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The Path to Sustainable Aviation

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1. Introduction

In the field of carbon emissions mitigation, it is now widely recognised that aviation is one of the key hard-to-abate areas. Decarbonisation via direct electrification, particularly for long-haul flights, is not a practical option using batteries, and this sector of the aviation industry must rely on the use of high-energy density jet fuels. With this in mind aviation is gradually assuming a leading role, and will continue to do so in the decades ahead, in global efforts to decarbonise human activities. The last ten to twenty years have seen a surge in research and development of technologies towards the production of man-made carbon-based high-energy density jet fuels. These products, more generally known as **Sustainable Aviation Fuels** (SAFs), are hydrocarbons which fall in the kerosene carbon range C_8-C_{16}.

Currently the technological development of SAFs has proceeded along two primary lines of action: bio-SAF and electro or e-SAF. The former relates to utilisation of bio-resources including cooking and plant oils (leading to hydro-processed esters and fatty acids (HEFA)), agricultural waste, municipal solid waste, woody biomass and dedicated energy crops. A fundamental requirement in the use of these resources is that they do not impede or influence primary agricultural needs (notably food). In a recent IATA report (1) it has been acknowledged that, at most, approximately 60% of SAF needs will be accounted for by the bio-route by mid-century and that the remaining 40% (e-SAF sourced through Power-to-Liquid or PtL) will need to come from synthetic processes involving green hydrogen (produced primarily by electrolysis of water) and CO_2 derived from carbon capture.

While the carbon required for SAF production is plentiful, a hard-ceiling faced by biogenic CO_2 arises due to:

(a) Limited feedstock availability, competing land use and seasonal variability;

(b) The carbon required is not necessarily in the same location where SAF is to be produced and,

(c) As already alluded to, an increase in biogenic carbon for SAF purposes cannot arise without placing pressure on agrifood production.

The focus of this chapter will be on e-SAF and hence the utilisation of non-biogenic carbon whose origins primarily include (i) point source emissions from power generation and industry and (ii) distributed domestic, commercial and transport emissions. Both of these sources have their disadvantages and advantages:

Point source CO_2

(a) Limitations on gas purity

(b) Require transport to the location of e-SAF production

(c) With the drive to net-zero, it is expected that point source emissions will fall significantly in the coming decades.

(d) The primary advantage of point source CO_2 capture at this time is its relative ease and hence moderate cost.

Distributed current and legacy atmospheric CO_2 emissions:

(a) Because of its dilution (430 ppm) CO_2 can be costly to remove from the atmosphere via direct air capture (DAC).

(b) A major advantage however is that DAC can be located directly at the site of e-SAF production.

(c) The captured atmospheric CO_2 is very pure.

In the following we will focus only on those e-SAF technologies which will require development in the coming decades to utilise purified CO_2 (sourced primarily from atmospheric emissions) and H_2 derived via electrolysis of water (e-H_2 or green Hydrogen). These technologies have been summarized by Neto et al (2) and include:

(1) Indirect CO mediated hydrocarbon production: CO is produced via the Reverse Water Gas Shift (RWGS) reaction followed by Fischer-Tropsch Synthesis (FTS):

$$CO_2 + H_2 = CO + H_2O(g) \quad (\Delta H^o = +41.2 \text{ kJ/mol } CO_2 \text{ (RWGS)}) \qquad [1]$$

$$nCO + 2nH_2 = C_nH_{2n}(g) + nH_2O(g) \quad \text{(FTS)} \qquad [2]$$

$$nCO + (2n+1)H_2 = C_nH_{2n+2}(g) + nH_2O(g) \quad \text{(FTS)} \qquad [3]$$

The FTS standard heats of reaction in the range $8 < n < 16$ are highly exothermic and approximately -163 kJ/mol CO_2

(2) Indirect Methanol mediated route: CO_2 reduced to Methanol followed by olefin synthesis, oligomerization and finally hydrogenation to hydrocarbons as expressed in compact form by reaction [5] below:

$$CO_2 + 3H_2 = CH_3OH(g) + H_2O(g) \quad (\Delta H^o = -49 \text{kJ/mol } CO_2) \qquad [4]$$

$$nCH_3OH(g) + H_2 = C_nH_{2n+2}(g) + nH_2O(g) \qquad [5]$$

The standard heat of reaction in the latter case is approximately -73 kJ/mol CO_2.

(3) Direct route: CO_2 converted directly with H_2 to hydrocarbons with either CO or methanol formed as intermediates on the surface of a single catalyst:

$$nCO_2 + (3n+1)H_2 = C_nH_{2n+2}(g) + 2nH_2O(g) \qquad [6]$$

with a standard heat of reaction ~ -122 kJ/mol CO_2.

2. Indirect RWGS-FTS

The Fischer-Tropsch Synthesis (FTS) process was developed approximately a century ago to convert solid fuels (primarily coal) into liquid fuels. The FTS process itself, now at a TRL (Technology Readiness Level) of 9 came into being as a means to provide much needed hydrocarbon feedstocks during depletion of national resources and/or difficult trading periods. FTS requires synthetic gas (syngas composed of H_2 and CO) to achieve this transformation and in recent years steam reforming or gasification at high temperatures (> 1000K) of solid carbonaceous materials is being partially replaced by utilising non-biogenic CO_2 with e-H_2 in the syngas production step (operating at high temperatures ~ 800-1200K and moderate pressures ~ 1-25 bar). While the latter approach is still at a relatively low TRL (~5-6) it is a key component in the drive towards net-zero with abundant atmospheric CO_2 serving as a valuable feedstock in the production of sustainable fuels.

With the requisite CO_2 and H_2 feeds the combined RWGS-FTS process in its most rudimentary form is as depicted in Figure 1.

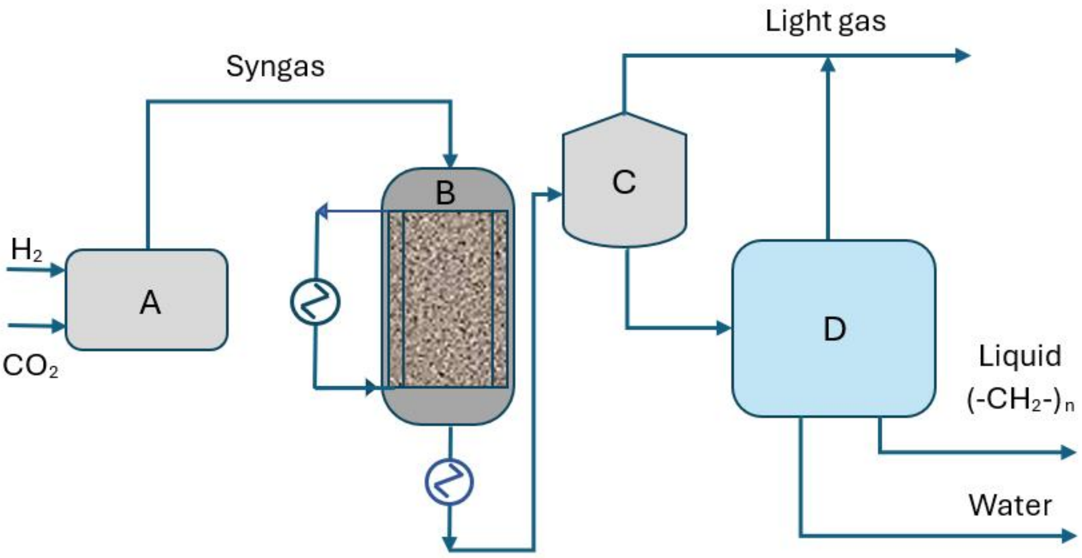

Figure 1. *The elements of the RWGS-FTS process. A. The RWGS reactor. B The FTS reactor. C. Gas-liquid separator. D Liquid separation unit.*

The conversion into liquid hydrocarbons occurs using a catalyst (Fe or Co) and the FTS reactor (typically a fixed or fluidized bed of porous particles) is operated at 200-300 °C and 10-50 bar depending on product requirements. As indicated by the nature of the highly exothermic reactions taking place in the reactor (reactions [2] and [3] above) significant levels of heat will be generated and heat management is an important design consideration in the process (e.g. the endothermic RWGS reaction is supported by the heat evolved in FTS). Lowering the temperature favours heavier hydrocarbons while higher temperatures leads to lighter hydrocarbons. Downstream from the reactor the products are cooled, separated and ultimately refined. To satisfy the need for high yield production of C_8-C_{16} (e-kerosene) the FTS reactor operating conditions are typically ~ 250 °C and ~25 bar using an iron based catalyst (see for example Dell'Aversano et al (3), Li et al (4)).

A key aspect of the hydrocarbon synthesis in FTS is the product distribution and particularly the chain growth probability, α, which is subject to the Anderson-Schulz-Flory (ASF) chain length distribution

$W_n = n(1 - \alpha)^2 \alpha^{n-1}$

where W_n is the mass fraction of hydrocarbons with chain length n. Theoretically a low value of $\alpha \sim 0.5$ corresponds to a high weight fraction of C_2-C_4 hydrocarbons (undesirable for e-SAF) while a value greater than 0.8 will provide a high weight fraction of the desirable C_8-C_{16} hydrocarbons (5,6). The growth probability is increased towards the e-kerosene range by decreasing temperature, using a lower H_2/CO ratio in the feed to the FTS reactor, increasing the pressure and selecting appropriate catalysts with promoters. A clear disadvantage of the FTS process is the resultant wide distribution of hydrocarbon products requiring significant downstream upgrading to e-kerosene. Furthermore, while the lower molecular weight gases and liquids and the high molecular oils and waxes have value, in a production process targeted for jet fuel production they impose a significant burden on the provision of expensive e-H_2 (particularly with regard to energy). For example, Hirunsit et al (7) report that the energy intensity for the provision of H_2 alone to a simulated RWGS-FTS process can reach as high as 80.77 MWh/(tonne C_8-C_{16}) which should be compared with the 3.39 MWh/(tonne C_8-C_{16}) required to operate the individual RWGS and FTS processes themselves. This outcome is largely a result of the excess production of C_5-C_7 and C_{17+} hydrocarbons during FTS leading to an e-kerosene production rate of just 0.08 kg C_8-C_{16}/kg CO_2 feed. Future developments in catalyst design (see for example (8) and Section 3 below) will certainly improve this situation and significantly lower the cost of e-SAF production which, for RWGS-FTS, is currently priced at approximately €8/kg (depending on renewable energy/e-H_2 costs). Today Jet A1 costs approximately €1.1/kg.

3. Direct CO_2-FTS

Two primary drawbacks of RWGS-FTS technology are (i) the requirement of two separate reaction systems to produce hydrocarbons from CO_2 and H_2 and (ii) the moderate selectivity of the FTS process itself toward the production of hydrocarbons in the e-SAF range. Recent work on reactor system design has led to the development of FTS systems which are significantly less complex than RWGS-FTS and which can produce e-SAF range hydrocarbons more effectively. The key to this development has been research on tandem catalysts which can perform both the RWGS reaction and the hydrocarbon production steps simultaneously i.e. reaction [6] above. The work conducted to date has been primarily at laboratory scale (so the TRL level is still ~ 3-4) however a wide variety of catalyst materials have been fabricated which can target specific ranges of hydrocarbons in the FTS process as the need arises (9-18).

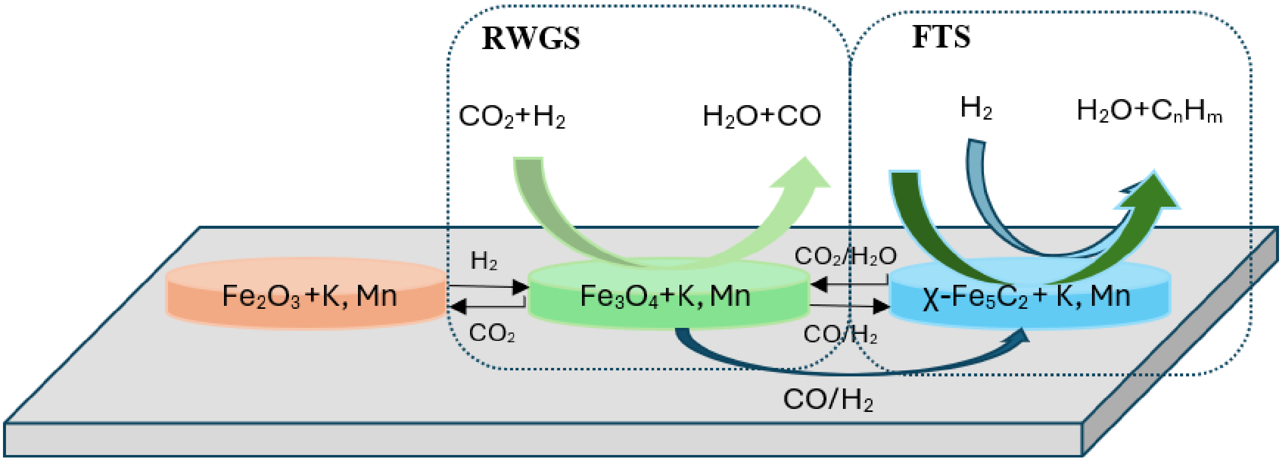

Figure 2. *Hematite nanoparticles with catalytic promoters K and Mn are activated to magnetite which forms the catalyst required to reduce* CO_2 *to CO in the RWGS reaction locally on the substrate surface. Magnetite itself is carburized forming the Hägg carbide which, in turn, provides the FTS catalytic pathway to the desired hydrocarbons (12).*

One proposed tandem scheme is illustrated above in Figure 2 (12). It is postulated that the Fe-K-Mn catalytic system depicted here (simplified for illustrative purposes) promotes the electronic transfers and molecular adsorption/desorption processes required to complete the direct CO_2-FTS process steps to hydrocarbons. The K and Mn promoters are specific to this system and may be replaced by other elements (e.g. Na, Cs or Cu, Zn) however the pairing K, Mn exhibited the highest selectivity (47.8%) for e-kerosene range hydrocarbons in the studies reported in (12). While tandem schemes of the kind illustrated in Figure 2 underpin the basis for achieving the hydrocarbon chain propagation required to enhance the yield of hydrocarbons in the desired e-SAF range (for example, C_{5+} selectivities approaching 60% have been observed in recent work (19)), the resulting distributions are still relatively broad and characterised by modified chain probability distributions similar in form to the ASF distribution (see for example (9, 19)). Molecular modelling (see for example (20)) is providing insights into the underlying mechanistic details of the catalytic processes taking place however Krausser et al (21) sound a word of caution from a process perspective with a more complete understanding of the impact of catalyst structure, activity and selectivity needed for optimal process design.

With the relative simplicity of the tandem catalytic concept it is clear that significant savings should be observed in CAPEX (for example, elimination of the separate RWGS reactor as employed in the indirect RWGS-FTS approach) and in OPEX (the endothermic RWGS reaction taking place side-by-side with the exothermic FTS reaction on a molecular scale allows for nanoscale heat management). While the above approach is still at an early stage of development, it is attracting considerable interest. In view of its relative immaturity, commercial development of the direct CO_2-FTS process is limited. Only one company (a spin-out from the University of Oxford (OXCCU) utilising the research reported in (12)) reports a roadmap towards commercialisation of modular systems, delivering approximately 10 ktpa by 2028 and proposals for licensing by 2030 with a projected 50% lower CAPEX than RWGS-FTS and reduced OPEX through efficient heat management (22).

4. Indirect CO_2-Methanol Mediated Route

In this approach (which is frequently referred to as methanol-to-jet fuel (MtJ)) the feedstocks CO_2 and H_2 are initially converted to methanol. Methanol is, itself, a very important industrial chemical feedstock for a wide variety of applications (e.g. plastics, adhesives, synthetic fabrics and pharmaceuticals), not the least of which is as an e-fuel in its own right (notably for use in the maritime sector). However, for the purpose of e-SAF production the methanol enters a process chain as depicted in Figure 3 below. Summaries of the process steps are provided in (2, 3) with extended details provided in (23) and involve, in series: (i) olefin synthesis; (ii) oligomerization and finally (iii) hydrogenation to predominantly C_8-C_{16} hydrocarbons using specialised catalysts.

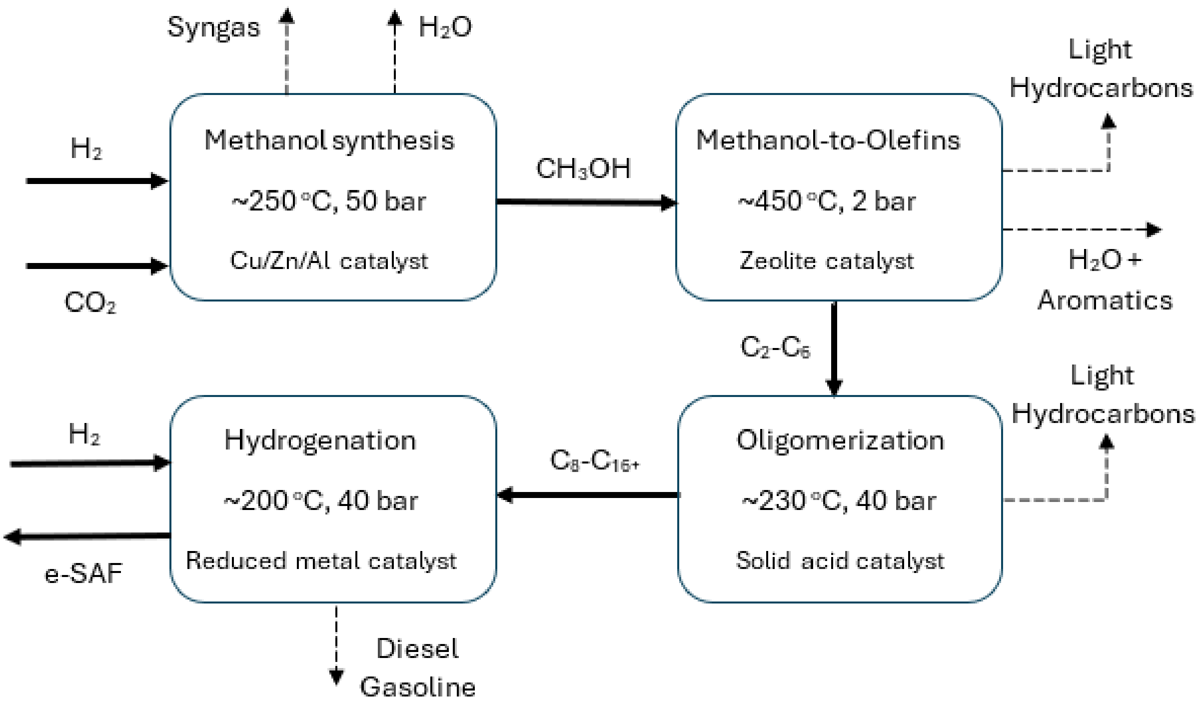

Figure 3. *A schematic of the Methanol-to-Jet fuel (MtJ) process*

The methanol synthesis reaction

$CO_2 + 3H_2 = CH_3OH(g) + H_2O(g) \quad (\Delta H^o = -49 \text{kJ/mol } CO_2)$ [7]

is conducted in the range 220-280 °C and 30-80 bar using Cu, Zn or Al catalysts.

Olefin synthesis

$2CH_3OH(g) = (CH_2)_2 + 2H_2O(g) \quad (\Delta H^o = -15 \text{ kJ/mol } CO_2)$ [8]

occurs at a significantly higher temperature but lower pressure using a microporous zeolite catalyst (e.g. H-ZSM-5).

Oligomerization

$(1/2)\, n\, (CH_2)_2 = C_nH_{2n}(g) \quad (\Delta H^o \sim -47.5 \text{ kJ/mol } CO_2)$ [9]

takes place with a return to lower temperature and higher pressure reaction conditions in the presence of, for example, a catalyst of the Nickel based silica-alumina or H-ZSM-5 class of materials.

Hydrogenation

$C_nH_{2n}(g) + H_2 = C_nH_{2n+2}(g) \quad (\Delta H^o = -10.4 \text{ kJ/mol } CO_2)$ [10]

also proceeds at conditions similar to oligomerization but using a reduced metal catalyst (e.g. Ni, Pd or Pt) supported on alumina.

Each of the reactions listed above are exothermic to a greater or lesser extent and hence favour lower temperatures and, with the exception of the olefin synthesis reaction with increasing moles in the products, higher pressures. The integrated process indicated in Figure 3 does not currently exist although each of the individual stages form part of the operations at a variety of sites globally. At this time a number of companies (e.g. ExxonMobil (24), Topsoe (25)) are investigating this technology with the possibility of commercial production within the next five years.

While the MtJ process is relatively complex involving multiple reaction systems and concomitant intermediate separation processes, it does possess a number of advantages as an approach toward the production of e-SAF. Most notably, MtJ is a ***targeted*** process (hence it is not subject to the broad ASF product distribution of FTS) producing e-SAF at high yields with only modest by-product formation outside the C_8-C_{16} range. A case in point is the simulation work of Hirunsit et al (7) which demonstrates that the energy intensity of the MtJ process is predicted to be 3.5 times lower than the RWGS-FTS process due to its efficient utilization of H_2 feedstock. Under optimum process conditions this route towards e-SAF can also limit production of undesirable compounds which appear in fossil based fuels and hence lead to a 'cleaner' aviation fuel.

5. Direct CO_2 (Methanol Mediated) Route

In contrast to the direct CO_2-FTS route, an approach in which all of the four steps outlined in Figure 3 are combined in some form using tandem catalysts (i.e. a CO_2-MtJ approach) has not yet been developed and is unlikely to be until the mid-2030s or early 2040s. This arises largely due of the complexity of the reaction route and kinetics, the sensitivity of the mechanism of the transformation of CO_2/H_2 to the desired e-SAF product range to the catalyst(s) properties including composition, active site distribution and support morphology, and finally the management of intermediates and by-products (e.g. CO and water) as they arise during the sequence of the reactions. A number of recent research studies in this area are summarised by Neto et al (2), and Elwalily et al (23) discuss possible opportunities for integrating pairs of the reaction steps [7] – [10] to simplify the process layout including the following:

1. Reactions [7] and [8] may be combined to achieve the direct conversion of CO_2 and H_2 to olefins thereby simplifying the overall process and reducing energy demand at a temperature and pressure intermediate to those for the individual steps (see for example (26) in which a mixed metal oxide/zeolite catalyst was employed). However, research is required to develop efficient multifunctional catalysts to limit the negative effects of by-product water on catalyst activity and to enhance carbon-carbon bond formation.

2. The oligomerization and hydrogenation steps may be integrated into one process utilising zeolite supported hydrogenation catalysts (e.g. Ni or Pd). The indicated operating conditions for the individual steps [9] and [10] imply this is achievable and can enhance energy efficiency. However it does eliminate recycling of low molecular weight olefins and can lead to by-product spread in the chain length distribution of final products.

Finally, in view of the value of methanol in its own right as a chemical feedstock for a variety of processes and as an e-fuel (.e.g. for shipping), it may also be worthwhile focussing on development of an integrated process involving just the three reactions [8]-[10] rather than the more complicated CO_2-MtJ approach.

Conclusion

Of the four approaches outlined here for the production of e-SAF from non-biogenic CO_2 and green hydrogen the indirect methanol-mediated route (MtJ) is the most mature although the direct CO_2-FTS approach is developing rapidly. At this time a key challenge in either case is the availability and cost of e-H_2. The route to green H_2 is predominantly through electrolysis of water with alkaline electrolyzers (AE), proton exchange membrane electrolyzers (PEM), solid oxide electrolyzers (SOEC) or anion exchange membrane electrolyzers (AEM). The IATA (1) projected requirements for e-SAF production worldwide by 2050 are 176 Mtpa. The e-H_2 requirement for this estimate to be met is in the range 25-50 Mtpa under ideal conditions (efficient H_2 feedstock utilization) which would in turn require between 1500 and 3000 TWh of electrical energy (based on the above electrolyzer technologies).

From the perspective of Ireland's needs, if we can assume that only long-haul flights to and from Ireland by mid-century operate using IATA's proposed mix of bio-SAF and e-SAF then to satisfy Ireland's annual e-SAF needs, the energy required to produce the e-H_2 required (based on today's jet fuel demands) would be ~ 1.9-3.1 TWh (~0.22-0.35GW). Employing the DAC technology developed by NEG8 Carbon to source pure CO_2 and with the availability of waste heat, then the CO_2 feedstock required for e-SAF (0.78 Mtpa) on long-haul flights would require 0.35 TWh electrical energy i.e. the feed to an e-SAF facility utilising e-H_2 efficiently would require between 2.25 and 3.45 TWh annually (5-10% of total electrical energy consumption) to maintain this component of the production process.

This is not an inconsiderable demand in energy and it is clear that significant efficiency improvements are required in the process technologies associated with e-SAF itself and the availability of abundant renewable energy (particularly low cost off-peak and/or dispatch-down power) if these requirements are to be met.

For more:

- **What is eFuel?**
- **What is eMethanol?**
- **Sustainable Aviation Fuel (SAF)**
- **Methanol-to-Jet Fuel for Sustainable Aviation**
- **Lift Off for NEG8 Carbon in Sustainable Aviation Fuel**
- **CORSIA vs EU ETS**

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