



Review

Repurposing Fischer-Tropsch and natural gas as bridging technologies for the energy revolution

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ABSTRACT

Immediate and widespread changes in energy generation and use are critical to safeguard our future on this planet. However, while the necessity of renewable electricity generation is clear, the aviation, transport and mobility, chemical and material sectors are challenging to fully electrify. The age-old Fischer-Tropsch process and natural gas industry could be the bridging solution needed to accelerate the energy revolution in these sectors – temporarily powering obsolete vehicles, acting as renewable energy's battery, supporting expansion of hydrogen fuel cell technologies and the agricultural and waste sectors as they struggle to keep up with a full switch to biofuels. Natural gas can be converted into hydrogen, synthetic natural gas, or heat during periods of low electricity demand and converted back to electricity again when needed. Moving methane through existing networks and converting it to hydrogen on-site at tanking stations also overcomes hydrogen distribution, storage problems and infrastructure deficiencies. Useful co-products include carbon nanotubes, a valuable engineering material, that offset emissions in the carbon nanotube and black industries. Finally, excess carbon can be converted back into syngas if desired. This flexibility and the compatibility of natural gas with both existing and future technologies provides a unique opportunity to rapidly decarbonise sectors struggling with complex requirements.

1. Introduction

The widespread adoption of renewable energy technologies, such as wind parks, solar farms, hydroelectric and geothermal power stations is undisputedly critical to the environmental security of the Earth and the future prosperity of its population [1,2]. Electrification of transport infrastructure and fossil fuel reliant industry to utilise this green energy is also necessary to reduce greenhouse gas (GHG) emissions and limit global warming [3]. However, the intermittent and seasonal nature of energy generated through renewable technologies, which require appropriate wind, sunlight, or tidal conditions to operate, still developing battery technologies and incompatible vehicles and infrastructure currently make reliable renewable energy supply, storage and use challenging [4].

In particular, decarbonising the transport and mobility sector is complicated due to the existing legacy vehicle fleet incompatible with new technologies and a lack of associated support infrastructure [5,6]. Most experts consider the use of liquid fuels, such as electrofuels (e-fuels) and biomass-derived fuels, necessary to achieve climate change goals within an acceptable timeframe [7]. This is especially relevant in the short and medium term where liquid fuels that are compatible with existing infrastructure and vehicles are required and for specific high energy density fuel applications for which large scale electrification is still challenging, such as commercial aircraft, ships, and trucks [8,9].

E-fuels are carbon-neutral synthetic fuels that store renewable electrical energy in chemical bonds of liquids and gases [10]. They are synthesised from hydrogen, typically sourced from water using electrolysis, and carbon dioxide sourced from the atmosphere, biomass, or

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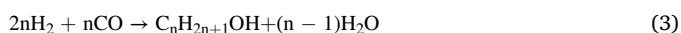
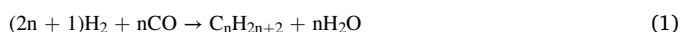
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industrial exhaust gases [11]. As the indirect electrification of an energy system, their main disadvantage is their very low efficiency compared to direct electrification, as seen in electric cars [12]. E-fuel powered conventional vehicles only utilise $\sim 10\%$ of the renewable electricity originally harvested and require 5 times more energy per km than an electric car, limiting viability through the supply and cost of renewable electricity [13]. The use of hydrogen in these fuels makes an increase in their use technologically akin to the direct use of hydrogen although e-fuels are easier to store and transport [14].

Alternatively, alkanes of various chain lengths (Equation (1)), including hydrocarbon fuels such as petrol, can be produced from any carbon-based feedstock, such as agricultural or forestry biomass (to make biofuels), plastic wastes [15] but also coal, and natural gas, using a series of chemical reactions known as the Fischer-Tropsch (FT) process [16]. Feedstock is converted into carbon monoxide and hydrogen, known as synthesis gas (syngas), by gasification and then processed into liquid hydrocarbons using conventional or renewable energy [17–20]. Most alkane products are straight chain and suitable as diesel fuel, however, competing reactions also generate small quantities of alkenes (Equation (2)), alcohols (Equation (3)) and other oxygenated hydrocarbons suitable for production of chemicals and polymers [21]. Products and yields can be controlled by adjusting the $H_2:CO$ ratio of the syngas, most commonly using the water–gas shift reaction to increase H_2 content at the expense of CO (Equation (4)) [22,23]. This process has been used for production of aviation fuel, petrol, diesel, chemicals, and polymers from coal and natural gas for coming up on a century but has been more recently coupled with biomass to make carbon-neutral biofuels [24] (Fig. 1).



Repurposing of the FT process for biofuel production has the major benefit of utilising an already well-established technology that requires at most minimal re-engineering of existing industrial scale refinery and petrochemical processes [18]. But more importantly the versatility of this process and its potential in the fight against climate change, as demonstrated in the case of biofuels, inspires reinvention of its current

scope of use and remaining unutilised opportunities.

Framework: We review and systematically compare the state of the art of traditional liquid fuels and their emerging greener alternatives (biofuels, e-fuels) with a focus on the GHG emissions associated with each option. We then introduce a bridging strategy utilising readily available existing natural gas infrastructure to satisfy hydrogen demand prior to the availability of completely green hydrogen and provide perspective on the acceleration of the adoption of hydrogen for storage of renewable energy and use in applications that cannot be easily electrified. Hydrogen production using natural gas pyrolysis yields considerable quantities of carbon co-products, whose use is discussed for energy storage, applications in engineering and advanced materials and production of legacy fuels or chemicals via Fischer-Tropsch synthesis. Recent advances in FT processes are reviewed and an outlook provided for the phasing out period of fossil fuels and the scope and duration of mid-term energy transitions in the transport sector.

2. A transitioning economy: Intermediary steps of a liquid-fuel dependent civilisation towards a sustainable future

2.1. Aviation fuel

The aviation sector is responsible for $\sim 2\%$ of global CO_2 emissions and demand for air travel is expected to more than double over the next 20 years [25]. Industry has responded with targets to reduce aviation CO_2 emissions to half of 2005 levels by 2050 [25]. Long term visions for the future of sustainable aviation rest on developing technologies, such as improved batteries, multifunctional composite materials which combine structural properties with electrical energy storage [26,27], and electrification of flight [28] or hydrogen fuel cells and turbines [14,29,30], however, the backbone of current aviation emission reduction strategies is the replacement of traditional jet fuel with sustainable aviation fuel (SAF) [31–33]. This biofuel of similar chemical composition to fossil-based fuel is produced using hydrogenated esters and fatty acids (HEFA) (in use), alcohol-to-jet fuel (ATJ), gasification-FT (commercial pilot) or power-to-liquid (e-fuel) technologies (under development) [33].

Synthetic paraffinic kerosene (SPK) produced from municipal solid waste (MSW), agricultural and forestry residues, wood and energy crops and non-renewable feedstocks such as coal and natural gas using the FT process was used as alternative jet fuel in 2009 [34], while FT-SPK with

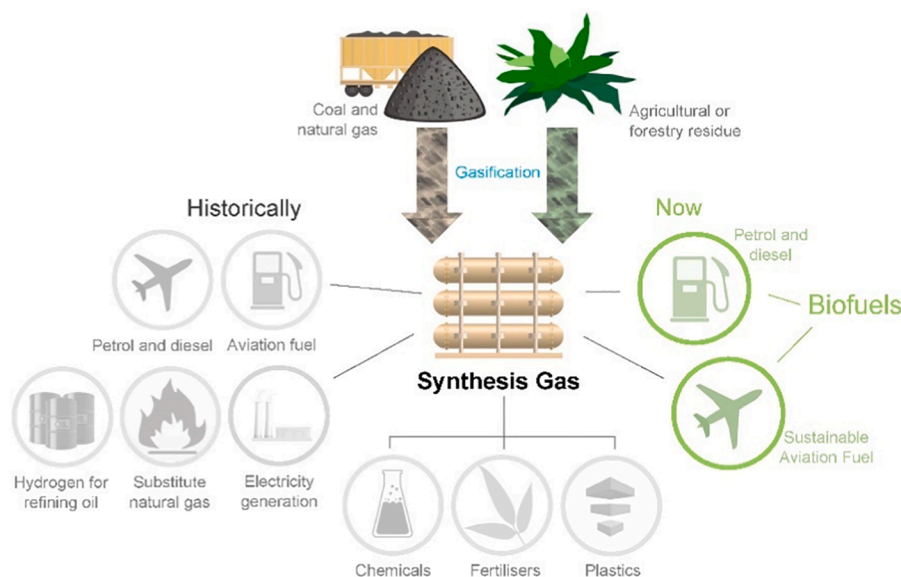


Fig. 1. The Fischer-Tropsch process is a series of chemical reactions used to convert carbon-based feedstock into hydrocarbon products. It can be used to produce hydrogen for refining oil, chemicals, fertilisers, plastics and to supplement natural gas or in electricity generation.

aromatics (FT-SPK/A), which is derived from the same feedstock, was approved in 2015 [35]. Conversely, ATJ-SPK is made from ethanol and isobutanol, while HEFA-SPK and catalytic hydrothermolysis synthesized kerosene (CH-SK, or CHJ) are derived from fatty acids and fatty acid esters, or more generally lipids derived from plant and animal fats, oils, and greases [36].

SAF is compatible with existing jet engine systems, in most cases blended with up to 50% conventional aviation fuel [37], although current commercial aviation trials are investigating 100% SAF-based aircraft propulsion and military trials have already been successfully completed [38]. Transient, ignition and extinction performance are comparable or superior to conventional jet fuels [39].

SAF has a lower viscosity than conventional jet fuels and contains no sulphur, resulting in 5–10% less CO [40], approximately half the sulphur dioxide and reduced SO_x emissions compared to petroleum-based jet fuel [41]. Nitrogen oxide (NO_x) emissions can vary, with some SAF having the same or higher values than traditional aviation fuel [41], however, reductions of up to 32% NO_x have been reported compared to Jet A-1 fuel due to a lower primary zone temperature [42]. The absence of aromatic rings in SAF also results in soot reductions of up to 87% compared to conventional fuel [42–44] and up to 95% lower particulate emissions at cruising conditions compared to JP-8 fuel due to a reduction in soot nuclei [41,45].

A product's environmental impact can be quantified using the climate change impact category of its life cycle assessment (LCA). This category is based on GHG emissions, most notably carbon dioxide, nitrous oxide, methane and halocarbons, which cause anthropogenic global warming. Global warming potential (GWP), expressed as kilogram CO₂ to air equivalent (kg CO₂ eq.) is used as a standardised expression of these different gases over a lifespan of 100 years [46]. Biomass-based FT aviation fuels have lower environmental impact than those derived from other sources, with GHG emissions of 9.3 g CO₂ eq./MJ for fuel produced from eucalyptus and 9.4 g CO₂ eq./MJ for fuel produced from sugarcane residues, both produced in a biorefinery setting combining an ethanol distillery with FT jet fuel production [47] (Table 1). These values can decrease in a standalone FT-plant to as low as 2.3 g CO₂ eq./MJ and 1.4 g CO₂ eq./MJ, respectively, if the feedstock is considered waste [48]. Fuel produced from corn stover is associated with GHG emissions of 5.4–13.0 g CO₂ eq./MJ [49] with variation-based process efficiency (46–60%) and fertilisation (mean global values compared to US values) [49,50]. Studies on forestry residues and short rotation forestry biomass such as willow and poplar report GHG emissions of 6.0–13.0 g CO₂ eq./MJ [49] and 9.0 g CO₂ eq./MJ [49], respectively. Higher values (32.4 g CO₂ eq./MJ) are reported if land use change (LUC) is included in the analysis for willow, which is comparable to wheat straw feedstocks including LUC (38.2 g CO₂ eq./MJ) and demonstrates how important this aspect is especially for the production

Table 1

Well-to-wheel (WtW) GHG emissions (g CO₂ eq./MJ) of e-fuel and biomass-based FT aviation fuels compared to oil- and natural gas derived references.

Type	Feedstock	GHG emissions (g CO ₂ eq./MJ)	Reference
Oil	Crude oil	80.7–109.3	[52]
Natural gas	Fossil fuels	99.8–105.8	[52–55]
	Fossil fuels (with carbon capture)	86.2–87.3	[52]
	Upgraded biogas	38.0	[36]
E-fuel	Water, CO ₂ , electricity	11.0–28.0	[56]
Biofuel	Forestry residues	–10.0–13.0	[49]
	Willow	–7.0–32.4	[36,49]
	Poplar	–6.0–10.0	[49]
	Corn stover	–3.0–13.0	[49,50]
	Switchgrass	–2.0–17.7	[53]
	Sugarcane	1.4–9.3	[47,48]
	Eucalyptus	2.3–9.4	[47,48]
	Municipal solid waste	32.9–62.3	[51]
	Wheat straw	38.2	[36]

of sustainable biomass feedstock [36]. Fuel derived from municipal solid waste using conventional or plasma gasification is associated with the greatest GHG emissions with values of 32.9–62.3 g CO₂ eq./MJ [51] but does reduce reliance on oil and utilise waste that may otherwise be destined for landfill.

The environmental impact of a fuel over its life cycle is influenced by several factors (e.g., choice of feedstock, including or excluding LUC), but especially by the methodical handling of co-products, which are generated during the production of the investigated fuel. This sensitivity is best illustrated by a study that reports carbon negative results for fuels derived from willow, poplar, corn stover and forestry residues (–7 g CO₂ eq./MJ, –6 g CO₂ eq./MJ, –3 g CO₂ eq./MJ and –10 g CO₂ eq./MJ, respectively) [49], perfectly demonstrating how complicated comparison of results based on different methodologies can be.

If omitting LUC from LCAs, GHG emission change associated with crop change (direct LUC) or land conversion from natural habitat into arable land for crop production (indirect LUC) is excluded from the analysis. In environmental assessment, carbon cost, storage and sequestration benefits are disregarded if LUC is excluded [57]. For energy crop production to be more sustainable than fossil fuels, the carbon reduction associated with the shift from one to the other must be greater than the carbon storage and sequestration loss associated with LUC. LUC greatly influences LCA results, even for assessments with the same conversion routes and biomass feedstocks, and its inclusion is strongly recommended when considering the environmental impact of biomass feedstocks [36]. That said, the methodology used in an analysis should be clearly justified for transparency, especially when LCAs are used for policy-related purposes.

More broadly, global biofuels potential is limited by the quantity of biomass that can be sustainably produced [58], with their cost and environmental footprint sensitive to life cycle factors, such as land clearance for crops, fertiliser use, crop processing and transportation [54]. Waste biomass feedstocks (2nd generation biofuels) omit environmental impact associated with cultivation and harvesting, providing higher GHG emission reductions (50–100%) than crop feedstocks (1st generation biofuels) (50–75%) [49,59–66]. However, possible changes in land use resulting from widespread adoption of biofuels draw criticism, such as cultivation of monocultures, competition of energy crops with food and fodder production on limited fertile land and deforestation and loss of biodiversity as arable land is expanded to accommodate the growing requirements for food and energy crops [53,67]. For all of the above reasons, it is challenging to make informed recommendations on optimal biomass sources for gasification and FT fuel production.

In terms of future potential, FT fuels remain more environmentally benign than e-fuels in the mid-term while the energy mix feeding the grid is transitioning to 100% renewables, if ever achieved. The environmental impact of e-fuels is highly dependent on the energy used to produce them; without the use of 100% renewable energy there is little environmental benefit in their use over FT fuels. Even projected energy mixes for 2030 are still too fossil fuel dependent to make e-fuels a greener option than FT fuels. This fact strongly demonstrates the importance of considerable acceleration in renewable energy production since e-fuels will undoubtedly be the environmentally preferred choice in the long term.

2.2. Petrol and diesel

Petrol- or diesel-powered light duty vehicles (LDV) are the primary method of transportation worldwide. Over 244 million LDVs are currently in service in the US alone with an additional 15 million new vehicles sold every year [68]. >90% of US families have at least one vehicle and on average each household owns two or more vehicles. Deteriorating air quality from the fine particulate matter, volatile organic compound, and GHG emissions of such large numbers of vehicles have prompted new legislation, such as the Energy Independence and Security Act (EISA), which mandates the annual production of 36

billion gallons (~ 136 billion L) of fuel from renewable resources (~ 25% of current US petrol demand) [69]. The revised EU Renewable Energy Directive (RED II) has also mandated that ~ 7% of LDVs be fuelled by biofuels by 2030 and a 70% reduction in GHG emissions achieved in vehicles running on non-renewable fuels [70].

Technologies, such as water (alkaline) electrolysis (AEL) for hydrogen production and direct air capture (DAC) of CO₂ from air can be coupled with FT synthesis via the water–gas shift reaction to produce petrol, diesel or waxes [71]. This would be viable if CO₂ emissions were appropriately priced. However, there is a general scientific consensus that DAC exhibits limited potential for large scale CO₂ capture due to the plant footprint required and its energy intensive operation [72]. These processes can be coupled with the chemical industry to offset plant capital costs [11,73] and are associated with GHG emissions of 64.1 g CO₂ eq./MJ if renewable wind energy is utilised during processing and 441.1 g CO₂ eq./MJ if the energy supplied is derived from the German gas grid [74]. E-fuels produced from renewable energy are typically associated with GHG emissions of 1–30 g CO₂ eq./MJ [53,56,75,76]. Future developments to make co-electrolysis (solid oxide electrolyzer cell, SOEC) available at scale and improvements in DAC technology could see these values drop to ~ 7 g CO₂ eq./MJ and ~ 100 g CO₂ eq./MJ by exclusively using renewable energy or the average grid mix, respectively, by 2030 [74]. However, the fact that the GHG emissions of even these improved technologies exceed values associated with traditional fossil fuel use when non-renewable energy is used during processing (100 g CO₂ eq./MJ compared to 83.8 g CO₂ eq./MJ) demonstrates the sensitivity of e-fuel emissions to energy source [53]. GHG emission reductions of up to 50% can be achieved by blending 70% e-diesel with conventional diesel [76].

Petrol and diesel biofuels can also be produced from biomass using FT technologies. High temperature FT processes are better suited to petrol production, yielding a high olefinicity product with a low boiling range (50% alkenes (olefins), 20–30% alkanes (paraffins), 10–15% oxygenates, 1–5% aromatics), while low temperature processes yield a much more paraffinic and straight-chain product (>70% alkanes, 15–20% alkenes, 5% oxygenates, <1% aromatics) and are used to produce diesel with a lower density by diesel standards [77,78]. Diesel is the focus of most literature due to the higher fuel efficiency of diesel engines, a factor which is expected to drive considerable growth in the diesel market [79,80].

Diesel produced from biomass or natural gas using the FT process is paraffinic, contains virtually no sulphur and has a low aromatic content [81,82]. It has a higher combustion yield than conventional diesel, which results in lower carbon monoxide and hydrocarbon emissions [83,84]. Notably, it also has a very high cetane number, which when coupled with a lack of polycyclic aromatic hydrocarbons provides reductions in particulate matter emissions for all injection timing variants [82,83]. FT diesel produces less soot than conventional diesel at mid and high engine loads and in response to variation in injection timing [85]. Its high-octane number also results in changes in combustion phasing that shorten ignition delay and in doing so reduce NO_x emissions and combustion noise [83,84,86]. Further reductions in NO_x emissions are possible without adverse smoke or thermal efficiency ramifications using exhaust gas recirculation and standard injection timing [84,86]. Critically, in addition to its environmental benefits FT diesel can also be used in conventional engines without major modifications [87] or loss in efficiency [83,86] or power [82]. It can also be blended with petroleum-derived diesel [88] or biodiesel [89] if desired.

FT fuels produced in a sugarcane biorefinery with integrated FT-synthesis from either a sugarcane/eucalyptus or energy-cane/eucalyptus mix were associated with 85–95% lower GHG emissions than their fossil fuel counterparts (diesel: 11.2 and 11.4 g CO₂ eq./MJ; petrol: 12.8 and 12.9 g CO₂ eq./MJ, respectively) [90]. Straw feedstocks can provide a CO₂ reduction of 98%, although they are associated with high water consumption and wastewater production [91]. Feedstocks, such as switchgrass, can provide additional environmental benefits by

converting soils depleted of carbon through agricultural practices into grasslands [92]. This consideration transforms traditional switchgrass derived FT diesel from a carbon positive fuel (18.2 g CO₂ eq./MJ) into a carbon negative fuel (−1.6 g CO₂ eq./MJ) [92]. Wood chip derived FT diesel exhibits 67–77% lower GHG emissions than fossil fuels (21–30 g CO₂ eq./MJ compared to 89 g CO₂ eq./MJ) [75,93], while short rotation forestry biomass, such as willow, ranges from 65 to 88 g CO₂ eq./MJ [94]. Even higher reductions (91–98%) can be achieved if renewable energy is used during processing [95,96].

Considerably less information is available on FT petrol compared to diesel, however, literature suggests that it has lower hydrocarbon, particulate matter, and carbon monoxide tailpipe emissions than conventional petrol [97]. Neither e-fuels nor biofuels are currently cost competitive with fossil fuels. However, their cost is expected to as much as halve in the EU between 2015 and 2050 [56] as they are scaled up, improvements are made in CO conversion, biomass collection and utilisation and they become more widely implemented [56,65,98,99]. Liquid fuels derived from natural gas using the FT process are already industrially produced by ExxonMobil in Qatar and by Shell in Malaysia and Qatar.

2.3. Chemicals and organic building blocks

Efforts to make polymer and chemical production less reliant on oil have focussed on the use of syngas, which can be processed into methanol, oxygenates, methane and higher alkanes as already noted by Fischer and Tropsch in 1925. Methanol is produced directly from syngas—primarily derived from steam reforming of natural gas but synthesised from coal gasification—over catalysts, such as Cu, ZnO, Al₂O₃ or MgO (Equation (5)):



Or from captured CO₂ and green H₂, for instance over a Pd/Cu catalyst (Equation (6)):



With a global demand of 70 million metric tonnes in 2015, methanol is one of the most important bulk chemicals in industry but can also be used as an additive in liquid fuels or as a safer and more easily transportable alternative to H₂ in fuel cells. It is used in the production of formaldehyde, methyl ether or ester products and acetic acid [17]. Methanol is also an alternative starting material for many existing petrochemical processes, such as methanol-to-olefin (MTO), benzene, toluene, xylenes (BTX) and many hydrocarbon production processes [17-19,54,100]. Dimethyl ether, olefins and petrol are already industrially manufactured from methanol [53,101,102].

Biomass-reliant production chains are considered short-cycle carbon systems and considerably more sustainable than long-cycle carbon systems based on fossil resources [103]. Dedicated energy crops, such as perennial grasses, agricultural and forestry by-products and vegetable waste are all good examples of high potential biomass for syngas production [104,105]. Methanol produced from CO₂ and renewable energy is usually associated with negative GHG emissions, with negative values ranging up to −1.4 kg CO₂ eq./kg methanol produced [17,106-109]. Conversion of methanol-to-olefins also has negative to near zero GHG emissions with values of 0.44 kg CO₂ eq./kg olefins using a municipal solid waste feedstock, −4.15 kg CO₂ eq./kg olefins for corn, −4.4 and −8.7 kg CO₂ eq./kg olefins for wood chips [110,111]. These results do strongly depend on the source of the CO₂ and energy used during electrolysis [20,112-117] with values rapidly rising to 4.6 kg CO₂ eq./kg methanol produced when coal- rather than wind-generated energy is used [106]. GHG emissions can be further reduced by introducing a CO₂ recovery unit [118] or incorporating the system within an existing production process, such as a pulp and paper mill with wood residue used as feedstock [106,119,120] or a biorefinery with e.g., methanol co-

produced alongside ethanol [121-123].

Most methanol is currently produced in China using coal gasification with GHG emissions of ~ 2.7 kg CO₂ eq./kg methanol [124,125]. Use of all bio-based alternatives or even traditional natural gas (0.8 kg CO₂ eq./kg methanol) would subsequently be radically more environmentally sustainable than current practice [106,118,121,124,125]. Additional benefits of biomass-based production of methanol include rural economic development, renewable resource utilisation and environmental protection. Methanol, ethylene, propylene, benzene, toluene, and xylene production from renewable resources should be accompanied by utilisation of low-carbon energy, electric heating and heat integration, high efficiency resource extraction and recycling, use of existing infrastructure and on-site processing with minimal transport distances to ensure meaningful GHG emission reductions are achieved [115,126].

3. Natural gas as bridging technology and supplement to biomass-based renewable processes

3.1. Sector coupling and energy storage opportunities: hydrogen and synthesis gas

Sector coupling of the two largest existing energy networks – electricity and natural gas – could present an immediately implementable and cost-effective bridging technology to aid the transition to renewable energy powered electricity grids and e-fuel or biomass-derived fuels in aviation and heavy goods vehicles until full electrification can be achieved (Fig. 2). Transport of natural gas is possible through well-established international grids, the capacities of which exceed even those of electricity grids [127]. Moving natural gas in line with supply and demand to supplement power networks is likely cheaper than expanding these grids [128]. The fuel and chemical sectors could also be coupled since many important organic building blocks and polymers are inherently produced during FT-synthesis of syngas feedstock [21,80].

Coupling the generation, distribution and storage infrastructure of multiple existing sectors substantially reduces capital and maintenance costs [7,54,129], increases reliability and flexibility, improves user acceptance rates for FT fuels and reduces the cost of transitioning to a decarbonised energy system [130,131].

Like pumped-storage hydroelectricity, natural gas could also be used for energy storage in gas storage facilities to alleviate downtime in renewable energy powered grids [132,133]. Natural gas can also be converted into synthesis gas via steam reforming or hydrogen and carbon via pyrolysis, all of which can be used for energy storage when electrical demand exceeds supply [134]. Natural gas, synthesis gas and carbon are all easily stored forms of potential chemical energy. Stored carbon can be converted into synthesis gas or hydrogen, which can be used in gas-to-power technologies, when energy demand is high and renewable energy supply low. Conversely, surplus renewable energy can be used to produce hydrogen through water electrolysis, stored in gas form for which there is already existing infrastructure, and the gas when needed then converted back into electricity in gas-fired powerplants with the produced power delivered through the electricity grid (power-to-gas-to-power, PtGtP) (Fig. 2). This is opposed to systems such as power-to-heat (PtH), in which excess renewable energy is converted into heat using resistors, boilers or heat pumps and stored as thermal energy for later use [8,9].

En route to a hydrogen economy, where water electrolysis is considered to be the best option if powered solely by renewable energy (Table 2), hydrogen can also be produced using the processes described above. Over the period for which a bridging solution based on natural gas would be relevant, it is unlikely that renewable energy would be the predominant or even a viable energy source for hydrogen production at the scale required during the energy transition [135]. In this case methane pyrolysis with further use of the carbon co-product outperforms steam reforming even when CCS is used to reduce the associated GHG emission footprint and could even be carbon negative if

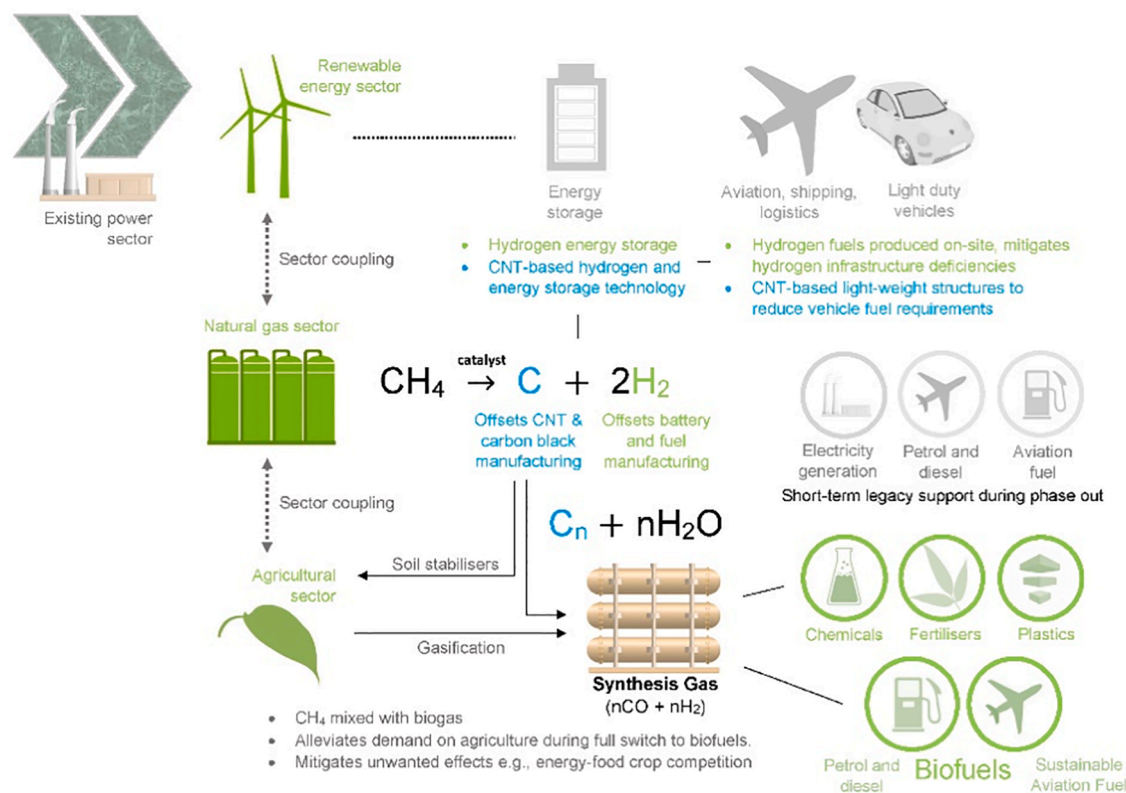


Fig. 2. Coupling of the renewable energy, natural gas, and agricultural sectors in conjunction with the Fischer-Tropsch process provides flexible pathways and inter-sector support during the transition to new fuel and energy technologies.

Table 2

Chemical equations and literature values for the associated greenhouse gas emissions (GHG, kg CO₂ eq./kg H₂) of various hydrogen production routes.

Production route	Equation	GHG emissions (kg CO ₂ eq./kg H ₂)	Reference
Steam methane reforming	CH ₄ + H ₂ O = CO + 3H ₂	10.0–15.9	[138,139]
Steam methane reforming + CCS	CH ₄ + H ₂ O = CO + 3H ₂	3.1–5.9	[138]
Methane pyrolysis	CH ₄ → C + 2H ₂	1.9–6.4	[135,140]
Water electrolysis (EU-27 energy mix)	2H ₂ O → 2H ₂ + O ₂	27.6	[141]
Water electrolysis (renewable energy)	2H ₂ O → 2H ₂ + O ₂	0.03–3.0	[142–147]

biomethane were used [136]. Arguably the main issue with methane pyrolysis is the 3:1 mass ratio of carbon to hydrogen produced during the process, which yields very large quantities of excess carbon. In 2020 global annual hydrogen demand was 90 Mt and demand is predicted to more than double by 2030 [137]. Subsequently, if total hydrogen demand was met using only methane pyrolysis technologies this would result in the generation of 270–540 Mt of carbon co-products. Potential uses for this excess carbon will be discussed further in Section 3.3.

3.2. Expanded use of hydrogen in transportation and mobility

Hydrogen is already an important feedstock in many industries and its use is likely to considerably grow in the future, especially as a fuel for transportation and mobility applications that are not easily electrified [148]. It is typically produced through photocatalysis, water splitting, steam reforming or hydrocarbon decomposition processes [149]. Water electrolysis processes are touted as the future of hydrogen production but consume large quantities of fresh water and renewable electricity, linking their cost and environmental sustainability with these factors [150].

Natural gas may provide an intermediate hydrogen generation platform to accelerate the adoption of hydrogen as a fuel for the transport and mobility sector [151]. Moving natural gas is a lot easier than moving hydrogen since it does not embrittle steel piping infrastructure, with hydrogen then produced at decentralised locations and transported to tanking stations. Moreover, natural gas can be used to generate power—although ideally this should be done with renewable energy—which can then be used to generate hydrogen via (alkaline) water electrolysis on site at tanking stations [132,134]. This technique would help to overcome the lack of hydrogen distribution and storage infrastructure currently available, making hydrogen powered vehicles and the networks servicing them technically and economically viable sooner [151,152]. Long-term, natural gas networks could be retrofitted to transport hydrogen as it becomes a more widely adopted fuel [153].

Steam reforming (Equation (7)) is a simple and efficient method to produce hydrogen from natural gas, however, CO_x by-products affect hydrogen purity [154] and the process is associated with considerable GHG emissions [155,156]. The methane thermal decomposition process (Equation (8)) is a simple and economical alternative to generate hydrogen from natural gas and does not require a water–gas shift reaction to purify the hydrogen [157]. It also results in the formation of carbon nanomaterials as a co-product. Carbon nanotubes (CNT) are currently produced in increasingly large volumes for engineering applications and have a growing associated carbon footprint of their own [158].



The simultaneous conversion of natural gas into hydrogen for fuel for the transportation and mobility sector or energy storage to alleviate

downtime in renewable energy powered electricity grids and carbon nanotubes for engineering applications would represent a range of very economically valuable products with versatile usage opportunities while offsetting and hence decarbonising the existing liquid fuel, battery, and carbon nanotube industries. The carbon footprint of hydrogen and carbon production can be reduced by up to 65% if heat integration and carbon use within the process is successful [73,159–162].

3.3. What to do with carbon co-products?

The advantage of methane pyrolysis over conventional hydrogen production routes such as steam methane reforming and coal gasification is that the only co-product is solid carbon in the form of carbon black, activated carbon, graphene or carbon nanotubes depending on the catalyst used (Ni, Fe and C-based catalyst [163]). These carbon co-products are stiff, light, flexible and have high surface area, thermal and electrical conductivity [164–167]. They are used in a wide range of applications, such as supercapacitors, photovoltaic devices, electrodes, lithium-ion batteries, computer and memory chips, conductive composites and films, organic light-emitting diodes, food packaging materials, drug delivery systems, energy, and hydrogen storage, scanning probes and various sensors [166–168].

The commercial value of these co-products (330–370 USD/tonne) also improves the economic viability of thermal decomposition of methane as a hydrogen production process when compared to steam reforming [162]. CNTs are currently the most valuable carbon co-product with a market value of 2.6 billion USD in 2019, representing a 26% market share in the nanomaterials industry [169]. Approximately 24% of this market exists in the US alone, although CNT manufacturing is rapidly expanding in China, Japan, Australia, and India [158]. The CNT market is expected to reach a value of 5.8–15 billion USD by 2027 [170]. CNT demand is driven by increasingly widespread use of lightweight polymer composites in structural applications, conductive fillers for electromagnetic shielding and antistatic materials for automotive, electronics and energy storage applications. They are also promising replacements for silicon transistors due to their high carrier mobility [158].

The largest demand for CNTs will, however, perhaps be derived from existing carbon black markets and applications, such as electric equipment, conductive components, tyre production or rubber reinforcing agents [171,172]. Carbon black is usually produced from crude oil residues or coal in a high temperature flame synthesis process. A 12.8 Mt market valued at 17.5 billion USD in 2018, carbon black is predominantly produced (~64%) and used (~62%) in the Asia Pacific region and primarily in China [173]. Manufacturing is projected to grow in this region reaching nearly 26.0 billion USD and a 65% market share by 2025 [173]. Shifting Chinese government environmental policies in 2020 have, however, caused surges in carbon black prices due to the industry's considerable environmental impact [174].

At least 30–50% of the existing carbon black market could be rapidly replaced by more environmentally friendly methane pyrolysis derived CNTs. In 2018, virgin carbon black production was associated with 30.3 Mt of CO₂ emissions (2.4–5.7 kg CO₂ per kg carbon black produced) [175,176]. A readily achieved 30–50% substitution of carbon black for CNTs would result in an up to 10 Mt reduction in CO₂ emissions [175]. Even higher GHG emission reductions could be achieved by replacing virgin activated carbons produced from hard coal with CNTs (up to 11 kg CO₂ eq. per kg) [177].

Nonetheless, while the current applications and market value of CNTs and carbon black do improve the economic viability of the methane pyrolysis process and represent opportunities for offsetting the environmental footprint of some existing industries it is highly unlikely that the current demand for these materials (15–20 Mt/year) would be sufficient to utilise all carbon co-products associated with the production of hydrogen to meet current demands using methane pyrolysis (270 Mt/year). New applications for carbon co-products must subsequently

be developed [178].

Solid carbon can be sequestered in concrete as a filler to improve compressive strength and reduce pour volume, which results in CO₂ emission reductions of 10% [179]. Structural engineering components could provide an opportunity for the utilisation of carbon co-products produced during methane pyrolysis [180,181]. However, while this may be possible in the future if high-performance carbon nanotube/nanofibre based structural fibres can be produced [182], such CNT/CNF fibres are currently incompatible with traditional carbon-based structural materials, which rely on carbon fibres. Carbon foams, on the other hand, could represent a timelier alternative to foamed concrete in the construction industry.

There has been a resurgence in interest in the potential of solid carbons as soil enhancer. CNTs increase soil water content, maximum dry density, plant resistance to disease and decrease optimal moisture content, heavy metal availability, non-CO₂ GHG emissions and plasticity index. Additionally, they affect hydraulic conductivity and improve soil water balance, especially in dry and hot areas [183–185]. Addition of solid carbon to soil increases plant CO₂ absorption and reduces CO₂ emissions into the atmosphere, forming a long-term carbon sink [180]. Nevertheless, negative or neutral soil and plant responses have also been reported and the long-term effects of biochar-soil-plant interactions are still unknown [180,186].

Surplus carbon co-products produced through methane pyrolysis could also be used in the steel industry, with 0.5 t carbon required per metric tonne of iron ore reduced to steel. Based on current global annual steel production (1877 Mt per year) [187], the carbon co-products produced by methane pyrolysis to satisfy all hydrogen demand for any year from 2020 to 2030 (270–540 Mt), could be fully utilised in steel manufacturing, with capacity for an additional 400–670 Mt per year.

3.4. Zero waste – excess carbon back to syngas

Alternatively, excess or undesired carbon material generated during hydrogen production can be converted back into syngas through a reaction with water if desired (Equation (9)). The syngas can then be used in the FT process to produce SAF, FT petrol and diesel and polymers and chemicals, including methanol [17–20,188,189]. These products will be especially important for short-term legacy support of obsolete aircraft, vehicles, and industries during the transition from liquid fossil fuels to electric and fuel cell technologies and to alleviate demand on the agricultural sector and energy-food crop competition during a full switch to biofuels [8,9]. The range of products and transformation pathways associated with the FT-system are also very useful as it means that CNTs produced on-site at facilities where their storage or use is impractical, such as tanking stations, can be converted back into more fitting products, such as other liquid fuels, at such sites. Nonetheless, the use of natural gas to generate liquid fuels amounts to little more than its traditional FT application.



Estimations of the GWP of the natural gas supply chain (in particular methane emissions) vary, with top-down estimates based on large-scale atmospheric sampling often exceeding bottom-up source-based emission inventory estimates [190]. The age and level of maintenance at gas extraction, processing, transport, and storage facilities is a key factor in the environmental impact of natural gas [190–192]. ‘Super emitters’ utilising dated, inefficient or poorly maintained equipment bear responsibility for a larger share of the environmental impact of natural gas, with the largest GHG emission source—methane venting and leakage during gas recovery—able to be reduced by 75–99% using modern equipment [191–194]. Methane emissions associated with shale gas production can be equal to [195] or disproportionately (30–90%) higher [190,196] than conventional gas due to methane emissions resulting from flow-back fluids and drilling operations.

Despite these considerations, jet fuel for aircraft [52–55,197], petrol and diesel for light vehicles [62,75,79,92,95,198,199] and olefins [200] derived from natural gas using the FT process are generally considered to have the same GWP as oil-based fuels and oil-to-olefin processes. Their use, while important during the transition phase, should be wound back, as should that of natural gas altogether, as battery technology improves, and full electrification is achieved. This can be supplemented as necessary with the use of fuels such as hydrogen, methanol and bio-fuels in heavy goods vehicles, shipping, aviation and other applications requiring high energy density fuels [8,9]. Regulators should promote the channelling of more traditional natural gas-derived fuels to the aviation sector over other transport sectors, where alternative energy sources, while still sparsely adopted, are at least more widely available [53]. Manure- or sewage sludge-derived biogas can also be mixed with natural gas to reduce the GWP of these fuels [36,201,202].

3.5. Reinventing Fischer-Tropsch in light of new complementary materials and technologies

The process to produce synthetic oil (methane homologs) from syngas (Wassergas) was first disclosed by Franz Fischer and Hans Tropsch in *Berichte der deutschen chemischen Gesellschaft* in 1926 [203,204] and became popular through the necessity to reduce dependence on oil reserves, producing fuels from hard coal via syngas. FT synthesis has been extensively used to produce automotive fuels, such as petrol and diesel, and more recently has been optimised for the production of non-automotive products, such as jet fuel, liquified petroleum gas, olefins, oxygenates, and aromatics [205,206]. The latter are key base chemicals and serve as building blocks for the production of polymers and pharmaceuticals etc. Recent research has either heavily focused on the development of more efficient catalyst systems, which address the limitations of the Anderson-Schulz-Flory product distribution (Equation (1), with n ranging from 1 to 20 and methane being undesirable) of standard FT fuels [207,208] or on improving product selectivity in favour of olefins or aromatics [206].

Fuel quality can be improved, and lower olefins and aromatics produced using reaction coupling and bi- or multifunctional catalysis. One challenge, which is still a topic of ongoing research, is the improvement of catalyst stability addressing catalyst deactivation [208]. While most research centres on metal and composite catalysts, such as metal oxides and zeolites, carbon supported metal (Co, Fe) catalysts, based on activated carbon, carbon nanotubes, nanofibers, spheres, and metal-organic framework derived carbonaceous materials are experiencing increased attention [209]. This is notable since these carbon products are readily available in large volumes from methane pyrolysis, with their co-product nature making them more economically viable in large scale operations (a limitation otherwise noted in the literature) [209] and also more environmentally friendly than might otherwise be possible since they can be held within a carbon gasification (C + H₂O → CO + H₂)-FT loop. Reactor design and optimisation of process parameters constitutes the other side of the coin required to optimise selectivity and desired product yields, especially given the variety of potential feedstocks for FT synthesis ranging from coal and gas to biomass [210].

Combining complementary processes to form integrated systems that exploit material and energy flows, such as the use of exothermic FT synthesis to heat water and liquify hydrogen for storage can further improve efficiency and resource utilisation [211]. The heat generated in FT reactions enable steam export to couple FT synthesis with steam reforming or gasification, water gas shift reactors, steam turbines or electrolysers, considerably improving their thermal efficiency [212]. Renewable energy technologies, such as solar heat, can also be introduced to power the process [213]. Furthermore, recent research highlighted the potential to use photocatalysts in FT synthesis to produce ‘clean’ fuels although such an implementation does require new reactors that permit irradiation of the entire photocatalyst bed [214].

While all these technologies represent considerable advances in

efficiency and resource utilisation, they do still require energy that is invariably associated with CO₂ emissions. A modified FT process route already allows for the utilisation of CO₂ as feedstock via a methanol-mediated pathway. New multifunctional catalyst systems are currently under investigation to enhance the efficiency of the hydrogenation of CO₂ via alkenes, their cyclization and dehydrogenation into aromatics [215]. High-octane fuel can also be produced in a two-stage FT process from low-octane gasoline by isomerisation of alkenes and methoxylation processes [216].

Recent process developments couple carbon capture and storage with FT in more flexible, small-scale reactors with direct CO₂-utilisation to produce light alkanes, naphtha, middle distillates, light and heavy waxes. During this process, CO₂ is converted to syngas using a reverse water gas shift reactor [217]. Conversely, direct CO₂ hydrogenation is also possible to produce methanol, resulting in lesser by-product formation and heat release but more water generation. This lowers the equilibrium conversion and makes new catalysts and reactor designs necessary [218].

The above strategies are also feasible using a biomass [210] or waste [219] feedstock for gasification and production of syngas. However, the utilisation of biomass as a feedstock is currently hindered by variations in quality, high transportation, feedstock preparation and plant capital costs [210]. Miniaturised FT reactors have been developed and tested [212]. This would enable decentralised applications allowing for the utilisation of local biomass streams, which would also mitigate the environmental footprint associated with transport. Syngas cleaning may also be required for some feedstocks; a factor that may affect the economic viability of the entire process [220].

4. Looking to tomorrow: Phasing out non-renewables

At current production and usage rates oil reserves would last ~ 50 years [221]. However, rising prices as supplies run low, increasing emphasis on environmental sustainability and diversification of energy portfolios will likely see demand for oil reduce in the future, somewhat extending this timeline [3]. In contrast, there are currently enough recoverable natural gas resources to last ~ 230 years at current production and usage rates [129]. The availability and versatility of natural/biogas in addition to its unique compatibility with many existing fossil fuel and future fuel cell technologies endows it with a critical role as a bridging technology in the transition from fossil fuels to renewable energy.

The U.S. Energy Information Administration (EIA), Gas Exporting Countries Forum (GECF), and Organization of the Petroleum Exporting Countries (OPEC) predict that gas production will continue to grow until 2050 driven by end use consumption and export opportunities. An increase in natural gas demand of 50% compared to 2019 levels is expected by 2050, driven by the increasing cost of oil, a growing population, rising focus on the environment and political support in many countries [222,223]. GECF suggests that natural gas use will overtake coal by 2025 and be the largest energy source by 2047 (~ 28%), with oil plateauing in 2040 and commencing an irreversible decline [222]. OPEC projects oil will continue to provide the largest contribution to the energy market (~ 27%) until 2045, followed by gas (~ 25%) and coal (~ 20%) [223].

Natural gas is nonetheless a short to medium term solution, with BP and Shell targeting a reduction of 40% in oil and gas production and investment over the next decade [224]. Oilfield service companies are now also investing in electric pumps, electrification of their road fleets and incorporation of carbon capture, utilisation, storage (CCUS) technologies. A mix of CCUS, hydrogen and bioenergy are projected to eventually meet the energy demands of most industries [222,225]. Gas will remain difficult to replace in most chemical and industrial sectors until at least 2035 but will be increasingly discontinued in power generation in Europe, Japan, and North America [226]. Biomethane (synthetic natural gas), which is predominantly used in heat and power

(40%), followed by transport and mobility (29%) and then residential and commercial use (20%) is projected to grow by 12% by 2025, especially in Europe and North America but is considerably more expensive than natural gas and currently represents just 0.1% of global gas supply [227].

Electrification of flight remains a key objective across the aviation industry, and it is expected that regional routes will be able to be serviced by electric aircraft by 2030 [228]. However, the high energy density to weight ratios required for flight and ongoing limitations of battery technologies currently make electrification of flights > 1500 km, which are associated with ~ 80% of aviation sector CO₂ emissions, inconceivable [54]. With a typical lifespan of 25–30 years [36], many aircraft built today will still be in service in 2050 [36,37], with the aviation sector likely reliant on liquid fuels until the decommissioning of these aircraft at the very earliest. Shipping and heavy-duty transport is also likely to rely on high energy density fuels for the coming decades [7,11].

This timeline highlights the need for the upscaling of natural gas and biomass derived SAF produced using FT synthesis to replace conventional jet fuels until such time that reliance on liquid fuel can be alleviated by new energy technologies [54]. A requirement of 450–500 million tonnes/year of SAF is estimated by 2050, a burden that can be managed without impacting food or water use through the utilisation of a wide range of biomass including MSW, agricultural and forestry residues, used cooling oil, jatropha, halophytes, algae, and non-biological alternative fuels, such as power-to-liquid and sun-to-liquid.

With fuel constituting nearly a quarter (23.7%) of total airline operating costs, the high production costs of SAF, in addition to its lower energy density and efficiency [229], are the major factors limiting its widespread adoption. Additional support and regulation are required to make SAF cost competitive with conventional jet fuels [36,49,53], a process that the International Air Transport Association (IATA) estimates the economic ramifications of climate change policies will achieve by 2030 [39]. The de-risking of SAF production facilities, reducing costs of renewable electricity generation, improving conversion efficiencies and increased investment in SAF technologies will also help to close the gap between SAF and conventional fuel prices [53,229,230].

Electrification and fuel cell technologies are also popular candidates for reducing light duty vehicle GHG emissions over short and medium distances. However, developing and adopting these technologies and their associated infrastructure will take time. With an average vehicle life of 9.3 years, it is estimated that just 6 million electric cars will be on the road by 2030 in Germany while the remaining 85% of vehicles will still utilise internal combustion engines powered by a suitable fuel [11]. Fuel cell electric vehicles (FCEVs) powered by hydrogen and the infrastructure required to fuel them are also in their infancy, with just 7500 FCEV sales worldwide by the end of 2019, although many countries plan to have millions of FCEVs on the road by 2030–2040 [231].

The production of bulk chemicals from renewable resources is expected to reach 113 million tonnes by 2050 suggesting that 38% of all organic chemicals would be derived from agricultural, forestry and other biomass-sourced syngas [103] and an appropriate FT synthesis route using tailored catalysts for the production of olefins for the production of polymers and aromatics [206]. The replacement of fossil-derived chemicals and materials, such as methanol, olefin and BTX, with bio-based alternatives is primarily limited by industry reluctance to adopt new production processes with 2–5 times the cost of existing optimised fossil-based processes [73,102,103,232,233]. This may be eased by a transition to natural gas, followed eventually by gasified biomass, both of which require FT processes to generate useful products, as the chemical sector in particular has raw material, heat and power requirements that require the use of gas [234]. Incorporation of carbon credits within production cost could help to make sustainable products economically competitive with their fossil-derived counterparts [232].

The duration of the use of natural gas as a bridging technology will depend on the rate at which existing infrastructure, vehicles and

industries can be upgraded or replaced and the rate at which renewable energy generation capacity grows. Nonetheless, natural gas will likely play a critical role in meeting climate change targets in a timely fashion and supporting the transition to renewable technologies for at least the next 30–50 years. Appropriate government support in the form of economic incentives, upscaling, carbon taxes or credits will be important over this period [56].

5. Conclusion

The world stands poised on the brink of a radical technological revolution necessary to protect the Earth's environment and hence its inhabitability. However, key sectors, such as aviation, mobility and transport, chemicals, and polymer materials are heavily reliant on liquid fuels and hydrocarbons. With these industries in their current form effectively incompatible with new technologies, bridging solutions will be necessary to decarbonise existing sectors in a timely fashion and sustain civilisation until new products and infrastructure are established. Natural gas provides conventional power generation options and can be used to make liquid fuels and other hydrocarbons using the FT process. These are more environmentally friendly than fossil-derived liquid fuels and compatible with existing processes, products, and infrastructure. Simultaneously, gas could be used as energy storage for renewable energy systems – converted into hydrogen, synthetic natural gas, or heat during periods of low electricity demand and then back into electricity when needed. It could also accelerate the adoption of hydrogen as a fuel in the transport and mobility sector, overcoming hydrogen distribution and storage problems and infrastructure deficiencies by moving methane through existing networks and converting it to hydrogen on-site at fuel stations. This hydrogen production is accompanied by generation of carbon (nano)materials and valuable engineering materials that further improve the environmental sustainability of the process by offsetting emissions in the carbon nanotube and black industries. Finally, excess or undesired carbon can be converted back into syngas as a legacy fuel for obsolete technologies and to alleviate demand on the agricultural sector and energy-food crop competition during a full switch to biofuels. This flexibility, coupled with the unique compatibility of natural gas with both existing technologies and future fuels makes it a critical bridging technology. When it comes to reliable real-world impact, we cannot wait for the promised miracle technologies of tomorrow. The safest bet is the devil we know. With time running out, what we need for now at least is technologies that can be immediately implemented with known, reliable, predictable, large-scale impact.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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