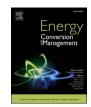


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Carbon-negative hydrogen: Exploring the techno-economic potential of biomass co-gasification with CO₂ capture



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ABSTRACT

The hydrogen economy is receiving increasing attention as a complement to electrification in the global energy transition. Clean hydrogen production is often viewed as a competition between natural gas reforming with CO_2 capture and electrolysis using renewable electricity. However, solid fuel gasification with CO_2 capture presents another viable alternative, especially when considering the potential of biomass to achieve negative CO_2 emissions. This study investigates the techno-economic potential of hydrogen production from large-scale coal/ biomass co-gasification plants with CO_2 capture. With a CO_2 price of 50 ℓ /ton, the benchmark plant using commercially available technologies achieved an attractive hydrogen production cost of 1.78 ℓ /kg, with higher CO_2 prices leading to considerable cost reductions. Advanced configurations employing hot gas clean-up, membrane-assisted water-gas shift, and more efficient gasification with slurry vaporization and a chemical quench reduced the hydrogen production cost to 1.50–1.62 ℓ /kg with up to 100% CO_2 capture. Without contingencies added to the pre-commercial technologies, the lowest cost reduces to 1.43 ℓ /kg. It was also possible to recover waste heat in the form of hot water at 120 °C for district heating, potentially unlocking further cost reductions to 1.24 ℓ /kg. In conclusion, gasification of locally available solid fuels should be seriously considered next to natural gas and electrolysis for supplying the emerging hydrogen economy.

1. Introduction

Increasing attention on the environmental footprint of fuels has led to accelerated research and development efforts in the direction of integrating practical and cost-effective carbon-free energy vectors in current energy systems. H₂ has great potential to decarbonize several industrial sectors, transportation, heating and power networks etc. as reflected in the IEA report [1]. Significant capital investments have taken place to generate "green H₂" from excess renewable energy sources. However, drastic cost reduction in electrolyzer technology must still be achieved in order for this technology to be competitive with respect to current fossil fuel-based generation options. This is further accentuated when cost of handling intermittent H₂ production from solar and wind power is accounted for in the comparison, as highlighted by Cloete et al. [2].

Currently, fossil fuel-based processes are responsible of more than 95% of the total H_2 produced, with Steam Methane Reforming (SMR) from natural gas feedstocks providing 50% of the supply, as detailed by Voldstund et al. [3]. When hydrogen produced from fossil fuels integrate

carbon capture and storage (CCS) technologies, it is referred to as "blue H₂". The high energy penalty imposed by post and pre-combustion capture technologies result in a decreased competitiveness with high CO₂ avoidance costs of around 100 €/ton for pre-combustion capture with MDEA absorption, as demonstrated by Spallina et al. [4]. Khojasteh et al. [5] report that CCS integration increases the cost of H₂ by 40 to 100% relative to the unabated thermochemical conversion process. To address these economic concerns, novel technologies utilizing natural gas for H₂ production with integrated CO₂ capture have been the focus of research recently. Spallina et al. [4] present a detailed assessment of Membrane Assisted Chemical Looping Reforming (MA-CLR) and Fluidized Bed Membrane Reactor (FBMR) based H2 plants yielding a negligible or even negative cost of CO2 avoidance, at equivalent H2 efficiencies of an SMR plant without CO2 capture. Subsequently, Spallina et al. [6] carried out a proof of concept of a CLR dynamically operated packed bed reactor with a promising performance. On the other hand, Cloete et al. [7] present a Membrane Assisted Autothermal Reforming (MA-ATR) concept with a Levelized Cost of Hydrogen (LCOH) of 1.54 €/kg when the membrane is assumed to operate to up to 800 °C. The cost of hydrogen production with CO₂ capture reported by

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| Nomenc | lature | LT | Low Temperature | |
|----------|--|--------------|--|--|
| | | LTS | Low Temperature Shift | |
| Acronyms | | MAWGS | Membrane-Assisted Water-Gas Shift | |
| ASME | American Society of Mechanical Engineers | MA-CLR | Membrane Assisted Chemical Looping Reforming | |
| ASU | Air Separation Unit | MA-ATR | Membrane Assisted Autothermal Reforming | |
| BEC | Bare Erected Cost | NET | Negative Emissions Technology | |
| BECCS | Bioenergy Carbon Capture and Storage | NG | Natural Gas | |
| CA | CO ₂ Avoided | NGCC | Natural Gas Combined Cycle | |
| CC | CO ₂ Captured | OC | Owner's Cost | |
| CCS | Carbon Capture and Storage | PT | Project Contingency | |
| CHH_2 | Combined Heat and Hydrogen | PS | Process Contingency | |
| CLC | Chemical Looping Combustion | PSA | Pressure Swing Adsorption | |
| CLR | Chemical Looping Reforming | TC | Total Contingency | |
| COT | Combustor Outlet Temperature | TIT | Turbine Inlet Temperature | |
| CPU | Cryogenic Purification Unit | TOC | Total Overnight Cost | |
| CSTR | Continuous Stirred Tank Reactor | TOT | Turbine Outlet Temperature | |
| DH | District Heating | SE | Sorption Enhanced | |
| EOS | Equation of State | SEA | Standardized Economic Assessment | |
| EPC | Engineering Procurement & Construction | SMR | Steam Methane Reforming | |
| FBMR | Fluidized Bed Membrane Reactor | S/C | Steam to Carbon | |
| FTR | Fired Tubular Reformer | RSC | Radiant Syngas Cooler | |
| GHG | Greenhouse Gases | WGS | Water Gas Shift | |
| GSR | Gas Switching Reforming | | 1.1 | |
| GT | Gas Turbine | List of Sy | | |
| HTS | High Temperature Shift | η_{H_2} | Hydrogen Efficiency | |
| IEA | International Energy Agency | $\eta_{El.}$ | Electrical Efficiency | |
| IEAGHG | International Energy Agency Greenhouse Gas R&D | C_{CO_2} | CO ₂ capture | |
| | Program | E_{CO_2} | Specific Emissions | |
| LHV | Lower Heating Value | <i>m</i> | Mass flow | |
| LP | Low Pressure | | | |
| | | | | |

Khan et al. [8] for a combination of chemical looping water splitting and combustion resulted in 1.68 \$/kg, significantly below the SMR benchmark with CO₂ capture. Nazir et al. [9] presents a technoeconomic assessment of Gas Switching Reforming (GSR), which overcomes the technological challenges of interconnected fluidized beds at high pressure and operation with membranes at high temperature, reaching CO₂ avoidance costs ranging from 15 to 26.4 \$/ton of CO2. Alternative process intensification pathways such as sorption enhanced steam methane reforming (SE-SMR) evaluated by Yan et al. [10] present LCOH values between 1.90 and 2.53 £/kg. Lee et al. [11] provide a cost estimation of a lab scale sorption enhanced membrane reactor (SEMR) H₂ production plant of 3.04 \$/kg H₂, indicating that such configuration achieves lower CO₂ emission rates relative to membrane and packed bed reactor designs. The different cost estimation bases and assumptions from these studies make it difficult to provide a consistent comparison between these advanced technologies. For perspective, the IEA report [12] indicates a H₂ cost from natural gas with conventional CO₂ capture of 1.73\$/kg (Europe, 2018), presenting a 34% levelized cost increase when CCS technology is integrated.

In parallel to natural gas feedstock plants, gasification technologies for H_2 and power co-production from solid fuels have also been gaining attention, as reflected in the IEAGHG report [13]. Such plants are an evolution of traditional pre-combustion CO₂ capture Integrated Gasification Combined Cycles (IGCC), where the generated syngas stream undergoes a series of water gas shift reactions and CO₂/H₂S removal by means of physical or chemical adsorbents for subsequent firing of a H₂ rich fuel in a Gas Turbine (GT) combustor. By integration of a Pressure Swing Adsorption (PSA) unit downstream the Acid Gas Removal plant, a substantial amount of the original heating value of the solid fuel can be retrieved as highly pure H₂ at pressurized conditions. The remaining PSA off-gas containing unconverted CO, CO₂ slip from the absorption unit and non-recovered H₂ is used for power generation in a Rankine cycle upon combustion with air to raise HP steam, generating enough electricity to satisfy the plant auxiliary demand. A block flow diagram of this reference plant is provided in Fig. 1:

Alternatively to this well-known pathway for H₂ generation from solid fuels, Pd-based membranes investigated experimentally by Basile et al. [14] and originally proposed by Uemiya et al. [15] show great potential to enhance H₂ production efficiencies with low investment costs by reducing the scope of the syngas treating section of the plant, as studied by Jordal et al. [16]. The WGS reaction takes place along a membrane tube with H₂ diffusing across its surface, maximizing CO conversion and yielding a CO₂/H₂O stream in the retentate which, after water knock out, can be boosted to CO₂ delivery pressure with minimal power requirements. Through cryogenic purification of this stream [17], unconverted CO and unrecovered H2 are retrieved and employed as fuel in a power cycle for electricity co-generation. Alongside membrane technology, high temperature contaminant removal from syngas is an opportunity to avoid the energy penalty derived from cooling and heating of syngas prior to the shift, as pointed out by Giuffrida et al. [18], while decoupling CO₂ and H₂S removal, resulting both in capital investment and auxiliary power reductions relative to low temperature syngas treating options [19].

A particularly attractive aspect of gasification is the possibility to use biomass feedstock for achieving negative CO_2 emissions, critical for reaching net-zero emission targets in a cost-effective manner. However, biomass gasification is challenging due to aggressive molten slag (Higman [20]), tar formation at low temperatures (Sikarwar et al. [21]) and potential biomass supply issues in the volumes required for a large-scale hydrogen economy. Depending of the bioenergy source, the assumption of negative emissions with CO_2 capture technologies (BECCS) can be questionable, as pointed out by Fajardy et al. [22], due to emissions associated to cultivation, harvesting, transportation and processing. Cogasification of a limited fraction of biomass with coal mitigates these

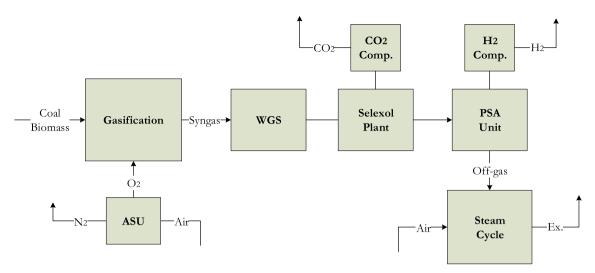


Fig. 1. Simplified block flow diagram of reference H₂ production plant. A detailed process flowsheet is provided in the Supplementary Material file.

challenges, while preserving the potential for negative emissions. Hetland et al. [23] highlight the advantages of co-firing biomass with coal in electricity generation plants with CCS. This is the route investigated in the current study, which presents a techno-economic assessment of a negative emissions technology (NET) for H₂ production employing an enhanced gasification system with a coal-biomass blend as feedstock, incorporating advanced technologies such as Pd-based membrane assisted water gas shift (MAWGS) reactors and hot gas clean up (HGCU) for syngas treating, conceptually represented in Fig. 2. This novel plant is benchmarked against the reference H₂ plant utilizing a GE gasifier with Selexol absorption for syngas treating and a PSA unit for H₂ purification, similar to the one presented in the IEAGHG report [13].

In the next section, a technological and modelling outline of the novel elements is presented, followed by detailed descriptions of the reference and advanced H_2 plant models. The energy and environmental metrics are defined, and the economic analysis methodology is presented. Subsequently, the model results for the different simulation cases are detailed, and the hydrogen production costs for the different configurations are reported. Sensitivity analyses to key process and economic assumptions are performed, to provide a holistic overview. Finally, the main conclusions from the study are presented, highlighting the principal impacts of the study in terms of the concept's

competitiveness and development challenges in the context of blue hydrogen production.

2. Methodology

In this section an outline of the process configurations for the reference and novel concepts is presented. Subsequently, the performance metrics in terms of energy, CO_2 emissions and economic analysis are outlined. The plants are modelled with the software Unisim Design R451 using Peng Robinson EOS for property calculation and ASME tables for steam. The Selexol unit in the reference H₂ plant is modelled using the thermodynamic parameters taken from Kapetaki et al. [24]. The MAWGS model is coded in Scilab using an in-house thermodynamic database, Patitug, for property estimation. The membrane reactor model is coupled to the stationary process flowsheet by means of a CAPE-OPEN unit operation. The energy balance relative error between the Scilab and Unisim platforms was below 0.1%.

2.1 Reference H₂ Plant

The reference plant model (Ref.) is developed based on the guidelines provided in an IEAGHG report [13], using a GE gasifier with an operating temperature of 1350 °C, and the maximum pressure indicated by Higman [20] (80 bar). A 95%mol O₂ stream is provided by a

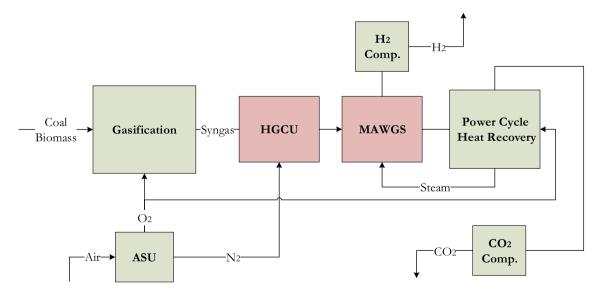


Fig. 2. Simplified block flow diagram of the advanced H₂ plant. A detailed process flowsheet is provided in the Supplementary Material file.

standalone low-pressure ASU consisting of a pumped liquid oxygen cycle, to avoid costly O₂ compressors and associated safety risks [25], to oxidize a blend of 30%w. of wood biomass [26] and Douglas Premium coal [27]. The feed slurry is assumed to be pre-heated to 250 °C with a resulting CGE of approximately 73%. A radiant syngas cooler (RSC) [28] is employed to generate HP superheated steam for syngas cooling to around 900 °C. A water quench achieves further syngas temperature reduction before it enters a saturator (modelled with three equilibrium stages) for fly ash and contaminant removal (NH₃, chlorides etc.). The resulting steam to carbon ratio is approximately 3 prior to a sour shift, composed of two intercooled adiabatic reactors operating at around 200-300 °C, achieving a final CO conversion of approximately 98%. The shifted syngas is routed to a Selexol plant after heat recovery to selectively remove H_2S and CO_2 . Given the high partial pressure of the latter species, a removal efficiency of around 95% is reached. Emissions originate from unconverted CO, CO₂ slip from the absorption column and a small fraction of CH4 present in the syngas, yielding an overall capture rate of 93%. The H₂ rich syngas is routed to a Pressure Swing Adsorption Unit (PSA) modelled with a recovery correlation presented by Nazir et al. [29], assuming a H₂ purity of 100%. The PSA off gas pressure for adsorbent regeneration is selected in such a way that the heat released upon combustion with an air stream in a dedicated boiler raises sufficient steam (110 bar and 550 °C) to achieve an electrically sufficient scheme upon expansion in a steam turbine, assuming a condenser pressure of 0.048 bar. This resulted in a PSA off-gas pressure of 1.85 bar and 90.5% H₂ recovery. Other steam sources such the RSC and WGS heat recovery units are integrated with the steam cycle to maximize power production. Further modelling assumptions of the plant units as well as a detailed process diagram and corresponding stream summary can be found in the Supplementary Material.

2.1. Advanced H₂ plants

Three advanced plant configurations are investigated: two models are designed assuming that all power consumption must be satisfied with electricity produced internally, and evaluate the trade-offs between different power cycles implemented to accomplish this. The final case assumes that some electricity can be imported outside battery limits, to maximize H_2 yield. A description of the advanced technology elements incorporated in each of these concepts as well as detailed process diagrams and stream summaries, together with further modelling assumptions are provided as Supplementary material.

- 1. Wnet0 ST: An electrically self-sufficient plant that employs a GE gasifier and HGCU treating to deliver syngas to a MAWGS reactor. Power is produced through oxy-combustion of the MAWGS retentate stream with pure O_2 from the ASU to generate additional steam for a steam turbine. Thus, this configuration produces electricity in an equivalent way to the Ref. model, with a significant portion of the steam delivered by an RSC.
- 2. Wnet0 GT: In this process concept, the benefits of a higher CGE from the E-gas gasifier (studied by Gräbner et al. [30]) with a second stage chemical quench instead of an RSC and slurry vaporization (experimentally proven by Aiuchi et al. [31]) are reaped by implementing a more efficient power cycle, i.e. a gas turbine (GT), for electricity generation. Fuel for the GT is obtained through cryogenic purification of the retentate stream, as shown by Jordal et al. [16].
- 3. MaxH₂: This configuration also integrates a two stage E-gas gasifier with slurry vaporization to maximize CGE. The power cycle is omitted and some electricity imports are necessary to satisfy the plant auxiliary power demand, designing the MAWGS to maximize H₂ extraction. The low heating value syngas in the retentate stream is combusted in the same way as in the Wnet0 – ST model, generating steam for the sweep after expansion in a back-pressure turbine, which somewhat reduces the net power required.

Some considerations should be made regarding the design of these advanced H₂ plants. Through the water quench cooling of the syngas coming out of the gasifier (from the RSC of the GE gasifier and the chemical quench for the E-gas depending on the case), the steam to carbon ratio is adjusted to a suitable value of 1.9, to carry out the WGS reaction downstream without catalyst deterioration. Prior to this step the syngas is fed to a hot gas desulphurization unit (HGCU) operating in the range between 400 and 500 °C to remove sulphur species and other contaminants. Clean syngas is subsequently cooled in a HP steam evaporator to approximately 320 °C and converted in a high temperature shift (HTS) stage, reaching around 75% CO conversion (assumed at equilibrium) and resulting in an adiabatic temperature rise to approximately 500 °C. The downstream effluent is further cooled producing HP superheated steam and sent to the MAWGS reactor, achieving overall CO conversion values above 90% and high H₂ recoveries per unit of area due to the large driving force, as the syngas stream is fed at around 70 bar. The MAWGS is operated with a counter-current steam sweep, which maximises both CO conversion (by quenching the exothermic reaction) and H₂ recovery (by decreasing its partial pressure), while obtaining a pressurized product stream, minimizing H₂ compression costs and auxiliary consumption. The inlet temperature of the syngas stream is controlled in each case by the amount of steam generation to limit the maximum temperature in the retentate side of the MAWGS to 500 °C.

Through a techno-economic optimization, it was determined that the permeate pressure which resulted in the lowest LCOH was approximately 15 bar, leading to a H₂ compression train comprising two intercooled stages for a delivery pressure of 60 bar, as suggested in the European Hydrogen Backbone site [32]. By increasing the n° of membrane tubes, larger sweep pressures could be imposed, but the reduction in H₂ compression costs did not outweigh the capital cost increase of the MAWGS reactor. On the other hand, decreasing the permeate pressure beyond the optimal value led to an additional compression stage to avoid a large temperature rise, resulting in higher overall costs. The amount of steam that was employed as sweep in the membrane for the Wnet0 - ST and Wnet0 - GT plant concepts was manipulated to reach a net electrical consumption of zero. Small variations from the optimal pressure for the nearest 500 membrane tubes were made to the models to reach the specified temperature approaches in the heat recovery network, which had small impacts in H₂ compression costs comparatively. Heat rejection for all cases was simulated with a cooling water tower (approximated to 3 equilibriums stages) to reach a cooling water temperature of 20 $^\circ$ C, and a process stream temperature after heat rejection of 25 °C.

For the Wnet0 - ST and MaxH₂ cases, the retentate outlet, with a small fraction of the heating value from the original syngas is combusted with pure O₂ produced by the ASU. Combustible species consist of unconverted CO, unrecovered H₂ and the CH₄ produced in the gasifier. Since their concentration can be low due to the large heating value extraction in the form of H2 upstream, in this work it is considered that oxidation can be enhanced by means of a metallic oxygen carrier (e.g., Cu/CuO or Ni/NiO) in a fluidized bed, to ensure complete conversion of the reactants. After heat recovery and cooling to ambient temperatures, traces of water which have not condensed in a knock-out vessel are removed in a dehydration unit resulting in a CO₂ stream with a purity close to 97%mol (the remaining consists of N2 and Ar from the ASU oxidant streams). Condensed water with some dissolved CO₂ is pumped, reheated and recycled to the syngas quench to achieve 100% CO2 capture. The outlet gas stream is boosted above supercritical pressure, and further pumped to 150 bar for transport and storage.

On the other hand, for the Wnet0 – GT case requiring a suitable fuel for a GT, the retentate stream oxy-combustion is not carried out: after heat recovery, water knock out and dehydration, the stream is purified in a double-flash cryogenic purification unit (CPU), similar to the one presented by Campanari et al. [17] and implemented by Jordal et al. [16]. The purge stream contains most of the combustible species and is sent to the GT combustor, leading to some unavoidable emissions originating from the CO, CH_4 and CO_2 present in this stream. The resulting CO_2 purity values were consistent with the guidelines recommended for transport and storage [33], although small fractions of CO, CH_4 and H_2 were present due to the lack of complete separation in the CPU, as opposed to the previous cases where the only source of impurities are inert N₂ and Ar from the oxidant streams delivered by the ASU.

2.2. Plant performance indicators

In this section, the metrics in terms of energy, environmental and economic performance are defined. The H_2 production plant concept can be schematically represented as shown in Fig. 3:

2.2.1. Energy and environmental

The energy performance of the plant is defined attending to the total fuel heating value input, the heating value of the H₂ product and the net electricity demand/production of the plant (\dot{W}_{net}) . In this work, the same efficiency weight is given to electrical and hydrogen efficiencies, based on the assumption that hydrogen prices will be similar to electricity prices in a future hydrogen economy. The hydrogen (η_{H_2}) , electrical (η_{El}) , and overall net (η_{net}) efficiencies are defined as shown in Eqs. (1)–(3), respectively:

$$\eta_{H_2} = \frac{\dot{m}_{H_2} LH V_{H_2}}{\dot{m}_{coal} LH V_{coal} + \dot{m}_{bio} LH V_{bio}} \tag{1}$$

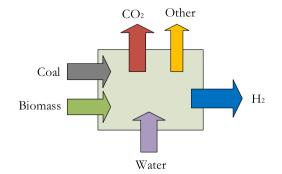
$$\eta_{El} = \frac{\dot{W}_{net}}{\dot{m}_{coal}LHV_{coal} + \dot{m}_{bio}LHV_{bio}}$$
(2)

$$\eta_{net} = \eta_{H_2} + \eta_{El} \tag{3}$$

 CO_2 capture(C_{CO_2}) is defined as the ratio between mass flow from the CO_2 pump discharge and the total CO_2 generated in the plant, both from biomass and coal origin (Eq. (4)). On the other hand, the net CO_2 capture rate $\left(C_{CO_2}\right)$ discounts from the denominator the CO_2 from biomass origin, returning values above 100% and negative specific emissions (Eq. (5)). The fraction of CO_2 originated from biomass is directly determined with the ultimate analysis on a moisture and ash free basis of the coal and biomass and their weight proportion in the blend. Finally, the specific emissions(E_{CO_2}) are determined with the total heating value of the H₂ product (Eq. (6)) and the net emissions flow rate. CO_2 emissions involved in electricity imports are neglected on the assumption that the electricity sector will be largely decarbonized by the time that these negative-emission plants are deployed at scale.

$$C_{CO_2} = \frac{\dot{m}_{CO_2,capt.}^{bio} + \dot{m}_{CO_2,capt.}^{coal}}{\dot{m}_{CO_2,capt.}^{coal} + \dot{m}_{CO_2,capt.}^{coal} + \dot{m}_{CO_2,capt.}^{coal} + \dot{m}_{CO_2,capt.}^{bio}}$$
(4)

$$\dot{C_{CO_2}} = \frac{\dot{m}_{CO_2,capt.}^{bio} + \dot{m}_{CO_2,capt.}^{coal}}{\dot{m}_{CO_2,capt.}^{coal} + \dot{m}_{CO_2,capt.}^{coal}}$$
(5)



$$E_{CO_2} = \frac{\dot{m}_{CO_2,emit.}^{coal} - \dot{m}_{CO_2,capt.}^{bio}}{\dot{m}_{H_2}LHV_{H_2}}$$
(6)

2.2.2. Economic analysis

The economic assessment methodology employed for the capital cost and operating & maintenance costs estimation and cash flow analysis is similar to the approach followed by the authors in a previous work [34]. The objective is to determine the levelized cost of hydrogen (LCOH), the price at which the product must be sold to arrive at a net present value (NPV) of 0 at the end of plant operation. NPV is determined through the discounted annual cash flow rates, which incorporate plant revenues, fixed and variable operating costs, capital costs and emissions taxes.

A Standardized Economic Assessment (SEA) Tool has been specifically developed by the authors in MS Excel with the aim to provide a transparent reproducible methodology for economic evaluation of chemical and energy plants [35]. A comprehensive user guide of this tool is available [36]. The full economic assessments of all the plants evaluated in this study are available for download [37], where details of the capital cost and operational expenditure estimations of each plant are provided. The most important function of the tool is to provide reliable and convenient estimates for the Bare Erected Costs (BEC) of the different plant units. For this purpose, two estimation approaches are followed. For well-known units, equipment lists from the process simulation are elaborated and each unit's cost is assessed by means of correlations from Turton et al. [38]. For less conventional units, a cost capacity estimate is employed similarly to Anantharaman et al. [27]. All unit estimates are adjusted from the source cost basis to a specified target cost basis by means of location factors to account for differences in material and labour costs between regions provided by an IEAGHG report [39], exchange rates for currency and Chemical Engineering Plant Cost Index (CEPCI) [40] for different cost-year estimates. The target basis for the present economic evaluation is defined in Table 1:

As the main source for scaling cost estimation, the costs reported in the NETL report [41] were used, since both gasification technologies investigated in this work are presented in this assessment, to provide a consistent comparison. Given the disparity of cost estimations for gasification-based plant units available in literature [13,27,41,42], a dedicated sensitivity to the gasifier cost is performed. The SEA tool also presents the possibility to benchmark user estimates with reference values adjusted to the target cost basis. As a supplementary cost estimation reference the IEAGHG report [13] presents a detailed description and economic parameters for the evaluation of a GE gasification-based plant for H_2 production.

To reach the Total Overnight Costs (TOC) of the evaluated plant, the methodology described in Table 2 was applied. Engineering Procurement and Construction costs were taken to be 10%, within the range suggested by Roussanaly et al. [43]. The project contingency (PC) and owner's costs (OC) where taken to add up to 35%, slightly higher than the value assumed in previous studies by the authors (Szima et al. [34]). This high value relative to the recommendations given by Anantharaman et al. [27] is considered to reflect uncertainties related to construction of plants involving gasification technologies. Process contingencies are accounted for to reflect a relatively lower degree of technological maturity of specific units with respect to commercial and well-known processes. For the advanced plants, both the HGCU and MAWGS units assume a PC of 30%, whereas for the unconventional steam-power cycle, using fluidized boilers to carry out oxy-combustion, a 10% PC was taken. Furthermore, the advanced features considered for

Table 1Target basis for economic assessment.

| Location | Western Europe | | |
|-----------------|----------------|--|--|
| Cost Year Basis | 2018 | | |
| Currency | e | | |

Fig. 3. Schematic representation of the H₂ plants.

Table 2

Cost estimation methodology for the plant TOC.

| Component | Definition | |
|---|---|--|
| Bare Erected Cost (BEC) | Sum of all BEC costs of the units adjusted to the target basis in Table 1 | |
| Engineering Procurement and Construction (EPC) | 10% of BEC | |
| Process Contingency (PC) | 0-30% of BEC | |
| Project Contingency (PT) | 20% of (BEC + PS) | |
| Total Plant Costs (TPC) | BEC + EPC + PS + PT | |
| Owners Costs (OC) | 15% of TPC | |
| Total Overnight Costs (TOC) | TPC + OC | |

the E-gas gasification (high operating pressure and slurry vaporization), justify a 10% PC applied to this unit. Finally, for the Wnet0 – GT plant, a PC of 10% was applied to the CO_2 CPU.

The cash flow analysis methodology assumes a baseload H_2 production plant with the economic evaluation baseline presented in Table 3. A dedicated sensitivity to the capacity factor and discount rate was carried out, the latter being a critical variable in the economic metric for competitiveness of capital-intensive low-carbon plants, as highlighted by Hirth et al. [44].

The main assumptions for fixed and variable operating and maintenance costs are summarized in Table 4. Sensitivities of the LCOH to the fuel costs, CO_2 tax, electricity price, and membrane costs are presented in the following sections. CO_2 tax levels are of particular interest in this study since, for a given biomass price, the income resulting from negative emissions can at a certain CO_2 price outweigh the higher costs of biomass relative to coal (on an energy basis).

3. Results

This section presents the H_2 plant model results, with a detailed energy breakdown for the cases considered, followed by the energy & CO_2 environmental metrics defined earlier. Then, the results of the economic analysis is shown, followed by a sensitivity analysis of the LCOH to the key economic assumptions.

3.1. Energy & environmental results

The energy breakdown results of the configurations discussed in the previous section are presented in Table 5; the energy and environmental metrics defined in Section 2.3.1 are also detailed below. These models assume a co-gasification of biomass with coal with a fraction of 30% on a mass basis, with a total heat input of 1254.2 MW, analogously to the plants evaluated in the IEAGHG report [13].

The Ref. model shows the lowest H₂ efficiency (59.3%), due to the large auxiliary consumption incurred upon by the CO₂ compression and Selexol units. This result is around 4%-points above the values reported in the IEAGHG report [13], due to a lower gasifier performance relative to this study and because some electricity is produced in excess, reducing therefore the amount of H₂ product. A reduced capture rate results from CO₂ slip, CH₄ and unconverted CO in the absorption system, which are combusted in a boiler for power generation in a steam cycle. The Wnet0 – ST model integrates advanced elements such as HGCU and MAWGS, considerably decreasing the overall auxiliary consumption, as the CO₂ is obtained at high pressure, leading to a 3.6%-points H₂

 Table 3

 Cash flow analysis assumptions [13]

| Component | Value | | |
|--------------------------|----------|--|--|
| Economic Lifetime | 25 years | | |
| Construction Period | 4 years | | |
| Discount Rate | 8% | | |
| Capacity Factor 1st year | 65% | | |
| Capacity Factor 2nd year | 85% | | |
| | | | |

Table 4

Fixed and variable operating & maintenance cost assumptions.

| Fixed O&M | | | |
|---------------------------------------|-------------------------------|--|--|
| Maintenance | 2.5% of TOC [13] | | |
| Insurance & Taxes | 1.0 % of TOC [13] | | |
| Labour | 60,000 €/p-y [13] | | |
| Variable O&M | | | |
| Cooling water make-up | 0.325 €/m ³ [27] | | |
| Process water costs | 6 €/m ³ [27] | | |
| Selexol make-up | 5000 €/ton [27] | | |
| WGS catalyst | 16,100 \$/m ³ [9] | | |
| Oxygen carrier | 15\$/m ³ [45] | | |
| ZnO cost | 25,230 \$/m ³ [46] | | |
| Ash disposal cost | 9.73 €/m ³ [19] | | |
| Membrane replacement | 6000 €/m ² [47] | | |
| CO ₂ transport and storage | 10 €/ton | | |
| CO ₂ emissions tax | 50 €/ton | | |
| Electricity price | 60 €/MWh | | |
| Fuel Costs | | | |
| Biomass | 100 €/ton (6.1 €/GJ) [13] | | |
| Coal | 2.5 €/GJ [13] | | |

Energy and environmental results.

| Model/Item | Ref. | Wnet0 – ST | Wnet0 – GT | $MaxH_2$ | | | |
|-------------------------------------|-------|------------|------------|----------|--|--|--|
| H ₂ LHV (MW) | 744.2 | 788.2 | 846.8 | 915.6 | | | |
| H ₂ flow (kg/s) | 6.20 | 6.57 | 7.06 | 7.63 | | | |
| Auxiliary Electricity Consumption | | | | | | | |
| ASU (MW) | 58.3 | 62.8 | 42.4 | 51.8 | | | |
| H ₂ compressor (MW) | 0.0 | 17.3 | 18.5 | 17.3 | | | |
| CO ₂ compressor (MW) | 25.3 | 2.8 | 7.9 | 3.0 | | | |
| Pumps (MW) | 3.2 | 2.9 | 1.9 | 2.0 | | | |
| Gasifier aux. (MW) | 3.4 | 3.4 | 3.4 | 3.4 | | | |
| Syngas treating aux. (MW) | 25.3 | 0.7 | 0.7 | 0.7 | | | |
| Heat rejection (MW) | 2.1 | 2.4 | 1.3 | 1.7 | | | |
| Other (MW) | 0.7 | 0.0 | 0.0 | 0.0 | | | |
| Electricity Generators | | | | | | | |
| Gas turbine (MW) | 0.0 | 0.0 | 54.0 | 0.0 | | | |
| Steam turbine (MW) | 119.0 | 93.0 | 22.4 | 29.5 | | | |
| Energy Efficiency | | | | | | | |
| $\eta_{H_2}(\%)$ | 59.3 | 62.9 | 67.5 | 73.0 | | | |
| $\eta_{El}(\%)$ | 0.1 | 0.1 | 0.0 | -4.0 | | | |
| $\eta_{net}(\%)$ | 59.4 | 62.9 | 67.5 | 69.0 | | | |
| CO ₂ Emissions | | | | | | | |
| $C_{CO_2}(\%)$ | 93.8 | 100.0 | 91.7 | 100.0 | | | |
| $C_{CO_2}(\%)$ | 121.0 | 129.0 | 118.3 | 129.0 | | | |
| E_{CO_2} (kg/MWh-H ₂) | -95.7 | -124.9 | -72.3 | -107.8 | | | |

efficiency improvement. Increased ASU size and H_2 compression duty offset to a small extent these power reductions. However, for the Wnet0 -GT model utilizing a more efficient power cycle, the higher CGE of the gasifier leads to a further increase of H_2 efficiency of 4.6%-points. Overall, the auxiliary consumption is reduced because of a smaller ASU, despite the small increase in duty associated to the CO₂ CPU. Nonetheless, a significant reduction in capture ratio due to the purge of the CPU unit, which is employed as GT fuel, is observed. Finally, the MaxH₂ case presents the highest H_2 efficiency (13.7%-points above the Ref. model), albeit at a net power consumption of 4% (relative to the heat input) which must be provided through electricity imports. Given the large CO₂ fraction derived from the biomass fuel, all models present negative specific emissions and net capture rates above 100%.

3.2. Economic assessment results

This section presents the economic results of the base case models,

followed by a sensitivity study of to the main process and economic assumptions. The effect of varying the co-gasified biomass mass fraction is investigated and finally the maximum economic potential of the plant considering district heating (DH) hot water sales is determined.

3.2.1. Base case results

A summary of the base case results in terms of specific capital investment can be seen in Fig. 4. Full details can be accessed in the SEA tool datasheets of each model for detailed values of the unit's cost estimation [37]. Overall, it can be seen that the most capital-intensive items are the ASU and gasification units, which in all cases represent more than 50% of the total capital cost. Notably, the Ref. plant with GE gasification, WGS unit, Selexol plant and PSA purification presents the highest specific capital investment, as a result of the large costs of these units together with the lowest H₂ production efficiency. When integrating advanced elements such as HGCU and MAWGS reactors, with an oxy-boiler for steam generation in Wnet0 -ST case, the cost increase due to H₂ compression is largely outweighed by the cost reductions provided by these items and the somewhat lower cost of the power cycle. The auxiliary power demand is to an important extent curtailed due to the avoidance of the Selexol plant and minimization of the CO₂ compression train, leading to a smaller power cycle. When a more efficient power generation system is implemented (Wnet0 - GT), efficiency benefits from higher CGE in the E-gas gasifier with slurry vaporization are translated into higher H₂ production efficiency and in parallel, cost reductions are achieved both in the ASU (lower O2 consumption as no oxy-firing is implemented) and power cycle (GT). This results in an even lower overall specific capital investment for this case, despite the notable increase in cost of the CO₂ compression section, requiring a CO₂ CPU. Finally, for the MaxH₂ case where the power generation for auxiliary consumption is neglected assuming some electricity imports, the slight increase in the ASU, heat recovery section (oxy-boiler) and larger MAWGS reactor are outweighed by the large H₂ efficiency enhancement. Overall, the cost of the power cycle and heat recovery section (only generating some power with a backpressure steam turbine) is reduced relative to the previous case. Analogously to the Wnet0 - ST case, the cost associated to CO₂ compression is minimized, as a highly pure and pressurized CO₂ stream is obtained after water knock-out from the oxy-combusted retentate stream.

The specific operating & maintenance costs are presented in Fig. 5,

for each of the cases. Notably, the plants with lower CO_2 capture (Ref. & WneO-GT) present lower revenues from negative emissions while the plant MaxH₂ shows a higher variable operating cost due electricity imports. Relative to the Ref. plant, the configurations employing HGCU and MAWGS units show a slightly higher VOM due to the costs associated to membrane and sorbent replacement, as well as somewhat higher process water costs due to a larger H₂ production. FOM contribution to the overall operating costs are smaller for the advanced plants given the lower estimated capital investments.

The LCOH distribution for all the base cases is shown in Fig. 6. As discussed, CO₂ tax creates a revenue for the plant (due to negative CO₂ emissions). Based on these results, the Wnet0 - ST plant can produce H₂ at a cost of 0,15 €/kg (8.6%) cheaper than the Ref. plant. This reduction is extended to 0,22 €/kg (12.2%) when a more efficient gasification and power generation systems are employed (Wnet0 - GT). As observed earlier, LCOH reduction due to CO₂ tax revenues for this plant and the Ref. model is comparatively smaller to the rest, due to lower CO₂ capture. When electricity imports are allowed (MaxH₂), the combination of enhanced H₂ production and lower capital expenditure yield a LCOH which is 0,28 €/kg (15.7%) cheaper than the Ref. plant. For this last case, the notable increase in VOM costs results from electricity imports. The relatively low costs of H₂ achieved for the plants of this study, despite their high capital intensity, arise from the benefits of the economies of scale given the large H₂ production capacities, compared to the smaller natural gas based plants [12].

3.2.2. Sensitivity to economic parameters

Given the methodological and market uncertainties in this assessment, several sensitivity analyses of the LCOH to key assumptions of the evaluation were carried out. These cases, which are presented in Fig. 7, can be classified in three main blocks: operational expenditure (cost of biomass and coal, CO_2 tax, electricity price), capital costs (gasifier cost, membrane costs), and market factors (capacity factor, discount rate).

Even though biomass is more than twice as expensive per unit of energy as coal, the sensitivity to the biomass price is lower than to the coal price because it provides only 22% of the LHV energy input to the plant. However, this flexible fuel plant could still benefit from adjusting the coal/biomass ratio in response to price signals of these two fuels. The MaxH₂ plant shows a moderate sensitivity to the electricity price, but it remains cheaper than the electrically self-sufficient plants even at an

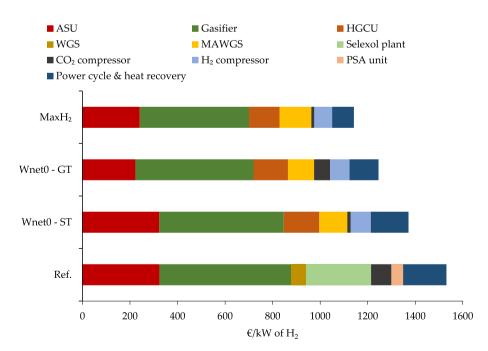
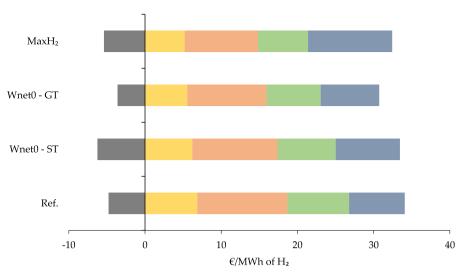


Fig. 4. Specific capital investment for the base case models.



■ FOM ■ Coal ■ Biomass ■ VOM ■ CO₂ tax

Fig. 5. O&M costs per MWh of H₂ for the base case models.

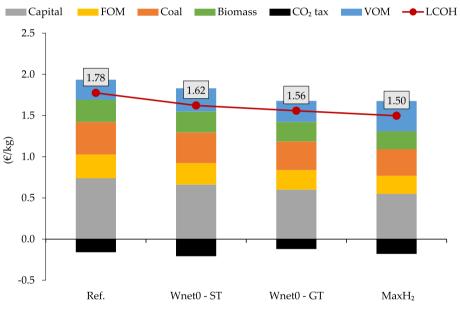


Fig. 6. LCOH results for the base case models.

electricity price of 80 €/MWh. Higher CO₂ prices have a large positive effect on all four plants due to their negative emissions, as will be discussed later. However, plants with capture rates below 100% (Ref. & Wnet0 – GT) present a comparatively lower sensitivity to CO₂ taxes, with the Wnet0 – GT plant resulting in a higher LCOH compared to the Wnet0 – ST plant at CO₂ prices of 100 €/ton, despite employing a more efficient power generation cycle. If electric self-sufficiency is important in an environment of high CO₂ taxes, a cost-effective alternative could be to combust some of the H₂/steam mixture from the membranes in the MaxH₂ case using a power cycle configuration similar to the Wnet0 – GT plant.

The discount rate has a significant effect on these relatively capitalintensive plants. The $MaxH_2$ plant shows the lowest sensitivity due to its lower specific capital cost (Fig. 4). A significant sensitivity to the capacity factor is also observed. Unlike thermal power plants that are increasingly required to balance variable renewables, thermochemical hydrogen plants are likely to operate maximizing the operating hours per year. The gasifier is most likely to dictate the achievable capacity factor in practice.

Being the costliest component in the plant, the relatively uncertain gasifier bare erected cost also has a significant effect on the LCOH. In general, a \pm 30% cost variation in the gasifier BEC has roughly the same effect as a \pm 30% cost variation in the coal price. Given the importance of gasifier technology both for plant capital costs and capacity factors, optimization of this component is key for the success of these negative-emission hydrogen plants. Finally, varying the membrane cost over an order of magnitude had a smaller effect than the \pm 30% gasifier cost variation, although the MaxH₂ case with a greater membrane surface area to maximize H₂ permeation shows a slightly steeper sensitivity. Various uncertain membrane performance metrics can also influence this sensitivity, including lifetime, permeability (influencing the required membrane surface area), and cost recovery (re-using some parts of the spent membranes).

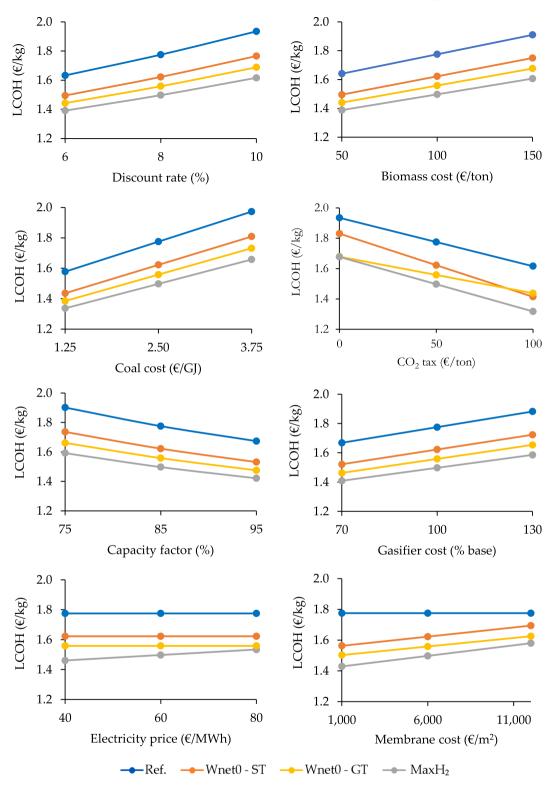


Fig. 7. Sensitivity analysis results.

3.2.3. Effect of biomass fraction

The extra cost of biomass fuel per unit of energy relative to coal can be outweighed by the increased revenue resulting from larger negative CO_2 emissions. For the biomass cost and CO_2 price levels considered in the base case models, the LCOH variation for the MaxH₂ model for biomass fractions ranging from 0 to 40%w. of the feed is shown in Fig. 8, with the relative contributions of each cost item. As shown, higher biomass fractions are economically beneficial under the base assumptions, despite a mild reduction in H₂ production efficiency is due to decreasing CGE and increasing methane fraction (which cannot be converted to H₂ in the MAWGS reactor) when the biomass weight fraction increases. Electricity consumption slightly decreases for a larger biomass content, somewhat reducing the power imports and hence operating costs.

As the coal fraction reaches 100%, a smaller ASU is needed due to the decreased O_2 consumption in the gasifier. On the other hand, the O_2

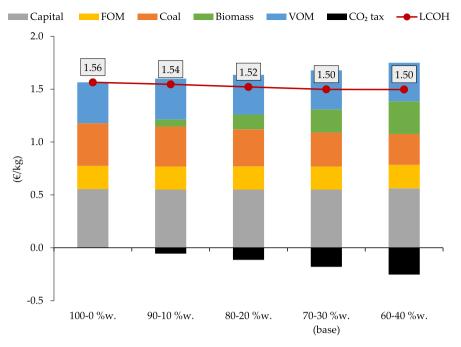


Fig. 8. LCOH of the MaxH₂ plant for different coal-biomass fractions in %w.

demand for the oxy-combustion of the retentate stream begins to increase as the biomass fraction rises due to a larger methane content in the syngas, slightly reducing H₂ efficiencies, which slightly reduces the economic benefit of higher biomass co-gasification fractions. However, these effects are small, and the main determinant of the economic attractiveness of higher biomass fractions is whether the CO₂ price cancels out the price premium of biomass. In Fig. 9, the break-even cost of biomass for different CO₂ prices is represented, as well as the base case assumptions (CO₂ price of 50 ϵ /ton and biomass cost of 100 ϵ /ton). It reflects the price region for biomass and CO₂ at which it is profitable to maximize the fraction of biomass intake to the plant, under the assumption that capital, FOM and other VOM costs remain constant.

If H₂ generated with this technology were to replace the European Union natural gas consumption of 2019 (around 17 EJ, based on the data from BP [48]), this would imply a yearly removal of approximately 0.6 gigatons of CO₂ from the atmosphere – fully 18% of total CO₂ emissions from 2018 [48]. Sufficient biomass from wood crops can be effectively supplied for this purpose based on several forecasts for Europe (de Wit et al. [49] Panoutsou et al. [50]).

3.2.4. Maximum economic potential

The previous assessments of advanced configurations accounted for

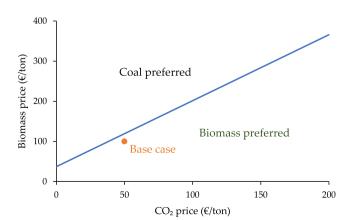


Fig. 9. Break-even biomass and CO₂ prices.

process contingency factors in those plant units which present a low technological maturity. In this section, to unravel the maximum economic potential of MaxH₂ case, an assessment considering no process contingency is presented. On top of that, the MaxH₂ model is modified to account for potential revenues from hot water production for district heating (DH) which can be produced from residual heat resulting in a combined heat and hydrogen generation plant (CHH₂). Such residual heat is available in the condensing enthalpy of the permeate stream, roughly consisting of 50%mol of steam, which cannot be effectively retrieved for useful purposes within the original plant scheme and was inevitably rejected to the ambient. Given the permeate pressure of around 15 bar and the composition, condensation of the steam takes place in a wide temperature, starting at approximately 160 °C. This allows for an efficient heat exchange with pressurize water (16 bar) to around 120 °C (specifications for DH [51]), as illustrated in Fig. 10. The water from district heating at low temperature of 60 °C is pressurized again and sent to the heat recovery unit. 5% of the heat is assumed to be lost due to transmission. A large uncertainty relative to the selling price of hot water exists, and in this study a reasonable value of 30€/MWh is taken (including transportation costs).

Utilizing this remnant heat to generate hot water has a direct effect on the size of the cooling water tower and auxiliary consumption for heat rejection, presenting therefore a somewhat lower capital

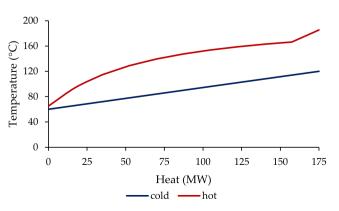


Fig. 10. Heat exchange profile in hot water generator.

investment for this item and smaller make-up water requirements relative to the original case. On the other hand, some extra power consumption is need for liquid pumping to overcome the pressure losses of the system. In Fig. 11, the LCOH breakdown for the cases discussed is presented. Avoiding process contingencies under the assumption that the more uncertain plant elements have reached a sufficient degree of technological maturity has a notable influence in the LCOH of the scheme, achieving a reduction of around 0.07 €/kg of H₂ (4.7% cheaper). Furthermore, when revenues from DH are accounted for, an additional cost reduction of around 0.19 €/kg of H₂ is achieved (17.3% overall reduction), which is even more significant, considering that the heat selling price is half of the electricity price assigned to the power imports for this plant.

4. Summary and conclusions

A detailed techno-economic assessment of four negative emission technologies (NETs) for H_2 production from co-gasification of coal and biomass was carried out in the present study. The main results are summarized below:

- The reference plant employed a GE gasifier, Selexol selective H₂S and CO₂ removal unit, PSA H₂ purification, and a steam power cycle, achieving a levelized cost of hydrogen (LCOH) of 1.78 €/kg and 93.8% CO₂ capture.
- Replacement of cold gas clean-up, Selexol absorption and PSA unit with hot gas clean-up (HGCU) and a membrane-assisted water–gas shift (MAWGS) reactor with subsequent oxycombustion of the retentate reduced the LCOH to 1.62 ℓ/kg and achieved 100% CO₂ capture.
- Introduction of a more efficient E-gas gasifier with slurry vaporization to increase CGE, and power generation with a gas turbine fuelled by the purge stream of the CO₂ cryogenic purification unit (CPU) after the MAWGS, led to a LCOH to 1.56 €/kg while the CO₂ capture decreased to 91.7%.
- When the power cycle was removed, allowing for electricity imports and carrying out oxycombustion of the retentate to raise steam, hydrogen production could be maximized to reach a 1.50 \notin /kg LCOH achieving 100% CO₂ capture.

LCOH sensitivity to a wide range of economic assumptions was evaluated, with CO_2 price being the most influential parameter. All

plants benefitted strongly from higher CO₂ prices, particularly those which achieved 100% CO2 capture. Under the base-case biomass and CO₂ prices of 100 and 50 €/ton, respectively, maximizing the biomass feed fraction was beneficial, decreasing the LCOH by 0.07 €/kg when the feed fraction increased from 0 to 40%w. Break-even biomass and CO2 prices were determined, revealing a great potential for this bio-CCS technology to remove CO₂ from the atmosphere competitively by partially replacing coal as fuel. A final case to determine the maximum economic potential assumes no process contingencies for the less mature process units to reach a LCOH of 1.43 $\ell/kg.$ Furthermore, when revenues from hot water production for district heating are accounted for (generated using the permeate stream condensation enthalpy), the LCOH reduces to 1.24 €/kg (~30% below the reference plant). Such a combined heat and hydrogen plant would be suitable to be deployed close to populated areas given the minimal emission levels attained through advanced gas clean-up systems, water quenching and NOx-free combustion in the oxygen carrier assisted combustor.

Key to the realization of this promising economic performance is the commercialization of high temperature syngas clean up, reliable operation of membrane technology, successful implementation of oxygen carrier-assisted combustion of the retentate, while achieving the solid fuel gasification enhancements (slurry vaporization, complete tar-heavy hydrocarbon conversion) that maximize cold gas efficiency, thereby limiting the partial oxidation losses and by-product formation. However, the relative simplicity of the MaxH₂ plant topology relative to the other cases suggests that this concept would be comparatively easier to build and to operate, provided that these technological hurdles are overcome. Further studies exploring the potential of increasing biomass feed fractions beyond the 30%-level assumed as the base-case are also recommended to unlock further cost reductions.

In conclusion, the NET plants evaluated in this study show great promise for providing a carbon-negative energy vector (H_2) using abundant locally available solid fuels. Even the reference plant presents an economic performance that can be attractive in an environment with high CO₂ and moderate biomass prices. The advanced concepts further improve competitiveness relative to published works evaluating novel H_2 production technologies with CCS from natural gas feedstocks [4,7,9]. The cost-effective negative emissions from these plants can allow the hydrogen economy to play a leading role in achieving net-zero in a timely and economical manner.

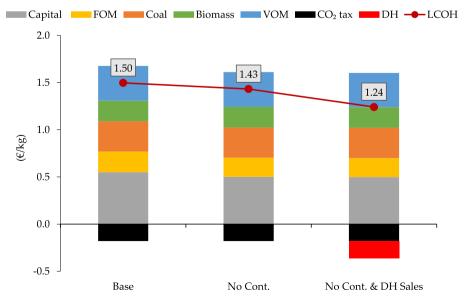


Fig. 11. LCOH for the MaxH₂ plant assuming no process contingency and revenues from DH.

CRediT authorship contribution statement

Carlos Arnaiz del Pozo: Conceptualization, Methodology, Formal analysis, Investigation, Writing - original draft. **Schalk Cloete:** Conceptualization, Methodology, Formal analysis, Writing - review & editing, Funding acquisition. **Ángel Jiménez Álvaro:** Writing - review & editing, Supervision.

Declaration of Competing Interest

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

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Appendix A. Supplementary data

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