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Research Article

Technoeconomic and Environmental Assessment of Biomass Chemical Looping Gasification for Advanced Biofuel Production

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Chemical looping gasification is a promising biomass conversion technology that could produce sustainable liquid transportation fuels on a large scale to reduce fossil fuel dependency. The current paper examines the technical, economic, and environmental performance of a biomass-to-liquid (BtL) process based on chemical looping gasification and Fischer-Tropsch synthesis. Two biomass feedstocks, i.e., pine forest residues and wheat straw, are selected for assessing the complete BtL production chain. The results of process simulations showed that both biomass types are suitable gasification feedstocks, with an overall energy efficiency of 53% and 52% for pine residues and wheat straw, respectively. The economic results show that the breakeven selling prices (BESP) are €816 and €781 per m³ for the pine forest residues and wheat straw pellets, respectively. However, if low-grade excess heat valorisation and CO₂ credits are considered, the BESPs could meet or become lower than the target value of €700 per m³, making the BtL plant competitive with other biofuel plants. The CO₂ avoidance cost is estimated at €74.4/tCO₂ for pine residues and €61.3/tCO₂ for wheat straw, when replacing fossil fuels. The results of the life cycle assessment study showed that the produced biofuels fulfil the requirements of the EU Renewable Energy Directive II, achieving the reduction in greenhouse gases emissions of up to 79% without carbon capture and storage (CCS) and up to 264% with CCS compared to fossil fuels.

1. Introduction

The emission levels of CO₂ in the atmosphere are currently unprecedentedly high (420 ppm CO₂ on average, in 2022). The latest IPCC report stresses the need to greatly and rapidly reduce fossil fuel consumption to limit the extent and impact of global warming [1]. Globally, the CO₂ emission from the transport sector account for almost 20% of the total CO₂ emissions, with 22.2% of them coming from sectors like aviation and maritime transport, where electrification is not a viable decarbonisation solution. The EU target is to achieve a 10% share of biomass-derived biofuels in the transportation sector by 2020 has not been reached yet [2]. Therefore, the research into new approaches to produce sustainable

liquid fuels from renewable biomass to displace fossil feedstocks is timely and important.

Gasification is one of the most promising biomass conversion technologies that can be employed on a large scale [3]. The syngas resulting from biomass conversion (rich in carbon monoxide and hydrogen) can be further processed into liquid fuels and value-added chemicals, such as methane, methanol, ethanol, and diesel-like fuels or, alternatively, used for heat and/or power generation [4]. The synthetic fuels obtained via Fischer-Tropsch (FT) synthesis can be blended with fossil fuels or used directly in existing transportation fleets, without the need for any engine modifications [3]. The biomass-to-liquids (BtL) route is one of the few renewable options for difficult-to-decarbonise sectors such as aviation [5] and maritime transport [6].

Several studies have investigated the technical, environmental, and economic feasibility of BtL processes via gasification and FT synthesis, showing that the technology is viable [7], and the resulting biofuels are environmentally superior to fossil fuels [8]. One of the economic aspects that hinders a higher acceptance of gasification for liquid fuel production is the high cost of the gasification agent: oxygen. Oxygen gasification is generally regarded as a more advantageous solution, compared to air or steam, as it produces a higher calorific value syngas [9]. However, oxygen gasification requires an expensive air separation unit (ASU) and high electricity consumption. An ASU can account for between 10 and 15% of the fixed capital investment [10] and even exceed the cost of the gasification reactor [11]. Similarly, the electricity consumption of ASU can represent up to 20% of the total plant electricity requirement, as shown in Banaskiewicz and Chorowski [12].

Chemical looping gasification (CLG) of biomass involves using two connected reactors: a fuel reactor, where the oxygen carrier is reduced during the gasification reactions, and an air reactor, where the reduced oxygen carrier material is regenerated via air oxidation. In CLG, the selected solid oxygen carrier circulates between two reactors and provides the oxygen for feedstock gasification to yield concentrated syngas. This avoids the need for an ASU and its associated energy and capital penalties. Besides from the above-mentioned benefit, the air reactor gives nearly zero CO₂ emissions. This is because only low carbon contents enter the air reactor. However, the CLG concept requires a second reactor and the use of an oxygen carrier, which incur both capital and operational costs. Reportedly, the syngas generated through CLG has a lower tar [13] and a higher hydrogen content and yield [14] compared to conventional fluidised bed gasification [15], due to the oxygen carriers' catalytic activity. The concept of the chemical looping gasification has been successfully tested at laboratory scale for numerous biomass feedstocks (wheat straw [16], wood [17] and wood residues [18], rice husks [19], sewage sludge [20], etc.). Another chemical looping pathway that has been proposed is the chemical looping hydrogen technology, where an additional steam reactor is employed for hydrogen production in a separate stream [21]. The hydrogen obtained can be directly employed in ammonia [22] or methanol [21] synthesis, showing a promising reductions in energy consumption and CO₂ emissions compared to the base technology. Furthermore, ammonia production via chemical looping has also been proposed [23], where the nitrogen is supplied via an ASU.

The work presented in the paper is part of the results of the Horizon 2020 CLARA project. The novelty of the wider project is the demonstration of the full-process chain using a 1 MWth chemical looping gasification unit [24] combined with a FT unit and the scaling up of a 200 MWth industrial plant using models validated at pilot scale. Biomass CLG has not yet been applied on a large scale, partly due to concerns regarding continuous operation, caused by the oxygen carrier interaction with biomass ash and volatile compounds [25]. In addition to solving these operational challenges, the economic and environmental impact of CLG must be

understood to facilitate its transition to an industrial scale. Only a few studies examined the techno-economic aspects of biomass CLG and showed that the technology is outperforming several existing processes for power generation [26, 27] and methanol production [28]. For power generation, Mohamed et al. [27] performed a techno-economic analysis for a potential plant, with a capacity of 650 MW using wood pellets as biomass feedstock and hematite as oxygen carrier. The estimated cost of electricity for the 650 MW facilities is reportedly 21.7 ¢/kWh, when a CCS solution is implemented, and 18.4 ¢/kWh in its absence. Similarly, Sarafraz and Chisto [26] performed a techno-economic analysis for the CLG of waste carbon black using a novel liquid indium-based chemical looping system for the production of electricity. They report a levelized cost of energy (LCOE) between \$11.46/GJ and \$29.3/GJ, according to biomass prices ranging from \$-10/GJ to a maximum value of \$6/GJ. For methanol production, Sun and Aziz [28] performed a comparative techno-economic assessment of CLG for methanol production, proposing two chemical looping configurations: one using a traditional gasification approach (for syngas production) and one for targeted hydrogen production using the chemical looping approach. They found the latter one to be more economical and able to produce electricity in addition to methanol.

For FT syncrude production, only Kumar et al. [29] and Saeed et al. [30] have so far investigated the techno-economic feasibility of using the CLG instead of conventional gasification, using lignocellulosic residues, at a scale of 100 and 80 MW (thermal input), respectively. Neither of the papers mentioned above considered the environmental impact of the fuel synthesis plant, which is an important feature of the novelty of this paper. While several studies indicate that biomass-derived FT fuels can be profitable when the market price of fossil fuels is high [10] or when suitable environmental credits/taxation are applied [31], they do not offer any quantitative insight into the environmental performance of the BtL plants.

The novelty of the present paper lies in the comprehensive techno-economic and environmental life cycle assessment (LCA) of a 200 MWth BtL plant to produce F-T products, whose performance has been validated against experimental data of a 1 MWth CLG unit. Another novel aspect of the work is establishing the cost of CO₂ emissions avoidance, by replacing transportation fuels with carbon-neutral or carbon-negative fuels obtained via the proposed BtL route. The CO₂ avoidance cost is a useful tool for policymakers to establish adequate taxation mechanisms or for investors to compare different biofuel production technologies. In this manner, the paper defines a benchmark for the performance of future chemical gasification systems. By integrating the techno-economic and environmental analysis and computing the cost of avoided CO₂ from the use of fossil transportation fuels, the paper provides novel insights on the sustainability of large-scale BtL processes based on CLG and FT synthesis.

2. Material and Methods

The BtL process chain assumes that biomass pretreatment is performed in decentralised plants and transported to the

CLG process; furthermore, the FT syncrude hydrocracking will be performed in existing oil refineries. This approach reduces both biomass transportation and FT syncrude upgrading costs. The fixed capital investment and annual operating costs are estimated for a potential commercial plant configuration of the proposed BtL scheme. Based on this, the breakeven selling point (BESP) is determined for the liquid fuels produced using the discounted cash flow rate of return methodology. An economic sensitivity analysis is performed to investigate how uncertainties in capital and operating cost estimations can influence FT fuel prices. An environmental LCA study evaluates the environmental impacts of biofuel production from biowastes.

The capacity considered for the gasification unit is 200 MW thermal input, based on the biomass lower heating value. This ensures the fuel synthesis plant takes advantage of the benefits of scale [32] while not being limited by biomass availability.

2.1. Biomass Selection and Pretreatment. The biomass required for the gasification plant is collected from various sites around the EU and transported to smaller decentralised facilities for processing, and this minimises transport requirements. It is assumed that the fuel synthesis plant is only fuelled by a single-biomass residue. The two biomass sources considered in the analysis are pine forest residues (from logging operations) and wheat straw, which are widely available in Europe.

The feedstock pretreatment considered includes drying, grinding and palletisation, and additive addition. Drying is required as the initial pine wood moisture content is around 40%, while the required moisture content for CLG is below 10%. This is because the CL gasifier is designed to operate with low moisture biomass content, as high moisture content can lead to higher tar formation. In addition, for high-moisture biomass feedstock, a significant amount of energy is required to evaporate the water present in the biomass. This energy consumption reduces the overall efficiency of the gasification process. The aim of grinding and palletisation is to improve biomass bulk density and thus feedstock handling and lowering transport requirements. Wheat straw has a significantly higher ash content than pine forest residue; thus the wheat straw requires an additive to increase its melting temperature. The additive selected in this work was calcium carbonate, as it displayed the best ash-related performance in experimental studies [33]. The biomass price at the gate of the BtL plant is estimated at €110.2/tonne for pine forest residue and €76.9/tonne for wheat straw [34].

The ultimate and proximate analyses of the biomass feedstocks are presented in Table 1.

2.2. Process Chain Description. The entire process chain considered in this study is schematically depicted in Figure 1. The biomass, received in pellet form at the BtL plant gate, is stored in a short-term storage facility, is transported using a conveyor belt to the gasifier unit, and fed to the fuel reactor, where the gasification reactions occur.

2.2.1. Chemical Looping Gasification. The chemical looping gasification concept is realised using two interconnected

TABLE 1: Biomass feedstock ultimate and proximate analysis [33, 34].

	Units*	Pine forest residue	Wheat straw
Proximate analysis			
Moisture content	% (wb)	9.5	10.0
Volatiles	% (db)	73.2	77.7
Fixed carbon	% (db)	24.8	13.0
Ash	% (db)	2.0	9.3
Ultimate analysis			
Carbon	% (daf)	52.7	49.6
Hydrogen	% (daf)	6.4	6.2
Nitrogen	% (daf)	0.39	0.42
Oxygen	% (daf)	40.5	43.6
Sulphur	% (daf)	0.05	0.11
Chlorine	% (daf)	0.01	0.05
Lower heating value	MJ/kg (daf)	20.20	18.5

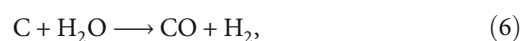
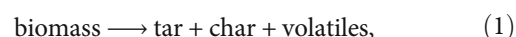
*wb =wet basis; db = dry basis; daf = dry ash free basis.

reactors (the fuel and air reactors), with the solid oxygen carrier circulating between them. The fuel reactor is fluidised with steam and CO₂ (captured downstream of the gasification unit), while oxygen is supplied by the oxygen carrier. The syngas and the spent oxygen carrier exit the fuel reactor and pass through a cyclone, separating the solid carrier which is sent to the air reactor. In the air reactor, the oxygen carrier reacts with preheated air to regenerate its oxygen content. The regenerated carrier exits the air reactor together with the oxygen-depleted air, passing through a cyclone before being returned to the fuel reactor. The heat of the syngas and oxygen-depleted air is recovered and used to preheat the combustion air, and the steam required for the fuel reactor.

The oxygen carrier chosen in this study is ilmenite (an iron-titanium mineral, in its most reduced form FeTiO₃) due to its high tensile strength, good oxygen transfer capacity, high-melting point, and minimal environmental concerns regarding its exploitation, use, and disposal [35]. Previous chemical looping experiments performed using ilmenite have shown good gasification behaviour and stable operation [36, 37].

The overall chemical reactions considered in the air and fuel reactor are presented in equations (1)–(9) [38].

Fuel reactor is as follows:



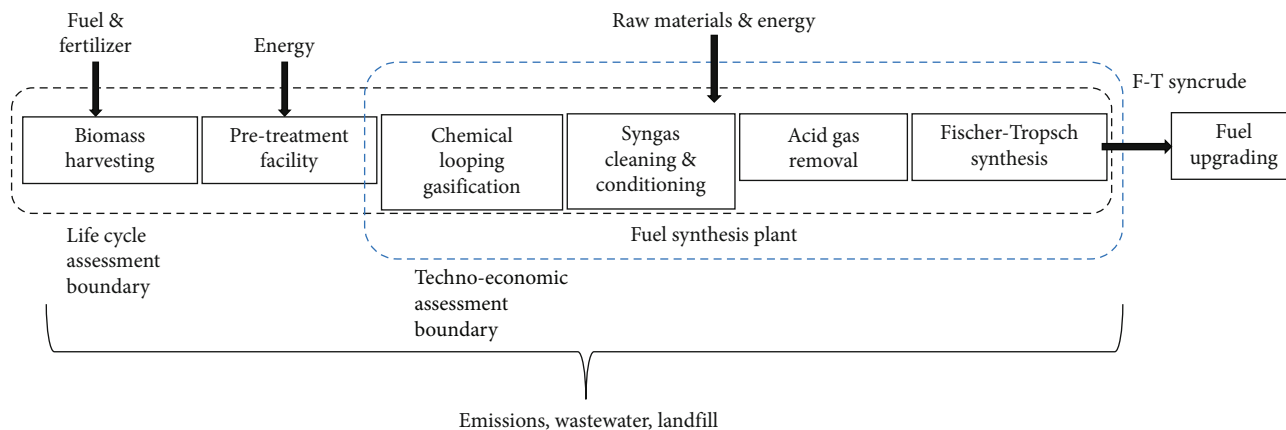
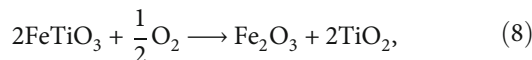


FIGURE 1: Biomass-to-liquid process chain diagram and boundaries considered in the analysis.



Air reactor is as follows:



For the kinetic parameters of ilmenite, we have used the values proposed in previous works from the CLARA consortium partners, using chemical looping setups with a capacity of 100 kWth [39] and 1 MWth [40].

2.2.2. Syngas Cleaning and Conditioning. The syngas cleaning unit consists of a ceramic dust filter, a water scrubber, a three-stage compression train, a water-gas shift reactor (WGSR), a COS hydrolysis reactor, and a tar scrubber. Firstly, the dust filter removes the larger particulates (i.e., ash, spent oxygen carrier), operating at atmospheric pressure and a temperature of 300°C. The gas is then passed through the water scrubber, which partially removes the inorganic gas impurities and condensable tar compounds. The scrubber further cools down the gas to approximately 70°C. The syngas is then compressed in a compression train, with interchange cooling, up to a final pressure of 35 bar. Part of the syngas is sent to a gas shift reactor (WGSR), operating at 350°C and 35 bar, to adjust the CO:H₂ ratio for the FT synthesis process. The entire gas stream is treated in a COS hydrolysis reactor before it finally passes on through a bio-oil scrubbing tower to remove tar to values below 1% (volume).

2.2.3. Acid Gas Removal. Following the final tar removal cleaning step, CO₂ and H₂S are separated from the gas in a methyl di-ethanol amine (MDEA) scrubber. The MDEA process is chosen instead of the more established Rectisol process, as MDEA solvents have been shown to remove both CO₂ and H₂S with great efficiency, without the stringent refrigeration requirements of the Rectisol process [41]. The MDEA is regenerated in a desorption tower, while the CO₂-rich stream is sent to a sulphur recovery unit. To achieve a ppb-level concentration of sulphur compounds,

the CO₂-free syngas is further washed with a sodium hydroxide solution in a caustic washing tower. The CO₂ stream is partly reused as a gasification agent in the fuel reactor.

2.2.4. Fischer-Tropsch Synthesis. In the fuel synthesis process, the conditioned syngas (containing CO, H₂, and CH₄) is converted into liquid fuels, in the presence of a Co-based catalyst, at a temperature of 280°C. The chain of polymerisation reactions produces a wide mix of hydrocarbons, and process conditions are optimised to produce the maximum amount of usable FT syncrude. After the FT reactor, a two-stage separator is employed to separate the gas (methane, ethane, and propane) and liquid fractions (water, light, and heavy hydrocarbons). The gas fraction is cooled and partly recirculated to the FT reactor, while the remaining gas stream is sent to a steam methane reforming (SMR) unit, which produces a mix of CO and H₂, recirculated to the FT reactor. The liquid fraction is treated to remove excess water, and the resulting stream is further distilled to obtain naphtha, diesel, and wax fractions.

2.3. Process Simulation. The entire process was simulated in Aspen Plus™, using the “IDEAL” property package for the chemical looping gasification unit, the “ELEC-NRTL” package for the syngas cleaning and acid gas removal units, and the “NRTL” package for the FT synthesis unit. The modelling approach for each main process unit is detailed in Table 2.

2.4. Cost Estimation Methodology. The economic analysis performed in this paper estimates the fixed capital costs (costs related to plant construction—equipment purchase cost, erection cost, building, and site development cost), working capital costs, project investment, and annual operating costs.

2.4.1. Capital Cost Estimation. Based on the results of the ASPEN Plus™ simulations, the performance of the 200 MW BtL plant is replicated using the ECLIPSE process simulator, which enables the economic assessment (capital cost and BESS calculations) [42]. The process flow diagrams for all the process units are regenerated within ECLIPSE.

TABLE 2: Modelling approach and assumptions for the main process equipment.

Equipment	Process conditions & assumptions	Modelling approach	Comments
Fuel reactor	935°C, 1.01 bar Steam to biomass ratio: 0.3 Heat loss <1%	RSTOIC reactor with cyclone, embedded calculator containing CFB hydrodynamics as well as reaction kinetics	Operating temperature is chosen to ensure high carbon conversion.
Air reactor	1010°C, 1.01 bar Air to fuel ratio: 0.3 Heat loss <1%	RSTOIC reactor with cyclone, embedded calculator containing CFB hydrodynamics as well as reaction kinetics	Air-to-fuel ratio is optimised to ensure autothermal operation; temperature is fixed to ensure sufficient heat transfer to the fuel reactor.
Hot gas filter	300°C, 0.97 bar	Cyclones and solid ceramic filter	—
Water scrubber	70°C, 1 bar 100% HCl removal	RADFRAC column module	Water recirculation with purge stream, partial NH ₃ and heavier tar compounds removal.
Compression train	A train of 3 compressors, to gradually increase the syngas pressure to 35 bar, with interstage cooling	Compressors	—
WGSR	350°C, 35 bar	REQUIL reactor module	Syngas flowrate redirected to WGSR is optimised to obtain the required H ₂ :CO ratio for FT synthesis.
COS hydrolysis reactor	210°C, 35 bar	REQUIL reactor module	Conversion of COS to H ₂ S.
Bio-oil scrubber	30°C, 34 bar Tar final concentration <1% (ν) in syngas	RADFRAC column module	The wash oil absorbs the hydrocarbon components and is sent for regeneration to the bio-oil desorber column.
Bio-oil desorber	70°C, 33 bar	RADFRAC column module	The wash oil solvent is regenerated and sent back to the absorber column. The desorber top product containing captured tars (benzene, toluene, and xylene) is sent back to the CLG unit.
MDEA scrubber	45°C, 33 bar >99.5% capture of CO ₂ & H ₂ S	RADFRAC column module	The MDEA solvent absorbs the acid gas components (CO ₂ & H ₂ S) and is sent for regeneration to the amine desorber.
MDEA desorber	105°C, 1.5 bar	RADFRAC column module with reboiler	The generated acid gas at the top is directed to the sulphur recovery section. The regenerated amine solvent re-enters the amine scrubber.
NaOH scrubber	40°C, 32 bar	RADFRAC column module	Ultimate gas fine cleaning eliminates remaining sulphur traces prior to entering the FT unit.
Sulphur recovery	40°C, 1.2 bar	RADFRAC column module	The CO ₂ -rich stream is contacted with a KMnO ₄ solution, in which H ₂ S is converted with high efficiency.
FT reactor	250°C, 30bar	RYIELD reactor, embedded calculator based on the Anderson-Schulz-Flory distribution, 60% CO conversion per pass	Cobalt catalyst, high chain growth probability ($\alpha >85\%$) favoring the formation of long-chain hydrocarbons.
SMR	850°C, 30bar Steam to carbon molar ratio = 3	RGIBBS reactor (free energy Gibbs minimization)	The heat requirement is provided by a combustor working at 950°C, which processes a fraction of the FT gas.
FT separator block	30°C, 24 bar	RADFRAC column module	

The stream inputs and operating conditions and requirements of individual modules are specified to allow the enthalpy calculations for each stream. The simulator is also able to compute the energy consumption of individual utili-

ties and compounds and estimate the cost of the plant components.

The results of the mass and energy balances represent the basis for the cost estimation efforts: the capital cost of

equipment is estimated using manufacturers' quotes, literature-published prices, or historical project data related to a specific size parameter of the corresponding equipment, and scaled-up or -down using a known value of the cost of similar equipment of different size, using [43]

$$\text{Cost X} = \text{Cost Y} \left(\frac{\text{Size X}}{\text{Size Y}} \right)^{\text{Scaling factor}}, \quad (10)$$

where Cost Y is the reference cost of equipment having capacity Size Y and Cost X is the computed cost of the equipment of interest, having a capacity Size X. The scaling factor takes values from 0.60 to 0.80, depending on the equipment type.

The installed equipment price, the cost of installation, piping, control instrumentation, etc., are calculated as a percentage of the purchased equipment, depending on the equipment type.

The installed prices obtained are normalised to the year 2020, using the average annual value for the 2020 Chemical Engineering Plant Cost Index (596.2) [44].

2.4.2. Annual Operating Costs. The annual operating costs include fixed (labour, maintenance and repair, plant insurance, and overheads) and variable costs (raw materials, catalysts, utilities, and disposal charges).

For the fixed costs, the maintenance and repair costs are estimated at 3.5% of the total capital investment, and the plant insurance represents an additional 1.5% of the total capital investment. The labour costs are detailed in Table 3, including personnel numbers, salaries, and overheads.

The variable annual costs are computed using the results of the mass and energy balances and the market/literature values for the price of utilities, consumables, and disposal services. The full list of consumables and prices used in this analysis is presented in Table 3, together with the assumptions used for calculating labour costs and the parameters of the economic analysis.

2.5. Technoeconomic Analysis. For the economic analysis, the net present value of the plant and the breakeven selling price of the diesel-naphtha mix are calculated using the discounted cash flow rate of return methodology. The values for the economic parameters used in the economic analysis are presented in Table 3.

A sensitivity analysis is performed to determine the most influential economic parameters and their effect on the breakeven selling price of syncrude.

2.6. Life Cycle Assessment. The life cycle assessment performed in this work is attributional and uses a cradle-to-gate approach: the modelled processes begin with biomass growth and harvesting and end when the syncrude is transported to the conventional refinery for fuel upgrading. A cradle-to-gate LCA approach is employed because assessing the vehicular emissions of using FT biofuels is an extremely challenging task. Several studies showed that the emissions resulting from FT fuel combustion are lower than those of conventional diesel in terms of CO₂, CO, hydrocarbons

TABLE 3: Economic parameters for the study.

<i>Economic parameters</i>		
Plant operating hours	8000	Hours
Discounted cash flow rate	6	%
Interest rate for debt financing	2.5	%
Inflation	3.5	%
Construction period	3	Years
Tax rate	0	%
Contingencies	15	% (EPC)
Working capital	5	% (EPC)
Commissioning cost	5	% (EPC)
Project life	25	Years
<i>Fixed operating costs</i>		
Maintenance	3.5	% (TCI)
Insurance	1.5	% (TCI)
<i>Labour</i>		
Personnel	Shifts	Salary
(i) Technical: 12/shift	3	€62,000/year
(ii) Admin: 8/shift	2	€44,000/year
Overheads	35%	
Total labour cost	3.9	M€/year
<i>Prices for utilities, waste disposal, and raw materials</i>		
Material stream	Unit price (€/tonne or €/MWh)	Reference
Fresh water	2	[45]
Pine forest residue (pellets)	110.2	[33]
Wheat straw (pellets)	76.9	[33]
MDEA	1,500	[46]
Oxygen carrier (ilmenite)	300	[47]
Water-gas shift catalyst	16,000	[48]
Fischer-Tropsch catalyst	35,000	[32]
Wastewater discharge	4	[45]
Ash disposal	25	[48]
Electricity	80	[49]
Wax (selling price)	1800	[32]

(especially PAHs), particulate matter, and NO_x emission profiles [50]. However, most of the studies concern coal or natural gas-derived FT fuels. There are only a couple of studies specifically analysing biomass-derived FT fuel behaviour [51, 52]. Moreover, the emissions associated with fuel upgrading and distribution would be the same for the two cases analysed, so the relative difference between the environmental scores of the FT fuels obtained in the analysed scenarios would remain constant.

The LCA study is conducted according to the guidelines of ISO 14040 and ISO 14044 standards, using the SimaPro™ software. The 4 stages of the LCA are followed: goal and

scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and results interpretation.

The goal of the environmental analysis is to evaluate the environmental impact of FT fuels produced using chemical looping gasification of waste biomass and compare the sustainability of the two feedstock materials to produce transport biofuels.

The functional unit of the study is 1 MJ of mixed naphtha and diesel fractions produced at the plant, and the boundary of the system is presented schematically in Figure 1. The upgrading and distribution of the FT products that are common to both regardless of the initial feedstock have not been included. The study does not consider the influence of capital goods or plant construction.

For the LCI, background processes (wastewater treatment, biomass feedstock supply) are modelled using data from the ecoinvent database (available through SimaPro) [53]. For the foreground processes (biomass consumption, emissions, and waste disposal), the mass and energy balance results were used for modelling. Allocation is avoided by treating the wax fraction as an avoided product, replacing the production of petroleum wax slack, and grouping together the naphtha and diesel fractions.

During the LCIA stage, the set of emission, material, and energy streams assembled during the inventory stage are translated into a number of impact scores using characterisation factors. Two main types of characterisation factors are employed in LCA studies: midpoint and endpoint. The midpoint characterisation factors are further used to derive the endpoint indicators, which are directly correlated to the environmental areas of protection. Several impact assessment methods have been developed and are available in SimaPro, using different characterisation factors and environmental areas of protection [54].

In the present paper, the ReCiPe 2016 method is used for the LCIA stage, employing midpoint environmental indicators and the heuristic approach [55]. The heuristic approach uses a “medium” time horizon for modelling the environmental impacts of the activities considered for 100 years.

The fourth stage in the LCA study is the results’ interpretation, which is presented in the “Results and Discussion” section.

3. Results and Discussion

3.1. CLG Model Validation. The predictions of the CLG mathematical model are verified using experimental data obtained from the 1 MWth pilot plant, operating at the TU Darmstadt, whose layout has been presented in [56]. The experimental campaign was performed as part of the CLARA Horizon 2020 project. The experimental conditions in which the results are obtained are presented in the Supplementary Materials, Table S1.

The results of the model validation efforts are presented in Figure 2, for the pine forest residues (Figure 2(a)) and wheat straw (Figure 2(b)).

The results show a good agreement between model predictions and experimental values, for both biomass samples, especially for the hydrogen and carbon monoxide behaviour.

The highest deviation between model results and experimental data is observed in the case of methane (standard deviation: 14.1% for pine forest residue case and 14.3% for the wheat straw sample) and carbon dioxide (standard deviation: 10.7% for pine forest residue case and 9.4% for the wheat straw sample).

It must be mentioned that successfully validating the mathematical model on laboratory or pilot scale does not necessarily imply that the model would perform well when applied to a larger scale. It is however an essential step to be able to confidently simulate the performance of a large scale CLG unit.

3.2. Mass and Energy Balance. A simplified version of the process flow diagram is presented in Figure 3, showing the BtL plant layout and selected results of the mass balance for the main process units.

The composition of the different syngas streams, as well as those for the three FT syncrude fractions (and their associated lower heating values) obtained are presented in the Supplementary Material, Table S2.

The cold gas efficiency (CGE) of the gasification unit has been computed, according to equation (11), where G_m is the mass flow rate of syngas/biomass (t/h), and LHV represents the lower heating value of syngas/biomass (MJ/kg).

$$CGE = \frac{LHV_{\text{syngas}} \cdot G_{m,\text{syngas}}}{LHV_{\text{biomass}} \cdot G_{m,\text{biomass}}} \cdot 100, \% \quad (11)$$

For the wheat straw case, the computed CGE was 78.2%. For the pine forest residue, the CGE was 80.4%.

The results of the heat balance over the fuel synthesis plant are presented in Figure 4(a) for the pine forest residue case and Figure 4(b) for the wheat straw scenario.

In both scenarios, the excess of ≈ 17 MW useful (low grade) heat could be sold to industrial consumers or used for district heating.

For the entire plant, the energetic fuel efficiency (EFE) was computed, similar to the cold gas efficiency for the gasification unit but considering the lower heating value of the syncrude (equation (12)) where G_m is the mass flow rate of syncrude/biomass (kg/s), and LHV represents the lower heating value of syncrude/biomass (MJ/kg).

$$EFE = \frac{LHV_{\text{syncrude}} \cdot G_{m,\text{syncrude}}}{LHV_{\text{biomass}} \cdot G_{m,\text{biomass}}} \cdot 100, \% \quad (12)$$

The results show that the proposed plant has a EFE of 52.3% in the case of wheat straw and 53.4% in the pine forest residue scenario.

3.3. Cost Estimation Results

3.3.1. Fixed Capital Cost. The breakdown of the capital cost of the BtL plant (including the integration cost) is presented in Table 4.

The total equipment installation price is €165.3 million, to which the integration and start-up cost (€28 million), and the building and land cost (€9.87) million are added.

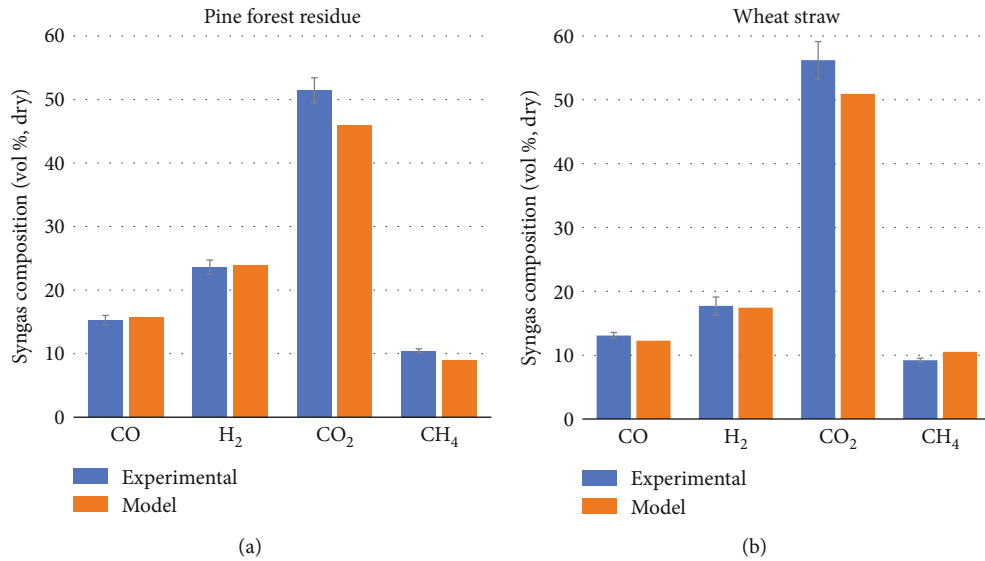


FIGURE 2: Model validation results for (a) pine forest residue and (b) wheat straw.

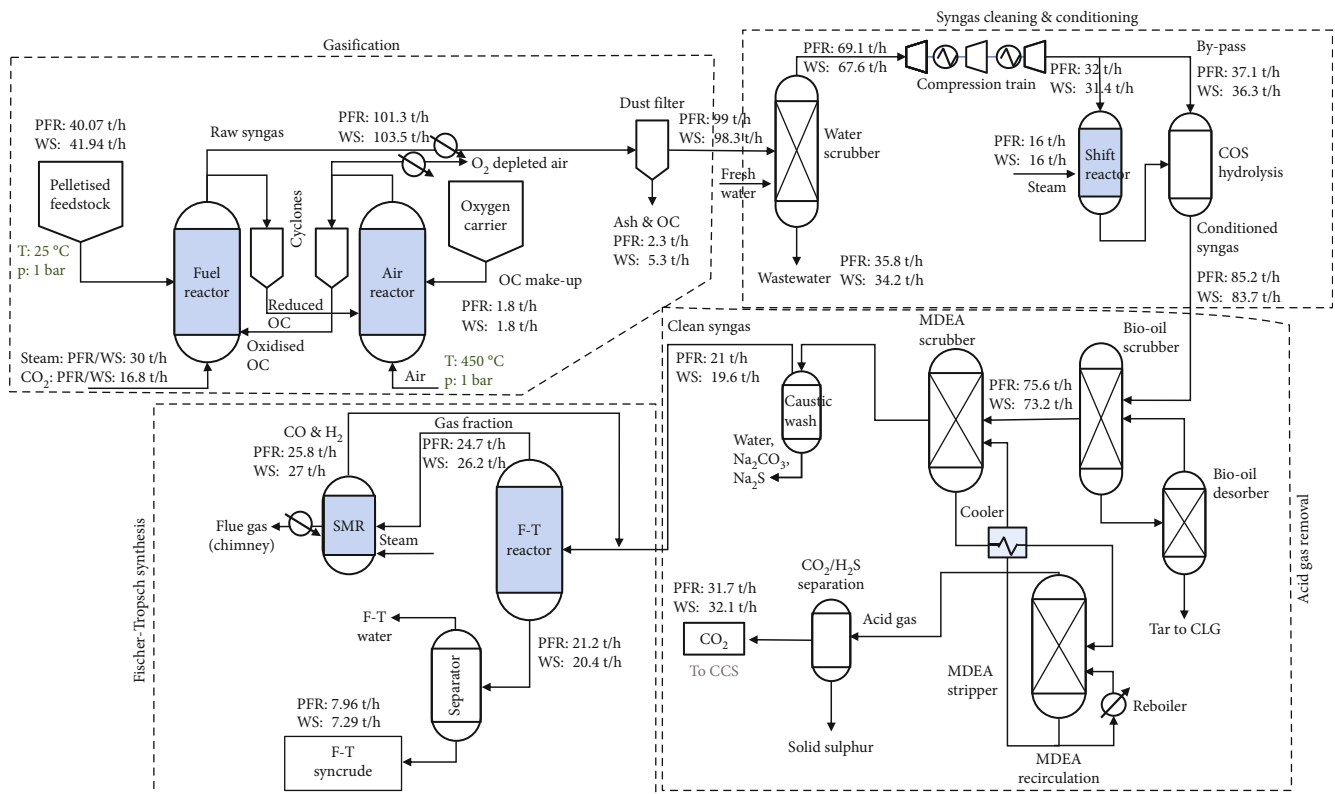


FIGURE 3: Simplified schematic diagram of the process and mass balance results.

A breakdown of the equipment costs for each process unit is provided in Figure 5.

The total project investment required for the BtL plant is estimated at approximately €273 million, including owner cost, contingencies, and the interest charges for the construction period. The largest proportion of the cost is incurred by the gasification unit (€68.9 million), which represents approx-

imately 34% of the total installed cost. The air and fuel reactors are the costliest equipment, accounting for 48.8% of the costs of the gasification unit. The second most expensive unit is the FT synthesis, in which the main cost components are the FT reactor and the SMR unit, while the acid gas removal unit and the syngas cleaning and conditioning account for less than 37% of the total costs.

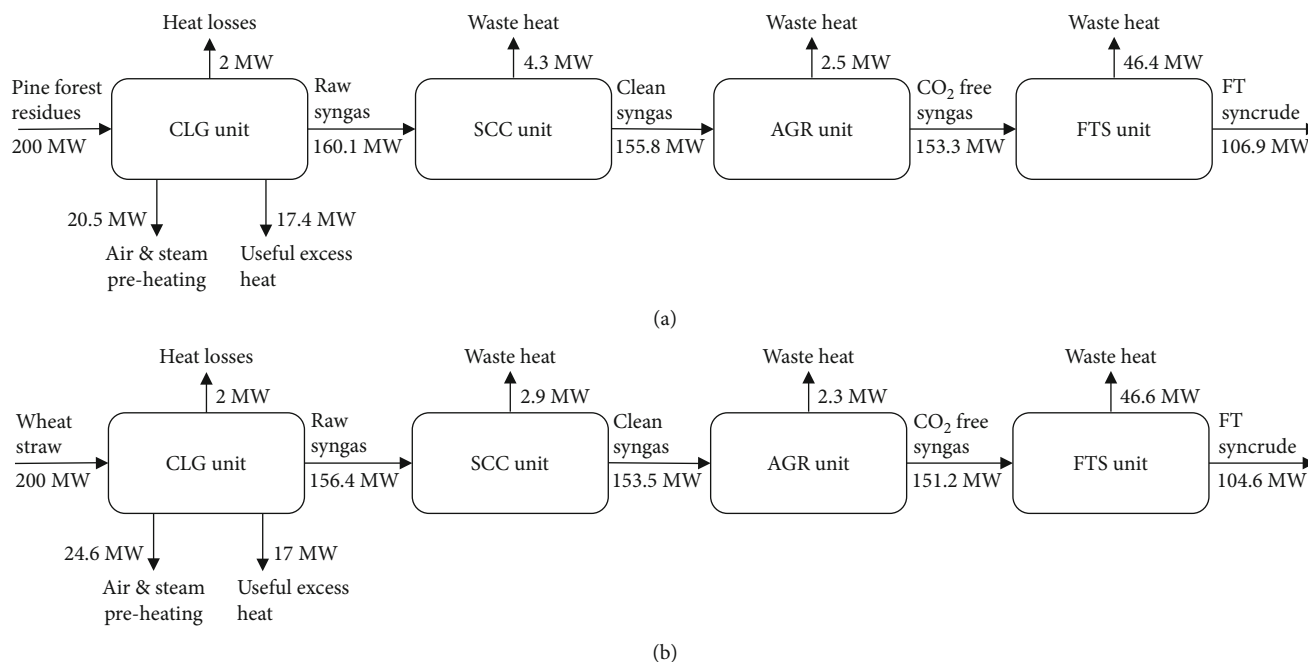


FIGURE 4: Overall heat balance for (a) pine forest residue and (b) wheat straw.

TABLE 4: Capital cost estimation results.

Equipment installed cost	(M€)
Chemical looping gasification	55.89
Syngas cleaning and conditioning	31.06
Acid gas removal	34.96
F-T synthesis	43.42
<i>Total equipment installed cost</i>	<i>165.33</i>
Start-up and integration cost	
Chemical looping gasification	12.73
Syngas cleaning and conditioning	3.73
Acid gas removal	5.24
F-T synthesis	6.51
Buildings & land	9.87
<i>Total installed cost</i>	<i>203.42</i>
<i>Owner cost</i>	<i>20.34</i>
<i>Contingency</i>	<i>30.51</i>
<i>Interest charges</i>	<i>18.69</i>
<i>Total project investment</i>	<i>272.97</i>

3.3.2. *Annual Operating Costs.* Using the assumptions detailed in the previous section and the results of the mass balance, the annual operating costs are calculated for the two cases and shown in Figure 6. The flowrates of raw materials, utilities, catalyst consumption, and waste disposal are presented in the Supplementary material, Table S3. The total operating costs of the BTL plant range from €68.1 million in the case of pine forest residues to €58.9 million for the wheat straw case. In both cases, the biomass supply represents the highest proportion of the operating costs: 52% for the pine forest residue case and 44% for the wheat

straw scenario. Due to the significantly lower price of wheat straw pellets, the operating costs in this scenario are 13.5% lower than for the case of pine forest residues.

The fixed operating costs represent 21% and 25% of the annual operating costs in the case of pine forest residues and wheat straw, respectively. The consumables (solvents, oxygen carrier make-up) and utilities account for the remainder of 27% and 31% for the pine forest residues and wheat straw, respectively, with electricity accounting for the largest proportion of these costs.

In both scenarios, the operating costs are balanced by the revenue obtained from FT wax selling. In the case of pine forest residue, the annual revenue is approximately €57.8 million, while for the wheat straw, the value is €52.7 million.

3.4. *Breakeven Selling Price.* The breakeven selling price (BESP) represents the price at which biofuel must be sold, so that, at the end of the plant lifetime, its net present value is equal to zero.

Using the net present value analysis, the computed value for the BESP of the FT syncrude using pine forest residues is €0.82 per litre and €0.78 per litre for wheat straw. If expressed with respect to the energy content of the FT products, the BESP becomes €84.5 per MWh and €80.8 per MWh for pine forest residue and wheat straw, respectively. Reportedly, production costs for FT crude vary widely, from 42 to €140/MWh, depending on plant scale and capacity, gasification technology, process conditions, economic assumptions, etc. However, direct comparisons with other TEA studies are difficult, due to differing economic assumptions and TEA methodologies, as well as the scale of the analysed installation. The study of Saeed et al. [30] (using a CLG configuration and pine forest residues) established a levelized cost of FT syncrude of €110/MWh. However, the biofuel plant

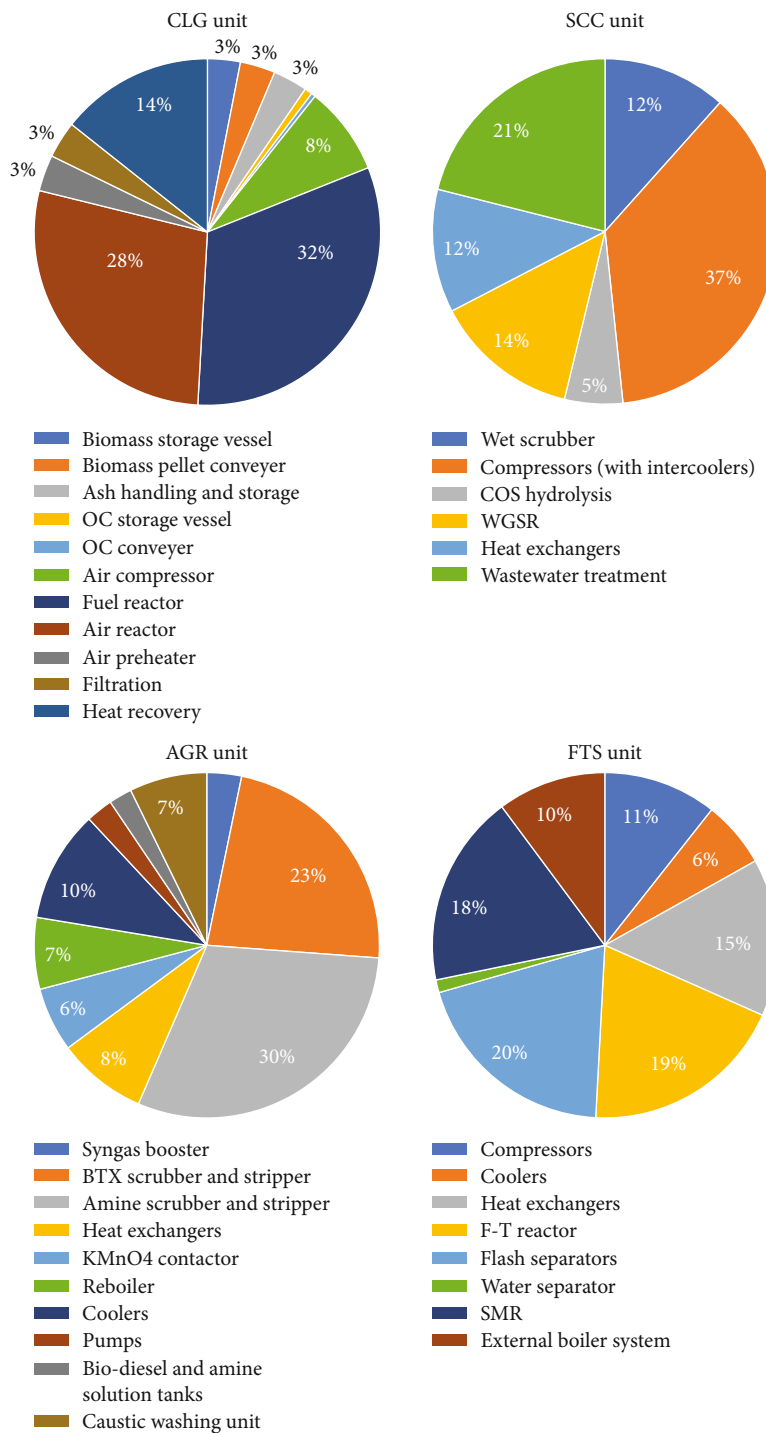


FIGURE 5: Installed cost breakdown for the main process units and equipment.

analysed had a lower biomass capacity, which can account for the increase in fuel costs. Using a similarly lower plant capacity, Kumar et al. [29] obtained values ranging between €119.3 and 146.9/MWh, in the absence of CO₂ capture tax credits and between €57.6 and 77.9/MWh when considering CO₂ credits.

The study conducted by Habermeyer et al. [32] assessed the economic perspectives of a 200 MWth circulating fluidised bed (CFB) gasification facility for the conversion of

forest residue chips to transport fuels. They obtained a production cost of €0.66 per litre for FT syncrude, lower than the BESF values determined in this study. This is attributed to differences in the considerations of the economic parameters and assumptions taken in both studies.

For a more accurate and fair economic comparison between chemical looping gasification and a conventional gasification setup, the BESF was computed from an equivalent plant design employing a CFB reactor. For the equivalent

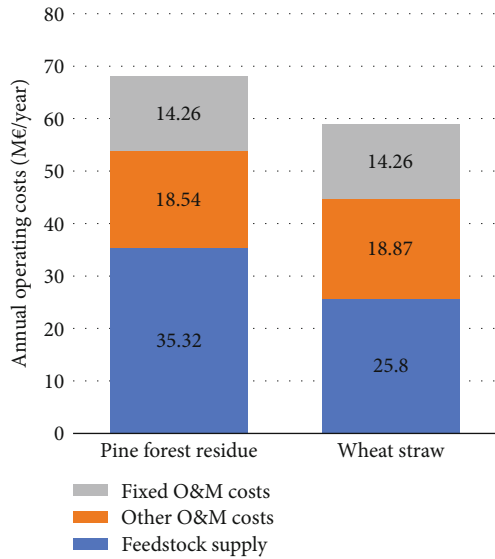


FIGURE 6: Cost breakdown of the annual operating costs.

plant design, it was assumed that the two CLG reactors would be replaced by a CFB reactor and ASU, maintaining the same gasification performance (i.e., syngas yield and composition, with negligible differences regarding steam, air, and CO₂ requirements). All other equipment costs and economic assumptions remain the same as for the CLG design, while the operational cost is increased due to the additional electricity requirement for the ASU. The calculated capital and operating cost estimation methodology is presented in the Supplementary Material (Tables S4 & S5), and the results of the analysis are summarized in Table 5.

For the pine forest residue scenario, the BEP value obtained using the conventional oxygen CFB gasification (including the air separation unit) is €0.89 per litre FT fuels, showing a 9% increase compared to the BEP produced by the CLG plant configuration.

3.5. Sensitivity Analysis. To quantify the impact of uncertainties regarding costs and economic conditions on the BEP value, a sensitivity analysis has been performed. The influence of variations in capital cost, biomass feedstock cost, plant availability and lifetime, discounted cash flowrate, wax selling price, electricity price, and contingency value was investigated. Figure 7 presents the results of the sensitivity analysis for the pine residue scenario; those for the wheat straw case are presented in the Supplementary material.

The most significant economic parameters are the wax selling price, plant availability, fixed capital investment, and feedstock price. The wax selling price variation has the most significant influence: the proposed $\pm 11.1\%$ variation in the wax selling price produces a $\pm 19.8\%$ deviation in BEP values. Possible plant disruptions, reflected in a 20% lower plant availability, lead to an increase of 26.7% of BEP, to values above €1 per litre.

The ability to source cheaper biomass (€88.5/tonne) would lower the BEP to only €0.61/L, amongst the lowest estimations found in the literature. In contrast, if the

TABLE 5: Comparison of project capital and operating costs using CLG and CFB gasification.

Biomass: Pine Forest residue	CLG	CFB
Total project investment (M€)	273.0	288.4
Annual O&M costs (M€/year)	68.1	69.9
BEP (€/m ³)	816	894

biomass price increases to €130.6/tonne, the BEP can reach €1 per litre FT syncrude. Additionally, a 30% lower capital investment would lead to BEPs under €0.6/L, while an increase of 30% would lead to values close to €1.1/L.

The electricity price, the discounted cash flow rate, and the project lifetime have a moderate influence on the computed BEPs, while high uncertainties in contingency estimations produce only slight variations in the F-T fuels' BEP.

3.6. CO₂ Avoidance Cost. If grown in a sustainable manner, the use of biomass for biofuel production, which is considered to produce no net CO₂ emissions in its life cycle, and as a replacement for fossil fuels in energy or power generation systems is one of the most effective ways of reducing CO₂ emissions. If the BtL plant is equipped with a CCS facility, negative CO₂ emissions will be achieved. A CCS facility would require additional compressors, leading to an increase in the plant capital investment costs. The operating costs would also increase to account for increased electricity consumption and CO₂ transport requirement. The total installed cost of the carbon capture and storage (CCS) plant is about €10 million, and the operating and maintenance costs associated with the CO₂ transport are about €6/tonne CO₂ [57].

The CO₂ avoidance cost is calculated according to the following [58]:

$$\text{€/t CO}_2 \text{ avoided} = \frac{\text{€/MWh} - \text{bioFT} - \text{€/MWh} - \text{fossil}}{t_{\text{CO}_2\text{-fossil}}/\text{MWh} - t_{\text{CO}_2\text{-bioFT}}/\text{MWh}}, \quad (13)$$

where €/MWh is the BEP or market price of FT syncrude and diesel/gasoline, respectively, expressed with respect to the energy content; and $t_{\text{CO}_2\text{-fossil/bioFT}}$ are the emissions (tonne of CO₂) corresponding to 1 MWh of fossil fuels or FT syncrude, respectively.

The CO₂ avoidance cost is given by the ratio between the difference of the BEP of the FT syncrude from the BtL plant and of diesel/gasoline and the difference of the relative CO₂ emissions.

To account for the emissions during the biomass pretreatment process the electricity requirement is estimated at 0.12 MWh and 0.10 MWh per tonne of treated biomass for PFR and WS, respectively; the average CO₂ intensity of grid electricity in the EU is approximately 0.275 tonne CO₂/MWh.

For feedstock transportation-related CO₂ emissions, it is assumed the average CO₂ intensity of 56.5 gCO₂ per tonne-kilometre (tkm) [59] and the average transportation distance

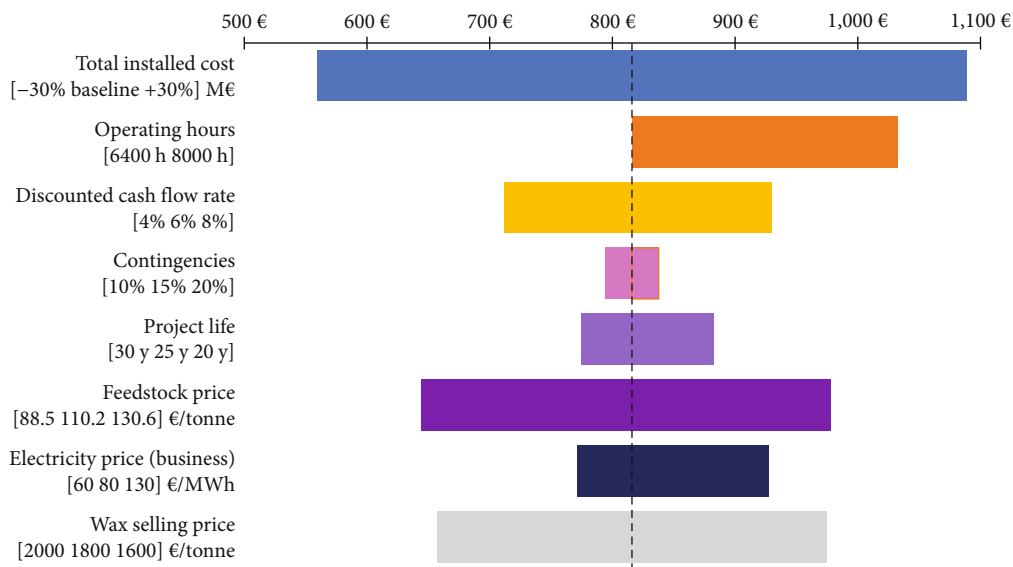


FIGURE 7: Sensitivity analysis of different economic parameters for pine forest residue-derived F-T fuels (dashed line refers to the base case study).

TABLE 6: CO₂ emissions' inventory to produce 1 MWh of FT syncrude*.

	Pine Forest residue (PFR)	Wheat straw (WS)
Indirect CO ₂ emissions derived from onsite electricity consumption (tCO ₂ /MWh)	0.079	0.086
Indirect CO ₂ emissions derived from electricity consumption (feedstock pretreatment plant) (tCO ₂ /MWh)	0.027	0.025
CO ₂ emissions derived from feedstock transport (tCO ₂ /MWh)	0.0047	0.0053
CO ₂ captured (tCO ₂ /MWh)	0.661	0.725
Net CO ₂ emissions (tCO ₂ /MWh)	-0.551	-0.609

*the average calorific value of the FT syncrude is around 9.7 MWh/m³.

TABLE 7: LCIA results.

Impact category	Unit	Pine forest residue	Wheat straw
Global warming	kg CO ₂ eq.	1.94E-02	2.64E-02
Stratospheric ozone depletion	kg CFC ₁₁ eq.	-3.73E-09	2.14E-07
Ionizing radiation	kg Co-60 eq.	1.43E-02	1.51E-02
Ozone formation, human health	kg NO _x eq.	6.18E-05	4.47E-05
Fine particulate matter formation	kg PM _{2.5} eq.	5.14E-06	5.12E-05
Ozone formation, terrestrial ecosystems	kg NO _x eq.	6.27E-05	4.28E-05
Terrestrial acidification	kg SO ₂ eq.	-2.24E-05	3.03E-04
Freshwater eutrophication	kg P eq.	3.24E-05	3.81E-05
Marine eutrophication	kg N eq.	1.54E-05	2.00E-05
Terrestrial ecotoxicity	kg 1,4-DCB	5.84E-03	-1.47E-02
Freshwater ecotoxicity	kg 1,4-DCB	5.12E-03	1.10E-02
Marine ecotoxicity	kg 1,4-DCB	6.98E-03	1.52E-02
Human carcinogenic toxicity	kg 1,4-DCB	2.02E-03	2.18E-03
Human noncarcinogenic toxicity	kg 1,4-DCB	1.78E-01	3.98E-01
Land use	m ² a crop eq.	2.88E-01	7.34E-03
Mineral resource scarcity	kg Cu eq.	3.43E-03	3.69E-03
Fossil resource scarcity	kg oil eq.	-1.92E-02	-1.87E-02
Water consumption	m ³	7.46E-04	7.85E-04

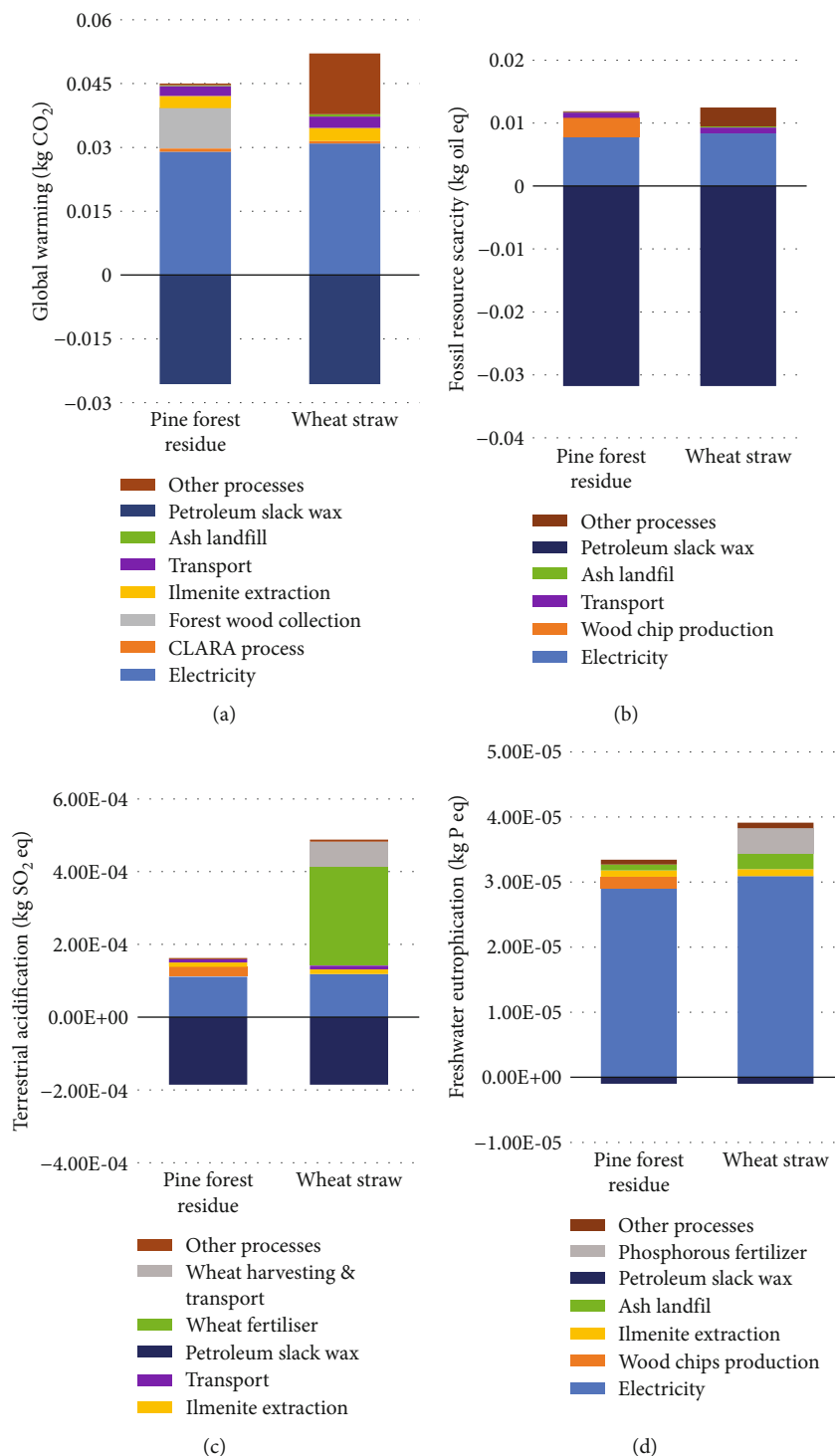


FIGURE 8: LCIA results for the global warming (a), fossil resource scarcity (b), terrestrial acidification (c), and freshwater eutrophication (d) categories.

for the biomass from the decentralised pretreatment facilities to the gasification plant of 100 km.

The resulting CO₂ emissions per MWh FT syncrude produced are presented in Table 6.

To estimate the CO₂ avoidance cost for fuel switching an average carbon intensity of petrol and diesel are assumed to be 0.338 tCO₂/MWh (or 94 gCO₂/MJ), specified in the

Renewable Energy Directive (RED II) [60]. In 2019, the average EU price (excluding taxes and tariffs) of diesel was €0.61 per litre, while that of gasoline was €0.56 per litre. In 2020, the average EU price of diesel dropped to €0.48 per litre, while that of gasoline was €0.45 per litre. In 2021, the average EU diesel price increased to €0.65 per litre and that of gasoline reached €0.64 per litre. Making a similar mix of

diesel and gasoline as the one in the FT syncrude produced, it is assumed that the average fossil fuel price ranges from €46.6 to €64.5 per MWh for the reference fuel.

When the CCS system is installed on the BtL plant and biomass feedstocks which are recognised as carbon neutral, the specific CO₂ emission intensities are -0.551 tCO₂/MWh for the PFR scenario and -0.609 tCO₂/MWh for the WS scenario. If the price of the mix of diesel and gasoline (excluding taxes and tariffs) is €46.6 per MWh, the CO₂ avoidance costs are €74.4/tCO₂ for the PFR scenario and €61.3/tCO₂ for the WS scenario. If the price of the mix of diesel and gasoline goes up to €64.5 per MWh, the CO₂ avoidance costs are reduced to €46.7/tCO₂ for the PFR scenario and €35.8/tCO₂ for the WS scenario.

3.7. Life Cycle Assessment. The results of the life cycle assessment are presented in Table 7. The scores for each of the 18 impact categories considered by the ReCiPe method are listed for 1 MJ of fuel mix, derived from pine forest residue or wheat straw. The full life cycle inventory, containing emission data, is listed in the Supplementary material, Table S6.

3.7.1. Global Warming Potential. The global warming (GW) potential is one of most important impact categories that produces contradictory results across multiple LCA studies of biofuel production systems [61]. When biomass is considered carbon neutral without CCS, the score in the GW category is 19.4 g CO₂ eq. per MJ for the pine forest residue derived fuels and 26.4 g CO₂ eq. per MJ for the wheat straw derived fuels. Furthermore, adding CCS to the BtL plant reduces the score to -144.5 g CO₂ eq. per MJ for the pine forest residue derived fuels and -154.2 g CO₂ eq. per MJ for the wheat straw derived fuels. It is worth noting that the scores obtained in this study do not consider fuel upgrading, distribution, and use. However, Iribarren et al. [8] showed that the impact of fuel upgrading and distribution was minor compared to that of syngas generation and FT synthesis.

For comparison, the average carbon intensity of petrol and diesel supplied in the EU is still assumed to be 94 g CO₂ eq. per MJ. Thus, the use of the fuel mix obtained from either wheat straw or pine forest residues can achieve the reduction in GHG emissions of up to 79% without CCS and 264% with CCS.

Figure 8(a) shows that the main process responsible for the negative environmental impacts in the GW category is the high electricity consumption required in the biomass pretreatment and fuel synthesis process.

3.7.2. Fossil Resource Scarcity, Acidification, and Eutrophication. In the fossil resource scarcity (FRS) category, both fuels show a negative result (positive environmental impact), due to displacing the production of paraffin wax (Figure 8(b)). The effect is slightly more pronounced for the pine forest residue case, due to the higher wax yield in this scenario. The electricity use has the most important negative contribution to the FRS score.

In the terrestrial acidification (TA) category, where most biofuels generally perform poorer than conventional fossil fuel alternatives, the pine forest case shows a positive environmental impact (Figure 8(c)). This is mainly due to the

avoided production of petroleum wax. In contrast, in the wheat straw case, the use of fertilizer and the harvesting and transport processes outweigh the benefits of wax production from renewable resources.

In both scenarios, the freshwater eutrophication category (Figure 8(d)) shows a positive environmental score due to the electricity consumption and (to a lesser extent) that of the use of fertilizers, ash landfill, and ilmenite extraction.

4. Conclusions

Chemical looping gasification followed by the Fischer-Tropsch synthesis is a promising alternative for the production of liquid transportation fuels, still in its early stages of pilot scale demonstration. To facilitate the transition of this technology to industrial scale, several technical and economic aspects must be first elucidated.

This work has examined the technical.

The energetic fuel efficiency obtained using the proposed plant configuration is approximately 52-53%, comparable with other proposed plant configurations using conventional gasification systems.

The plant requires a total project investment of €272.97 million, with annual operating costs ranging from €58.9 to €68.1 million for the wheat straw and pine residue scenarios, respectively.

The resulting breakeven selling prices for the FT fuels are €0.82 per litre for the pine residue scenario and €0.78 per litre for the wheat straw case. These prices are approximately 9% lower than those produced using a conventional gasification setup, but still higher than fossil fuel prices reported between 2013 and 2021. However, the prices of automotive fuels reported in 2022 (average values of €1.07 and €1.01 for diesel and gasoline) make the prospect of biowaste-derived fuels extremely attractive.

The sensitivity analysis showed that reducing capital investment, sourcing cheaper biomass feedstocks, and achieving higher revenues from the selling of FT wax are the most effective ways to achieve BESPs comparable with fossil fuel prices.

Additionally, the export of excess heat to industrial consumers can further offset the prices of FT fuels and increase the profit margin of the BtL plant. Outfitting the plant with CCS capability and using carbon neutral biomass will lead to carbon negative emissions. The CO₂ avoidance cost varies between €74.4/tCO₂ for the pine forest residues and €61.3/tCO₂ for the wheat straw pellets. In a high cost fossil fuel scenario, the avoidance costs can reach values as low as €46.7/tCO₂ and €35.8/tCO₂ for the pine forest residues and wheat straw scenarios, respectively.

The environmental analysis showed that the biofuels obtained using the CLARA concept would be suitable under the framework of the RED, achieving up to 79% reduction without CCS and up to 264% with CCS in the GHG emissions compared to conventional fossil fuels.

Data Availability

Data used for the article is available upon request.

Disclosure

A preprint version of this article has been published [62].

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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Supplementary Materials

The supplementary material contains the experimental conditions for the model validation experiments (Table S1), the composition and heating values of the syngas and FT syncrude streams (Table S2), the mass flow rates used for computing the variable operating costs (Table S3), the cost estimations sources (Table S4) and gasification unit cost (Table S5) used for the comparison between CLG and CFB gasification options, the data for the inventory stage of the LCA (Table S6), and the results of the economic sensitivity analysis for the wheat straw case (Figure S1). (*Supplementary Materials*)

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