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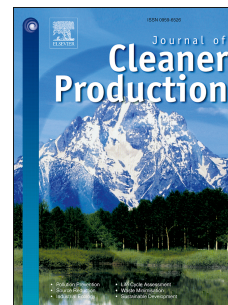
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Credit Author Statement

Rémi Demol: Conceptualization, Methodology, Software, Writing, Visualisation

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Yann Rogaume: Supervision, Funding acquisition

Guillain Mauviel: Conceptualization, Methodology, Writing, Supervision, Funding acquisition

1 **Production of Purified H₂, Heat and Biochar from Wood:**
2 **Techno-Economic and Life Cycle Assessment of Small Scale Units**

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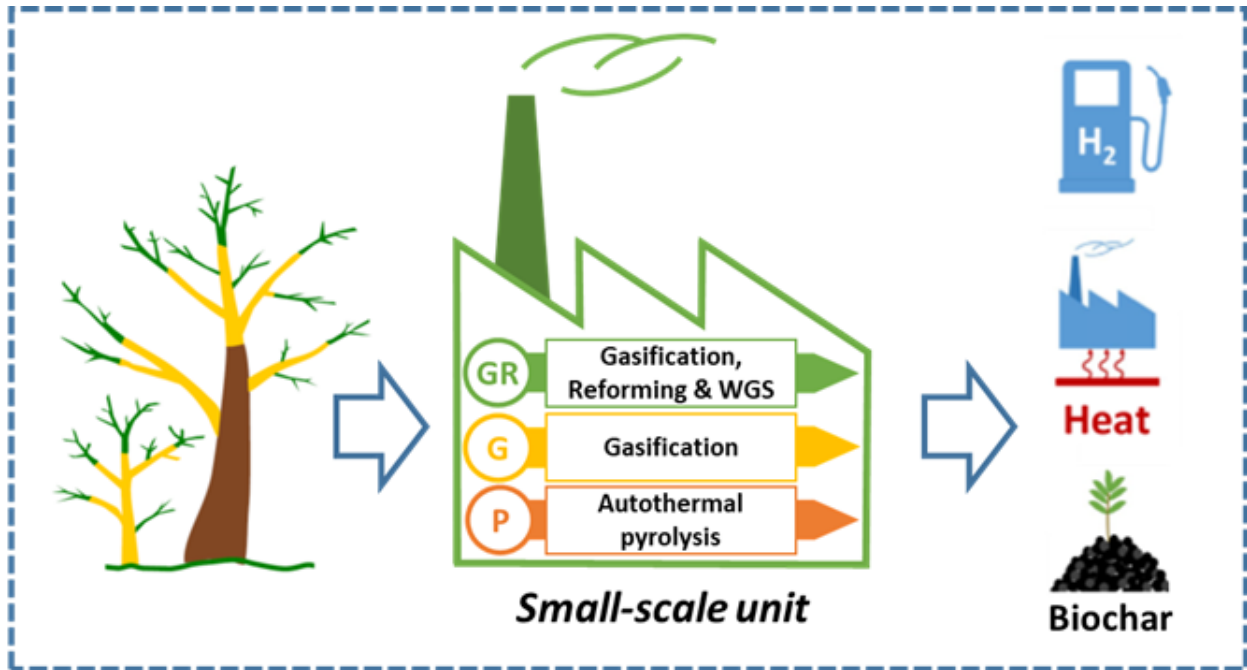
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Techno-Economic Assessment
Life Cycle Assessment



Subsidies required
Environment relevance

Production of Purified H₂, Heat and Biochar from Wood: Techno-Economic and Life Cycle Assessment of Small Scale Units

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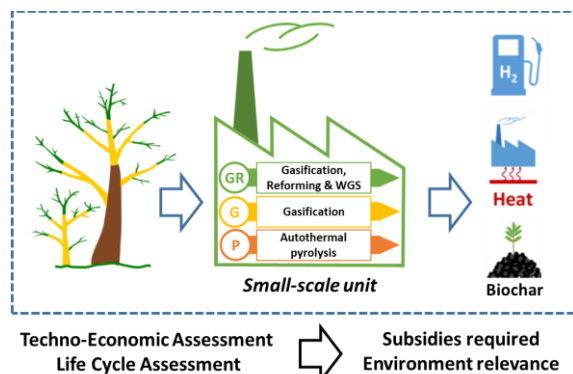
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HIGHLIGHTS

- Gasification of woodchips and reforming to produce hydrogen below 6 €/kg_{H₂}.
- Autothermal pyrolysis for hydrogen production and carbon sequestration.
- Far lower emissions than SMR (up to 15 times less for H₂, 7 times less for heat).
- Cost of subsidies per ton of CO₂ avoided between 0 and 400 €/t_{CO₂ avoided}.

GRAPHICAL ABSTRACT



ABSTRACT

Hydrogen is forecasted to contribute to the energy transition. This study evaluates the production of purified H₂, heat and biochar from woodchips. Three pyrogasification processes are compared: gasification (O₂/H₂O) with or without catalytic reforming and water gas-shift, autothermal pyrolysis. Techno-economic and life cycle assessments were conducted based on detailed mass and energy balances. On the economic front, the historical market prices of hydrogen, heat and biochar are not sufficient to reach profitability. The subsidies required to reach profitability and the avoided CO₂ emissions from reference steam methane reforming (SMR) process were evaluated. It is estimated between 130 and 310€/t_{CO₂ avoided} which can be reduced with higher price of hydrogen. On the environmental front, the pyrogasification processes are more attractive than SMR regarding global warming, ozone depletion layer and fossil fuel consumption potentials. Meanwhile, the acidification potential and the photochemical oxidant formation are higher than SMR.

KEYWORDS

Gasification, Pyrolysis, Hydrogen, Biochar, Reforming.

1. INTRODUCTION

The intergovernmental panel on climate change (IPCC), in his sixth assessment report, sounded once again the alarm on the consequences of the anthropogenic emissions of greenhouse gases [1]. To mitigate global warming, carbon-neutral or even carbon-negative processes in the energy sector could contribute to the reduction of these emissions. As an energy vector, hydrogen presents a growing interest for its ability to decarbonize the industry sector and for future mobility applications. Yet, the production of hydrogen mainly relies nowadays on fossil fuels: natural gas steam reforming (71%) or coal gasification (27%) [2]. Other means of production are investigated: thermochemical water splitting, biochemical processes and especially water electrolysis [3]. In this case, the CO₂ emission problem is transferred to the electricity production. As an example, in France, the electric mix highly nuclear-based contained 34 g_{CO2}/kWh in 2020 [4]. As a result of its low energetic efficiency (40 to 70%), electrolysis produces H₂ that “contains” between 48 and 85 g_{CO2}/kWh (lower if the heat produced is valorized). This is clearly better than H₂ from CH₄ with 357 g_{CO2}/kWh [5] or even higher, 560 g_{CO2}/kWh, considering methane emissions (during methane extraction and transport to consumer) [6], or from coal gasification (339 g_{CO2}/kWh) [5]. In countries where the electric mix is richer in carbon (over 300 g_{CO2}/kWh), electrolytic H₂ is clearly not a good solution to reduce the greenhouse gases emissions [7,8].

Hydrogen can also be produced from other renewable resources such as woody biomass [9,10]. This gas can be produced by thermochemical processes through pyrolysis and gasification [11–13]. In this case, the hydrogen efficiency is around 35 and 50% [7]. These processes are by nature multi-products oriented: the heat produced along the process could be recovered to fulfill the heat requirement of the process or to supply a heat network. Biochar could be produced for soil fertilization and as a carbon sink [14,15] or it could be burned to replace fossil carbon combustion. Considering all these products, the global energy efficiency can reach 70-80% [13].

Several studies investigated the potential of producing electricity of biofuels from biomass gasification [16,17]. Shahabuddin reviewed some techno-economic assessment of biomass and waste gasification to produce hydrogen [18]. They collected levelized costs of H₂ between 2.3 and 5.2 USD/kg_{H2} at 10 MW_{th} feedstock based, whereas it is evaluated between 2.8 and 3.4 USD/kg_{H2} at scale above 250 MW_{th} for biomass. These costs can be competitive with low-carbon production pathways. Table 1 presents previous techno-economic analysis of the literature. The final cost of hydrogen could vary a lot from one study to another depending notably on the size of the unit (Figure 1), but also on the hypothesis on the processes. All these studies do not give the same details on the hypothesis considered. For instance, the chosen

feedstock price has a major impact on the cost of hydrogen. Besides, the hydrogen produced does not always present the purity or pressure.

Table 1: Non-exhaustive list of techno-economic analysis of biomass gasification in literature.

	Technology	Biomass	Biomass throughput	H ₂ Efficiencies & production	H ₂ specification and price	Ref.
Iwasaki, 2003	Pyrolyser, cracker, CO-shift, PSA ^b , engine	Woody biomass	100 t/d 18.6 MW _{LHV}	H ₂ = 47.9% _{HHV} (net) 5.9 t/d	99.99% - 200 bar 4.28 \$ ₂₀₀₃ /kg ^a	[19]
Spath et al., 2005	DFB ^b , reformer, wet scrubber, LO-CAT ^b , ZnO bed, steam reformer, WGS ^b , PSA ^b	Hybrid poplar wood chips	2000 t/d 434 MW _{LHV}	H ₂ = 49.8% _{LHV} (net) 152 t/d	99.9% - 70 bar 1.38 \$ ₂₀₀₂ /kg	[11]
				H ₂ = 55.3% _{LHV} (net) 163 t/d	99.9% - 70 bar 1.24 \$ ₂₀₀₂ /kg	
Lv et al., 2008	Downdraft gasifier (O ₂), CO-shift, PSA ^b , engine	Forest residues	6.4 t/d 1.4 MW _{LHV}	H ₂ = 51.5% _{LHV} (gross) 0.52 t/d ^d	1.69 \$ ₂₀₀₈ /kg ^a	[20]
Parks et al., 2011	First unit, gasifier, reformer, WGS, PSA ^b	Woody biomass	500 t/d 109 MW _{LHV} ^c	H ₂ = 43.8% _{LHV} (gross) 32.4 t/d	99.99% 5.40-7.70 \$ ₂₀₀₉ /kg	[21]
	N th unit.		2000 t/d 434 MW _{LHV} ^c	H ₂ = 45.7% _{LHV} (gross) 135 t/d	99.99% 2.80-3.80 \$ ₂₀₀₉ /kg	
Sara et al., 2016	Indirect FB ^b , catalytic candles, WGS ^b , PSA ^b	Almond shells	4.8 t/d ^d 1 MW	H ₂ = 46-50% _{LHV} ^d 0.033-0.036 t/d	9.5-13 € ₂₀₁₆ /kg ^a	[22]
Sentis et al., 2016	Indirect FB ^b , catalytic candles, WGS ^b , PSA ^b	Almond shells	4.80 t/d ^d 1 MW	H ₂ = 20% _{LHV} ^d Global = 30% _{LHV} 0.14 t/d ^d	6 bar 5.6-7.1 € ₂₀₁₆ /kg ^a	[23]
			48.0 t/d ^d 10 MW	H ₂ = 20% _{LHV} ^d Global = 30% _{LHV} 1.4 t/d ^d	6 bar 2.7-2.9 € ₂₀₁₆ /kg ^a	
Salkuyeh et al., 2018	High-pressure EF ^b oxygen-blown, ASU ^b , LO-CAT, WGS ^b , PSA ^b	Canadian pine wood	5 840 t/d 1200 MW _{LHV} ^d	H ₂ = 54% _{LHV} ^d Global = 56% _{LHV} 454 t/d	3.4 \$ ₂₀₁₈ /kg ^a	[24]
	With carbon capture			H ₂ = 50% _{LHV} ^d Global = 50% _{LHV} 454 t/d	3.5 \$ ₂₀₁₈ /kg ^a	

	Atmospheric indirectly-heated FB, tar reformer, scrubber, LO-CAT, WGS ^b , PSA ^b			H ₂ = 42% _{LHV} ^d Global = 45% _{LHV} 454 t/d	3.1 \$ ₂₀₁₈ /kg ^a
	With carbon capture		7 380 t/d 1500 MW _{LHV} ^d	H ₂ = 41% _{LHV} ^d Global = 41% _{LHV} 454 t/d	3.5 \$ ₂₀₁₈ /kg ^a

^aYear cost assumed.

^bPSA: pressure swing adsorption, DFB: dual fluidized bed, WGS: water gas shift, FB: fluidized bed, ASU: air separation unit, EF: entrained flow.

^cAssuming a LHV of 18.7 MJ/kg.

^dEstimate.

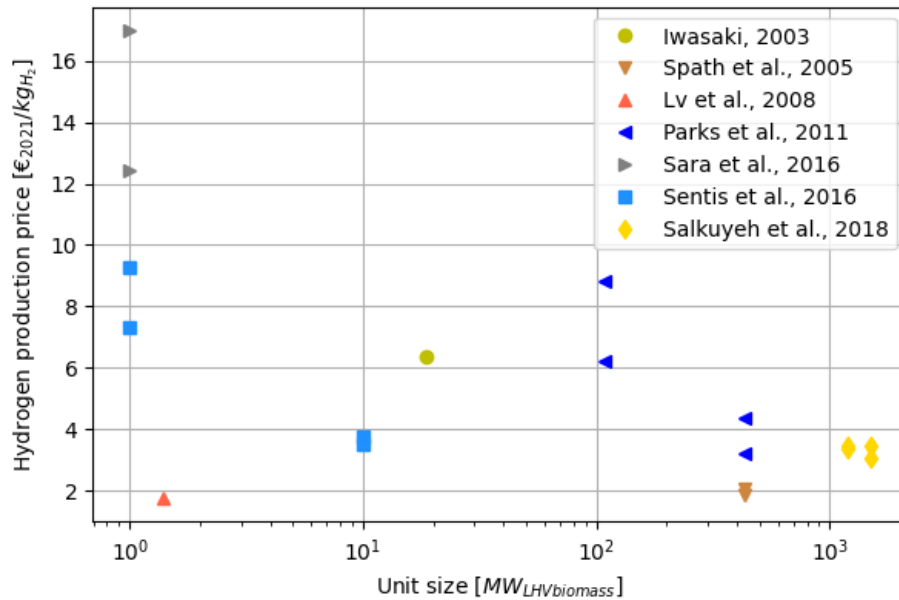


Figure 1: Summary of the hydrogen production cost estimates in the literature as a function of unit size. Note that this is only a simplistic way of comparison, the underlying assumptions as feedstock price are not the same from one study to another.

Spath et al. [11] produced a fundamental work on the economic evaluation of large-scale production of hydrogen (434 MW_{biomass,LHV}). They concluded to a very low price of 1.8-2.1 €₂₀₂₁/kg_{H2}. This price was then reviewed by Park et al. to 3.2-4.4 €₂₀₂₁/kg_{H2}. Salkuyeh et al. on an even larger scale (1200-1500 MW_{biomass,LHV}) evaluated this price at 3.0-3.5 €₂₀₂₁/kg_{H2} without and with CO₂ capture [24]. Such large scale does not really correspond to the availability of the feedstock considered (wood-based). Sentis et al.

evaluated the hydrogen cost on smaller scales (1 to 10 MW) and concluded to 9.3 to 3.5 €₂₀₂₁/kg_{H₂} respectively [23].

The production price of hydrogen should not be the only criteria to evaluate the interest of a process. The environmental impacts generated by such processes can be evaluated through a life cycle assessment (LCA) framework.

Valente et al. [25] reviewed LCA conducted on hydrogen energy production systems. The functional unit frequently chosen is energy or mass-based. Particularly for thermochemical processes, the production of multi-products put the problem of the attribution of the impacts on the various products. The first option consists of the system expansion in which the avoided production of the co-products in the baseline process are considered. This approach requires additional data for the inventories of the baseline process [26]. A second option is to allocate the impacts to the products on a mass, energy, exergy, volume or economic basis. Half of the studies reviewed chose the system expansion, the other half preferred the allocation with a predominance of energy/exergy basis. The most studied impacts are by order of occurrence: the global warming potential (GWP), the acidification potential (AP), the cumulative energy demand (CED), the ozone layer depletion (ODP), the photochemical oxidant formation (POFP), the fossil energy consumption. In less than 20% of the studies reviewed, the energy consumption (nonrenewable), the abiotic depletion, the human toxicity and the land use were also considered. The methods used are IPCC [27] for GWP, VDI [28] or GREET [29] for CED and CML [30] for all other indicators.

As the system definition is crucial in the comparison of various hydrogen production processes, Valente et al. worked on the harmonization of GWP, CED and AP impacts [31–33]. To ensure consistency of impact evaluation, comparison between different hydrogen production systems, the purity and the compression of hydrogen should be at the same level.

Our group has worked previously on the global warming potential of combustion technologies [34] and on gasification processes to produce heat and electricity [35]. In this present study, we investigate the economics and potential impacts of pyrogasification units at small scale to produce hydrogen, heat and biochar. This work is based on a detailed mass and energy balance presented in a previous article [13].

Due to the nature of the feedstock, small gasification units are investigated in this work to use local resources and to minimize the transport distance and the comings and goings of truck near the plant.

The aim of this work is to evaluate the economic conditions to produce H₂ from biomass at relatively small scale and to evaluate the environmental impacts resulting in this production.

The originality of this work relies on: 1) The comparative techno-economic and life cycle assessment of small-scale process of production of purified hydrogen, heat and biochar from a detailed heat and mass balance. 2) A carbon-negative process based on autothermal pyrolysis process is evaluated for the first time. 3) These processes are compared with a common indicator: the ratio between the amount of subsidies required and the CO₂ emission avoided.

2. MATERIAL AND METHODS

2.1. Scenarios investigated

Five scenarios of production of hydrogen from woodchips are investigated in this study (Figure 2). The unit size chosen is 20 MW-woodchips LHV basis to ensure a woodchips feeding with a 50 km-average transport distance in a 27 t-moving floor truck. They correspond to the three processes modeled in detail, under Aspen Plus process modeling software, in a previous paper [13]. The gasification conditions were chosen to maximize the production of hydrogen from wood (steam and oxygen as gasification agent). This study completes the evaluation of these processes with a techno-economic and a comparative life cycle assessment.

The scenarios (Figure 2) and its variations are hereafter denominated as: GR-100, GR, G, P and P-S. A common pretreatment of the woodchips consisted in drying and solid conveying. The GR (gasification reforming) scenario is designed to maximize hydrogen production. It corresponds to a steam-oxygen gasification combined with a partial oxidation (POX) unit and followed by a reformer and water gas shift (WGS) reactors to promote the production of hydrogen. After a wet scrubber, the hydrogen is separated from the syngas by a pressure swing adsorption (PSA H₂), the usual separation technology. Heat for heat network is produced from the cooling of the syngas along the process and from a gas boiler in which the PSA H₂ tail gas is burnt. In addition to the 20 MW case, a scenario with a larger scale of 100 MW (noted as GR-100), is considered in order to study the impact of the scale on the same process.

The G (gasification) process is very similar but without reformer and water gas-shift reactors to avoid costly catalytic reactors and increase the robustness of the process. Because the hydrogen is more diluted in the syngas than in case GR, a two-stage separation process is implemented for its purification. The first step consists of a membrane separation used to concentrate the hydrogen before entering the second step made of a PSA H₂ [13]. This option produced less H₂ but in counterpart more heat than GR.

The P and P-S (pyrolysis) scenarios consist of a pyrolysis running under autothermal conditions by adding a smaller amount of oxygen than in gasification. As a result, the reactor temperature is decreased and biochar is produced [36]. This is a staged process, the solid phase (char) is recovered after pyrolysis and the gas phase produced is further oxidized in a POX unit to promote pyrolysis vapors conversion and hydrogen production. The syngas produced from the POX follows the same treatment as in G scenario. For an economic and environmental comparison, two alternatives are considered: 1) in case P the biochar is sold and not considered as permanently sequestered, 2) in case P-S biochar is permanently sequestered

and not sold. The biochar sequestration may enable the collection of CO₂ credits from other companies, this is considered in case P-S.

In these scenarios, the products are hydrogen, heat and biochar for autothermal pyrolysis options. Heat is recovered along the process and from the gas boiler. It provides the heat required in the process. The heat surplus can be sold as a product in a heat network. Hydrogen is produced at 70 bars and 99.99% purity, a level chosen for industrial applications.

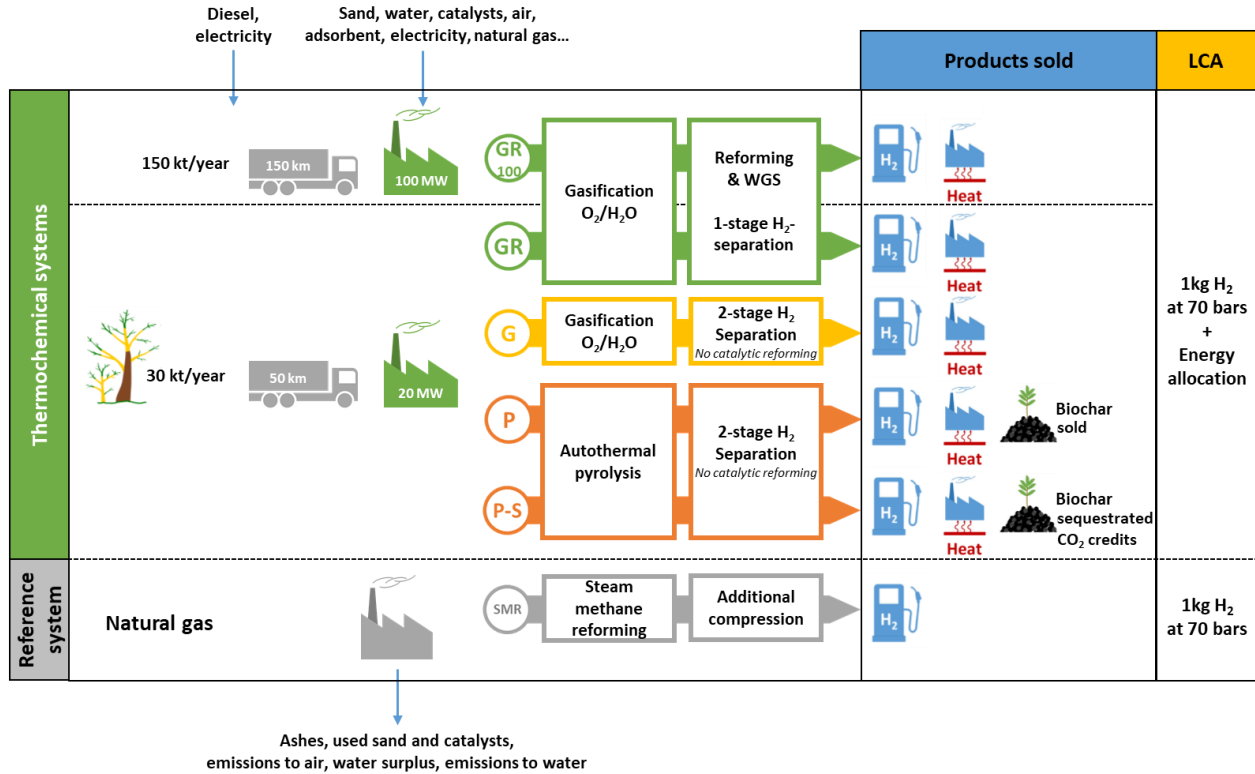


Figure 2: Scenarios investigated and their system boundaries for the life cycle assessment.

2.2. Techno-Economic Model

In order to compare these scenarios on an economic basis, few figures need to be estimated: the fixed capital cost, the manufacturing cost and the revenues from product sales. The plant is assumed to be located in France. In the following, if not specifically mentioned, all prices are given in €₂₀₂₁.

2.2.1. Fixed Capital Investment

The fixed capital investment (FCI) can be estimated from the equipment cost. The individual costs of equipment were estimated with abacus [37,38] or scale-up power law based on capacity according to equation (1) [11,39–50].

$$C_{new} = C_0 \cdot \left(\frac{Q_{new}}{Q_0}\right)^n \quad (1)$$

$$C_{2021} = C_y \cdot \left(\frac{CEPCI_{2021}}{CEPCI_y}\right) \quad (2)$$

C_{new} is the equipment cost for a Q_{new} capacity estimated from a reference unit cost C_0 for a Q_0 capacity, n is the exponent factor specific to the equipment. The prices were adjusted to the year 2021 with equation (2) using the chemical engineering plant cost index (CEPCI). Its value for 2021 is $CEPCI_{2021} = 708$ [51]. C_{2021} and C_y are the cost updated to 2021 and the reference cost of the year y . Prices in US dollars were also converted to euro with the mean value of exchange rate 0.846 €//\$ in 2021 [52].

Various costs are attributed to a plant, either direct (DC) or indirect costs (IC). The direct costs are relative to equipment purchase, installation, instrumentation, piping, electrical, insulation, painting & buildings, process, auxiliary & service facilities and yard improvement. The indirect costs are relative to engineering, supervision, construction expenses, legal expenses, contractor fees and contingencies. At this stage of factored estimate study ($\pm 30\%$ accurate), the sum of direct and indirect costs (i.e. the fixed capital investment FCI) can be estimated by multiplying the purchased equipment cost by a global factor: the fixed capital investment factor (FCIF) (3) [37]. When costs are taken from the literature, this coefficient is closely linked to the source of the original reference cost. The use of various costs source requires to carefully check the type of costs taken into account.

$$FCI = DC + IC = \sum FCIF \cdot Purchased\ equipment\ cost \quad (3)$$

The costs of all major equipment determined in this study can be found in supplementary material 4. In the absence of an industrial quote, and when available, abacus estimations were preferred because these estimations are given with domains of validity. This is usually not the case with scale-up power law, the potential error on the calculated cost is then higher, especially when the scale ratio between the estimate and the reference unit is far from unity.

2.2.2. Cost of manufacturing *COM*

The economic assessment provides the cost of manufacturing to take into account the cost of the raw material, the utilities and consumables, the labor, the maintenance and repairs and other costs related to the operations.

The cost of woodchips was supposed to be 17 €/MWh on an LHV-basis corresponding to an average market price for woodchips of medium to coarse granulometry and 40%-humidity in 2021 in France [53]. This price does not include the transport cost from the preparation site of woodchips to the unit. This transport cost was evaluated with the article of Yordanova and Migette (2017) [54] by assuming a 150 km distance for the case GR-100 and 50 km for the other scenarios (detailed in supplementary material). The overall cost of woodchips was estimated to be 26.9 and 22.0 €/MWh at the entrance of the pyrogasification facility including transport and taxes for the 100 MW (GR-100) and the 20 MW-unit, respectively. In scenario GR-100, 21 trucks (27t-moving floor type) are required with an average of 30 unloading per day. In others scenarios, only 2 trucks are required with an average of 6 unloading per day resulting from the smaller transport distance of the 20 MW-unit in comparison with the 100 MW-unit (50 km vs. 150 km). This lower traffic would probably be more easily accepted by the neighborhood population. More details can be found in supplementary material 1.

The cost of labor was calculated by estimating the number of workers required and the mean wages in France. The hypothesis can be found in Table S2 in supplementary material. The number of truck drivers is also reported in this table even if their salaries are already taken into account in the woodchips transport cost.

In the pyrolysis scenarios, the biochar is either sold (P) at market price and not supposed sequestered or sequestered (P-S) in the long term as soil amendments or stored in a former mine [55]. In case of carbon sequestration, the plant benefits of CO₂ credits from another company. Relevant hypotheses on the feedstock, utilities and consumables costs and price products are gathered in Table 2.

Table 2: Prices of products and costs of feedstock and utilities.

Feedstock cost	Year: 2020	2021	2022	Unit	Ref.
Woodchips (40% moisture)	17		20	€/MWh	[53]
Transport of woodchips:					
- 50 km (GR, G, P, P-S)		5.0		€/MWh _{LHV-basis}	[54]
- 150 km (GR-100)		9.9		€/MWh _{LHV-basis}	[54]
Product market prices					

Heat for industry (water at 80°C)	30			€/MWh	[56]
Hydrogen (99.99%v, SMR at large scale)	0.88	2.59	4.78	€/kg	[2,57–60]
Bio-char:					
- sold and not sequestered	500			€/t _{char}	[61]
- sequestered (CO ₂ credits)	550 (150 €/t _{CO2})			€/t _{char}	[61]
Utility costs					
Electricity	86	86	125	€/MWh	[56,62]
Natural gas (start-up)	9.5	47	93	€/MWh	[63]
Fresh water	0.5			€/m ³	
Bed material	230			€/t ^a	[11]
Catalyst reformer (Ni-based)	50			€/kg	[64]
Catalyst HTS	20			€/kg ^a	[43]
Catalyst LTS	20			€/kg ^a	[43]
Membrane cost	40			€/m ²	[48]
Membrane replacement	25			€/m ²	[48]
Adsorbent	6			€/kg ^a	[43]
Waste disposal					
Landfill	90			€/t	
Ash under cyclone and fabric filter	250			€/t	

^aCost from original reference updated to 2021 with CEPCI index.

2.2.3. Cost of H₂ from SMR

Historically, the price of H₂ produced from steam methane reforming (SMR) varied from 1-3 €/kg_{H₂} for large-scale consumer delivered with pipeline (ammonia, petrochemical, methanol or chemical processes) to 5-7 €/kg_{H₂} for smaller consumers delivered by truck [57–60]. This hydrogen price is directly linked to the fossil gas cost. As a consequence of the post-covid pandemic and the geopolitical context, the European market price of natural gas increased from around 20 €/MWh to more than 150 €/MWh (supplementary material 2). The production cost of hydrogen followed this rise. The Table 3 gives the estimated production cost of hydrogen from large-scale SMR as a function of the natural gas averaged price. These production costs of hydrogen rose from 1.2 €/2017/kg_{H₂} to 4.8 €/2022/kg_{H₂}.

Table 3: Hydrogen production price estimated in Europe.

Year	Natural gas price ^b [€/MWh]	No CCUS ^c (efficiency 76%)			With CCUS ^c (efficiency 69%)		
		OPEX ^a [€/kg _{H₂}]	CAPEX ^a [€/kg _{H₂}]	Total [€/kg _{H₂}]	OPEX ^a [€/kg _{H₂}]	CAPEX ^a [€/kg _{H₂}]	Total [€/kg _{H₂}]
2017	17.2	0.151	0.302	1.21	0.328	0.541	1.70
2020	9.5	0.157	0.313	0.88	0.341	0.562	1.36
2021	46.7	0.179	0.359	2.59	0.391	0.644	3.29
2022	93.4	0.228	0.456	4.78	0.497	0.819	5.83

H₂ production cost = CAPEX + OPEX + Fuel

^aOriginal cost in \$₂₀₁₇ [2], converted to € and updated with CEPCI index.

^bNatural gas corresponding to market annual price average value (PEG index), first semester for 2022 [63].

^cCCUS: carbon capture, utilization and storage.

2.2.4. Financial options and economical criterion

To evaluate the profitability of the plant, the net profit after taxes of the year k is calculated from equation (4). R is the revenue from sales, COM the cost of manufacturing, d_k the depreciation cost and t the taxation rate.

$$NET\ PROFIT_k = (R - COM - d_k) \cdot (1 - t) \quad (4)$$

The resulting cash flow of the year k is deduced with equation (5).

$$CASH\ FLOW_k = (R - COM - d_k) \cdot (1 - t) + d_k \quad (5)$$

To estimate the opportunity of the investment, the net present value (NPV) can be determined with equation (6). This method considers the time value of money, assuming a discount rate i and is particularly relevant for multi-product processes.

$$NPV = -TCI + \sum_{k=1,n} \frac{CASH\ FLOW_k}{(1 + i)^k} \quad (6)$$

The minimum selling price of H₂ to reach a given internal rate of return (IRR) is the minimum price of hydrogen that gives an NPV equal to zero at the end of the plant life considering the discounting rate equal to the IRR. The IRR expected is estimated to 7% [65]. The economic hypotheses are given in Table 4.

Table 4: Financial options.

Options	
Operation time	7500 h/year
Discounting rate i	7%
Taxation rate t	30%
Project life n	20 years
Construction period	1 year
Depreciation period	15 years (linear)
Loan	50% of total capital investment. Interests rate 10% per year [21,24].
CAPEX estimation	

Working capital WC	$WC = 15\% \cdot FCI$ [43]
Total capital investment	$TCI = FCI + WC$
OPEX estimation	
Raw materials & utilities	From process simulation and Table 2.
Operating labor	See Table S2 in supplementary material.

2.3. Life Cycle Assessment

2.3.1. Definition of the system and life cycle inventories

A comparative life cycle assessment was conducted to evaluate the five pyrogasification scenarios to produce hydrogen. As a reference system for hydrogen production, the steam methane reforming (SMR) from natural gas was chosen.

The functional unit consists of 1 kg of hydrogen with 99.99 vol% purity at 70 bars. A cradle-to-gate system boundary was set to cover the woodchips production or extraction of natural gas to the final product: hydrogen (Figure 2).

The life cycle inventory (LCI) of the pyrogasification scenarios were calculated from mass and energy balances of our previous study [13]. For SMR reference scenario, the LCI was taken from Susmozas et al. [66], in which no evaluation of methane emission from leaks or unburns was considered. In this study, the hydrogen was produced at 28 bars [66], the additional electrical consumption to increase the pressure to 70 bars in compressors was evaluated with AspenPlus® and included in its LCI for a better consistency. The detailed LCI for each case are presented in supplementary material 6.

Hydrogen is the only product of the SMR process, whereas the pyrogasification processes generate two or three products (H₂, heat and biochar). The relative impacts were allocated on an energy basis (LHV).

2.3.2. Method and Hypotheses

The carbon contained in the wood before harvesting was considered carbon-neutral and the related CO₂ emissions from wood pyrogasification as biogenic emissions. The different inputs and outputs of the LCI were evaluated from Ecoinvent database. The calculations were conducted with Simapro 9 software.

The method chosen for the evaluation of the impacts is the midpoint method CML-IA baseline V3.05 [30], a common method used for hydrogen systems [25]. This method evaluates the following impacts: abiotic

depletion (AD), abiotic depletion of fossil fuels (ADff), global warming potential for 100 years (GWP) estimated from the IPCC guidelines [27], acidification potential (AP), ozone layer depletion potential (ODP), photochemical oxidation formation potential (POFP), eutrophication potential (EP) along with impacts related to toxicity for human (HTTP), fresh water and marine aquatic ecotoxicity (FAETP, MAETP), terrestrial ecotoxicity (TETP).

3. RESULTS AND DISCUSSION

3.1. Techno-Economic Assessment

Figure 3 presents the fixed capital investment in each case with the contribution of each sub process. The detailed equipment costs for each case is presented in supplementary material 5.

GR-100 presents an economy of scale in comparison with GR. The fixed capital investment required for 100 MW_{LHV wood}-scale is lower than 5 times those of 20 MW_{LHV wood}-scale (1.11 vs. 1.63 M€/MW_{LHV wood}). This was expected since the exponent used in equipment cost is lower than 1 for scale ratio (equation 1).

Cases G (1.11 M€/MW_{LHV wood}) and P (1.04 M€/MW_{LHV wood}) have lower capital requirements than GR (1.63 M€/MW) due to the absence of catalytic reactors in the syngas cleaning and upgrading sub-process. The hydrogen separation sub-process has also a relatively smaller cost despite the two-stage separation process in P and G. This unexpected trend is explained because the first stage using membrane separation is used at lower pressure (5 bars) than PSA (27 bars). As the compressor investment cost is a function of the power required, it leads to a smaller investment cost for compressors than in GR. The following PSA H₂ unit is also smaller than in GR. This separation architecture taken from our previous work [13] was designed to minimize the specific separation cost of hydrogen.

An additional consequence of this architecture is a bigger flow rate of tail gas dedicated to the boiler in G and P than in GR. This explains the more important investment required for heat generation (gas boiler).

Case P (1.04 M€/MW_{LHV wood}) is the cheapest of all options due to a lower syngas production of the process that reduce the investment of syngas compression and H₂ separation sub processes (-18% between G and P). A lower feed rate of oxygen for autothermal pyrolysis than in gasification also reduce the capital investment of oxygen separation sub-process (-26% between G and P).

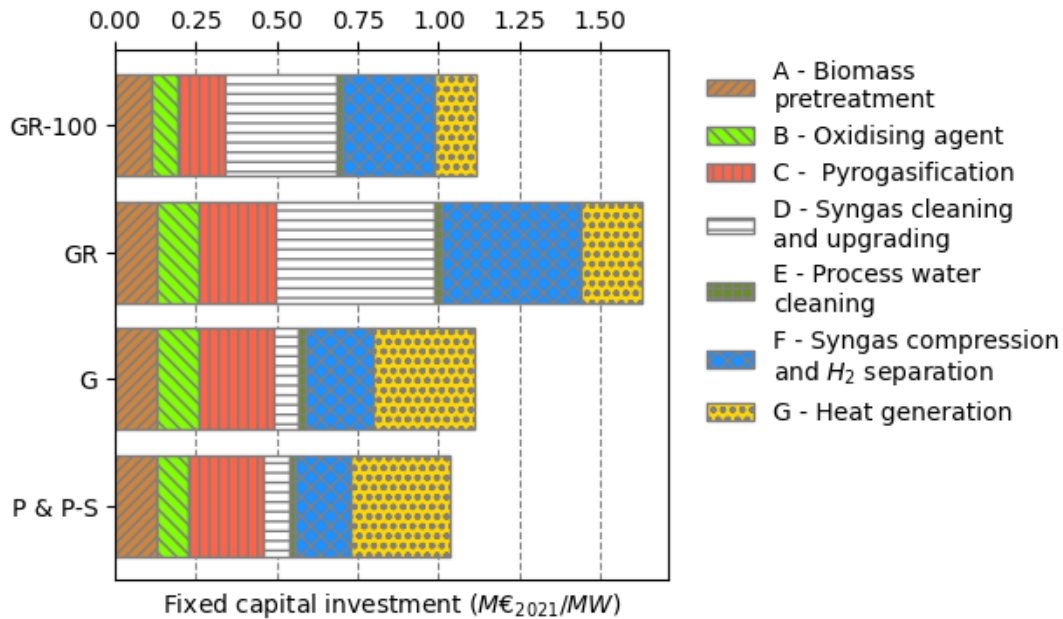


Figure 3: Fixed capital investment (in M€₂₀₂₁/MW_{LHV wood}) of the different scenarios (more details in supplementary material 5).

Figure 4 presents the operational expenditure calculated in each case. The main operational cost of these processes are driven by the cost of woodchips feedstock (38 to 45%). The transport of the woody feedstock represents 22% in GR-100 for 150 km in comparison with 12 to 13% for 50 km in other cases. The second main cost (21 to 16%) is due to the consumption of electricity mainly for syngas compression in the hydrogen separation sub-process.

The relative cost of labor was estimated higher at the 20 MW_{LHV wood}-scale (10%) compared to a bigger 100 MW_{LHV wood}-scale (4%).

All in all, the relative OPEX of the case GR-100 (44.2 €/MWh_{LHV wood}) equals those of the case GR (44.3 €/MWh_{LHV wood}), whereas cases G and P yield lower operating costs (39.2 €/MWh_{LHV wood} and 37.6 €/MWh_{LHV wood} respectively).

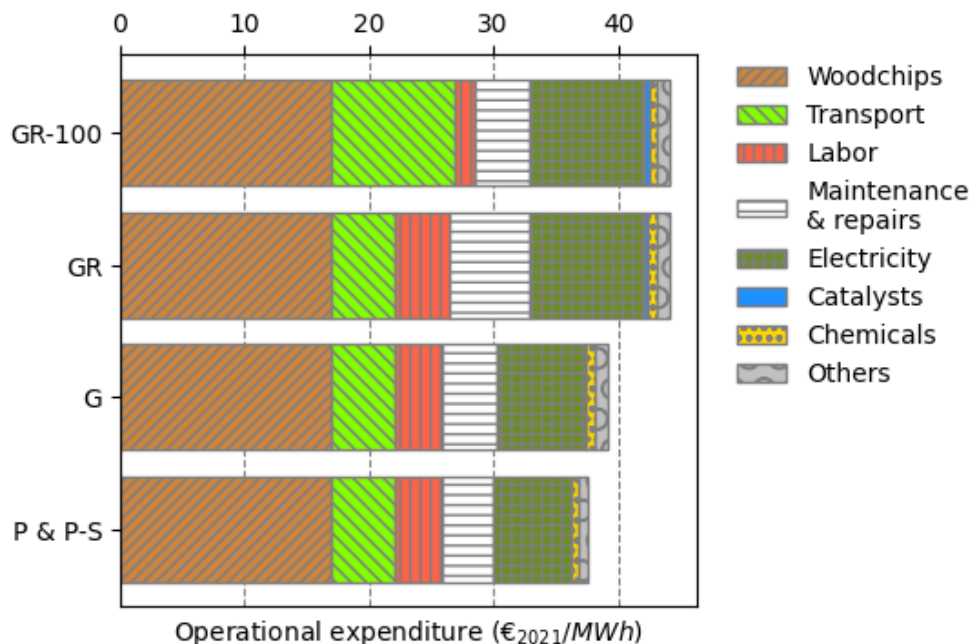


Figure 4: Operational expenditure of the different scenarios (in €₂₀₂₁/MWh_{LHV wood}).

Figure 5 presents the minimum selling price of hydrogen to reach an internal rate of return of 7% considering the prices of Table 2 for heat and biochar. The post-covid pandemic had a major impact on the energy market (especially natural gas for hydrogen from SMR) and the global chemical engineering economics (supplementary material 2). The uncertainties of economic conditions made the forecast of future conditions hazardous. The Figure 5 presents the results of the economic analysis of the last three years, 2020-2022 with the corresponding prices of natural gas, electricity, wood and CEPCI index for the evaluation of CAPEX. The minimum selling price of hydrogen range from 3.8 to 9.1 €₂₀₂₁/kg_{H2} and even 4.7 to 12.6 €₂₀₂₂/kg_{H2} with the most recent economic indicators (CEPCI index June 2022). On the other side, the hydrogen production cost from SMR was estimated to 2.5 €₂₀₂₁/kg_{H2} and 4.8 €₂₀₂₂/kg_{H2} in 2021 and 2022, respectively.

GR and GR-100 are the cheapest processes to produce hydrogen because H₂ production is promoted by steam reforming and water gas-shift reactors. The results show 16% difference between case GR at two scales (3.84 and 4.47 €₂₀₂₁/kg_{H2} for 100 MW_{LHV wood} and 20 MW_{LHV wood} respectively). The economy done at a bigger scale can counterbalance the higher woodchips transport cost. This big scale production price is close to the estimated SMR production cost, especially in the 2022 estimate, where GR-100 could be at the same price as the fossil-based production (4.78 €₂₀₂₂/kg_{H2}).

Cases G and P presents higher hydrogen costs: 8.0 and 9.0 €₂₀₂₁/kg_{H₂}, respectively. With this method, all the extra-cost required to reach the targeted internal rate of return is attributed to hydrogen, although this is not the only product. The yield of hydrogen is smaller in cases G and P than in GR. The smaller capital cost of these scenarios and the increase of the revenues related to heat and bio-char do not compensate for the large reduction in H₂ yield. This may be different if the heat and bio-char produced from renewable resources could be sold at a higher price (see sensitivity analysis section).

The minimum selling price of hydrogen at large scale (GR-100, 3.84 €₂₀₂₁/kg_{H₂}) is close to the value, converted in euros 2021, mentioned by Park et al. (3.2-4.4 €₂₀₂₁/kg_{H₂}) [21]. Salkuyeh et al. mentioned 3.0-3.5 €₂₀₂₁/kg_{H₂} on a scale more than ten times bigger without and with CO₂ capture [24]. On a smaller scale (10 MW_{LHV_wood}), Sentis et al. evaluated the hydrogen cost to 3.8 €₂₀₂₁/kg_{H₂} [23]. If a lower purity of hydrogen is targeted, the resulting minimum selling price could be reduced.

As a comparison, the international renewable energy agency (IRENA) estimated the production cost of green hydrogen from electrolysis around 2 to 5 €₂₀₂₀/kg_{H₂} [67]. The underlying assumption of this estimation assumed an electricity produced from wind farm or solar panel at a price between 17 to 46 €₂₀₂₀/MWh_e. These electricity prices are very low in comparison with the prices reached in 2022 with the post-covid context in the European market.

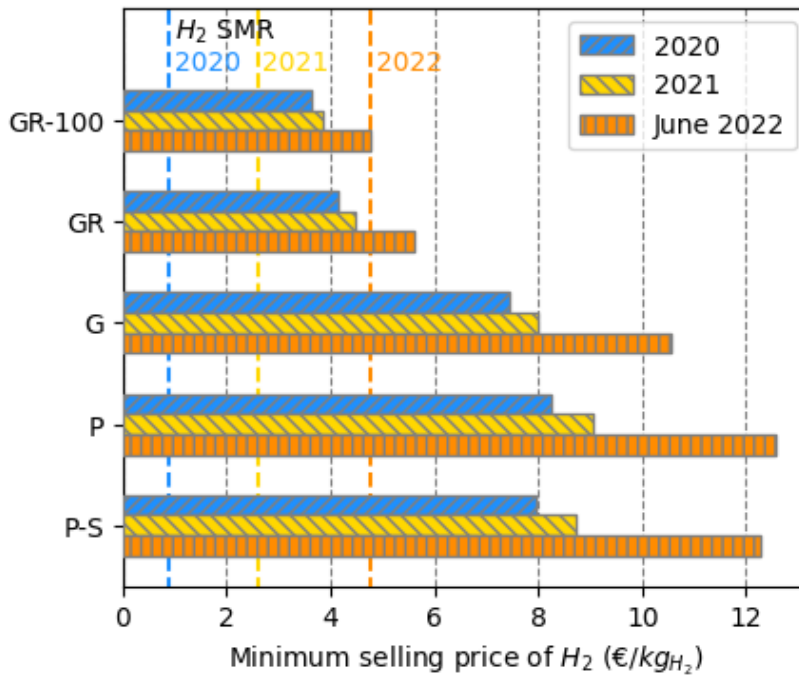


Figure 5: Minimum selling price of hydrogen for each case and hydrogen production cost from SMR [2]. Prices adjusted to the market conditions of year 2020, 2021 and 2022 (June) (natural gas, electricity, wood prices and CEPCI indexes (see Table 2)).

3.2. Life Cycle Assessment

3.2.1. LCA results on impact factor of the different options

The results of the five pyrogasification processes (GR-100, GR, G, P, P-S) and the SMR reference process are presented in Figure 6 for five impacts (GWP, AP, ADff, ODP and POFP). These impacts were selected as they are also discussed in other studies [31–33]. The contribution of the inputs and emissions of the processes are also presented. The detailed results are available in supplementary material 7.

First of all, the pyrogasification processes gives a lower impact than the reference SMR system for the global warming potential (GWP), the abiotic depletion of fossil fuels (ADff) and the ozone depletion layer potential (ODP). Meanwhile, the acidification potential (AP) and the photochemical oxidant formation potential (POFP) are higher than the SMR process. The emissions related to the process (solid waste to landfill and emissions to air and water), the electricity consumption and the production of woodchips are the main players for the impacts considered, whereas it is essentially the natural gas and its associated emission of CO₂ in the SMR case. Among the pyrogasification processes, P and P-S generate less impacts for the functional unit (1 kg H₂) because they produce less gas products and therefore generate less water and gaseous emissions.

The slight differences between GR-100 and GR only results from the additional woodchips transport distance in GR-100 (150 km vs. 50 km). For these considered distances and 27 t-moving floor trucks, the transport of woodchips does not represent a major source of impact. Yet, the social acceptance is not evaluated. It is likely to be lower with bigger-scale production. The 100 MW unit generates an average of 30 truck deliveries per day in comparison with only 6 trucks at 20 MW.

The GWP of the functional unit (1 kg of H₂) of the pyrogasification processes are more than ten times smaller than the SMR reference system, with the biogenic emissions hypothesis for the wood-based carbon. The Table 5 presents the allocated value on energetic basis for each product. The P-S process provides energetic vectors (H₂ and heat) along with biochar which can be considered as a carbon sink. The pyrolysis option is even negative for GWP with the biochar produced sequestered in the long term in former mines [55] or used as a soil amendment. In comparison, the GWP emissions from electrolysis based

hydrogen can be evaluated to 2.84-3.79 $\text{kg}_{\text{CO}_2\text{eq}}/\text{kg}_{\text{H}_2}$ for the electrical consumption only, assuming an electrolysis efficiency between 50 and 66% [8] and an average emission factor for the French electricity grid at 56.9 $\text{g}_{\text{CO}_2\text{eq}}/\text{kWh}_e$ in 2021 [68]. Furthermore, the effective GWP emissions from SMR could be underestimated in the study of Susmozas et al. [66]. Howarth et al. evaluated these emissions at 18 $\text{kg}_{\text{CO}_2\text{eq}}/\text{kg}_{\text{H}_2}$ by considering the methane leaks along the process [6].

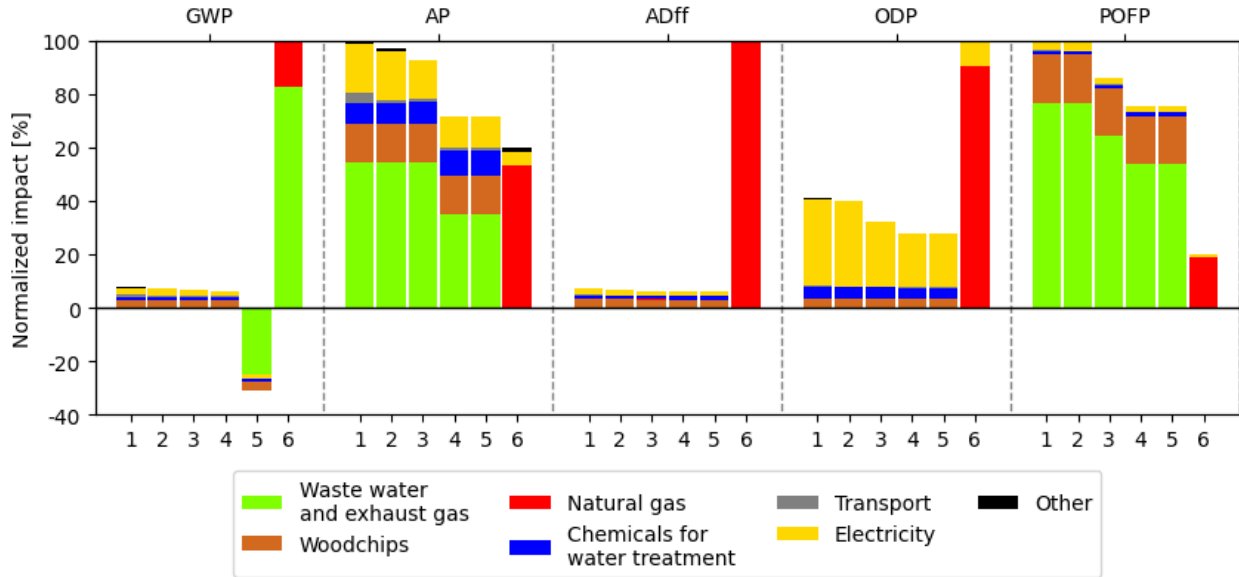
The acidification potential (AP) calculated is higher for the pyrogasification processes than in the reference process. Gaseous emissions (SO_2) play the main role on this impact. For example, all the sulfur content of wood was assumed to be released as SO_2 [13], a major molecule for acidification. In the SMR process, no SO_2 emissions were considered [66].

The impact on abiotic depletion of fossil fuels (AD_{ff}) was confirmed to be 13 to 17 times less important for pyrogasification than SMR since fossil fuels does not play a major role contrary to SMR. Fossil fuels are used for starting-up, woodchip production and transport and grid electricity production.

The ozone layer depletion potential (ODP) is mainly driven by the consumption of natural gas and the production of electricity (itself partially produced from natural gas). The pyrogasification scenarios are globally 2 to 4 times less emissive than SMR.

The photochemical oxidant formation potential (POFP) shows a higher impact for pyrogasification scenarios.

In order to minimize the impacts on acidification and the formation of photochemical oxidants, the gaseous and liquid emissions could be reduced by capturing sulphur dioxide and polycyclic aromatic hydrocarbons emitted during syngas burning and an effective waste water treatment system. Additional work could be done regarding to the water treatment unit i.e. to analyze in details the emissions of this process.



1: GR-100, 2: GR, 3: G, 4: P, 5: P-S, 6: SMR. ADff: Abiotic depletion (fossil fuels), GWP: Global warming, ODP: Ozone layer depletion, POFP: Photochemical oxidation, AP: Acidification. Other: catalysts, bed material, adsorbents for PSA H₂, process water.

Figure 6: Relative impact comparison attributed to H₂ production between the different scenarios and steam methane reforming (SMR) with CML-IA baseline V3.05 method. In P, biochar is sold as a product, in P-S biochar is sequestered.

Table 5: Global warming potential attribution to each product per functional unit (FU).

Product		GR-100	GR	G	P	P-S	SMR
H ₂	kg _{CO₂ eq} /FU	0.785	0.737	0.669	0.632	-2.55	10.3
	Reduction/SMR	-92%	-93%	-94%	-94%		
Heat	kg _{CO₂ eq} /FU	0.353	0.332	2.25	2.56	-10.2	
Bio-char	kg _{CO₂ eq} /FU				0.998	-3.96	
Total	kg _{CO₂ eq} /FU	1.14	1.07	2.91	4.20	-16.8	10.3
Emissions per unit of energy ^a	g _{CO₂ eq} /kWh	19.8	18.6	16.9	15.9	-63.7	259

^aThe CO₂ emissions per unit of energy are equal for the three products (H₂, heat, biochar). This is a consequence of the energy allocation of the impacts.

3.2.2. Influence of the assessment method

A sensitivity analysis of the assessment method was conducted on LCA indicators by testing four methods (RecipeH, CML, IMPACT2002+ and ILCD). The results are presented in supplementary material 8 and shows very close results and similar trends. For the GWP impact: the three first methods concluded to the same results because the method used for this impact is the same (IPCC). For the AP impact, only slight

differences are found between the four methods. For the ADff impact, the three first methods are in good agreement, except for SMR scenario and method IMPACT2002+. The method ILCD does not include ADff impact. For the ODP impact, a good agreement is found for the four methods. The CML method, chosen in this paper, is then adapted for the life cycle assessment presented.

3.3. Cost per CO₂ avoided

The production of hydrogen in 20 MW_{LHV,wood}-scale units is dedicated to feed with pipelines small consumer industry or captive H₂-vehicle fleet, thus avoiding the cost of H₂ transport by truck. The estimated production price of hydrogen in SMR around 2.6 €₂₀₂₁/kg_{H₂}, which is too low to match economic profitability for most of 20 MW-scale pyrogasification units. Yet, these scenarios have a beneficial impact by providing hydrogen and heat from renewable and local resources. An alternative way of considering the relevance of such processes is to estimate the over-cost required to develop these industries. The amount of subsidies required to reach profitability can be related to CO₂ avoided emissions with actual means of production. In this case hydrogen from SMR. This “cost of action” can be calculated as the ratio of extra-cost (subsidies) divided by the emissions avoided by a classical production process. The avoided emissions are related to the hydrogen production and the heat production. The GWP for heat dedicated to heat network was estimates to 116 kg_{CO₂}/MWh as the average value for 2018 [69]. To estimate the relevance of a process regarding the investment priorities to tackle emissions issues, this cost is estimated to 250 €/t_{CO₂ avoided} in 2030 and 500 €/t_{CO₂ avoided} in 2050 in France according to a report [70] prepared to meet the Paris agreement targets.

The results are presented in Figure 7. Because of the actual uncertain market conditions already mentioned, three years for costs are considered: 2020, 2021 and 2022. 2020 was characterized by a low natural gas average price (9.45 €₂₀₂₀/MWh) during the beginning of the covid pandemic. As a consequence, the resulting estimated production price of hydrogen was low (0.88 €₂₀₂₀/kg_{H₂}). The year 2022 is on the contrary characterized by historically extremely high costs of natural gas: 93 €₂₀₂₂/MWh in the first semester 2022. The index (CEPCI) used for economic calculations rise with the same trend leading to an estimated price of hydrogen at around 4.78 €₂₀₂₂/kg_{H₂} (Table 3). These market conditions are taken into account in Figure 7.

The inflation context of the market and on energy reduces the need for subsidies in all scenarios between 2020 and 2022 (except for the pyrolysis scenarios P and P-S), despite the increasing capital investment in

the same period. Among the 20 MW processes, P-S has the lowest cost of action due to its ability to sequester carbon (except in the context of 2022 with a high cost of natural gas and therefore the market price of hydrogen from SMR). Scenario GR-100 almost do not require subsidies in the 2022 context, and the subsidies in scenario GR are significantly reduced. The other options fall below, or near, the 2030 target of 250 €/t_{CO2 avoided}.

The rising price of heat resulting from the rising prices of fossil fuels will be beneficial to scenarios G and P. Indeed, these scenarios produce more heat (and less H₂) than GR. Furthermore, fewer investments are required with simpler technologies involved. It should be stressed out that the cost of action is divided by two between scenarios P and P-S. At 150 €/t_{CO2} of carbon credit, the carbon sequestration is more favorable than carbon use from an economic standpoint.

A high level of energy market would definitely improve the profitability of such processes if the wood availability is ensured and its price does not follow the same rising trend. This increase could be driven by higher taxation of carbon emissions. On the other hand, the cost of action i.e., the need for subsidies, would decrease for these low carbon emissions processes.

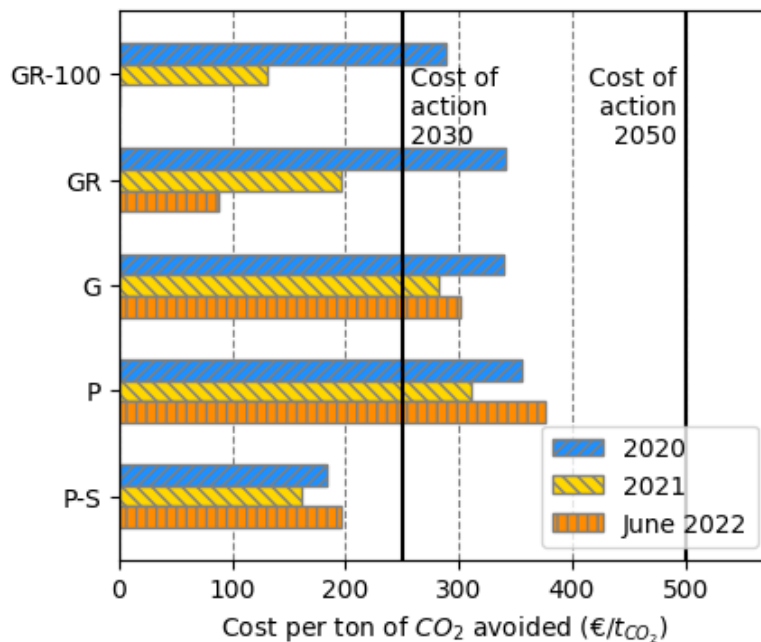


Figure 7: Cost of action, amount of subsidies per ton of CO₂ avoided.

In case P, biochar is sold at the market price (500 €/ton) and not sequestered. In case P-S, biochar is sequestered and carbon credits can be sold at 550 €/ton of char (for 150 €/t_{CO2 avoided}). H₂ market prices from Table 3, other costs from Table 2, CEPCI adjusted for years 2020-2021-2022.

3.4. Sensitivity Analysis

To determine the main uncertainties of the techno-economic analysis, a sensitivity analysis is presented in Figure 8, Figure S10 and Figure S11 for cases GR, P and P-S respectively. From the result obtained from reference year 2021, multiple parameters were varied by $\pm 50\%$: the capital investment, the selling market price of hydrogen, heat and biochar, the CO₂ credit, the costs of electricity, woodchips, catalysts and the number of operators.

By evaluating various cost estimation methods, van Amsterdam found that the results can differ hugely from one method to another [71]. Thus the 30% error expected in such a method could increase a lot. This inaccuracy can somehow be decreased with the number of equipment in a so-called damping effect.

In GR case, the main parameters affecting the cost of action is the hydrogen selling price (the main product of the plant), the woodchip price and the capital investment. The loan interest, electricity cost and heat selling price have a lower impact. If the hydrogen could be sold at 4.47 €₂₀₂₁/kg (i.e. the minimum selling price of hydrogen) the facility does not require subsidies.

In P case, the hydrogen selling price is only the fifth main parameter because this case produces less hydrogen. The main parameters are the woodchips cost, the heat selling price and the capital investment. The biochar selling price is the fourth main parameter. In P-S case, the role of biochar selling price is replaced by the CO₂ credit received.

It is important to put the biomass cost into perspective. The variation considered ($\pm 50\%$) is certainly overestimated. In contrary to natural gas, the wood energy costs are more stable upon the time on the market [53].

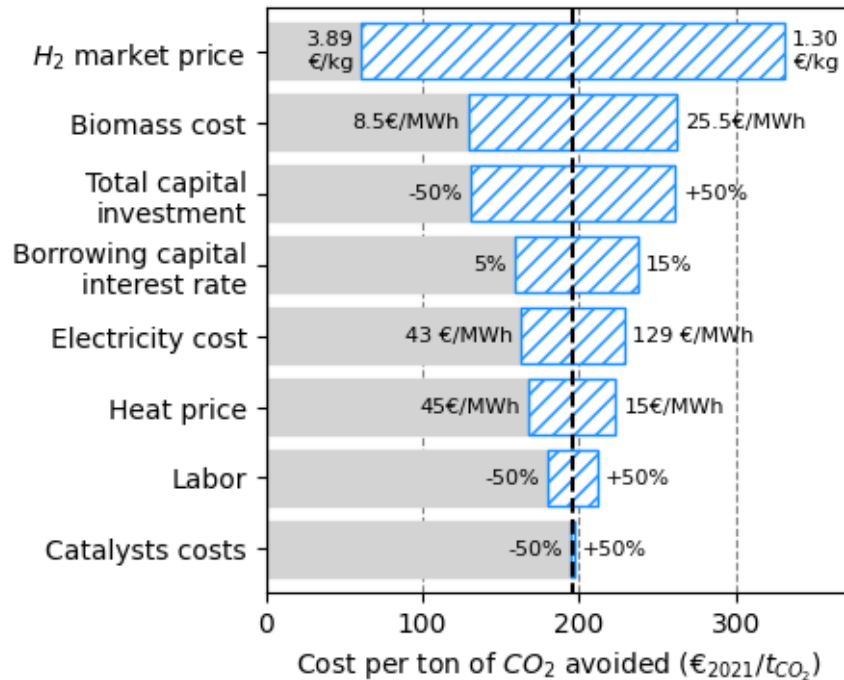


Figure 8: Sensitivity analysis on case GR (reference year 2021).

4. CONCLUSION

This article presents a detailed techno-economic analysis of three processes of hydrogen production from biomass. This hydrogen at 70 bars and 99.99% purity could be used for small consumer industries or captive H_2 -vehicle fleet. A life cycle assessment was also conducted to evaluate the environmental impacts.

The scenarios presented are more than 10 times less emissive of greenhouse gases than the reference production of hydrogen from steam methane reforming. A carbon-negative process based on autothermal pyrolysis is also proposed and evaluated on a techno-economic basis.

Depending on the scenario, the hydrogen production cost was evaluated between 4 and 9 $\text{€}_{2021}/\text{kg}_{H_2}$. In the economical context of 2020-2022, the market prices of SMR hydrogen, heat and biochar were too low to make these processes profitable. Additional subsidies are required. We evaluate this cost between 130 and 310 €_{2021} per ton of CO_2 avoided in comparison with steam methane reforming for the production of hydrogen and the average global warming potential of heat networks in France. This cost could decrease or even become unnecessary if the market price of natural gas stays permanently high in the long term (as

it is in 2022). Alternatively, a higher market cost for renewable based hydrogen could also reduce the amount of subsidies required.

Other results from LCA show less impact for biomass-to-H₂ processes on the ozone layer depletion and consumption of fossil fuels than steam methane reforming resulting from natural gas. The acidification and photochemical oxidant potential are estimated to be higher in the pyrolysis and gasification processes than the reference system because of electricity consumption and process emissions (waste water and exhaust gas). Other points of these processes can be further studied to assess their impacts: the detailed characterization of emissions to water and gaseous emissions from the industrial process. The regeneration of activated carbon used for water treatment could also decrease these impacts. Therefore, based on these different scenarios analysis, a potential better process for a cleaner production of H₂ (based on LCA) must reduce emissions to water generated during syngas washing.

From the results presented: 1) GR can be considered as the cheapest process, 2) P and P-S present the lowest environmental impacts, 3) P-S requires less subsidies per amount of CO₂ avoided. A trade-off must be made between maximizing H₂ production from gasification and producing biochar from pyrolysis leading to carbon sequestration. The social aspects (like community perception, employments, value-chains, available skills in the territory, etc.) are other important criteria which may be further assessed.

5. FUNDINGS

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HIGHLIGHTS

- Gasification of woodchips and reforming to produce hydrogen below 6 €/kg_{H2}.
- Autothermal pyrolysis for hydrogen production and carbon sequestration.
- Far lower emissions than SMR (up to 15 times less for H₂, 7 times less for heat).
- Cost of subsidies per ton of CO₂ avoided between 0 and 400 €/t_{CO2 avoided}.

Declaration of interests

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.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: