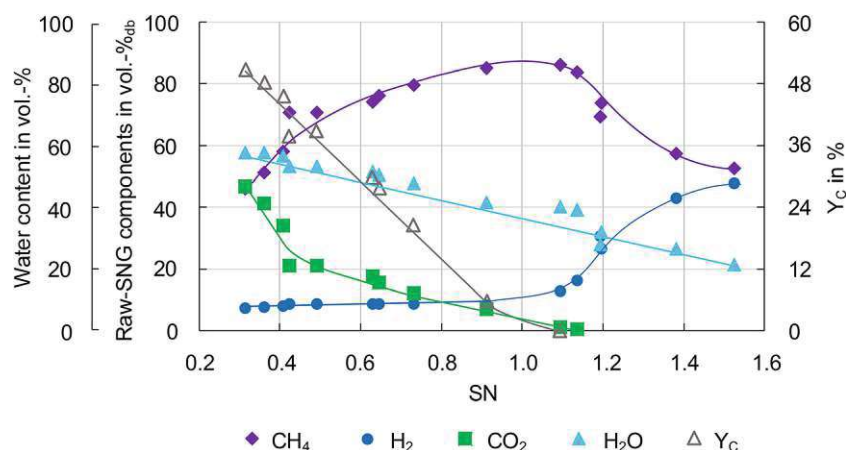


Fig. 6 Product gas composition over gasification temperature for the 100 kW_{th} DFB gasifier at TU Wien for softwood and olivine as fuel and bed material, respectively (from [61])

trend for CO₂, H₂O, and the amount of carbon formed for an increasing *SN*. CH₄ has a maximum at a *SN* slightly above 1. At the same point, carbon formation declines to 0 and the small incline in H₂ turns into a sharp increase for higher *SN*. CO is only present in trace amounts (0.14–614 ppm_{v,db}) and is not displayed here. From a thermodynamic point of view, the feed gas with a *SN* of 1.09 results in a raw-SNG with the most favorable composition for the methanation at 300 °C and 1 bar_a. A *SN* of 1.09 corresponds to a gasification temperature of about 680 °C. The associated compositions for the feed gas and the raw-SNG as well as the key figures are depicted in Table 3. Both CO and CO₂ are almost completely converted and therefore no CO₂ separation step is necessary. Compared with feed gas no. 1 the H₂ content is lower but for grid feed-in the H₂ still needs to be separated. A pressure increase to 4 bar_a lowers the H₂ content below 10 vol.-%, and the raw-SNG could be directly utilized as SNG without further purification if the loosened H₂ restriction in the gas grid is assumed. This would be an economic improvement because no H₂ separation step is necessary. Additionally, the CH₄ yield and the CO₂ conversion increase and the CO content decreases. The according raw-SNG composition and the key figures at 4 bar_a and 300 °C are also displayed in Table 3. Different operation conditions of the methanation might favor other feed gas compositions from Fig. 6 and vice versa. In order to find the most suitable feed gas composition for deviating methanation conditions, reiterations of the thermodynamic equilibrium calculations would have to be carried out.

Fig. 7 Raw-SNG gas composition and Y_C over SN at 300 °C and 1 bar_a for the feed gas compositions according to Fig. 6 in the thermodynamic equilibrium



3.5 Comparison of all investigated feed gases with the carbon utilization efficiency

Table 4 compares the investigated feed gases (feed gas nos. 1–6) by means of the carbon utilization efficiencies (η_C , $\eta_{C,DFB}$) as well as the H_2 and CO_2 contents in the raw-SNG at 300 °C and 1 bar_a. η_C is the highest for the product gas from the gasification of LI (feed gas no. 3) and the lowest for the product gas from the CO_2 gasification of RSC (feed gas no. 5). All other values for η_C are in a similar range between 34.6 and 37.9%. The comparison of η_C and $\eta_{C,DFB}$ reveals that the carbon utilization for the SER product gases (feed gas no. 1 and 6) is governed by the carbon utilization in the DFB system. The excess carbon (in the form of CO_2), which is still in the raw-SNG in case of conventional gasification (like feed gas nos. 2–4), is already removed within the SER process by the increased transport of carbon from the fuel to the flue gas. This results in a low $\eta_{C,DFB}$ but a similar value for η_C compared with feed gas nos. 2 and 4 because nearly a complete carbon utilization is achieved in the methanation section. Additionally, no CO_2 separation step is required as the

possibility to adjust the stoichiometric number SN is inherent to the process. Further savings result from the fact that no steam addition to the feed gas is necessary and the fact that the composition of the feed gas can be adjusted (cf. Fig. 6). Despite the high flexibility, a H_2 separation is nevertheless required under current regulations. If 10 vol.-% of H_2 would be allowed, the SER process seems economically advantageous because neither a CO_2 nor a H_2 separation unit or a steam addition to the feed gas is required under the right process conditions (e.g. feed gas no. 6 at 300 °C and 4 bar_a). The CO_2 separation alone was estimated to account for 13–22% of the total fixed capital investment costs of a biomass-to-SNG plant [19].

The highest η_C is reached with feed gas no. 3, which originates from the gasification of lignin with olivine as bed material. The high η_C results from the high value for $\eta_{C,DFB}$. Almost 93% of the carbon in the fuel is relocated to the gasifier product gas. The lowest η_C results from feed gas no. 5, which originates from the gasification of rapeseed cake with olivine as bed material and a CO_2/H_2O mixture as gasification agent. The gasification with a CO_2 admixture to the

Table 3 Feed gas and raw-SNG composition and key figures for the feed gas with a SN of 1.09 at 300 °C and 1 bar_a as well as 300 °C and 4 bar_a in the thermodynamic equilibrium

| Parameter | Unit | Feed gas | Raw-SNG at 1 bar _a | Raw-SNG at 4 bar _a |
|------------|----------------------|----------|-------------------------------|-------------------------------|
| CH_4 | vol.-% _{db} | 13.3 | 86.1 | 90.0 |
| H_2 | vol.-% _{db} | 67.8 | 12.8 | 9.8 |
| CO | vol.-% _{db} | 7.3 | 0.005 | 0.0008 |
| CO_2 | vol.-% _{db} | 9.8 | 1.1 | 0.2 |
| C_2H_4 | vol.-% _{db} | 1.7 | 0 | 0 |
| H_2O | vol.-% | 0 | 40.2 | 41.6 |
| Y_{CH_4} | % | - | 98.8 | 99.7 |
| Y_C | % | - | 0 | 0 |
| X_{CO} | % | - | 100 | 100 |
| X_{CO_2} | % | - | 95.8 | 99.1 |

Table 4 Comparison of the carbon utilization efficiencies and the H₂ and CO₂ contents in the raw-SNG for feed gas nos. 1–6 at 300 °C and 1 bar_a

| Parameter | Unit | Feed gas number | | | | | |
|-------------------------|----------------------|-----------------|------|------|------|------|------|
| | | 1 | 2 | 3 | 4 | 5 | 6 |
| $\eta_{C, DFB}$ | % | 36.5 | 72.8 | 92.6 | 86.9 | 78.0 | 38.4 |
| η_C | % | 36.5 | 37.0 | 47.0 | 34.6 | 23.1 | 37.9 |
| H ₂ content | vol.-% _{db} | 22.0 | 7.3 | 7.3 | 6.5 | 5.5 | 12.8 |
| CO ₂ content | vol.-% _{db} | 0.1 | 45.5 | 45.6 | 56.2 | 66.4 | 1.1 |

gasification agent therefore cannot be used advantageously for the production of SNG if no external hydrogen is provided. For feed gas nos. 2–5, a CO₂ separation and a H₂ separation is required. If the 10 vol.-% H₂ threshold is applied, the H₂ separation can be avoided (e.g. feed gas no. 4 at 320 °C and 5 bar_a). Even the 4 vol.-% H₂ threshold can be met if the operation conditions are adapted (e.g. feed gas no. 4 at 280 °C and 10 bar_a), but kinetic effects at these low temperatures most likely need to be considered. For these feed gases (feed gas nos. 2–5), the carbon utilization efficiency can be increased by the addition of H₂ from external sources (e.g. electrolysis) which allows the methanation of the leftover CO₂. From a technical and ecological point of view, the addition is advantageous since η_C can be maximized. The availability and the expenditures for the additional hydrogen on the other hand need to be eyed critically. In this paper, this concept is not discussed any further but some relevant studies were already referred above [50, 52].

In general, the calculated results are in good agreement with literature values. The GoBiGas plant reached a η_C of about 30%, which is slightly lower as most of the calculated values. The slightly lower values seem justified, since this study is based on thermodynamic calculations and therefore the results need to be seen as maximum values. The gasification section of the GoBiGas plant reached a $\eta_{C,DFB}$ of about 70% as can be calculated from the results in [63]. This value is similar to feed gas no. 2 but lower compared with all other feed gases. The discrepancy possibly arises from the small scale and good performance of the pilot plant as well as the difficult scalability of the carbon utilization efficiency as explained in the methodology section. Taking the results from the modelling study of Heyne and Harvey [19], a η_C of 35% can be calculated, which is very close to the calculated values in this paper. Also, the raw-SNG composition with 45 vol.-%_{db} of CH₄, 47 vol.-%_{db} of CO₂, and 4 vol.-%_{db} of H₂ is close to the calculated values. Similar values were also reported by Gassner et al. [64] who calculated a raw-SNG composition with 45 vol.-%_{db} CH₄, 45 vol.-%_{db} CO₂, and 6 vol.-%_{db} H₂. Both studies assumed similar operating conditions at approximately 300 °C and 1 bar_a. Experimentally, Seemann et al.

[58] confirmed a similar raw-SNG composition. They reconstructed the feed gas composition of the Güssing gasifier and reached slightly lower CH₄ concentrations at approximately 40 vol.-%_{db} CH₄, 47 vol.-%_{db} CO₂, and 4 vol.-%_{db} H₂. The 1 MW_{SNG} methanation plant in Güssing, however, could not meet the 4 vol.-% threshold, and a two-stage membrane separation process was necessary, whereas in Gothenburg, no H₂ separation unit was required [10, 31].

4 Conclusion and outlook

In this work, the suitability of various product gases from the 100 kW_{th} DFB gasifier for methanation in a fluidized bed reactor was evaluated from a thermodynamic point of view. It was shown that a complete methanation of CO and CO₂ is only possible for SER product gases. For all other presented product gases, only the methanation of CO is possible, whereas CO₂ might even constitute the main raw-SNG component. Additionally, gases from conventional steam gasification or gasification with CO₂ admixture to the gasification agent (H₂O + CO₂) are subject to carbon depositions in the methanation reactor. Therefore, up to 55 vol.-% of H₂O needs to be added to the feed gas for a stable operation. Furthermore, the influence of different operation conditions of the methanation on the raw-SNG composition was visualized. By the careful choice of operation conditions, energy savings and/or less effort for further gas upgrading can be accomplished. A comparison between upstream and downstream CO₂ separation revealed that only a downstream CO₂ separation results in the required SNG quality if a single fluidized bed methanation reactor with understoichiometric feed gases is utilized. A further investigation of the SER product gases revealed that it is also possible to adapt the gasification process to suit certain methanation conditions. A SER product gas with a stoichiometric number of 1.09, which corresponds to a gasification temperature of 680 °C, was shown to be the most suitable feed gas for methanation. No CO₂ separation step and no H₂O addition to the feed gas was necessary, which clearly indicated an economic advantage. However, under current regulations, a H₂ separation unit could not be avoided for the raw-SNG from the SER product gas. An increase of the allowed H₂ content in the natural gas grid to 10 vol.-% would therefore increase the degrees of freedom of the whole system. In turn, this would result in improved operating points, which would simplify the overall process and reduce costs. This would apply for all investigated feed gases, but especially the SER process would benefit from these loosened restrictions. For example, the SER product gas (feed gas no. 6) could be methanated at 300 °C and 4 bar_a to gas grid quality without a CO₂ or H₂ separation step nor a H₂O addition to the feed gas.

A comparison of the carbon utilization efficiencies revealed that the gasification of lignin resulted in the highest

overall value of 47%. Apart from one exception, all other values including the SER product gases range between 34.6 and 37.9%. Only if CO₂ is added to the gasification agent, the carbon utilization factor drops to 23%. The addition of H₂ from an external source would allow a much more efficient conversion of the carbon, but the availability and the economic implications would need to be considered.

It should be noted that all investigations in this paper are based on thermodynamic equilibrium calculations. Catalyst poisoning due to insufficient gas cleaning, kinetic limitations concerning carbon deposition, methanation of CO₂, the high feed water content, or low temperatures as well as possible heat or mass transfer limitations necessitate experimental investigations. These issues are subject of further investigations with the bench-scale fluidized bed methanation setup at TU Wien.

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Abbreviations BA, bark; bar_w , bar absolute; C, carbon; CNG, compressed natural gas; db, dry basis; DFB, dual fluidized bed; eq, equilibrium; feed, in the feed gas; gasif, in the gasification agent; ΔH_R^{300} , molar reaction enthalpy at 300 °C; H_2O_{feed} , volume fraction of H₂O in the feed in vol.-%; L, limestone; LI, lignin; O, olivine; N_i , number of carbon atoms in species i ; n_i , molar flow of species i in mol/s; η_C , overall carbon utilization efficiency; $\eta_{C, DFB}$, carbon utilization efficiency over the DFB gasifier; OL, olivine/limestone mixture; PSA, pressure swing adsorption; PSI, Paul Scherrer Institute; PtG, power-to-gas; raw-SNG, raw synthetic natural gas after methanation/before gas upgrading; RSC, rape-seed cake; SER, sorption enhanced reforming; SN, stoichiometric number; SNG, synthetic natural gas; SS, sewage sludge; SW, softwood; v, volumetric; vol.-%, volumetric percent; wt.-%, weight percent; X_{CO} , carbon monoxide conversion in %; X_{CO_2} , carbon dioxide conversion in %; Y_C , carbon yield in %; Y_{CH_4} , methane yield in %; y_i , molar fraction of species i

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PAPER VI

Bartik A., Benedikt F., Fuchs J., Hofbauer H., Müller S.

SNG PRODUCTION VIA GASIFICATION AND CATALYTIC METHANATION: LATEST DEVELOPMENTS AT TU WIEN

in: Proceedings of the 7th International Conference on Renewable Energy Gas Technology, Weimar, Germany, September 20, 2021

Responsibility of the author:

- Mentored the evaluation of the experimental campaign and the related test procedures
- Supported the creation of the simulation model used to validate the experimental data
- Supported the interpretation of the results achieved by key performance indicators

PAPER VII

Müller S., Groß P., Rauch R., Zweiler R., Aichernig C., Fuchs M., Hofbauer H.

PRODUCTION OF DIESEL FROM BIOMASS AND WIND POWER – ENERGY STORAGE BY THE USE OF THE FISCHER-TROPSCH PROCESS

in: Biomass Conversion and Biorefinery, Vol. 7, pp. 275–282, 2017

doi: 10.1007/s13399-017-0287-1

Responsibility of the author:

- Evaluated the experimental campaign and the related test procedure
- Created the simulation model used to validate the experimental data
- Interpreted and visualized the results
- Corresponding author independent of mentoring support

PAPER VIII

Hammerschmid M., Müller S., Fuchs J., Hofbauer H.

EVALUATION OF BIOMASS-BASED PRODUCTION OF BELOW ZERO EMISSION REDUCING GAS FOR THE IRON AND STEEL INDUSTRY

in: Biomass Conversion and Biorefinery, Vol. 11, pp. 169–187, 2021

doi: 10.1007/s13399-020-00939-z

Responsibility of the author:

- Designed the investigation concept and methodology
- Created the simulation model used for the calculations
- Interpreted the results
- Scientific mentoring



Evaluation of biomass-based production of below zero emission reducing gas for the iron and steel industry

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Abstract

The present paper focuses on the production of a below zero emission reducing gas for use in raw iron production. The biomass-based concept of sorption-enhanced reforming combined with oxyfuel combustion constitutes an additional opportunity for selective separation of CO₂. First experimental results from the test plant at TU Wien (100 kW) have been implemented. Based on these results, it could be demonstrated that the biomass-based product gas fulfills all requirements for the use in direct reduction plants and a concept for the commercial-scale use was developed. Additionally, the profitability of the below zero emission reducing gas concept within a techno-economic assessment is investigated. The results of the techno-economic assessment show that the production of biomass-based reducing gas can compete with the conventional natural gas route, if the required oxygen is delivered by an existing air separation unit and the utilization of the separated CO₂ is possible. The production costs of the biomass-based reducing gas are in the range of natural gas-based reducing gas and twice as high as the production of fossil coke in a coke oven plant. The CO₂ footprint of a direct reduction plant fed with biomass-based reducing gas is more than 80% lower compared with the conventional blast furnace route and could be even more if carbon capture and utilization is applied. Therefore, the biomass-based production of reducing gas could definitely make a reasonable contribution to a reduction of fossil CO₂ emissions within the iron and steel sector in Austria.

Keywords Iron and steel · Low-carbon steelmaking · Direct reduction · Biomass · Sorption-enhanced reforming · Oxyfuel combustion

1 Introduction

Today the iron and steel industry in EU-28 is responsible for 200 million tons of carbon dioxide [1] which amounts to a share of 5% of the total carbon dioxide equivalent (CO₂e) [2] emissions [3]. These numbers show that especially the transformation of heavy load industries like the iron and steel industry towards low-carbon technologies will be challenging. In Austria the iron and steel industry also contributes to a significant share concerning greenhouse gas emissions. In 2017, 8.1 million tons of crude steel were produced in Austria [4], which are responsible for around 16% of the total greenhouse gas emissions [5]. Technological development

has enabled to improve the energy efficiency and to reduce CO₂ emissions in this sector. However, the principles of steelmaking have not changed fundamentally over the years. In 2017, over 91% of the Austrian crude steel was produced within oxygen-blown converters, which were fed with hot metal from blast furnaces. The remaining share was produced within electric arc furnaces [4]. According to the EU Roadmap 2050 [6], the CO₂ emissions within the iron and steel industry must be reduced by around 85%. To accomplish this major goal, a complete conversion towards low-carbon steelmaking technologies has to be done.

Numerous researchers and international institutions investigate alternative low-carbon steelmaking routes. Especially, the ULCOS program [7, 8] has evaluated the CO₂ reduction potential of over 80 existing and potential technologies. Several investigations are working on further optimization of fossil fuel-based state-of-the-art processes like the coke and pulverized coal-based-integrated blast furnace route [9–11]. All this optimization steps to reduce the consumption of fossil fuels are limited [12]. For reaching the previous described

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climate goals within the iron and steel sector, a fundamental change of steelmaking is necessary. The ULCOS program [7, 8] identified four technologies with CO₂ emission reduction potentials of more than 50%. The technologies within this program, which are based on carbon capture and storage (CCS) or utilization (CCU), are the top-gas recycling within the blast furnace (BF-TGR-CCS/U), a novel bath-smelting technology (HISARNA-CCS/U) [13, 14], and a novel direct reduction process (ULCORED-CCS/U). Only the novel ULCOLYSIS [15] process, which is characterized by melting iron ore through electric direct reduction, is not based on CCS or CCU. In addition to the research activities in Europe, the COURSE50 program in Japan, POSCO in Korea, AISI in the USA, and the Australian program are some international examples for investigations regarding CO₂ reduction in the iron and steel industry [16]. The COURSE50 program [8, 16, 17] is focused on H₂-based reducing agents in blast furnace (BF) for decreasing the fossil coke consumption and technologies for capturing, separating, and recovering CO₂ from the BF gas. POSCO [8, 16, 18] in Korea is working on the adaptation of CCS and CCU to smelting reduction processes, like the FINEX and COREX process. Furthermore, POSCO is researching in bio-slag utilization, pre-reduction and heat recovery of hot sinter, CO₂ absorption using ammonia scrubber, hydrogen production out of coke-oven gas (COG), and iron ore reduction using hydrogen-enriched syngas. AISI [8, 16] is working on the molten oxide electrolysis, which is similar to the ULCOLYSIS concept and iron making by hydrogen flash smelting. The research programs regarding breakthrough iron and steelmaking technologies in Brazil, Canada, and Australia [19] are all strongly focused on biomass-based iron and steel production routes for replacing fossil coal and coke by use of biomass-derived chars as substitutes [8, 16, 20].

Summing up, there are a lot of investigations going on around the world to reduce the CO₂ footprint of the iron and steel industry.

The most of the previous described concepts apply CCS or CCU to reach a CO₂ reduction potential over 50% in comparison to the conventional integrated BF route. Nevertheless, the implementation of CCS requires a fundamental investigation due to storage sites and long-term response of the environment. Beside the CCS or CCU-based approaches, the replacement of fossil fuel-based reducing agents by biomass-based substitutes or the use of hydrogen as reducing agent are promising approaches for reaching the climate targets within the iron and steel sector. Furthermore, some electric direct reduction processes like ULCOWIN, MOE, and ULCOLYSIS are under investigation. One possible CO₂ reduction path could also be the rise of the share of steel production through electric arc furnaces. Therefore, enough high-quality scrap must be available.

With respect to the estimates regarding biomass potential in the next decades [20, 21], in Austria beside the rise of the

share of steel production through scrap-based electric arc furnaces, another possible synergetic transition option seems to be the replacement of the integrated blast furnace route with the direct reduction of iron ore based on biomass-based reducing gas. The Austrian steel manufacturing and processing group, voestalpine AG, is already operating one of the biggest direct reduction plants, based on the MIDREX concept and reformed natural gas as reducing agent in Texas [22]. This approach would combine the gained expertise within the field of direct reduction with the Austria-developed concept of dual fluidized bed steam gasification [23]. Within the present work, a biomass-based production of biogenic reducing gas through dual fluidized bed steam gasification, which allows the replacement of steam reformed natural gas, is investigated. At this stage, it remains unclear if the investigated process is competitive with respect to other production routes for the supply of reducing gas for iron ore reduction.

So far, following question has not been answered sufficiently:

How can the production of biomass-based reducing gas via dual fluidized bed steam gasification enable a reasonable contribution to a reduction of fossil CO₂ emissions within the iron and steel sector?

The following paper describes the results of the investigated process enabling the production of a below zero emission reducing gas by applying the biomass-based dual fluidized bed steam gasification technology in combination with carbon capture and utilization. The investigations are based on experimental results combined with simulation work. The present paper discusses:

- The *comparison of different iron- and steelmaking routes regarding their CO₂ footprint*
- The *proposed process concept* for the production of biomass-based reducing gas
- *Experimental and simulation results* achieved
- The results of a *techno-economic assessment*

2 Concept and methodology

With regard to the techno-economic assessment of the selective separation of CO₂ technology OxySER, a plant concept for the integration in a direct reduction process has been developed. Beforehand, a short overview and comparison of primary and secondary iron and steelmaking routes regarding their CO₂ footprints will be given. Furthermore, the application of dual fluidized bed steam gasification with respect to the combination of sorption-enhanced reforming and oxyfuel combustion will be explained.

2.1 Comparison of iron and steelmaking routes regarding their CO₂ footprint

Two main steelmaking processes can be distinguished. The primary steelmaking route converts virgin iron ores into crude steel (CS). Secondary steelmaking is characterized by the recycling of iron and steel scrap in an electric arc furnace [8, 24]. Table 1 gives an overview of chosen iron and steelmaking routes and the comparison regarding CO₂ footprint. First of all, the primary steelmaking integrated blast furnace (BF) route, which is predominant in Austria. Thereby, steel production takes place at an integrated steel plant, where iron ores are reduced into hot metal through the use of reduction agents such as coke or coal. Afterwards, the hot metal is converted into steel by oxygen injection in a basic oxygen furnace (BOF). As result of the high energy demand of 11.4 GJ/t_{CS} on fossil reducing agents, the CO₂ footprint of the BF-BOF route is with 1.694 t CO₂e/t_{CS} very high [25]. Furthermore, the secondary steelmaking electric arc furnace (EAF) route is used in Austria. Therein, the major feedstock is ferrous scrap, which is melted mainly through the use of electricity. However, increasing the share of EAF steel is constrained by the availability of scrap, and the quality requirements for steel grades have to meet [8]. The smelting reduction route belongs also to the state-of-the-art iron and steelmaking routes. Within this route, iron ores are heated and pre-reduced by the off-gas coming from the smelter-gasifier. The pre-reduction step could be realized in a shaft kiln (COREX) or a fluidized bed reactor (FINEX). Pre-reduced iron ores are then melted in the smelter-gasifier. The smelter-gasifier uses oxygen and coal as a reducing agent. Afterwards, the hot metal is also fed to the BOF for steelmaking. Another possibility of steelmaking is the primary direct reduction (DR) route. MIDREX is one of the used direct reduction technologies. It is characterized by the reduction of iron ores into solid direct reduced iron (DRI) within a shaft kiln. The direct reduction technologies could also work within a fluidized bed reactor. Examples include the FINMET and CIRORED process [38]. The direct reduction is driven by the fed of a reducing gas. Currently, the commercial used reducing gas is based on the reforming of natural gas. For extended information regarding the fundamentals of iron and steelmaking routes, a reference is made to [8, 24, 39].

Beside the previous described state-of-the-art iron and steelmaking routes, some innovative developments and investigations are compared with the conventional routes regarding their energy demand, CO₂ footprint, merit, and demerit in Table 1. Therein, the integrated blast furnace route (BF and BOF) which is predominant in Austria is set as reference regarding CO₂ emissions. Recycling of the blast furnace top-gas in combination with CCS or CCU (BF-TGR-CCS/U and BOF) or the replacement of fossil coal by biogenic substitutes reduces the fossil reducing agent demand and decrease the

CO₂ footprint of integrated blast furnace routes up to 50% [7, 16, 26, 30, 31].

The replacement of the BF by smelting reduction processes like the COREX or FINEX process would raise slightly the CO₂ footprint due to the high consumption of fossil coal. An ecologically favorable operation of smelting reduction processes only could be realized by the use of CCS or CCU [8, 16, 18]. The use of a smelting reduction technology based on bath-smelting (HISARNA-CCS/U and EAF) in combination with CCS would reduce the CO₂ emissions up to 80% [7, 16].

Direct reduction plants enable a big CO₂ emission saving potential in comparison with the integrated BF route due to the present used reformed natural gas as reducing agent. Reformed natural gas consists to a large extent of hydrogen, which results in lower CO₂ emissions due to the oxidation of hydrogen to steam within the reduction process [12]. The replacement of the integrated BF route by the state-of-the-art MIDREX plant, which is based on the reduction of iron ore within a shaft kiln by the use of reformed natural gas, would decrease the CO₂ emissions by 50% in comparison with the reference route [12, 32, 33]. The economic viability of direct reduction-based routes, which are based on reformed natural gas, strongly depend on the natural gas price which is in Europe much higher than in North America [33]. Within the ULCOS project, a novel direct reduction process (ULCORED-CCS/U) based on partial oxidized natural gas is investigated [7, 8]. By the reduction of the required amount of natural gas and the application of CCS or CCU, the CO₂ emissions could be decreased up to 65% compared with the reference route. The dual fluidized bed steam gasification process, based on the bed material limestone, which is called sorption-enhanced reforming (SER), produces a biomass-based hydrogen-rich gas, which allows the replacement of the steam reforming unit for reforming of natural gas. The application of SER to produce a biomass-based reducing gas for the MIDREX process (MIDREX-BG-SER) reduces the CO₂ footprint compared with the integrated BF route up to 80%. The combination of SER with oxyfuel combustion (OxySER) enables an in situ CO₂ sorption within the reducing gas production process. Beside the production of biomass-based reducing gas, a CCU or CCS ready CO₂ stream is released. Therefore, a below zero emission reducing gas due to the application of CCU or CCS is generated. Another direct reduction breakthrough technology could be the HYBRIT process, which is based on the reducing agent hydrogen, produced by electrolysis [16, 26, 34, 35]. Therefore, the emissions within the HYBRIT process are mostly caused by the CO₂ footprint of the electricity mix. With regard to the Austrian electricity mix, with a CO₂ footprint of 0.218 kg CO₂e/kWh_{el} [36], a CO₂ emission saving potential up to 50% could be reached with the HYBRIT process.

Table 1 Overview of different iron and steelmaking routes including their energy demands and CO₂ emissions [16]

| Iron and steelmaking route ¹ | Description of the technology | Reducing agent | Total energy demand [GJ/t _{cs}] | Energy demand reducing agent [GJ/t _{cs}] | N et power demand [GJ/t _{cs}] | CO ₂ emissions ² [t CO ₂ e/t _{cs}] | Savings potential ² [%CO ₂ e] | Advantages | Disadvantages | Literature |
|-----------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------|-------------------------------------------|----------------------------------------------------|-----------------------------------------|-------------------------------------------------------------------------------|-----------------------------------------------------|-------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------|
| Integrated Blast Furnace route | | | | | | | | | | |
| BF and BOF | Fossil reducing agents (coke and pulverized coal) are used as reducing agents; mixture of sinter, pellets, and additives are fed to the BF | Coke/coal | 19.2 | 15.3 | 0.6 | 1.694 | – | Production of HM in existing BF for steelmaking is the most cost-efficient technology to-day | State of the art BFs are operated near their theoretical minimum energy limit, further CO ₂ reductions are difficult | [25–27] |
| BF-TGR-CCS/U and BOF | Upgraded and recirculated BF gas is used as reducing agent, parts of pulverized coal and top-charged coke are replaced, CCS can be used for further CO ₂ reduction | Coke/coal/recirculated BF gas | 20.0 | 11.6 | 1.4 | 0.813 | – 52% | Top-gas recycling captures CO ₂ and enables CCS, Reduction of fossil reducing agent demand | Higher total energy demand because of additional energy demand for carbon capture, higher net energy demand due to the lack of power recovery from BF gas | [7, 15, 26, 28, 29] |
| BF-Bio-Char and BOF | Charcoal replaces fossil coal by 100% in the BF | Coke/charcoal | 19.2 | 15.3 | 0.6 | 1.220 | – 28% | Replacement of fossil coal by charcoal could be quite straightforward | Charcoal is more expensive than fossil coal, handling, transportation and storage is more difficult compared with fossil coal | [16, 26, 30, 31] |
| Smelting reduction route | | | | | | | | | | |
| COREX and BOF | Combination of pre-reduction in a shaft kiln and smelter-gasifier | Coal | 17.7 | 15.9 | 0.6 | 1.975 | + 17% | No need for coke oven plant | Restrictions for non-coking coal quality, customer for export gas necessary for economic viability | [12, 16, 26] |
| FINEX and BOF | Combination of pre-reduction in a fluidized bed reactor and smelter-gasifier | coal | – | – | – | 1.910 | + 13% | No need for coke oven plant, pelletizing, sintering or agglomeration of iron-bearing materials | Technology not wide-spread | [12, 16] |
| HISARNA-CCS/U and EAF | Bath-smelting technology which combines coal | Coal | 18.0 | 15.0 | 2.5 | 0.330 | – 81% | No need for coke oven and | Technology at demonstration stage, | [7, 16] |

Table 1 (continued)

| Iron and steelmaking route ¹ | Description of the technology | Reducing agent | Total energy demand [GJ/t _{cs}] | Energy demand reducing agent [GJ/t _{cs}] | N et power demand [GJ/t _{cs}] | CO ₂ emissions ² [t CO ₂ e/ t _{cs}] | Savings potential ² [%CO ₂ e] | Advantages | Disadvantages | Literature |
|------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------|-------------------------------------------|----------------------------------------------------|-----------------------------------------|--------------------------------------------------------------------------------|-----------------------------------------------------|-----------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------|
| Direct reduction route MIDREX and EAF | pre-heating and partial pyrolysis in a reactor, a smelter vessel is used for final ore reduction and a melting cyclone for ore smelting | Reformed natural gas | 16.6 | 10.0 | 2.8 | 0.835 | -51% | sinter/pellet plant. Use of non-coking coal qualities. Economic viable even at small size | more net power demand because of EAF and CCS | [12, 32, 33] |
| ULCORED-CCS/U and EAF | DRI production based on shaft furnace with reformed natural gas as reducing agent. Based on lump/pellet ore. | Partial oxidized natural gas | - | - | - | 0.600 | -65% | No need for coke oven and sinter plant Reduction of natural gas consumption helps to reduce OPEX | OPEX strongly depend on natural gas price, which is very different around the world Requires pure oxygen instead of air. Technology at demonstration stage | [7, 8] |
| MIDREX-BG-SER and EAF | Reformed natural gas is replaced by biomass-based reducing gas produced by dual fluidized bed steam gasification | Biomass-based reducing gas | 16.6 | 10.0 | 2.8 | 0.280* | -83% | Replacement of reformed natural gas by biomass-based reducing gas could be quite straightforward | OPEX strongly depend on biomass price | Captured within this paper |
| MIDREX-BG-OxySER-CCS/U and EAF | Reformed natural gas is replaced by biomass-based reducing gas produced by dual fluidized bed steam gasification and in situ CO ₂ capture and utilization | Biomass-based reducing gas | - | 10.0 | - | Below zero** | More than 100% reduction | Replacement of reformed natural gas by biomass-based reducing gas could be quite straightforward | OPEX strongly depend on biomass price. CCS or CCU approach requires pure oxygen instead of air as fluidization agent in the fluidized bed system | Captured within this paper |
| HYBRIT and EAF | Hydrogen produced with water electrolysis is used as reducing agent | Hydrogen | 14.7 | 6.8 | 12.6 | 0.800 | -53% | CO ₂ emissions very low if renewable energy sources | Hydrogen production is quite expensive with current technologies and | [16, 26, 34, 35] |

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Table 1 (continued)

| Iron and steelmaking route ¹ | Description of the technology | Reducing agent | Total energy demand [GJ/t _{cs}] | Energy demand reducing agent [GJ/t _{cs}] | Net power demand [GJ/t _{cs}] | CO ₂ emissions ² [t CO ₂ e/t _{cs}] | Savings potential ² [%CO ₂ e] | Advantages | Disadvantages | Literature |
|-----------------------------------------|----------------------------------------------------------------|----------------|-------------------------------------------|----------------------------------------------------|----------------------------------------|-------------------------------------------------------------------------------|-----------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------|------------|
| | within a direct reduction plant | | | | | | | are used. Almost zero direct emissions from production | requires a lot of electrical energy. Technology at demonstration stage | |
| Secondary steelmaking route EAF | Instead of DRI, scrap is used as iron source | Electricity | 3.3 | - | 2.1 | 0.190 | -89% | DRI production is replaced by recycling of ferrous scrap. Almost zero direct emissions from production | Increasing the share of EAF steel is constrained by the availability of high quality scrap | [12, 26] |
| Other steelmaking routes ULCOLYSIS | Melting iron ore at 1600 °C by using electric direct reduction | Electricity | 15.0 | - | 13.0 | 0.800 | -53% | CO ₂ emissions very low if renewable energy sources are used. Almost zero direct emissions from production | Technology at lab scale. High net power demand | [7, 16] |

¹ Energy values and CO₂ emissions includes material preparation, ironmaking, steelmaking, and casting/based on 80% hot metal or DRI and 20% scrap except from secondary steelmaking route: 100% scrap in EAF

² CO₂ emission factor for grid/calculation model: 0.218 kg CO₂e/kWh_{el} (Austrian electricity mix)/saving potential in comparison with BF and BOF route as reference [36]

* Assumptions: energy values are the same as within the MIDREX route, and the CO₂ emissions are calculated by the difference between the emissions from the MIDREX route based on natural gas and the caused emissions only through natural gas as reducing gas [37]

** Assumptions: energy values are the same as within the MIDREX route with CCS [7] and the CO₂ emissions are below zero because of the combination from the use of a biomass-based feedstock with CCS or CCU

Further possibilities are the rise of the share of steel production through scrap-based electric arc furnaces. This steel-making route enables CO₂ reduction potentials up to 90%, because of the replacement from ironmaking processes with scrap. The EAF-based routes are strongly depended on the availability of high-quality scrap [12, 26]. Furthermore, some novel electric direct reduction processes, like the ULCOLYSIS project, are under investigation [7, 16]. Similar to the HYBRIT process, the electric direct reduction processes are strongly depended on the CO₂ footprint of the national electricity mix, because of the high-net power demands.

Several technologies provide the possibility of additional carbon-emission reduction by sequestration of CO₂. The use of post-combustion capture technologies, like pressure swing adsorption or amine scrubber, is the possibility for the sequestration of CO₂ within iron and steelmaking routes [40]. Within the OxySER process, through the in situ CO₂ sorption, a CCU or CCS ready CO₂ stream is produced. Further explanations regarding CO₂ sequestration can be found in [41–43]. The selective separated and purified CO₂ could be used in further process steps as raw material, *carbon capture and utilization*, or stored in underground deposits, *carbon capture and storage* [43, 44].

Today around 230 million tons of carbon dioxide per year are globally utilized materially. One hundred thirty million tons are used in urea manufacturing and 80 million tons for enhanced oil recovery [45]. With the assumption that hydrogen for the ammoniac production is produced by water electrolysis, which is beside CO₂ the primary energy source for urea production, external CO₂ is necessary for the urea synthesis. In Linz, near to one of the main sites for iron and steel production, a urea synthesis plant with a production rate of around 400,000 t per year of urea is located [46]. Therein, around 300,000 t CO₂ per year are required for the production of the given amount of urea [46]. Further utilization possibilities could be CO₂-derived fuels, like methanol or FT-synthesis and power to gas. Furthermore, the utilization within CO₂-derived chemicals beside urea, like formic acid synthesis, or CO₂-derived building materials, like the production of concrete, could be promising alternatives [45].

Beside the CCU technologies, CO₂ can also be stored in underground deposits. CCS is banned in Austria except research projects up to a storage volume of 100,000 t of CO₂ [44]. For further information regarding CCU and CCS, a reference is made to [40, 45, 47–49].

Since biomass releases the same amount of CO₂ as it aggregates during its growth, the utilization of biogenic fuels can contribute significantly to a reduction of CO₂ emissions. Therefore, the main focus of the paper lies on the production of a below zero emission reducing gas by the use of oxyfuel combustion in combination with sorption-enhanced reforming. This technology for the selective separation of

CO₂ uses as fluidization agent a mix of pure oxygen and recirculated flue gas. Therefore, the nitrogen from the air is excluded from the combustion system [42].

2.2 Combination of oxyfuel combustion and sorption-enhanced reforming

A promising option for the selective separation of CO₂ from biomass and the generation of a hydrogen-rich product gas at the same time is the sorption-enhanced reforming process in combination with oxyfuel combustion (OxySER). The sorption-enhanced reforming (SER) is based on the dual fluidized bed steam gasification process. The main carbon-related (gas-solid) and gas-gas reactions are shown in Table 2. Test runs at the 100 kW pilot plant at TU Wien showed calculated overall cold gas efficiencies of around 70% [51, 52]. Detailed information regarding the dual fluidized bed steam gasification process can be found in literature [37, 51–54].

The combination of oxyfuel combustion and sorption-enhanced reforming combines the advantages of both technologies. Figure 1 represents the concept of the combined technology [44]. First of all, biomass, residues, or waste materials are introduced in the gasification reactor. Limestone is used as bed material which serves as transport medium for heat but also as carrier for CO₂ from the gasification reactor (GR) to the combustion reactor (CR) by adjusting the temperature levels in the reactors correctly. Within the OxySER process, steam serves as fluidization and gasification agent in the GR. Therein, several endothermic gasification reactions take place in a temperature range between 600 and 700 °C [37]. Residual char is transferred with the bed material from the GR to the CR. Due to the combination of SER with oxyfuel combustion, pure oxygen instead of air is used as fluidization agent in the CR, which is operated within a temperature range between 900 and 950 °C. By combustion of residual char in the CR, heat is released. This suitable temperature profiles in the GR and CR ensure that the bed material (limestone) is first calcined to calcium oxide (CaO) at high temperatures in the CR (13). Then the CaO is carbonized in the GR with the carbon dioxide from the product gas (12). Thus, in this cyclic process, a transport of CO₂ from the product gas to the flue gas appears [52]. The use of steam in the gasification reactor and the water gas shift reaction (8) in combination with in situ CO₂ sorption via the bed material system CaO/CaCO₃ enables the production of a nitrogen-free and hydrogen-enriched product gas [37, 56]. Due to the combination of SER with oxyfuel combustion, in addition to the nitrogen-free and hydrogen-enriched product gas, a CO₂-enriched flue gas is generated caused by the use of pure oxygen as fluidization agent in the CR instead of air [57].

The CO₂ equilibrium partial pressure in the CaO/CaCO₃ system and the associated operation conditions for the

gasification and combustion can be found in [52]. By the use of renewable fuels and a continuous selective separation and storage or utilization of CO₂, an improved CO₂ balance can be achieved [44, 57].

Table 3 represents a comparison between the product and flue gas compositions of conventional gasification, SER, and OxySER. The results are based on test runs with the 100 kW pilot plant at TU Wien and the 200 kW pilot plant at University of Stuttgart [37, 57]. As mentioned above, the carbon dioxide content of the product gas could be reduced through the SER method. Furthermore, the hydrogen content is higher in comparison with conventional gasification. The possibility of adjusting the H₂/CO ratio over a wide range makes the SER process very flexible according to product gas applications [52]. The catalytic activity of limestone enables a reduction of tar at the same time [37, 44, 58]. The comparison between the SER and OxySER process illustrates that a CO₂-enriched flue gas in the OxySER test rig in Stuttgart was obtained [57]. In Table 4 the proximate and ultimate analyses of used wood pellets for gasification test runs with the 100 kW pilot plant at TU Wien are listed.

However, OxySER implies the following advantages in comparison to the conventional gasification:

- Selective CO₂ transport to flue gas
- Decrease of tar content in product gas
- High CO₂ content in flue gas > 90 vol.-%_{dry} [57]
- Smaller flue gas stream because of flue gas recirculation
- Nitrogen free flue gas

These assumptions according to experimental results serve as a basis for the conception of an industrial application.

2.3 Integrated OxySER concept for the production of below zero emission reducing gas

The OxySER plant concept for integration in a direct reduction plant is illustrated in Fig. 2. The plant concept is designed for a product gas power of 100 MW. For the production of 100 MW product gas, 50,400 kg/h of wood chips with a water content of 40 wt.-% are required [37]. The wood chips are treated in a biomass dryer. Afterwards the biomass is fed in the gasification reactor. The bed material inventory (limestone) of the system contains 25,000 kg. In the gasification reactor, a H₂-enriched product gas with a temperature of 680 °C is produced. Subsequently, the dust particles are removed from the product gas by a cyclone. Besides ash, these dust particles contain still carbon. This is the reason why the particles are recirculated to the combustion reactor. Afterwards, the product gas is cooled down to 180 °C. The released heat can be used for preheating of the biomass dryer air [44]. Furthermore, the product gas filter separates further fine dust particles from the product gas stream and conveys

them back to the combustion reactor. After that, tar is separated in a scrubber, and water is condensed. Biodiesel (RME) is used as solvent. The product gas exits the scrubber with a temperature of 40 °C. Afterwards, it is compressed in a blower, before it is dried to a water content of 1.5% and fed to the compression and preheating of the direct reduction plant. The CO₂-enriched flue gas leaves the combustion reactor with a temperature of 900 °C. The flue gas is cooled down to 180 °C by the steam superheater and a flue gas cooler. Steam is heated up to 450 °C in a countercurrent heat exchanger. Fly ash is removed out of the system by a flue gas filter. A partial flow from the flue gas is recirculated and mixed with pure oxygen. Pure oxygen is produced by an air separation unit. The remaining flue gas stream is compressed in the flue gas blower, and water is condensed in a flue gas dryer. The cleaned CO₂-rich gas can be used in different CCU processes, like urea or methanol synthesis [44].

The integration approach offers the advantage to use existing equipment, like the air separation unit from the steel-making facility. Furthermore, the generated product gas can be used directly in the direct reduction plant, as reducing gas [44]. For this application, a compression up to approx. 2.5 bar and preheating of the product gas up to 900 °C are necessary.

2.4 Simulation of mass and energy balances with IPSEpro

The calculation of mass and energy balances for different operation points with the stationary equation-orientated flow sheet simulation software IPSEpro enables the validation of process data. All data which cannot be measured during experimental test runs can be determined by the calculation of closed mass and energy balances. These equations are solved by the numerical Newton-Raphson Algorithm [59, 60]. Therefore, no models regarding kinetic or fluid dynamic approaches are considered. The used simulation models within the software IPSEpro are based on model libraries, which were developed at TU Wien over many years [61]. All experimental results from the pilot plant at TU Wien, presented within this publication, were validated with IPSEpro. Uncertainties are given by the accuracy of measurement data which relies on used analysis methods. The measurement accuracy of the ultimate and proximate analysis is listed in Table 4. The validation percentage error of the gasification model is covered by the range of values which are listed in Table 3. For further information regarding IPSEpro, a reference is made to [61, 62]. Due to the validation of the results from the pilot plant at University of Stuttgart, a reference is made to [57].

The simulation results for the OxySER concept for the production of below zero emission reducing gas presented in Section 2.3 are based on scale up of the experimental results of the pilot plants. The simulation model of the dual fluidized

Table 2 Important gas-solid and gas-gas reactions during thermochemical fuel conversion [50]

| | | | |
|----------------------------------------------------------------|---------------------------------------------------------------------|---------------------|------|
| Important heterogeneous reactions (gas-solid) | | | |
| Oxidation of carbon | $C + O_2 \rightarrow CO_2$ | Highly exothermic | (1) |
| Partial oxidation of carbon | $C + \frac{1}{2} O_2 \rightarrow CO$ | Exothermic | (2) |
| Heterogeneous water-gas shift reaction | $C + H_2O \rightarrow CO + H_2$ | Endothermic | (3) |
| Boudouard reaction | $C + CO_2 \rightarrow 2 CO$ | Endothermic | (4) |
| Hydrogenation of carbon | $C + 2 H_2 \rightarrow CH_4$ | Slightly exothermic | (5) |
| Generalized steam gasification of solid fuel (bulk reaction) | $C_x H_y O_z + (x-z)H_2O \rightarrow x CO + (x-z + \frac{y}{2})H_2$ | Endothermic | (6) |
| Important homogeneous reactions (gas-gas) | | | |
| Oxidation of hydrogen | $2 H_2 + O_2 \rightarrow 2 H_2O$ | Highly exothermic | (7) |
| Homogeneous water-gas shift reaction | $CO + H_2O \rightarrow CO_2 + H_2$ | Slightly exothermic | (8) |
| Methanation | $CO + 3 H_2 \rightarrow CH_4 + H_2O$ | Exothermic | (9) |
| Generalized steam reforming of hydrocarbons | $C_x H_y + x H_2O \rightarrow x CO + (x + \frac{y}{2})H_2$ | Endothermic | (10) |
| Generalized dry reforming of hydrocarbons | $C_x H_y + x CO_2 \rightarrow 2x CO + \frac{y}{2} H_2$ | Endothermic | (11) |
| Important reactions of active bed material (limestone) for SER | | | |
| Carbonation | $CaO + CO_2 \rightarrow CaCO_3$ | Exothermic | (12) |
| Calcination | $CaCO_3 \rightarrow CaO + CO_2$ | Endothermic | (13) |

bed steam gasification system is based on an exergy study of T. Pröll [63].

2.5 Techno-economic assessment with net present value calculation

The techno-economic assessment regarding the net present value (NPV) calculation serves as decision-making tool for the valuation of upcoming investments. The NPV is a function of the investment and operating costs. The operating costs are multiplied by the cumulative present value factor, which includes the interest rate and the plant lifetime. Therefore, the NPV calculation helps to compare expected payments in the future with current payments. Further information can be found in [54, 64]. Cost rates have been updated to the year 2019 by using data from a chemical engineering plant cost index (CEPCI) database [65]. For the calculation of the investment costs, the cost-scaling method was used [66].

The techno-economic analysis is based on the following *business case* that an operator of a direct reduced iron plant would like to build a new reducing gas supply unit driven by a biogenic feedstock. The goal to produce 100 MW reducing gas should be achieved with regard to CO₂ emissions. The reference option (option 0) is the production of reducing gas by steam reforming of natural gas. Furthermore, three biogenic alternative options (options 1–3) are compared with the reference option:

- *Option 0* (reference case): Production of 100 MW reducing gas through steam reforming of natural gas
- *Option 1*: Production of 100 MW reducing gas through gasification of wood chips by SER
- *Option 2*: Production of 100 MW reducing gas through gasification of wood chips by an integrated OxySER plant
- *Option 3*: Production of 100 MW reducing gas through gasification of wood chips by a greenfield OxySER plant

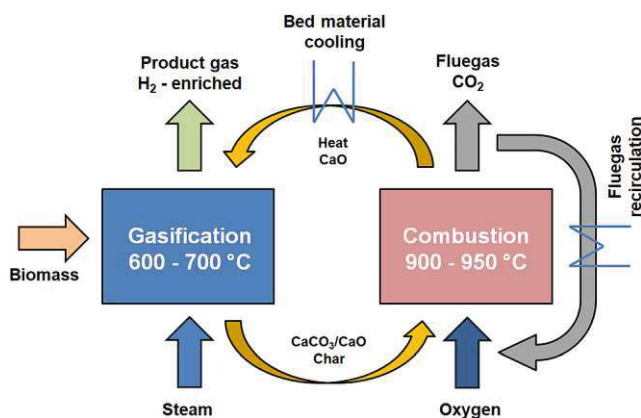


Fig. 1 Concept of OxySER [55]

The SER process in option 1 requires no pure oxygen, consequently no ASU for operation. However, the flue gas of the SER process cannot be exploited in further utilization steps because of the high nitrogen content in the flue gas. The alternative option 2 is based on the SER process in combination with oxyfuel combustion implemented in an existing iron and steel plant facility. The process heat is used for preheating of the reducing gas. The required oxygen is delivered from an existing ASU within the iron and steel plant facility. Furthermore, the OxySER process is based on the assumption that the CO₂ is sold as product for utilization to a urea synthesis plant. Option 3 is based on the OxySER process without the benefits from option 2.

Table 3 Comparison product and flue gas composition of conventional gasification, SER, and OxySER [37, 57]

| Parameter | Unit | Conventional gasification (100 kW) | Gasification by SER (100 kW) | Gasification by SER (200 kW) | Gasification by OxySER (200 kW) |
|---------------------------------------------------------|-----------------------|------------------------------------|------------------------------|------------------------------|---------------------------------|
| Plant location | | TU Wien | TU Wien | University Stuttgart | University Stuttgart |
| Reference | | [37] | [37] | [57] | [57] |
| Fuel | | Wood pellets | Wood pellets | Wood pellets | Wood pellets |
| Bed material | | Olivine | Limestone | Limestone | Limestone |
| Particle size | mm | 0.4–0.6 | 0.5–1.3 | 0.3–0.7 | 0.3–0.7 |
| Product gas composition | | | | | |
| Water (H ₂ O) | vol.-% | 30–45 | 50–65 | 50 | 50 |
| Hydrogen (H ₂) | vol.-% _{dry} | 36–42 | 55–75 | 69–72 | 70 |
| Carbon monoxide (CO) | vol.-% _{dry} | 19–24 | 4–11 | 8–11 | 8 |
| Carbon dioxide (CO ₂) | vol.-% _{dry} | 20–25 | 6–20 | 5–7 | 8 |
| Methane (CH ₄) | vol.-% _{dry} | 9–12 | 8–14 | 11–12 | 11 |
| Non cond. hydrocarbons (C _x H _y) | vol.-% _{dry} | 2.3–3.2 | 1.5–3.8 | 2–3 | 3 |
| Dust particles | g/Nm ³ | 10–20 | 20–50 | n.m. | n.m. |
| Tar | g/Nm ³ | 4–8 | 0.3–0.9 | 14 | 6 |
| Flue gas composition | | | | | |
| Water (H ₂ O) | vol.-% | n.m. | n.m. | 14 | 30 |
| Oxygen (O ₂) | vol.-% _{dry} | n.m. | n.m. | 7 | 9 |
| Nitrogen (N ₂) | vol.-% _{dry} | n.m. | n.m. | 46 | - |
| Carbon dioxide (CO ₂) | vol.-% _{dry} | n.m. | n.m. | 47 | 91 |

n.m., not measured

This means that, in option 3, the costs for pure oxygen are higher in consideration to the use of a greenfield ASU. Furthermore, no earnings through CO₂ utilization are considered.

Furthermore, a payback analysis has been done by solving the following equation, where A are the savings minus the operation and maintenance costs, P is the present worth capital costs, and IR is the interest rate. The variable n represents the number of years to return the investment in comparison with the reference case [67].

$$A = P * \frac{IR * (1 + IR)^n}{(1 + IR)^n - 1}$$

3 Results and discussion

Based on experiences of the pilot plant from the TU Wien and the University of Stuttgart, combined with the previously

Table 4 Proximate and ultimate analyses of used wood pellets for gasification test runs [51]

| Parameter | Unit | Meas. accuracy (%) | Wood pellets (100 kW) |
|----------------------------------|----------------------|--------------------|-----------------------|
| Water content (H ₂ O) | wt.-% | ± 4.3 | 7.2 |
| Ash content (550 °C) | wt.-% _{dry} | ± 9.2 | 0.2 |
| Carbon (C) | wt.-% _{daf} | ± 1.0 | 50.8 |
| Hydrogen (H) | wt.-% _{daf} | ± 5.0 | 5.9 |
| Nitrogen (N) | wt.-% _{daf} | ± 5.0 | 0.2 |
| Sulfur (S) | wt.-% _{daf} | ± 7.5 | 0.005 |
| Chlorine (Cl) | wt.-% _{daf} | ± 7.5 | 0.005 |
| Oxygen (O)* | wt.-% _{daf} | - | 43.1 |
| Volatile matter | wt.-% _{daf} | ± 0.45 | 85.6 |
| Lower heating value, moist | MJ/kg | ± 1.0 | 17.4 |

* Calculated by difference to 100 wt.-%_{daf}

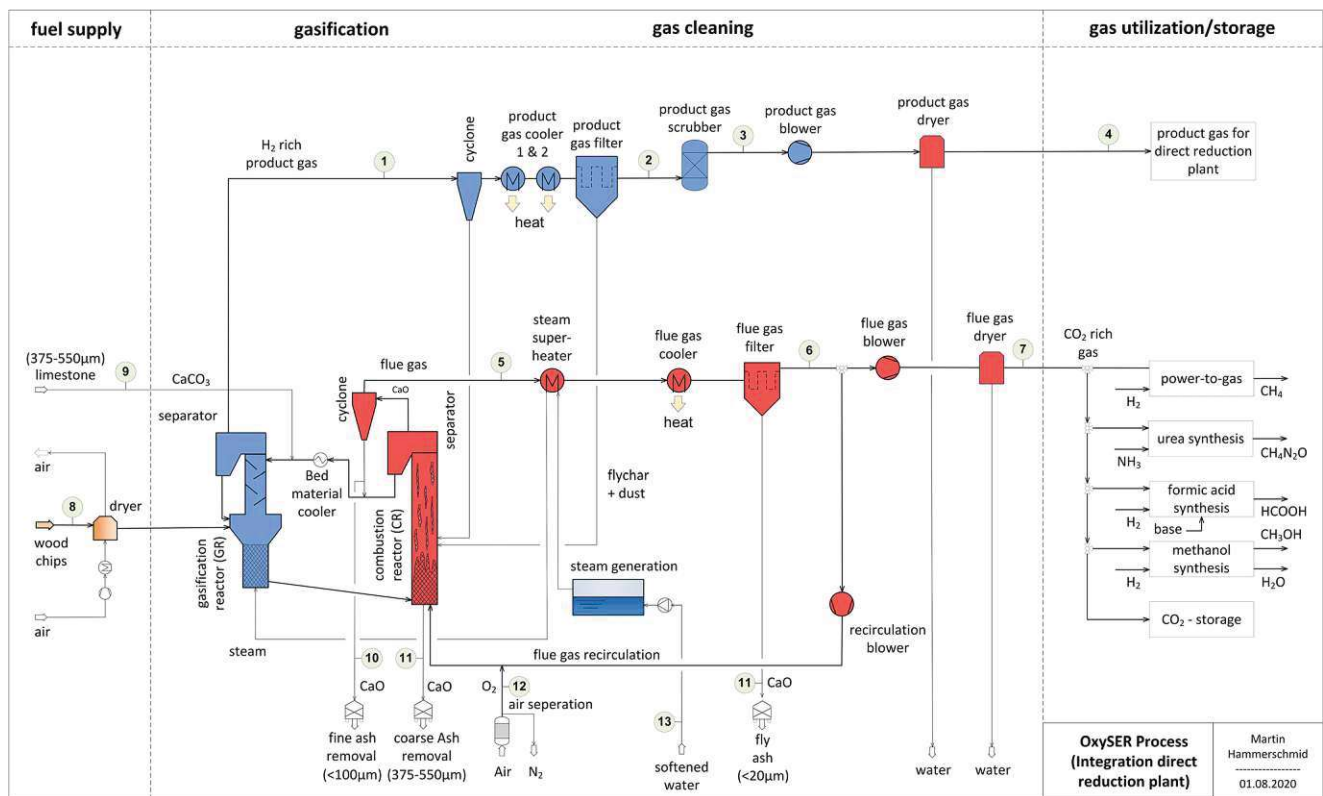


Fig. 2 OxySER plant concept with 100-MW product gas power for the production of reducing gas as feedstock for a direct reduction plant

described concept, mass and energy balances for the OxySER plant concept for integration in a direct reduction plant were calculated. Furthermore, mass and energy balances are the basis for a techno-economic assessment. In Table 5 the most important streamline data of chosen flow streams, marked in Fig. 2, are shown. Table 6 and Table 7 represent the input and output data and operating parameters of an OxySER plant.

Table 6 shows the input and output flows of an OxySER plant with 100 MW product gas energy. It can be seen that 50,400 kg/h of wood chips and 11,020 Nm³/h of pure oxygen are required for the generation of 28,800 Nm³/h product gas. The product gas is used as reducing gas in the direct reduction route. Furthermore, 36,100 kg/h of CO₂ can be recovered for further utilization. The costs for final disposal of 1050 kg/h of ash and dust have been taken into account.

In Table 8, the main requirements on the product gas for the utilization in the direct reduction plant are listed. The comparison illustrates that the generated below zero emission product gas out of the OxySER plant meets, except from the temperature and pressure, all the requirements. The concept is based on the assumption that the reducing gas is compressed and preheated before it is fed to the direct reduction plant. Therefore, the required temperature and pressure are reached after compression and preheating of the product gas.

The techno-economic assessment relies on the results of the IPSEpro simulation. Table 9 represents the fuel prices for chosen fuel types and cost rates for utilities. It is thus

evident that the European natural gas price with 25 €/MWh is more expensive than in other continents. Exemplary, the costs for one employee per year are assumed to 70,000 €/a, and the expected plant lifetime of an OxySER plant is 20 years.

Table 10 represents the investment cost rates for the NPV calculation. The presented investment costs are based on total capital investment costs of realized fluidized bed steam gasification plants driven as combined heat and power plants reduced by the costs through the gas engine. Furthermore, this investment costs are updated by CEPCI and scaled with the cost-scaling method. For the integrated OxySER plant, the assumption was made that the oxygen from the air separation unit (ASU) of the iron and steel plant is used. For the green-field OxySER plant, the whole investment costs for an ASU were added.

The techno-economic analysis is based on the Section 2.5 that described *business case*, wherein an operator of a direct reduced iron plant would like to build a new reducing gas supply unit driven by a biogenic feedstock. The NPV calculation, which is shown in Table 11, serves as decision-making tool. The goal to produce 100 MW reducing gas should be achieved with regard to CO₂ emissions. The reference option (option 0) is the production of reducing gas by steam reforming of natural gas. Furthermore, three biogenic alternative options (options 1–3), which are described in Section 2.5, are compared with the reference option.

Table 5 Streamline data of the OxySER concept according to Fig. 2

| Parameter | Unit | Product gas streams | | | | Flue gas streams | | |
|---------------------------------------------------------|-----------------------|--------------------------|------------------------------|--------------------------------|-------------------------|-----------------------|---------------------------|---------------------|
| | | Product gas after GR (1) | Product gas after filter (2) | Product gas after scrubber (3) | Reducing gas for DR (4) | Flue gas after CR (5) | Flue gas after filter (6) | Flue gas to CCU (7) |
| Streamline in Fig. 2 | – | | | | | | | |
| Pressure | Bara | Ambient | Ambient | Ambient | Ambient | Ambient | Ambient | Ambient |
| Temperature | °C | 675 | 150 | 40 | 60 | 950 | 160 | 160 |
| Mass flow rate | kg/h | 26,000 | 25,500 | 16,000 | 15,800 | 93,200 | 92,600 | 36,100 |
| Volume flow rate | Nm ³ /h | 40,500 | 40,000 | 28,800 | 28,400 | 53,000 | 52,500 | 20,500 |
| Water content | wt.-% | 35.0 | 35.0 | 8.0 | 1.5 | 15.0 | 15.0 | 5.0 |
| Hydrogen (H ₂) | vol.-% _{dry} | 69.2 | 69.2 | 69.2 | 69.2 | 0 | 0 | 0 |
| Carbon monoxide (CO) | vol.-% _{dry} | 9.1 | 9.1 | 9.1 | 9.1 | 2.8 | 2.8 | 2.8 |
| Carbon dioxide (CO ₂) | vol.-% _{dry} | 6.5 | 6.5 | 6.5 | 6.5 | 91.2 | 91.2 | 91.2 |
| Methane (CH ₄) | vol.-% _{dry} | 11.0 | 11.0 | 11.0 | 11.0 | 0 | 0 | 0 |
| Non cond. Hydrocarbons (C _x H _y) | vol.-% _{dry} | 2.4 | 2.4 | 2.4 | 2.4 | 0 | 0 | 0 |
| Oxygen (O ₂) | vol.-% _{dry} | 0.1 | 0.1 | 0.1 | 0.1 | 6.0 | 6.0 | 6.0 |
| Nitrogen (N ₂) | vol.-% _{dry} | 1.7 | 1.7 | 1.7 | 1.7 | 0 | 0 | 0 |
| Dust particle | g/Nm ³ | 10 | 0.025 | 0 | 0 | 20 | 0 | 0 |
| Tar content* | g/Nm ³ | 4 | 0.5 | 0.025 | 0.025 | 0 | 0 | 0 |

*Tar is considered in the simulation model as naphthalene (main component in the DFB product gas) [51]

Table 11 represents the net present value calculation for the production of 100 MW reducing gas. Therein, the fuel energy per year, the investment costs including interest and fuel costs per year are listed. Beside the fuel costs, Table 11 shows also all other consumption-related costs. Costs for CO₂ emission certificates are paid only for the use of fossil fuels (reference case). The relative NPV represents the profitability of alternative production routes in comparison with the reference case and the payback period for return of investment. The NPV of all alternative options

(1–3) shows negative values. This means that the operation of SER and OxySER with wood chips based on the expected plant lifetime of 20 years is less profitable than the reference option. The techno-economic comparison between SER and OxySER shows that in option 2, the earnings through carbon dioxide are higher than the oxygen costs. In option 3, no earnings through CO₂ utilization and no benefits regarding oxygen costs have been considered. Therefore, an extremely negative NPV in option 3 is the result. The payback analysis shows that only option 2

Table 6 Input and Output data of an OxySER plant with 100 MW product gas energy

| Input | | | | | Output | | | | |
|-------------------------------------|----------------------|--------------------|--------|----------|--------------------------|----------------------|--------------------|--------|------|
| Parameter | Streamline in Fig. 2 | Unit | Value | Ref. | Parameter | Streamline in Fig. 2 | Unit | Value | Ref. |
| Bed material inventory | - | kg | 25,000 | [37] | Product gas | (3) | Nm ³ /h | 28,800 | IPSE |
| Fuel (wood chips) | (8) | kg/h | 50,400 | [37] | Flue gas | (8) | Nm ³ /h | 53,000 | IPSE |
| Fresh bed material | (9) | kg/h | 1770 | [37, 64] | Ash and dust | (11) | kg/h | 1050 | [37] |
| Cooling capacity in % of fuel power | - | % | 5–20 | [68] | Bed material | (10) | kg/h | 1000 | [44] |
| Electricity consumption | - | kW | 2800 | [37] | Carbon dioxide (for CCU) | (7) | kg/h | 36,100 | [37] |
| Oxygen | (12) | Nm ³ /h | 11,020 | [37] | | | | | |
| Fresh water | (13) | kg/h | 378 | [37] | | | | | |
| Scrubber solvent (RME) | - | kg/h | 200 | [37] | | | | | |
| Flushing gas | - | Nm ³ /h | 500 | [37] | | | | | |

Table 7 Operating parameters of an OxySER plant with 100 MW product gas energy

| Parameter | Unit | Value | Ref. |
|-----------------------------------------|--------|---------|------|
| Lower heating value, moist (wood chips) | MJ/kg | 9.53 | [37] |
| Water content (wood chips) | wt.-% | 40 | [37] |
| Combustion temperature | °C | 900–950 | [69] |
| Gasification temperature | °C | 625–680 | [69] |
| Particle size (bed material) | µm | 375–550 | Asm. |
| Coarse ash | µm | 375–550 | Asm. |
| Fine ash | µm | < 100 | Asm. |
| Very fine ash | µm | < 20 | Asm. |
| Water content (PG to DR) | vol.-% | 1.50 | IPSE |
| CO ₂ recovery rate* | % | >95 | IPSE |

$$*CO_2 = \frac{CO_2 \text{ volume flow flue gas}}{CO_2 \text{ volume flow total (FG+PG)}}$$

could return the investment regarding the expected interest rate in comparison with the reference case. However, the payback time of 24 years is very long and would not be profitable. Option 1 and option 3 could not return the investment in comparison to the reference case.

Furthermore, the reducing gas production costs of the four different routes were calculated. As can be seen from Table 11, the production costs (LCOP) of the reference case are with 39.0 €/MWh as the lowest followed by the integrated OxySER process with 39.4 €/MWh. Figure 3 represents the discounted expenses and revenues, divided in the main cost categories. It can be seen that the fuel costs are the main cost driver in the process. The techno-economic comparison points out that the production costs of a below zero emission reducing gas could only be in the range of steam-reformed natural gas, if generated CO₂ can be utilized and the pure oxygen is delivered by an integrated ASU. Otherwise, the production of biomass-based reducing gas via the SER process is preferable. A further reduction of the production costs of the biomass-based reducing gas could be reached by the use of cheaper fuels.

Table 8 Requirements on product gas for the utilization in the direct reduction plant [22, 70]

| Parameter | Unit | Requirement reducing gas | Value product gas |
|---------------------------|--------------------|--------------------------|-------------------|
| Temperature | °C | > 900 | 60 |
| Pressure | bara | 2–4 | 1.05 |
| H ₂ /CO ratio | - | 0.5 - ∞ | 7.6 |
| Gas quality* | - | > 9 | 9.8 |
| Methane | vol.-% | > 3.5 | 11.0 |
| Sulfur (H ₂ S) | ppm | < 100 | < 20 |
| Soot | mg/Nm ³ | < 100 | - |

$$*Gas \text{ quality} = (\%CO + \%H_2) / (\%CO_2 + \%H_2O) [70]$$

Additionally, a sensitivity analysis of the NPV calculation has been created. The results for the sensitivity analysis based on the NPV of option 2 are shown in Fig. 4. The sensitivity analysis shows that the fuel prices of natural gas and wood chips are the most sensitive cost rates. The fuel cost rates depend very much on the plant location. Furthermore, the NPV in this techno-economic comparison is also sensitive to the investment costs of the reducing agent production route, the revenues through CCU, the price of CO₂ emission certificates, the plant lifetime, the operating hours, and the interest rate. The revenues through CCU depend on the availability of consumers. The sensitivity to operating hours and plant life time reaffirms high importance to a high plant availability during the whole plant life cycle. Cost rates for operating utilities, maintenance, and employees are less sensible to the results.

Finally, a comparison of the production costs of the biomass-based reducing gas with other reducing agents like reformed natural gas, hydrogen, or coke has been done. The comparison in Fig. 5 shows that the production of biomass-based reducing gas via OxySER (option 2) and SER is more than twice as expensive as the production of coke in a coking plant, but it is in the same range than the production of reducing gas via steam reforming of natural gas. All fuel costs are based on European price levels. Especially, the natural gas price strongly depends on the plant site. For example, the natural gas price in Europe is four to five times higher than in North America [33]. This is the reason why most of the existing direct reduction plants are built in oil-rich countries [33]. The production of hydrogen using water electrolysis is currently economically not competitive. On the ecologic point of view, the use of biomass-based reducing gas without CCU decrease the CO₂ emissions of the whole process chain for the production of crude steel down to 0.28 t CO₂e/t_{CS}. This amounts to a reduction of CO₂ emissions in comparison with the integrated BF-BOF route by more than 80%. Further on, the use of CCU within an OxySER plant could create a CO₂ sink, since biomass releases the same amount of CO₂ as it aggregates during its growth.

With regard to 8.1 million tons of crude steel production in Austria, in the year 2017 [4], and an estimated woody biomass potential of around 50 PJ in the year 2030 [21], 13 biomass-based reducing gas plants (OxySER or SER) with a reducing gas power of 100 MW could be implemented. This would result in the production of around 35 Mio. GJ of biomass-based reducing gas for the direct reduction process, which is sufficient for the production of 3.5 Mio. tons of crude steel. One of the biomass-based reducing gas plants could be operated via the OxySER process with regard to the CCU potential from the nearby urea synthesis plant of 300,000 t CO₂ per year [46]. Further CCU potential could be arise through the production of CO₂-derived fuels or chemicals [41].

Table 9 Cost rates for utilities and NPV calculation

| Utility cost rate | Unit | Value | Ref. | NPV cost rate | Unit | Value | Ref. |
|----------------------------------|---------------------|--------|-------------|---------------------------------------------|------|---------|----------|
| Wood chips (Austria) | €/MWh | 15.7 | [71] | Maintenance costs per year | %/a | 2.00 | [54] |
| Natural gas (Austria) | €/MWh | 25.0 | [72] | Insurance, administration, and tax per year | %/a | 1.50 | [73] |
| Electricity | €/kWh _{el} | 0.04 | [64] | Number of employees (integration) | - | 3 | [64] |
| Limestone | €/t | 35 | [64] | Number of employees (greenfield) | - | 7 | [44, 64] |
| Nitrogen | €/Nm ³ | 0.003 | [64] | Expected plant life time | a | 20 | [73] |
| Fresh water | €/t | 0.02 | [64] | Annual operating hours | h/a | 7500 | [64] |
| Solvent (RME) | €/t | 960 | [64] | Interest rate (IR) | % | 6 | [74] |
| Oxygen (air separator available) | €/Nm ³ | 0.022* | [44] | Costs of one employee per year | €/a | 70,000* | [64] |
| Oxygen (greenfield) | €/Nm ³ | 0.075* | [37] | | | | |
| Emission allowances certificate | €/t _{CO2} | 23 | [75] | | | | |
| Costs for ash disposal | €/t | 90 | CHP Güssing | | | | |
| CO ₂ expenses | €/Nm ³ | 0.03 | [76] | | | | |

4 Conclusion and outlook

The scope of this publication was the investigation of a concept for the production of a below zero emission reducing gas for the use in a direct reduction plant and whether it has a reasonable contribution to a reduction of fossil CO₂ emissions within the iron and steel sector in Austria. The gasification via SER allows the in situ CO₂ sorption via the bed material system CaO/CaCO₃. Therefore, a selective transport of carbon dioxide from the product gas to the flue gas stream is reached. The use of a mix of pure oxygen and recirculated flue gas as fluidization agent in the CR results in a nearly pure CO₂ flue gas stream. Through the in situ CO₂ sorption, CO₂ recovery rates up to 95% can be reached. The CO₂ could be used for further synthesis processes like, e.g., the urea synthesis. Therefore, a below zero emission reducing gas could be produced.

The experimental and simulation results show that the produced below zero emission OxySER product gas meets all requirements for the use in a direct reduction plant. The use of the biomass-based reducing gas out of the SER process within a MIDREX plant would decrease the emitted CO₂ emission by 83% in comparison to the blast furnace route. The use of a below zero emission reducing gas out of the OxySER process by the use of CCU would create a CO₂ sink. The results of the techno-economic assessment show that the production of reducing gas via sorption-enhanced reforming in combination with oxyfuel combustion can compete with the natural gas route, if the required pure oxygen is delivered by an available ASU and if CCU is possible. Otherwise, the SER process is more profitable. Furthermore, the sensitivity analysis of the cost rates exhibited that the fuel and investment costs are strongly dependent on the profitability of the OxySER plant and in consequence the direct reduction plant.

Table 10 Investment costs for NPV calculation

| Parameter | Unit | Value | Ref. |
|-----------------------------------------------------------------------------------|--------|-------|---------------------------|
| Investment costs SER plant (total capital investment costs)* | Mio. € | 85 | [37] adapted by CEPCI |
| Investment costs integrated OxySER plant (SER plus maintenance ASU)** | Mio. € | 91 | [37, 66] adapted by CEPCI |
| Investment costs greenfield OxySER plant (SER plus total investment costs ASU)*** | Mio. € | 115 | [37, 66] adapted by CEPCI |
| Investment costs Steam Reformer natural gas | Mio. € | 54 | [77] adapted by CEPCI |

*Investment costs are based on scaled total capital investment costs of realized dual fluidized bed steam gasification plants driven as combined heat and power plants reduced by the costs of the gas engine/investment costs updated with CEPCI [37]

**Investment costs are based on costs SER plant raised by a third of the ASU maintenance costs (2% of the investment costs per year with an expected lifetime of 20 years)/assumption: 50% of ASU is used for OxySER plant and 50% for iron and steel plant

***ASU investment costs: approx. 30 Mio. € [66] adapted by CEPCI

Table 11 Net present value calculation for the production of 100 MW reducing gas

| Parameter | Unit | Steam Reforming (100% natural gas) | SER (100% wood chips) | Integration OxySER (100% wood chips) | Greenfield OxySER (100% wood chips) |
|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------|------------------------------------|-----------------------|--------------------------------------|-------------------------------------|
| | | Option 0 | Option 1 | Option 2 | Option 3 |
| Boundary conditions | | | | | |
| Reducing gas for direct reduction | MW | 100 | 100 | 100 | 100 |
| Natural gas consumption | MWh/a | 750 000 | | | |
| Wood chips consumption | MWh/a | | 997 500 | 997 500 | 997 500 |
| Investment costs incl. interest | € | 54 000 000 | 85 000 000 | 91 000 000 | 115 000 000 |
| Expenses | | | | | |
| Fuel costs natural gas | €/a | 18 750 000 | | | |
| Fuel costs wood chips | €/a | | 17 010 000 | 17 010 000 | 17 010 000 |
| CO ₂ emission certificates | €/a | 3 450 000 | | | |
| Maintenance, insurance, etc. | €/a | 1 890 000 | 2 975 000 | 3 185 000 | 4 025 000 |
| Employee costs | €/a | 70 000 | 210 000 | 210 000 | 490 000 |
| Auxiliaries | €/a | | 1 916 000 | 1 916 000 | 1 916 000 |
| Electricity costs | €/a | 356 000 | 835 500 | 835 500 | 835 500 |
| Ash disposal costs | €/a | | 709 000 | 709 000 | 709 000 |
| Oxygen costs | €/a | | | 1 818 000 | 6 198 500 |
| Sum of expenses per year | €/a | 24 516 000 | 23 655 500 | 25 683 500 | 31 184 000 |
| Earnings | | | | | |
| Earnings CO ₂ utilization | €/a | | | 4 102 000 | |
| Sum of earnings per year | €/a | | | 4 102 000 | |
| Net present value calculation | | | | | |
| Expenses - Earnings | €/a | 24 516 000 | 23 655 500 | 21 581 500 | 31 184 000 |
| Additional investment costs (P) (compared to reference option) | € | 0 | 31 000 000 | 37 000 000 | 61 000 000 |
| Operating expenses savings (A) | €/a | 0 | 860 500 | 2 934 500 | -6 668 000 |
| Relative Net Present Value | € | 0 | -21 128 000 | -3 340 000 | -137 500 000 |
| Payback analysis (Return of investment period compared to reference case) | | | | | |
| Payback time (n)* | a | - | ∞ | 24 | ∞ |
| Production costs reducing gas (LCOP) | | | | | |
| Production costs reducing gas (LCOP)** | €/MWh | 39.0 | 41.4 | 39.4 | 54.9 |
| | €/GJ | 10.8 | 11.5 | 10.9 | 15.3 |
| * Payback analysis: $A = P * \frac{IR * (1+IR)^n}{(1+IR)^n - 1}$ [67] | | | | | |
| ** LCOP = $\frac{\text{Sum of discounted (expenses-earnings)}}{\text{Discounted Delivered reducing gas}} = \frac{\text{€}}{\text{MWh}} = \frac{\text{€}}{\text{GJ}}$ [78, 79] | | | | | |

Fig. 3 Relative net present value

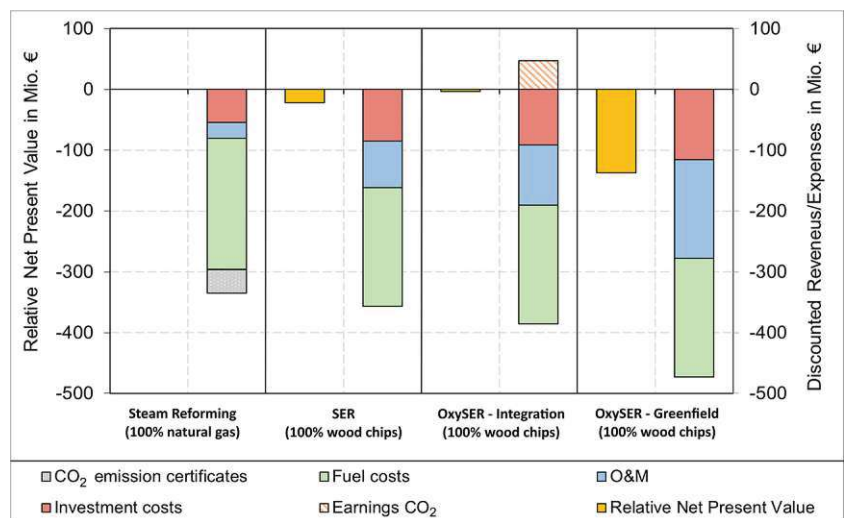
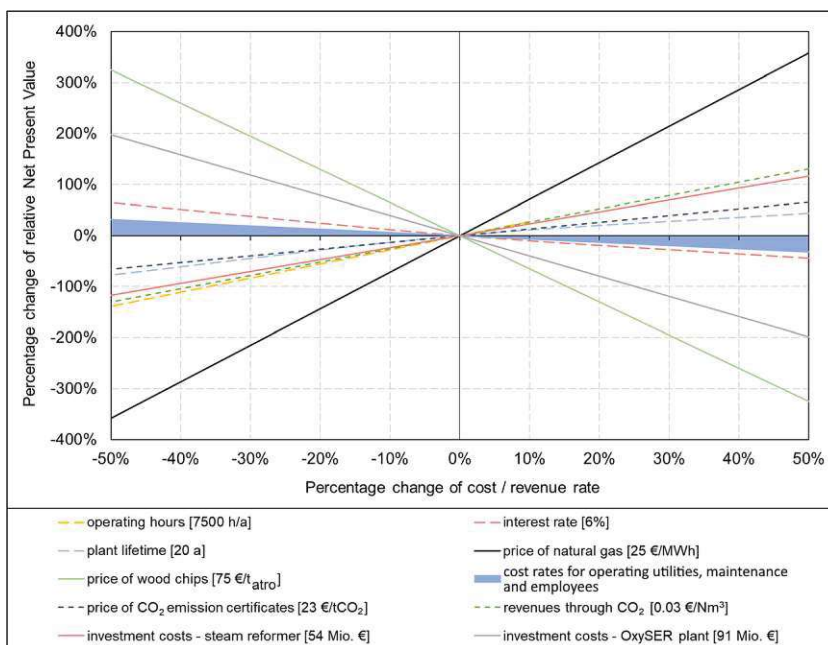


Fig. 4 Sensitivity analysis of the NPV calculation

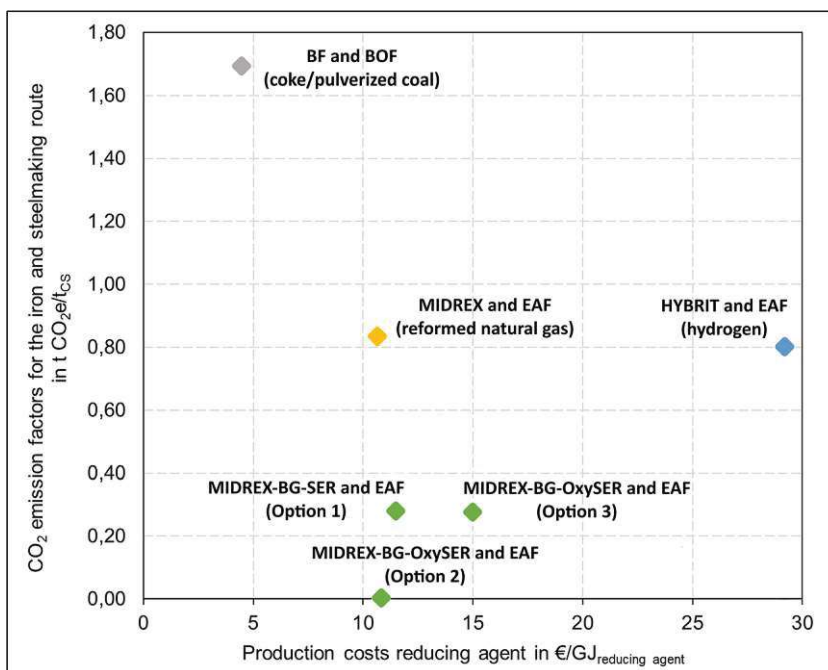


The production costs of the biomass-based reducing gas are more than twice as high as the fossil coke, which is used mainly in the blast furnace route.

Summing up, the presented integrated concept and the calculated results enable valuable data for further design of the proposed concept. Beforehand a demonstration at a significant scale is recommended. Further on, the implementation of the energy flows from an iron and steel plant within the simulation model could improve the current model regarding to efficiency. The profitability of the direct reduction with a biomass-

based reducing gas or natural gas is strongly dependent on the availability of sufficient fuel. With regard to the woody biomass potentials in Austria in the year 2030, the production of 3.5 Mio. tons of crude steel by the use of biomass-based reducing gas could be reached. Due to the substitution of the integrated BF and BOF route by the MIDREX-BG-SER and EAF route, the reduction of 6.8 Mio. tons of CO₂e could be reached. This amount would decrease the CO₂ emissions within the iron and steel sector in Austria by 50%. Concluding, the production of biomass-based reducing gas

Fig. 5 Economic and ecologic comparison of different Iron and Steelmaking routes [12, 20, 25, 34, 80]



could definitely help to contribute on the way to defossilization of the iron and steelmaking industry in Austria.

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Data availability The data that support the findings of this study are available from the corresponding author, M. Hammerschmid, upon reasonable request.

Compliance with ethical standards

Conflicts of interest The authors declare that they have no conflict of interest.

Code availability Not applicable.

Abbreviations AISI, American Iron and Steel Institute; Asm., assumption; ASU, air separation unit; BF, blast furnace; BG, biomass-based reducing gas; BOF, basic oxygen furnace; C, carbon; CaCO₃, calcium carbonate; CaO, calcium oxide; CCS, carbon capture and storage; CCS/U, carbon capture and storage or utilization; CCU, carbon capture and utilization; CEPCI, chemical engineering plant cost index; CH₃OH, methanol; CH₄, methane; CH₄N₂O, urea; CHP, combined heat and power; CIRCORED, novel direct reduction technology; CO, carbon monoxide; CO₂, carbon dioxide; CO₂e, carbon dioxide equivalent; COG, coke oven gas; COREX, smelting reduction technology; COURSE50, CO₂ ultimate reduction steelmaking process by innovative technology for cool Earth 50 located in Japan; CR, combustion reactor; CS, crude steel; C_xH_y, non condensable hydrocarbons; DR, direct reduction; DRI, direct reduced iron; dry, dry basis; EAF, electric arc furnace; EU-28, member states of the European Union (until January 2020); FG, flue gas; FINEX, smelting reduction technology; FINMET, direct reduction technology; GR, gasification reactor; H₂, hydrogen; H₂O, water; H₂S, hydrogen sulfide; HCOOH, formic acid; HISARNA, novel bath-smelting technology; HM, hot metal; HYBRIT, Hydrogen Breakthrough Ironmaking Technology; IPSEpro, software tool for process simulation; LCOP, leveled costs of products; MIDREX, state-of-the-art direct reduction technology; MOE, molten oxide electrolysis; N₂, nitrogen; NH₃, ammonia; NPV, net present value; O₂, oxygen; OPEX, operational expenditure; OxySER, sorption-enhanced reforming in comb. with oxyfuel combustion; PG, product gas; POSCO, iron and steelmaking company located in Korea; Ref., reference; RME, rapeseed methyl ester; SER, sorption-enhanced reforming; t_{CS}, tons of crude steel; TGR, top-gas recycling; ULCOLYSIS, novel electric direct reduction technology; ULCORED, novel direct reduction technology; ULCOS, ultra-low CO₂ steelmaking; ULCOWIN, novel electric direct reduction technology; vol.-%, volumetric percent wet; vol.-%_{dry}, volumetric percent dry; wt.-%, weight percent wet; wt.-%_{daf}, weight percent dry and ash free; wt.-%_{dry}, weight percent dry

Symbols %CO, volume percent of carbon monoxide within reducing gas; %CO₂, volume percent of carbon dioxide within reducing gas; %H₂, volume percent of hydrogen within reducing gas; %H₂O, volume percent of water within reducing gas; A, savings minus the operation and

maintenance costs; *IR*, interest rate; *n*, payback period; *P*, present worth capital costs

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PAPER IX

Müller S., Theiss L., Fleiß B., Hammerschmid M., Fuchs J., Penthor S., Rosenfeld D.C.,
Lehner M., Hofbauer H.

DUAL FLUIDIZED BED BASED TECHNOLOGIES FOR CARBON DIOXIDE REDUCTION – EXAMPLE HOT
METAL PRODUCTION

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Responsibility of the author:

- Investigated different concepts and methodologies
- Created the simulation model used for the calculation of data
- Interpreted the results
- Corresponding author independent of mentoring support



Dual fluidized bed based technologies for carbon dioxide reduction — example hot metal production

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Abstract

The present work describes the results achieved during a study aiming at the full replacement of the natural gas demand of an integrated hot metal production. This work implements a novel approach using a biomass gasification plant combined with an electrolysis unit to substitute the present natural gas demand of an integrated hot metal production. Therefore, a simulation platform, including mathematical models for all relevant process units, enabling the calculation of all relevant mass and energy balances was created. As a result, the calculations show that a natural gas demand of about 385 MW can be replaced and an additional 100 MW hydrogen-rich reducing gas can be produced by the use of 132 MW of biomass together with 571 MW electricity produced from renewable energy. The results achieved indicate that a full replacement of the natural gas demand would be possible from a technological point of view. At the same time, the technological readiness level of available electrolysis units shows that a production at such a large scale has not been demonstrated yet.

Keywords Carbon dioxide reduction · Oxyfuel combustion · Sorption enhanced reforming · Biomass gasification

1 Introduction and short description

In the past, large amounts of easily accessible primary energy resources accompanied by an efficient energy infrastructure enabled the development of pleasant wealth in Europe. At the same time, limited resources in Europe itself led to significant dependency on energy imports. The energy strategy of the European Union for the future aims at pretending secure, safe and affordable energy. Furthermore, the energy strategy includes the utilization of local available resources, a reduction of greenhouse gas emissions, and the development of new innovative energy technologies, as new high performance low-carbon technologies [1, 2].

The production of hot metal causes significant fossil carbon dioxide (CO₂) emissions. Therefore, numerous researchers investigate the reduction of fossil carbon dioxide emission in the surrounding of hot metal production. Hereby, the replacement of fossil energy carriers should not impair the quality of final products or lead to a reduced availability of the applied production process.

So far, the following question:

Which setup in an existing integrated hot metal production would enable the most reasonable reduction of fossil carbon usage based on available dual fluidized bed based technologies?

has not been answered. The following paper describes the results of investigations aiming at reasonable changes with respect to the reduction of fossil carbon usage of a hot metal production process. At the beginning, a short review about the potential application of dual fluidized bed based technologies is carried out [3]. Afterwards, an application for hot metal production is investigated. This step bases on the creation of an optimized industrial model of the integrated hot metal production. Within the present work, a description of the production process is carried out. Furthermore, the present paper discusses:

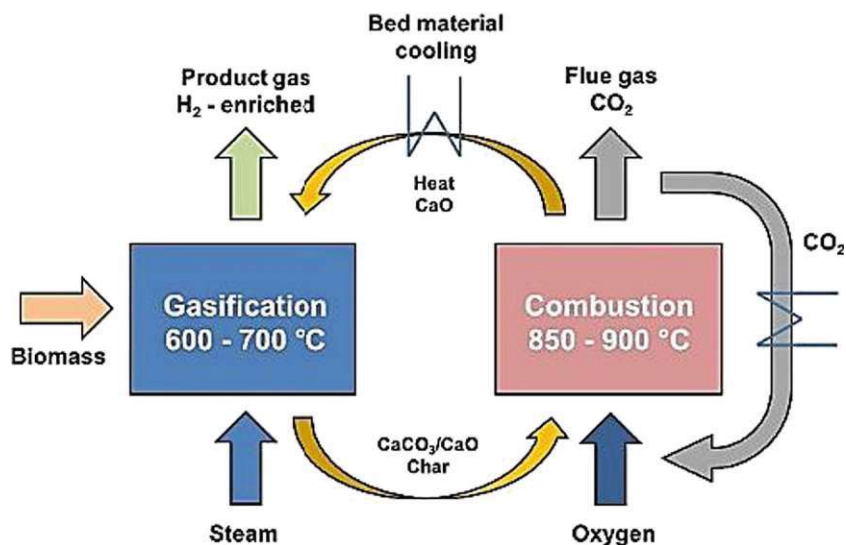
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Fig. 1 Sorption enhanced reforming with oxyfuel combustion (OxySER)



- The application of dual fluidized bed technology for carbon dioxide reduction,
- The used concept for the reduction of fossil-based carbon emissions,
- The creation of an industrial plant model,
- The simulation results achieved,
- And an interpretation of the achieved results with respect to a large-scale implementation.

Herby, within the present work, an industrial plant information model is defined as a simplified reproduction of the reality of an industrial plant focused on the investigation of important existing relations. This includes beside technical parameters such as fundamental chemical and thermodynamic relations also practical aspects for the operation such as operation control, economic performance, ecologic impact and legal aspects relevant for an optimized future operation. Following this definition, the focus of the illustrated model within the present work lies on the mass-

and energy balances of a large-scale plant with respect to an ecologic optimization by a reduction of the fossil carbon utilization.

2 Technological review

The dual fluidized bed (DFB) technology offers a broad range of applications for the utilization of CO₂ neutral energy carriers like biomass. Conventional biomass steam gasification is an already well-known technology. Based on this technology the sorption enhanced reforming process (SER) was developed and enables the in-situ removal of CO₂ from the product gas. Consequently, the chemical equilibrium of the product gas is shifted and a high hydrogen (H₂) content can be obtained in the product gas. By applying oxyfuel combustion in the combustion reactor (OxySER), an almost pure CO₂ stream can be obtained [3]. Thus, both gas streams can contribute to a CO₂

Fig. 2 CO₂ and steam gasification within a DFB reactor system

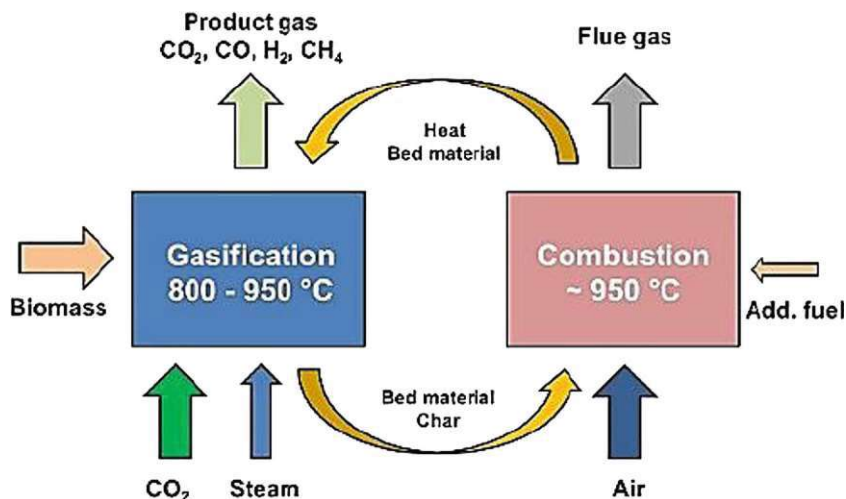
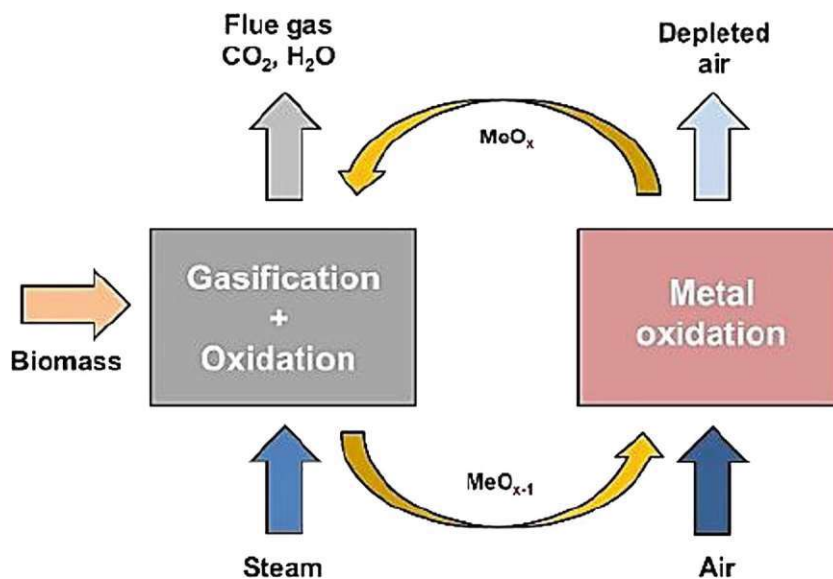


Fig. 3 Chemical looping combustion of solid biogenic fuels within a DFB reactor system



reduction in the atmosphere: On the one hand, a hydrogen-rich product gas is generated, which can be used directly as a reducing agent in steel industry or as basis for synthesis processes like methanation. On the other hand, a pure CO_2 stream is produced, which either can be stored or, again, used for synthesis processes like methanation [4]. A scheme of the process can be found in Fig. 1 [5].

Another variation of a DFB process is the gasification with CO_2 instead of steam shown in Fig. 2. The usage of CO_2 leads to a product gas with high carbon content. This seems to be contradictory, since decarbonization is the overall aim of energy intensive industries. Nevertheless, carbon, although it might be provided as CO_2 or CO respectively, is an important element for many processes and could be reused in this way to generate a product gas as basis for further utilization [6, 7].

Last but not least, a DFB reactor system can also be used for the so-called chemical looping combustion of solid biogenic fuels (BioCLC) process, which has promising

potential for capturing CO_2 due to its low energy demand. The principle of the chemical looping process is shown in Fig. 3 and is based on the use of a metal oxide as bed material and oxygen carrier. This oxygen carrier is used to burn the gas, which is produced by the gasification reactions of the biomass with steam. The oxygen carrier itself is oxidized in the air reactor again. This procedure enables the production of a nitrogen-free flue gas. Therefore, the gas consists mainly of CO_2 and H_2O [8].

Several experimental campaigns have been conducted with the advanced 100 kW_{th} pilot plant at TU Wien to investigate the different technological approaches. The mentioned pilot plant consists of a gasification reactor and a combustion reactor with an overall height of about 7 m. More details about the latest reactor-design can be found in literature [9]. Figure 4 shows pictures of the described test plant.

The three very different dual fluidized bed processes offer different advantages and disadvantages. Table 1

Fig. 4 Upper part of the 100 kW_{th} dual fluidized bed test plant (left), lower part (right) [10]



Table 1 Comparison of presented processes regarding different parameters

| | OxySER [11, 12] | CO ₂ and steam gasification of biomass [13, 14] | BioCLC [15, 16] |
|-----------------------------------------------------|-----------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------|-----------------------------------------------------------|
| Achieved products | Hydrogen-rich product gas*, CO ₂ and heat | Carbon monoxide-rich product gas* and heat | Carbon dioxide and heat* |
| Technical readiness | Pilot scale | Pilot scale | Pilot scale |
| Advantages | Product gas with high H ₂ content and pure CO ₂ for further utilization | Utilization of CO ₂ as gasification agent enabling the reuse of carbon. | Pure CO ₂ from biomass for further utilization |
| Disadvantages | Air separation unit necessary for oxygen supply | limited CO ₂ conversion efficiency | No chemical energy supply |
| CO ₂ -avoidance (t/MWh _{out})* | ~0.35 | 0.25–0.50 | 0.25–0.50 |
| CO ₂ -capture (t/MWh _{out})* | ~0.25 | - | ~0.4 |
| * Reference | Investigated case in present work | Synthesis gas from coal | Conventional combustion process |

provides and overview about different parameters of these processes. Typically, the gasification processes OxySER and CO₂ gasification produce a product gas, which contains chemical energy, whereas the CLC process as a typical combustion process provides heat. Especially, the OxySER process offers advantages, but also disadvantages. On the one hand, a hydrogen-rich product gas is produced; on the other hand, carbon from biomass is selectively transported to a separate CO₂ stream. Since biomass is the only renewable carbon source, the OxySER concept must be implemented wisely. This means the integration in an environment where H₂ on the one hand, but pure CO₂ on the other hand is needed. Therefore, within the present work, the application of OxySER in the surrounding of hot metal production is investigated.

3 Methodology for application in hot metal production

Figure 5 shows the used concept for the creation of an industrial plant model enabling a full replacement of the natural gas supply of a hot metal production. As can be seen, the proposed concept consists of a biomass gasification system, an electrolysis unit, a carbon dioxide removal unit and a methanation unit. The used biomass gasification system is operated as dual fluidized bed gasification system converting woody biomass into a hydrogen-rich gas. Besides, the used operation mode enables the production of a carbon dioxide-rich gas to follow a carbon capture perspective and enable further utilization. Figure 6 gives a detailed illustration of the basic principle of this process, which has been investigated intensively in the

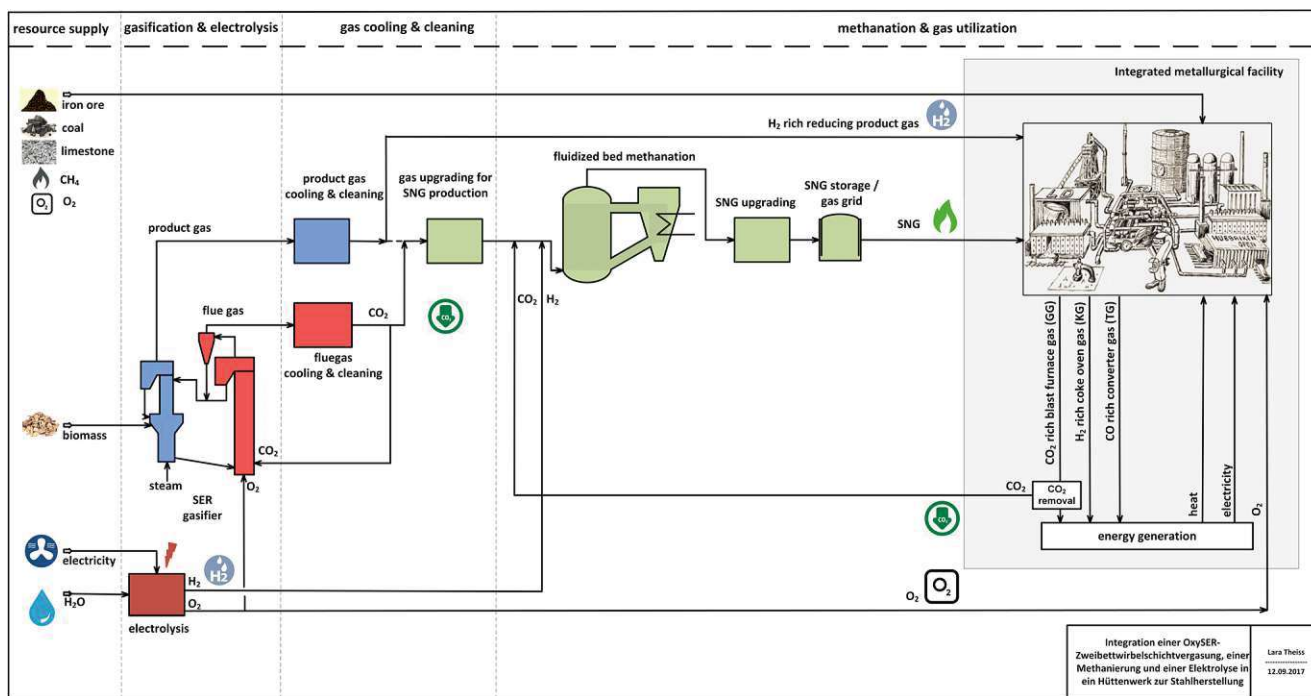


Fig. 5 Concept for full replacement of the natural gas demand of a hot metal production

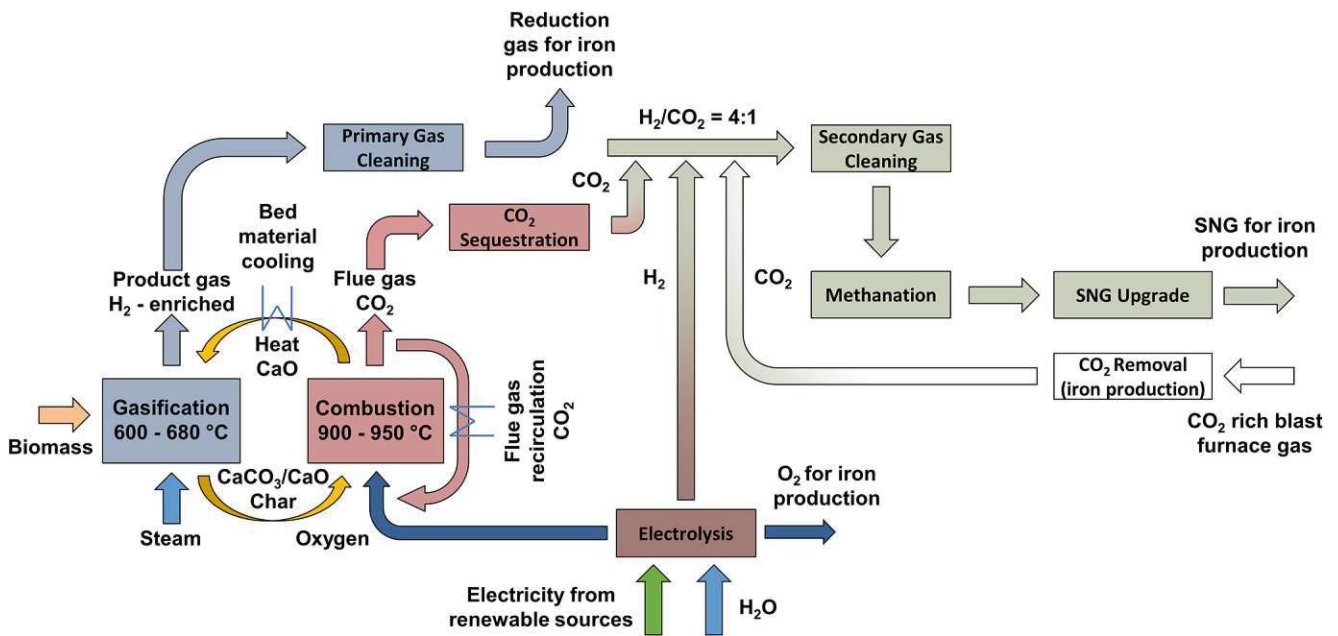


Fig. 6 Schematic illustration of sorption enhanced reforming in combination with oxyfuel combustion (OxySER), subsequent gas cleaning, electrolysis, and CO₂ separation

last years [17–19]. A detailed description of the so-called OxySER process can be found in literature [20, 27, 28].

Furthermore, the described concept in Fig. 5 includes an electrolysis unit. The electrolysis unit is used to provide

Table 2 Comparison of main important gas streams within the investigated process model

| Parameter | Unit | Coke oven gas | Converter gas | Blast furnace gas | Product gas OxySER [19] | Flue gas OxySER [20] | Requirements methanation [26] |
|------------------------------------------------|----------------------------------|---------------|---------------|-------------------|-------------------------|----------------------|-------------------------------|
| Hydrogen (H ₂) | vol.-% _{db} | 66 | 0.6 | 3.7 | 70 | - | 30–90 |
| Carbon monoxide (CO) | vol.-% _{db} | 5.8 | 51.8 | 25 | 8 | - | 0–25 |
| Carbon dioxide (CO ₂) | vol.-% _{db} | 1.2 | 20 | 23 | 8 | 91 | 0–30 |
| Methane (CH ₄) | vol.-% _{db} | 22 | - | - | 11 | - | 0–100 |
| Nitrogen (N ₂) | vol.-% _{db} | 3 | 27.6 | 48.3 | - | - | <3 |
| Non condensable C _x H _y | vol.-% _{db} | 2 | - | - | 3 | - | x |
| Oxygen (O ₂) | vol.-% _{db} | - | - | - | - | 9 | x |
| Dust particles | mg/Nm ³ | 1.3 | 0.8 | 0.6 | 30 | x | <0.5 |
| Sulfur: H ₂ S, COS, CS ₂ | mg/Nm ³ _{db} | 250 | - | 208 | 122 | - | <0.4 |
| Nitrogen: NH ₃ , HCN | mg/Nm ³ _{db} | 510 | - | 0.4 | 13 | - | <0.8 |
| Halogens: HCl, HBr, HF | mg/Nm ³ _{db} | - | - | 5.8 | 12 | - | <0.06 |
| Alkali metals: K, Na | mg/Nm ³ _{db} | - | - | - | x | - | <1 |
| Tar | mg/Nm ³ _{db} | x | x | x | 25 | - | <0.1 |
| Nitrogen oxide as NO ₂ | mg/Nm ³ _{db} | x | x | x | - | 900 | x |
| Sulfur dioxide (SO ₂) | mg/Nm ³ _{db} | x | x | 11.9 | - | <3 | x |
| Volume flow | Nm ³ /h | 75,000 | 50,000 | 1,000,000 | | | |
| Lower heating value | MJ/Nm ³ _{db} | 17.3 | 6.8 | 4.0 | 14.7 | | |

x not available and experimental determination recommended

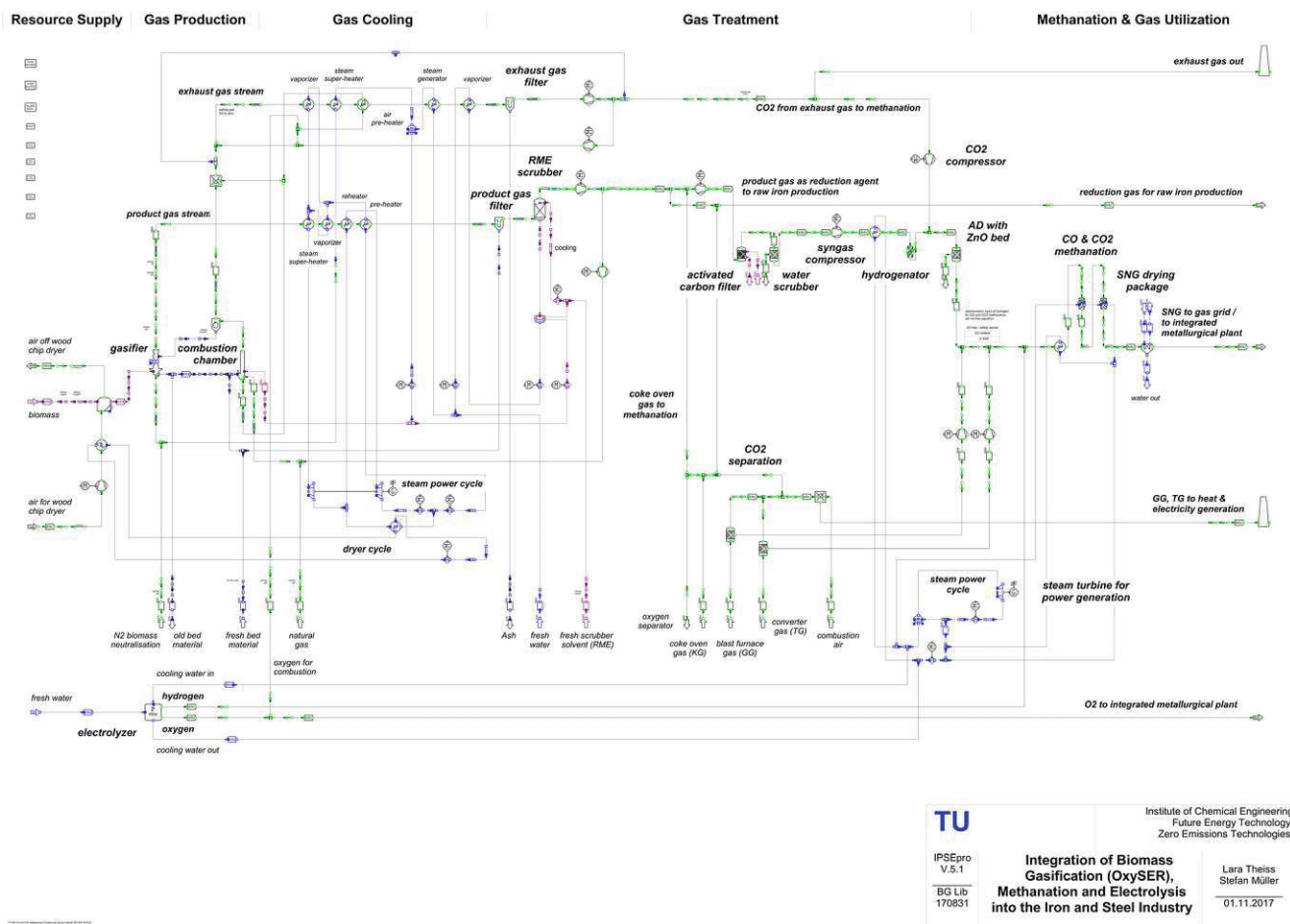


Fig. 7 Flowchart of the created process simulation model

hydrogen to the overall process allowing methanation according to stoichiometric ideal conditions accounting present biogenic carbon as well as recycled carbon dioxide from a blast furnace. A carbon dioxide removal unit is used to recycle carbon dioxide from the CO₂-rich blast furnace gas as a feedstock for the methanation process. A detailed description of the process units can be found elsewhere [21–23]. Figure 6 shows a simplified sketch of the process concept.

Hereby, the applied biomass gasification system and the electrolysis are used to replace most parts of the fossil energy demand of a hot metal production [24]. To achieve this, the electricity demand for electrolysis is delivered by renewable sources like wind, water or sun power. As a result, a hydrogen-rich reducing gas as well as a synthetic natural gas (SNG) is supplied to the production process. At this point, it must be mentioned that the illustrated concept is not influenc-

Table 3 Results of the mass and energy balance calculations

| Input parameter | Unit | Value | Output parameter | Unit | Value |
|-----------------------------------|------|--------|----------------------------------|------|--------|
| Mass balance | | | | | |
| Wood chips dry | kg/h | 37468* | H ₂ -rich product gas | kg/h | 15,671 |
| Carbon dioxide (CO ₂) | kg/h | 40,247 | Synthetic natural gas | kg/h | 28,206 |
| Fresh water | kg/h | 60,365 | Oxygen (O ₂) | kg/h | 96,325 |
| Others | kg/h | 5791 | Others | kg/h | 3669 |
| Energy utilization: | | | | | |
| Wood chips wet | MW | 132 | H ₂ -rich product gas | MW | 100 |
| Electricity electrolysis | MW | 571 | Synthetic natural gas | MW | 385 |

*50 t/h before dryer

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Integration of Biomass Gasification (OxySER), Methanation and Electrolysis into the Iron and Steel Industry

Lara Theiss
 Stefan Müller
 01.11.2017

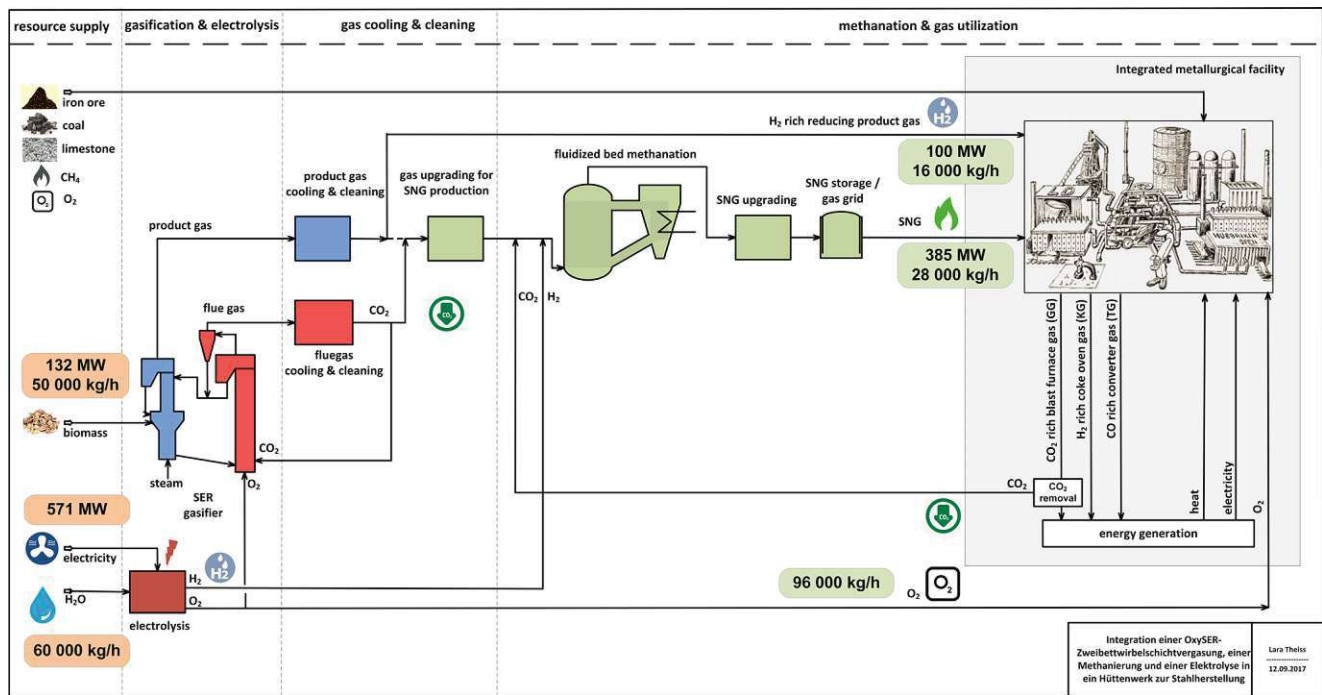


Fig. 8 Optimized industrial plant model with reduced fossil carbon usage

ing the existing core production process and therefore ensures the desired quality of final products. The described process concept has been used for the creation of a digital industrial plant information model (IPIM) [25] allowing the calculation of all relevant information with respect to the investigated process concept. The executed work included:

- A data analysis of existing mass and energy streams,
- Modeling of an optimized conceptual design (cf. Fig. 4),
- The creation of an industrial plant information model for the
- Calculation of simulation results,
- As well as an interpretation of results achieved.

4 Results and discussion

Table 2 gives an overview about main relevant data with respect to existing gas streams in an integrated steel mill. As can be seen, product gas from dual fluidized bed biomass gasification shows similar properties like coke oven gas. Therefore, the product gas represents a valuable gas for a direct utilization within an integrated steel mill. In comparison, the converter gas and blast furnace gas show significantly lower heating values. Therefore, this gas streams can be considered containing low value with respect to its utilization possibilities. So, this gas streams are only used for the generation of process heat. At the same time, the blast furnace gas represents the

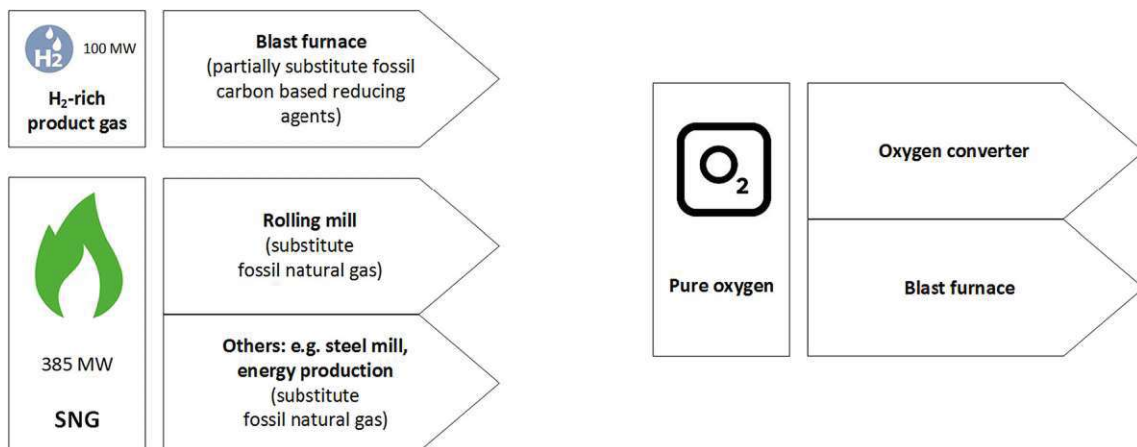


Fig. 9 Applications of the generated product streams

largest off-gas stream containing a high share of carbon dioxide. A process configuration, which is reusing the carbon dioxide from this stream, would enable a high carbon recycling rate and lead to significantly lower carbon dioxide emissions. The described circumstances formed the basis for the created concept displayed in Fig. 5.

Figure 7 shows a process flow diagram of the created process simulation model. It contains all relevant process units and parameters in the scale of an existing hot metal production. The used model library has been described by [29] with modifications and additions made by the authors of the present work [30]. The calculation of the OxySER process bases on experimental data from TU Wien and Stuttgart. Besides, it takes into account made experiences with the biomass gasification plants in Güssing, Oberwart, and Senden [25]. The calculation of the electrolysis was carried out based on provided data from literature [21]. The electrolysis unit supplies additional oxygen (O₂), which can be supplied to the hot metal production to cover internal demand. The calculation of the methanation of reused carbon dioxide with hydrogen produced by water electrolysis as well as the carbon dioxide from the OxySER has been carried out based on experiences provided by several authors from literature [31]. Described sources have been used for the creation of the simulation model.

Within the simulation model, available high temperature heat is used to produce electricity via a steam cycle process. The created simulation model was implemented into the surrounding of a hot metal production. As a result, Table 3 summarizes the main calculation results achieved and Fig. 8 shows a simplified illustration of main calculation results. 50 t/h of wood chips and 571 MW of electricity are necessary to enable a substitution of the present natural gas demand of 385 MW. The products obtained by the OxySER process, methanation, and excess oxygen from electrolysis as can be seen in Fig. 9 can be fully used within the hot metal production plant:

- i. The OxySER product gas serves as reducing agent within the blast furnace and thereby replaces fossil pulverized coal injection (PCI) coal.
- ii. The produced SNG replaces the total natural gas demand of the hot metal production. Natural gas is mainly used in the rolling mill, steel mill and for energy production in an integrated hot metal production.
- iii. The excess oxygen from electrolysis is used in integrated metallurgical facility for example in an oxygen converter and the blast furnace.

The process corresponds to a possible equivalent CO₂ reduction of 77 t/h due to the replacement of fossil natural gas. Additionally, 100 MW of a valuable hydrogen-rich product

gas can be supplied to the hot metal reduction process, which could reduce equivalent fossil CO₂ emissions by 33 t/h due to the replacement of PCI coal [32]. 40 t/h of carbon dioxide needs to be recycled from the blast furnace gas to supply enough carbon for the methanation unit. The implemented reuse of carbon dioxide would represent an interesting approach for the recycling of fossil carbon following the idea of a circular economy action plan [33]. As by-product from the electrolysis, 96 t/h of pure oxygen are produced. Illustrated numbers indicate the expected size of the proposed process concept if a full replacement of the present natural gas demand is the aim of further implementation steps.

5 Conclusion and outlook

The present work was executed to determine possible modifications to enable a significant reduction of fossil carbon dioxide emissions of a hot metal production. Biomass gasification, electrolysis, and carbon dioxide separation was identified as possible short-term modifications to enable a full replacement of the present natural gas demand. The proposed integration of the identified process units was calculated by the use of a process simulation model. As a result, the proposed modifications are not expected to cause negative influence on the quality of final products. As a further result, the carried out calculations show that:

- 50 t/h of woody biomass,
- 60 t/h of water,
- And 571 MW of electricity

would be necessary to replace fossil natural gas. This would represent about 1% of the primary energy usage of woody biomass in the energy sector in Austria [34]. Besides, the recycling of 20,000 Nm³/h of carbon dioxide (CO₂) is required to operate the proposed process configuration. This would represent a reuse of consumed fossil and biogenic carbon sources. The investigated process within the present work could enable a saving of about 800,000 tCO₂/a. An estimation indicates that this represents a share of up to 1% of the overall CO₂ emissions in Austria [35]. The achieved results show that the natural gas demand of an integrated hot metal production can be replaced by the use of electrolysis, biomass gasification and a methanation system. The created model indicates valuable data for the design of proposed modifications. The following steps:

- Up-scaling of OxySER to 150 MW,
- Up-scaling of electrolysis to 600 MW,

and long-term tests of the methanation step with real gas from hot metal production are recommended to be executed before an implementation at a larger scale. These steps are

necessary, as so far, no electrolysis unit at this scale is available, whereas, the largest dual fluidized bed biomass gasification system so far has been built in the scale of 32 MW in Gothenburg, Sweden.

Furthermore, a detailed analysis of the economic circumstances is recommended in near future. Recently published results indicate important economic aspects with respect to a further implementation of the presented concept. Main findings show that biomass based concepts could be economically feasible if the biomass price, the natural gas price and the price for CO₂ emission certificates provide a reasonable development with respect to the political aims formulated by the European Commission [36, 37]. A quick acceleration of accompanying implementation steps is demanded, if there should be any chance to reach the climate targets from the latest Paris Agreement.

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Abbreviations BioCLC, chemical looping combustion of solid biogenic fuels; CO₂, carbon dioxide; DFB, dual fluidized bed; DME, dimethyl ether; H₂, hydrogen; IPIM, industrial plant information model; O₂, oxygen; OxySER, sorption enhanced reforming in combination with oxyfuel combustion; PCI, pulverized coal injection; SER, sorption enhanced reforming; SNG, synthetic natural gas

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