Identifying the ideal process configuration for a green methanol production plant dependent on economic boundary conditions

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Abstract:

The production of second-generation biofuels from agricultural waste products represent a sustainable option for future fuel and chemical industry. Yet, due to the local biomass availability, the identification of process designs dependent on the site-specific boundary conditions is mandatory to assess their potential contribution to a sustainable transition. While biomass-based routes have the advantage of mature technologies, the combination with renewably-generated hydrogen provides the opportunity to significantly increase the extent of carbon utilisation. Via dual fluidized bed gasification, gas cleaning, and a subsequent methanol synthesis and purification, grade AA methanol can be synthesised. With its ability of a direct CO_2-conversion at a high selectivity towards methanol, the methanol synthesis constitutes a promising option to chemically store fluctuating renewable energy. The potential of a biomass-to-liquid concept is analysed incorporating economic constraints into the process design to allow the identification of regionally adjusted process designs. The production costs of the concepts are estimated by setting up a detailed flowsheet simulation in AspenPlus®. A techno-economic evaluation methodology has been extended by an automated utility integration to identify European sweet spots. The conducted evaluation includes two different process configurations for two different biogenic feedstocks, including the integration of hydrogen produced through renewable sources. A correlation between economic boundary conditions such as the electricity and heat market, and the process design will be presented. Finally, the potential contribution of the investigated (power&)biomass-to-liquid process to a green methanol economy is discussed.

Keywords:

Techno-economic analysis, Green methanol, Second-generation biofuels, Process design

1. Introduction

The Intergovernmental Panel on Climate Change (IPCC) is once again urging the energy, transport, and agriculture sectors to take decisive action to achieve the Paris climate targets of reducing greenhouse gas emissions by 80 to 95 % by 2050 compared to 2010 [1]. To promote the use of renewable energy sources in the transport sector, the European Union has adopted the Renewable Energy Directive (RED) in 2009 [2], and its 2018 revision (RED II) [3], which require EU member states to achieve minimum shares of alternative and renewable energy sources in the transport sector (RED: 10 % by 2020, RED II: 14 % by 2030). The Fuel Quality Directive (FQD) [2] is another instrument used to monitor energy carriers in the transport sector, which aims to reduce the lifecycle greenhouse gases of fuels and defines greenhouse gas reduction targets for fuels placed on the market [2]. Fuel suppliers are obligated to report GHG emissions for the fuels they place on the market. As a result, there is an increased interest in renewable alternative fuels that can be produced from renewable power sources or waste materials derived from municipal, forestry, or agricultural wastes.

Methanol synthesis from biomass is a promising approach to meet the future demand for renewable fuels. Synthetic methanol can be produced through biomass-to-liquid (BtL) processes using a methanol synthesis and purification system, to achieve high methanol purity [4].

Renewable power-based methods are facing challenges in scaling up their technologies, particularly in water electrolysis and running a chemical production plant with varying feed sources [5, 6]. In contrast, biomass-based methods have a higher technology readiness level and require limited adaptations to use biomass as a feedstock, benefiting from the experience gained from coal gasification and large-scale fuel synthesis in coal-to-liquid plants like Sasol in South Africa [7-9]. However, there are also scalability concerns with biomass-to-liquid (BtL) processes due to the availability and transportation distance of biomass, which impacts overall greenhouse gas emissions and production costs. To overcome these challenges, BtL plant size can be kept small to medium or located near waste material sources. Alternatively, an efficient and ecologically friendly method to provide enough biomass is needed to benefit from economies of scale.

https://doi.org/10.52202/069564-0129

Based on the gasification and gas cleaning track described in [10] a decentralised process configuration for a biomass-to-methanol process is set up and extended by a proton exchange membrane electrolyser. For the methanol synthesis two different reactor concepts are investigated in order to analyse their performance for different boundary conditions.

In contrast to the work of Poluzzi et al. [11], two different feedstocks are investigated. Nevertheless, since the process setups are very similar to the ones modelled by Poluzzi et al. they are used as a benchmark for the conducted analyses.

Other studies looking into the biomass-to-liquid process often have Fischer-Tropsch syncrude as their product since it can easily integrated into the existing refinery structure [12, 13]. Another difference is the plant scale, since the biomass transport is a main driver for the feedstock's costs and its global warming potential, the investigated plant size is limited to 200 MW_{th} [14].

Simply evaluating pre-defined process configurations for multiple boundary conditions to find economically viable plant locations may not result in optimal designs, as there is no way to adjust the process itself. Therefore, this study aims to analyse the potential of multiple configurations under different economic boundary conditions while ensuring flexibility. This has been enabled by changed way of how TEPET accesses the AspenPlus® simulation files and to allow the modification of certain parameters through TEPET. To evaluate the process potential and its ideal configuration for achieving the lowest production costs, data was gathered on regional biomass potential, prices for straw and bark, other raw material prices, labour and transportation costs, and applicable revenues in the heat market.

2. Methodology

To evaluate the different process design a detailed flowsheet model [10] is utilized and adapted for the conducted investigation of methanol production. The process models are linked to the DLR inhouse tool TEPET [15] and its extension described in [10].

2.1. Flowsheet model

The basic flowsheet model is well described in the previous work of Maier et al. [10] which is based on the work within the EU-project COMSYN [16] and the experimental and simulative work of [17, 18]. The process schemes for the biomass-to-liquid and the power&biomass-to-liquid process can be seen in Figure 1.

The main difference is the introduction of an electrolyser into the model to provide some additional hydrogen for the methanol synthesis and to replace the air for the reformer by pure oxygen.

While the settings for the gasification and gas cleaning steps are well described by Maier et al. [10], the electrolyser, the methanol synthesis, and the purification units are described in the following chapters. The two investigated biomass types, bark and straw, differ the most in the assumed moisture content and a slight difference in the carbon, oxygen and ash contents. While bark is received with 50 wt.% moisture and its dry matter analysis results in 52.5 wt.% carbon, 38.5 wt.% oxygen and 2.6 wt.% of ash, straw is received with 10 wt.% moisture and 47.3 wt.% carbon, 41.4 wt.% oxygen and 4.7 wt.% ash. Hence, the bark is firstly led into a drying section to reduce its moisture content to 12 wt.% [10, 19]. In case of using straw as the feedstock, a certain amount of chlorine content is assumed which needs to be removed before the reformer by quenching the raw syngas before the filtration step from 730 °C to 550 °C.

Electrolyser

The PEM electrolytic cell was modelled through a black box approach with conversion factor 1 (with regard to water) and operating conditions of 40 bar and 80 °C. The electrical energy demand of 61.6 kWh per kg of H_2 was used for electrolysis corresponding to an electrical efficiency (power to HHV_{H2}) of approximately 64 % [20]. It was assumed that pure oxygen can be separated from the anode, which is utilized to operate the reformer with pure oxygen, significantly reducing the amount of inerts in the syngas. The amount of hydrogen is adjusted to achieve a H/C ratio of 3 at the entrance of the MeOH synthesis loop.

Methanol synthesis

The methanol section consists of a compression to the 90 bar operating pressure, a plug flow reactor model (RPlug) with an isothermal cooling liquid at 240 °C. The reactor model is representing a tube-and-shell reactor concept as it is commonly used for large methanol production plants. The entering temperature is set to 235 °C. The pressure drop is calculated using the Ergun equation [21]. A recycle with a purge rate of 5 % ensures that impurities such as nitrogen are not accumulating within the recycle loop. For the reaction modelling the simplified kinetic model from Van-Dal and Bouallou [22] of the in-depth model of Vanden Bussche and Froment [23] with the readjusted parameters from Mignard and Pritchard [24] is implemented. The whole section is modelled with the IDEAL property method as suggested by Graaf et al. [25].

Methanol purification

For the purification a two-column approach is applied. While the first column divides the gaseous side-products from the methanol, the second column separates the methanol from the water. The methanol is purified until

https://doi.org/10.52202/069564-0129

a purity of grade AA (99.85 wt.%) is achieved. The methanol purification section is modelled using ELECNRTL to achieve a better description of the azeotropic methanol-water separation.

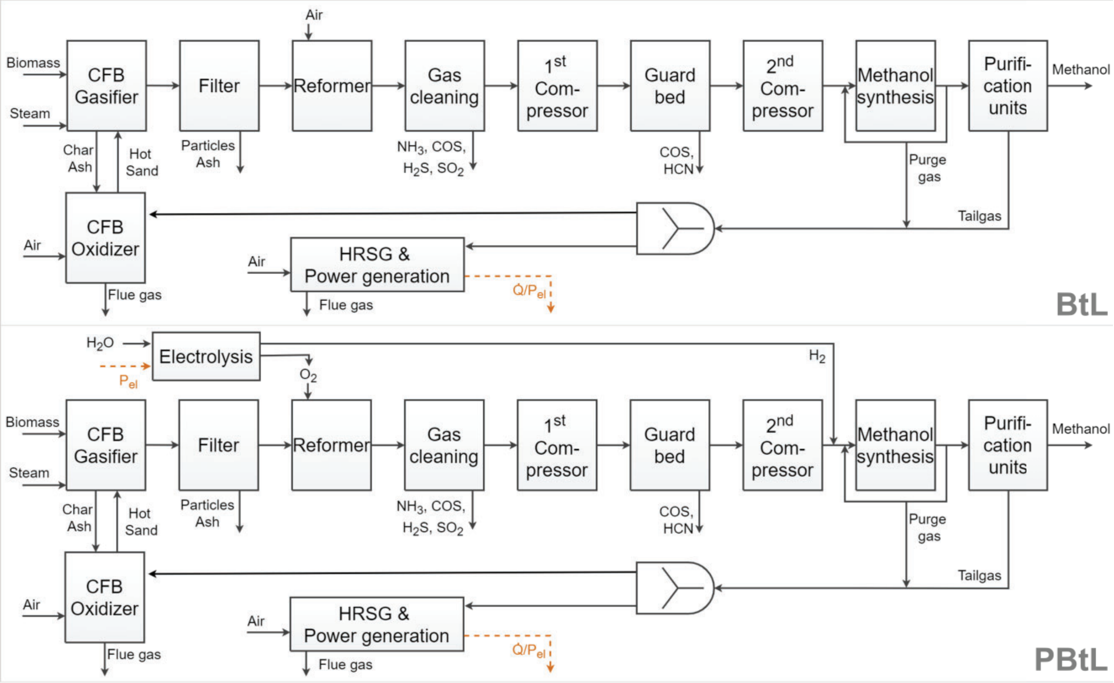

Figure 1. Process schemes for the biomass-to-liquid (BtL) and the power&biomass-to-liquid (PBtL) process.

2.2. Technical analysis

The process configurations are evaluated according to their ability to convert the stored energy of the biomass into the desired product methanol. To compare the different setups according to this performance indicator, the biomass-to-liquid efficiency is defined as follows:

Biomass-to-liquid efficiency: $\eta_{PBtL} = \frac{\dot{m}_{CH_3OH} \cdot LHV_{CH_3OH}}{\dot{m}_{biomass}\, LHV_{biomass} + P_{el}}$

2.3. Techno-economic analysis

The economic evaluation of the concept follows the approach of Peters, Timmerhaus, and West [26], which has been utilized in several previous techno-economic studies [15, 27, 28]. To apply this methodology, a database of reference equipment costs is needed to account for changes in the process configuration and its impact on capital and operational expenses. The AspenPlus® process model previously described is used to scale equipment sizes, as well as raw material and by-product streams, based on the material and energy flows. For the main equipment the cost functions of Maier et al. [10] are applied to the current biomass-to-methanol process concepts. The formulas utilized to calculate the production costs are listed below [15]:

- Equipment costs: $EC_i = EC_{ref,i} \cdot \left(\frac{D_i}{D_{ref,i}}\right)^{d_i} \cdot \left(\frac{CEPCI_{2020}}{CEPCI_{ref}}\right)$
- Fixed capital investment: $FCI = \sum_{i=1}^{m} EC_i \cdot \left(1 + \sum_{j=1}^{12} F_{eco,i,j}\right),\ TCI = \frac{FCI}{0.9}$
- Annualized capital costs: $ACC = FCI \cdot IR \cdot \left(\frac{(1+IR)^{PL}}{(1+IR)^{PL}-1} + \frac{1}{1-0.1} - 1\right)$
- Net production costs: $NPC = \frac{ACC + \sum OPEX_{ind} + OPEX_{dir} + N_P \cdot c_{labour}}{\dot{m}_{fuel}}$

The equipment costs are calculated by setting the actual unit capacity *D* into relation with a reference equipment capacity D_{ref} and its reference costs EC_{ref} and applying a defined digression factor *d* to it. To account for price differences between the reference and the base year the Chemical Engineering Plant Costs Index (CEPCI) is applied to the equipment costs. After including the Lang factors [26] for additional costs such as piping, engineering etc. the fixed capital investment (FCI) is estimated. To calculate the final net production costs, the direct and indirect operational expenditures (OPEX) are sum up together with the annualized capital costs and the labour costs. Finally, the whole yearly expenses are divided by the product output per year.

3. Results and discussion

Assessing the different process concepts regarding their biomass-to-methanol efficiency, it becomes clear that the power&biomass-based process routes yield in a higher energetic efficiency (Table 1). By enabling additional product formation, more biogenic carbon is converted to methanol, which increases the overall process efficiency. Furthermore, it can be seen, that the choice of the biomass feedstock has a significant impact on the technical potential of the process. Due to the higher ash content and higher oxygen content in the straw, the resulting syngas has a lower potential [10], since more hydrogen is required to remove the oxygen in form of water out of the system. Furthermore, due to the potential chlorine in the straw, the particle filter after the gasifier has to be operated at 550 °C which leads to further energy losses, since part of the reaction enthalpy in the autothermal reformer is used to heat the syngas up to 850 °C again. The disadvantageous composition is also the reason for the additional electricity demand in case of the straw-based PBtL concept, since more hydrogen is required to achieve the desired H/C ratio for the MeOH synthesis of 3.

Table 1. Technical results for the BtL and PBtL concepts with bark and straw as their feedstock

		BtL-B	PBtL-B	BtL-S	PBtL-S
Biomass moisture content	wt.%	50	50	12	12
Biomass input (mass flow)	kg/h	39.1	39.1	24.4	24.4
Biomass input (energy content)	MW_{th}	100	100	100	100
Electricity consumption	MW_{el}	14.6	103.9	15.8	121.4
Product output (mass flow)	kg/h	10,210	18,604	8,478	18,532
Product output (energy content)	MW_{th}	64.9	118.3	53.9	117.9
PBtL-efficiency	%	56.7	58.0	46.6	53.2

Compared to previous studies such as Peduzzi et al [4] the proposed biomass-to-liquid concept presents an 5% higher BtL efficiency for the bark case. The calculated efficiency for the bark-based PBtL case are in very good alignment with the work of [11], who estimated an efficiency of 57.5% for their process setup. Yet, the conducted analyses of this work highlight the potential of a power-enhanced setup for other biomass feedstocks. While for bark the PBtL efficiency has been increased by only 1.3%, the efficiency for converting straw into methanol can be increased by 6.6% which shows that especially for biogenic waste materials with a disadvantageous composition, the implementation of additional hydrogen generation is highly beneficial for its conversion rate.

To investigate the different concepts economically, the net production costs (NPC) are calculated (Figure 2) with the economic boundary conditions given in Table 2. With Germany as the selected region the biomass-to-methanol plant's costs are calculated for a country with relatively high raw material prices compared to other European regions such as Poland, Czech Republic or Slovenia [10].

Table 2. Economic boundary conditions.

Base year	-	2020	
Plant size	MW_{th}	100	
Interest rate (IR)	%	10	
Persons per shift (N_P)	-	10	
Full load hours (flh)	h/a	8260	
Plant lifetime (PL)	a	20	
Plant location	Germany		
Electricity costs/revenue (c_{EL})	€/MWh_{el}	106.5	[29]
Biomass costs ($c_{Bio,b}$)	€/GJ	8.4	[30]
District heating revenue (r_{DH})	€/MWh	21.0	[31]
Process steam revenue (r_{PS})	€/MWh	36.0	[31]
Labor costs (c_L)	€/h	29.0	[32]

Due to the process configuration, which contains an internal recycle within the methanol synthesis but no further re-activation of the process' tailgas, the process yields in a high amount of excess heat. Its utilisation is very dependent on the given boundary conditions. In the conducted techno-economic analysis, it is assumed that high pressure steam, as well as district heating can be provided. Hence, the methanol production costs can be decreased by achieving a quite high by-product revenue on the heat market. While the prices for

https://doi.org/10.52202/069564-0129

electricity and biomass are extracted from literature, the revenue for potential heat market products is calculated based on the assumption that they have to compete with a provision based on biomass- or natural-gas fuelled boilers. Therefore, the costs are calculated according to Ulrich et al. [31], where the price for the fuels is assumed to be the average between the local natural gas price and the biomass price [10].

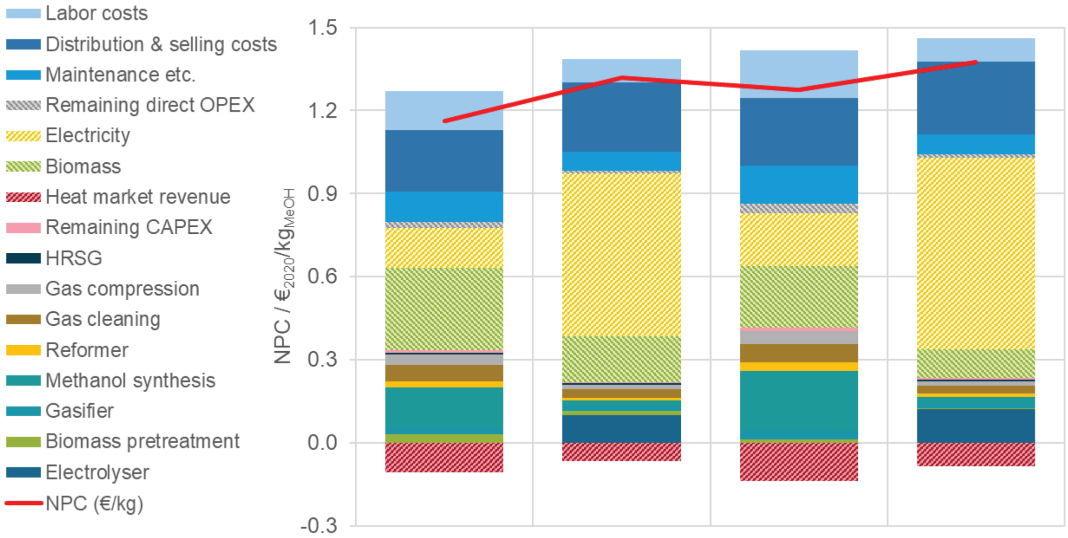

Figure 2. Net production costs for straw and bark for the two different process concepts biomass/power&biomass-to-liquid

In contrast to the prior technical results, the economic analysis of the concepts shows that the BtL process is more economic than the PBtL concepts. The main reasons can be found in the high additional expenses for the required electrical power and the additional investment for the electrolyser. The estimated NPC for all concepts differs within a range of 0.22 €$_{2020}$/kg$_{\mathrm{MeOH}}$.

In order to identify at which conditions the PBtL concept might be able to compete with the BtL process, a sensitivity analysis with bark as feedstock is conducted and shown in Figure 3.

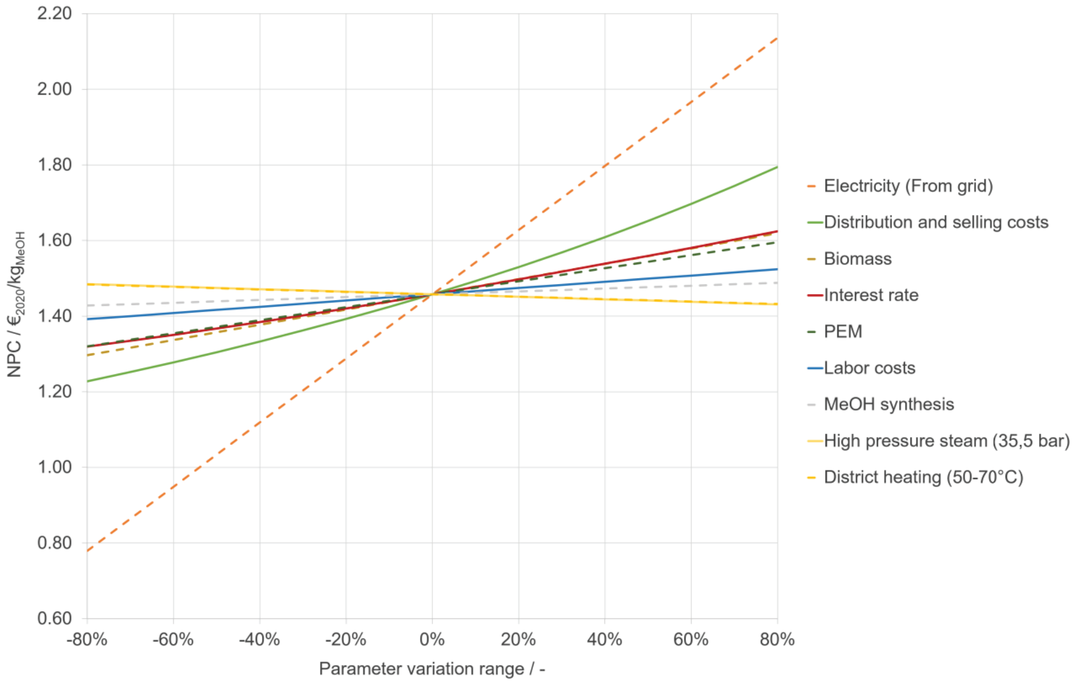

Figure 3. Sensitivity study for power&biomass-to-methanol concept using bark as feedstock

The presented sensitivity analysis is performed by varying each of the most relevant cost drivers within a range of -80% and +80%. Since the additional costs for distribution and selling of the product is dependent on the overall production costs and the interest rate affects the annualized capital costs their variations result in non-linear impacts on the net production costs. The remaining raw materials, utilities and equipment costs are having a linear effect on the calculated net productions.

In order to identify at which boundary conditions which concept is the favourable a comprehensive case study of the raw materials is conducted and shown in Figure 4. In case of bark, due to the lower benefits of the power-enhanced process design lower electricity prices are required to reach the required trade-off between additional CAPEX and OPEX and the higher product output. In case of straw utilization, the switch between BtL and PBtL is between 62 and 101 $€_{2020}/MWh_e$ in dependence on the biomass prices, while it is between 47 and 85 $€_{2020}/MWh_e$ for bark.

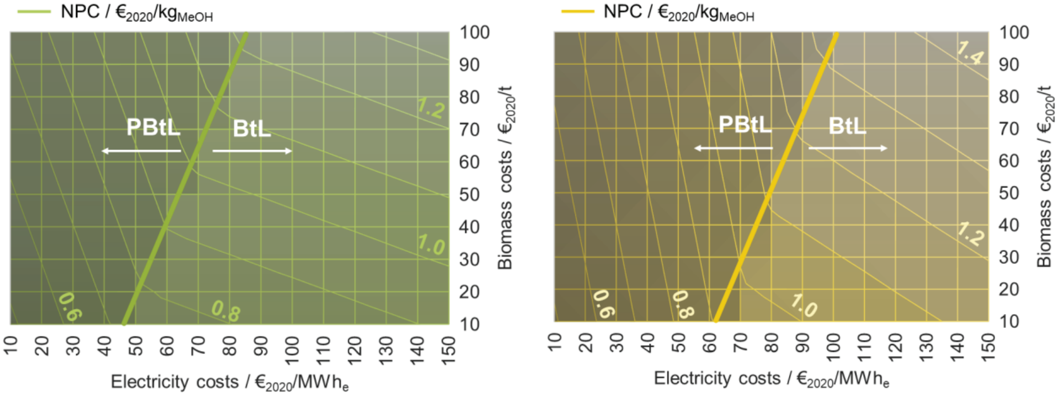

Figure 4. Production cost mapping for the two feedstocks bark (l) and straw (r) in dependence on their raw material costs

Future reduction of renewable electricity costs might favour the power-assisted biofuels production in the coming years. Furthermore, the increase of renewable energy in the electricity mix is required to achieve a positive environmental impact with the PBtL cases. Recent studies of Habermeyer et al. [27] and Weyand et al. [14] showed that utilizing the electricity from the German grid to generate the hydrogen, yields in an even higher greenhouse gas emission than using the fossil alternatives. Hence, PBtL concepts imply a potential alternative for an increased production of alternative fuels by an increased carbon efficiency but strongly depend on the availability of renewable and inexpensive electricity. Until then, only off-grid solutions may contribute to a reduction of the global warming potential or the exclusive use of biogenic waste materials as a feedstock together with a reduction of society's fuel consumption.

4. Conclusion and outlook

The study presented a promising approach for the production of sustainable methanol using two biogenetic waste materials, straw and bark. By combining highly efficient process units and by avoiding a recycle to the reformer stage, the equipment sizes can be kept small, while still achieving (power&)biomass-to-liquid efficiencies of up to 58.0%. An AspenPlus® flowsheet model was set up to model multiple process configurations which were evaluated using the DLR software-tool TEPET. Through automated heat integration, different process configurations were assessed, and a trade-off was identified between maximizing the BtL efficiency and minimizing net production costs. Even though the PBtL concepts come with high process efficiencies, the additional investment and costs for electricity lead to higher production costs in Germany in 2020. Furthermore, the shown work shows the potential of a hydrogen-enhanced biomass-to-liquid process especially for preliminary unattractive biogenic waste materials such as straw.

With an increasing investment into renewable energy sources and a ramp-up of cheap renewable electricity generation technologies, the PBtL concept might contribute to a carbon-neutral domestic transport by ensuring that the biogenic carbon is utilized best possible. To minimize the investment risk associated with the introduction of the technology, one approach could be to start the market introduction with biomass-to-methanol plants. These plants could then be expanded to include a hydrogen production plant and a second methanol synthesis reactor once a sufficient supply of renewable electricity is available.

Appendix A

Heat integration has been conducted with the automated heat and utilisation feature of the DLR-inhouse tool TEPET [10]:

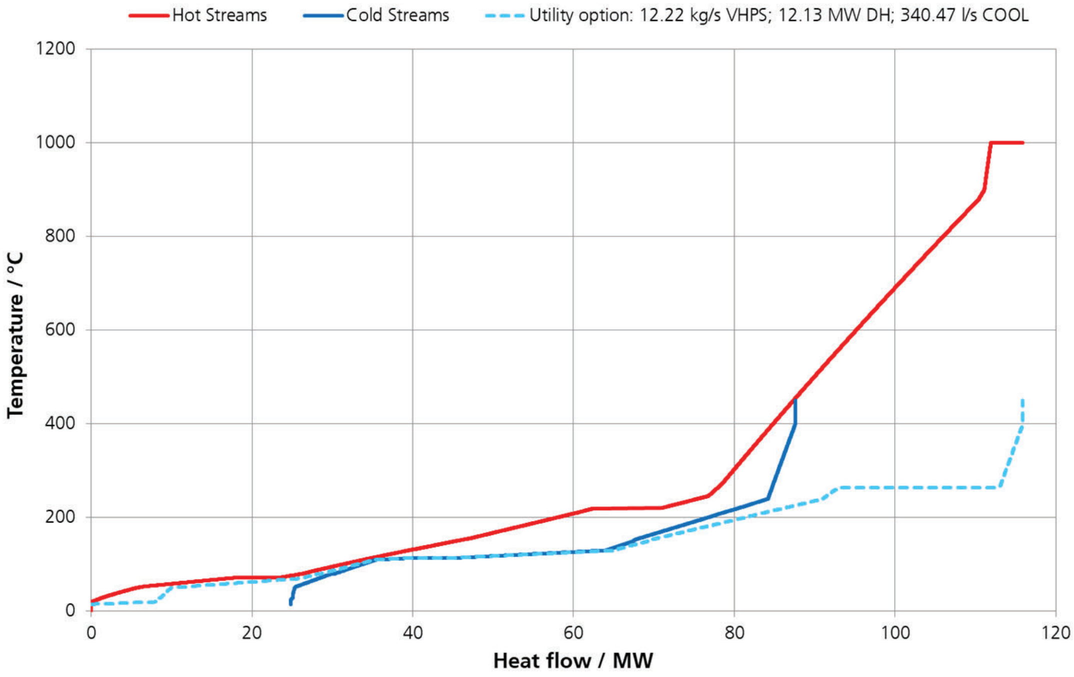

Figure 5. Heat integration for the bark-based biomass-to-liquid process.

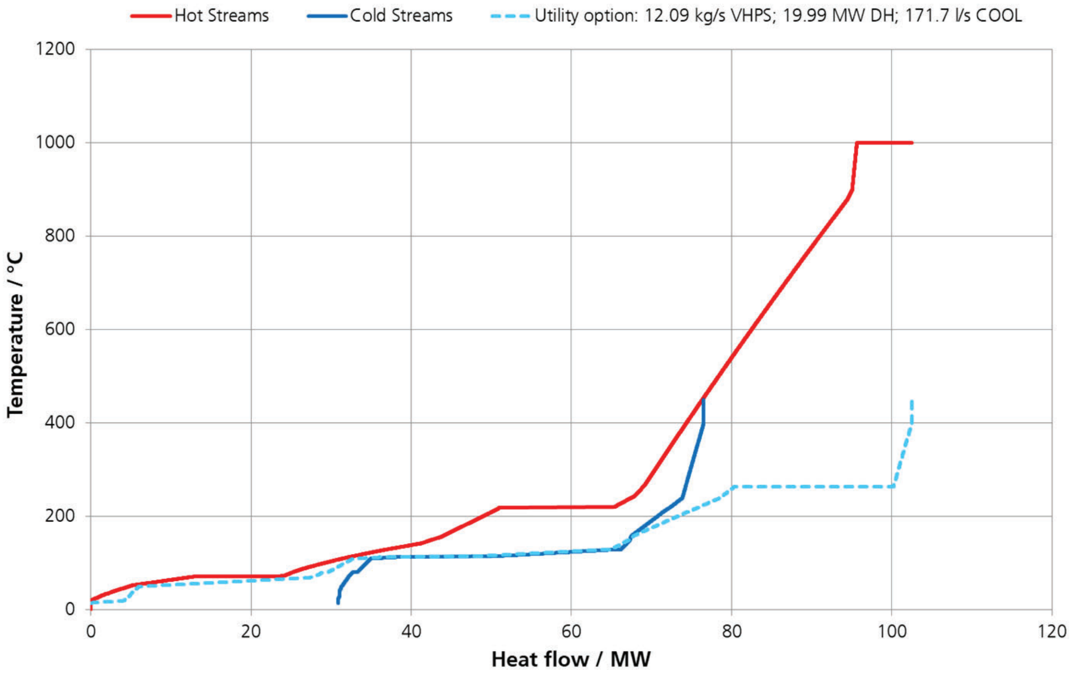

Figure 6. Heat integration for the bark-based power&biomass-to-liquid process.

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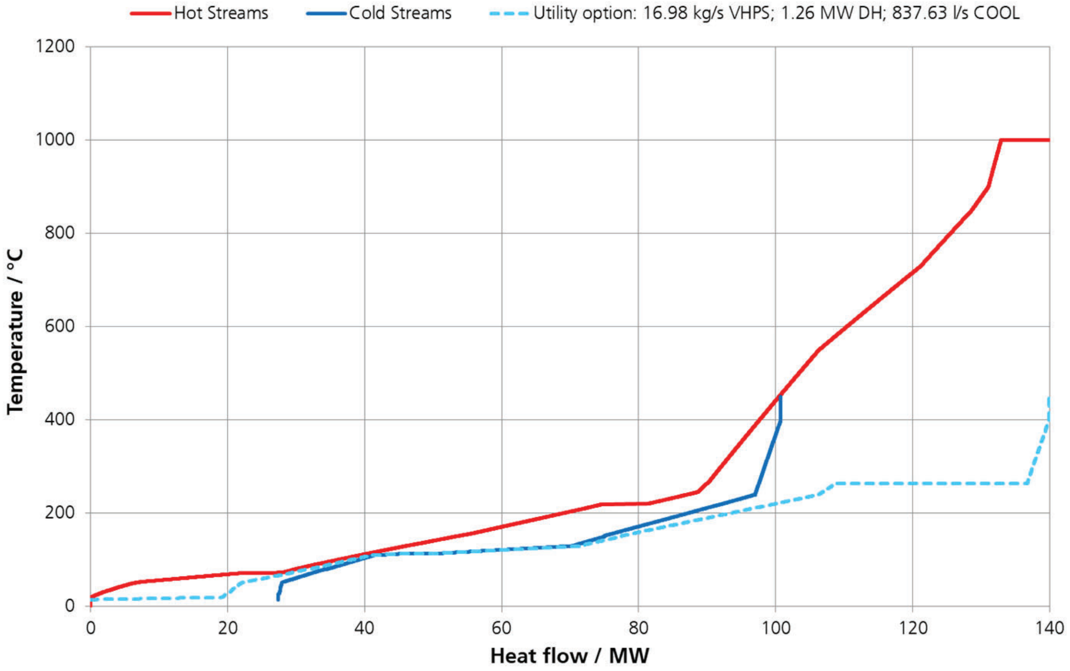

Figure 7. Heat integration for the straw-based biomass-to-liquid process.

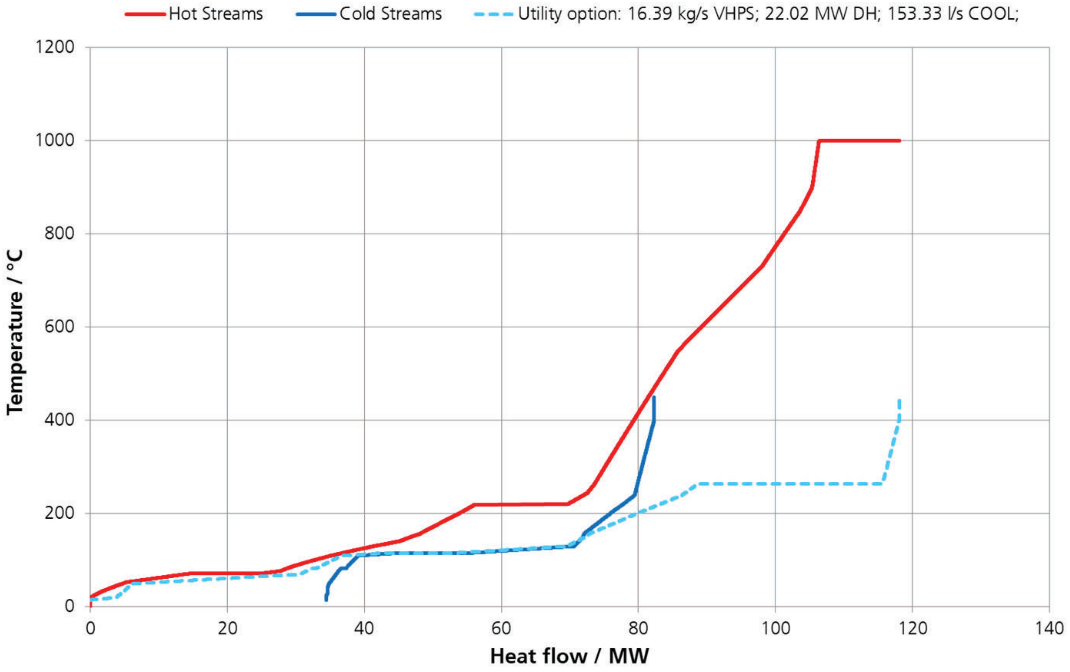

Figure 8. Heat integration for the straw-based power&biomass-to-liquid process.

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Nomenclature

Abbreviations	
CAPEX	Capital investment expenditures
OPEX	Operational expenditures
Acronyms	
ACC	Annualized capital costs
BtL	Biomass-to-Liquid
DFB	Dual fluidized bed
DH	District heating
EC	Equipment cost
FCI	Fixed capital investment
FLH	Full load hours
HHV	Higher heating value
IR	Interest rate
LHV	Lower heating value
NPC	Net production costs
PL	Plant lifetime
PBtL	Power&Biomass-to-Liquid
TCI	Total capital investment
TEPET	Techno-Economic Process Evaluation Tool
Greek letters & variables	
α	Capital cost factor for utility cost calculation
β	Operational cost factor for utility cost calculation
η	Plant efficiency
c_i	Costs for i = [raw material, heat, power]
C_{plant}	Plant capacity, MW_{th}
d_i	Degression factor
D_i	Equipment capacity
$\dot{m}$	Mass flow, t/h
N_P	Number of persons per shift
P_e	Power, MW_e
r_i	Revenue for i = [by-product, heat, power]

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