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Techno-economic and environmental assessment of BECCS in fuel generation for FT-fuel, bioSNG and OMEx†

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This study focuses on bioenergy with carbon capture and storage (BECCS) in fuel generation and assesses the potential of biofuel generation to decarbonise the fuel economy by reducing CO₂ emissions to the atmosphere. The research investigates the technical, economic, and environmental performances of three biofuel production routes, namely Fischer–Tropsch synthesis (FTS), bio-synthetic natural gas (bioSNG) and oxymethylene ethers (OMEx) synthesis using flowsheets developed in Aspen Plus. It constitutes the first attempt to holistically evaluate both the techno-economic performance and the environmental benefits of employing BECCS in fuel generation. For an input of 1020 dry tonnes per day of woody biomass, the FTS route yields 275 t d^{−1}, the bioSNG route yields 238 t d^{−1} and the OMEx route yields 635 t d^{−1} of fuel and the energy efficiency is in the range of 44.9% to 59.7% without CCS and 44.0% to 58.2% with CCS. In addition, negative emissions can be achieved for all routes with CCS in the range of 301 000 to 519 000 tCO₂ per year. For economic viability, the minimum selling price for FT-fuels, bioSNG, and OMEx production with CCS have been calculated as £23.4 per GJ, £14.5 per GJ and £26.5 per GJ, respectively. However, competition with conventional fossil-derived fuels is not possible without the combination of existing financial incentives and a proposed carbon pricing. With carbon credit as the only financial incentive, carbon pricing in the range of £48 to £86 per tCO₂ needs to be applied to achieve feasibility. Also, more negative emissions need to be generated to decrease the value of this range and reasonably phase out dependence on fossil-derived fuels. Parametric studies identified as crucial parameters to be improved the fuel output, CAPEX, operating hours and feedstock cost.

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1 Introduction

The concentration of greenhouse gases in the atmosphere is on the increase and as human civilisation relies heavily on energy, it is of utmost importance that the drastic effects of these gases in the atmosphere should be avoided. A proposed solution to mitigate emissions is the employment of negative emissions technologies (NETs).¹ The contribution of NETs in meeting the 2050 targets, set by the Paris Agreement in 2015, needs to be better understood.² One such technology is bioenergy with carbon capture and storage (BECCS). Based on the concept of negative emissions, BECCS can play a significant role in limiting global warming when deployed under the appropriate conditions. Previous and current BECCS research has focused on its role in power generation neglecting the possible benefits in fuel generation. Compared to BECCS in power generation, carbon capture is intrinsic in the fuel generation plant, thus

creating an initial economic advantage. BECCS In fuel generation can assist the decarbonisation of the transport and heating sectors.

To this end, the current study examines the technical, economical, and environmental feasibility of three fuel production routes using a second-generation biomass, *i.e.*, lignocellulosic woody biomass, as a potential BECCS technology. As many economies move towards decarbonisation, conventional fuels cannot be displaced due to their importance, instead they can be replaced by alternatives that can help meet the goal. Suitable alternatives involve conversion of biomass by various routes. Biomass is an important fuel in a low-carbon economy because of its composition and similar processes to fossil fuels that it can undergo. Also, it should be noted that in this work, since carbon capture is an integral part of each production process, carbon capture and storage (CCS) refers to the captured CO₂ compression, transportation, and storage. The use of second-generation biomass in this study is rooted in the fact that the production of this class of biomass does not compete with food production and also that the technology for these routes are readily available but not developed to a commercial scale.^{3–5} Previous studies^{6–10} on the biofuels

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production from second-generation biofuels has solely focused on the need to increase biofuel production to displace fossil-fuels but the economic and environmental impact of employing CCS has not been fully considered.

Biomass is considered carbon-neutral because the CO₂ released during combustion is absorbed to regenerate more biomass. This is assuming that no CO₂ emissions are given off during the process but in reality, emissions are given off during the planting, harvesting, transporting and processing of biomass so the use of biomass results in many cases in net positive emissions.¹¹ Hence, while biofuels have the potential to remove CO₂ emissions from the atmosphere, a less than optimal route could result in net positive emissions to the atmosphere. This study focuses on the use of biomass in addition to CCS to produce biofuels while removing CO₂ from the atmosphere. In addition to the technical and economic feasibility of the routes examined, the environmental impact is determined in terms of the mitigation potential. The routes considered are the common Fischer–Tropsch synthesis (FTS) to produce hydrocarbons, methanation to produce bio-synthetic natural gas (bioSNG) and oxymethylene ethers synthesis to produce OMEx. All three production routes are thermochemical processes and selected based on a common gasification process. Fischer–Tropsch synthesis fuel (FT-fuel) is chosen because it is an alternative for the most common liquid fuels in transportation – gasoline and diesel. BioSNG is chosen as the alternative for the most common gaseous fuel – natural gas and OMEx is chosen because it is a relatively novel biofuel which is derived from methanol and it has shown potential in diesel engines; one of which is its ability to serve as an almost carbon-neutral component when 24% (wt) is blended with diesel.¹²

The FTS is not a new process and has been in existence since the 1920s and it has been commercially used to produce liquid transportation fuels from coal or natural gas.¹³ The process involves reacting hydrogen (H₂) and carbon monoxide (CO) over a catalyst to form a range of hydrocarbon chains of varying length. The most common ones are the South African SASOL coal-to-liquid (CTL) technology and the Shell gas-to-liquid (GTL) technology.¹⁴ The use of biomass in FTS has since been researched and a commercial scale plant producing 15 000 tonnes of biofuel per annum was established by CHOREN. However, it requires producing a large volume of 100 000 tonnes for profitability.¹⁵ Methanation is the conversion of CO and carbon dioxide (CO₂) to methane (CH₄) and water *via* hydrogenation. The UK has commissioned a commercial bioSNG plant using up to 175 000 tonnes of bio-resources, including unrecyclable wood and refuse-derived fuel (RDF).¹⁶ The Swedish GoBiGas project which produced 20 MW of bioSNG demonstrated good results but plans to move to large-scale production were terminated.¹⁷ Oxymethylene ethers (OMEx) synthesis, is an emerging technology and interest into this has increased over the past decade.¹⁸ While OME₁ studies as fuel is more common, interest in higher OMEs (OME₃–OME₅) are being considered due to their behaviour in diesel engines. The main precursor for OMEx is methanol which can be produced from syngas. While methanol itself is a useful fuel, its low flammability makes it unsuitable in compression engines

and its high toxicity increases operating expenditure.¹⁹ The production process includes methanol synthesis and formaldehyde synthesis which are established processes.

Techno-economic assessments provide information to support the deployment of technologies and identify areas of improvement. Most of the existing literature related to BECCS deals with the power generation sector.^{20–25} Some of the literature consider the addition of CCS in fuel production, albeit without much information on the environmental impact and the required policy framework necessary for the biofuel routes to be competitive with fossil fuels. Tagomori *et al.*²⁶ performed a techno-economic assessment of FT-diesel production from eucalyptus and pine residues with the CCS case increasing production costs by 1%. Song *et al.*²⁷ carried out an assessment on four different agriculture residues to produce methane and the addition of CCS increased the production cost in the range of 3–4%. Michailos *et al.*²⁸ investigated the feasibility of coupling CCS with syngas biomethanation and concluded that the addition of CCS increases the production costs by roughly 17%. De Álamo *et al.*²⁹ studied the techno-economic feasibility of adding CCS to FTS and bioSNG and estimated that production costs increase by 10% and 14%, respectively. The current study updates the work in the literature and adds much more information on the economic and environmental impact of deploying BECCS in the investigated fuel production routes. The results of this study answer the following questions:

- Is biofuel generation suitable to create substantial negative emissions?
- Are there opportunities present in current processes to maximise carbon removal?
- Are the biofuels produced competitive with conventional fossil fuels in transportation and what tools can help in transitioning to a low-carbon economy?

2 Methodology

2.1 Process simulation and description

All the examined processes were modelled using the sequential-modular approach in the Aspen Plus V10 software. Aspen Plus is capable of simulating large complex processes even those involving solids and non-ideal components such as biomass. Thermodynamic models were used to solve the mass and energy balances in the system. Generally, the Peng Robinson equation of state with Boston Mathias alpha function (PR-BM) was chosen to estimate phase equilibria and properties, and steam tables for the power generation section.³⁰ The enthalpy of formation, specific heat capacity and density of the non-conventional components (biomass and ash) were estimated using the HCOALGEN and DCOALGIT property methods.³¹ Modelling techniques and approaches applied in this study along with respective process flow diagrams and process conditions are detailed in the ESI.†

In Aspen Plus, the gasification process can either be equilibrium modelled or kinetic modelled. The complexity of the involved reactions prevents the simulation of the gasification process as a single unit operation model in Aspen Plus, so the process is simulated using various reactor models. In this work,

equilibrium modelling^{32,33} is applied across all processes due to its suitability for providing good estimates as is the scope of this work and create a solid foundation for techno-economic assessments. The acid gas removal (AGR) section with MEA is not modelled in Aspen Plus but instead the mass and energy requirements are calculated using equations provided in the gas processors suppliers association (GPSA) Engineering Data-book.³⁴ There is little information on OME_x modelling which is mostly based on kinetics so as to simplify the OME_x reactor, mass yield fractions are implemented based on the work of Schmitz *et al.*³⁵ In the gasification section, two types of gasifiers are considered based on the final product. An entrained flow gasifier (EFG) is used in FTS and OME_x synthesis while a dual fluidised bed (DFB) gasifier is used in methanation. This is because a DFB gasifier promotes the formation of methane thereby maximising the final output (bioSNG) while the entrained flow gasifier destroys hydrocarbons such as methane formed during the gasification process. The cryogenic air separation unit for the EFG is not modelled in this work. Instead it is represented by an MCOMPR block and the power requirements are calculated based on the report in ref. 36 that a cryogenic air separation system consumes about 260 to 340 kW h of energy per tonne of oxygen produced with 90% used by the main compressor depending on the plant capacity.

2.1.1 Biomass preparation. Wood is used as the main feedstock in all production routes. The estimated amount of waste wood biomass used in the UK in 2019 was 3.98 million tonnes per year³⁷ and based on the figures in previous years, this figure is expected to increase year on year. In 2013, the overall potential availability of wood fibre in Britain was 15.6 million tonnes per year and this figure is expected to grow up to over 18 million tonnes per year by 2029.³⁸ Currently in the broad energy sector, wood is utilised for CHP, while studies³⁹ have proposed due to sufficient availability the production of biofuels too. The selected feed rate of wood to the plant of 1020 dry tonnes per day (dt d⁻¹) is well below the national wood availability and it is in line with previous biofuel techno-economic studies.^{8,40,41} This size corresponds to large scale production and as such the project can benefit from the economies of scale. In this study, it is assumed that the wood is received without any pre-processing. All drying and processing is performed on the plant and the wood moisture content on a wet basis (wb) is 15%. The composition of the wood is detailed in Table 1.

In the preparation section of the plant, wood is dried and milled to 1 mm for EFG and 6 mm for the DFB gasifier. For the entrained flow gasifier, a chopper is first employed to reduce the size of the wood to 12 mm and then a grinder is used to further reduce the size to 1 mm. The electrical requirement for the chopper is scaled from Swanson *et al.*⁸ while the electrical requirement of the grinder is calculated using the regressions from Mani *et al.*⁴³

The chopped wood is sent to the drier to reduce the moisture level to 10%. This is achieved by steam drying and the reaction is modelled in an RSTOIC reactor based on the equation:

Table 1 Proximate and ultimate analysis of woody biomass.⁴²

White wood pellet	wt%
Proximate analysis	
Moisture (wb)	15
Volatile matter (db)	83.7
Fixed carbon (db)	15.55
Ash (db)	0.75
Ultimate analysis (db)	
C	51.89
H	6.79
N	0.16
S	0.02
Cl	0.01
O	40.38
Ash	0.75
LHV (MJ kg ⁻¹)	16.28
Price (£ per tonne)	50

A calculator block using FORTRAN statements defined in Aspen Plus determines the amount of steam to the reactor and the extent of drying (the final moisture content is 10%).

2.1.2 Biomass gasification. The gasification process involves several steps which can be split into different zones in the gasifier. The steps include the drying stage where the moisture content of biomass is reduced and removed in the form of steam; the pyrolysis stage where the biomass feed is thermally decomposed in the absence of an oxidising agent releasing volatile matter, tar and char; and the oxidation stage where the volatile matter and some of the char undergo oxidation with limited oxygen present in the gasifying media supplied to the gasifier.

The gasifying media could be air, oxygen (O₂), steam (H₂O) or carbon dioxide (CO₂). However, the choice of gasifying agent affects the heating value of the product gas. Air is the poorest choice as it results in the lowest heating value range of 4–7 MJ Nm⁻³ (ref. 44) because the high amount of nitrogen present in the product gas dilutes the quality of the product gas. Steam is a better gasifying agent which gives a product gas with a heating value in the range of 10–18 MJ Nm⁻³; this higher heating value is because of the increased H/C ratio. Pure oxygen results in a product gas with a heating value in the range of 12–28 MJ Nm⁻³ (ref. 44) however, caution needs to be exercised when using oxygen; Ghassemi and Shahsavan-Markadeh⁴⁵ observed that while increasing the O/C content would increase the cold gas efficiency and heating value of the product gas, there is a threshold beyond which the effect becomes negative.

Gasification occurs in different types of gasifiers – fixed or moving bed, fluidised, and entrained flow bed. The choice of gasifier is dependent on the range of application and desired outcome. Fixed or moving bed gasifiers include the updraft gasifier, downdraft gasifier and crossdraft gasifier, which are suitable for applications in the range of 10 kW to 10 MW but it experiences problems such as tar production and entrainment, and difficulty maintaining uniform temperature due to poor mixing. The fluidised bed gasifier includes the bubbling fluidised bed and circulating fluidised bed gasifiers which are

capable of perfect mixing and maintaining a uniform temperature. Fluidised bed gasifiers operate at moderately high temperatures (1000 °C) but experiences char entrainment in the product gas; a cyclone is used to solve this problem. The entrained flow gasifier (EFG) is commonly used in large scale operations and basically acts like a plug flow reactor. Conditions in the gasifier are such that the residence time of biomass particles in the reactor is short, carbon conversion is up to 99.5% and the product gas contains very little or even no tar. These conditions include high temperatures of up to 1600 °C and high pressure, up to 80 bar, and finely reduced biomass below 75 µm.⁴⁴

The gasification module consists of a combination of unit operations. An RYIELD reactor present in both gasifiers decomposes the non-conventional biomass into conventional components based on the ultimate analysis. The entrained flow gasifier is modelled after the GE gasifier which is approximately at equilibrium using an RGIBBS reactor, which estimates the composition of the syngas product by minimising the Gibbs free energy. The option to restrict equilibrium by specifying the temperature of individual reactions is employed and applied to the water-gas shift reaction, see (eqn (4)). The equations modelled in the gasifier are given as follows:

Steam and oxygen are used as the gasifying agents. The oxygen to carbon mole ratio is set at 0.25 while the steam to carbon mole ratio is set at 0.75 based on the stoichiometric equation for syngas formation.⁴⁶

Oxygen at 95% purity from an air separation unit is fed in at 149 °C and 28 bar while steam generated on the plant is fed in at 120 °C and 28 bar. Before the dried biomass goes into the Gibbs reactor, it is pressurised in a lock hopper using CO₂, an inert gas, produced from the AGR section at 0.09 kg kg⁻¹ dry biomass as reported by Higman and van der Burgt⁴⁷ for gasifiers at 25 bar. The EFG operates at 1300 °C and 28 bar.

The DFB gasifier is based on the Göteborg biomass gasification project (GoBiGas) plant in Sweden that produced bioSNG from woody biomass gasification.⁴⁸ In the DFB gasifier, gasification occurs in one fluidised bed and the heat for gasification

is generated in the other fluidised bed. In this gasifier, since air is introduced in a different zone, the syngas is practically nitrogen free. The gasifier model is validated against experimental data from Alamia *et al.*⁴⁹ (see ESI, S.2.1†). While thermodynamic modelling is suitable for first estimates and determining the limits of the system, it underestimates hydrocarbons (C₁, C₂, tars) production and overestimates hydrogen production from gasification.^{32,50,51} In this model, to reduce the inaccuracies from thermodynamic modelling, non-ideal corrections are applied to account for the formation of hydrocarbons especially methane and tar. This is based on similar equilibrium modelling techniques in ref. 52–54.

After the RYIELD reactor, a separator is used to remove a portion of the char (assuming incomplete conversion of carbon) that is burnt in a combustor to provide heat for the gasifier. The rest of the biomass goes to an RSTOIC reactor where hydrocarbon formation is simulated. The hydrocarbons considered are C₂H₂, C₂H₄, C₂H₆, C₆H₆, C₇H₈ and C₁₀H₈ (tar). The fractional carbon conversion of each reaction is set to match the experimental data in Alamia *et al.*⁵⁵ After this reactor, the hydrocarbons formed are separated from the biomass stream to prevent destruction in the Gibbs reactor which is where the rest of the stream is directed to form the remaining components of the resulting syngas – H₂, CO, CO₂, CH₄, NH₃, HCl, H₂S and H₂O. Steam is introduced to the gasifier for steam to biomass (dry ash free) mass ratio of 0.5. The DFB gasifier operates at 1.24 bar and 870 °C. The syngas composition for both gasifiers are presented in Table 2.

2.1.3 Syngas cleaning and conditioning. Syngas leaving the gasifier contains impurities such as tar, particulate matter (PM) and poisonous gases including hydrogen sulphide (H₂S), sulphur dioxide (SO₂), hydrogen chloride (HCl) and ammonia (NH₃). These contaminants need to be removed before downstream applications to prevent hazards including plugging, deactivation of catalysts, and corrosion which would consequently affect the process efficiency.^{56,57} The gas cleaning method is based on the cleaning temperature – hot gas cleanup and cold gas cleanup.

In this study, the cold gas cleanup method is applied as it is the most common method across fuel production processes.

Table 2 Syngas compositions from EFG and DFB gasifier (vol% dry basis)

	EFG gasifier	DFB gasifier
H ₂	47.5	45.4
CO	40.8	23.1
CO ₂	8.34	16.5
N ₂	3.35	0.09
H ₂ O (vol% wet)	21.5	4.37
CH ₄	1.42 × 10 ⁻²	8.24
NH ₃	6.75 × 10 ⁻⁷	2.85 × 10 ⁻⁸
HCl	6.4 × 10 ⁻⁷	5.14 × 10 ⁻⁷
H ₂ S	3.54 × 10 ⁻⁷	0.01
C ₂ H ₂	—	0.12
C ₂ H ₄	—	1.98
C ₂ H ₆	—	0.18

For the EFG, the syngas is cooled by direct-contact water quench to 203 °C and then slag from melted ash is removed from the syngas using a separator. Wet scrubbing using water is applied in a flash unit to simulate removal of impurities such as NH₃, HCl and Cl₂. In FTS, the H₂/CO ratio is 1.17 which quite low. To adjust the ratio to the optimal value of 2.1 for FTS, a sour water-gas shift (SWGS) reactor is used. This reaction is modelled in an REQUIL reactor. For suitable WGS activity, and to reduce the volume of the SWGS reactor, only a portion of the syngas undergoes the reaction. To achieve the optimal ratio, a design specification to set the temperature of the reactor and a calculator block to set the steam flow rate to the reactor at 3 times the flow rate of CO in syngas to the reactor are used. In OMEx synthesis, the H₂/CO ratio is adjusted to 3 for methanol synthesis in an SWGS reactor. This is achieved in an REQUIL reactor and a design specification where the flow rate of steam is varied.

The adjusted syngas is further cooled, and water removed in three flash units before the acid gas removal (AGR) section. In the AGR section, CO₂ and H₂S are removed using 30 wt% MEA. This is not modelled in Aspen Plus but the reboiler duty, cooling duty and pump requirements are calculated based on the equations in the Engineering Data Book³⁴ and results are presented in Table 3. In OMEx synthesis, the amount of CO₂ removed is set by the stoichiometric ratio of (H₂ – CO₂)/(CO + CO₂). This is set at 2.1 as the optimal value should be greater than 2.⁴⁰ Also, this keeps the CO₂ composition in the syngas at ≤7% in order to improve the methanol productivity.⁵⁸ In FTS, CO₂ and H₂S removal are set at 96% and 99%, respectively.

In methanation, after the dual fluidised bed gasifier, tar is condensed from the raw syngas in a three-phase flash unit and recycled to the combustor to be burnt alongside char for heat generation.

After this, the cyclic hydrocarbons–benzene and toluene—are removed from the syngas and also recycled to be burnt with tar and char. This occurs when the raw syngas is passed through an activated carbon bed modelled by a separator unit. Before removing the rest of the impurities, the syngas is compressed to the methanation pressure of 16 bar then cooled to 40 °C for acid gas removal using 30 wt% MEA. CO₂ and H₂S removal are set as the same in FTS. A guard bed located after the AGR section and before the water-gas shift (WGS) reactor absorbs impurities to prevent possible contamination of catalysts upstream. In the WGS reactor, a design specification to maintain a desired H₂/CO ratio of 3.5 in the clean syngas is used. This is due to the

optimal value of the methane synthesis being greater than 3 as seen in the equation:

2.1.4 Fuel synthesis

2.1.4.1 Fischer–Tropsch synthesis. In the FTS, syngas is compressed to 25 bar and heated to 200 °C before passing through a zinc-oxide guard bed modelled using a separator unit to reduce H₂S to 200 ppb/50 ppb to avoid catalyst poisoning in the FTS reactor. Before the FTS reactor, a pressure swing absorber (PSA) isolates a stream of hydrogen from a fraction of the syngas for hydroprocessing of wax downstream of the FTS reactor. The FTS reactor operates at 25 bar and 200 °C using a cobalt-based catalyst⁸ and is modelled as a RSTOIC reactor. The product distribution was estimated by the Anderson–Schulz–Flory (ASF) model (described in Song *et al.*⁵⁹) with a chain growth factor of 0.9 and carbon monoxide per-pass conversion of 40%.⁸ This was implemented using a calculator block with FORTRAN statements. The output of the FTS reactor consisted of unconverted syngas, light gases and hydrocarbons. The hydrocarbons produced were all alkanes from C₁–C₂₀ and C₃₀. Alkanes from C₅ to C₁₀ were grouped as naphtha, C₁₁ to C₂₀ were grouped as diesel and C₂₀ to C₃₀ was wax. The products were separated in a series of flash units and vacuum distillation (RADFRAC) columns. Wax was further cracked to naphtha and diesel using the hydrogen stream isolated earlier on in a RSTOIC reactor operating at 370 °C and 0 bar. The unconverted syngas was recycled to the process with one stream going from product recovery to the FTS reactor, and another stream going to the syngas cleaning section. A small portion of the unconverted syngas stream was sent to the power generation to prevent accumulation in the FTS reactor and generate power for the plant. The mass fraction that returns to the FTS reactor is 0.75; this figure is not optimised and it was selected in order for the FTS case to be a near zero net electricity scenario.

2.1.4.2 Methanation. The methanation process is modelled after the GoBiGas setup.⁴⁹ In a premethanation reactor, hydrocarbons present in the syngas are cracked using steam while some of the CO and CO₂ present are converted to methane. The steam to hydrocarbon ratio is set at 0.5.⁶⁰ Syngas goes into methanation at 16 bar and 300 °C. Methanation takes place in a series of four reactors without recycle based on the Topsøe Recycle Energy-efficient Methanation Process (TREMPE) using the MCR catalyst.⁶¹ Coolers are situated between reactors due to the exothermic nature of the reaction and to maintain catalyst activity. The heat removed is recovered later in the process to generate steam for parts of the plant. Steam is added to the first reactor to prevent the formation of carbon on the catalyst. The methanation reactors are modelled as RGIBBS reactors with a pressure drop of 0.5 bar in each reactor. BioSNG is upgraded to recover unconverted H₂ and reduce CO₂ in the final product.

2.1.4.3 Oxymethylene ethers synthesis. OMEx is synthesised from methanol and formaldehyde so the first step in this process is the conversion of syngas to methanol. Methanol synthesis was modelled after the ICI Syntex Methanol process

Table 3 Acid gas removal (AGR) estimated parameters

Acid gas parameter	FT synthesis	Methanation	OMEx synthesis
Circulation rate (m ³ h ^{−1})	537.4	382.5	634.7
Absorber pressure (bar)	22.1	15.6	24.8
Stripper pressure (bar)	1.01	1.01	1.01
Reboiler duty (MW)	49.9	35.5	59.0
Condenser duty (MW)	20.8	14.8	24.6
Amine cooler duty (MW)	10.4	7.4	12.3

available from the Aspen Plus database. Syngas is compressed to 80 bar in two stages, and then cooled to 230 °C before the reactor. In the ICI Syntex process, the methanol reactor is modelled as four RPLUG reactors in series with fresh syngas introduced between each reactor for cooling. In this work, the reactor is modelled as a single REQUIL reactor operating at 250 °C and 80 bar (see the ESI, S.6†). The reactions occurring in the reactor include the WGS reaction in eqn (4) and the following:

Crude methanol is cooled to 38 °C and flashed where most of the methanol leaves in one stream. The other stream containing unconverted syngas and some methanol is compressed and recycled to the reactor. However, a fraction of this purged and sent to power generation to prevent build up. The methanol stream is sent to methanol recovery where a couple of flash units and distillation (RADFRAC) columns are used to separate methanol from CO₂ and H₂O. The CO₂ recovered in this part of the plant is sent to compression in the CCS plant while H₂O is sent to wastewater treatment.

In an RSTOIC reactor operating at 200 °C and 3 bar, methanol (MeOH) is converted to formaldehyde (FA) using air at a conversion rate of 87%. Based on the stoichiometric reaction and required conversion, the air flow to the reactor is determined by a calculator block. The product contains 37 wt% FA. In the OMEx reactor, H₂O, FA and MeOH go through a series of reactions forming hemiacetals and glycols, which in turn react with FA and MeOH to form OMEx. The reaction pathways are detailed in Schmitz.⁶² In this work, this is modelled as an RYIELD reactor operating at 96 °C and 3.04 bar over Amberlyst-36 catalyst based on mass fractions calculated from the mass balance in the model by Ai.⁶³ Ai⁶³ modelled the OMEx synthesis and this consisted of a CSTR and distillation columns to separate the products and recycle OME_{1-2,4-8}, FA, and MeOH. The energy requirements in distillation of the products (modelled as separator units) were calculated from this work at 44.7 MJ kg⁻¹ of OMEx produced for the heating duty and 47.1 MJ kg⁻¹ for the cooling duty. The final OMEx product is a mix of OME₃–OME₅.

2.1.5 Power and steam generation. Heat recovery from syngas cleaning is used to generate steam for different parts of the plant. In FTS, unconverted syngas is burnt in a combustor with 10% excess air to fully combust the purge gas. The flue gas generates electricity after passing through a gas turbine before heat recovery from the flue gas to generate steam for the drier, gasifier and SWGS reactor. Also, heat from the syngas cooling section is recovered to generate steam for the steam turbine. In methanation, a portion of the bioSNG produced (20% in both cases) is sent to a combined cycle where electricity is produced to meet the plant demand. Heat is recovered from the flue gas in a similar manner in order to generate steam for parts of the plant (gasification, WGS reactor, methanation) and the steam turbine. Other streams for heat recovery include the flue gas from the dual gasifier and intermediate streams from the

methanation reactors. In OMEx synthesis, methanol purge streams from the methanol synthesis are burnt and used to generate steam to drive the steam turbine.

2.1.6 CO₂ compression. For the cases with CCS, the CO₂ captured from the amine plant needs to be compressed and liquefied (supercritical fluid) before pipeline transportation and geological storage. This is achieved by multiple compressors with interstage cooling, and then a pump and a final cooler. CO₂ is compressed up to 110 bar⁶⁴ and cooled to 25 °C (see ESI, S8†). This final condition is based on the phase diagram of CO₂,⁶⁵ at this pressure and temperature, CO₂ exists as a supercritical fluid and can be easily transported using pipelines to a final underground storage site. It is essential that impurity and water levels are kept below the recommended levels⁶⁵ to prevent corrosion of the transportation pipelines. To achieve this, CO₂ entering the CO₂ compression undergoes gas conditioning to minimise the concentration of impurities and water level.

2.2 Economic assessment

An economic model was built to determine the feasibility of production of each product as well as monitor the effect of certain parameters on the price. Discounted cash flow rate of return (DCFROR) analysis, net present value (NPV) break-even analysis to estimate the minimum selling price (MSP) of the biofuels. The major economic assumptions are listed in Table 4. These assumptions are well aligned with previous studies^{8,41,66} while a discount rate of 10% constitutes a conventional and legitimate market assumption. Also any salvage value is assumed to be fully offset by decommissioning costs.

2.2.1 Capital, operating and maintenance expenditures. The total capital investment (TCI) estimates are commonly based on the total purchased equipment costs (TPEC), installation factors and contingency. The methodology for TCI estimation is listed in Table 5. Non-installed direct costs are broken down into 29% buildings, 12% yard improvements and 6% land while indirect costs are broken down into 32% engineering and supervision, 34% construction expenses and 23% contractor's fee and legal expenses.^{8,41}

In this work, the TCI for each plant was calculated using the factorial estimation method and the costs for the major

Table 4 Process economic assumptions

Parameter	Value
Location	United Kingdom
Currency	GBP
Base year	2018
Project lifetime (years)	20
Construction period (years)	2.5
Start-up time (years)	0.5
Capacity factor (%)	85
Tax rate (%)	30
Equity/Debt (%/%)	100/0
Discount rate (%)	10
Depreciation	Straight-line
Depreciation period (yr)	10
Salvage value (£)	0

Table 5 Total capital investment (TCI) estimation methodology

Parameter	Method
Total purchased equipment cost (TPEC)	Aspen process economic analyzer, open literature
Total installed cost (TIC)	TPEC installation factor
Non-installed direct costs (NDC)	47% of TPEC
Indirect costs (IC)	89% of TPEC
Contingency (CC)	20% of TPEC
Fixed capital investment (FCI)	TIC + CC + NDC + IC
Working capital (WC)	15% of FCI
Total capital investment (TCI)	FCI + WC

components were taken from the open literature. Other unit operations that are not available in the open literature were estimated using the Aspen Process Economic Analyzer (APEA). However, due to the installation factors by APEA being generally low, an overall installation factor of 3.02 was used as suggested by Peters *et al.*⁶⁷ for solid-liquid plants. Equipment costs were calculated using eqn (14).

$$\text{Cost} = \text{Cost}_0 \times \left(\frac{\text{Scale}}{\text{Scale}_0} \right)^n \quad (14)$$

where 'Cost₀' and 'Scale₀' represent the cost and capacity of the base unit; 'Cost' and 'Scale' represent the estimated cost and actual size of the plant equipment; and 'n' is the scaling factor. The base capacity and cost for units obtained from the open literature are listed in Table 11 in the Appendix. After converting currencies using the average yearly exchange rate of the corresponding years, the costs were converted to GBP using the Chemical Engineering Plant Cost Index (CEPCI). The scaling factors were obtained from Tagomori *et al.*, Thunman *et al.*, and Sinnott and Towler.^{26,68,69}

The operating and maintenance (OPEX) costs (summarised in Table 12 and Table 13 in the Appendix) covers every other cost for the day-to-day running of the plant. This includes labour costs, maintenance, insurance and purchase of raw materials. For this study, the fixed costs were estimated using guidance from Peters *et al.*, and Sinnott and Towler.^{67,68} The labour costs were estimated using data from Glassdoor and Payscale.^{70,71} The number of employees and shifts were derived from Phillips *et al.*⁷² In FTS and methanation, there are 8 maintenance technicians and 20 shift operators while in OMEx synthesis, there is an extra section for methanol synthesis, increasing the number of maintenance technicians and shift operators to 12 and 30, respectively. The cost of CO₂ transport and storage is taken as variable cost and equal to £19/tCO₂.⁹⁰

2.2.2 Levelised cost of fuel (LCOF). The levelised cost of fuel represents the cost of producing a unit of fuel. This includes the amortized capital investment over the life of the plant and the yearly operating and maintenance costs. For each process, it is calculated as follows:

$$\text{LCOF (£ per GJ)} = \frac{(\text{FCF} \times \text{TCI}) + \text{OPEX} - \text{Revenues from by-products}}{Q_{\text{fuel}} \times \text{CF} \times 8766} \quad (15)$$

$$\text{FCF} = \frac{i(1+i)^n}{(1+i)^n - 1} \quad (16)$$

where 'FCF' is the fixed charge factor determined by the discount rate (*i*) and plant lifetime (*n*); 'TCI' is the total capital investment in GBP, 'OPEX' is the yearly operating costs including the cost of feedstock in £ per year; 'by-products' is the revenue generated from sale of by-products such as electricity in £ per year; 'Q_{fuel}' is the fuel energy output in GJ h⁻¹; 'CF' is the plant availability; and 8766 represents the total number of hours in a year.

2.2.3 Minimum selling price (MSP). After determining the capital, operating and maintenance costs, the minimum fuel selling price is determined using a DCFROR analysis. The MSP is the calculated at the selling price of fuel when the NPV is equal to zero at the fixed discount rate over the plant lifetime. This is achieved using the NPV function and solver in Excel. The equation for the DCFROR is outlined as follows:

$$\sum_{n=1}^{n=20} \frac{\text{CF}_n}{(1+i)^n} = 0 \quad (17)$$

$$\text{CF}_n = P(1 - t) + Dt \quad (18)$$

where 'CF_n' is the cash flow in year *n*; 'P' is the gross profits in year *n*; 't' is the tax rate; and 'D' is the depreciation.

2.2.4 CO₂ avoidance cost

The CO₂ avoidance cost represents the minimum carbon tax to be paid when the CCS plant is compared to a similar plant without CCS. In comparison to a similar plant, the cost is calculated on the basis of transportation and storage costs as capture is included in all scenarios. The avoidance cost is calculated as follows:

$$\text{CO}_2\text{avoided (£ per tCO}_2\text{)} = \frac{\text{LCOF}_{\text{CCS}} - \text{LCOF}_{\text{ref}}}{(\text{tCO}_2 \text{ per MW h})_{\text{ref}} - (\text{tCO}_2 \text{ per MW h})_{\text{CCS}}} \quad (19)$$

where LCOF is the levelised cost of fuel and (tCO₂ per MW h) is the emission factor to the atmosphere. In this study, the CO₂ avoidance cost is compared to two plants; the first plant is a conventional fossil-derived fuel production plant such as the production of natural gas or diesel (Plant A), and the second plant is a biorefinery where biomass is converted to a biofuel

(Plant B). On both plants, CO₂ is not transported and stored, rather it is released to the atmosphere.

2.2.5 Financial tools

In the UK, there are several laws and policies that have been adopted to fight climate change. Most of these laws and policies put financial incentives in place to support the roll out of energy efficient and greenhouse gas reduction technologies. Two of these policies are considered in this study for transport fuels. The first one is the Renewables Transport Fuel Obligation (RTFO)⁷³ which was introduced to support the supply of transport fuels from renewable energy sources in to support the UK's government policy to reduce GHG emissions from vehicles and drive the supply of renewable fuels. Under this scheme, suppliers that meet the eligibility criteria are issued Renewable Transport Fuel Certificates (RTFCs) for each litre or kg of biofuel produced. The second one is the non-domestic Renewable Heat Incentive⁷⁴ which provides financial support for generating renewable heat for non-domestic purposes. Another financial tool considered in this study is carbon pricing which is commonly used in emissions trading schemes.

2.3 Process performance indicators

The performance of each plant can be quantified by several indicators. These performance indicators determine how efficient a production process is. The performance of the gasifier is determined by the cold gas efficiency which is calculated as the energy content of the resulting syngas against the energy content of dry ash free biomass sent to the gasifier.

$$\text{CGE}\% = \frac{m_{\text{syngas}} \times \text{LHV}_{\text{syngas}}}{m_{\text{biomass}} \times \text{LHV}_{\text{biomass}}} \times 100 \quad (20)$$

where m is the mass flowrate in kg s⁻¹ and LHV is the lower heating value in MJ kg⁻¹.

Another important technical parameter considered is the overall energy conversion efficiency and it represents the amount of biomass energy present in the fuel generated. This value also considers the electricity input and output on the plant.

$$\eta = \frac{Q_{\text{fuel}} + \text{El}_{\text{out}}}{Q_{\text{biomass}} + \text{El}_{\text{in}} + Q_{\text{in}}} \quad (21)$$

where Q is the energy content in MW and El_{in} is the net electricity imported, El_{out} is the net electricity exported from the plant both in MW and Q_{in} is purchased heat in MW.

3 Results and discussion

This section covers the technical, environmental, and economic results and interpretation of the three production routes. For each route, with and without CCS scenarios are investigated. Then the results are compared to the conventional fossil-derived fuel counterparts. In addition, financial analysis is presented for all cases and a sensitivity analysis has been carried out for the CCS cases.

3.1 Mass balance

The diagrams in Fig. 1 show the mass balance on the plant based on the carbon mole flows. In both FTS cases, 43.9% of carbon in the feedstock to plant is stored in the resulting FT-fuels. In the FTS + CCS case, 46.8% of carbon is captured, and 9.3% is vented in the flue gas from the power generation while 0.1% is lost in wastewater; in the FTS case, 56.1% is vented to the atmosphere. In both bioSNG cases, 32.6% of carbon is stored in bioSNG; in the bioSNG + CCS case 32.5% is captured and 34.9% is vented in the flue gas from the power generation and gasification; without CCS, 67.4% is vented to the atmosphere. In both OMEx synthesis cases, 52.9% of carbon to process is stored in OMEx. In OMEx + CCS, 45.4% of carbon is captured and 1.7% is vented to the atmosphere from waste streams; without CCS, 47.1% of carbon is vented to the atmosphere. The system with the least CO₂ venting is in the OMEx production while the most venting occurs in bioSNG production due to the flue gas generated from the dual fluidised gasifier.

3.2 Energy balance

In all production routes, the common sources of heating demand include the distillation columns, gasifier, dryers, intermediate heaters, and power generation unit. The sources of cooling demand include distillation columns and intermediate coolers. Suitable heat integration was performed to recover useful heat from different streams and minimise heat waste on the plant. This allowed manufacture of low pressure to high pressure steam where necessary for use in different locations in each plant. Where further heating was required, fired heat from natural gas was used. Further cooling was achieved with air, cooling water, and refrigerants. Electricity was imported from the grid to meet power requirements in the case that a plant is not self-sustaining. Table 6 shows a breakdown of the power usage and generation for each production route.

3.3 Plant efficiency

The cold gas efficiency, given in eqn (20) is a technical parameter that represents the efficiency of the EFG and DFB gasifiers. A heat loss from the reactor is assumed as 1% of the biomass energy input. The efficiency of the EFG was calculated from Aspen at 87.2% and that of the DFB gasifier was calculated at 70.1%. The difference in these efficiencies lies in the operation of the gasifiers. In the DFG gasifier, further efficiency is lost due to char and tar formation when compared to the EFG which operates near equilibrium and destroys tar and char formed during the gasification process. The summary of the results from the process simulation are presented in Table 7.

For all the biofuels considered, without CCS, the bioSNG route has the highest energy efficiency at 59.7% while the FT-fuel route has the lowest at 44.9%. With the CCS route, it is the same with bioSNG at 58.2% and the FT-fuel route at 44%. Across all three cases, the addition of CCS results in no more than a 1.5-percentage point drop. Energy efficiency loss in routes using the EFG (FTS and OMEx synthesis) is due to the cooling method of the raw syngas. Quench cooling is used and,

Fig. 1 Carbon balance investigated for the production routes (a) Fischer–Tropsch synthesis, (b) methanation and (c) oxymethylene ether synthesis.

in this way, sensible heat that would otherwise be recovered for heat integration is lost. Cool gas cleanup is used with the EFG and this induces energy penalties on the plant but switching to hot gas cleanup can reduce waste streams and costs while the overall plant efficiency is improved when compared to cold gas

cleanup. To improve the efficiency, radiant cooling is more suitable, but the capital costs will increase. For the bioSNG route, the product yield decreases due to a portion being sent off to generate power for the plant demand; this in turn reduces the energy conversion efficiency.

Table 6 Power requirements and generation for three production routes without and with CCS^a

Power (MW)	Fischer-Tropsch		Methanation		Oxymethylene ethers	
		CCS		CCS		CCS
USAGE						
Chopper	0.26	0.26	0.26	0.26	0.26	0.26
Grinder	1.36	1.36	0.32	0.32	1.36	1.36
Lock hopper system	0.10	0.10	—	—	0.10	0.10
AGR pumps	1.02	1.02	0.78	0.78	1.47	1.47
Syngas booster compressor	0.87	0.87	7.72	7.72	—	—
PSA compressor	0.08	0.08	—	—	—	—
Methanation compressor	—	—	0.18	0.18	—	—
BioSNG compressor	—	—	1.01	1.01	—	—
Naphtha pump	3.30×10^{-4}	3.30×10^{-4}	—	—	—	—
Diesel pump	7.10×10^{-4}	7.10×10^{-4}	—	—	—	—
MeOH synthesis compressors	—	—	—	—	4.86	4.86
MeOH cleaning pumps	—	—	—	—	2.20×10^{-3}	2.20×10^{-3}
OMEx pump	—	—	—	—	3.10×10^{-3}	3.10×10^{-3}
Air compressor (OMEx)	—	—	—	—	1.48	1.48
Hydroprocessing	0.21	0.21	—	—	—	—
Air compressor (GT)	2.82	2.82	7.88	7.88	—	—
Air separation unit	5.00	5.00	—	—	5.00	5.00
Oxygen compressor (ASU)	2.38	2.38	—	—	2.38	2.38
Water pumps (steam generation)	0.05	0.05	0.02	0.02	0.31	0.31
CO ₂ compression	0.00	5.00	0.00	3.60	0.00	4.68
Refrigeration	1.82	1.82	0.00	0.00	0.00	0.00
Total	16.2	21.2	18.2	21.8	17.7	22.4
Generation						
Gas turbine	10.7	10.7	17.6	17.6	4.8	4.8
Steam turbine	4.6	4.6	3.7	3.7	0.7	0.7
Net electricity	−1.0	−6.0	3.18	−0.43	−12.3	−16.9

^a *negative net electricity signifies that electricity is imported.

In terms of mass yield, the OMEx route has the highest product yield at 53%. Due to the recycling in OMEx synthesis to optimise the process, the OMEx yield from methanol increases from 38% reported in Zhang *et al.*,⁷⁵ to 83%, as proposed by Ai *et al.*⁶³ This 83% mass yield corresponds to a methanol conversion of 99.9%. Also, the syngas conversion to methanol process is 95%. Therefore, the methanol yield from biomass is compared with previous studies. In the present work, the stoichiometric ratio of $(\text{H}_2 - \text{CO}_2)/(\text{CO} + \text{CO}_2)$ in the syngas is set at 2.1 in this work as advised by E4Tech⁴⁰ and this gives methanol yield of 67% (from biomass). In the models on

OMEx production by Zhang *et al.*⁷⁵ and Oyedun *et al.*⁶⁶ this ratio is set between 0.29 and 0.55 resulting in methanol yields between 29% and 52%. Also, in Phillips *et al.*⁵⁸ a stoichiometric ratio of 1 results in a methanol yield of 49%.

The FT-fuel yield of 22.9% is comparable to values reported in the literature; Tagomori *et al.*²⁶ reports a yield of up to 15% for FT-liquids from forestry residue while Dimitriou *et al.*⁴¹ reports a yield of 20.4% using an entrained flow gasifier and woody biomass to produce FT-fuel. In FTS, the single pass conversion is 40%; by recycling unconverted syngas, this goes up to 72.7%. The initial yield of bioSNG at 25% is consistent

Table 7 Summary of the process simulation results

	FTS	FTS + CCS	BioSNG	BioSNG + CCS	OMEx	OMEx + CCS
Product yield (%)	22.9	22.9	19.8	19.8	52.9	52.9
Energy produced (MW)	111.1	111.1	131.8	131.8	140.4	140.4
Energy conversion (%)	44.9	44.0	59.7	58.2	48.5	47.7
Net heat input (MW)	20.5	20.5	0	0	51.3	51.3
Electricity produced (MW)	15.1	15.1	21.3	21.3	5.5	5.5
Electricity demand (MW)	16.2	21.2	18.2	21.8	17.7	22.4
Net electricity (MW)	−1.0	−6.0	3.18	−0.43	−12.3	−16.9

with the value of bioSNG yield of 24% obtained using a gasifier operating at 1 bar as reported by Vitasari *et al.*⁷⁶

3.4 Capital, operating and maintenance expenditures

The total capital investment breakdown for all cases is presented in Fig. 2. The capital cost is in the range of £140 to £296 million. The only contribution costs of CCS to the capital cost is associated with CO₂ compression as a capture unit is present in all cases. Likewise, the contribution costs of CCS to the operating and maintaining expenditure is associated with transport and storage. The highest capital cost is encountered in OMEx production. In this process, the major contribution to the capital cost is the fuel synthesis which is 41% of the entire cost. This fuel synthesis includes OMEx production from methanol and trioxane. In essence, this includes 4 synthesis units – formaldehyde, trioxane, methylal and OMEx syntheses.

For the FTS, gasification is the most significant contributor to the capital cost accounting for 24% of the total investment. In bioSNG production, syngas cleaning and the power generation section are the biggest contributors to the capital cost, each accounting for 25% of the overall cost. In bioSNG without CCS, excess electricity from burning a fraction of the product to meet plant demand is sold to generate revenue. In OMEx production, the fuel synthesis section accounts for 41% of the overall cost as a result of the additional synthesis steps required in the production process.

The cost of the entrained gasification units used in FTS and OMEx synthesis is noticeably higher than the cost of the dual fluidised gasifier used in methanation. The operating conditions of the gasifiers influence the gasification unit cost. The EFG operates at a significantly higher pressure of 28 bar than the DFB gasifier operating at atmospheric pressure. Also, the EFG operates at 1300 °C while the DFB operates at 870 °C. These are the conditions considered when designing the pressure vessel for the gasifier as these factors determine the thickness of the vessel and amount of materials required to build the gasifiers. In all cases, the cost of additional CO₂ compression is

Fig. 3 Operating and maintenance expenditure of three production routes without and with CCS.

between 1% and 2% of the total investment, thus indicating that the additional investment for CCS is minimal.

The annual operating cost and maintenance costs are presented in Fig. 3. The feedstock cost is the same across all processes as the feed input is fixed at 1020 dt d⁻¹. In terms of the fixed cost which includes labour, insurance, and rent; the methanation process has the lowest annual cost while OMEx has the highest cost.

This is the same trend as the total capital investment as the fixed costs are a fraction of the fixed capital investment. The feedstock cost is the major contributor to the operating costs in all cases except OMEx production where the variable cost dominates. The addition of transport & storage minimally affects the overall OPEX contributing to between 5.3% and 7.3% of this cost. While the variable cost is constant in FTS and methanation with the addition of CCS, the variable cost in OMEx production increases due to an increase in electricity demand in the OMEx plant which is not self-sufficient. The OMEx production clearly is the most expensive to run. This is due to several reasons such as the daily cost of electricity; the use of fired heat compared to the other plants to vaporise water for steam generation; and also, daily wastewater treatment and

Fig. 2 Total capital investment breakdown of three production routes without and with CCS.

cooling water for heat exchange on the plant is significantly higher than the other plants due to more synthesis units required.

3.5 Levelised cost of fuel (LCOF)

The levelised cost of fuel production is presented in Fig. 4. It represents the costs – capital, operating and maintenance – required to produce one unit (GJ) of fuel based on the lower heating value. The cost of production of these biofuels is in the range of £12.2 to £22.9 per GJ without CCS and £13.4 to £24.4 per GJ with CCS.

For all routes, the increase in the production costs of the CCS cases is associated with storing CO₂; this includes the cost of additional compressors to prepare CO₂ for transportation, pipeline costs and storage costs. In FT-fuel, an increase value of approximately 10% is observed (from £20.03 per GJ to £22.01 per GJ), 9.7% in bioSNG production (from £12.21 per GJ to £13.40 per GJ), and 6.8% in OMEx (from £22.86 per GJ to £24.42 per GJ). The bio-SNG production, even without by-product (electricity) credits in the CCS case, it still has the least production costs in both the with and without CCS pathways while the OMEx route has the highest production cost. The production cost is a function of the capital cost, fixed charge factor, by-products and fuel output as presented in eqn (15).

Based on the trend of the CAPEX and OPEX, it would be expected that the highest production cost would be for OMEx while the least would be for bioSNG and this is what is observed. Further, the results are in good agreement with the literature for all routes. Brown *et al.*⁷⁷ have reported bioSNG and FT-fuel production costs that lie in the range of £15.3 to £27.5 per GJ

and £18.3 to £35.3 per GJ, respectively. Comparing the OMEx production cost, Oyedun *et al.*⁶⁶ reported a value of £1.24 per L when processing 500 tonnes per day of woody biomass day. While Schmitz *et al.*³⁵ reported a value £0.41 per L (from \$614.8 tonnes assuming OMEx density of 1097 kg m⁻³ and exchange range values for 2014) when producing OMEx at 1 million tonnes per year. For the present work, the values are £0.47–£0.50 per L when processing 1020 dt d⁻¹ for an output of 0.2 million tonnes per year of OMEx.

3.6 Greenhouse gas (GHG) emissions

Greenhouse gas (GHG) emissions are a source of environmental concern when establishing a production plant. In this subsection, the GHG emissions for each production route are characterised by emission factors. The emission factor estimates how much greenhouse gases in CO₂ equivalent is released per unit of energy output; this also measures the potential for negative emissions. This emission factor includes the biomass harvesting and processing emissions, natural gas for fired heat on the plant, electricity imported to the plant, and CH₄ and N₂O emissions in the resulting biofuels. It basically covers the lifetime emissions of biomass use but excludes emissions from transportation and distribution of the biofuels.

The emission factors used in this study are GHG conversion factors based on the outline from the UK Department of Business, Energy and Industrial Strategy (BEIS).⁷⁸ The BEIS framework categorises the emissions based on activities into three scopes – scope 1, scope 2, and scope 3. Scope 1 covers direct emissions from the plant itself such as fuel combustion which is zero for CO₂ emissions in the case of biomass and biofuels

Fig. 4 Levelised cost of fuel production for three production routes without and with CCS.

Table 8 Emission factors used in greenhouse gas reporting in this study.⁷⁸

Fuel	Scope 1 (kg CO ₂ e kW h ⁻¹)	Scope 2 (kg CO ₂ e kW h ⁻¹)	Scope 3 (kg CO ₂ e kW h ⁻¹)	Overall emission factor (kg CO ₂ e kW h ⁻¹)
Wood pellets	0.01563	—	0.03744	0.05307
Natural gas	0.20428	—	0.02657	0.23085
Electricity	—	0.2556	—	0.2556

due to its status as CO₂ neutral. However, N₂O and CH₄ released are not absorbed in biomass regeneration so scope 1 covers these gases. Scope 2 covers indirect emissions from the plant such as imported electricity and heat purchased to cover internal needs. Scope 3 covers indirect emissions from sources not owned or controlled by the plant such as the emissions due to harvesting, refining, and transporting biomass. The values used in this study are presented in Table 8. For the wood pellets and the natural gas, the summation of scope 1 and scope 3 were used.

The calculated emission factors for the three routes investigated with and without CCS are presented in Fig. 5. Without CCS, all the production routes are net positive for GHG emissions. With CCS, there is a significant decrease of over 250% in the emission factors. While adding CCS is minimal to production costs (economics) – as seen in previous section – the effect on the environment is significant in terms of huge emission savings.

Adding CCS to FT-fuel production results in negative emissions equivalent to a GHG mitigation potential of 519 000 tCO₂ per year; for bioSNG production, there exists a mitigation potential equivalent to 301 000 tCO₂ per year; and for the OMEx route, there is a mitigation potential equal to 303 000 tCO₂ per year. The OMEx route without CCS has the highest emission potential at 201 000 tCO₂ per year and this is mainly due to the indirect emissions from the purchased electricity. The OMEx route purchases the most electricity from the grid. This also limits the emission saving potential with CCS applied. The

bioSNG route has the least emission savings potential with CCS. This is mainly due to the amount of captured and stored carbon; looking at the carbon balance in Fig. 1, the most amount of carbon is vented within this process due to the required gasification technology and power generation. While the OMEx route imports much electricity which contributes to emissions, a higher energy output and CO₂ capture helps in reducing the emission factor. The emission factor is also a function of energy output, so to decrease emissions and improve environmental performance, improving the plant efficiency, which results in a higher output is one way. Other ways include decreasing the amount of CO₂ vented to the atmosphere (especially with the bioSNG route) and limiting electricity imports from the grid by coupling to renewable energy power generation.

3.7 CO₂ avoidance cost

The CO₂ avoidance cost (in the CCS cases) is presented in Table 9. In this study, this value is calculated based on two reference plants. Plant A refers to a conventional fossil fuel plant (natural gas for bioSNG, and diesel for FT-fuel and OMEx) while plant B refers to the biomass-based plant without CCS. These values represent the minimal carbon tax to be paid on CO₂ emissions. With the conventional plant, the values are much higher due to the lower cost of fuel production using fossil fuels. A maximum avoidance cost of £119 per tonne CO₂ is reported in this study and it is comparable to the 2011 values reported for the UK. Charles *et al.*⁷⁹ reported an abatement cost of biofuels of £115

Fig. 5 Emission of production routes without and with CCS.

Table 9 CO₂ avoidance cost in £ per tonne CO₂ with reference to a conventional plant and a biorefinery

	Plant A	Plant B
FTS	75	21.7
BioSNG	91	22.4
OMEx	119	25.7

per tonne CO₂ for bioethanol and £154 per tonne CO₂ for used cooking oil diesel.

Fuel switching to biomass resources results in a reduced avoidance cost as seen in the case of plant B. Across the three routes, production of OMEx generally has the highest cost due to its relatively low emission factor as more emissions to the atmosphere needs to be paid for. In the bioSNG route, although a higher percentage of carbon is vented in the process, minimal electricity is purchased and there is no heat or steam purchase to indirectly increasing the CO₂ emissions from the process. The FT-fuel route has the lowest avoidance cost even with electricity and heat imports; however, the amount of emissions from these imports are offset by a higher amount of CO₂ capture in the process. Note that electricity and heat imports are less than half of what is required in the OMEx route. This is because the avoidance cost is a function of the emission factor and the more negative emissions achieved by a plant, the lower is the cost. Alternatively, increasing the price of conventional fossil-derived fuels will lower the avoidance cost. Finally, including BECCS in carbon trading systems will further result in reduced avoidance costs.

3.8 Comparison with fossil-derived fuels – diesel and natural gas

In this section, the minimum selling price (MSP) of each biofuel is compared to the market price of conventional fossil fuels. Since OMEx is a diesel additive, its production route will also be compared to the diesel production cost. Using a DCFROR and NPV break-even analysis, the MSP was determined for all routes and presented in Table 10.

The average 2018 UK wholesale price of natural gas was £5.5 per GJ (ref. 80) while that of diesel excluding UK fuel duty was £13.23 per GJ (ref. 81) (for a diesel energy density of 32 MJ L⁻¹). The MSP for bioSNG with and without CCS is at least 2.5 times the cost of producing natural gas while the MSP of FT-fuels and OMEx is up to two times the price of conventional diesel price. This means that the three routes investigated cannot compete

Table 10 Minimum biofuel selling price of three production routes without and with CCS

	Without CCS (£ per GJ)	With CCS (£ per GJ)
FTS	21.8	23.4
BioSNG	13.4	14.5
OMEx	24.6	26.5

with natural gas and diesel without process improvements to decrease costs and incentives such as policy schemes.

To encourage the production of these biofuels to compete with and phase out the dependence on fossil-derived fuels, policy instruments such as subsidies and carbon taxes are required. The next section covers the existing policy schemes within the UK that support the production of biofuels and suggest a possible scheme to assist with decarbonisation.

3.9 Financial tools analysis

The UK government introduced the Renewable Transport Fuel Obligation (RTFO) in 2008 with the aim of reducing GHG emissions from fuel used for transport purposes. It lays out an obligation for fuel suppliers to meet a target of renewable fuel in each obligation period. Renewable Transport Fuel Certificates (RTFCs) are issued for each litre or kg of biofuel produced. For certain biofuel sources such as waste, double RTFCs are issued. As such, these RTFCs can be traded and the price is dependent on demand and supply. In the past, the trade prices have varied between £0.09 and £0.20 per certificate.^{82,83} For this analysis, an RTFC price of £0.145 per RTFC will be used. Based on the RTFO guidelines, for each litre of biofuel produced, 1 RTFC has been issued.

The Renewable Heat Incentive (RHI) established in 2009, provides financial support for renewable heat technologies. A tariff is paid to the renewable heat generator for each unit of energy generated. Currently, for biomethane produced and injected to the grid, the current tariff is an average of £22.16 per MW h;⁸⁴ this is the value assumed for the plant's lifetime. BioSNG is also eligible for RTFCs at 1.9 RTFCs for each kilogram of biomethane, using feedstocks that are wastes or residues or dedicated energy crops doubles the RTFCs to 3.8 RTFCs kg⁻¹. In this case, biomass feedstock is assumed to be woody biomass and thus qualifies for the 1.9 RTFCs kg⁻¹ and with preliminary calculations, the RHI provides more savings. The RHI is applied to bioSNG while the RTFO is applied to FT-fuels and OMEx.

Fig. 6 depicts the effect of the RTFCs and the RHI on biofuel production. While the application of the RTFCs is beneficial to the production routes without and with CCS, it is still not enough to comfortably compete with the conventional diesel, only the LCOF of FTS without CCS is close enough; which entails that tax exemptions may also be required for the feasibility of the biorefineries.

The implementation of the RTFO results in a decrease of the MSP of up to 28% for FT-fuels and up to 24% decrease for OMEx. While the implementation of the RHI to bioSNG production decreases the minimum selling price by 41% without CCS and 36% with CCS, it is still not profitable and competitive with natural gas production. Further, decreasing the MSP of all production routes will require switching to waste as feedstock so that the bioSNG process can qualify for 3.8 RTFCs kg⁻¹ under the RTFO and the FT-fuels and OMEx can qualify for 2 RTFCs/L; also, this switch could result in a decrease in the feedstock price but at the same time the technical feasibility of waste to fuels is more challenging and less mature compared to virgin wood.

Fig. 6 Effect of RTFO and RHI on the production cost and minimum fuel selling price of three production routes without and with CCS.

Carbon price is an effective tool in reducing GHG emissions when applied effectively. In the current EU emissions trading scheme applied by the UK, there is no credit for negative emissions. Applying a sufficient carbon price to close the carbon pricing gap will assist renewable technologies to fairly compete with fossil-fuel technologies and eventually decrease the dependence on fossil fuels by lowering the levelised cost of fuel of biofuels whilst increasing that of fossil-based fuels. Introducing a carbon price for negative emissions could boost the feasibility of producing biofuels because while revenue will be generated through this means, the price of fossil-derived fuels will increase as a result of paying the environmental price for positive emissions. The emission factors used for fossil derived diesel ($0.26880 \text{ kg CO}_2\text{e kW}^{-1}$) and natural gas ($0.20428 \text{ kg CO}_2\text{e kW}^{-1}$) in this section were the scope 1 factors reported by the BEIS.⁷⁸

Fig. 7 illustrates the dependency of the minimum selling price for the CCS cases on the carbon price (RTFO and RHI payments are not included). In this study, a break-even carbon price of £48 per tCO_2 is required for FT-fuels to compete with fossil-derived diesel and a price of £86 per tCO_2 is required for OMEx to compete. Higher carbon prices will discourage the production of diesel and favour the production of FT-fuels with CCS and OMEx with CCS as well as drive the development and deployment of this technology. With bioSNG, even with the increase in natural gas prices where a carbon price is introduced, a carbon price up to £63 per tCO_2 is required to break even. The major barrier to the deployment of the bioSNG route is the very low price of natural gas. These break-even carbon prices are dependent on the emission factor of the production route. To further decrease the minimum carbon price required, the routes, especially the

Fig. 7 Effect of negative emission credit on minimum selling price of biofuels. The fossil-derived fuel prices are presented for comparisons.

OMEx route will need to generate more negative emissions; doubling the emission factor halves the break-even carbon price.

Natural gas production currently enjoys a lot of benefits as it produces 50% less CO₂ emissions than coal⁸⁵ and is currently being used as a transition fuel. However, now is the time to prepare and cut the costs associated with bioSNG production to boost its feasibility as economies move towards decarbonisation.

3.10 Sensitivity analysis

The production cost of biofuel is dependent on different parameters. In this section, a sensitivity analysis is performed to pinpoint the parameters that have a significant effect on the production cost. This analysis highlights the parameters to be focused on making each route feasible and competitive. Generally, based on previous studies^{86,87} the parameters (CAPEX, plant life, biomass cost, CCS) are changed by $\pm 30\%$ of the base case scenario with the exceptions of the fuel output at $\pm 10\%$, the interest rate at $\pm 20\%$ and the operating hours where the capacity factor is between 70% and 90% as not to exceed the maximum hours in a year. Also, this analysis is only carried out for the CCS cases as the focus is on BECCS technologies and it is only the CCS cases that provide negative emissions. The results of the analysis are displayed in Fig. 8.

The bars in the Fig. 8 represent the sensitivity of each parameter to the production cost. Longer bars depict higher sensitivity while shorter bars depict lower sensitivity. From this, the most significant factor that will affect the production cost is the operating hours. The plant operating hours in a year is dependent on the capacity factor. Increasing the capacity factor to 90% while other factors are kept constant, decreases the production cost by 6% while decreasing the capacity factor to 70% results in a 21% increase in the production cost. Apart from maintenance, the capacity factor is affected by the feedstock availability. Where there is a shortage of supply, the capacity factor is reduced, thus resulting in decreased output. It is important that the supply of the biomass is sustainable and secure for each production route; this should also be applied for the biomass price to avoid fluctuations in price (at 30% uncertainty of feedstock price, fluctuations in production cost are -6% to $+10\%$). Alternatively, considering a mix of biomass sources could be beneficial.

The next factors that significantly affect the production cost are the capital investment and the fuel output. Fluctuations of -15% to $+15\%$ in production cost are seen for an uncertainty of 30% in the capital investment while fluctuations of -9% to $+11\%$ are seen for an uncertainty of 10% in the fuel output. The possible decrease in production cost highlights the importance of improving the plant efficiency to increase the fuel output. As the data for the capital investment is acquired from factorial estimation and open literature, it is difficult to accurately predict the capital expenditure. Also, the capital investment is subject to location; importation of equipment

Fig. 8 Sensitivity analysis on the production cost for three production routes with CCS (a) Fischer–Tropsch synthesis, (b) methanation and (c) oxymethylene ether synthesis.

attracts import duties and could result in increased CAPEX while producing within the country will not have such duties. Accurate data will be available where there are existing commercial plants. Until then, the initial estimates provided in this chapter are suitable for preliminary feasibility studies. Other parameters – CO₂ compression, transport & storage costs, and plant life – have minimal effect on the production cost.

4 Conclusion

BECCS is an integral concept to stabilizing and eventually reducing CO₂ levels in the atmosphere. Fuel generation already has an advantage due to the intrinsic capture unit and pure stream of CO₂ available providing an opportunity for easy CCS retrofitting. This work investigated the technical, economic, and environmental performance of three biofuel

production routes that have the potential to be BECCS technologies. The fuel synthesis routes – Fischer–Tropsch synthesis, methanation and oxymethylene ethers synthesis – were modelled using Aspen Plus. A sensitivity analysis was performed to highlight the parameters for process improvements.

The energy conversion was in the range of 44.9% to 59.7% without CCS and 44.0% to 58.2% with CCS. BioSNG production had the highest efficiency due to high CO conversion to CH₄ as well as optimised heat integration with no external heat input. FT synthesis had the least efficiency due to the relatively low CO conversion, *i.e.* 72.7%. In all three cases, the energy conversion decreases by less than 1.5 percentage points when CCS is added to the plant due to intrinsic nature of CO₂ capture in biofuel generation.

Regarding the effect on the environment, adding CCS results in substantial negative emissions for the three routes considered. Overall, 301 000 to 519 000 tCO₂ can be removed from the atmosphere per year using the investigated routes. The OMEx route has the least emission factor at $-0.2892 \text{ kg CO}_2 \text{ kW h}^{-1}$ due to purchased heat, steam, and electricity to meet the plant demand. The bioSNG route has a lower emission factor than the FT-fuel route due to the amount of CO₂ that is vented to the atmosphere during the process especially in the gasification section that uses dual fluidised bed gasifier to maximise the product yield. However, more negative emissions can be achieved by capturing CO₂ in the flue gas and limiting electricity imports which increase indirect emissions, thus maximising carbon removal.

The production costs are in the range of £12.2 to £22.9 per GJ without CCS and £13.4 to £24.4 per GJ with CCS. BioSNG is relatively the cheapest fuel to produce while OMEx is relatively the most expensive to produce as a result of the high capital

investment and the high operating expenditure. The addition of CCS does not have a drastic effect on the production costs while providing huge environmental benefits. However, in comparison to fossil-derived fuels on the market, the biofuel counterparts cannot feasibly compete without financial incentives due to minimum selling prices of the biofuels being at least two times more than fossil-derived fuels. With the application of the current RTFO and the RHI prices, competition is still not feasible with the current feedstock – woody biomass. Applying carbon pricing as an economic tool, a price in the range of £48 to £86 per tCO₂ is required to break-even with the current market price of the fossil-derived fuels. These figures are a function of the emission factor hence more negative emission credits need to be generated by the plants to achieve a lower carbon price. Combining the current financial incentives (RTFO and RHI) with carbon pricing could result in a much lower breakeven price. The sensitivity analysis highlights the capacity factor as the critical process parameter affecting the production cost. Other important parameters include the fuel output, the capital expenditure and feedstock costs.

Overall, the three routes with CCS are promising BECCS technologies but cannot currently compete with fossil-fuels without carbon pricing. Process improvements to boost the product yield and capture more CO₂ are required. Also, more aggressive policies and incentives focused on decarbonisation are needed to drive development and deployment and eventually favour competition to phase out fossil-derived fuels.

Appendix

Table 11 Major process equipment references and scaling factors

Process unit	Design variable	Base capacity	Base cost (£ million)	Base year	Scaling factor	References
Pretreatment	Biomass input	2200 dt d ⁻¹	11.34	2007	0.77	8 and 26
Gasification (EFG)	Biomass input	2200 dt d ⁻¹	33.87	2007	0.66	8 and 26
Gasification (DFB)	LHV (MW _{SNG})	20 MW	2.61	2014	0.66	69
Syngas cleaning	Syngas output	3823 t d ⁻¹	16.74	2007	0.7	8 and 26
Primary cleaning	SNG output	20 MW	2.11	2014	0.67	69
WGS & cracking	SNG output	20 MW	2.09	2014	0.7	69
FT synthesis	Biomass input	2200 dt d ⁻¹	24.68	2007	0.7	8 and 26
Methanation	SNG output	20 MW	2.18	2014	0.7	69
Methanol synthesis	Methanol output	44.3 t d ⁻¹	2.18	2011	0.8	88
OMEx synthesis	OMEx output	1 million t y ⁻¹	166.29	2014	0.65	35
Pressure swing adsorption	PSA input	797.6 t d ⁻¹	8.37	2015	0.7	26 and 89
Hydrocracking	Flowrate	378 t d ⁻¹	16.49	2007	0.67	8 and 26
Upgrading unit	SNG output	20 MW	0.44	2014	0.7	69
Power generation	Power	35.9 MW	29.83	2015	0.75	8
Air separation	Air input	2903 t d ⁻¹	12.14	2007	0.8	8 and 26
Centrifugal compression	Power	500 kW	0.91	2010	0.6	68
Reciprocal compression	Power	500 kW	0.36	2010	0.75	68
Cooling system	Flowrate	10 000 L s ⁻¹	3.45	2010	0.9	68

Table 12 Estimated labour costs for operating costs

Position	Salary	Number	Total
Plant manager	£60 000	1	£60 000
Plant engineer	£40 000	2	£80 000
Maintenance supervisor	£30 000	1	£30 000
Lab manager	£38 000	1	£38 000
Shift supervisor	£26 000	5	£130 000
Lab technician	£22 000	4	£88 000
Maintenance technician	£29 000	8/12	£232 000/£348 000
Shift operators	£29 000	20/30	£580 000/£870 000
Yard employees	£20 000	12	£240 000
Clerks & secretaries	£20 000	3	£60 000
Total			£1 538 000/£1 944 000

Table 13 Operating and maintenance costs data^{4,5,19–22}

Parameter	Value
Fixed costs	
Insurance, taxes & rent	2.5% fixed capital investment
Maintenance and repairs	2% fixed capital investment
Operating supplies	15% maintenance & repairs
Laboratory charges	10% operating labour
Overhead costs	50% operating labour
Variable costs	
Feedstock	£50 per dry tonne
Ash disposal	£19 per tonne
Wastewater treatment	£4.52 per m ³
MEA	£1.92 per tCO ₂
WGS catalyst	£16 per kg
FTS catalyst	£30 per kg
Methanation catalyst	£0.02 per GJ of SNG
Methanol catalyst	£18.8 per kg
OMEx catalyst	£33.2 per kg
Formaldehyde catalyst	£437 per kg
Activated carbon	£10.4 per kW h
Boiler feed water	£0.50 per tonne
Cooling water	£0.023 per tonne
Fired heat	£8.06 per MW h
Electricity to grid	£58 per MW h
Electricity from grid	£104 per MW h
LO-CAT chemicals	£160 per tonne S
Hydroprocessing	£3.63 per barrel produced
PSA	£3.31 per kg
CO ₂ transportation and storage	£19 per tCO ₂

Conflicts of interest

There are no conflicts to declare.

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