

Towards carbon-neutral and clean propulsion in heavy-duty transportation with hydroformylated Fischer–Tropsch fuels

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Clean transport requires tailored energy carriers. For heavy-duty transportation, synthetic fuels are promising but must fulfil the key challenges of achieving carbon neutrality while reducing air pollution and ensuring scalability through compatibility with existing infrastructure. Here we show that hydroformylated Fischer–Tropsch (HyFiT) fuels composed of optimized alkane–alcohol blends simultaneously address these challenges. First, the design of the HyFiT fuel process flexibly closes the carbon cycle by employing biomass or carbon dioxide as feedstock, while being scalable through mature technologies. Second, fuel testing shows that HyFiT fuels comply with global fuel standards. Material compatibility is demonstrated for two standard sealing materials, enabling the retrofit of today’s vehicle fleets. Third, vehicle testing shows that HyFiT fuels substantially reduce combustion-induced particulate matter and nitrogen oxides. Fourth, a well-to-wheel life cycle assessment finds that HyFiT fuels enable the transition to net-zero greenhouse gas emissions, showing simultaneously a favourable profile in other environmental parameters. HyFiT fuels can thus complement electrification for heavy-duty transportation.

Despite the urgent actions that have been recognized as required to meet climate goals¹, greenhouse gas (GHG) emissions from fossil fuel combustion have risen sharply in the global transport sector, from 5.1 GtCO₂e in 1990 to almost 8.0 GtCO₂e in 2022 (ref. 2). Fossil fuel combustion in the transport sector is also one of the main emitters of the urban air pollutants particulate matter (PM) and nitrogen oxides (NO_x; ref. 3). A key concept for reducing GHG emissions and urban air pollution of the transport sector is renewable power generation combined

with direct electrification of power trains². However, hard-to-electrify transport subsectors such as aviation, shipping, agricultural machinery and heavy-duty trucks in long-haul applications are predicted to rely to a large extent on liquid energy carriers in the future⁴. These applications are envisaged to transition from air-polluting fossil-based fuels towards cleaner bio-based or electricity-based synthetic fuels with improved emission profiles for reduced air pollution⁵. Synthetic fuels benefit from the existing infrastructure for liquid energy carriers and

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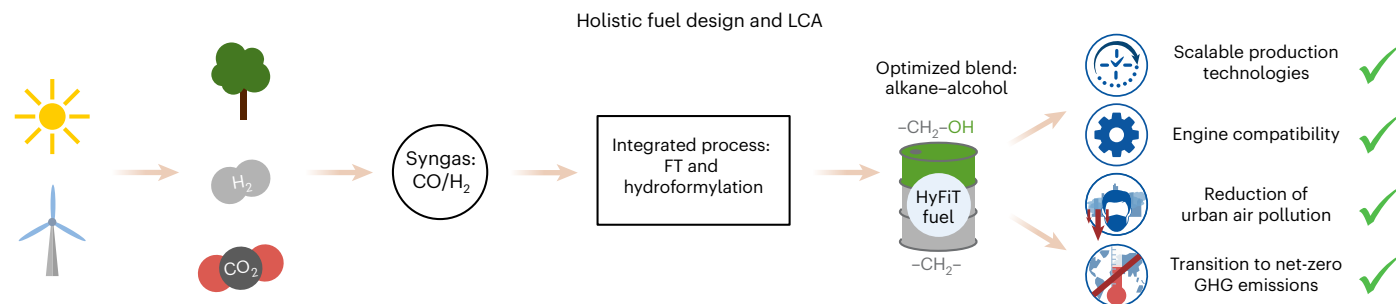


Fig. 1 | Holistic approach of HyFiT fuels. The approach comprises the fuel design and LCA of HyFiT fuels to validate their potential as possible diesel substitutes.

harvest renewable energy from global regions with the potential for low-carbon energy⁶.

In the scientific literature and energy scenarios of future mobility, several synthetic fuels are discussed as promising substitutes for diesel in heavy-duty applications (Supplementary Note 1). Gaseous energy carriers such as hydrogen, methane or dimethyl ether offer high molecular energy densities⁷ but are not compatible with existing infrastructures and power trains⁸. Liquid synthetic fuels include fatty acid methyl ester, hydrogenated vegetable oil, Fischer–Tropsch (FT) fuels, polyoxymethylene ethers and long-chain alcohols. Ideally, their application requires only minor or even no adaptation of existing engines and reduces pollutants from fuel combustion^{9–13}. These fuels are largely compatible with existing fuel infrastructure^{9,14,15}; however, the technological maturity of their production processes varies substantially for large-scale application^{7,16}.

In summary, the design and development of synthetic fuels as drop-in solutions providing additional benefits requires four key factors: first, scalability of the synthesis processes; second, compatibility with fuel standards and components of current engine technology; third, reduction of urban air pollutants; and fourth, transition to net-zero GHG emissions with flexible feedstocks while avoiding shifting the burden into other environmental parameters. Synthetic fuels need to address these four key challenges simultaneously to accelerate the transition towards a clean transport sector.

FT diesel consisting mostly of alkanes already meets several challenges by showing good compatibility with existing engines and fuel infrastructure¹⁴. FT processes can also be operated with syngas from biomass or CO₂ (ref. 14), whereby production can flexibly adapt to local feedstock availability. The GHG reduction potential of FT fuels has already been evaluated in many life cycle assessments (LCAs) with feedstocks of biomass^{17,18} and CO₂ (Supplementary Note 2)^{19,20}. Reported GHG reductions range between 70% and 98% compared to fossil diesel^{17–20}. The low aromatic content and high cetane number of FT diesel enable the reduction of engine-out pollutant emissions, particularly PM (up to 40%) and NO_x (up to 30%)²¹. These engine-out emissions can be further reduced with state-of-the-art engine and exhaust after-treatment technologies.

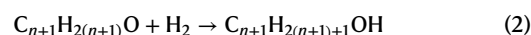
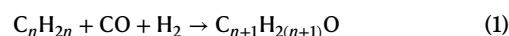
The beneficial emission behaviour of FT diesel can be further improved by exploiting the opportunity to regard fuel composition as an optimization variable⁵ and adding C₆–C₁₀ alcohols, as demonstrated by some of this study's authors and their coworkers^{22,23} as well as other groups^{24,25}. For example, 1-octanol shows exceptionally low PM emissions upon combustion: PM emissions are halved compared to fossil diesel at high part load and almost reduced completely at lower part load¹³. However, 1-octanol is not suitable in pure form as diesel fuel due to its low cetane number²⁶. Blending FT diesel with long-chain alcohols can synergistically combine the advantages of the two options and reduce the drawbacks; that is, suitable fuel properties are maintained while reducing PM emissions by up to 94% compared to fossil diesel^{22,23}.

Here we validate the potential of alkane–alcohol blends as possible diesel substitutes in a holistic approach by, first, developing a conceptual process design for the production of alkane–alcohol blends; second, comprehensively studying their compatibility with fuel standards and engines by fuel testing; third, assessing their combustion properties and emission profiles in heavy-duty vehicle tests; and lastly, carrying out a detailed well-to-wheel LCA using as benchmarks both fossil diesel and battery electric vehicles (BEVs; Fig. 1). The innovative process concept combines at its core the FT synthesis with hydroformylation, that is, two chemical transformations using synthesis gas (CO and H₂) that are already used in different ways on an industrial scale today. With syngas being accessible from biomass, CO₂ or waste and using renewable energy, the feedstock-flexible production process can be fully 'de-fossilized'²⁷. This interdisciplinary study demonstrates the high potential of the resulting HyFiT fuels as a technological solution to harvest renewable energy for hard-to-electrify propulsion systems.

Production process

The proposed process concept for HyFiT fuel production (Fig. 2; details in Supplementary Note 3) aims at producing an optimal alkane–alcohol blend with minimum energy input and maximum carbon yield. The carbon yield describes how much of the carbon present in the input streams is converted to the desired product, that is, the HyFiT fuel. Current scientific efforts to generate alcohols from syngas focus mainly on the design of modified catalysts for the FT synthesis to facilitate the insertion of CO into the growing alkyl chain at the catalytic active centre. Substantial progress has been made with this approach, but alcohol selectivities remain moderate even at low CO conversion and products are limited to the C₂–C₅ range, which is not suitable for diesel-type fuels^{24,25}.

By contrast, our HyFiT production process targets the olefin fraction of FT synthesis as substrates for hydroformylation. In this concept, FT synthesis produces a mixture of mainly saturated hydrocarbons (alkanes) and unsaturated hydrocarbons (olefins). The olefins present in this mixture are further transformed to the corresponding C₁-elongated alcohols by catalytic hydroformylation, that is, the addition of syngas to the C=C double bond (equation (1)), and subsequent hydrogenation (equation (2)). This sequence produces the required amount of alcohols as oxygen carriers in the blends, directly minimizing the need for intermediate separation.



Thus, HyFiT production differs from current FT processes that are optimized to produce saturated hydrocarbons (alkanes) for further upgrading, and instead builds on technically mature FT variants yielding olefin–alkane mixtures as primary products in the C₅–C₂₀ range²⁸.

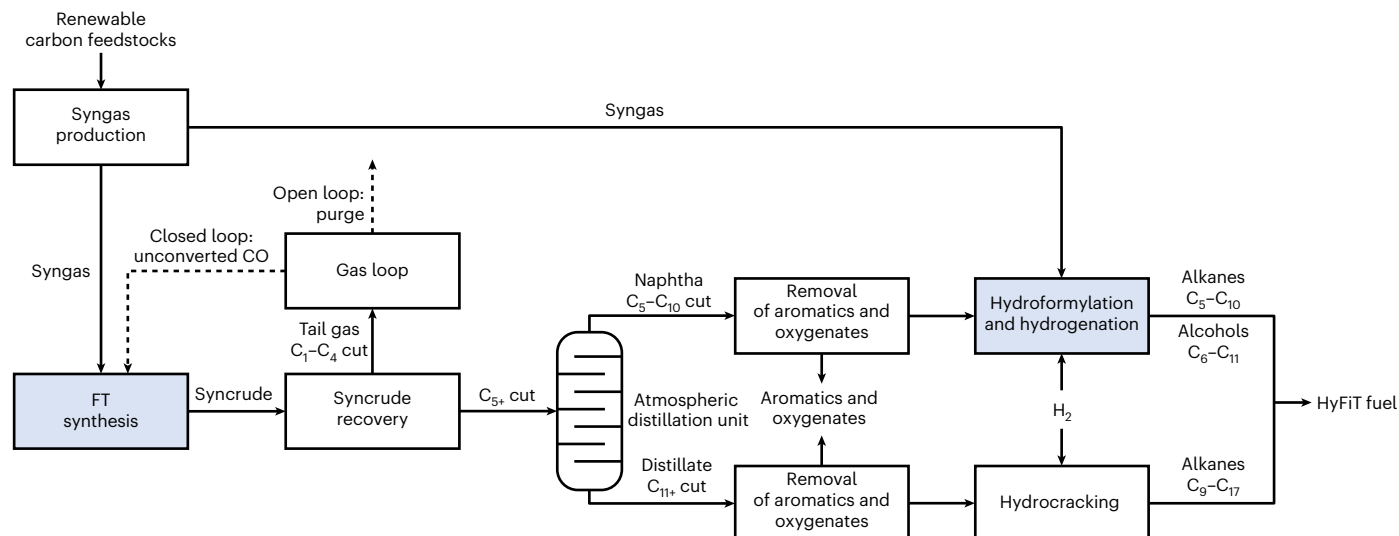


Fig. 2 | Simplified process concept for the production of HyFiT fuels. Process steps are depicted as boxes while arrows denote mass flows. The process concept combines at its core the FT synthesis and hydroformylation (light blue boxes). In the gas loop, tail gas is either purged (open loop) or treated (closed loop) to recycle unconverted carbon monoxide (CO; dashed arrows). Depending on the

supply chain optimization, by-products, for example, aromatics and oxygenates, are either incinerated for heat recovery followed by optional CO₂ capture or flared. Note that we do not differentiate between the LT-FT and HT-FT syntheses for the sake of better readability. Detailed process flow charts of all involved process steps are in Supplementary Note 4.

Isolated pure olefins are converted industrially today to alcohols by catalytic hydroformylation and hydrogenation²⁹. Research by some of us in the framework of the EU project REDIFUEL^{30,31} demonstrated that alkane-olefin product streams from FT synthesis can be hydroformylated directly using industrially relevant catalytic systems^{32,33}.

These developments are exploited in the proposed process design that thus uses syngas as the starting material in its two synthesis steps: syngas is produced from the renewable carbon sources of biomass, CO₂ or a combination thereof. Hydrogen from water electrolysis is used for the conversion of CO₂ and also employed in all subsequent upgrading steps. The FT synthesis is tailored in such a way that its product spectrum is shifted from alkanes towards olefins in the naphtha cut (C₅–C₁₀), since these olefins are desired intermediates to produce alcohols via subsequent hydroformylation. The olefin content in the mixture is converted directly to the C₇-elongated alcohols by selective hydroformylation and hydrogenation without prior separation. Hydrocarbons in the distillate cut (C₁₁+) are sent to a hydrocracking unit and remixed with the product stream, such that the resulting HyFiT fuel consists of C₆–C₁₁ alcohols and C₅–C₁₇ alkanes.

To adjust the alcohol/alkane ratio via the olefin content in the crude FT product (syncrude), the conceptual process model combines two types of FT synthesis: low-temperature (LT)-FT synthesis and high-temperature (HT)-FT synthesis. LT-FT synthesis uses cobalt-based catalysts to provide high chain growth probability α , yielding a syncrude of mostly long-chain alkanes²⁸. Iron-based catalysts are usually used for HT-FT synthesis, resulting in a higher amount of olefins yet at the expense of chain growth. By combining LT-FT and HT-FT syntheses, the alcohol/alkane ratio can be varied in response to the composition optimization in our studies on engine compatibility, combustion emissions and environmental impacts (vide infra). Complementary to our approach, future research in FT synthesis may exploit process improvements through catalyst design^{34,35}, as recently demonstrated by some of us³⁶.

The overall carbon efficiency of the process concept is strongly affected by the fate of unconverted CO from the FT step, which is treated in the gas loop²⁸. This study considers the two extremes of the gas loop: an open-loop design and a closed-loop design. The main text focuses on the open-loop design without tail gas recycling. Results for the closed-loop design are presented in Supplementary Note 14.

To enable rapid deployment, the proposed HyFiT fuel production integrates mature technologies in a new design framework. As a consequence, the resulting HyFiT fuels match and even exceed the carbon efficiency of previous FT fuels, in particular for the bio-based route (Supplementary Note 25). At the same time, the resulting yields of up to 83% at CO conversions above 95% match and even exceed those of C₂–C₅ alcohols formed with state-of-the-art systems with modified catalysts for the FT synthesis (Fig. 3)²⁵.

Engine compatibility

As a potential drop-in fuel, the HyFiT fuel has to fulfil current fuel standards and material requirements. This match was achieved by optimizing the HyFiT fuel's alcohol/alkane ratio in a range from 15 wt% (LT-FT synthesis) to 65 wt% (HT-FT synthesis) to tailor its fuel properties (Fig. 4a–e) and material compatibility (Fig. 4f–h) with common elastomers used in standard engine systems. Experiments were conducted for both pure and additized HyFiT fuels.

A higher alcohol content in the HyFiT fuel increases density and viscosity while the lower heating value (LHV), derived cetane number (DCN) and lubricity decrease. The LHV is not limited in fuel standards and is in the range of fossil diesel (42.8 MJ kg^{−1}) for alcohol contents between 15 and 40 wt%. For the DCN, an alcohol content of 40 wt% or less allows it to stay above the European Standards (Euronorm, EN) 590 limit of 51. While the density values do not comply exactly with the limits of the EN 590 norm, they are still within the range of common global fuel standards, for example, the EN 15940 standard for paraffinic diesel. The density may also be increased by mixing with biodiesel if desired. In terms of viscosity, alcohol contents of 20 wt% or greater meet the requirements of EN 590, while lower alcohol contents are still acceptable for the broader range of fuel standards. The lubricity values can be adjusted to the EN 590 standards with the fuel additive Infineum R655 while the other properties are not affected.

Material compatibility is shown through the change of mass, hardness and volume for the reference elastomers fluorine rubber (FKM), nitrile butadiene rubber (NBR) and hydrogenated NBR (HNBR), which are common in diesel and gasoline systems. While mass and volume change only slightly with increasing alcohol content, hardness is affected. Still, the tolerated limits are practically fully realized

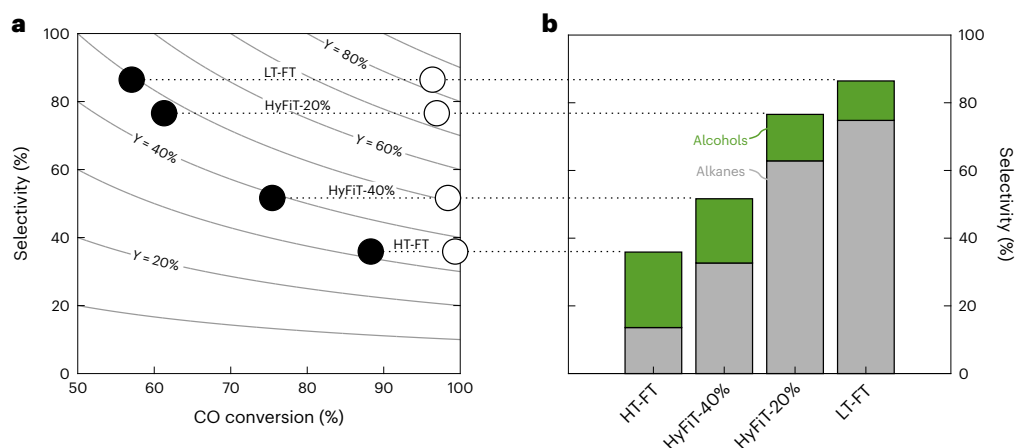


Fig. 3 | Selectivity and yield of HyFiT fuels. a, Selectivity as a function of carbon monoxide (CO) conversion, with yield (Y) values as contour curves. Results are presented for the open (black circles) and closed (white circles) gas loop designs. Dotted lines are guides to the eye. **b**, Alcohol and alkane selectivities of HyFiT fuels. Further details are in Supplementary Note 26.

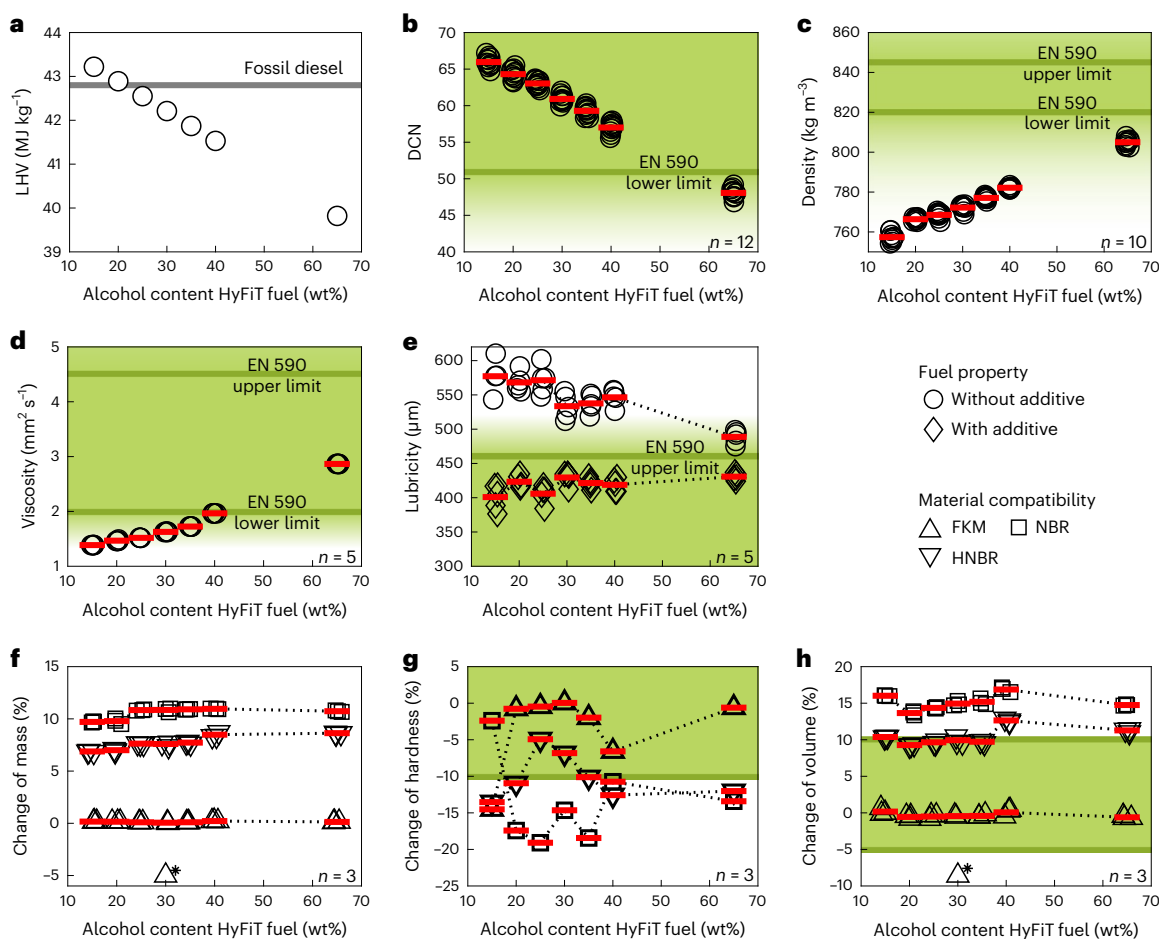


Fig. 4 | Fuel properties and material compatibility of HyFiT fuels at 23 °C. For experimental values, the average of the constituent data points is shown as red horizontal lines, with the corresponding sample size (n) in the bottom right corner. Dotted curves are guides to the eye. **a–e**, Fuel properties include the LHV (**a**), DCN (**b**), density (**c**), viscosity (**d**) and lubricity (**e**). The properties are compared to the EN 590 diesel standard (green area) and a range of common global standards (green colour gradient) for alcohol contents between 15 and 65 wt%. Standards are listed in Supplementary Note 6. **f–h**, Material compatibility

includes the change of mass (**f**), hardness (**g**) and volume (**h**) of FKM, NBR and HNBR for alcohol contents between 15 and 65 wt%. Note that, for change of hardness, solely the mean is shown to ease readability; Supplementary Fig. 10 contains separate plots for each elastomer. Limits are taken from Richter⁵⁰. The measurements for the change of mass and volume of FKM and 30 wt% alcohol gave inconsistent results. This data point is marked with an asterisk and not included in the further analysis. Numerical values are in Supplementary Note 9.

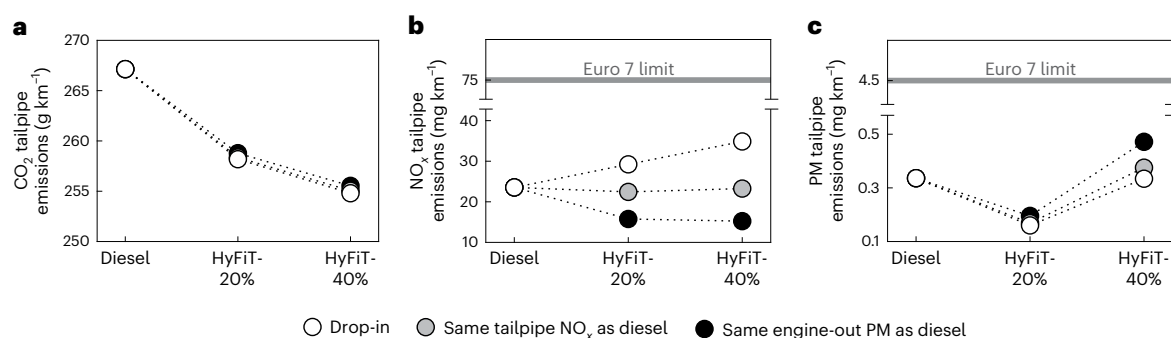


Fig. 5 | Tailpipe emissions from the combustion of diesel and HyFiT fuels.

a–c, Tailpipe emissions of diesel, HyFiT-20% and HyFiT-40%: CO₂ (**a**), NO_x (**b**) and PM (**c**). Results are shown for additized HyFiT fuels and three engine calibration cases: measured emissions from vehicle tests with the manufacturer's engine calibration, denoted as 'drop-in' (white circles); and estimated emissions of HyFiT fuels with an engine calibration that targets either the same tailpipe NO_x

emissions as diesel (grey circles) or the same engine-out PM emissions as diesel (black circles). Limits of the upcoming Euro 7 legislation are shown as grey horizontal (**b** and **c**). Dotted curves are guides to the eye. Numerical values of emissions are presented in Supplementary Table 9. An alternative figure showing PM over NO_x tailpipe emissions is presented in Supplementary Fig. 15.

with the elastomer FKM and at least partly with HNBR. Consequently, compatible reference elastomers for HyFiT fuels are FKM and, with limitations, HNBR.

Overall, the range between 20 and 40 wt% appears most promising for the parameters investigated. In the following, we therefore focus on the HyFiT fuels with 20 wt% alcohol (HyFiT-20%) and 40 wt% alcohol (HyFiT-40%) for the analyses of combustion emissions and environmental impacts.

Combustion emissions from vehicle testing

Synthetic fuels should retain the engine efficiency of diesel while reducing the pollutant emissions of PM and NO_x. Both pollutants are therefore evaluated for diesel and additized HyFiT fuels (HyFiT-20% and HyFiT-40%) within a heavy-duty van for three engine calibration cases (Fig. 5): emissions are measured in vehicle tests with the manufacturer's calibration, denoted as 'drop-in', and the NO_x reduction potential of HyFiT fuels is estimated with two additional calibrations described below.

Even under drop-in calibration (white circles in Fig. 5), HyFiT fuels reduce CO₂ emissions by 3–5% compared to diesel (Fig. 5a), mainly due to the more favourable carbon-to-hydrogen ratio of 0.45 for HyFiT fuels compared to 0.53 for diesel³⁷. Under these conditions, HyFiT fuels increase engine-out NO_x emissions by around 8% (Supplementary Fig. 14). In combination with lower average exhaust gas temperatures and a correspondingly slower warm-up of the exhaust gas after-treatment system, higher engine-out NO_x emissions result in higher NO_x tailpipe emissions for HyFiT-20% (29 mg km⁻¹) and HyFiT-40% (35 mg km⁻¹) than for diesel (24 mg km⁻¹; Fig. 5b). However, these NO_x tailpipe emissions are still far below the limits of the upcoming Euro 7 legislation (75 mg km⁻¹).

The main advantage of HyFiT fuels is substantially reduced PM engine-out emissions (Supplementary Fig. 14): by 55% for HyFiT-20% and by 70% for HyFiT-40% compared to fossil diesel. PM is reduced for three reasons. First, HyFiT fuels benefit from a much higher oxygen content (2.5–5.0 wt%) than fossil diesel (0.8 wt%)³⁷ due to the incorporated alcohols. In addition, HyFiT fuels contain no aromatics due to the FT chain growth mechanism and feature high volatility for better mixture formation. The PM reduction is a similar order of magnitude to that in studies with mixtures of fossil diesel and 1-octanol²². Therefore, the increased oxygen content is a decisive effect here, due to the incorporated alcohols. In combination with a standard particulate filter, almost no PM emissions can be measured in the exhaust gas after the tailpipe (Fig. 5c).

The reduced PM engine-out emissions enable the reduction of NO_x emissions from HyFiT fuels below diesel level by adapting the exhaust gas recirculation rate. We thus adapt this rate until HyFiT fuels have

either the same tailpipe NO_x (grey circles in Fig. 5) or engine-out PM emissions (black circles) as diesel. In the second case, NO_x tailpipe emissions are reduced to 16 mg km⁻¹ (HyFiT-20%) and 15 mg km⁻¹ (HyFiT-40%), corresponding to a reduction of 33–38% compared to diesel. While both calibrations reduce NO_x at the cost of slightly increased CO₂ and PM, HyFiT-20% can reduce all three emissions substantially compared to diesel.

In summary, a drop-in of HyFiT fuels into current engines would already lead to tailpipe emissions of PM and NO_x far below the limits of the upcoming Euro 7 legislation. An adapted exhaust gas recirculation strategy allows one to exploit the potential of HyFiT fuels to reduce NO_x emissions substantially below the diesel level. HyFiT-20% is favourable over HyFiT-40% since it yields lower PM and NO_x tailpipe emissions with only slightly higher CO₂ emissions. Consequently, for the proceeding analysis of environmental impacts, we focus on HyFiT-20% with an adapted exhaust gas recirculation rate for 'same engine-out PM as diesel'.

Carbon footprint and environmental impacts

The central motivation for synthetic fuels is reducing the well-to-wheel carbon footprint from transportation, while avoiding other major negative environmental impacts. The following LCA therefore assesses the well-to-wheel environmental impacts of using HyFiT-20% in a hybrid heavy-duty van under the energy system scenario for the year 2030 within a global setting. The environmental impacts are compared with two benchmarks: a hybrid heavy-duty van powered by fossil diesel as today's benchmark and a fully battery electric van as a future benchmark. The functional unit of this LCA is 'the provision of 1 km of transportation in a heavy-duty van'. We focus on climate change by optimizing HyFiT fuel supply chains for minimal carbon footprint. A potential burden shift to other environmental impacts is analysed for these climate-optimal supply chains in Supplementary Note 16.

The system boundary contains the supply of feedstocks and utilities, and the production of syngas, HyFiT fuel and the battery, as well as driving the van to provide the service of transportation (Fig. 6). Syngas can be produced from biomass or CO₂ (ref. 38). The presented open-gas-loop design results in the lowest carbon footprint; Supplementary Note 14 contains results with the closed-gas-loop design and HyFiT-40%.

The carbon footprint of bio-based HyFiT fuel is lower than that of diesel and even competitive with that of the BEV over the entire range of electricity impacts (Fig. 7a). This result is robust with respect to variations of battery lifetime and capacity as well as land use change emissions from biomass (Supplementary Notes 22 and 23). The carbon footprint of bio-based HyFiT fuel also depends much less on low-carbon

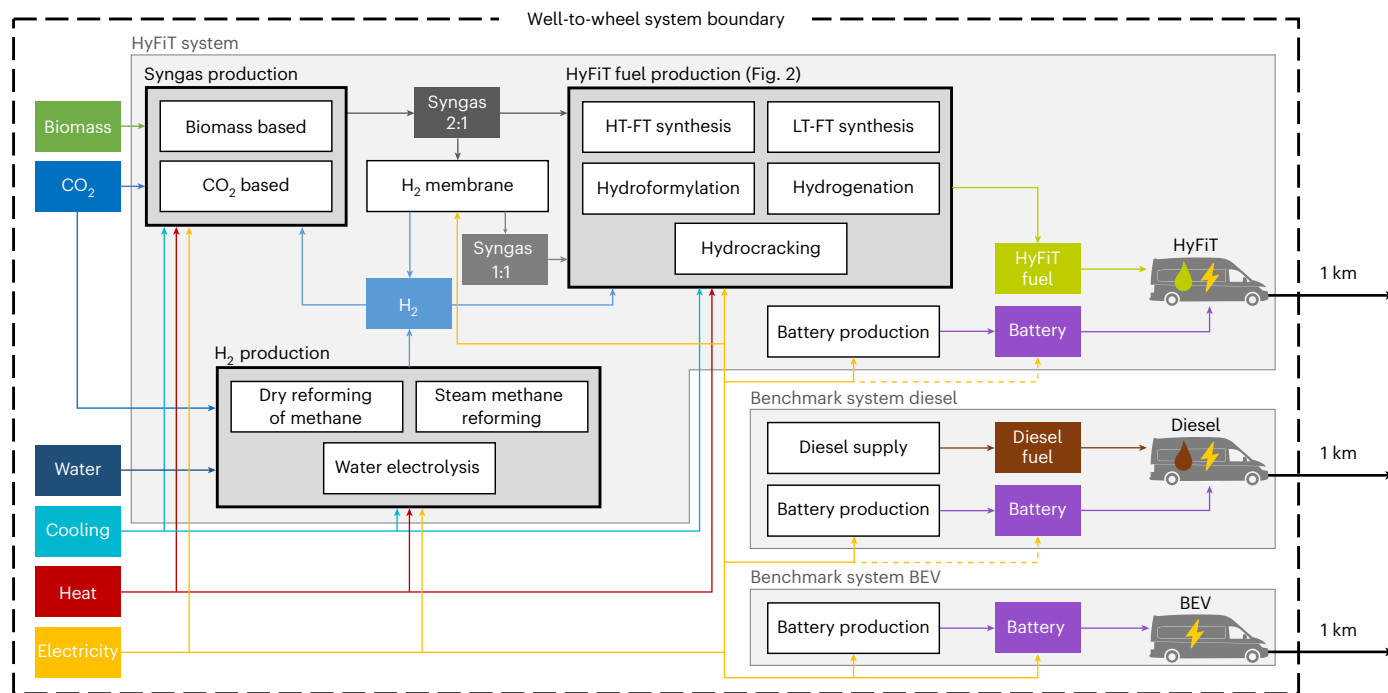


Fig. 6 | System boundary of the well-to-wheel LCA. Systems under study are the HyFiT system (top), benchmark diesel system (middle) and benchmark BEV system (bottom). The functional unit for all three systems is ‘the provision of 1 km of transportation in a van’. In the HyFiT and benchmark diesel systems, the hybrid van has a battery with a capacity of 14 kWh in addition to its internal combustion

engine. By contrast, in the benchmark BEV system, the fully electric van has a battery capacity of 153 kWh. Syngas for HyFiT fuel production is produced from biomass or CO₂. Note that the battery is recharged by fuel combustion at the end of a hybrid vehicle’s test cycle, which is why electricity for battery charging is shown as a dashed arrow for the hybrid vans.

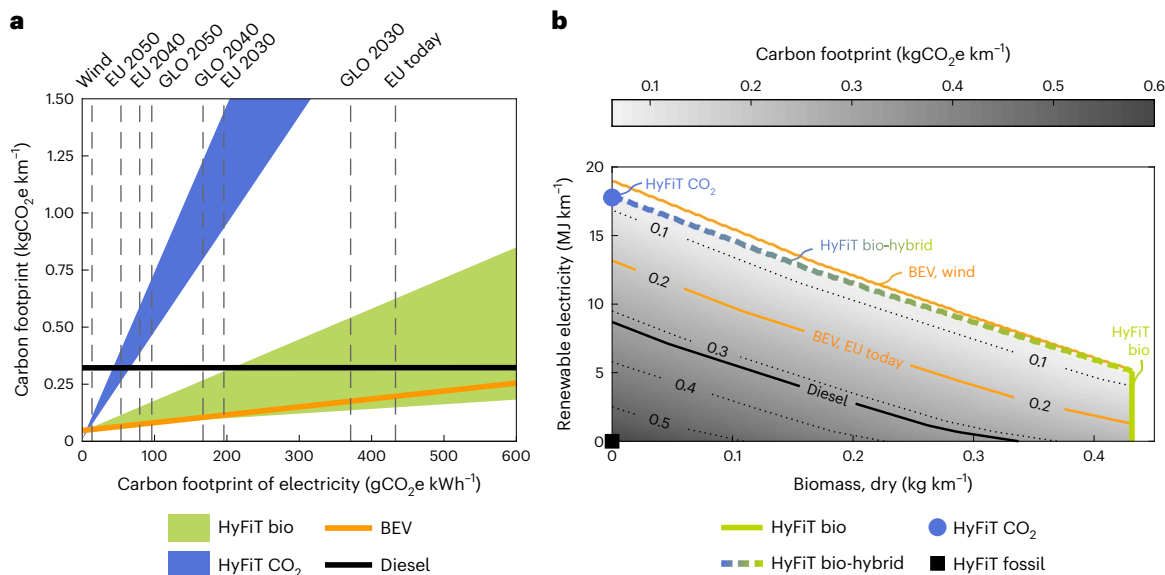


Fig. 7 | Well-to-wheel carbon footprints of HyFiT fuels and the benchmark systems. **a**, The well-to-wheel carbon footprint of the van powered by bio-based or CO₂-based HyFiT fuel as a function of the carbon footprint of electricity. The multiple production routes for each feedstock are represented as coloured ranges. Technologies of the supply chain designs are described in Supplementary Note 13. Benchmark results are additionally shown for diesel and BEV. EU, European Union; GLO, global. **b**, The well-to-wheel carbon footprint of HyFiT fuels as a function of available renewable electricity and biomass. A purely

bio-based HyFiT fuel supply chain is indicated by the green vertical line, whereas the dashed curve represents a bio-hybrid, that is, bio-based and CO₂-based, supply chain. The circle and rectangle show a HyFiT fuel supply chain purely based on CO₂ and fossil fuels, respectively. If renewable electricity is limited, additional electricity is taken from the electricity grid. For reference, contour curves are shown for the carbon footprint of a diesel and BEV with either electricity from today’s EU grid or wind power.

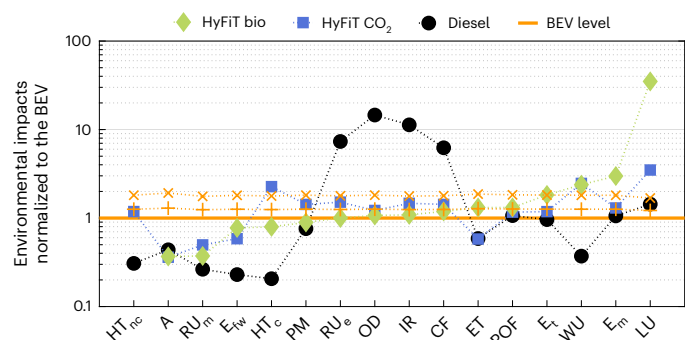


Fig. 8 | Well-to-wheel environmental impacts of HyFiT fuels and the benchmark systems. The impacts of bio-based (green) and CO₂-based (blue) HyFiT-20% fuel and diesel (black) are normalized to the environmental impacts of the BEV (orange). The impact categories on the x axis are sorted by the normalized impacts of the bio-based HyFiT fuel. Results are shown for the supply chains with electricity from wind power as a best-case proxy for a future low-carbon grid (Fig. 7a). The BEV's battery capacity is varied to 200 kWh (+ symbol) and 300 kWh (× symbol), corresponding to driving ranges of 550 km and 850 km, respectively. Dotted curves are guides to the eye. Contribution and sensitivity analyses are in Supplementary Notes 18–22. HT_{nc}, non-carcinogenic human toxicity; A, acidification; RU_m, resource use of minerals and metals; E_{tw}, freshwater eutrophication; HT_c, carcinogenic human toxicity; PM, particulate matter; RU_e, resource use of energy carriers; OD, ozone depletion; IR, ionizing radiation; CF, carbon footprint; ET, ecotoxicity; POF, photochemical ozone formation; E_e, terrestrial eutrophication; WU, water use; E_m, marine eutrophication; LU, land use.

electricity than does the carbon footprint of CO₂-based HyFiT fuels. This reflects that the carbon in the bio-feedstock is already chemically reduced as compared to CO₂. As a result, CO₂-based HyFiT fuel requires low-carbon electricity below 65 gCO₂e kWh⁻¹ to break even with diesel. While this target is even lower than in the forecasted EU grid mix in 2050, production could be powered by off-grid renewable energy or in favourable geographic regions.

The analysis shows that the availability of either the carbon source biomass or the power source low-carbon electricity defines the limiting factors for large-scale applications of bio-based and CO₂-based HyFiT fuels (Fig. 7b), respectively. However, the flexibility of the HyFiT fuel concept enables adapting the supply chain design regarding the feedstock used if input availability is limited (Supplementary Note 15). Thereby, optimized 'bio-hybrid' supply chain designs can be identified that yield carbon footprints in the same range as those of a BEV using electricity from wind power or today's EU grid.

The results further show that reducing GHG emissions does not lead to major burden shifting for most other environmental impacts (Fig. 8). In long-haul applications, bio-based HyFiT fuel reduces environmental impacts compared to a purely battery-driven power train in 10 or 12 of the 16 impact categories of the Joint Research Centre of the European Commission's Environmental Footprint 3.0 if driving ranges greater than 550 km or 850 km are required, respectively. The BEV's environmental impacts are dominated by its large battery in these applications, which is required to achieve such long driving ranges. Similar conclusions can be drawn if reduced battery life times are assumed (Supplementary Fig. 24a). A notable trade-off results in the land use due to the biomass demand. Land use can be again reduced by a 'bio-hybrid' approach combining CO₂ and biomass as the carbon feedstock.

Discussion

Tailored HyFiT fuels are found in this comprehensive study to meet the key requirements for 'de-fossilized' drop-in fuels with improved properties. First, the conceptual process design combining FT synthesis and hydroformylation allows flexible production of HyFiT fuels from

biomass and CO₂ based on mature technologies. Second, fuel testing demonstrates that the main HyFiT fuel properties lie within the range of common global fuel standards and show compatibility with engine components such as standard reference elastomers. Third, HyFiT fuels allow for efficient propulsion and show exceptional emission behaviour, as shown by vehicle testing: both PM and NO_x emissions can be reduced substantially and are far below the limits of the upcoming Euro 7 legislation even when used as drop-in fuels. Lastly, the potential of HyFiT fuels to enable the transition to net-zero GHG emissions is confirmed in the well-to-wheel LCA. The burden shift to other environmental categories is similar to or even below that of an electrified power train in long-haul applications. Consequently, HyFiT fuels are promising energy carriers for retrofitting today's vehicle fleets.

The integrated process concept starting from synthesis gas enables flexible value chains to produce HyFiT fuels depending on the regional availability of renewable energy or feedstocks. The HyFiT fuels based on biomass as feedstock require comparably less energy input and can compete with fully battery electric vans in terms of the electricity required to reduce the carbon footprint even with today's electricity grid mix of the EU. While biomass can be a limiting resource and land use is a critical factor, CO₂ is an abundant carbon source even in post-fossil scenarios. HyFiT fuels based on CO₂ require large amounts of low-carbon electricity to compete with BEVs, however, similar to other electricity-based fuels^{39,40}. On a systemic level, combining the two options could balance these trade-offs.

Any technology targeting low-carbon propulsion systems will impact other environmental categories such as resource use of minerals and metals, water and land. These trade-offs must be considered in future technology development. Notably, bio-based HyFiT fuels show a lower impact compared to a battery electric van in 10 or 12 of 16 impact categories, if driving ranges greater than 550 km or 850 km are required, respectively. These characteristics illustrate the potential of HyFiT fuels as a complementary, rather than competing, approach to the enormous challenge of CO₂ reduction in all areas of mobility and transportation. Their compatibility with existing infrastructure for the distribution and storage of liquid energy carriers renders HyFiT fuels a potential drop-in fuel employing drop-in technology. In our view, the HyFiT fuel concept should encourage the industry to invest in synthetic fuels and build the required infrastructure, and to dislodge the impression that synthetic fuels serve only as a distraction to prolong the use of fossil fuels.

Future studies should carefully assess the cost and capacity development timeline of HyFiT fuels—two key challenges we did not touch on in this study due to scope. On the one hand, the early adoption of bio-based synthetic fuels has been shown to provide both environmental and economic benefits immediately in the existing infrastructure with current propulsion technology, which is expected to remain an important part of future mobility globally⁴¹. To enable a fast capacity development timeline, one design criterion for the HyFiT fuels was combining individual production technologies that have already proven their robustness and scalability in today's industrial reality. On the other hand, recent studies highlight that particular electricity-based synthetic fuels, such as CO₂-based HyFiT fuels, would require strong policy support to render their timely market penetration economically viable⁴². Such policies should favourably allocate synthetic fuels to those applications where liquid energy carriers are expected to remain the preferred option⁴³, for example, heavy-duty trucks in long-haul transportation. Consequently, this application area was the focus of our LCA study.

Methods

Production process

In Supplementary Note 3, we give an in-depth description of the proposed process concept for HyFiT fuel production, comprising all involved process steps. Detailed process flow charts are presented in

Supplementary Note 4. Supplementary Note 5 provides the modelled fuel compositions of HyFiT fuels with varying alcohol/alkane ratios.

Engine compatibility

For the additized fuel, 2,000 ppm of the additive R655 is blended with the fuel, which is within the range suggested by Luecke and Zigler⁴⁴.

Fuel properties. Determined fuel properties are the LHV and, as standardized in the EN 590, density, viscosity, lubricity and DCN. While the HyFiT fuel's LHV is calculated, the other properties are measured in experiments. The density is determined by buoyancy tests using the Archimedes principle. The viscosity is measured using an Ubbelohde-type viscometer, according to Deutsches Institut für Normung (DIN) standard 51562. For the lubricity, high-frequency reciprocating rig tests were conducted, as described in DIN, EN and International Organization for Standardization (ISO) standard 12156-1. The DCN was determined using the Advanced Fuel Ignition Delay Analyzer (AFIDA), following the ASTM International D6890 and D8183 standards. More details are in Supplementary Note 7.

Material compatibility. The experiments on compatibility with sealing materials follow DIN and ISO standard 1817. Therein, the change of mass, hardness and volume is evaluated for the three reference elastomers FKM, NBR and HNBR, according to DIN and ISO standard 13226. More details are in Supplementary Note 8.

Combustion emissions

For vehicle testing, a light commercial van was investigated with a vehicle mass of 3.3 t, a plug-in hybrid power train with a battery capacity of 14 kWh and an electric motor with 95 kW peak power^{45,46}. Based on its maximum vehicle mass of 5.5 t when fully loaded, the baseline conventional vehicle was certified according to the Euro 6d standard for heavy-duty vehicles. Emissions from fuel combustion were measured for diesel and additized HyFiT fuels in the worldwide harmonized light-vehicles test cycle with the maximum speed limited to 120 km h⁻¹. To ensure the reproducibility of the provided emission results in the worldwide harmonized light-vehicles test cycle and under these testing boundary conditions, the vehicle tests were repeated several times for each fuel, in order to primarily eliminate the driver's effect on the results. For plug-in hybrid vehicles, the test procedure provides a charge sustain mode: the difference between the state of charge of the battery at the beginning and end of the test cycle must not exceed 1%. However, state of charge deviations greater than 1% occurred for two reasons: the LHV values of HyFiT fuels do not correspond to those of diesel, and the vehicle calibration was not adjusted to the HyFiT fuels. Consequently, fuel consumption, PM emissions and NO_x emissions were corrected according to these state of charge deviations to maintain comparability between different fuels. More details on the state of charge correction are presented in Supplementary Note 10.

Life cycle assessment

The methodology of the LCA is standardized in the ISO 14040 and 14044 standards and enables the holistic evaluation of environmental impacts of production systems and products along their entire life cycle.

Goal and scope. The goal of this LCA is to assess the well-to-wheel environmental impacts of using HyFiT fuels in a hybrid heavy-duty van in 2030 within a global setting. These environmental impacts are compared with two benchmarks: a hybrid heavy-duty van powered by fossil diesel as today's benchmark and a fully battery electric van as a future benchmark. The applied system boundary contains the supply of feedstocks and utilities and the production of syngas, HyFiT fuel and the battery, as well as driving the van to provide the service of transportation (Fig. 6). We neglect vehicle construction except

for the battery since the environmental impacts of fuel-powered and battery electric vans differ mostly in terms of direct emissions, battery production and the supply of energy carriers for driving, that is, fuel or electricity⁴⁷.

Functional unit and impact assessment. Comparing production systems consistently requires a common basis. This common basis is called a functional unit in an LCA. Here we define the functional unit as 'the provision of 1 km of transportation in a heavy-duty van'. We characterize all material and energy flows exchanged with the environment according to the method 'Environmental Footprint 3.0' recommended by the Joint Research Centre of the European Commission⁴⁸. Thereby, we consider all impact categories covered by the Environmental Footprint 3.0 method (Supplementary Note 12). Note that we use the term 'carbon footprint' when referring to climate change.

Life cycle inventory. The life cycle inventory is described in detail in Supplementary Note 12, along with all considered LCA datasets in Supplementary Table 11.

Supply chain optimization. The HyFiT fuels' potential for net-zero GHG emissions is analysed by minimizing the carbon footprint of the HyFiT fuel supply chain (Fig. 7a). As HyFiT fuels can be produced from multiple feedstocks, we optimize the supply chain for each feedstock, that is, biomass and CO₂. Previous studies have shown that the carbon footprint of CO₂-based fuels depends strongly on the carbon footprint of electricity^{39,40}. In the optimization, we thus vary the carbon footprint of electricity to assess its influence on the potential of, in particular, CO₂-based HyFiT fuels. For the carbon footprint minimization with limited availability of renewable electricity and biomass (Fig. 7b), we do not predefine feedstock-specific technology sets. Thus, the optimizer can freely choose and combine all included HyFiT fuel production routes. Additional electricity is taken from the grid if renewable electricity is limited. In Supplementary Note 16, we additionally evaluate the carbon-footprint-optimal supply chains for environmental impacts beyond carbon footprint in detail.

Data availability

All unrestricted data used and generated in this study are provided in the Article, Methods and Supplementary Information or via Zenodo at <https://doi.org/10.5281/zenodo.10390761> (ref. 49). However, third-party data used from ecoinvent for the LCA and IHS Markit for processes in the synthesis gas production network are proprietary data from commercial sources. These third-party data cannot be published, as user licences are required to access the underlying data. To gain access, ecoinvent and IHS Markit can be contacted via <https://ecoinvent.org/> and <https://ihsmarkit.com>, respectively. Source data are provided with this paper.

Code availability

The code to conduct the analysis is publicly available via Zenodo at <https://doi.org/10.5281/zenodo.10390761> (ref. 49). However, running the entire code requires proprietary third-party data from ecoinvent and IHS Markit. These third-party data are not included in the repository, as user licences are required to access the underlying data (as described in the 'Data availability' statement).

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Author contributions

S.V., M.B., L.M., B.L., A.V., S.P., W.L. and A.B. conceived the concept. S.V., N.G., M.B. and L.M. developed the methodology. S.V. and N.G. designed the process model for fuel production, calculated the key performance indicators and LHV, performed the LCA and visualized the results. M.B. provided the process model of the synthesis gas production network. M.N. carried out experiments to determine the DCN values and analysed the results. T.K. and P.M. performed vehicle testing to determine the combustion emissions and analysed the results. M.H. carried out experiments to determine the lubricity, density, viscosity and material compatibility and analysed the results. B.L., K.S., S.P., W.L. and A.B. supervised and obtained funding and resources for the project. S.V., N.G., L.M., T.K., B.L. and M.H. wrote the paper. S.V., M.B., B.L., A.V., K.S., S.P., W.L. and A.B. revised the paper.

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Competing interests

The authors declare the following competing interests: S.P. and W.L. have a patent pending for producing a fuel for internal combustion engines that combines FT synthesis and hydroformylation (applicant: Rheinisch-Westfälische Technische Hochschule (RWTH) Aachen; inventors: W.L., Benedikt Heuser, S.P.; patent number: EP3649098). A.V. and W.L. have a patent pending for producing long-chain alcohols also for fuels that integrates FT synthesis and hydroformylation in one synthetic step (applicant: Max-Planck-Gesellschaft zur Förderung der Wissenschaften e.V.; inventors: A.V., W.L., Gonzalo Prieto, Kai Jeske, Thorsten Rösler, Maurice Belleflamme; patent number: WO 2023/280720 A1). A.B. served on review committees for research and development at ExxonMobil and TotalEnergies, oil and gas companies that are also active in fuel production. A.B. has ownership interests in firms that render services to industry, some of which may produce fuels. The other authors declare no competing interests.

Additional information

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