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Power-to-Liquids as Renewable Fuel Option for Aviation: A Review

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Supporting Information
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Severe reductions in the greenhouse gas intensity of aviation fuels are required to get a growing aviation sector on a flightpath compliant with the Paris Climate Agreement. Introducing renewable energy into aviation is challenging. The power-to-liquid (PtL) pathway uses renewable electricity, CO_2, and water to synthesize a sustainable alternative fuel that chemically resembles conventional jet fuel. The state-of-art of key technologies for PtL fuel production as well as the environmental and techno-economic performance of the resulting fuel in comparison with fossil and biomass-derived jet fuel are reviewed.

Keywords: Jet fuel, Life cycle assessment, Power-to-liquids, Sustainability, Techno-economics

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1 Introduction

The target of limiting the increase in the global average temperature “well below 2 °C”, as agreed on in the 2015 United Nations Climate Change Conference (COP 21) held in Paris [1], requires severe reductions in greenhouse gas (GHG) emissions in all sectors [2]. This also includes civil aviation that has defined a set of non-binding targets aiming for climate protection [3], in particular a long-term goal of halving the entire fleet’s annual net CO_2 emissions by 2050 relative to the level of 2005. However, even under optimistic scenarios of technological development, it is expected that future growth in air traffic [4, 5] will outpace the targeted gains in efficiency [3, 6]. Such an emissions gap, i.e., the gap between targeted and projected GHG emissions of the sector, has also been concluded in several scenario studies on future developments in air traffic demand and fuel burn efficiency [7 – 9].

Unless air traffic demand is cut dramatically, this emissions gap can only be closed by transforming the aviation sector’s energy base from fossil to renewable. In this context, so-called “drop-in capable” synthetic liquid hydrocarbon fuels, that physically and chemically resemble conventional jet fuel and that can be used without prior adaption of fuel and combustion systems, represent a viable option for the short to medium-term future.

This understanding has led to an increasingly diverse landscape of production pathways towards renewable jet fuels, defined by different energy sources, types of feedstock and conversion technologies [10]. However, the potential impact of specific renewable fuel options on the aviation sector’s carbon footprint varies strongly. A high potential impact requires that two key criteria are met: Firstly, the considered renewable fuel option has to show strongly reduced lifecycle greenhouse gas emissions, compared to conventional jet fuel. Secondly, the respective pathway has to offer a production potential sufficiently large vis-à-vis (aviation’s) transport fuel demand, specifically in terms of feedstock and energy availability. Additionally, renewable production pathways have to be economically competitive (but not necessarily cost-competitive) with conventional fuels, which is also subject to regulatory boundary conditions.

So far, most attention has been attracted by pathways based on biogenic raw materials (biofuels) [11 – 13]. Despite promising biofuel technologies under development, large-scale utilization of biogenic materials for fuel production generally poses risks regarding land-use change and other sustainability issues [14]. It is questionable, if biofuels can be produced in sufficient quantities to close the aviation sector’s emissions gap in a sustainable way [15, 16], let alone

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to facilitate the energy transition in the entire transport sector. These concerns have given rise to R&D efforts devoted to renewable production pathways that do not rely on biomass as feedstock. Examples of such renewable non-biogenic technologies are solar-thermochemical fuels [17, 18] and power-to-liquids (PtL) [19, 20].

This paper discusses PtL as a sustainable and scalable technology and as enabler for the transition from a fossil to a renewable energy base in civil aviation. Important PtL process steps and technology options are described, in particular the aspect of providing the required renewable energy and the feedstock carbon dioxide (CO_2). Recent progress in technology development and industrial deployment of PtL is presented and environmental as well as techno-economic performance potentials of PtL fuels are discussed.

2 The Power-to-Liquid Process

The three main constituents of PtL are electricity, water and carbon dioxide (CO_2). The generic PtL production process consists of the following key steps as depicted in Fig. 1: First, hydrogen is produced in an electrolyzer (see Sect. 2.1) using renewable electricity and water as educts. Then, hydrogen and CO_2/CO are synthesized to hydrocarbons and converted into specified hydrocarbon target products.

Two main pathways are currently discussed for the production of PtL: the Fischer-Tropsch (FT) pathway (depicted in Fig. 2 and detailed in Sect. 2.2) and the methanol pathway (depicted in Fig. 3 and further detailed in Sect. 2.3).

The state-of-development of key steps and the two PtL production pathways are described in the following sub-

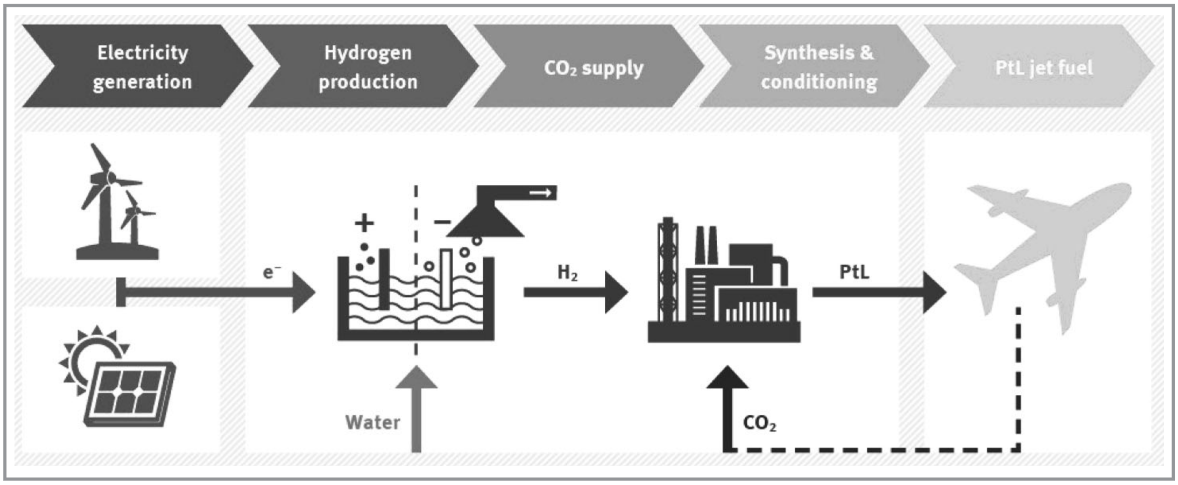

Figure 1. Generic PtL production scheme [19].

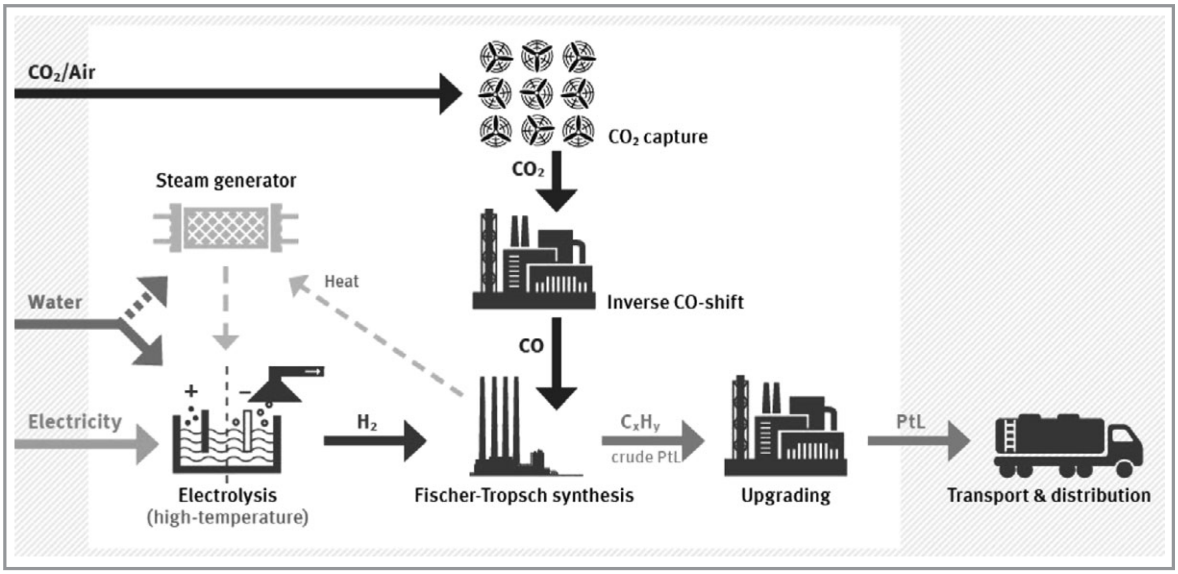

Figure 2. Fischer-Tropsch pathway to produce PtL (incl. the option of using high-temperature electrolysis) [19].

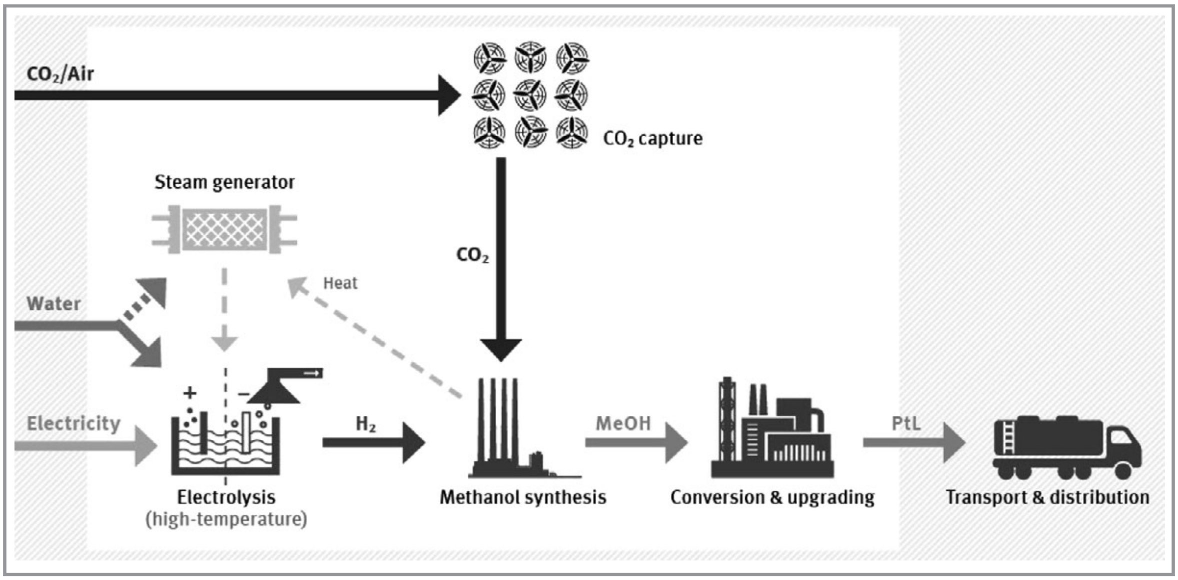

Figure 3. Methanol (MeOH) pathway to produce PtL (incl. the option of using high-temperature electrolysis) [19].

sections. An overview of relevant PtX processes is given in Tab. S1 in the supporting Information.

2.1 Electrolysis

The history of electrolyzer applications dates way back to 1891, when Paul la Cour coupled a windmill with an electrolyzer to store energy as hydrogen [21]. In the late 1980s, German research institution DLR coupled a 350 kW_e electrolyzer directly to a solar power plant in Riyadh, Saudi Arabia. Initiated by Ludwig Bölkow, a 300 kW_e power-to-hydrogen plant for larger-scale integration of a 360 kW photovoltaic power park was opened 1991 in the Bavarian Forest in the context of the *Solar-Wasserstoff-Projekt*.

Historically, electrolyzers once already had been a commercial technology at industrial-scale. In the 1900s in Norway, Egypt, India, and Canada, several large-scale electrolyzer plants with up to 150 MW_e each connected to hydro power supplied over decades hydrogen for synthetic fertilizer production. During the past decades' domination of fossil resources, the main electrolyzer application had been in the kW-scale for on-site supply of hydrogen as generator coolant in thermal power plants.

With the advent of renewable power generation, the industrialization of polymer electrolyte membrane (PEM) fuel cells and the market development of power-to-gas applications, PEM electrolyzers closed up with traditional alkaline electrolysis technology. Today, electrolyzers with few W to many hundred kW per unit capacities are technically proven and commercially available. Products in the lower single-digit MW range have become standard lately. Design concepts and detail engineering for multi-unit power-to-hydrogen production plants up to 60 and 100 MW have been laid out. For example, the electrolyzer manufacturer Nel entered an industrial-scale power-to-gas framework agreement with the French company H2V PRODUCT. The first 100 MW hydrogen plant has a contract value of approximately NOK 450 million or about NOK 4500 per kW of electricity input ($\sim$490 € kW_e^{-1}) [22].

For comparison, for a PtL plant producing 100 kt of jet fuel per year requires some 600 MW_e of installed electrolyzer production capacity, assuming a low-temperature electrolysis system operating 3750 equivalent full load h a^{-1} with an efficiency of 71 % for the electrolysis plant, an efficiency of 75 % for the FT plant (including upgrading), and a lower heating value of 43 MJ kg^{-1} of jet fuel.

Specific investment for uninstalled low-temperature electrolysis has fallen from several thousand € kW_e^{-1} (PEM) to a few hundred € kW_e^{-1} (alkaline and PEM) over the last 10 a [23 – 25]. Further cost reductions can be expected through upscaling of single units and complete system capacities. Similar to photovoltaics or Li batteries, electrolyzers are a surface related technology that can be fabricated in industrial environments, and thereby benefit from typical economies of scale for industrial production. Between 1980 and 2015, PV achieved cost-reduction of 24 % in average with every doubling of PV cumulated installed capacities [26], while battery packs for electric vehicles achieved 6 – 9 % cost-reduction following a cumulative doubling of production [27]. In reference [28] a learning rate of 13 % for PtX plants is assumed. For our estimation of future electrolyzer cost, we apply this learning rate both for alkaline and PEM electrolyzers.

High-temperature (steam) electrolysis uses solid oxide electrolyzer technology (SOEL). Using high-temperature heat, e.g., from the exothermic reaction of FT or methanol synthesis, reduces the electrical energy demand for producing PtL. However, in case of CO_2 extraction using direct air capture via temperature-swing adsorption technology (e.g., from Climeworks), heat is required which reduces the efficiency advantage of high temperature electrolysis. The commercialization of SOEL is in an earlier stage compared to PEM and alkaline electrolysis (see Section 2.5 for current examples).

High-temperature co-electrolysis of water and CO to form hydrogen and CO is in an early stage of research and development. This technology would render the use of reverse water-gas-shift or the development of CO_2-tolerant FT synthesis void (see Sect. 2.2 on the FT pathway).

2.2 Fischer-Tropsch Pathway

FT synthesis was developed in the 1920s by Franz Fischer and Hans Tropsch with the original purpose to produce liquid hydrocarbon fuels from coal. The process is fed by synthesis gas (syngas), a mixture of carbon monoxide (CO) and hydrogen (H_2) that can be generated via gasification of coal, and results in a mixture of saturated hydrocarbons (here indicated as (-CH_2-) unit:

$$CO + 2H_2 \rightarrow (-CH_2-) + H_2O \quad \Delta H_R = -165 \text{ kJ mol}^{-1} \tag{1}$$

The relevance of FT synthesis for PtL and other fuel production pathways is based on the fact that syngas can be generated from virtually any carbonaceous feedstock. Apart from coal, this includes methane (natural gas, flare gas or biogas from fermentation) in the gas-to-liquids (GtL) pathway as well as dry biomass, but also syngas electrochemically generated from CO_2 and water, as in case of PtL.

Although FT synthesis has a long history of industrial application, new production pathways give rise to new developments in FT technology. The main difference between the conventional FT processes and new enterprises is scale: Conventional liquefaction plants have production capacities of tens of thousand barrels per day (bpd). Relying on coal and natural gas as low-cost feedstock of vast local availability, CtL and GtL plants can take advantage of a highly positive effect of economies of scale [29].

For example, Shell's GtL facility located in Bintulu, Malaysia, has a production capacity of 14 700 bpd [30, 31]. For a PtL plant, this corresponds to annual electricity consumption for the electrolysis alone of about 17.5 TWh, or 3 % of the 2015 German electricity demand of 521 TWh [32], and requires an upstream electrolysis plant of above 2 GW installed power. However, such a scale is not realistic for early PtL projects as the investment into individual facilities would be associated with tremendous financial risks. Similar challenges exist for other emerging FT-based pathways that utilize biomass, waste streams or limited resources of natural gas, where the local feedstock availability is insufficient to supply a large-scale FT unit or the collection of sufficient quantities would impose prohibitively high cost.

In order to enable implementation of new value chains, various companies are developing FT reactor systems of substantially reduced production capacities. An overview of developing small-scale FT technologies can be found in references [33, 34].

Several of the industrial small-scale FT projects are based on a microstructured reactor design. Such reactors consist of parallel arrays of microchannels, with channel diameters typically ranging from 0.1 to 5 mm, about 1 to 2 orders of magnitude smaller than conventional reactor tubes [35]. The reactor channels are interleaved with coolant channels.

The small diameter of microchannels leads to increased surface area in relation to the reactor volume, strongly enhancing heat transfer and improving temperature control. This is particularly important for strongly exothermic processes, such as FT synthesis. Moreover, mass transfer is substantially improved. Therefore, microchannel FT reactor systems enable a process intensification through reduced specific reactor volume (volume per production capacity), process acceleration by a factor of 10 to 1000 and increase of catalyst productivity (formed product quantity per hour per catalyst volume) [35]. This process intensification holds the potentials of cost-effective production at substantially smaller scales compared to conventional FT units.

Examples of companies driving the commercialization of microchannel FT technology are Velocys Inc. and Ineratec GmbH. While Velocys' commercialization activities are currently focused on GtL [36] and BtL [37] projects, Ineratec's FT technology has recently been deployed in a PtL demonstration plant in Finland as part of the Soletair project [38] (Fig. 4). This project aims at demonstrating the complete PtL process chain, including renewable electricity generation, hydrogen production through PEM electrolysis, CO_2 provision from direct air capture (DAC), RWGS for CO generation, FT synthesis and refining to fuel products.

Figure 4. Chemical synthesis unit of the Soletair project with Ineratec's FT reactor (Copyright by Ineratec/VTT).

An important advantage of FT-based fuel synthesis lies in the excellent suitability of the produced fuels for applications in various transport sectors. With respect to aviation, FT-based synthetic jet fuel has been approved for use in commercial aviation in blends of up to 50 % with conventional jet fuel [39]. Importantly, this approval is not restricted to certain types of feedstock, but valid for all FT-based pathways, as the long as the fuel is "wholly derived from synthesis gas via the Fischer-Tropsch (FT) process using Iron or Cobalt catalyst" and subsequently refined to meet the defined specifications [39].

2.3 Methanol Pathway

An alternative to FT synthesis as liquefaction process is the production of methanol as intermediate product. Today, methanol is industrially produced from synthesis gas, typically generated from natural gas or coal, using the ternary Cu-ZnO-Al_2O_3 catalyst according to the following equation [40]:

$$CO(g) + 2H_2(g) \rightarrow H_3COH(l) \quad \Delta H^0 = -131.6\ \text{kJ mol}^{-1} \tag{2}$$

The reaction is significantly accelerated in the presence of small quantities of CO_2 of about 10 % in the feed stream [41]. Recent research efforts focus on the development of catalysts supporting direct hydrogenation of CO_2, without requiring prior RWGS reaction to generate CO [42]:

$$CO_2(g) + 3H_2(g) \rightarrow H_3COH(l) + H_2O \quad \Delta H^0 = -137.8\ \text{kJ mol}^{-1} \tag{3}$$

Direct conversion of CO_2 poses several technical challenges, particularly with respect to required pressures that often exceed 30 MPa for promising reaction performance [43]. Nevertheless, the company Carbon Recycling International (CRI) operates a commercial methanol production plant based on direct reaction of CO_2 with electrolytically generated hydrogen [44]. The CRI facility in Iceland is operational since 2012 and yields 4000 t of methanol per year [45].

Methanol is a versatile raw material for the chemical industry and represents an important commodity with an annual production of close to 80 Mt in 2016 [46] and, correspondingly, large markets. It is also of importance in the production of transport fuels, such as methyl ethers (e.g., dimethyl ether (DME)) or biodiesel. Methanol can also be used as blendstock in gasoline [47], and direct use of pure methanol as marine fuel is tested in commercial pilot projects [48].

An interesting option, especially from the aviation sector's point of view, is the conversion of methanol into liquid hydrocarbon products that are potentially suitable as synthetic jet fuel. In addition to the more commonly known methanol-to-gasoline (MtG) process that is currently deployed in several commercial plants, Mobil has also demonstrated the conversion of methanol into middle distillate fuels (diesel and kerosene).

Both, the MtG process as well as production of distillate fuels from methanol, rely on shape-selective zeolite catalysts [49], particularly ZSM-5 zeolite catalyst [50]. The distillate fuels pathway (depicted in Fig. 5) comprises the conversion of methanol into light olefins and the subsequent oligomerization of light olefins with mild hydrotreatment and fractionation. The process yields products in the distillate as well as in the gasoline range and can be adjusted to shift yields of the desired fraction (distillate or gasoline) to above 80 % [50].

Both processes, methanol to olefin conversion and subsequent distillate synthesis, have been demonstrated in Mobil facilities in the 1980s with favorable reported yields, efficiencies and selectivities [49, 50]. Analysis of the quality of the distillate fractions showed that selected requirements of diesel and jet fuel specifications are fulfilled [49]. The process also generates aromatic compounds, rendering Mobil's methanol to distillate pathway potentially suitable for producing 100 % drop-in capable synthetic jet fuel, as a minimum content of 8 % aromatics is required according to current fuel standard ASTM D7566. But Mobil's methanol-to-distillate process has not yet been commercialized and the methanol pathway has not undergone the American Society for Testing and Materials (ASTM) approval procedure. Hence, more comprehensive analyses are required to assess the drop-in quality for diesel or jet fuel applications, even though preliminary results [49] do not give rise to expect a lesser suitability than in case of other already approved synthetic jet fuels.

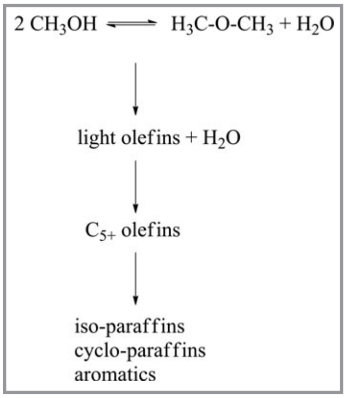

Figure 5. Basic process scheme of Mobil's distillate fuels production from methanol: Light olefins form in a mixture of methanol and dimethylether (DME), further growing to higher olefins, followed by oligomerization to higher branched or cyclic species and mild hydrotreatment, yielding branched (iso) and cyclic paraffins as well as aromatic compounds [50].

2.4 CO_2/CO Supply

There are three major groups for sourcing concentrated CO_2, namely from (i) fossil origin, such as flue gas of coal, oil or gas firing units and industrial processes, such as blast furnace gas, coke-oven gas, calcination of limestone; (ii) mineral origin like geothermal sources and cement production; (iii) renewable origin, such as biogas upgrading, breweries, ethanol plants, flue gas from biogas/solid biomass combustion, and extraction from air.

The extraction of CO_2 from concentrated sources or the air can be conducted via, e.g., scrubbing with amines (state-of-the-art technology for CO_2 separation from concentrated sources) [51, 52]; scrubbing with potassium or sodium hydroxide and subsequent electrodialysis (mainly proposed for direct air capture of CO_2) [53 – 55]; or temperature swing adsorption (mainly developed for direct air capture of CO_2) [56].

For the cost and efficiency calculations in this paper, direct air capture via temperature swing adsorption/desorption (TSA) is taken into account with regeneration of the sorbent using low temperature heat (95 °C). The technical and economic data have been derived from the Swiss company Climeworks [57].

CO_2 liquefaction and buffer storage is considered to purify the CO_2 feed and to support decoupling the (fluctuating) renewable electricity supply from PtX production.

FT synthesis requires CO and hydrogen as reaction educts. CO_2, thus, has to be converted into CO via reverse water-gas-shift (RWGS). Today, this process is established at smaller scales only, and scale-up of the RWGS to industrially relevant capacities is needed to facilitate commercial PtL production. Alternatively, co-electrolysis of water and CO_2 to directly form H_2 and CO is a technology option being researched [58]. For the synthesis of methanol as primary PtL product, CO and CO_2 can be both applied as reactants (see Sect. 2.3). Sustainability aspects associated with different CO_2 sources are discussed in Sect. 5.

2.5 PtX Pilots, Demonstration and Up-Scaling

To-date, no complete power-to-jet fuel pathways has been demonstrated yet. However, PtL system integration is progressing in Iceland [59], Finland [60], Germany [61], and Norway [62].

In Tab. S1 an overview of selected current PtX upscaling and industrialization projects is given. The examples are depicted by technologies/process steps that are at the same time key enablers for the development towards industry-scale power-to-liquids production pathways.

To-date, the majority of power-to-x plants are located in Europe, especially in Germany, although PtX is also gaining momentum internationally as a vector for bulk energy imports and exports [63 – 65]. As per October 2017, there are more than 50 PtX pilot and demonstration projects worldwide in operation or under construction [66]. About 40 projects are currently in operation in Europe [67], thereof more than 20 power-to-gas (H_2, CH_4) research and pilot plants in Germany [68]. The electrolyzer is at the heart of PtX pathways. Concept designs and basic engineering for two to three-digit multi-megawatt power-to-hydrogen plants are presented by industry stakeholders like Areva H_2Gen [69], Hydrogenics [70], ITM [71], Nel [72], Siemens [73], among others.

3 Techno-Economic Performance

In this chapter, the current and long-term costs for the production of PtL jet fuel 'well-to-tank' from renewable sources are considered. Tab. 1 shows the technical and economic data for PtL fuel production based on today's cost data supplied by manufacturers. Today, there are only a few small-scale PtL plants operational that generate liquid hydrocarbon products, including kerosene. However, the key process steps of PtL production chains, most notably water electrolysis, hydrogen storage vessels, FT and methanol synthesis and refining technologies, are already mature and industri-

Table 1. Techno-economic data for PtL fuel production plant with low temperature electrolysis today.

Today		Methanol pathway		Fischer-Tropsch pathway	
CO_2 source		Direct air capture	Concentrated source	Direct air capture	Concentrated source
Technical key data					
Electricity input [MW]		76	60	73	60
Fuel output	$[MW_{LHV}]$	29	29	28	28
	$[t\,h^{-1}]$	2.4	2.4	2.4	2.4
	$[kt\,yr^{-1}]$	10	10	9	9
Efficiency [%]		39	48	39	47
Investment					
Electrolysis (low temp.) [M€]		28	28	28	28
H_2 storage [M€]		3	3	30	30
CO_2 supply [M€]		55	8	55	8
Synthesis & conditioning [M€]		19	19	18	18
Total [M€]		105	58	131	84
Specific costs					
Jet fuel	$[€\,GJ_{LHV}^{-1}]$	95.0	71.0	97.7	75.2
	$[€\,t^{-1}]$	4100	3062	4215	3245
Sensitivity (optimistic [a]) [%]		–28	–30	–27	–28
Sensitivity (pessimistic [b]) [%]		+4.1	+5.5	+4.0	+5.2

[a] Electricity costs: 6.6 cent kWh^{-1} instead of 10.3 cent kWh^{-1}. [b] Investment electrolyzer, synthesis & conditioning: +50 %.

ally applied. Only the RWGS reactor has yet to be demonstrated at relevant scale and is currently tested at Sunfire in Dresden [74].

The investment for the methanol synthesis, MtSynfuels plants, and the FT synthesis plants have been derived from biomass-to-liquid (BtL) plants with a capacity of some 100 kt a^{-1} based on data from various manufacturers. A scaling exponent of 0.7 has been applied for down- or upscaling. For today it has been assumed that the capacity per unit is one tenth of that in 2050. The CO_2 supply consists of the DAC plants and the CO_2 liquefaction and storage. In case of CO_2 from concentrated source it has been assumed that the CO_2 comes from an existing biogas plant with biogas upgrading. As a result, only investment for CO_2 compression, liquefaction and storage is required. The electricity generation costs have been derived from a mix of renewable energy sources plus electricity transport and distribution.

The projected costs of PtL fuel production via the FT and methanol pathway are presented in Tab. 2, along with underlying process parameters and assumptions. The cost for renewable electricity is assumed to be 40 € MWh_e^{-1} for an equivalent full-load period of 3750 $h_{eq}a^{-1}$. If a successful commercialization of high temperature electrolysis was assumed, the resulting costs could be slightly lower (still within the error bar range) compared to the low temperature case depicted in Tab. 2.

The cost of renewable power is the main driver for the overall PtL production costs [19, 57, 75, 76]. Consequently, the efficiency of the power-to-fuel conversion is crucial for the overall economic performance. Increasing efficiencies are a key objective of R&D efforts dedicated to PtL technologies. The potentials for PtL conversion efficiencies for several process configurations are presented in Tab. 2 in comparison to the state-of-the-art efficiencies achievable today in Tab. 1. The efficiency mainly depends on the efficiency of the electrolysis plant and the CO_2 source. The heat from the exothermal synthesis reaction can be used for heat supply for the temperature swing adsorption plant for DAC and in case of high temperature electrolysis for the heat demand of the SOEL plant. Efficiencies of up to 63 % could be realized in a PtL process with high-temperature electrolysis if concentrated CO_2 from renewable point sources are used and up to 46 % with CO_2 from DAC [19].

Today's efficiency figures can be robustly achieved using available technologies at smaller scales [19, 77]. For bulk PtL jet fuel production, improved production efficiencies and reduced capital costs are beneficial, i.e., further optimize low-temperature electrolysis, adapt the Mobil process to a methanol-to-jet fuel conversion and industrialize high-temperature (co-)electrolysis and the RWGS process.

The short-term costs of sustainable alternative jet fuel production (including PtL jet fuel) exceed the current cost of conventional jet fuel [19, 20, 75]. With PtL jet fuel from renewable sources, the avoidance of external costs for, e.g., climate change are priced-in (except aviation's climate impacts from high-altitude emissions [78]) while the costs of environmental impacts from fossil jet fuel use are externalized. Economies of scale and volume production are important elements to close the cost gap; another approach is using PV-wind-hybrid renewable power plants to increase the capacity factor for PtL production ([75] based on analyses of Lappeenranta University). Policy frameworks and support instruments will be needed to address the (initially

Table 2. Techno-economic data for PtL fuel production plant with low temperature electrolysis 2050 [19].

2050		Methanol pathway		Fischer-Tropsch pathway	
CO_2 source		Direct air capture	Concentrated source	Direct air capture	Concentrated source
Technical key data					
Electricity input [MW]		760	594	729	588
Fuel output	[MW_{LHV}]	319	319	310	310
	[t h^{-1}]	26.6	26.6	25.8	25.8
	[kt yr^{-1}]	100	100	97	97
Efficiency [%]		42	54	42	53
Investment					
Electrolysis (low temp.) [M€]		140	140	140	140
H_2 storage [M€]		3	3	30	30
CO_2 supply [M€]		359	45	359	45
Synthesis & conditioning [M€]		100	100	94	94
Total [M€]		602	288	622	308
Specific costs					
Jet fuel	[€ GJ_{LHV}^{-1}]	39.8	28.0	42.7	31.3
	[€ t^{-1}]	1719	1206	1841	1352
Sensitivity (optimistic[a]) [%]		–44	–43	–41	–39
Sensitivity (pessimistic[b]) [%]		+4.9	+7.0	+4.6	+6.2

[a] Electricity costs: 2.1 cent kWh^{-1} instead of 4.0 cent kWh^{-1} based on cost data [91] and an interest rate of 4 %. [b] Investment electrolyzer, synthesis & conditioning: +50 %.

substantially) higher specific costs of renewable jet fuel and bridge the economic valley of death [19, 79, 80].

4 Environmental Performance

Key performance indicators for the environmental assessment of alternative fuels are GHG emissions, water demand, and land requirement. Most importantly, specific GHG emissions of PtL fuels can be close to carbon neutral and substantially below fossil and most biogenic fuel options (Tab. 3).

The GHG balance of biomass-based fuels is highly sensitive to the production of the biogenic feedstock, especially with regards to direct and indirect land-use change. Even if conservatively assuming today's fossil-dominated processes and energy mixes for material sourcing and construction of facilities and power plants, the specific GHG emissions of PtL jet fuel are reduced by at least 70 % compared to the conventional jet fuel reference. In a world increasingly using renewable power, the GHG balance of renewable PtL jet fuel can be expected to develop towards a > 95 % reduction.

Furthermore, being of synthetic origin, PtL jet fuel burns cleaner than conventional kerosene, thereby reducing volatile and non-volatile particle emissions, as has been shown in studies on synthetic jet fuels produced from biogenic feedstock by NASA, DLR and others [81 – 83].

The critical issue of water consumption associated with energy conversion processes is vividly discussed as water-energy nexus [84 – 87]. A widely accepted metric to quantitatively assess water consumption along process chains, the concept of water footprint has been developed and laid down as ISO 14046.

In case of biofuels, substantial quantities of water are consumed for biomass cultivation, e.g., through evapotranspiration of plants and water run-off from fields. In PtL fuel production, however, water is mainly needed as hydrogen source in the water electrolysis step, where it is needed as feedstock in quantities that are defined by the reaction stoichiometry of the fuel synthesis. The water demand for PtL fuel production is listed in Tab. 4 in comparison to several biogenic fuels. For both, the methanol and FT pathway, product water from synthesis and upgrading processes is used as input for the water electrolysis.

The water footprint of biofuels depends on the cultivated species, agricultural practices, local climatic conditions and soil properties. Nevertheless, the data clearly show that the water footprint of PtL fuel is by several orders of magnitude, i.e., by a factor of 400 to 15 000, lower than in case of biofuels.

The PtL water footprint translates into a net water consumption of 170 000 m^3 water per year for a production of 100 $kt_{PtL}a^{-1}$. Even though this can be considered negligible compared to water demands of biofuel production, local water availability and supply options are relevant aspects that are typically addressed in local environmental impact assessments for plant approval.

Table 3. Specific greenhouse gas emissions of PtL fuel in comparison to various fossil and biogenic jet fuel options [19].

Jet fuel pathway	GHG emissions without land-use change [$g_{CO2eq}MJ_{fuel}^{-1}$]	GHG emissions including direct land-use change [$g_{CO2eq}MJ_{fuel}^{-1}$]
Crude oil (reference)	87.5	–
Crude oil (ultra-low sulfur)	89.1	–
Oil sand (e.g., Canada)	103.4	–
Oil shale (in situ)	121.5	–
Natural gas (GtL)	101.0	–
Coal (CtL)	194.8	–
Switchgrass (BtL)	17.7	–2.0[a]
Soybean oil (HEFA)	37	97.8 – 564.2
Palm oil (HEFA)	30.1	39.8 – 698.0
Rapeseed oil (HEFA)	54.9	97.9
Jatropha oil (HEFA)	39.4	–
Algae oil (HEFA)	50.7	–
PtL (wind/PV in Germany, renewable world embedding)	~1	–
PtL (wind/PV in Germany, today's energy landscape in material sourcing and construction)	11 – 28[b]	–

[a] Negative value because soil carbon from former vegetation is lower compared to soil carbon for switchgrass. [b] Including construction of power plants and production facility (today).

Table 4. Water demand for biofuel and power-to-fuels production. Colum 2: Green water = Precipitation on land that is stored in the vegetation, in the soil, or stays on top of the soil. Colum 3: Blue water = Water consumption from surface and groundwater.

Feedstock/pathway	Green water $[m^3GJ^{-1}]$	Blue water $[m^3GJ^{-1}]$	Sum $[m^3GJ^{-1}]$	Sum $[l_{H2O}l_{jet\text{-}fuel\text{-}eq}{}^{-1}]$	Reference
Jatropha oil	239	335	574	19914	Weighted global Ø [92]
Palm oil	150	0	150	5204	Weighted global Ø [92]
BtL from poplar[a]	107	6	112	3892	[93]
Algae oil (open pond with water recycling)[a)]	0	14	14	497	[94]
Algae oil (open pond w/o water recycling)[a)]	0	53	53	1839	[94]
PtG hydrogen (wind, PV)	0	0.076	0.076	2.63	[19]
PtL via FT pathway (wind, PV, CSP[b)])	0	0.040	0.040	1.38	[19]
PtL via methanol pathway (wind, PV, CSP[b)])	0	0.038	0.038	1.33	[19]

[a)] In moderate climate zones, e.g., Europe, Northern USA, Southern Canada. [b)] Concentrated solar power via solar-thermal steam turbine with dry cooling system.

Land demand represents another performance indicator of environmental and social relevance. Tab. 5 depicts the specific gross area demand and achievable air mileage from various feedstocks. As can be clearly seen, the area-specific fuel yield of PtL is generally high and superior to the yields achieved with biofuels. Importantly, this comparison is drawn based on the gross area demand, with PV and especially wind power having substantially lower land coverage than agricultural biomass production (land coverage near 100 %). This means that especially in case of wind power, the occupied land can still be used for other purposes.

It is also important to acknowledge that it is not only the amount of land area required for production that has to be considered, it is also the type of land. Renewable power generation in principle does not depend on arable land, with desert regions, e.g., offering highly suitable conditions for photovoltaic or solar-thermal power generation. Consequently, the risk of competition between energy and food production is strongly reduced.

Table 5. Area-specific yield and achievable air mileage related to gross area [19].

Production pathway	Jet fuel yield[a)] $[GJ\,ha^{-1}a^{-1}]$	Achievable air mileage[b)] $[km\,ha^{-1}a^{-1}]$
Jatropha oil (HEFA)	15 – 50	124 – 425
Palm oil (HEFA)	162	1379
Algae oil (HEFA)	156 – 402	1327 – 3422
Short rotation forestry (BtL)	47 – 171	398 – 1456
PtL (photovoltaic electricity)	580 – 1070[c)]	4950 – 9080
PtL (wind electricity)	470 – 1040[d)]	4040 – 8860

[a)] Bandwidth resulting from moderate vs high-yielding production locations and CO_2 sources available. [b)] Assuming an Airbus A320neo with a specific air mileage of 0.37 km $kg_{jet\text{-}fuel}{}^{-1}$. [c)] Includes working space and distance between panel rows to avoid (partial) shading. [d)] Most of the area can still be used for other purposes because only some 2.6 – 3.4 % of the area is actually covered (5500 m^2 for foundation, working space and access roads related to a gross land area of 163 216 – 211 600 m^2 per wind turbine).

5 Requirement of Renewable Sources of Electric Energy and CO_2

The environmental analysis shows that both, renewable electricity generation and renewable CO_2, are necessary for sustainable PtL fuels. While the requirement of renewable electricity is generally accepted, the public debate about the environmental performance of different sources of CO_2 for PtL fuels (or generally of fuels from processes based on CO_2 as feedstock) is still ongoing. The need for renewable electricity and renewable CO_2 for synthetic fuel production through processes using CO_2 as feedstock was rigorously shown by life cycle assessment (LCA) analyses [88, 89] and is quantitatively shown here in a straightforward way on the basis of general energy and mass balances.

As a starting point the specific CO_2 emissions resulting from the combustion of liquid hydrocarbon fuels, termed as ($-CH_2-$) for simplicity are quantified:

$$2(-CH_2-) + 3O_2 \rightarrow 2CO_2 + 2H_2O \qquad (4)$$

The combustion of 1 kg_{-CH2-} (14 amu) results in the emission of about 3.14 kg_{CO2} (44 amu). Jet fuel has an average specific energy content of about 43.2 MJ kg^{-1}, thus the specific CO_2 emission (CE) from fuel combustion are about CE_{fuel} = 262 $g_{CO2}kWh_{fuel}{}^{-1}$. To quantify tolerable levels for the specific carbon emissions of the electricity feedstock a break-even condition is defined by multiplying with the electricity-to-fuel energy conversion efficiency:

$$\mathrm{CE}_{\mathrm{breakeven}} = \mathrm{CE}_{\mathrm{fuel}}\eta_{\mathrm{el}\rightarrow\mathrm{fuel}} \tag{5}$$

For typical PtL energy conversion efficiencies $\eta_{\mathrm{el}\rightarrow\mathrm{fuel}}$ (Tabs. 1 and 2) specific break-even emissions range from about 100 $\mathrm{g_{CO2}kWh_{el}^{-1}}$ to 140 $\mathrm{g_{CO2}kWh_{el}^{-1}}$. A deep reduction of GHG emissions compared to conventional fuels is only possible, if the specific emissions of the electricity input are significantly lower. However, typical specific CO_2 emissions of fossil electricity generation are much higher, ranging from 800 – 1300 $\mathrm{g_{CO2}kWh_{el}^{-1}}$ for coal-fired plants and 380 – 600 $\mathrm{g_{CO2}kWh_{el}^{-1}}$ for power plants fueled by natural gas [90]. Therefore, PtL-based fuel production from fossil-derived electric energy is highly unsustainable and even few percent of fossil share in the electricity mix can spoil the GHG balance of PtL fuels.

Now, the impact of the CO_2 source on the GHG balance of PtL fuel production is discussed based on the energy and mass balance of an exemplary process chain using atmospheric CO_2 in comparison to the utilization of fossil CO_2 captured from a coal-fired power plant (Fig. 6). For generality, any carbon loss along the production chain is neglected. Clearly, the carbon cycle can only be closed using atmospheric CO_2, while fossil derived CO_2 inevitably accumulates in the atmosphere and contributes to the greenhouse gas effect. The overall CO_2 emissions need to be allocated between both product streams (177 $\mathrm{kJ\,mol_C^{-1}}$ electricity and 602 $\mathrm{kJ\,mol_C^{-1}}$ fuel) to prevent double counting. Choosing energy allocation, about 77 % of the overall emissions need to be attributed to the fuel product, and 23 % to the electricity output, leading to about 56 $\mathrm{g_{CO2}MJ_{fuel}^{-1}}$ resulting from final fuel combustion alone. Accounting for additional contributions in a proper LCA (e.g., fugitive CO_2 emission during flue gas capture or gas-to-liquids conversion) can easily result in specific GHG emissions that exceed that of conventional fuel. Furthermore, the specific renewable electricity consumption (> 1000 $\mathrm{kJ\,mol_C^{-1}}$) greatly exceeds the corresponding conventional electricity generation (177 $\mathrm{kJ\,mol_C^{-1}}$); consequently, it is much more effective to reduce overall emissions by substituting the fossil power plant with renewable electricity. Thus, PtL fuels production with a coal-fired power plant as a CO_2 source is not sustainable due to a very unfavorable energy balance that arises from the mass balance along the fuel production chain.

The above arguments are tailored to coal power plants. An in-depth analysis of all major CO_2 sources is beyond the scope of this manuscript, but basic trends if to achieve long term carbon-neutrality [2] are indicated in Tab. S2: PtL fuels produced from fossil CO_2 sources are unsustainable in the long run, because CO_2 from transportation fuel combustion accumulates in the atmosphere. Biogenic sources are a pragmatic choice for early PtL projects as CO_2 can be recovered at competitive cost from fermentation processes (biogas upgrading, ethanol production). In the long term, the availability of biogenic CO_2 sets severe limits to the potential scale of PtL fuels production. Thus, truly sustainable PtL fuel production at the scale of future fuel demand requires the development of CO_2 extraction from air.

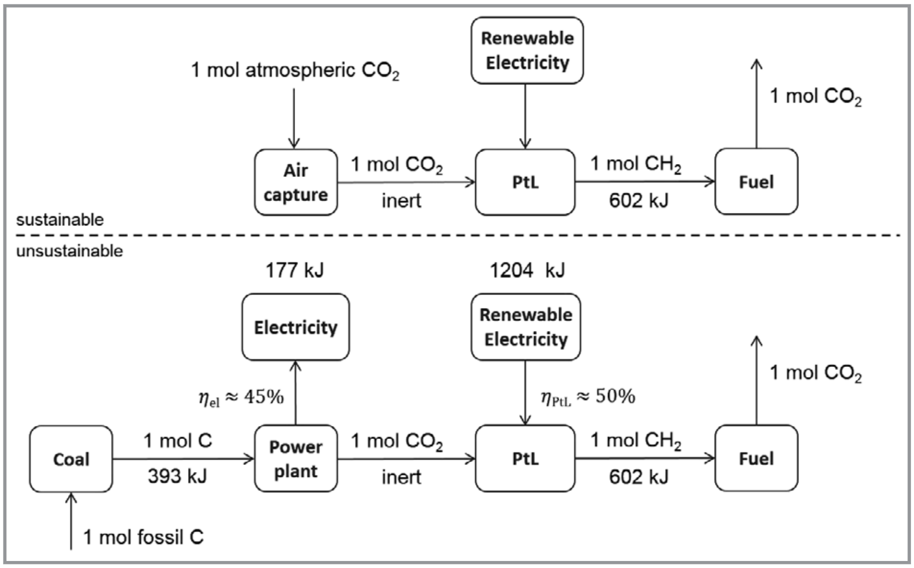

Figure 6. Basic process scheme for 1 $\mathrm{mol_C}$ atoms along a fuel production chains in a mass and energy balance, zero carbon loss is assumed for clarity. Utilization of atmospheric CO_2 closes the carbon cycle, while utilization of fossil CO_2 contributes to GHG accumulation in the atmosphere. The specific energy content of the product streams (602 $\mathrm{kJ\,mol_C^{-1}}$ fuel vs 177 $\mathrm{kJ\,mol_C^{-1}}$ electricity) call for an allocation of total emissions towards the fuel product. The renewable electricity demand for fuel synthesis (1204 $\mathrm{kJ\,mol_C^{-1}}$) is much larger than the conventional electricity output.

6 Conclusions

The power-to-liquid pathway offers a viable option for the transition of the aviation sector's energy basis from fossil to renewable, using existing aircraft systems. PtL fuels can potentially meet future demand and offer profound benefits in terms of resource demand (land and water) and greenhouse gas emissions. It should be noted that when continuing burning (alternative sustainable) jet fuels, complementary options are required to address aviation induced climate effects from, e.g., high-altitude and black carbon emissions. Furthermore, all key technologies along the PtL fuel production pathway have already achieved sufficient technological readiness to proceed with process integration and industrial scale-up.

The main requirement towards large-scale implementation is a continued cost reduction of renewable hydrogen production from water electrolysis powered by solar and wind energy. On that front, recent interest in various power-to-x schemes led to a steep growth in the number of renewable electrolysis projects at multi-MW scale to drive down electrolysis cost. So far, further processing of renewable hydrogen to kerosene-range PtL fuels is limited to few pioneering research and development projects at small product volumes. Promising PtL options include the Fischer-Tropsch synthesis as liquefaction step, converting syngas into hydrocarbon products. Alternatively, methanol can be generated as liquid intermediate product, to be subsequently converted into hydrocarbon fuels, including jet fuel.

Further development of PtL fuel production will require industrial projects, ASTM approval in case of the methanol route, and appropriate sustainability safeguards to ensure the use of electricity and CO_2 from renewable sources via a monitoring, reporting and verification (MRV) system.

Looking at the tremendous gap between aviation fuel use and environment targets, it is high time for the sector to engage in the development and ramp-up of PtL jet fuel production using renewable sources. Effective regulatory frameworks have to be installed to enable economic competitiveness and, thus, large-scale uptake of sustainable jet fuels.

This paper builds on results from several research studies financed by the German Ministry for Transport and Digital Infrastructure (BMVI) in the context of the Mobility and Fuel Strategy of the German Government, the German Environment Agency (UBA), the Research Association for Combustion Engines (FVV), and the German Association of the Automotive Industry (VDA).

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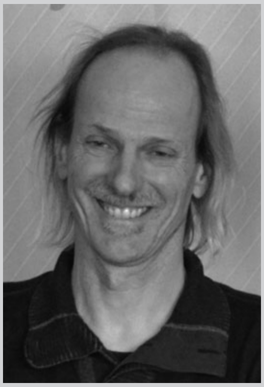

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Symbols used

CE	$[g_{CO2}kWh_{fuel}^{-1}]$	CO_2 emission

Abbreviations

ASTM	American Society for Testing and Materials
BtL	biomass-to-liquid
CRI	Carbon Recycling International
DAC	direct air capture
FT	Fischer-Tropsch
GHG	greenhouse gas
GtL	gas-to-liquids
LCA	life cycle assessment
MRV	monitoring, reporting and verification
MtG	methanol-to-gasoline
PEM	polymer electrolyte membrane
PtL	power-to-liquids
RWGS	reverse water-gas-shift
SOEL	solid oxide electrolyzer technology
TSA	temperature swing adsorption/desorption

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