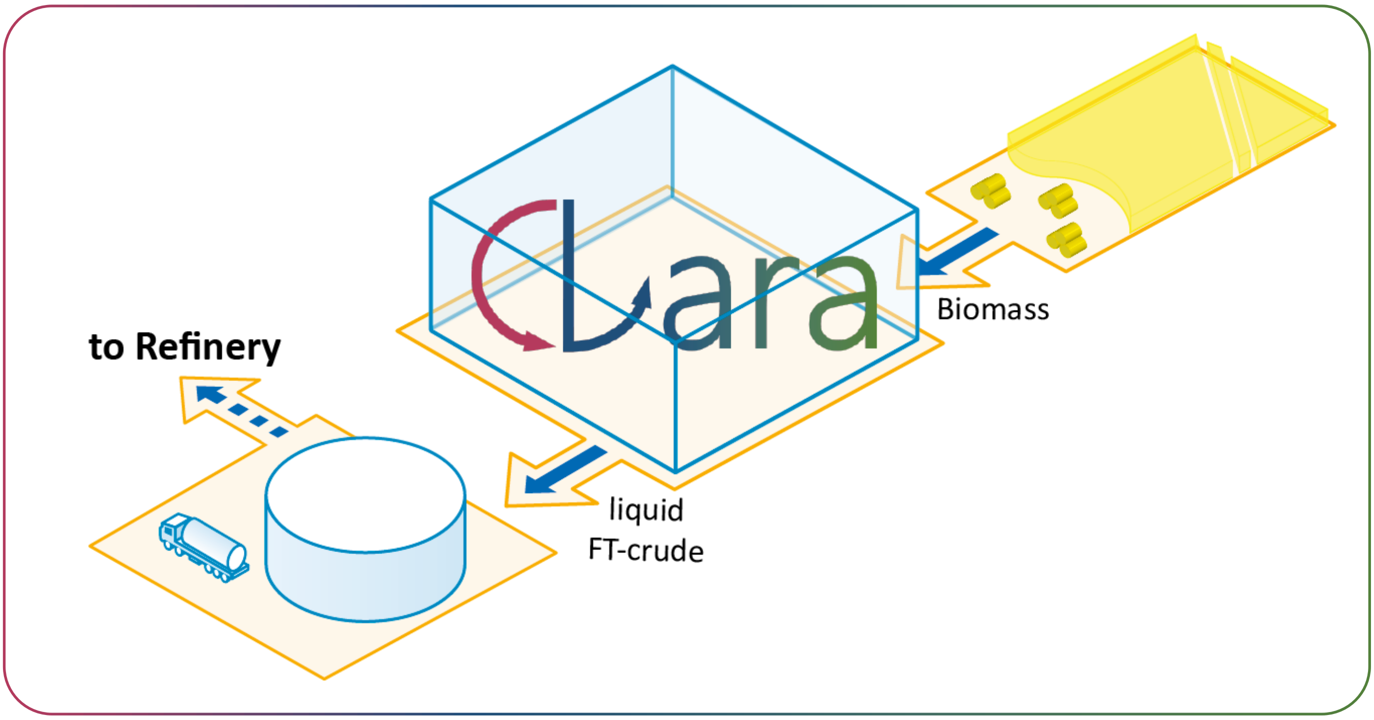
Chemical **L**ooping G**a**sification fo**r** Sustain**a**ble Production of Biofuels

H2020 Research and Innovation action
Grant Agreement no 817841

Public Report IV



Authors: Falko Marx[1], Paul Dieringer[1], Jochen Ströhle[1], Carl Linderholm[2], Nikos Detsios[3], Konstantinos Atsonios[3], Christian Aichernig[4], Thorsten Liese[5], Frank Buschsieweke[5], Ye Huang[6], Nadine Gürer[7].

Affiliation: [1]Technical University Darmstadt, [2]Chalmers University of Technology, [3]Centre for Research & Technology Hellas, [4]Aichernig Engineering GmbH, [5]RWE Power AG, [6]University of Ulster, [7]Vienna University of Technology

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 817841.

Publication date: 2023-06-13**,** Access the digital version via this link: https://clara-h2020.eu/public-reports/

Executive Summary

The final project year of the CLARA project had the partners focused on the chemical looping gasification (CLG) technology modeling (see Chapter 3), the full chain pilot testing of the previously defined Biomass-to-Liquid (BtL) process chain (see Chapter 4), and the subsequent validation of full process chain models (see Chapter 5). Moreover, the final assessment of the whole technology was carried out regarding risks (see Chapter 6) and techno-economic and life cycle assessments have been performed (see Chapter 7)

The 1.5D CLG model was further developed and optimized with results from the 50 kW CLG unit from *CSIC* and used for further optimization of the industrial scaled chemical looping gasifier design by *AE*. The resulting agreement of the finalized model with the experiments is presented in Chapter 3.

The full chain pilot plant including the 1 MW_{th} chemical looping gasifier located at *TUDA* was commissioned in March 2022 and subsequently used for three full chain pilot test campaigns using industrial wood pellets (IWP) as reference feedstock, pine forest residue (PFR), and wheat straw pellets (WSP). The PFR pellets and WSP were prepared by *ABT* according to pretreatment method developed by *CENER*. During the tests carried out by *TUDA* over 100 t of biomass were converted to syngas and the full process chain was operated demonstrating the technical possibility of the suggested BtL process chain including the novel sour gas cleaning process in a test-rig by *RWE* and the Fischer-Tropsch (FT) synthesis using a catalyst by *UNICRE*. The details of the pilot tests are available in Chapter 4 including details on the operation of the gas cleaning pilot plant.

Using the data generated during the full chain pilot testing *CERTH* performed model validation to ensure the alignment of the previously developed full chain process model with the experimental results obtained by *TUDA*. The model output was used for the definition and sizing of equipment required in an industrial application of the full BtL process chain. The comparison of the results from the model and the experimental pilot plant results are presented in Chapter 5.

Rounding up the efforts of the CLARA consortium is the assessment of the whole BtL process chain by *RWE*, *ULSTER*, *AE* and *TU WIEN*. Included in this report are the results from Health&Saftey (Chapter 6.1) and environmental (Chapter 6.2) risk assessment. Moreover, Technological risks have been assessed as well as the economic feasibility of the CLARA process which are detailed in Chapter 6.3 and 6.4 respectively. Complementing the assessment of the BtL process chain are techno-economic and life cycle analysis which can be found in Chapters 7.1 and 7.2.

Content

1 Introduction

Within the scope of the *Horizon 2020* project CLARA, a novel biomass-to-biofuel process chain is to be investigated. Through cutting-edge research and interdisciplinary cooperation, the CLARA consortium, consisting of thirteen international members including universities, research institutes and industrial partners, aims to investigate the complete process chain and bring the suggested technologies to market maturity.

Here, the advantages of utilizing locally availably biogenic residues and the economy of scale are combined, through decentralized feedstock pre-treatment facilities and a centralized fuel production plant in the scale of 100-300 MW_{th}. The fuel production plant itself consists of a chemical looping gasifier for the production of a raw syngas, a gas treatment train to provide the required syngas composition for the subsequent synthesis, and a Fischer-Tropsch (FT) reactor to covert the syngas into liquid FT-crude. This crude can then be purified and upgraded to ready-to-use second generation drop-in biofuels in existing state-of-the-art refineries. A schematic overview over the suggested biomass-to-biofuel process chain, with its four main subunits, is shown in Figure 1.

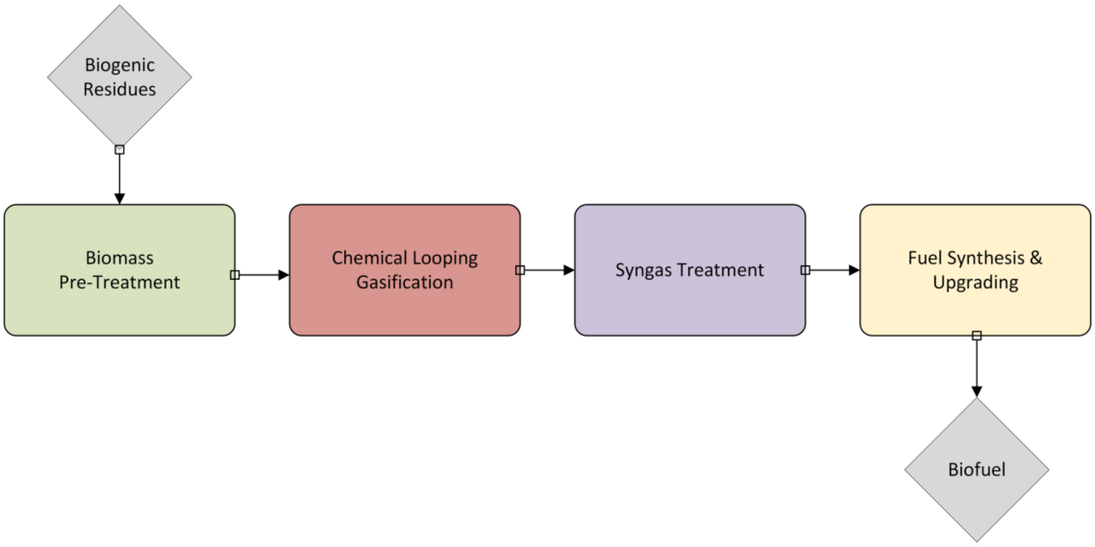


Figure 1: Simplified scheme of full biomass-to-biofuel process chain.

As an introduction, this report again informs the reader about the project motivation and the underlying goals (Chapter 2) as well as the some technology development carried out to achieve these (Chapter 3). Moreover, findings and highlights unveiled by the CLARA consortium during the last 18 months (Feb. 2022 – Apr. 2023) are presented in the following. This includes the pilot testing of the entire process chain in Chapter 4 and the modeling of the process chain (Chapter 5). Risk analysis and techno-economic and life cycle analysis are detailed in Chapters 6 and 7. A summary of this public report can be found in the last Section (Chapter).

In case you have any remarks or questions, do not hesitate to contact us (jochen.stroehle@est.tu-darmstadt.de). More details and updates can be found on our project website (https://clara-h2020.eu/).

2 Project Motivation & Project Goals

Significant reductions in greenhouse gas emissions are required to prevent a surge in global average temperatures, exceeding the much discussed 1.5 °C threshold of the Paris Agreement. Here, the de-carbonization of the transport sector, which utilizes over a third of the global final energy [1] and is responsible for almost one quarter of the European greenhouse gas emissions [2], is a key concern on the route to achieve this goal. Particularly, the substitution of fossil fuels in transport sectors for which electrification is presently not viable (e.g. road transport and aviation), remains a major challenge.

To tackle this issue, the European Union has set a target of a share of 14 % renewable energy in the transport sector by 2030 in the Renewable Energy Directive (RED II) in 2018 [3]. This requires the large-scale deployment of biofuels in addition to electrification and the increased deployment of rail transport. Since the wide-spread utilization of energy crops is being strongly criticized publicly, the utilization of biogenic residues, which do not impact food availability and prices negatively, in the energy and transport sector is to be intensified. Therefore, substantial advances in renewable fuel generation are required.

One route to achieve these objectives is the synthesis of advanced biofuels through thermochemical conversion of biomass-based residues. Gasification is a well-established thermochemical biomass conversion technology. Yet, its primary use is the production of heat and electricity, whereas industrial scale gasifiers for the synthesis of advanced biofuels are not available, hitherto [4].

Within the scope of the CLARA project, an efficient technology for the production of liquid fuels based on chemical looping gasification (CLG) of biogenic residues is being developed. The major objective is to further investigate and test CLG up to 1 MW_{th} scale in an industrially relevant environment, elevating the process to market maturity. Furthermore, the project aims at devising and optimizing innovative, cost-efficient technologies for biomass pre-treatment and syngas cleaning. These novel process steps will be supplemented by established fuel synthesis technologies (e.g. Fischer-Tropsch process), yielding the full biomass-to-biofuel process chain.

By focusing on biological non-food-grade precursors, CLARA contributes not only to a sustainable shifting from fossil to renewable resources, but also facilitates the large-scale economic production of biofuels, without detrimental effects on food availability and prices arising. This aspect, in combination with the projected advances in terms of process scalability, CO_2-reduction potential (net negative CO_2 emissions) and projected biofuel costs of 0.7 €/l, make the process investigated within the scope of CLARA an auspicious candidate for a key industry of the 21st century.

3 Chemical Looping Gasification Technology Development

During the previous year of the project, *CTH* and *CSIC* investigated the performance of the selected oxygen carriers after initial screening, ilmenite and LD slag, in the 10 kWth at *CTH* [5]–[7], and 1.5 kWth [8], [9] and 50 kWth [10], [11]at *CSIC*, in order to select the oxygen carrier to be used at higher scale in the 1 MW unit at *TUDA* [12].

To create the best possible conditions for technology evaluation, it is vital to combine experimental tests with theoretical studies. Modelling is a useful tool to cover the multiple phenomena happening during continuous conversion of solid biomass-based feedstocks to a high-quality syngas. *CSIC* developed a 1.5D fluidized bed model to simulate a high number of conditions in a relatively short period of time with low computing effort, but at the same time with the required complexity to consider the main processes affecting to the reaction of the biomass and the oxygen carrier, such as reactor fluid dynamics and the reaction pathway of biomass in the fuel reactor. The model considers the redox kinetics of the oxygen carrier, ilmenite in this case, and the fuel devolatilization and gasification kinetics previously determined both the intrinsic one and in the form of pellets as it is really used at high scale.

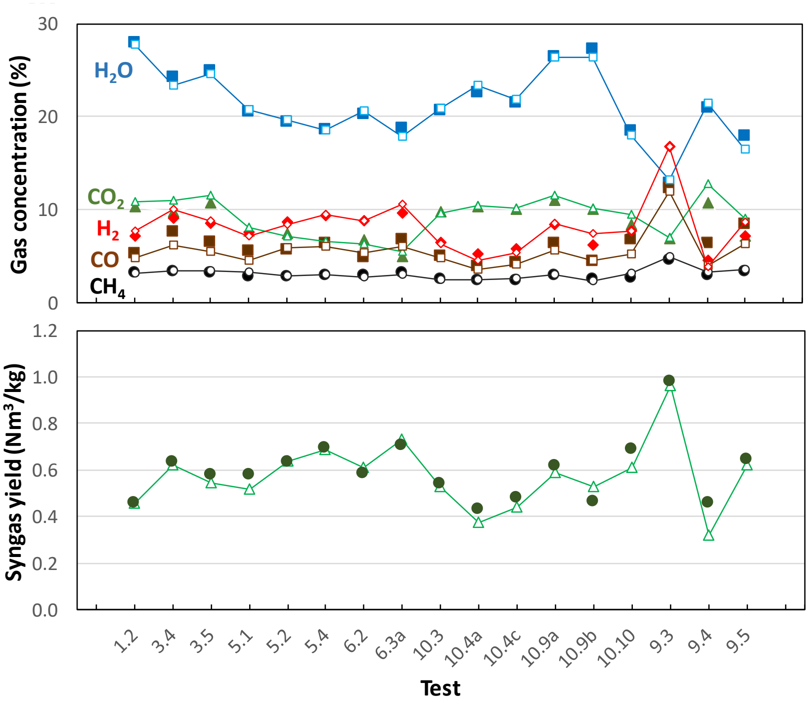


Figure 2: Gas concentration at the fuel reactor exit and syngas yield for tests performed at different operating conditions in the 50 kW CLG unit at CSIC. Open symbols: experimental results; Closed symbols: model predictions.

The model was validated against the data obtained during the experimental campaign (85 h of continuous operation) in the 50 kW CLG unit located at CSIC [10], [11] with good agreement between model and experimental results as depicted in Figure 2. Simulations done with the validated model were used to optimize the design and operating conditions of the Chemical Looping Gasification process, and more specifically to optimize the performance of the 200 MW_{th} chemical looping gasifier designed by *AE*. Results for the cold gas efficiency η_g, synas yield Y_{syngas}, char conversion X_{char}, and gas concentrations with temperature are visible in Figure 3.

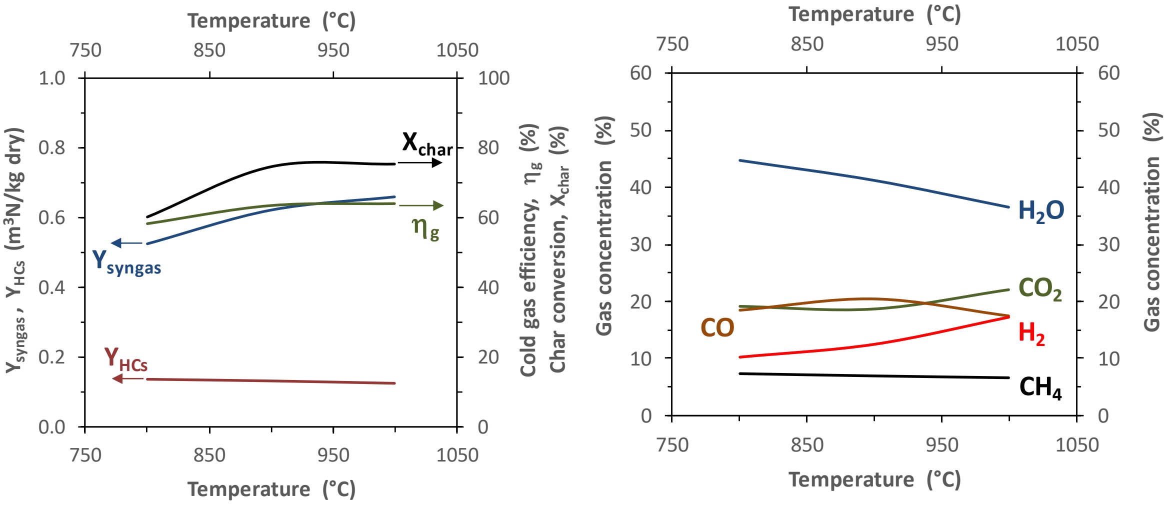


Figure 3: Effect of FR temperature on the gasification efficiency parameters, and gas composition.

4 Full-Chain Pilot Testing

The full chain pilot tests were started by *TUDA* with the hot commissioning of the chemical loping gasifier in Darmstadt [12] in March 2022. During three test campaigns between March and August 2022, more than 150 t of pelletized biogenic feedstock were gasified in the pilot plant. Generated syngas was treated in the gas-cleaning pilot plant and subsequently supplied to the Fischer-Tropsh synthesis test-rig to test synthesis with real syngas. Part of the separated sour gas was routed to the gas-washing test-rig to test the innovative Sulphur recovery with real sour gas. During the first test campaign, the focus was the commissioning of the newly erected equipment and first autothermal CLG pilot testing using commercially available wood pellets conforming to norm ENPlus A1. The second [13] and third test campaigns used pine forest residue (PFR) pellets and wheat straw pellets (WSP), respectively, to investigate more difficult residual feedstocks. Moreover, different particle size distributions of the oxygen carrier (OC) material ilmenite were investigated.

Figure 4: Overview over the 1 MW_{th} pilot plant at TUDA, consisting of the Chemical Looping Gasifier, Gas Treatment Pilot Plant, Gas Treatment Test Rig, and Fuel Synthesis Test Rig.

During the pilot tests, a new process control concept [14], developed in small pilot scale, was successfully tested allowing for efficient process control for the autothermal operation of the CLG process during all test campaigns. Here, significant insight into the intricate interaction between the CLG process and the deployed OC material was gained from the analysis of over 200 offline samples collected during the pilot tests.

The biogenic residue feedstocks were produced by *ABT* (Figure 5), during which it was discovered that the storage of raw, harvested wheat straw has a significant influence on the processing. As such a second batch of straw had to be incorporated to the pellet production as the first batch exhibited high loss of fines during processing. Pre-treatment in the form of additives according to the recipe developed by *CENER* were added to the wheat straw pellets. ABT produced over 80 t of WSP and 72 t of PFR pellets for the pilot tests.

The pilot test with PFR were hugely successful with over 170 h of stable CLG operation even surpassing the successful test of the first test campaign utilizing IWP. This showed that even difficult, or “residual”, biomasses can be utilized in the CLG process for the production of a high calorific syngas. During the pilot tests with the WSP some agglomeration of the bed material was observed in the bottom product material removed from the reactor and the samples taken from the loop seals (Figure 6). After shutdown of the pilot plant samples of the agglomerates, the OC material and the WSP were sent to *UNIVAQ* and *FZJ* to analyze the cause of the agglomerates. It was discovered, that the second batch of wheat straw used for the pellet production had some significant deviations in ash composition and thus ash melting behavior. Therefore, the additives added during feedstock production were insufficient for this batch to prevent agglomeration to occur. However, as this can be predicted and accounted for during feedstock production [15], this is not a “show-stopper” for the usage of wheat straw in the CLG process. Therefore, pilot testing validated that the production of liquid fuels from biomass feedstock of different sources is facilitated via the suggested BtL process chain.

Figure 5: Straw pellet production. Left: fine grinded straw, left pelletized straw

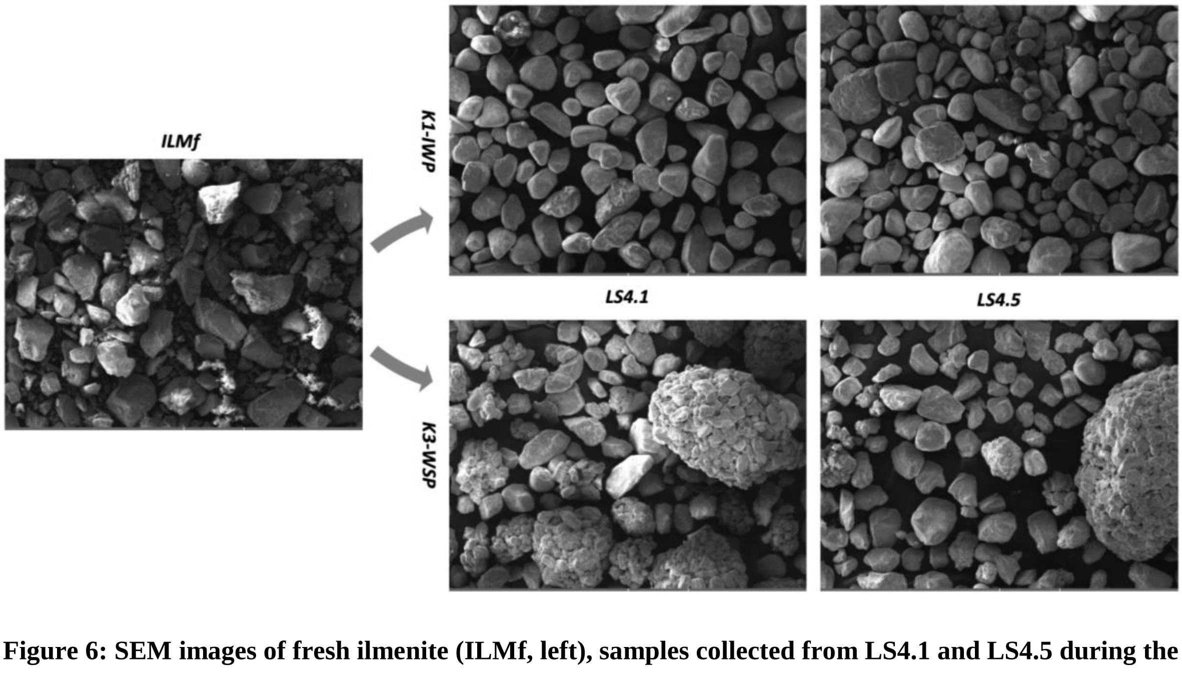


Figure 6: SEM images of fresh ilmenite (ILMf, left), samples collected from LS4.1 and LS4.5 during the wood pellets (top) and wheat straw (bottom) campaign at 70x magnification.

The gas cleaning pilot plant (Block Diagram in Figure 7) showed that successful cleaning of the syngas generated during CLG to a quality required for the FT-synthesis and the Sulphur recovery is possible using the proposed process scheme. After removal of dust and chlorine species in the initial filtration and washing steps, the steam is removed and the syngas compressed. A hydrolysis step converts COS to H_2S which can later be removed using the amine wash. Aromatic compounds are removed in the BTX absorber and finally, in the amine wash the sour gas is separated from the syngas. The change in gas composition at the individual process steps is depicted in Figure 8. The small amount of Nitrogen visible at the end of the gas treatment pilot plant is the result of instrument purges which are designed differently in a commercial unit.

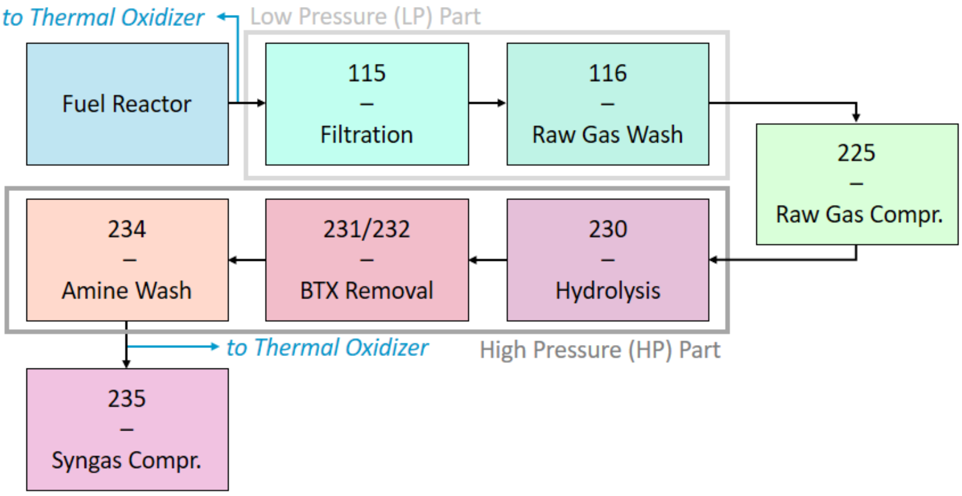


Figure 7: Block Diagram of TUDA Gas Cleaning Facility [4].

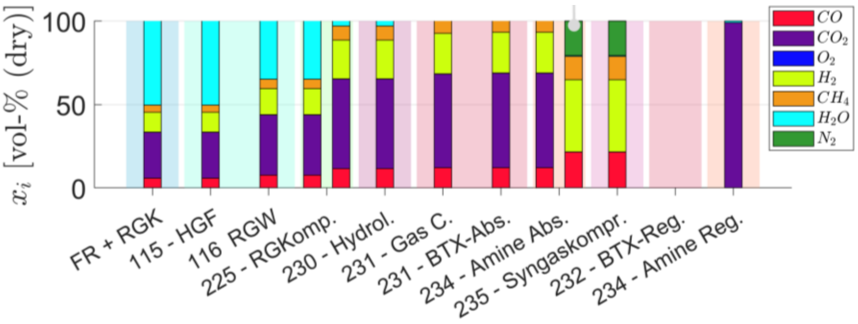


Figure 8: Syngas composition at equipment outlets in the gas cleaning pilot plant. FR: fuel reactor, RGK: raw gas cooler, HGF: hot gas filter, RGW: raw gas wasing, RGKomp: raw gas compression, Hydrol: hydrolysis, Gas C: gas cooler, Abs: absorber, Reg: regenerator.

5 Full Chain Process Modelling

The main goal of the full chain process model validation done by *CERTH* described in the following is the validation of the process model developed during the CLARA project against the experimental data generated from pilot testing at *TUDA (see Chapter 4)* to ensure consistency between the model and the real world. This evaluation procedure reinforces the validity of full-scale process simulations generated.

After the alignment of the model input parameters with the respective data of the pilot trials, interventions were performed for some assumptions (e.g. pyrolysis model) and calculation expressions (e.g. char gasification kinetics, OC reduction kinetics, water-gas shift reaction extent) of the model in order to approximate as much as possible the real behavior of the pilot tests. The objective was improvement of some model aspects to better reproduce the actual experimental findings. The data generated from the resulting model visualized in Figure 9 for the pilot tests with PFR showing good agreement of the model results with the experimentally recorded data.

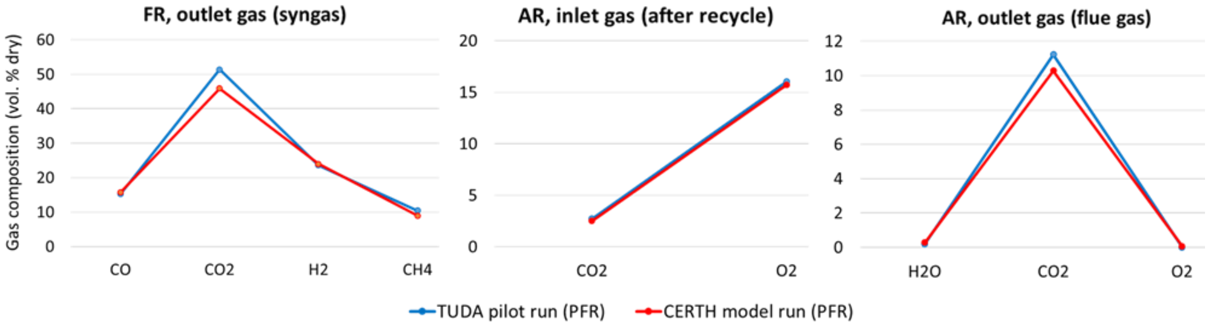


Figure 9: Validation of pilot/model CLG results for the main gas streams (PFR).

The gas cleaning pilot plant was validated as well to ensure proper alignment of the full process chain model with the experimental results obtained during pilot testing by *TUDA*. The gas composition of all major as compositions were compared and no significant deviations were found between the process simulation and the data obtained from pilot plant experiments. The reproduction of the experimental composition change over the different process steps is visualized in Figure 10.

The consistency of the model regarding some auxiliary streams from pilot testing by TUDA (e.g. fresh water in RGW, regenerated biodiesel in BTXABS, captured benzene in BTXREG, regenerated amine in AmineABS) was checked, and the complete alignment of the gas cleaning model with the experimental trial was secured. The developed FT-synthesis model is able to follow the experimental FT results in a very good manner and can be considered as a reliable tool for the prediction regarding the composition of the final products (Figure 11).

Therefore, it can be concluded that the model yielded reliable results and the data generated during the CLARA project can be used to reliable assess the process chain and associated risks. Moreover, the model can be used as a basis for upscaling and detailed engineering of demonstration plants and commercial units.

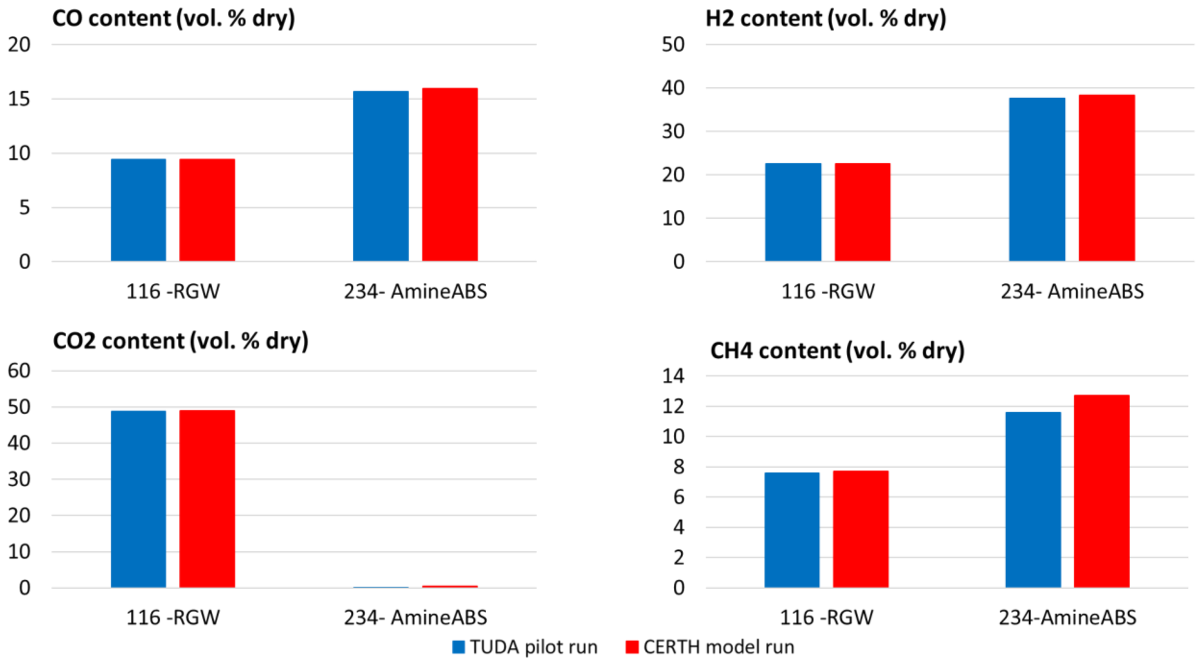


Figure 10: Validation of pilot/model gas cleaning results for the main gas species.

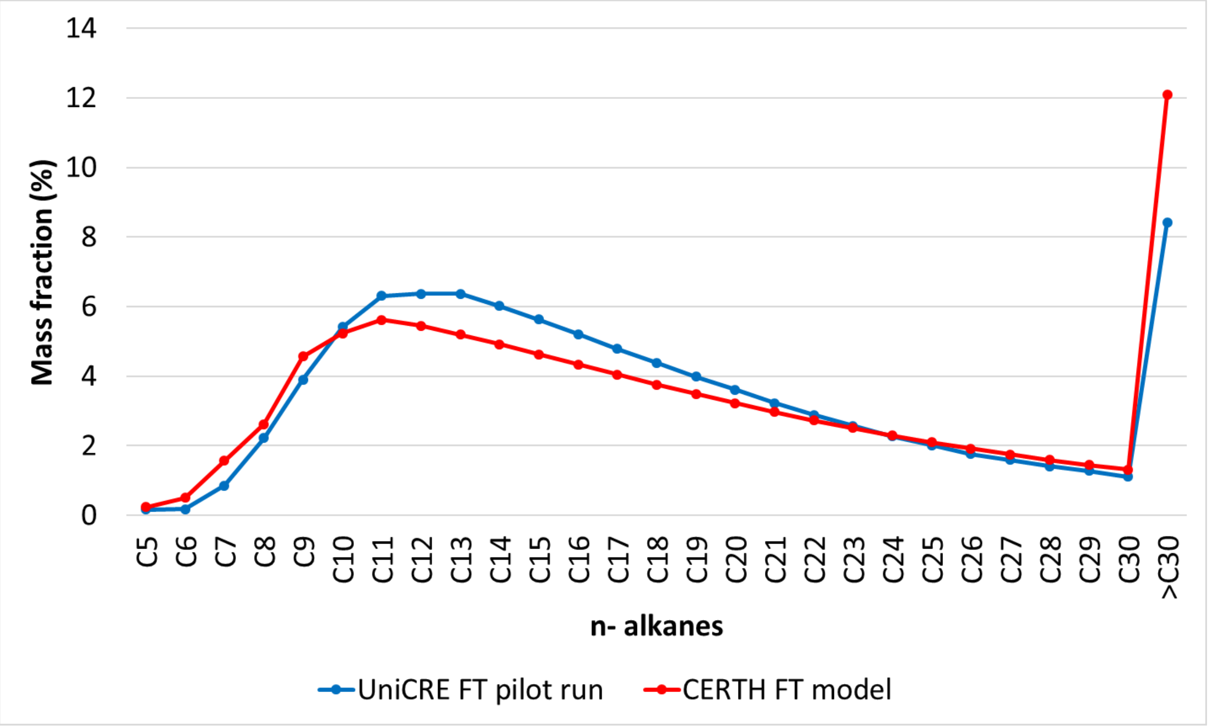


Figure 11: Validation of pilot/model FT-synthesis results.

6 Risk Assessment of the Full Process Chain

Risk assessment was carried out in order to find any possible risks which could potentially hinder the application of the CLARA process chain on a commercial scale and to identify suitable mitigation measures in case of potential risks. During risk assessment, the following aspects were investigated:

- Health and Safety
- Environment
- Society
- Technology
- Economy

6.1 Health & Safety Risk Assessment

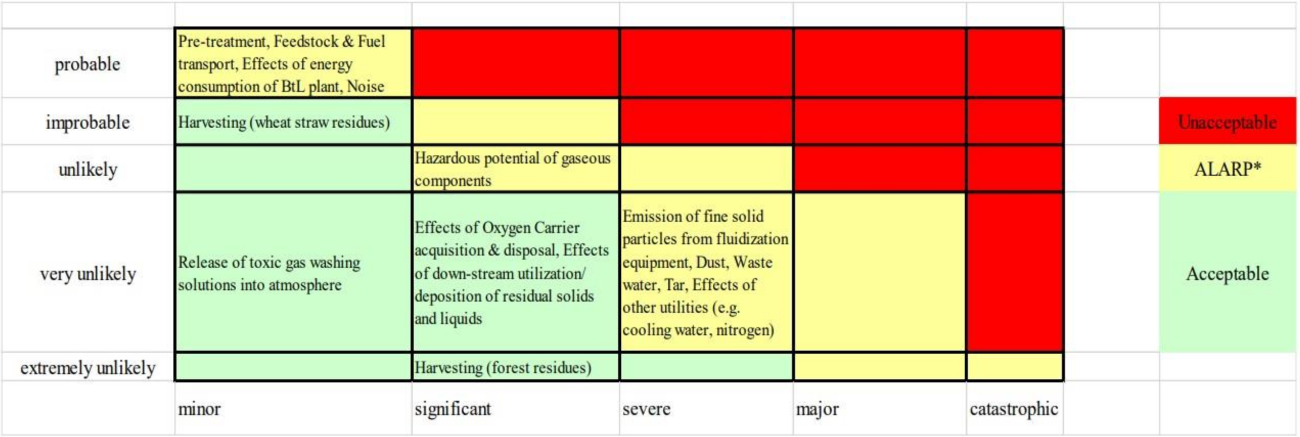
The *health and safety* issues were by *RWE* (with support from *CENER*, *TU WIEN*, *AE*, and *TUDA*) with respect to the full-scale biomass-to-end-use chain based on the processes defined at the beginning of the CLARA project. One focus is on the handled materials including solids (oxygen carrier, feedstock, dust, ash, etc.), liquids (biodiesel from gas cleaning unit, biofuel-products, solvents for gas washing, etc.) as well as gaseous components (H_2, CO, CO_2, etc.) and their hazardous potential for workers and local residents. Particular attention is drawn to the process itself and the potential safety risks in case of a malfunction of the unit and preventive measures. The partners agreed on the methodology in terms of looking section-wise into the overall process chain, identifying main risks based on typical key words. The identified risks have been evaluated qualitatively and mitigation measures are defined. As a main result no “show-stoppers” have been found, meaning that all risks can be handled, so that the plant can be operated in a safe mode comparable to other large-scale chemical plants.

6.2 Environmental Risk Assessment

TU Wien has developed a list of 14 environmental risks that were assessed by the entire project consortium, which included but were not limited to: risks related to land use and harvesting, soil preparation, soil contamination by pesticides/fertilizers, the effects of downstream utilization and/or deposition of residual solids and liquid, as well as the energy consumption of the Biomass-to-Liquid plant on the environment. In addition to the technology-specific risks, safety concerns, such as explosions, fire hazards, and operator failures have also been included in the analysis. The analysis has been carried out using a qualitative approach that was mainly based on a literature survey, as well as a series of interactive workshops.

This study came to the conclusion that no risks related to the CLARA project were found that are expected to be deemed “unacceptable”, i.e. pose a serious, irreversible threat to the environment and surrounding ecosystem. According to both the literature analysis and the interactive workshop with consortium members, most risks under study were in the acceptable range and a few, select risks were deemed to be in the “as low as reasonably practicable” (ALARP) range, i.e. classified as tolerable risks. According to the literature survey, potential environmental risks related to biomass pre-treatment, feedstock & fuel transport, effects of energy consumption of the BtL plant, and noise pollution were all risks that are highly likely but are also expected to have an insignificant effect on the environment. However, high likelihood does not equal high severity (and vice versa) and there will naturally be risks related to the pre-treatment and gasification of biomass that are unavoidable but pose a low environmental threat – thus they should not be the focus of mitigation efforts, but rather be kept to a minimum through existing best practices. Potential environmental risks related to the emissions of fine solid particles from fluidization equipment, dust, waste water, tar, and the effects of other utilities (e.g. cooling water, nitrogen) were deemed to be very unlikely but their potential effect was classified as severe. In case of these risks it is important to stress their low likelihood, given proper engineering practices and the strict implementation of EU norms. The similarity of the workshop results to the literature survey suggests that no overly pronounced resistance to such a plant may be expected from the local community.

The analyzed risks and their categorization according to likelihood and severity are summarized in Figure 12.



probable	🟨 Pre-treatment, Feedstock & Fuel transport, Effects of energy consumption of BtL plant, Noise	🟥	🟥	🟥	🟥		
improbable	🟨 Harvesting (wheat straw residues)	🟨	🟥	🟥	🟥		🟥 Unacceptable
unlikely	🟩	🟨 Hazardous potential of gaseous components	🟨	🟥	🟥		🟨 ALARP*
very unlikely	🟩 Release of toxic gas washing solutions into atmosphere	🟩 Effects of Oxygen Carrier acquisition & disposal, Effects of down-stream utilization/ deposition of residual solids and liquids	🟨 Emission of fine solid particles from fluidization equipment, Dust, Waste water, Tar, Effects of other utilities (e.g. cooling water, nitrogen)	🟨	🟥		🟩 Acceptable
extremely unlikely	🟩	🟩 Harvesting (forest residues)	🟩	🟨	🟨		
	minor	significant	severe	major	catastrophic		

Figure 12: Risk matrix depicting likelihood & severity of each risk.

6.3 Technology Risk Assessment

Technology risks were identified by *AE* (with support of *ABT*, *TUDA*, *CENER*, *RWE*) for each section of the technology chain. These risks were classified regarding likelihood and severity and a product of both was calculated to get an overall risk factor.

Out of a maximum of risk factor 9 (= very high likelihood and very high severity), only one risk with the risk factor of 6 occurred concerning the Sulphur poisoning of the shift catalyst where multiple mitigation options are available to reduce the risk to lower levels. All other risks had a factor of 4 or less. Overall, technological risks are not thus deemed similar as for other 2nd generation biofuel production routes.

Suitable mitigation actions were proposed for each risk. Some process steps have a lower TRL (5 or 6). Here additional research actions can reduce the technology risks in future plants. For other process steps with higher TRL suitable specification of equipment and good quality control during delivery and erection can reduce the risks.

6.4 Economic Risk Assessment

The aim of the economic risk assessment performed by *ULSTER* is to determine the likelihood of loss on the investment, helping investors to understand the economic feasibility of the plant, the main factors affecting it, and possible mitigation strategies. The assessment was carried out using Monte Carlo simulations.

The main types of economic risks within the present assessment:

- Risk on the project capital cost estimating
- Risk on Net Present Value and Internal Rate of Return coming from uncertainties in:
    - Supply cost for feedstock
    - Fisher-Tropsch products selling price (i.e. naphtha, biodiesel, and wax)
    - Oxygen carrier cost
    - Purchasing electricity price
    - Carbon tax/credits price

The results from the investigation of the economic risk in the chemical looping gasification show, that to have a 95% probability of not overrunning the budget estimated, a reserve (contingency budget) equal to 14% of the budget is needed.

Regarding the risk on the NPV and IRR, for both the PFR and the WS, the results found that without the use of CCS, which allows for selling carbon credits, there is a high risk (higher than 40%) of having a negative NPV or an IRR below the discount rate, making the investment not feasible. As possible mitigation strategies for the specific case, investors should look at long-term bilateral contracts to fix the price of wax and biodiesel, which are responsible for more than 90% of the variation of the NPV, and to identify a strategy for utilizing the carbon captured that is ready to be used. The addition of CCS allows a critical reduction of the economic risk to values below 15%. Moreover, selling the excess heat produced by the plant provides extra revenues that can further reduce the risk of negative NPV for the PFR and WS to values below 10%. If hydro-processing to produce biodiesel from FT-products is included in the investment, the risk increases due to an increase in the investment cost and a reduction in the volume of final products but using CCS and the possibility of selling heat guarantee to keep the risk of negative NPV and IRR below 6% at values below 20%, making the investment attracting.

7 Techno-Economic and Life Cycle Assessment

Based on the full scale layout developed a techno-economic and life cycle analysis has been carried out in order to assess the viability for investment and the required selling prices of the generated bio-fuels as well as the overall carbon emission of the produced bio-fuels. The evaluation focused on decentralized feedstock pre-treatment facilities with a centralized CLG system with a thermal input of 200 MW_{th} and subsequent gas treatment and FT-Synthesis.

7.1 Techno-Economics Analysis

As cost base year 2020 was selected for a 25 year project lifetime. For the mass and energy balances and the equipment sizes established in the project a total capital investment of approx. 254 million € was estimated with annual operating and maintenance cost of around 68 million € for the PFR case and around 59 million € for the WSP case. Based on this, a brake even selling price (BESP) of the raw FT-crude produced of 816 €/t and 781 €/t were calculated, respectively.

It was further analyzed that the employed MDEA based case cleaning leads to a lower BESP than what could be achieved in a currently state-of-the-art Rectisol plant. The BESP can be decreased further if waste heat can be marketed and if the CO_2 is further stored with selling of carbon credits as can be seen in Figure 13 for the WS case. Moreover, the analysis showed, that integrating a hydro processing unit into the plant would increase the price of final bio-fuel products by 34.9 % to 1.06 €/l. Therefore, the suggested option of further processing the produced FT-crude in existing refineries also poses an advantage from an economic perspective.

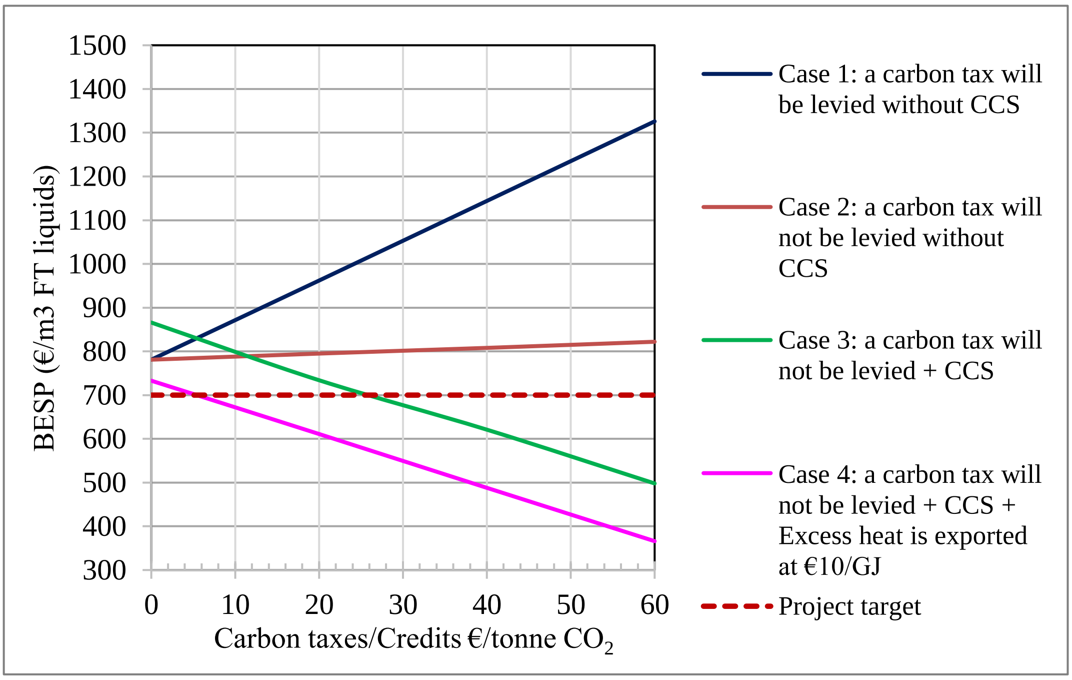


Figure 13: Impact of CO_2 tax/credits on BESP of FT fuels for the WS scenario.

7.2 Life Cycle Analysis

The goal of the life cycle analysis for the BtL process chain is to assess the greenhouse gas emission, environmental impacts, resource consumption, and efficiency while comparing the different feedstocks.

The life cycle analysis was carried out according to the norms ISO 14040:2006, ISO 14044:2006, and ISO 14025:2006 and was following the Product Environmental Footprint (PEF) Guide. During analysis the fuel production activity was investigated for a plant location in Germany utilizing the local electricity mix with a cradle to gate approach including the following stages:

- Pretreatment
- Chemical Looping Gasification (CLG)
- Gas Cleaning (GC)
- Fuel Synthesis (FS)
- Carbon Capture and Storage (CCS)

The two indicators analyzed in detail are climate change and fossil energy usage. It was found that electricity is the main driver for carbon dioxide emission and climate change potential in the production of bio fuels using the CLARA BtL process chain. As such the local electricity mix has an impact on the climate change potential, i.e. the results will differ with the location of a BtL plant. The resulting CO_2 emissions obtained for the production of 1 t of liquid FT crude product are negative and are visualized in Figure 14 for the three feedstocks tested in CLARA and show a clear negative climate change impact. For the output of the proposed 200 MW_{th} BtL plant this results in a negative climate change potential of approx. 130 million tons per year.

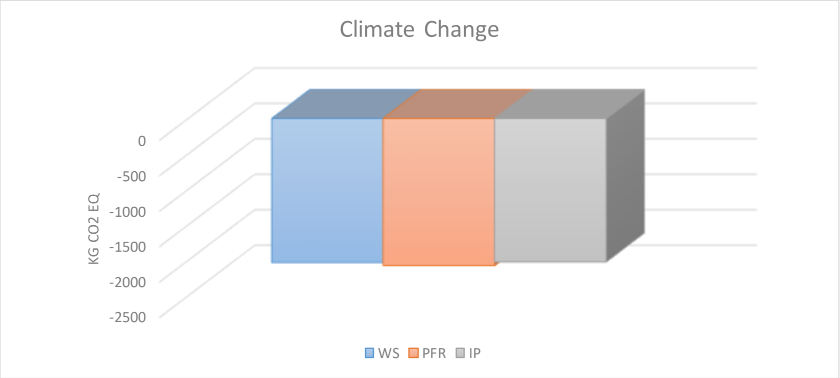


Figure 14: Comparison of kg $CO2_{eq}$ per ton of FT crude by Feedstock.

8 Summary and Conclusions

The major advances and most important findings made within the fourth year of the CLARA project with regard to technology development (see Chapter 3), full chain pilot testing (see Chapter 4), full chain process modeling (see Chapter 5) and the investigation of the BtL chain risks (see Chapter 6) are summarized below.

- The insights from small-scale laboratory investigations in the CLG process were used to optimize an existing 1.5D process model. This model was then used to predict the operation of a 200 MW gasification plant and the subsequent sizing of the corresponding gasifier by AE.
- The entire CLARA process chain was demonstrated at Darmstadt during three full chain pilot test campaigns. More than 100 t of biomass pellets were converted to syngas in the 1 MW chemical looping gasifier using a novel process control concept developed by TUDA. Over 150 t of biogenic residue feedstocks used during pilot testing were successfully produced by *ABT* in preparation of the experiments with methods devised by *CENER*. The generated syngas was successfully cleaned in the gas cleaning pilot plant at *TUDA* and the purified syngas fed to the innovative sour gas cleaning test rig from *RWE* and the FT test rig for further processing.
- The data generated during pilot testing was used by *CERTH* to validate the full chain process model used for the modeling of the full process chain. The model showed good agreement with the CLG unit, the gas cleaning pilot plant, and the FT-reactor.
- The output of full scale modeling and the corresponding layout was used for analysis and assessment of the full-scale BtL process chain. No severe risks making the CLARA process chain infeasible were identified
- Techno-economic analysis showed that the BESP of the produced FT products is starkly dependent on carbon tax/credit prices.
- Life cycle assessment showed a clear negative climate change potential of approx. 130 million tons CO_2 equivalent per year for the suggested BtL plant.

The CLARA consortium was able to demonstrate the technological feasibility of the whole process chain by full chain pilot testing. The results were used for the definition and optimization of process sub-units scaled at industrial size which formed the basis for the final assessment of the full BtL process chain. Here, no “show stoppers” could be found and the economic assessment showed investment in the process chain to be an interesting economic endeavor.

Summarizing the whole project, the entire CLG-BtL process chain was investigated during the CLARA project. Starting from technology development for CLG and the fine cleaning of the sour gas, to upscaling the whole process chain, the feasibility of the process chain was demonstrated and the TRL of relevant technologies advanced. Especially the CLG process was demonstrated for the first time under autothermal conditions during the project. Various assessments of the Technology and the final product have been carried out in parallel to investigate risks, climate change potential, and economic viability. All results suggest that a demonstration plant should be the next step in developing the technology towards market maturity.

9 Disclaimer

10 References

[1] International Energy Agency, *Key World Energy Statistics 2018*. in Key World Energy Statistics. OECD, 2018. doi: 10.1787/key_energ_stat-2018-en.

[2] “Transport emissions - A European Strategy for low-emission mobility,” *Transport emissions*. https://ec.europa.eu/clima/policies/transport_en (accessed Aug. 23, 2019).

[3] “DIRECTIVE (EU) 2018/ 2001 OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL - of 11 December 2018 - on the promotion of the use of energy from renewable sources,” p. 128.

[4] J. E. Carrasco, A. Monti, J. Tayeb, J. Kiel, and F. Girio, “Strategic Research and Innovation Agenda 2020,” EERA. Accessed: Jun. 01, 2019. [Online]. Available: http://www.eera-bioenergy.eu/publications/#sria-2020

[5] F. Hildor, H. Leion, and T. Mattisson, “Steel Converter Slag as an Oxygen Carrier—Interaction with Sulfur Dioxide,” *Energies*, vol. 15, no. 16, p. 5922, Aug. 2022, doi: 10.3390/en15165922.

[6] F. Hildor, A. H. Soleimanisalim, M. Seemann, T. Mattisson, and H. Leion, “Tar characteristics generated from a 10 kWth chemical-looping biomass gasifier using steel converter slag as an oxygen carrier,” *Fuel*, vol. 331, p. 125770, Jan. 2023, doi: 10.1016/j.fuel.2022.125770.

[7] I. Gogolev, A. H. Soleimanisalim, D. Mei, and A. Lyngfelt, “Effects of Temperature, Operation Mode, and Steam Concentration on Alkali Release in Chemical Looping Conversion of Biomass─Experimental Investigation in a 10 kW th Pilot,” *Energy Fuels*, vol. 36, no. 17, pp. 9551–9570, Sep. 2022, doi: 10.1021/acs.energyfuels.1c04353.

[8] O. Condori, F. García-Labiano, L. F. de Diego, M. T. Izquierdo, A. Abad, and J. Adánez, “Biomass chemical looping gasification for syngas production using ilmenite as oxygen carrier in a 1.5 kWth unit,” *Chem. Eng. J.*, vol. 405, p. 126679, Feb. 2021, doi: 10.1016/j.cej.2020.126679.

[9] O. Condori, F. García-Labiano, L. F. de Diego, M. T. Izquierdo, A. Abad, and J. Adánez, “Biomass chemical looping gasification for syngas production using LD Slag as oxygen carrier in a 1.5 kWth unit,” *Fuel Process. Technol.*, vol. 222, p. 106963, Nov. 2021, doi: 10.1016/j.fuproc.2021.106963.

[10] O. Condori, F. García-Labiano, L. F. de Diego, M. T. Izquierdo, A. Abad, and J. Adánez, “Syngas production via Biomass Chemical Looping Gasification (BCLG) in a 50 kWth unit using ilmenite as oxygen carrier,” *Proc. Fluid. Bed Convers. Conf. 2022*, p. 10, 2022.

[11] O. Condori, A. Abad, M. T. Izquierdo, L. F. de Diego, F. García-Labiano, and J. Adánez, “Assessment of the chemical looping gasification of wheat straw pellets at the 20 kWth scale,” *Fuel*, vol. 344, p. 128059, Jul. 2023, doi: 10.1016/j.fuel.2023.128059.

[12] F. Marx, P. Dieringer, J. Ströhle, and B. Epple, “Design of a 1 MWth Pilot Plant for Chemical Looping Gasification of Biogenic Residues,” *Energies*, vol. 14, no. 9, p. 2581, Apr. 2021, doi: 10.3390/en14092581.

[13] F. Marx, P. Dieringer, J. Ströhle, and B. Epple, “Solid flux measurement in dual fluidized bed processes based on solid samples,” *Fuel*, vol. 341, p. 127589, Jun. 2023, doi: 10.1016/j.fuel.2023.127589.

[14] P. Dieringer, F. Marx, F. Alobaid, J. Ströhle, and B. Epple, “Process Control Strategies in Chemical Looping Gasification—A Novel Process for the Production of Biofuels Allowing for Net Negative CO2 Emissions,” *Appl. Sci.*, vol. 10, no. 12, p. 26, Jun. 2020, doi: 10.3390/app10124271.

[15] F. Lebendig and M. Müller, “Effect of pre-treatment of herbaceous feedstocks on behavior of inorganic constituents under chemical looping gasification (CLG) conditions,” *Green Chem.*, vol. 24, no. 24, pp. 9643–9658, 2022, doi: 10.1039/D2GC02906E.

Abbreviations

ALARP	As Low As Reasonable Possible
BESP	Brake Even Selling Point
BtL	Biomass to Liquid
BTX	Benzene, Toluene, Xylene
CCS	Carbon Capture and Storage
CLG	Chemical Looping Gasification
FT	Fischer-Tropsch
ILM	Ilmenite
IRR	Internal Rate of Return
IWP	Industrial Wood Pellets
MDEA	Methyl diethanolamine
NPV	Net Present Value
OC	Oxygen Carrier
PFR	Pine Forest Residue
REDII	Renewable Energy Directive
TRL	Technology Readiness Level
WSP	Wheat Straw Pellets

Project Consortium

TUDA	Technische Universität Darmstadt	*TU WIEN*	Vienna University of Technology
AE	AICHERNIG Engineering GmbH	*FJZ*	Forschungszentrum Jülich
CTH	Chalmers Tekniska Högskola AB	*ABT*	AB Torkapparater
CSIC	Agencia Estatal Consejo Superior de Investigaciones Cientificas	*ULster*	University of Ulster
RWE	RWE Power AG	*CERTH*	Centre for Research & Technology Hellas
CENER	Centro Nacional de Energías Renovables	*UniCRE*	Unipetrol Centre for Research and Education, a.s.
UNIVAQ	University of L'Aquila