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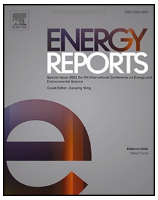

Research paper

Production of methanol and electricity from biogenic waste integrating hydrothermal carbonization, sorption enhanced gasification, hot gas conditioning, plasma CO2 conversion and SOFC

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ABSTRACT

This study presents the development and preliminary integration of Hydrothermal Carbonization (HTC), sorption-enhanced gasification (SEG), hot-gas cleaning (HGC), dielectric barrier discharge (DBD) plasma-assisted CO_2 conversion, oxygen-selective membranes, methanol synthesis via membrane, and solid oxide fuel cells (SOFC) to produce methanol and electricity from biogenic waste. The system was investigated within the EU Horizon 2020 project GICO, combining laboratory and pilot-scale data and modelled in Aspen Plus for a 1 MWth biomass input. This paper builds upon previous work by extending the process model to include methanol synthesis and SOFC units. The focus is on both the development of individual subsystems and their preliminary integration within a unified process configuration. Initial outcomes show promising performance: the SEG process generates 21.2 kg/h of hydrogen (118 g/kgBiomass), which is utilized as an intermediate for methanol synthesis and SOFC operation. The system achieves a conversion efficiency based on the LHV of the input biomass of 28 % for methanol production (250 g/kgBiomass) and generates of 292 kWe and 279 kWth via SOFC, corresponding to 24 % electrical and 23 % thermal global LHV-based efficiencies, respectively (not considering the DBD consumption that can lead also to electricity requirement than generation) The corresponding global efficiencies reaches 52 %, increasing to 74 % with thermal recovery. These results represent an intermediate advancement in the development of production of methanol and electricity from biogenic waste, highlighting the

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potential of integrated biomass valorization pathways and underscoring the need for further optimization and experimental validation.

1. Introduction

In recent years, fossil fuels have diminished in their appeal as an energy source, primarily due to concerns about global warming, climate change, and the imperative for countries to establish self-sustaining energy resources. A reliable substitute for fossil fuels is biomass, a renewable resource abundantly available worldwide. From an environmental standpoint, biomass is considered favourable due to its net carbon balance, where the carbon dioxide emitted during combustion is largely offset by the CO_2 consumed during photosynthesis. Utilizing organic waste from industrial, agro-forestry, and municipal activities presents a cost-effective and sustainable strategy that avoids competition between food and fuel and reduces pollution caused by waste through its repurposing and valorisation (Kuchler and Linnér, 2012; Pala et al., 2017; Bocci et al., 2014; Thapa et al., 2017; Büyüktahtakın and Cobuloglu, 2015).

Gasification emerges as a promising technology for converting biogenic waste to generate heat, electricity, and other biofuels with high efficiency and reduced environmental impact (Campoy et al., 2014; Liao et al., 2014). It involves the partial oxidation of solid feedstock, converting its chemical energy through the production of a gas stream, known as syngas, composed of hydrogen, carbon monoxide, carbon dioxide, water vapor, and methane, alongside unwanted by-products like tar and inorganic compounds (Campoy et al., 2014, Marcantonio et al., 2020). Compared to air gasification and other energy conversion processes such as combustion and anaerobic digestion, steam, oxygen and CO_2 gasification offer key advantages. These include the production of high-quality syngas (without nitrogen dilution), high electrical efficiencies (due to the compatibility with fuel cells, turbines and internal combustion engines), suitability for fuel synthesis, and broad applicability to various feedstocks due to the higher conversion efficiencies of thermochemical over biochemical processes (Adams and McManus, 2014; Fryda et al., 2008, Vitale et al., 2024). In particular, steam gasification (SG) is widely adopted in biomass conversion due to the greater reactivity of water as a gasifying agent. The presence of steam enhances hydrogen production and facilitates hydrocarbon reforming, resulting in a syngas with higher hydrogen content and overall superior quality compared to that produced by alternative gasification technologies (Papa et al., 2025). Specifically, the H_2-rich syngas produced, with lower CO_2 content and higher low heating value (LHV), represents a suitable intermediate for biogenic waste-to-fuel processes. However, SG is also more energy-intensive, as significant heat input is required to generate steam and maintain the desired working temperature. In addition, direct biomass gasification encounters several challenges, especially related to high moisture content, ash content, and poor grindability of certain feedstock.

Hydrothermal carbonization (HTC) emerges as a promising technology to upgrade wet biomass without requiring a drying step prior to gasification. HTC involves the thermochemical transformation of biomass in the presence of water, resulting in hydrochar production, along with valuable liquid by-products (Zeng et al., 2021; Zhuang et al., 2022). The resulting hydrochar is more suitable for gasification treatment, owing to its higher carbon content and lower oxygen content compared to raw biomass (Papa et al., 2024). Furthermore, its use is expected to reduce the formation of aromatic hydrocarbons during the gasification process.

The integration of carbon capture and utilization (CCU) with SG, specifically through the use of CaO-based sorbents in place of inert materials in an indirectly heated fluidized bed reactor, can further enhanc the biomass exploitation process. This approach, known as Chemical Looping Gasification (CLG), and in particular Sorption Enhanced Gasification (SEG), offers several advantages. SEG enables local CO_2 capture enhancing syngas quality and hydrogen yields, shifting the composition from approximatively 40 % H_2, 23 % CO and CO_2 and 13 % CH_4, to around 90 % H_2, 5 % CO, 3 % CH_4 and 2 % CO_2 on a dry volume basis. Moreover, CaO-based sorbent into a SEG process represents a solid carrier for the CO_2 that can be further delivered and converted in other sections of the process. In particular, pure CO_2 produced by the calcination can be reduced in CO by dielectric barrier discharge (DBD) or directly used into chemical synthesis. Although it is not considered in this paper, there is the possibility to store part of this CO_2 since the composition is more than 95 % mol compatible with CO_2 storage in deep geological formation, with the aim of turning the GICO process into a net-zero system.

Furthermore, by operating at lower gasification temperatures compared to conventional gasification, SEG enables the use of biomass with low ash melting points and reduces the external energy demand required to sustain the process (Dai and Whitty, 2022; Martínez et al., 2020; Martínez et al., 2022; Parvez et al., 2021). Moreover, the gasifier doubles as a carbonator and the combustor as a calciner, thereby simplifying the system and reducing the need for solid circulation. Nevertheless, lower operating temperatures lead to increased unreacted char and tar content (Chen et al., 2020; Majoumerd et al., 2012; Gholamian and Zare, 2016).

Among power to gas (P2G) technologies, non-thermal plasma methods are gaining interest for their potential to convert gaseous streams (e.g. CO_2 into CO and O_2 Bongers et al., 2017; Goede, 2018) because they use low-temperature operation (chemical reactions without heating the entire gas volume and no need of materials withstanding extreme conditions as in the thermal plasma), selective activation (excite or ionize specific molecules meanwhile the high temperatures of thermal plasma can lead to unwanted byproducts), fast response time (ideal for dynamic or intermittent energy sources as renewable electricity), no need for catalysts (avoiding issues like catalyst deactivation, poisoning, electrode degradation typical of the catalytic and electrolysis processes) but they still suffer for low energy efficiency (less than thermal or catalytic processes due to energy losses in plasma generation), complex reactor design (for the high-voltage equipment and careful control of plasma conditions) and limited scalability (respect to thermal, catalytic and electrolysis process). Among non-thermal plasma technologies DBD reactors are particularly promising due to their low-cost components, high wall-plug efficiency, scalability, and inherent thermal stability (Mei et al., 2015, 2016; Mei and Tu, 2017; Ramakers et al., 2015; Mori et al., 2016; Kogelschatz et al., 1997; Brandenburg, 2018; Renninger et al., 2022).

At the laboratory scale, early studies reported high CO_2 dissociation energy efficiencies, up to 80 % for subsonic and 90 % for supersonic flow, under optimized conditions of gas pressure, electron density, and electron energy in microwave-excited (MW) plasmas (Friedman, 2008). However, high-frequency (HF) DBD plasmas, operating in the 100 kHz range (Kogelschatz et al., 1997), are considered more promising for practical implementation compared to MW plasmas, due to several advantages: low cost, high wall-plug-to-plasma efficiency, availability of low-cost components for high average power drivers, avoidance of MW matching networks, and ease of scale-up to industrial sizes (e.g., ozonisers; Brandenburg, 2018). Unlike direct current glow discharges (Staack et al., 2005), DBD plasmas can be stably operated at atmospheric and higher pressures (Renninger et al., 2022), due to the intrinsic ability to prevent thermal runaway on electrode surfaces by limiting the charge transferred per cycle.

Various plasma systems have since been applied to CO_2 valorisation, including DBD (Renninger et al., 2022), glow discharges (Staack et al., 2005), gliding arcs, and rotating arcs (Zhang et al., 2018), targeting both direct CO_2 dissociation and downstream conversion via methanation (Zhang et al., 2018) or dry reforming (Zhang et al., 2023) when catalytic systems are integrated into the discharge region. To the best of authors knowledge, the highest reported energy efficiency for CO_2 conversion thus far is approximately 43 %, achieved with a glow discharge—specifically, a rotating arc system (Renninger et al., 2022). However, none of these studies have explored the use of high-power (kW-scale), water-cooled DBD systems.

A persistent challenge identified since early research is the trade-off between CO_2 conversion rate and energy efficiency (Friedman, 2008). High energy efficiencies are typically observed at low conversion rates, primarily due to the recombination (back-reaction) of dissociated CO and O_2 within the plasma zone. This phenomenon leads to stringent requirements on the separation of discharge products, particularly the removal of O_2 from CO, to enable downstream utilization such as hydrogen production via the water–gas shift (WGS) reaction. Accordingly, integrating O_2-selective membranes within the plasma system has been suggested to address this issue (Chen et al., 2020).

To ensure the purity of syngas and eliminate impurities, hot gas cleaning (HGC) processes, such as filter candles and sorbent reactors as a secondary conditioning system, have proved to be effective. Bed gasifier sorbents and catalytic filter candles work synergistically, because the catalytic filter in the freeboard of the gasifier not only avoids any particle to exit the gasifier, increasing the residence time of the gasifier, but also brings ashes and solid carbon that act as catalysts and sorbents to reduce tar as well as sulphur and chlorine compounds (Papa et al., 2023). Moreover, secondary hot sorbents reactors reduce sulphur and chlorine compounds within the limits required for SOFC and methanol synthesis (Marcantonio et al., 2020a). In details, the organic contaminants, tars, from biomass gasification are mainly composed of benzene, toluene, naphthalene and can contribute to electricity generation in SOFCs through reforming, nevertheless these components expose the anode to the risk of degradation due to carbon deposition (Liu and Aravind, 2014) and so Ni deactivation, voltage drop and pressure increase also at 10 ppm(v) of naphthalene and 24 ppm(v) of toluene (Jeong et al., 2019; Papurello et al., 2016). Regarding H_2S and HCl (Błesznowski et al., 2013) pointed out that the limit for correct SOFC operation is 1 ppm for H_2S. For HCl the limit for feasible SOFC operation was discovered to be 10 ppm at 750°C.

The cleaned syngas (rich in H_2) and the CO-enriched gas from the cleaned flue gas converted through the DBD unit can be used for methanol synthesis, increasing the potential of the plant.

According to the stoichiometric ratio of $(H_2\text{-}CO_2)/(CO+CO_2) = 2$ required by methanol production, the process leads to CO surplus production. The excess of CO and methanol synthesis by-products can be used in solid oxide fuel cell (SOFC), which operating at high temperature produce electricity and heat through chemical energy conversion with high electrical efficiency (up to 60 %) and low emissions (Marcantonio et al., 2020b, 2022; Wasajja et al., 2020; Saadabadi et al., 2019). Although extensive research exists on individual units, integrated systems combining HTC, SEG, DBD plasma, HGC, methanol synthesis, and SOFC remain underexplored. Investigating such a system aligns with the EU Strategic Energy Technology (SET) Plan and circular economy objectives, aiming to analyse plant performance (e.g. yields, efficiency) and contribute to a more sustainable energy perspective. Combining these technologies could lead to a system capable of producing methanol and electricity from biogenic waste at competitive costs and lower environmental impacts. In particular, this paper developed within the four-years GICO Horizon 2020 project (2020–2024), extends the model developed in previous work (Marcantonio et al., 2023) by integrating methanol synthesis and SOFC units.

The work expands the process modelling furnishing an updated configuration of the system. Indeed, the previous work, that used out of use wood as biomass feedstock with constant flow rate set to 180 kg/h (1 MW th input size, considering HHV), pointed out that the products of the HCG unit had a dry syngas composition of approximately 70 % H_2, 22 % CO, 7 % CO_2, and 1% CH_4, with H_2S concentrations below 0.05 ppm and tar content below 1 g/Nm^3 , while the DBD plasma unit produced 45 kg/h of CO. These previous findings offer the possibility of further exploiting the outlet gas through fuel synthesis and power generation and in this work methanol synthesis and SOFC units were implemented. These implementations naturally request further process improvements such as the integration of Na_2CO_3 and ZnO sorbents reactors required for the reduction of the chlorine and sulphur compounds to the very low admissible levels required by SOFC and methanol synthesis.

The process presented in this work represents a significant advancement over previous configurations, as all process streams have been effectively valorised. The resulting process model integrates multiple technologies to enable the following study and optimization for the co-production of electricity and methanol from biomass, thereby maximizing resource efficiency and overall system value.

2. Materials and methods

2.1. Overall process scheme

As described in Fig. 1, in section a), the biogenic waste together with steam enters the HTC reactor, producing hydrochar, which in turn, together with additional steam and fresh sorbent feeds the SEG. This process produces a syngas very rich in hydrogen and with a low CO_2 content, but still containing CH_4 (not useful for methanol synthesis) and contaminants such as particulate matter, CH_4, tar, sulphur and chlorine compounds, and alkali. Ammonia is not considered due to its low concentration in the gas, as the nitrogen content in biomass is significantly lower than that of C, H, O, and the fact that the formation of ammonia from nitrogen and hydrogen requires catalysts and high pressure. Moreover, ammonia formed is converted by the nickel-based catalyst both inside the hot ceramic candles and in the secondary tar reforming stage, in nitrogen and hydrogen. In addition, ammonia is less detrimental and can even act as a fuel in SOFCs. These contaminants are eliminated by the hot gas ceramic candle and HGC unit, and the cleaned syngas is entirely sent to the methanol membrane reactor to produce CH_3OH. The Ca-based sorbent, after capturing the CO_2 in the SEG at 650 °C, along with the char, is sent to the calciner at a higher temperature, where it releases CO_2 and is subsequently recirculated into the SEG. Part of the CO_2, after treatment in the HGC, is sent to section b) together with the exhausted sorbent to a carbonator to produce bricks. Another portion of CO_2 instead after temporary storage, is sent in section c), where it is processed in a plasma reactor assisted by a membrane system. This system converts CO_2 into CO and O_2, which are contextually separated, thus achieving both CO_2 valorisation and oxygen recovery. The resulting O_2 stream, although insufficient to meet the total oxygen demand, reduces the amount of externally supplied pure oxygen required for the endothermic calcination process.

The plasma reactor is powered by renewable energy. The right amount of H_2, CO and CO_2, such that the molar ratio $(H_2+CO_2)/(CO+CO_2)$ equals 2, as required for methanol synthesis, is obtained by adding the DBD output to the syngas coming from section a). This mixture is sent to the membrane reactor operating at 240 °C and 27 atm without any unconverted reactants recycle, see section d), as described in paragraph 2.7 and chapter 3. The CH_3OH can be used for biofuel production, and the unreacted flow containing H_2, CO and CO_2, together with the separated H_2O, is directly sent to the SOFC to produce energy.

2.2. Feedstock selection and HTC

First, a selection of biomass wastes with the best prospects as feedstocks for sorption enhanced gasification (SEG) and subsequent CO_2 conversion and oxygen separation (e.g. sorbents, catalysts and membrane) was carried out. As shown in Tables 1 and 2, a large variety of

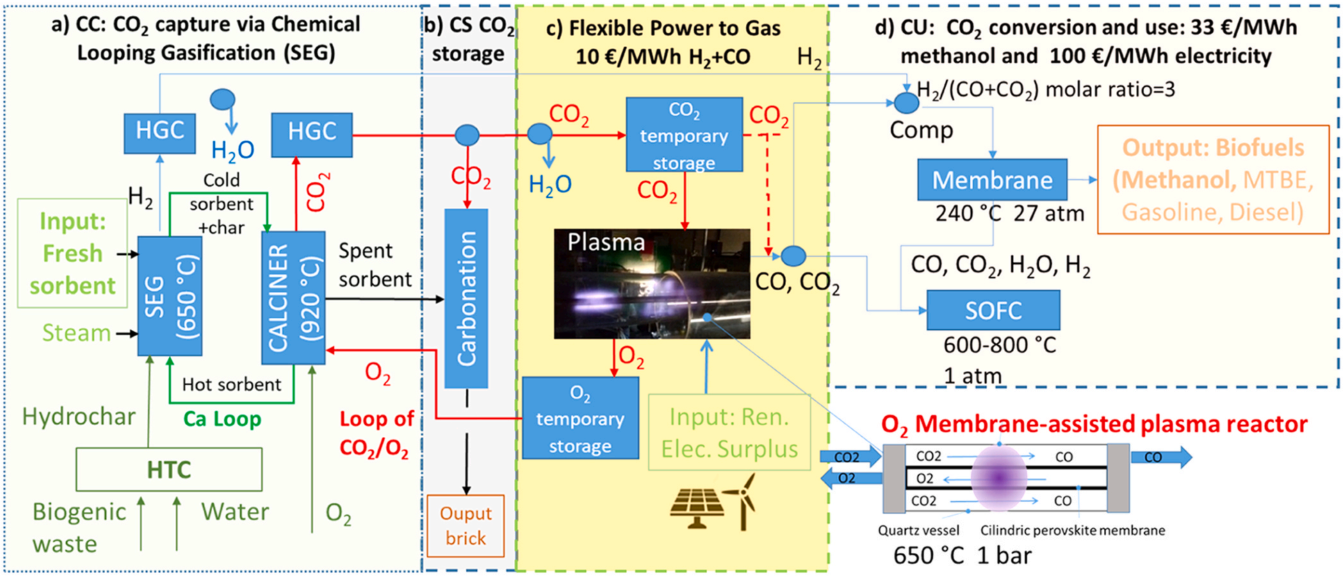

Fig. 1. Overall process scheme.

Table 1
Biogenic residues cost.

Biomass Category	Roadside Price (€/t)	Roadside Price + 100 km transport (€/t)	Maximum cost (€/t)
Primary residues from forest	35	37	63
Agricultural residues	28	30	70
Secondary residues from wood industries	35	37	63
Secondary residues utilising agricultural products	55	57	43
Waste from wood (no hazardous)	15	17	83
Digestate (collection)	66	68	98

biomasses have been identified in terms of type, techno-economic feasibility and availability (t/year in EU).

Based on the combination of a diversity of criteria and taking into account partners' databases and results quoted in the literature, residues (singles or mixtures) with higher potential to be used in gasification have been selected.

Such preliminary approach has been subsequently refined by a comprehensive assessment of the physicochemical features of the diverse biomass feedstocks, resulting in the final choice of 10 wastes for direct gasification and 10 residues for which a prior treatment by hydrothermal carbonization could reduce detrimental effects on SEG and CO_2 sorbents and catalysts (see D 2.1 – Feedstock selection and D2.2 Feedstock characterization GICO public deliverables (GICO Project, 2024). After the evaluation of 20 feedstocks, 5 feedstocks were treated with HTC as shown in Fig. 2. The carbon content increases along with the heating value, while the ash content decreases owing to the ash dissolved in water, but energy and water are required; for this reason, e. g. whey has been considered as reaction medium (Kopsch et al., 2024).

2.3. SEG process

SEG was tested at laboratory scale using olivine and dolomite as bed materials, with steam to biomass ratios ranging from 0.5 to 1 and operating temperatures between 600 and 700 °C. Both unpretreated and hydrochar were used as feedstocks Experimental results showed that residual char content was higher when hydrochar was used, but it decreases with increasing S/B ratio. The simultaneous increase of dolomite and steam led to a reduction in both residual char and contaminants, while improving the overall gas yield. At pilot scale, tests were conducted in a rotary drum gasifier at 650°C using olive pomace as feedstock and dolomite fed together with biomass, with a biomass-to-CaO ratio of 1:1. Due to the longer residence time typical of this reactor configuration, a stable syngas composition was achieved. The dry syngas composition was as follows (see D 4.2 – Integrated GICO process test, GICO Project, 2024, Dai and Whitty, 2022; Martínez et al., 2020; Martínez et al., 2022; Parvez et al., 2021):

- $H_2 = 71.2 \pm 0.9$ %
- $CO_2 = 10.5 \pm 1.0$ %
- $CH_4 = 6.6 \pm 1.2$ %
- $CO = 10.2 \pm 0.2$ %
- Light hydrocarbons $= 1.5 \pm 0.1$ %.

In the flue gas, the CO_2 concentration reached 90 %.

2.4. 2.4 Hot gas ceramic candles

In order to apply hot gas ceramic candles filter with Ni catalyst in the gasifier freeboard, laboratory scale tests using Alumina-Silicate ceramic filter candles with commercial catalyst has been performed. The tests show complete removal of particulate matter from the gas, which remains in the reactor and almost complete conversion of tar compounds at 650 °C, even with 20 ppm of H_2S, under a Gas Hourly Space velocity (GHSV) of 3000 h^{-1}. The reduction in GHSV (and the corresponding increase in residence time) led to improved tar conversion at both 20 and 50 ppm of H2S in the syngas. If 20 ppm of H2S is achieved (by using a CaO-based sorbent), a GHSV of 3000h-1 enables 99 % tars conversion (from around 10 g/Nm3 to approximately 100 mg/Nm3), as shown in Fig. 3 (Nacken et al., 2024). Furthermore, in the secondary conditioning system (downstream of the gasifier), a secondary Ni-based catalytic tar reforming reactor is used to ensure further tar conversion, reaching SOFC compatible levels (below 10 ppmv; (Marcantonio et al., 2022)).

Finally, the filter candles, made of alumina, also promote the hydrolysis of COS to H_2S due to the presence of steam in the gasifier atmosphere.

Table 2
Biogenic residues availability.

Biomass category	Biomass type	Quantity (kton/year) [dry basis]	Candidate
Agricultural residues	2211 - Rice straw and husk	3218.15	
	2212 - Cereals straw	167,181.94	◙
	2213 - Oil seed rape straw	18,029.48	◙
	2214 - Maize stover	43,370.94	◙
	2215 – Sugar beet leaves	8044.03	
	2216 - Sunflower straw	12,389.47	◙
	2221 - Residues from vineyards	2818.70	◙
	2222 - Residues from fruit tree	3091.60	
	2223 - Residues from olives tree	5897.37	◙
	2224 - Residues from citrus tree	944.66	
Secondary residues from wood industries	4111 - Sawdust (conifers)	11,302.45	◙
	4112- Sawdust (non-conifers)	1579.12	
	4113 - Other residues (conifers)	21,040.79	◙
	4114-Other residues (non-conifers)	3274.37	
	4121 - Industries ofwood-based panels	3503.86	
	4122 - Residues of further wood processing	16,068.34	◙
	4131 - Bark	5465.81	
	4132 - Black liquor	25,671.72	◙
Secondary residues from industrial processing of agricultural products	4211 - Olive-stones	1187.14	◙
	4213 - Rice husk	630.68	◙
	Empty shells of dried fruit	500 (18)	◙
	4214 - Pressed grapes dregs	484.50	
	4215 - Cereal bran	27,224.78	
Municipal waste	5111 – Bio-waste non-separately collected	54,101.43	◙
	5112 – Bio-waste separately collected	35,662.09	◙ with and without HTC
Waste from wood	5211 - Hazardous post-consumer wood	4044.82	
	5212 – Non-hazardous post-consumer wood	22,373.40	
Digestates from biogas production	Digestate from Biogas production	12,634.60	◙
Livestock manures	Bovine dry matter	139,079	◙
	Swine manure	9247	◙
	Poultry manure	24,892	◙

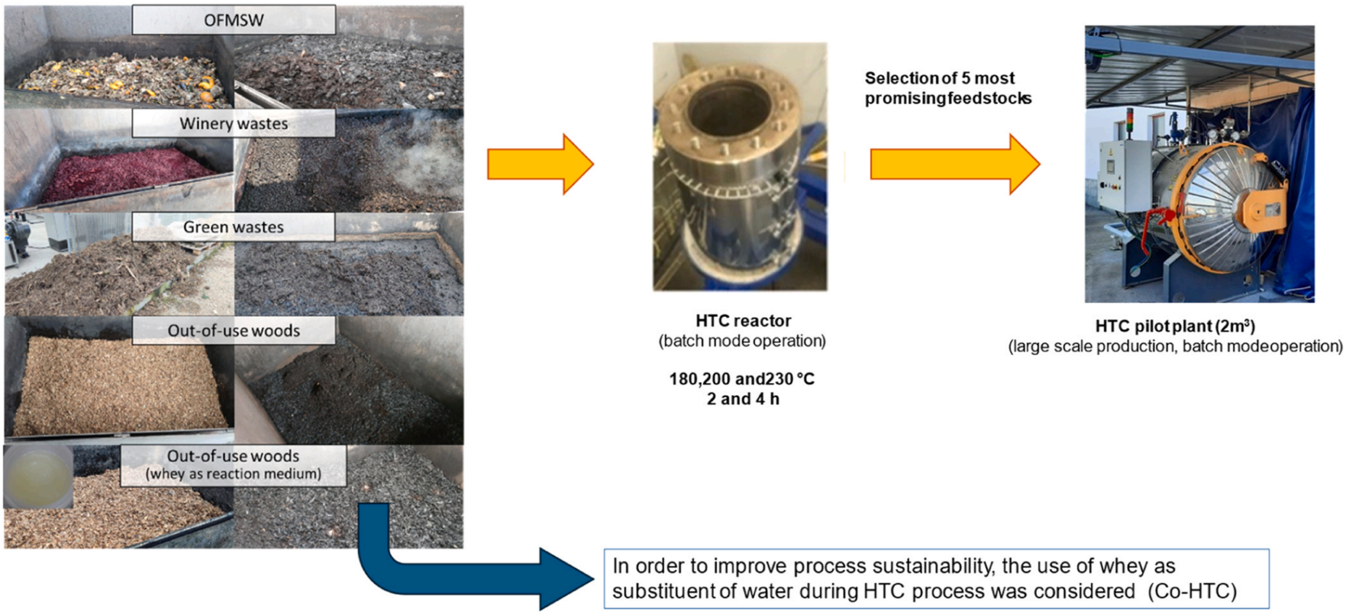

Fig. 2. HTC feedstock treated.

2.5. *Sorbents for removal of detrimental inorganic species and HGC*

Thermodynamic calculations and dedicated experiments on the adsorption behaviour of Alkali, Sulphur and Chlorine compounds at 650 °C within syngas atmosphere and at 900 °C within flue gas atmosphere have been conducted to select the best high temperature sorbents for removal of detrimental inorganic species. Using a mass spectrometer, it was possible to record the H_2, CO_2, H_2S, and HCl concentrations during the sorption experiments simultaneously, although the input concentrations of the gas components (H_2: 72 vol%, CO_2: 1 vol%, H_2S: 60 ppmv and HCl: 40 ppmv) were significantly different. Zinc titanate, a conventional sulfur sorbent, only reduced the H_2S concentration to around 7 ppmv. Conversely, Ba-, Sr-, and Ce-containing sorbents were able to maintain H_2S concentrations below 1 ppmv for several hours, which

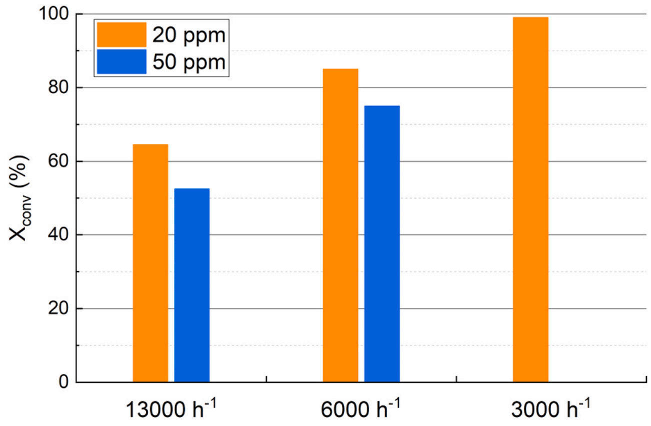

Fig. 3. TAR conversion by hot gas ceramic candles with catalyst.

should be sufficient to prevent poisoning of the nickel catalyst. Six aluminosilicates, Bauxite, Clinoptilolite, Kaolin, Bentonite, Montmorillonite, and Cat litter, proved effective in reducing KCl concentration below 1 ppmv.

Due to economic reasons, it is preferable to operate the HGC unit at approximately 500 °C. For HCl removal at this temperature, Na-based sorbent can be used (e.g. Na_2CO_3 is applied) as well as lime-based sorbents for SO_x removal. A hot gas filter can trap critical alkali compounds like KCl at 500 °C without a dedicated alkali sorption system. However, at temperature above 650 °C, the sorption of HCl is much more complex, as Na-based sorbents may release part of the chlorine as NaCl (GICO project, 2024). At these elevated temperatures, ZnO remains effective in reducing H_2S concentrations below 1 ppmv. Further details can be found in literature as results of the GICO project (Ciro et al., 2023; Hatunoglu et al., 2021; Kopsch et al., 2024, Kopsch et al., 2025). An Additional heat exchanger is required to reheat the gas before entering the catalytic tar cracking reactor, which operates at a minimum of 650 °C for tar reforming with nickel catalysts. Thus, the HGC unit shown in Fig. 4 has been designed and constructed accordingly.

2.6. DBD-membrane for CO2 conversion to CO

The Dielectric discharge barrier membrane reactor concept is shown in Fig. 5.

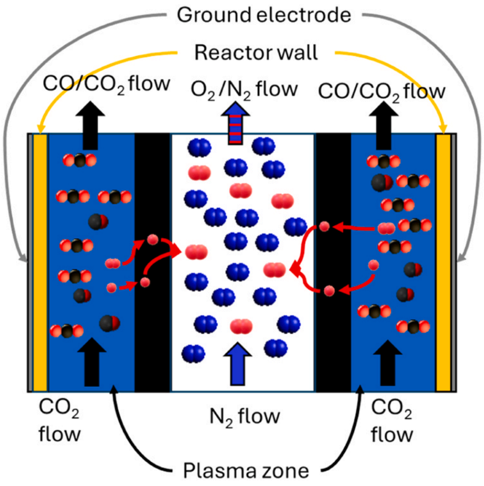

Fig. 5. DBD membrane concept.

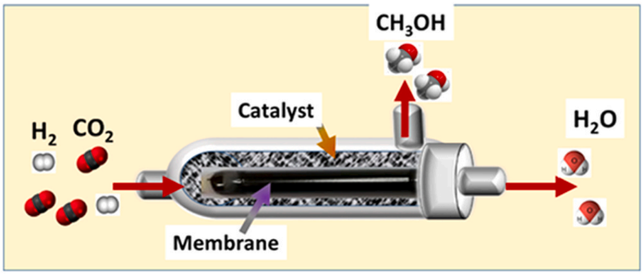

Fig. 6. Methanol reactor concept.

In literature can be found conversion of CO_2 higher than 25 %, but the efficiency decrease (Lisi et al., 2023). Indeed, according to the literature and to the current GICO results, high conversion requires a "high power - low flow" regime and high efficiency "low power - high flow", this is due to two reasons: the first is the back-reaction of CO and O_2 in the plasma, the second is the more filamentary and less homogeneous electric discharge that typically occurs at high power. However, as

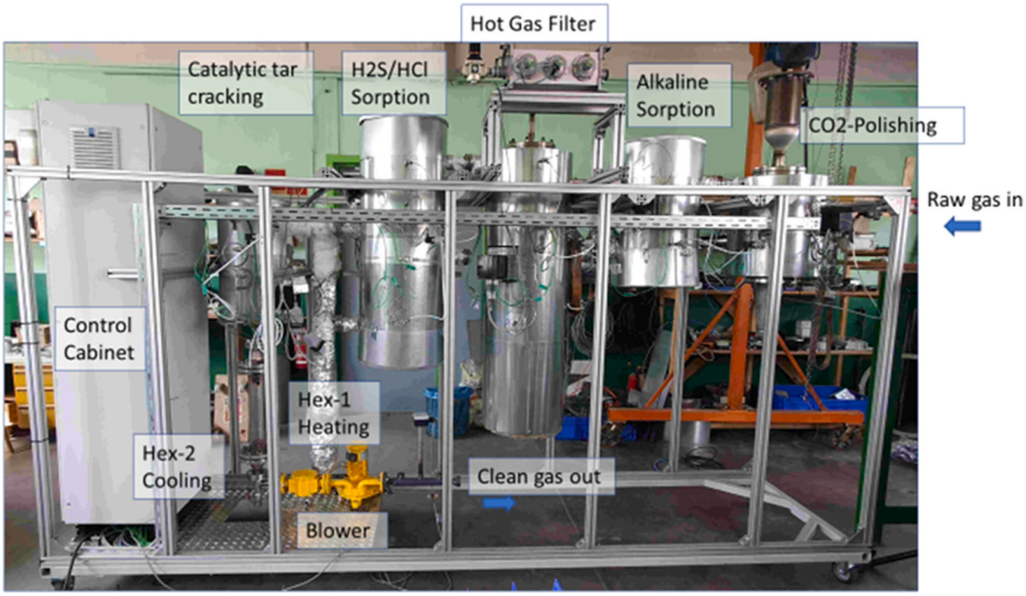

Fig. 4. HGC system.

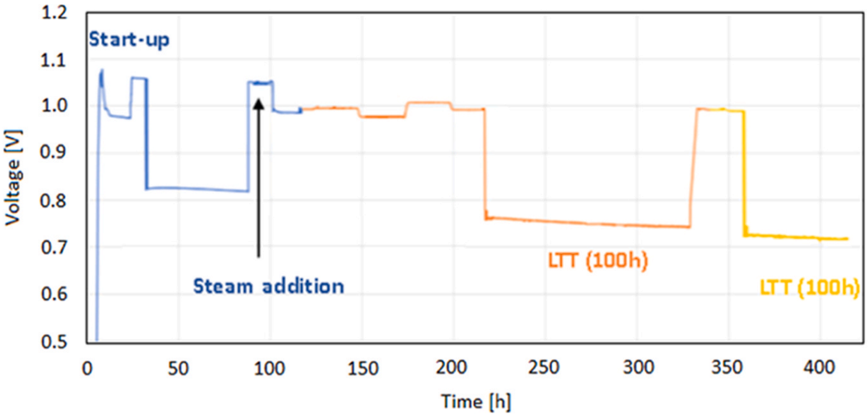

Fig. 7. Voltage decreases from pure H_2 in blue, syngas in orange, syngas+H_2S in yellow.

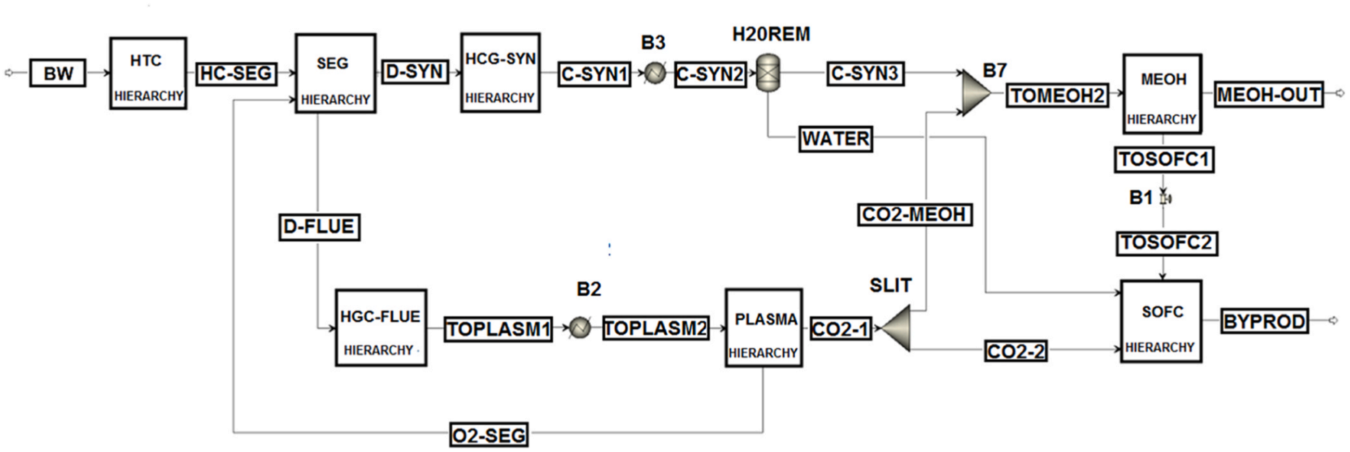

Fig. 8. ASPEN global process scheme.

afterwords explained the conversion of 25 % has been the highest value used for the simulation in this paper, because it is more often found in literature (Cimini et al., 2005; Mei et al., 2015; Mei and Tu, 2017; Lisi et al., 2023; Lisi and Laverdura, 2024) and, also, to be more conservative.

2.7. Carbon membranes for methanol synthesis and SOFC

Methanol synthesis is an equilibrium system (see reaction 1 and 2 in Section 3 - Process modelling). The process is normally carried out at high pressure, typically in the range of 50–100 bar to favour methanol production (Mäyrä and Leiviskä, 2018). In this study, an operating pressure of 27 bar is considered. Such pressure conditions are compatible with those reported in the literature for methanol from syngas, especially for intensified configurations with Pd/Zn- and Cu/Zn/Al-based catalyst (Liao et al., 2017; Previtali et al., 2020).

Following the Le Chatelier's principle, the selective removal of H_2O shift the equilibrium toward methanol formation. Thus, the process is evaluated with a membrane reactor able to remove water, as shown in Fig. 6. The resulting methanol yield ranges from 10 % to 25 % (Richard et al., 2025; Poto et al., 2022; Gallucci and Basile, 2007).

Regarding methanol purity, the model assumes ideal separation downstream of the synthesis reactor, where unreacted gases and by-products are removed to obtain a purified methanol stream. In particular, the reactor is a methanol membrane reactor and so methanol is produced within reagents, and it is separated via the membrane alongside a few reagents that can be easily separated via condensation). While the focus of the present study is on process feasibility and energy integration rather than separation optimization, these assumptions reflect commonly adopted practices in process simulations and are consistent with literature dealing with similar configurations.

To accurately assess the impact of syngas quality on SOFC performance in the process modeling, button cell tests were carried out to determine the voltage drop caused by feeding syngas instead of pure H_2, as well as th effect of 3 ppm of H_2S in the syngas. The observed voltage reduction trends are reported in Fig. 7 (Tamburrano et al., 2024; Wasajja et al., 2022; Steiger et al., 2019).

3. Process modelling

Following the Fig. 1, a process scheme was developed in ASPEN, shown in Fig. 8. As already mentioned, the HTC, SEG, HGC, and DBD plasma reactor were already developed and discussed in a previous work of the authors (Papa et al., 2021; Bartolucci et al., 2023; Marcantonio et al., 2023).

For clarity, a brief description of the main details of the model developed in the previous work in reported below:

- HTC process

The HTC process was tailored within specific guidelines (Ischia and Fiori, 2021), which include:

1) Neglecting reaction kinetics.
2) Simplifying the thermodynamics of HTC reactions using data gleaned from experimental pilot plants, outlined in (Marcantonio

et al., 2023). This involved imposing the proximate and ultimate analyses of the two biomasses while mandating the ultimate analysis of hydrochar as output.
3) Disregarding mass transport and fluid-dynamics phenomena.
4) The Aspen Plus modelling of the HTC process utilized an RYield reactor. The input data for determining the yield of outlet products was derived from experimental data, following the methodology outlined by Akbari et al. (Akbari et al., 2019).
5) HTC process simulations were executed at 200°C (compared to an experimental temperature of 195°C) and 14 bar pressure, with a whey to biomass ratio of 4 (Kopsch et al., 2024; Amado-Fierro et al., 2023, 2025).

- SEG process

  The SEG process was based on principles of mass-energy conservation and chemical equilibrium. The key assumptions made are as follows:
  1) The process operates under steady-state and isothermal conditions (Ye, et al., 2009).
  2) Volatile products primarily comprise H_2, CO, CO_2, CH_4, and H_2O (Sadaka, et al., 2002).
  3) The char is assumed to be 100 % carbon (Demirbaş, 2001).
  4) All gases exhibit ideal behaviour.
  5) Pressure drops and heat losses are neglected.

The gasifier was modelled as a Gibbs reactor using restricted chemical equilibrium to reflect the actual experimental data. According to the minimization of Gibbs free energy, this approach allows to change the equilibrium temperature for each reaction to match the experimental results. The temperature of the gasification process was fixed at 650 °C (Marcantonio et al., 2023). Calcium oxide (CaO) is introduced into the gasifier for in-situ CO_2 adsorption. Steam serves as the oxidizing agent, with a steam to biomass (S/B) ratio of 0.6. The CaO to biomass ratio is maintained at 2 (Li et al., 2020). Subsequently, the $CaCO_3$ exiting the gasifier undergoes regeneration in the calcination reactor. Operating at 950°C, the calcination reactor is modelled as a Gibbs reactor. The reactions taking place within the gasifier and the calcinator are detailed in (Marcantonio et al., 2023).

- DBD plasma reactor

  The DBD plasma reactor was simulated using a stochiometric reactor and two separator blocks. Although literature reports higher fractional conversion of CO_2, the value was set at about 25 % according in particular to (Cimini et al., 2005; Mei et al., 2015; Mei and Tu, 2017; Gico Project, 2025). The two separator blocks are used for: 1) O_2 separation, for which an ideal separation efficiency of 100 % is assumed; 2) splitting CO and CO_2 streams.

- HGC process

The proposed HGC process was composed of: 1) In-bed use of calcined dolomite; 2) Catalytic filter candles in the freeboard of gasifier; 3) Adsorption of H_2S at 450 °C using ZnO as sorbent.

Assuming that by-products from reactions with calcined dolomite are negligible, conversion rates for the three step were taken from literature (Ischia and Fiori, 2021) and used in the simulation. These values comes from experimental data in the temperature range 750–850 °C and atmospheric pressure.

In the present paper, the modelling of methanol synthesis and SOFC system are added to the previously developed sections. The methanol synthesis was simulated as a RGibbs reactor at 240 °C and 27 bar, imposing the reactions:

$$CO + 2H_2 \rightarrow CH_3OH \tag{1}$$

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \tag{2}$$

The SOFC system was modelled as in a previous paper by the authors, via a Gibbs reactor to simulate the anode and a separator for the cathode (Marcantonio et al., 2020b).

4. Results and discussion

Considering input hydrochar stream of 180 kg/h to the SEG section (LHV equals to 22.8 MJ/kg), the hydrogen production achieves approximately 118 $g_{H2}/kg_{Hydrochar}$ (21.2 kg/h of H_2 in the clean syngas after the HGC section). This H_2 yield corresponds to an energy efficiency η (Cold Gas efficiency in hydrogen) of 62 % according to the following equation:

$$\eta = \frac{\frac{kg_{H_2}}{kg_{biomass}} \times LHV_{H_2}}{LHV_{hydrochar}} \tag{3}$$

Of course, when considering the input energy other than biomass of the HTC, SEG and HGC processes, the overall efficiency is lower. However, the energy inputs of each process are mainly constituted of thermal energy that can be provided by thermal energy outputs of other units via a global optimization of the mass and energy flows. This optimization will be addressed in a forthcoming paper, along with detailed mass and energy balance, because is foreseen for the end of the GICO project. Anyway, the simulated yields obtained are very promising.

Indeed, Table 3 summarizes the H_2 yields reported in the literature. The experimental H_2 yield obtained in this study—within a steam-to-biomass ratio range of 0.17–1.7 and a temperature range of 630–1220 °C—is comparable to or even higher than those reported in previous works. This is particularly significant given that many of the literature results were achieved under more favorable conditions, such

Table 3
Comparison of H_2 yields from different biomass conversion processes.

Type of gasification	Feedstock	Gasifier	S/B	Temperature (°C)	H_2 yield (gH_2/kg biomass)	Ref.
Steam gasification	Wood sawdust		0.17–0.51	630–830	98–101	(Abuadala and Dincer, 2010)
Steam gasification	Palm oil	Fixed bed	0.6	800	121	(Li et al., 2009)
Steam gasification	Mixed sawdust		0.5–1.5	730–1200	80–130	(Li et al., 2020)
Steam gasification	Pellet wood	Fluidised bed	1.7	850	128	(Turn et al., 1998)
Steam gasification	Miscanthus X giganteus	Fluidised bed	1	800	73	(Michel et al., 2011)
Pyrolysis/ reforming	Pine wood/ pyrolysis volatiles	Spouted bed/fluidised bed	1.7	500/600	110	(Arregi et al., 2016)
Bio-oil reforming	Raw bio-oil	Fluidised bed	10	700	102	(Kan et al., 2010)

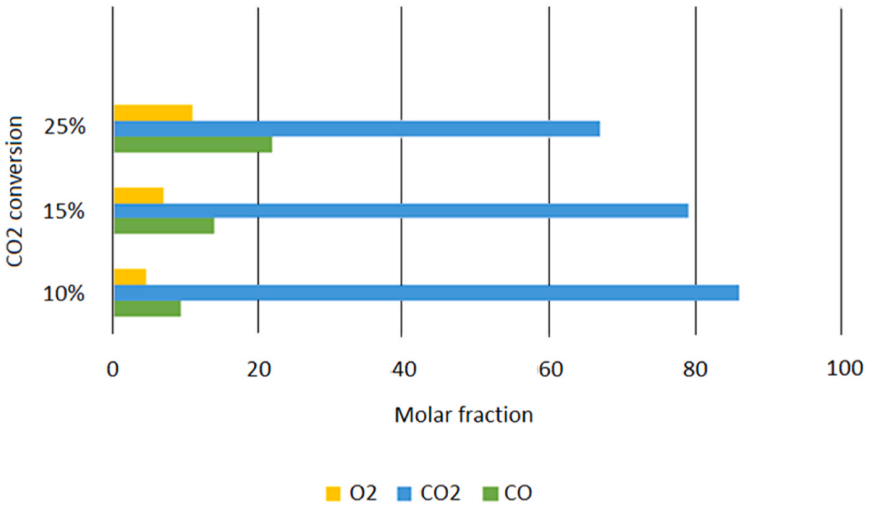

Fig. 9. CO_2, CO and O_2 trend out of stoichiometric plasma reactor for CO_2 conversion of 10 %, 15 % and 25 %.

as higher steam ratios, higher temperatures, or cleaner feedstocks. In contrast, the present system demonstrates similar performance under less severe operating conditions and with more challenging feedstock, highlighting its robustness and potential.

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The DBD reactor splits the inlet stream into CO and O_2, according to reaction (4):

$$CO_2 \rightarrow CO + 0.5O_2 \tag{4}$$

As previously mentioned, the highest fractional conversion of CO_2 considered for a DBD reactor is about 25 % and this or lower values were set in the simulation. Fig. 9 shows the resulting molar fraction of CO and O_2 for a CO_2 conversion of 10 %, 15 % and 25 %.

Assuming a 25 % fractional conversion of CO_2 and an ideal 100 % efficiency for the membrane separator that splits O_2 and CO, it is possible to obtain 46 kg/h of CO, which can be utilized for other applications, such as methanol synthesis. However, this value is an overestimation due to the assumption of ideal membrane behavior and the use of a thermodynamic equilibrium model. In practice, experimental data show less conversion efficiency due to CO reconversion to CO_2 and residual oxygen depending on flow and pressure differences, as well as membrane sealing leakage.

Finally, very promising yields per kg of biogenic waste, and therefore high conversion efficiencies, are obtained:

- Cold gas efficiency of the synthetic fuel production equals to 28 % (62 kg/h of methanol, 250 g/kg biomass).
- SOFC electrical and thermal efficiency based on LHV of the input biomass equal to 24 % and 23 %, respectively (292 kW_e and 279 kW_{th} from SOFC fed by 13 kg/h of H_2 and 42 kg/h of CO)

Thus, a global energy conversion efficiency (based on the LHV of the input feedstock and not taking into account the consumption of DBD) in terms of methanol and electricity production results 52 %, leading to 74 % when considering also the recovery of the heat produced by the SOFC. As for hydrogen yields, the overall efficiencies would be lower than the presented conversion efficiencies when considering the additional energy inputs beyond the biomass feedstock, which are not included in this preliminary analysis. In particular, the system may exhibit a net electricity demand if the DBD electricity consumption exceeds the power generated by the SOFC.

The system shows potential to convert biomass into methanol and electricity while maintaining significant flexibility in terms of feedstock variety and final product selection. HTC allows the treatment of wet biomass meanwhile SEG introduces in situ CO2 capture, thereby increasing the hydrogen content of the syngas and enabling localized CO_2 removal. The captured CO_2, being of high purity, can be either used as a reagent for synthesis or stored permanently. Moreover, the implementation of a DBD plasma system offers a rapid and flexible route to convert this CO_2 into CO, enhancing its chemical utilization.

The coupling of methanol synthesis with SOFC introduces further operational flexibility, allowing the system to adjust methanol and electricity outputs according to feedstock characteristics and process requirements. The electricity produced can be used to meet the internal energy demands of the system, particularly of high-consumption units like the DBD plasma reactor, or, if in surplus, be exported to the grid, contributing to overall energy valorization.

Finally, it is worth noting that the total recoverable heat from the system is likely to be even higher when considering the exothermic heat released during methanol synthesis, as well as the thermal energy recovered from syngas cooling and compression units. This additional heat could be effectively integrated into the overall energy management strategy of the system.

Despite the advantages of the integrated system, it is also important to acknowledge several challenges associated with the combination of multiple technologies. These include the production of wastewater from the HTC process and the high energy demand of the DBD system. Moreover, syngas contaminants and their impact on the stringent operating conditions required for methanol synthesis and SOFC operation must be carefully considered, especially when evaluating the use of different feedstocks.

5. Conclusions

This work presents a preliminary assessment of a novel integrated system for the conversion of biogenic waste into synthetic fuels and power. The system investigates, via laboratory-pilot-scale tests and Aspen Plus simulation, the production of methanol and electricity generation from biogenic waste. Although still preliminary and subject to further refinement and optimization, the results provide valuable insights into the performance of the combined technologies.

In particular, the simulation results indicate that the SEG process can yield up to 118 g/kgBiomass of hydrogen, with a cold gas efficiency of 62 %. Assuming 25 % CO_2 conversion efficiency and ideal gas separation, up to 46 kg/h of CO can be recovered, contributing to methanol synthesis. The system achieves the production of 61 kg/h of methanol (250 g/kgBiomass) with 28 % energy conversion efficiency (based on the lower heating value of the input biomass) and combined SOFC outputs of 292 kWe and 279 kWth, corresponding to 48 % energy generation efficiency of the SOFC unit. The global system energy efficiency reaches 52 %, increasing to 74 % when thermal recovery is included. However, the electricity consumption of auxiliary unit such as the DBD plasma reactor has not been accounted for; this consumption could offset or even exceed the electricity produced by the SOFC, leading to a net electricity requirement for the system instead of a production of electricity.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Data availability

Data will be made available on request.

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