



Fischer-Tropsch products from biomass-derived syngas and renewable hydrogen

Hannes Gruber^{1,2} · Peter Groß¹ · Reinhard Rauch^{2,3} · Alexander Reichhold¹ · Richard Zweiler⁴ · Christian Aichernig⁵ · Stefan Müller⁶ · Nabeel Ataimisch⁷ · Hermann Hofbauer^{1,2}

Received: 29 March 2019 / Revised: 31 May 2019 / Accepted: 4 June 2019
© The Author(s) 2019

Abstract

Global climate change will make it necessary to transform transportation and mobility away from what we know now towards a sustainable, flexible, and dynamic sector. A severe reduction of fossil-based CO₂ emissions in all energy-consuming sectors will be necessary to keep global warming below 2 °C above preindustrial levels. Thus, long-distance transportation will have to increase the share of renewable fuel consumed until alternative powertrains are ready to step in. Additionally, it is predicted that the share of renewables in the power generation sector grows worldwide. Thus, the need to store the excess electricity produced by fluctuating renewable sources is going to grow alike. The “Winddiesel” technology enables the integrative use of excess electricity combined with biomass-based fuel production. Surplus electricity can be converted to H₂ via electrolysis in a first step. The fluctuating H₂ source is combined with biomass-derived CO-rich syngas from gasification of lignocellulosic feedstock. Fischer-Tropsch synthesis converts the syngas to renewable hydrocarbons. This research article summarizes the experiments performed and presents new insights regarding the effects of load changes on the Fischer-Tropsch synthesis. Long-term campaigns were carried out, and performance-indicating parameters such as per-pass CO conversion, product distribution, and productivity were evaluated. The experiments showed that integrating renewable H₂ into a biomass-to-liquid Fischer-Tropsch concept could increase the productivity while product distribution remains almost the same. Furthermore, the economic assessment performed indicates good preconditions towards commercialization of the proposed system.

Keywords Fischer-Tropsch synthesis · BtL · Energy storage · Excess electricity · ASF distribution · Economic assessment

Abbreviations

FT	Fischer-Tropsch	BtL	biomass-to-liquid
FTS	Fischer-Tropsch synthesis	PtG	power-to-gas
¹ H	hydrogen-nuclear magnetic resonance	ICE	internal combustion engines
NMR	spectroscopy	HC	hydrocarbon
		C _n	hydrocarbon with <i>n</i> carbon atoms
		DFB	dual fluidized bed
		BTX	benzene, toluene, xylene
		SV	space velocity
		NI	standard liter
		Nm ³	standard cubic meter
		h	hour
		SBCR	slurry bubble column reactor
		P/O	paraffin/olefin ratio by weight
		ASF	Anderson-Schulz-Flory
		RWGS	reverse water-gas shift
		GC	gas chromatography
		FID	flame ionization detector
		TCD	thermal conductivity detector
		ROI	return on investment

✉ Hannes Gruber
hannes.gruber@tuwien.ac.at

¹ Institute of Chemical, Environmental & Biological Engineering, TU Wien, Vienna, Austria

² Bioenergy 2020+, Güssing, Austria

³ Engler-Bunte-Institut, Karlsruhe Institute of Technology, Karlsruhe, Germany

⁴ Güssing Energy Technologies GmbH, Güssing, Austria

⁵ Aichernig Engineering GmbH, Vienna, Austria

⁶ EC Engineering GmbH, Vienna, Austria

⁷ Bilfinger Bohr- und Rohrtechnik GmbH, Vienna, Austria

1 Introduction

1.1 The need for decarbonization

At this year's United Nations Framework Convention on Climate Change Conference, a strategy for a functional Paris agreement was negotiated. It is going to be challenging to limit global warming to 1.5 or 2 °C above preindustrial levels, as proposed by the Paris Agreement. Today, anthropogenic global warming accounts already for about 1 °C, reaching 1.5 °C between 2030 and 2050. To reach either of these climate goals, net zero for anthropogenic CO₂ emissions needs to be accomplished in this century [1, 2].

In Germany, the share of renewable energy consumed by transportation settled to 5.2% with stagnating tendencies in 2017. Compared to 2007 rates (7.5%), a decrease was observed in this regard. On the other hand, the renewable share of gross electricity consumed increased at a high pace from 14.3% in 2007 to 36.2% in 2017 [3].

Thus, a high potential to reduce fossil CO₂ emissions in transportation is evident. Fischer-Tropsch synthesis (FTS) produces hydrocarbons ranging from CH₄ to hydrocarbons with more than 60 carbon atoms. The products of an integrated FTS biorefinery designed to produce renewable transportation fuel could be premium diesel and kerosene [4, 5]. Feasible upgrading steps and final applications of these renewable fuels derived by FTS were discussed in complementary literature [4–13].

The addition of renewable power production capacities is predicted to increase in the next years [14]. This expansion will correlate with the availability of fluctuating excess electricity. Power-to-gas (PtG) systems are a suitable technology to convert and subsequently store excess electricity [15]. Gahleitner [16] gives an overview on realized PtG facilities. One of the main issues regarding the quick integration of H₂ or CH₄, produced via PtG technologies, as transportation fuel is the lack of cars powered with these fuels. In Germany, over 45 million registered vehicles are powered by diesel or gasoline, whereas less than one million are powered by alternative power trains, in 2018 [17]. Even though new legislation may benefit the development and distribution of alternative powertrains, a complete shift away from internal combustion engines (ICE) will take time. To reach a rapid reduction of CO₂ emissions in transportation, alternative fuels will be needed to power existing ICEs. The Winddiesel technology provides a solution to this dilemma. Groß et al. [18] presented the concept in 2015. The basic idea of this technology is shown in Fig. 1. Biomass gasification, using CO₂ and steam as gasification agent, produces CO-rich product gas [19–22]. CO₂ is removed from the product gas stream. If excess electricity is available, CO₂ is recycled as gasification agent to the gasifier promoting the reverse water-gas shift (RWGS) reaction. CO₂ that is not fed to the gasifier might be utilized using a RWGS

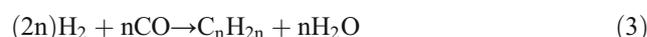
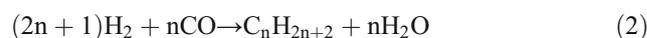
unit as proposed by Pastor-Pérez et al. [23]. H₂, produced by excess electricity, is mixed with the CO-rich product gas from biomass gasification. Subsequently, FTS is applied to convert the cleaned and conditioned syngas to hydrocarbons. If integrated without storage capacities, the fluctuating H₂ implies load changes to FTS. Compared to a conventional biomass-to-liquid (BtL) FT concept, the FT capacity is enlarged, an electrolyzer is added to enable the utilization of excess electricity, and CO₂ is recycled. The desired products naphtha, diesel, and wax are fractionated and if required refined. After purification, the product water can be recycled as feed for electrolysis.

Müller et al. [24] validated experimental data using process simulation. This investigation showed that the integration of renewable H₂ produced by excess electricity into FTS fuel production is a suitable possibility to store energy. Further investigation regarding efficiency and economic aspects regarding Winddiesel were proposed. Nikparsa et al. [25] investigated FT syncrude regarding olefin distribution using ¹H NMR technology.

This work is a summary and review of experimental and economic data published so far in the course of the Winddiesel project. Additionally, new aspects regarding substance-class distribution of FTS syncrude, effects of load changes on productivity, and economic aspects were added and discussed.

1.2 Why Fischer-Tropsch synthesis?

First insights on the hydrogenation of CO were reported by Sabatier et al. [26] in 1902. Subsequently, Fischer and Tropsch [27] described the production of synthetic oil from CO and H₂. Later research and the commercialization are summarized by Steynberg [28]. FTS produces a mixture of hydrocarbons (syncrude) with different chain lengths. The catalytic conversion of H₂ and CO to hydrocarbons is described as stepwise reaction. CH₂ intermediates (Eq. 1) are formed from H₂ and CO. The chain growth and the production of paraffins (Eq. 2), olefins (Eq. 3), and oxygen-containing components by FTS are summarized by Schulz [29]. Further summaries regarding FT reactions were published by other authors [30, 31].



The Anderson-Schulz-Flory (ASF) distribution describes the distribution of FT products. De Klerk [31] described the ASF distribution and summarized deviations from it. The mass fraction W_n of hydrocarbons with n carbon atoms can be determined experimentally by gas chromatography. Subsequently, the chain growth parameter α can be

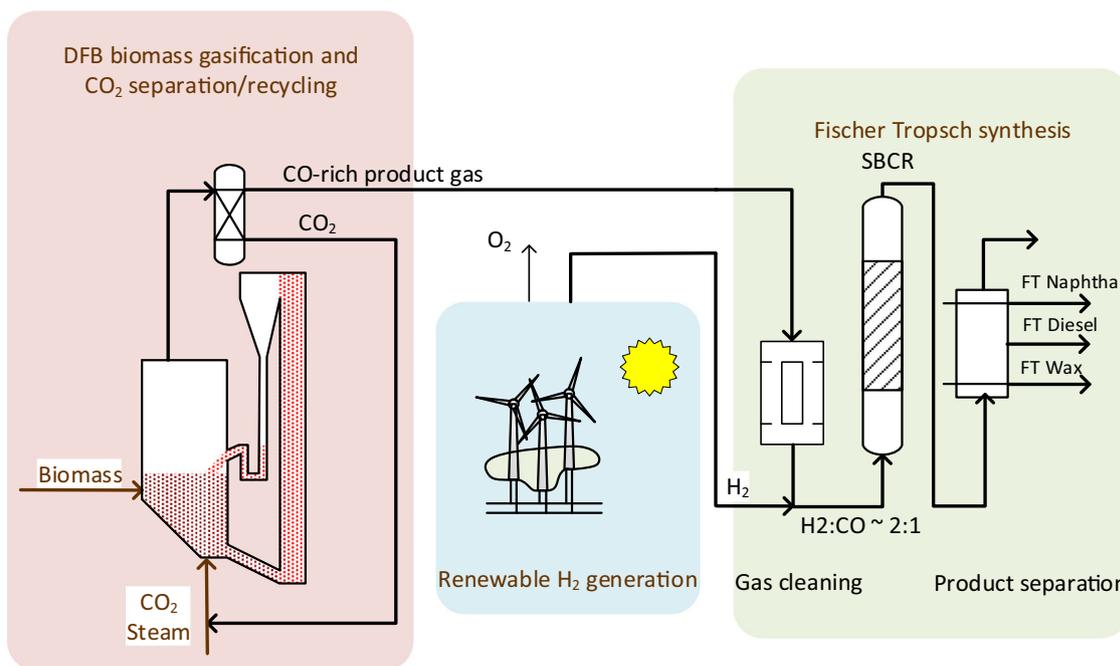


Fig. 1 Basic concept of the “Winddiesel” technology

determined from the straight-line proportion of the ASF plot using Eq. 4 [32].

$$\log\left(\frac{W_n}{n}\right) = n \cdot \log(\alpha) + \log\left(\frac{(1-\alpha)^2}{\alpha}\right) \quad (4)$$

Anderson [30] gives an overview on suitable catalysts for FTS. Industrial case studies were summarized by Yong-Wang and de Klerk [33]. Possible products from FTS are gasoline, diesel, jet fuel, paraffinic waxes, lubricants, petrochemical base products, and so on [34].

FTS has the capability to convert biomass-based syngas and renewable H₂ from excess electricity to hydrocarbons with different chain lengths. The product flexibility might be one of the biggest benefits of FTS. If the production of renewable fuels becomes unprofitable, FT products have other high-value applications such as 1-olefins [35] (naphtha), white oil blend [36] (diesel), and solid synthetic paraffin [37] (wax). Subsequently, this work summarizes the effects of a varying load on FT performance [24, 38, 39] and presents new insights.

The controversial topic of biomass-based fuels was discussed in literature [40–45]. Different pathways to produce renewable fuels in agreement with current and future legislative and ethic aspects were analyzed. The European Union is currently promoting the production of second-generation biofuels produced from organic residues and waste while at the same time introducing the phasing out of first-generation biofuels [46, 47]. Alternative substitutions for the farmed

wood feedstock, which were used in this investigation, are discussed in the conclusion section.

2 Experimental

Seven long-term experimental campaigns were performed at Güssing’s biomass conversion research facility. This section gives an overview on materials and methods used to perform and evaluate the experiments.

2.1 Production of biomass-derived syngas

The product gas to perform the experiments was provided from the Güssing 8 MW_{th} biomass steam gasification plant using wood chips as feedstock. At the commercial plant, contaminants like particles, benzene, toluene, xylene (BTX), and tars are reduced to levels suitable for gas engine application. Nevertheless, this gas cleaning is not sufficient for FT synthesis and further cleaning steps were integrated in the FT unit.

Table 1 Ranges of the main components of the product gas [48]

Component	Unit	Composition
CO	v% _{dry}	20–30
H ₂	v% _{dry}	35–45
CO ₂	v% _{dry}	15–25
CH ₄	v% _{dry}	8–12

Typical ranges of the main components of the DFB gasifier's product gas are shown in Table 1 [48–50].

2.2 Fischer-Tropsch laboratory unit

The laboratory FT unit used to perform the experiments has been continuously improved, integrating know-how of over 10,000 operating hours. Several works [24, 32, 38, 51–53] described the extensive research performed, operating the laboratory FTS unit. The basic flow sheet of the FT unit is shown in Fig. 2. The three main plant sections are as follows.

The *gas cleaning and conditioning* unit removes tars, BTX, chlorine, and sulfur contaminants from the product gas. Sulfur components are reduced to levels below 10 ppb. This is essential to protect the cobalt catalyst, used for FT synthesis, from poisoning. Rauch et al. [54] summarized acceptable levels regarding impurities like sulfur components and alkali metals if cobalt catalysts are used.

The 20-l *slurry bubble column reactor (SBCR)* converts H_2 and CO to hydrocarbons with chain lengths ranging from C_1 to $>C_{60}$. The clean syngas enters the three-phase slurry reactor at the bottom. Products and non-converted gaseous components exit the reactor at the top.

The *three-step high-pressure product condensation* separates liquid FT products and FT water from gaseous components.

The laboratory FT plant is fully automatized. The H_2/CO ratio is monitored, and adjusted if additional H_2 is needed for ideal stoichiometric FTS conditions.

2.3 Analysis methods

The inlet and outlet gas composition of the SBCR was measured continuously by gas chromatography (GC). Gaseous hydrocarbons were determined using a flame ionization detector (FID) and other components with a thermal conductivity detector (TCD). GC was also used to analyze liquid and solid

FT products. In-house methods were applied to determine the hydrocarbon distribution. The GC was equipped with a FID and SimDist column. Sauciuc et al. [51] described the analysis method in more detail.

Furthermore, a substance class analysis was carried out. For low-molecular FT syncrude (C_6 – C_{20}), the content of n-paraffins, 1-olefins, and other hydrocarbons (other HC) was determined. The substance class of other HC summarizes i-paraffins, oxygenates, and other substance classes. For higher-molecular hydrocarbons, it was distinguished between n-paraffins and other HC.

The per-pass CO-conversion ($X_{CO,reactor}$) is defined by Equation 5 [24]. $X_{CO,reactor}$ is defined by the quotient of moles of CO consumed by FTS ($n_{CO, in} - n_{CO, out}$) and moles entering the SBCR ($n_{CO, in}$).

$$X_{CO,reactor} = \frac{n_{CO,in} - n_{CO,out}}{n_{CO,in}} \quad (5)$$

Note that Gruber et al. [38] defined $X_{CO,reactor}$ different to Eq. 5. Additionally, the system productivity regarding condensable FT syncrude (\dot{m}_{C5+}) was evaluated (Eq. 6). \dot{m}_{C5+} is derived by dividing the sum of naphtha ($\dot{m}_{naphtha}$), diesel (\dot{m}_{diesel}), and wax (\dot{m}_{wax}) mass flow by a mass catalyst ($m_{catalyst}$) applied to the system.

$$\dot{m}_{C5+} = \frac{\dot{m}_{naphtha} + \dot{m}_{diesel} + \dot{m}_{wax}}{m_{catalyst}} = \sum_{i=5}^{\infty} \dot{m}_i \quad (6)$$

Furthermore, the n-paraffin to 1-olefin (P/O) ratio (Eq. 7) was determined by the quotient of mass n-paraffin to mass 1-olefin produced per experiment.

$$P/O \text{ ratio} = \frac{m_{n\text{-paraffin}}}{m_{1\text{-olefin}}} \quad (7)$$

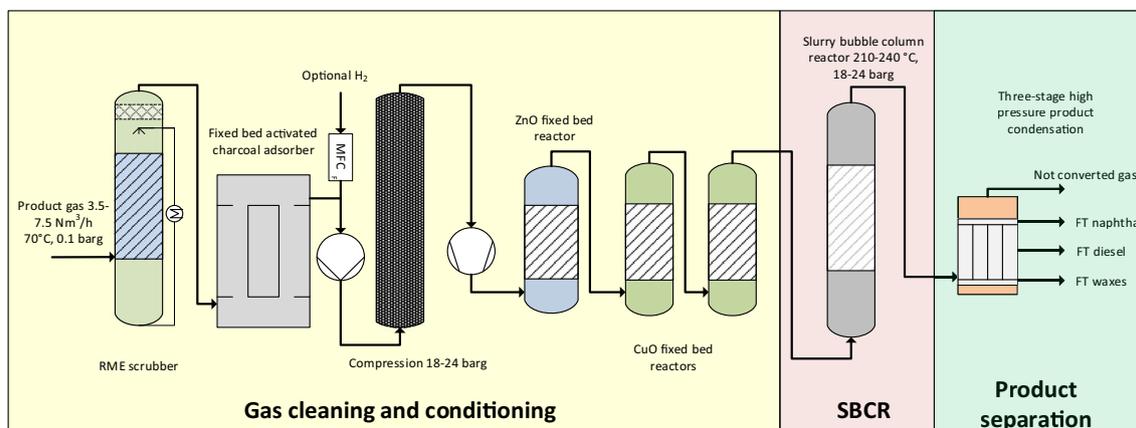


Fig. 2 Basic flowsheet of the laboratory-scale FT unit

2.4 Result summary

The integration and subsequent storing of renewable H_2 were investigated by applying load changes to the FT system. Figure 3 shows the volume flow applied to the SBCR simulating low capacity, benchmark operation, peak capacity, and load changes. The SBCR was designed to process 3.5 to 7.5 Nm^3/h , having a performance optimum at 5 Nm^3/h . Experiments performed at low feed rate (3.5 Nm^3/h) simulated low-capacity operation introducing a decreased space velocity (SV) to the synthesis step. Experiments performed at 5 Nm^3/h simulated benchmark operation. The experiments performed at 7.5 Nm^3/h were carried out to evaluate the performance of the SBCR at peak availability of excess electricity. At load change conditions, the feed rate changed its level every 10 min in the range of 3.5 to 7.5 Nm^3/h . Thus, this experimental work evaluated the effects of low capacity, high capacity, and load change operation on the SBCR. Compared to the 5 - Nm^3/h benchmark operation, the low-capacity and high-capacity experiments represent a load of 70% and 150% , respectively.

Table 2 summarizes published experimental results. In total, three campaigns (experiments 2 to 4) were carried out to investigate the influence of fluctuating renewable H_2 to FTS. Each campaign consists of a long-term base load and a load change experiment. Müller et al. [24] published five experimental data sets that were validated using process simulation. Gruber et al. [38] published additional experimental data. All test runs were performed with 2.5 kg of commercial-grade Co/ Al_2O_3 catalysts from different suppliers.

Experiments 2 and 3 showed a constant α -value comparing load change and base load operation. The α -value obtained from experiment 4 shows a slight decrease from 0.88 to 0.87 if load changes were performed. These results indicate that direct supply of renewable H_2 in to the FTS biorefinery might be realized without severe changes to the product distribution.

Compared to their associated base load experiments, 2B showed a decrease of 15% , and 3B a decrease of 17% regarding $X_{CO,reactor}$ if load changes were performed. Experiment 4B showed stable $X_{CO,reactor}$ compared to 4A. Due to the decreasing α -value experiment, 4B showed stagnating m_{C5+} production despite similar CO conversion and increased load compared to 4A. Thus, a higher share of gaseous FT products was produced in 4B. Furthermore, Müller et al. [24] showed that load changes introduce only marginal temperature deviations to the SBCR.

As part of experiment 4B, Gruber et al. [38] investigated the influence of low and high availability of renewable H_2 by applying low and high SV to the SBCR. Table 3 shows the operation conditions applied to perform the low and high SV experiments in the course of experiment 4B. The 72 h startup phase of the plant, performed at the beginning of each experiment to reach a steady $X_{CO,reactor}$, was not included in this analysis. At high SV, the SBCR feed was increased by 50% to 7.5 Nm^3/h , compared to baseload settings. Low syngas flow on the other hand represents low feedstock availability by decreasing the SBCR feed by 30% to 3.5 Nm^3/h .

Figure 4 shows the experimental results of high and low SV, compared to the associated base and load change experiments. Compared to base load and load change experiments, a slightly higher α -value of 0.89 was achieved by applying high SV. This indicates a higher selectivity towards condensable hydrocarbons. Additionally, m_{C5+} was enhanced by 30% if SV was increased. The trade-off by increasing the SV was the decline of $X_{CO,reactor}$ to 43% .

The low SV experiment showed an α -value of 0.88 , which is slightly lower than at high SV and similar to the chain growth probability at base load settings. By decreasing the SV, the $X_{CO,reactor}$ was enhanced to 57% , which is an augmentation of 10% compared to 52% at base load settings. However, m_{C5+} was decreased to 0.07 $g/h \cdot g_{catalyst}$.

Fig. 3 Feed rates applied to the SBCR

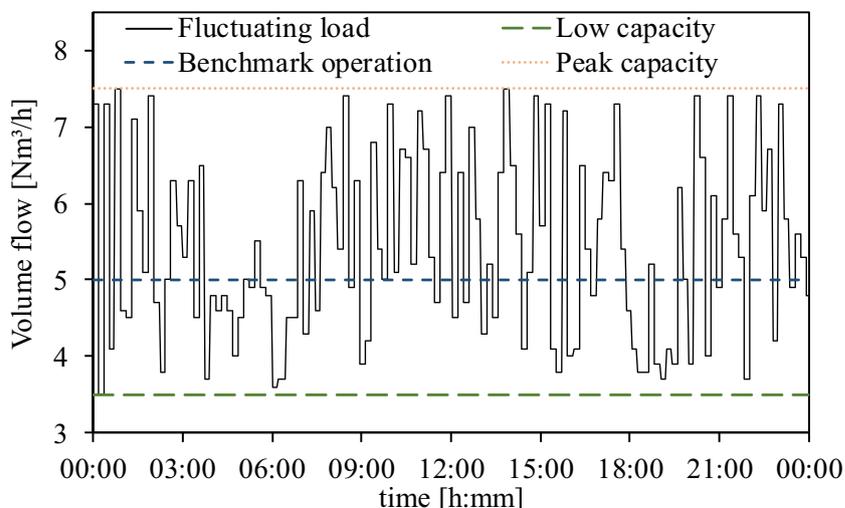


Table 2 Summary of Winddiesel experiments

Parameter	Unit	Benchmark [24]	Base load [24]	Load change [24]	Base load [24]	Load change [24]	Base load [38]	Load change [38]
Experiment	–	1	2A	2B	3A	3B	4A	4B
Duration	h	1000	500	500	500	500	500	225
SBCR temperature	°C	230 ¹						
SBCR pressure	barg	20 ¹						
Catalyst	–	O	A	A	B	B	C	C
Average SBCR feed	Nm ³ /h	4.5	4.6	5.6	4.8	5.2	5.0	5.4
SV	Nl/h·g _{catalyst}	1.80	1.84	2.24	1.92	2.08	2.0	2.2
H ₂ /CO ratio	–	2.40	1.80	2.00	2.40	3.10	2.1	2.1
m _{C5+}	g/h·g _{catalyst}	0.08	0.09	0.11	0.05	0.04	0.10	0.10
Product H ₂ O	g/h·g _{catalyst}	0.116	0.12	0.14	0.1	0.08	0.20	0.22
α-Value	–	0.90	0.93	0.93	0.90	0.90	0.88	0.87
X _{CO, reactor}	%	32	40	34	12	10	52	52

¹ Values applied in all experiments

Another important factor to evaluate the effects of syngas load changes to the slurry-based FTS reactor is the product analysis regarding substances produced. The main substance classes found were *n-paraffins*, *1-olefins*, and *other hydrocarbons* (HC). The substance class of other HC includes mainly *i-paraffins* and oxygenates. The liquid FTS fraction C₆–C₂₀ was analyzed due to its *n-paraffin*, *1-olefin*, and other HC content. The solid FTS product C₂₁₊ was analyzed regarding *n-paraffin* and other HC content. Within the solid fraction, the substance class of other HC also includes *1-olefins*. Figure 5 shows the cumulative substance class distribution of the FTS liquids and solids obtained after 200 operating hours at load change conditions in experiment 4B.

Figure 6 shows the results of the substance class analysis of liquid and solid FT products produced during experiment 4B. Hydrocarbons with a chain length of C₆ to C₂₀ were considered as liquid FT syncrude fraction. C₂₁₊ hydrocarbons are defined as solid FT fraction.

By applying load changes to the slurry-based FT system, the *n-paraffin* content rises from 75.3 to 77.7 wt.% in the liquid product and from 81.2 to 82.2 wt.% in the solid product, compared to base load operation. The base load experiment

showed the lowest and load change experiment and the highest *n-paraffin* content for both liquid and solid products. The liquid fraction had an O/P ratio of 9.69 at load change settings and 7.14 at base load operation. By applying low SV, the liquid and solid FT product had an *n-paraffin* content of 77.0 wt.% and 81.7 wt.%, respectively. A P/O ratio of 11.99 was obtained for the liquid FTS fraction at low syngas application. At high SV, a *n-paraffin* content of 75.9 wt.% was obtained for liquid FT products and 81.3 wt.% for C₂₁₊ hydrocarbons. The analysis of the liquid FT fraction produced at high SV showed a P/O ratio of 6.67.

3 Economic assessment

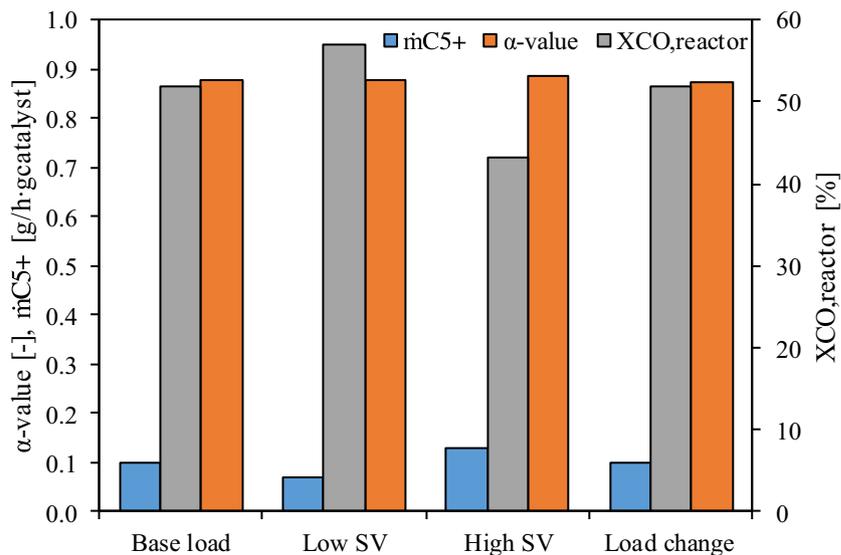
This chapter summarizes and discusses the economic assessment performed by Zweiler et al. [39]. A sensitivity analysis was performed to evaluate the effects on the economic performance, if key parameters were varied. Based on experimental results and simulations carried out within this project, a flexible economic model was acquired. The aim of this economic

Table 3 Operational settings of low and high SV experiments

Parameter	Unit	Low SV [38]	High SV [38]	Load change [38]	Base load [38]
Experiment	–	4B	4B	4B	4A
SBCR temperature	°C	230 ¹			
SBCR pressure	barg	20 ¹			
SV	Nl/h·g _{catalyst}	1.40	3.00	1.4–3.0	2
H ₂ /CO ratio	–	2.1	2.1	2.1	2.1
Catalyst	–	C ¹			
Experiment duration	h	81	26	46	500

¹ Values applied in all experiments

Fig. 4 Performance indicating parameter of high and low SV experiments with data from [38]



assessment was to evaluate the return on investment (ROI) of commercial Winddiesel biorefineries.

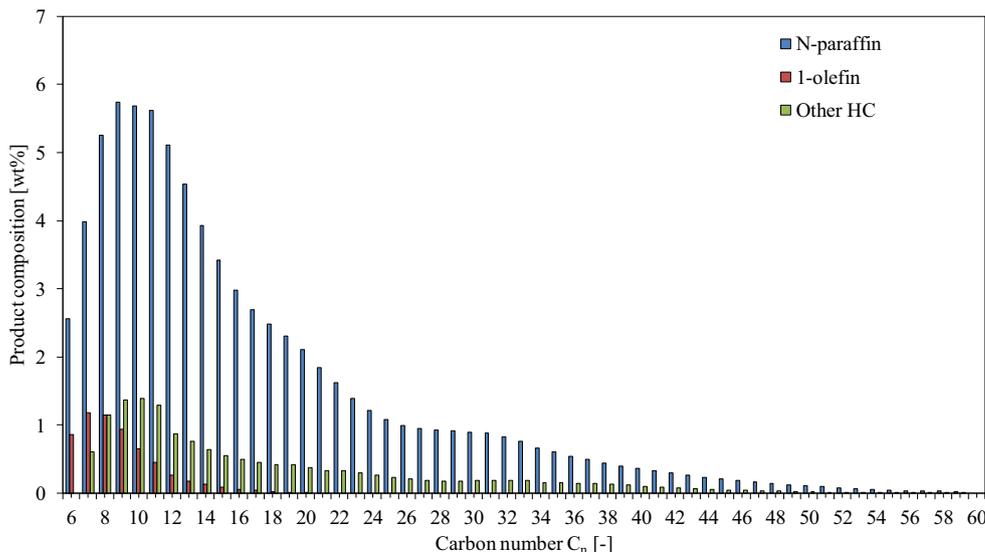
3.1 Data basis

The economic assessment includes operating and investment cost of essential plant units. The evaluated process chain includes the lignocellulosic biomass gasification plant, the H₂ production via electrolysis, the FT unit, and a steam cycle to convert excess heat to electricity. EC Engineering simulated mass and energy balances used in this assessment. Costs for insurance, wearing parts, maintenance, commodities, credit payback, and operating personnel were summarized as yearly operational costs. To process additional renewable H₂, the FT unit is designed to have a capacity of 170% compared to base load settings. Table 4 shows the assumed FT product

distribution and the assumed market price for the naphtha, diesel, and wax fraction. As can be seen from the table, waxes (C₂₁₊) are assumed to be the most valuable product. The operating conditions of the FT plant are assumed to be optimized for high wax production [39].

Performance-indicating parameters used in this assessment are summarized in Table 5. Two cases, a 50 MW_{th} (case A) and a 200 MW_{th} (case B) fuel input, were investigated. Case A represents a medium-scale biorefinery, producing about 20,000 t of condensable hydrocarbons (naphtha, diesel, and wax) per year. Case B, representing a reasonable upscale to case A, would produce about 88,000 t of condensable hydrocarbons per year. In both cases, a suitable location provides a secure supply of biomass feedstock and excess renewable electricity. Additionally, a suitable setup regarding product monetarization should be located at a close range to minimize

Fig. 5 Exemplary substance class distribution of FTS syncrude, at load change conditions after 200 operating hours



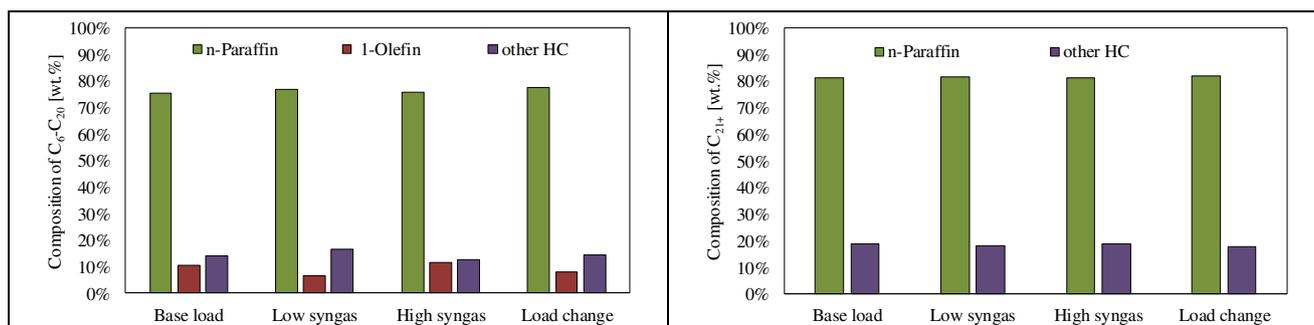


Fig. 6 Composition of liquid (left) and solid (right) FT products (experiments 4A and 4B)

transportation. Strategic factors for biomass conversion sites of similar size compared to the proposed system, such as economics, plant location, and biomass availability, were discussed in further studies [41, 56–60]. The assumption of the costs for excess electricity was based on prices listed at electricity exchanges. Because of the high degree of novelty and the remarkable potential to save CO₂ emissions, an investment subsidy rate of 50% is considered realistic for the first demonstration plant. Furthermore, external financing for investment cost is assumed. Sellable excess electricity, FT products, and oxygen from electrolysis are considered as income [39].

The economic performance was assessed by calculating the ROI (Eq. 8). The yearly profit was calculated by subtracting the operational costs from the income.

$$\text{ROI} = \frac{\text{yearly profit}}{\text{investment-funding}} \quad (8)$$

Considering the commercialization of the integrative energy-storing and fuel production concept, it needs to be more profitable than a standalone BtL plant including biomass gasification and FTS. For this reason, the ROI of such a standalone BtL plant was calculated with 8.9%. Subsequently, the breakeven point regarding the integration of excess electricity was evaluated. The breakeven point describes the minimal full-load operating hours necessary, to obtain a ROI equal to the standalone BtL concept. The minimal electrolysis operating hours to gain a ROI of 8.9% at energy storing modus are listed in Table 5. The full-load operating hours of the electrolysis unit were kept constant for

Table 4 Assumed product distribution and their estimated marked prices [39]

Parameter	Unit	FT naphtha	FT diesel	FT waxes
Price	EUR/l	0.78	0.74	2 ¹
Product distribution	wt.%	13.00	37.35	49.65

¹ Price in (EUR/kg) assumption based on [55]

each sensitivity analysis performed in this economic assessment.

3.2 Parameter variation

To gain insights on how key parameters affect the economic performance of the proposed system, a parameter variation was performed. Table 6 shows the parameters that were considered in this sensitivity analysis. Wood chips with a price of 20 EUR/MWh were assumed as standard fuel input in this assessment. If organic residuals or waste could be utilized to fuel the integrative biorefinery, a much lower fuel price could be assumed. Thus, the fuel price was varied from 0 to 40 EUR/MWh. Feedstock with a price < 15 EUR/MWh is considered as low-cost feedstock. A price of 40 EUR/MWh was considered for premium fuels. The uncertainty regarding the price for excess electricity was covered by varying it from – 50 to + 50 EUR/MWh. The funding rate was varied from 0 to 70%.

The results achieved by varying the fuel price are shown in Fig. 7 (top-left). Case A showed that, if the fuel price rises to 30 EUR/MWh, a ROI > 6% could be obtained. Case B showed that, despite that premium fuels are applied, a ROI

Table 5 Data summary used to perform the economic assessment [39]

Parameter	Unit	Case A	Case B
Fuel input	MW	50	200
Water content of wood chips	wt.%	40	40
Wood chip price	EUR/MWh	20	20
Basis price electricity	EUR/MWh	70	70
Price excess electricity	EUR/MWh	25	25
Full-load hours of FT base load	h	7500	7800
Full-load hours of electrolysis	h	3460	4209
El. power input of electrolysis	MW	33.5	134.1
Electrical own consumption	%	10.0	8.2
Gasification cold gas efficiency	%	70.0	70.0
Investment funding	%	50.0	50.0
Operational costs	kEUR/a	26,552	107,275
Income	kEUR/a	33,276	143,917
Investment costs	kEUR	150,350	412,250

Table 6 Parameter variation

Parameter	Unit	Range
Fuel costs	EUR/MWh	0 to 40
Surplus electricity	EUR/MWh	- 50 to 50
Investment funding	%	0 to 70

> 10% could be obtained. The fluctuating excess electricity is designated to power the electrolysis for H₂ production. The price for surplus electricity was varied between - 50 and 50 EUR/MWh. The results obtained by varying the price for excess electricity is shown in Fig. 7 (top-right). Case A showed that, if the price for surplus electricity rises to 50 EUR/MWh, a ROI of 6.9% could be achieved. Moreover, Case B calculated a ROI of 14.1% if costs for surplus electricity rise to 50 EUR/MWh. To cover uncertainties regarding the funding rate, a parameter

variation from 0 to 70% investment subsidies was carried out. The results of this variation are shown in Fig. 7 (bottom-left). Case A showed a ROI of 4.4% and case B a ROI of 8.7%, if no investment subsidies will be granted.

4 Conclusion

Within this project, the effects of the fluctuating syngas flow on the FTS performance were investigated. Three long-term campaigns were performed comparing different load regimes applied to the SBCR regarding per-pass CO conversion, productivity, and product distribution. A direct utilization of excess electricity would imply higher and fluctuating loads to the FTS compared to conventional BtL operation.

The high SV experiments showed that an increased load of 50% in fact decreases the CO conversion by 17% but on the contrary increases the productivity by 30%. Considering

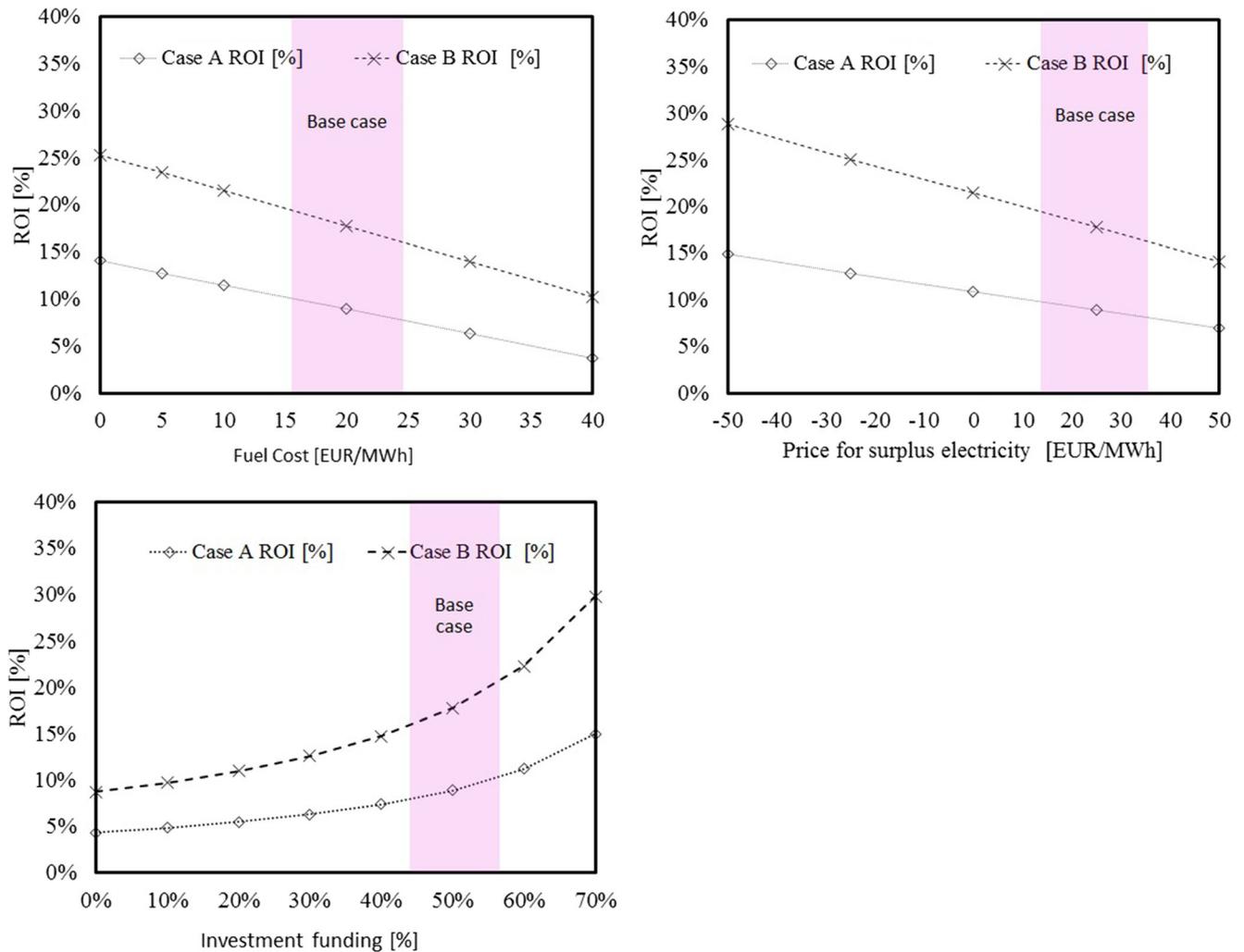


Fig. 7 Economic performance of the proposed biorefinery (top-left: fuel cost variation; top-right: price variation of excess electricity; bottom-left: variation of investment funding, based on data from [39])

experiment 2, the analysis of the chain growth probability factor α showed similar results in all performed operation modes. A per-pass CO conversion of up to 57% was reported at low SV settings. The substance class analysis showed that the solid FT products have a higher n-paraffin share compared to liquid FT products. In the solid FT fraction, a n-paraffin of up to 82.2 wt.% was determined, whereas in the liquid fraction the maximal reported n-paraffin was 77.7 wt.%. The P/O ratio of the liquid FT product was determined between 6.67 and 11.99. By comparing the performance of the applied commercial-grade cobalt-based FT catalysts, it became obvious that catalyst choice will be a major factor towards a stable and economic plant operation. This experimental work indicates that the utilization of excess electricity via electrolysis, CO₂ gasification, and FTS could increase the productivity compared to conventional BtL operation and at the same time maintain the product distribution in the same range.

The improvement of fuel flexibility will be one of the main economic challenges for industrial gasification plants in the future. Lignocellulosic residues, sewage sludge, or plastic residues might be easily obtainable, low-cost feedstock alternatives. Germany, for example, produced about 1.7 million tons of sewage sludge (dry substance) in 2017 [61]. Additional 27.1 million tons of plastic waste was generated in Europe in 2016 [62]. However, the challenges of the utilization of plastic were reviewed by Lopez et al. [63]. Increasing costs for waste management and waste disposal, if not prohibited by law, might be a benefactor that promotes the utilization of these materials via gasification [64]. The applicability of gases produced by sewage sludge or plastic gasification [64–66] for FTS needs to be evaluated in further research work. A suitable location providing secure access to low-cost gasification feedstock and nearby product consumers might be municipal waste management facilities. It seems realistic that investment funding will be granted to a demonstration plant. However, follow-up plants are not expected to be subsidized. Thus, the 0% investment funding case should be considered regarding further investments.

The experimental work and the economic assessment indicate that the utilization of excess electricity improves the productivity of the proposed biorefinery, compared to conventional BtL FT setups. Further experimental work should evaluate the benefit of adding a gas loop and a steam reformer to the FT unit. Additionally, the feasibility to implement a RWGS unit to utilize surplus CO₂ should be investigated.

Symbols α chain growth probability

- W_n mass fraction of hydrocarbons with n carbon atoms
- n_i moles of substance i
- m_i mass of substance i
- \dot{m}_i mass flow of substance i

Acknowledgments The project Winddiesel_klienIF is executed in cooperation with Energie Burgenland AG, Bilfinger Bohr- und Rohrtechnik

GmbH, Güssing Energy Technologies GmbH, REPOTEC GmbH & Co KG, Energy & Chemical Engineering GmbH, and the Institute of Chemical Engineering from TU WIEN.

Funding information Open access funding provided by TU Wien (TUW). This study was financially supported by the Austrian government through the “Klima- und Energiefonds”-financed Project Winddiesel_klienIF within the “e!Mission.at” funding scheme.

Open Access This article is distributed under the terms of the Creative Commons Attribution 4.0 International License (<http://creativecommons.org/licenses/by/4.0/>), which permits unrestricted use, distribution, and reproduction in any medium, provided you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made.

References

1. Masson-Delmotte V, Zhai P, Pörtner H-O, Roberts D, Skea J, Shukla PR, Pirani A, Moufouma-Okia W, Péan C, Pidcock R, Connors S, Matthews JBR, Chen Y, Zhou X, Gomis MI, Lonny E, Maycock T, Tignor M (2018) IPCC - Summary for Policymakers. In: Global Warming of 1.5°C. An IPCC Special Report on the impacts of global warming of 1.5°C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global resp. World Meteorological Organization, Geneva
2. United Nations (2019) The Katowice climate package: making the Paris Agreement work for all. <https://unfccc.int/process-and-meetings/the-paris-agreement/katowice-climate-package>. Accessed 6 Mar 2019
3. BMWi (2019) “Anteil Erneuerbarer Energien an der Energieversorgung in Deutschland nach Bereichen im Jahresvergleich 2007 und 2017.” - Statista - Das Statistik-Portal, Statista. [de.statista.com/statistik/daten/studie/153098/umfrage/anteil-erneuerbarer-energien-zur-energiebereitstellung/](https://www.statista.com/statistik/daten/studie/153098/umfrage/anteil-erneuerbarer-energien-zur-energiebereitstellung/). Accessed 6 Mar 2019
4. Krylova AY (2014) Products of the Fischer-Tropsch synthesis (a review). *Solid Fuel Chem* 48:22–35. <https://doi.org/10.3103/S0361521914010030>
5. Lappas A, Heracleous E (2016) Chapter 18 - production of biofuels via Fischer-Tropsch synthesis: biomass-to-liquids. In: Luque R, Lin C, Wilson K, Clark J (eds) *Handbook of biofuels production - processes and technologies*, 2nd edn. Elsevier Ltd, Amsterdam, pp 549–593
6. Ail SS, Dasappa S (2016) Biomass to liquid transportation fuel via Fischer Tropsch synthesis - technology review and current scenario. *Renew Sust Energ Rev* 58:267–286. <https://doi.org/10.1016/j.rser.2015.12.143>
7. Dry ME (2002) High quality diesel via the Fischer-Tropsch process - a review. *J Chem Technol Biotechnol* 77:43–50. <https://doi.org/10.1002/jctb.527>
8. Schablitzky HW, Lichtscheidl J, Hutter K, Hafner C, Rauch R, Hofbauer H (2011) Hydroprocessing of Fischer-Tropsch biowaxes to second-generation biofuels. *Biomass Convers Biorefinery* 1:29–37. <https://doi.org/10.1007/s13399-010-0003-x>
9. Leckel D, Liwanga-Ehumbu M (2006) Diesel-selective hydrocracking of an iron-based Fischer-Tropsch wax fraction (C15–C45) using a MoO₃-modified noble metal catalyst. *Energy Fuel* 20:2330–2336. <https://doi.org/10.1021/ef060319q>

10. Blakey S, Rye L, Wilson CW (2011) Aviation gas turbine alternative fuels: a review. *Proc Combust Inst* 33:2863–2885. <https://doi.org/10.1016/j.proci.2010.09.011>
11. Hudson L, Jefferson A, Bauen A, Nattrass L (2016) Chapter 14 – sustainable aviation: a UK roadmap for sustainable aviation fuels. In: *Biofuels for aviation*, pp 315–337
12. de Klerk A (2011) Part IV : synthetic transportation fuels: chapter 13–15. In: *Fischer-Tropsch refining*, pp 249–299
13. de Klerk A (2011) Part V : refining technology: chapter 16-23. In: *Fischer-Tropsch Refining*, pp 301–487
14. IEA Renewables 2018 - Market analysis and forecast from 2018 to 2023 - Power. <https://www.iea.org/renewables2018/power/>. Accessed 6 Mar 2019
15. Mazza A, Bompard E, Chicco G (2018) Applications of power to gas technologies in emerging electrical systems. *Renew Sust Energ Rev* 92:794–806. <https://doi.org/10.1016/j.rser.2018.04.072>
16. Gahleitner G (2013) Hydrogen from renewable electricity : an international review of power-to-gas pilot plants for stationary applications. *Int J Hydrog Energy* 38:2039–2061. <https://doi.org/10.1016/j.ijhydene.2012.12.010>
17. Kraftfahr-Bundesamt Bestand an Pkw in den Jahren 2009 bis 2018 nach ausgewählten Kraftstoffarten. https://www.kba.de/DE/Statistik/Fahrzeuge/Bestand/Umwelt/b_umwelt_z.html?nn=663524. Accessed 6 Mar 2019
18. Groß P, Rauch R, Hofbauer H, Aichernig C, Zweiler R (2015) Winddiesel technology – an alternative to power to gas systems. *Proc EUBCE* 23. doi: <https://doi.org/10.5071/23rdEUBCE2015-ICV.4.17>
19. Jeremiás M, Pohorely M, Svoboda K, Manovic V, Anthony EJ, Skoblia S, Beno Z, Šyc M (2017) Gasification of biomass with CO₂ and H₂O mixtures in a catalytic fluidised bed. *Fuel* 210: 605–610. <https://doi.org/10.1016/j.fuel.2017.09.006>
20. Valin S, Bedel L, Guillaudeau J, Thiery S, Ravel S (2016) CO₂ as a substitute of steam or inert transport gas in a fluidised bed for biomass gasification. *FUEL* 177:288–295. <https://doi.org/10.1016/j.fuel.2016.03.020>
21. Stec M, Czaplicki A, Tomaszewicz G, Słowik K (2018) Effect of CO₂ addition on lignite gasification in a CFB reactor : a pilot-scale study. *Korean J Chem Eng* 35:129–136. <https://doi.org/10.1007/s11814-017-0275-y>
22. Jeremiás M, Pohorely M, Svoboda K, Skoblia S, Beno Z, Šyc M (2018) CO₂ gasification of biomass : the effect of lime concentration in a fluidised bed. *Appl Energy* 217:361–368. <https://doi.org/10.1016/j.apenergy.2018.02.151>
23. Pastor-Pérez L, Baibars F, Le Sache E, Arellano-García H, Gu S, Reina TR (2017) CO₂ valorisation via reverse water-gas shift reaction using advanced Cs doped Fe-Cu/Al₂O₃ catalysts. *J CO₂ Util* 21:423–428. <https://doi.org/10.1016/j.jcou.2017.08.009>
24. Müller S, Groß P, Rauch R, Zweiler R, Aichernig C, Fuchs M, Hofbauer H (2018) Production of diesel from biomass and wind power – energy storage by the use of the Fischer-Tropsch process. *Biomass Convers Biorefinery* 8:275–282. <https://doi.org/10.1007/s13399-017-0287-1>
25. Nikparsa P, Mirzaei AA, Rauch R (2016) Impact of Na promoter on structural properties and catalytic performance of CoNi/Al₂O₃ nanocatalysts for the CO hydrogenation process: Fischer-Tropsch technology. *Catal Lett* 146:61–71. <https://doi.org/10.1007/s10562-015-1620-6>
26. Sabatier P, Senderens JB (1902) New synthesis of methane. *C R Hebd Seances Acad Sci* 134:514–516
27. Fischer F, Tropsch H (1923) Über die Herstellung synthetischer Ölgemische (Synthol) durch Aufbau aus Kohlenoxyd und Wasserstoff. *Brennstoff-Chemie* 4:276–285
28. Steynberg AP (2004) Chapter 1 - introduction to Fischer-Tropsch technology. In: Steynberg AP, Dry ME (eds) *Fischer-Tropsch technology*. Elsevier Science & Technology Books, Amsterdam, pp 1–63
29. Schulz H (1999) Short history and present trends of Fischer–Tropsch synthesis. *Appl Catal A Gen* 186:3–12. [https://doi.org/10.1016/S0926-860X\(99\)00160-X](https://doi.org/10.1016/S0926-860X(99)00160-X)
30. Anderson RB (1956) *Catalysis Vol. IV - hydrocarbon synthesis, hydrogenation and cyclization - chapter 1–3*. Reinhold Publishing Corp, New York
31. de Klerk A (2011) Chapter 4 - Fischer–Tropsch synthesis. In: *Fischer–Tropsch Refining*. Wiley-VCH Verlag & Co. KGaA, Hoboken, pp 73–104
32. Sauciuc A, Abosteif Z, Weber G, Potetz A, Rauch R, Hofbauer H, Schaub G, Dumitrescu L (2012) Influence of operating conditions on the performance of biomass-based Fischer–Tropsch synthesis. *Biomass Convers Biorefinery* 2:253–263. <https://doi.org/10.1007/s13399-012-0060-4>
33. Yong-Wang L, de Klerk A (2013) Chapter 5 - industrial case studies. In: Maitlis PM, de Klerk A (eds) *Greener Fischer-Tropsch processes for fuels and feedstocks*. Wiley-VCH Verlag & Co. KGaA, Hoboken, pp 107–128
34. de Klerk A, Maitlis PM (2013) Chapter 4 - what can we do with Fischer–Tropsch products? In: Maitlis PM, de KA (eds) *Greener Fischer-Tropsch processes for fuels and feedstocks*. Wiley-VCH Verlag & Co. KGaA, Hoboken, pp 81–106
35. Gu B, Khodakov AY, Ordonsky VV (2018) Selectivity shift from paraffins to a-olefins in low temperature Fischer–Tropsch synthesis in the presence of carboxylic acids. *Chem Commun* 54:2345–2348. <https://doi.org/10.1039/c7cc08692j>
36. NULL VK (2006) Patent : WO2006122979 – use of a Fischer-Tropsch derived white oil in food contact applications - Shell Internationale Research Maatschappij BV, pp 1–26
37. Bekker M, Louw NR, Van RVJJ, Potgieter J (2013) The benefits of Fischer-Tropsch waxes in synthetic petroleum jelly. *Int J Cosmet Sci* 35:99–104. <https://doi.org/10.1111/ics.12011>
38. Gruber H, Groß P, Reichhold A, Reinhard R, Abualmfafel R, Zweiler R, Aichernig C, Weber G, Loipersböck J, Binder M (2018) Laboratory scale experiments and evaluation of Fischer-Tropsch - Winddiesel experiments. *Proc 6th Annu Int Conf Chem Chem Eng Chem process - CCECP 2018*. https://doi.org/10.5176/2301-3761_CCECP18.14
39. Zweiler R, Goritschnig M, Kreuter K, Novakovits P, Pomper M, Hofbauer H, Groß P, Rauch R, Aichernig C, Müller S, Bosch K, Haider M (2017) Untersuchung des Lastwechselverhaltens eines Slurryreaktors zur Einkopplung von H₂ und Produktion von FT-Diesel. http://winddiesel.at/images/Downloads/13003-BB006a_Publizierbarer_endbericht_Winddiesel_klienIF.pdf. Accessed 6 Mar 2019
40. Naik SN, Goud VV, Rout PK, Dalai AK (2010) Production of first and second generation biofuels: a comprehensive review. *Renew Sust Energ Rev* 14:578–597. <https://doi.org/10.1016/j.rser.2009.10.003>
41. Kallio AMI, Chudy R, Solberg B (2018) Prospects for producing liquid wood-based biofuels and impacts in the wood using sectors in Europe. *Biomass Bioenergy* 108:415–425. <https://doi.org/10.1016/j.biombioe.2017.11.022>
42. Buchspies B, Kaltschmitt M (2018) A consequential assessment of changes in greenhouse gas emissions due to the introduction of wheat straw ethanol in the context of European legislation. *Appl Energy* 211:368–381. <https://doi.org/10.1016/j.apenergy.2017.10.105>
43. Tsalidis GA, El DF, Korevaar G, Haije W, de Jong W, Kiel J (2017) An LCA-based evaluation of biomass to transportation fuel production and utilization pathways in a large port's context. *Int J Energy Environ Eng* 8:175–187. <https://doi.org/10.1007/s40095-017-0242-8>

44. Cherubini F, Bird ND, Cowie A, Jungmeier G, Schlamadinger B, Woess-Gallasch S (2009) Energy- and greenhouse gas-based LCA of biofuel and bioenergy systems: key issues, ranges and recommendations. *Resour Conserv Recycl* 53:434–447. <https://doi.org/10.1016/j.resconrec.2009.03.013>
45. Hjulær SV, Hansen SB (2018) LCA of biofuels and biomaterials. In: Hauschild MZ, Rosenbaum RK, Olsen SI (eds) *Life cycle assessment*. Springer International Publishing, Cham, pp 755–782
46. European Union (2018) 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 (recast)
47. European Commission. (2015) Directive 2015/1513 of the European Parliament and of the Council of 9 September 2015 amending Directive 98/70/EC relating to the quality of petrol and diesel fuels and amending Directive 2009/28/EC on the promotion of the use of energy from renewable sou
48. Bohlhar-Nordenkamp M, Rauch R, Bosch K, Aichernig C, Hofbauer H (2002) Biomass CHP plant Güssing – using gasification for power generation. *Int Conf Biomass Util*
49. Hofbauer H, Rauch R, Bosch K, Koch R, Aichernig C (2002) Biomass CHP plant Güssing - a success story. In: *Expert meet pyrolysis Gasif biomass waste*, pp 527–536
50. Bolhár-Nordenkamp M, Hofbauer H (2004) Gasification demonstration plants In Austria. IV International Slovak Biomass forum, Bratislava 1:227–230. <https://doi.org/10.13140/2.1.4084.1280>
51. Sauciuc A, Potetz A, Weber G, Rauch R, Hofbauer H, Dumitrescu L (2011) Synthetic diesel from biomass by Fischer-Tropsch synthesis. *Renew Energy Power Qual J* 1:337–342. <https://doi.org/10.24084/repqj09.328>
52. Sauciuc A (2012) Biocombustible materials obtained from biomass using cobalt based catalysts - PhD Thesis Transilvania University of Brasov
53. Gruber H, Groß P, Rauch R, G. Weber, Loipersböck J, Niel J, Wahringer D, Reichhold A, Hofbauer H, Zweiler R, Aichernig C (2017) Fischer-Tropsch synthesis - effects of feedstock load changes regarding product quality and catalyst attrition. *Proc EUBCE* 25. <https://doi.org/10.5071/25thEUBCE2017-3AO.9.4>
54. Rauch R, Kiennemann A, Sauciuc A (2013) Chapter 12 - Fischer-Tropsch synthesis to biofuels (BtL process). In: Triantafyllidis K, Lappas A, Ströcker M (eds) *The role of catalysis for the sustainable production of bio-fuels and bio-chemicals*. Elsevier B.V., Amsterdam, pp 397–443
55. Jensen KL, Menard JR, English BC (2013) Market analysis for Fischer-Tropsch waxes
56. Hannula I, Kurkela E (2013) Liquid transportation fuels via large-scale fluidised-bed gasification of lignocellulosic biomass - online report. <https://www.vtt.fi/inf/pdf/technology/2013/T91.pdf>. Accessed 17 June 2019
57. Thunman H, Gustavsson C, Larsson A, Gunnarsson I, Tengberg F (2019) Economic assessment of advanced biofuel production via gasification using cost data from the GoBiGas plant. *Energy Sci Eng* 7:217–229. <https://doi.org/10.1002/ese3.271>
58. Dutta A, Talmadge M, Hensley J, Worley M, Dudgeon D, Barton D, Groendijk P, Ferrari D, Stears B, Searcy E, Wright C, Hess J (2011) Process design and economics for conversion of lignocellulosic biomass to ethanol - thermochemical pathway by indirect gasification and mixed alcohol synthesis - online report. <https://www.nrel.gov/docs/fy11osti/51400.pdf>. Accessed 17 June 2019
59. Brynolf S, Taljegard M, Grahn M, Hansson J (2018) Electrofuels for the transport sector: a review of production costs. *Renew Sust Energy Rev* 81:1887–1905. <https://doi.org/10.1016/j.rser.2017.05.288>
60. Natarajan K, Leduc S, Pelkonen P, Tomppo E, Dotzauer E (2014) Optimal locations for second generation Fischer Tropsch biodiesel production in Finland. *Renew Energy* 62:319–330. <https://doi.org/10.1016/j.renene.2013.07.013>
61. Bundesministerium für Umwelt Naturschutz und nukleare Sicherheit (BMU) Statistiken Klärschlamm. <https://www.bmu.de/themen/wasser-abfall-boden/abfallwirtschaft/statistiken/klaerschlamm/>. Accessed 21 May 2019
62. PlasticsEurope Kunststoffabfallaufkommen und recycelte Menge Kunststoff in Europa in den Jahren 2006 bis 2016 (in Millionen Tonnen). <https://de.statista.com/statistik/daten/studie/206843/umfrage/kunststoffabfallaufkommen-und-recycelte-menge-kunststoff-in-europa/>. Accessed 21 May 2019
63. Lopez G, Artetxe M, Amutio M, Alvarez J, Bilbao J, Olazar M (2018) Recent advances in the gasification of waste plastics. A critical overview. *Renew Sust Energy Rev* 82:576–596. <https://doi.org/10.1016/j.rser.2017.09.032>
64. Judex JW, Gaiffi M, Burgbacher HC (2012) Gasification of dried sewage sludge: status of the demonstration and the pilot plant. *Waste Manag* 32:719–723. <https://doi.org/10.1016/j.wasman.2011.12.023>
65. Schmid JC, Wolfesberger U, Koppatz S, Pfeifer C, Hofbauer H (2011) Variation of feedstock in a dual fluidized bed steam gasifier—influence on product gas, tar content, and composition. *Spec issue TC. Biomass* 31:205–215. <https://doi.org/10.1002/ep.11607>
66. Kern SJ, Pfeifer C, Hofbauer H (2013) Cogasification of polyethylene and lignite in a dual fluidized bed gasifier. *Ind Eng Chem Res* 52:4360–4371. <https://doi.org/10.1021/ie303453e>

Publisher's note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.