

Co-production of Hydrogen and Electricity from Lignocellulosic Biomass: Process Design and Thermo-economic Optimization

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Abstract

The thermochemical production of hydrogen from lignocellulosic biomass is systematically analyzed by developing thermo-environmental models combining thermodynamics with economic analysis, process integration techniques and optimization strategies for the conceptual process design. H₂ is produced by biomass gasification and subsequent gas treatment, followed by H₂ purification via CO₂ removal. It is shown how the overall efficiency is improved by considering process integration and computing the optimal integration of combined heat and power production. In the conversion process, electricity can be generated in steam and gas turbine cycles using the combustion of the off-gases and recovering available process heat. Additional electricity can be produced by burning part of the H₂-rich intermediate or of the purified H₂ product. The trade-off between H₂ and electricity co-production and H₂ or electricity only generation is assessed with regard to energy, economic and environmental considerations. Based on multi-objective optimization, the most promising options for the poly-generation of hydrogen, power and heat are identified with regard to different process configurations. The best compromise between efficiency, H₂ and/or electricity production cost and CO₂ capture is identified. Biomass based H₂ and electricity reveal to be a competitive alternative in a future sustainable energy system.

Keywords: Biomass, Hydrogen, Polygeneration, Process integration, Thermo-economic optimization, Life cycle assessment

Nomenclature

Abbreviations

BM	Biomass
CC	Carbon Capture
CFB	Circulating Fluidized Bed
CGC	Cold Gas Cleaning
COE	Cost Of Electricity
E_{imp}	Electricity import
FICFB	Fast Internally Circulating Fluidized Bed
GT	Gas Turbine
GWP	Global Warming Potential
HHV	Higher Heating Value
HP	Heat Pump
HTS	High Temperature Shift
IBGCC	Integrated Biomass Gasification Combined Cycle

IGCC	Integrated Gasification Combined Cycle
IPCC	Intergovernmental Panel on Climate Change
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LHV	Lower Heating Value
LTS	Low Temperature Shift
MDEA	Methyldiethanolamine
MEA	Monoethanolamine
MOO	Multi-Objective Optimization
NG	Natural Gas
NGCC	Natural Gas Combined Cycle
PC	Pulverized Coal
PG	Producer Gas
PM	Particulate Matter
PSA	Pressure Swing Adsorption
RME	Rape Methyl Ester
SMR	Steam Methane Reforming
WGS	Water-Gas Shift

Greek letters

Δh°	Lower heating value, kJ/kg
$\Delta \tilde{h}_r^0$	Standard heat of reaction, kJ/mol
Δk°	Exergy value, kJ/kg
ϵ_{eq}	Natural gas equivalent efficiency, %
ϵ_{H_2}	H ₂ efficiency, %
ϵ_{ex}	Exergy efficiency, %
ϵ_{tot}	Energy efficiency, %
η	Efficiency, %
η_{CO_2}	CO ₂ capture rate, %
θ_{wood}	Wood humidity, %wt
Y_{H_2}	H ₂ Yield, g _{H₂} /kg _{BM}

Roman letters

\dot{E}	Mechanical/electrical power, kW
\dot{m}	Mass flow, kg/s
P	Pressure, bar
\dot{Q}	Heat, kW
T	Temperature, °C or K

Superscripts

+	Material/energy stream entering the system
-	Material/energy stream leaving the system

1 Introduction

In a future clean and abundant energy system, hydrogen is to be considered as an alternative energy carrier. H₂ is a clean fuel that can be used in combustion engines and fuel cells for electricity generation without local CO₂ emissions. Being a secondary form of energy, H₂ does not freely exist in nature and consequently has to be manufactured. Today H₂ is produced essentially by steam methane reforming, coal gasification and in a lesser extent by water electrolysis [2, 3, 10]. The drawback of these processes is that they are using fossil fuels or electricity from non-renewable sources. Within the worldwide challenge of global warming mitigation and energy supply, alternative H₂ production processes from renewable resources have received considerable attention. Different renewable energy resources may be used like wind, biomass or solar energy

[2, 10]. Biomass-based technologies have a high potential because they emit no or very few net CO₂ emissions, if carefully managed, since the released CO₂ was previously fixed in the plant as hydrocarbon by photosynthesis. H₂ production from biomass can be divided into two categories; thermo-chemical processes (i.e. biomass gasification and pyrolysis) and biological processes (i.e. biophotolysis and fermentation). An overview of these H₂ production processes from fossil and renewable resources and its economics can be found in [3].

The economic surveys in [3, 28, 18] among with other studies assessing the energy and exergy efficiency of the biomass conversion into H₂, as well as the influence of operating conditions, show that it is a technical feasible process that could be promising on the future energy market [16, 30, 29, 7, 1, 33, 36]. H₂ yields in the range of 80-130g_{H2}/kg_{Biomass} are assessed in [1, 33]. Energy efficiencies between 51 and 60% on lower heating value basis and H₂ production cost ranging from 29 to over 40 \$/MWh_{H2} are reported for biomass based H₂ processes in [16]. The performance of some H₂ processes using fossil or renewable resources are compared in Table 1.

Instead of producing H₂ from biomass, electricity can be generated in an integrated biomass gasification combined cycle (IBGCC) [6, 21, 8, 19, 5]. Even if there are still some technological challenges, this option appears as promising for CO₂ mitigation when compared to fossil fuel based power plants with CO₂ capture as shown by the performances reported in Table 2. Carbon capture decreases the efficiency of fossil power plants by around 10% points and increases the electricity cost by nearly one third which yields CO₂ avoidance cost in the range of 13-75\$/t CO_{2,avoided} [24]. Polygeneration processes co-producing H₂ and electricity can also be considered as an alternative.

The different studies about biomass conversion into H₂ or electricity assessed the process performance either by the thermal efficiency [7, 5], the economics [30, 8] or by the life cycle impacts [6, 21]. In some of these researches performance analyses and cost or environmental assessment are combined. However, no consistent comparison and optimization considering efficiency, costs and environmental impacts at the same time is performed. The difficulty to choose the best concept without including these three dimensions is revealed in [20] making a comparison of natural gas power plants concepts with CO₂ capture based on efficiency and CO₂ emissions without including economics. Moreover, in these studies energy integration is most of the time not explicitly considered. This may lead to sub-optimal solutions from the energy efficiency point of view. In [11, 26] the advantage of applying energy integration in biomass conversion processes is demonstrated. Environmental objectives have been introduced in a multi-objective optimization strategy combining energy integration and life cycle assessment (LCA) in [4] to study the impact of CO₂ capture in NGCC plants. This approach was also successfully applied to reveal potential process improvements in biomass conversion into synthetic natural gas [15] but has not yet been applied for H₂ and power generation.

In this paper, the thermochemical conversion of biomass into H₂ and electricity is investigated and optimized with regard to energy, economic and environmental considerations. This is done by applying a consistent methodology [15, 12, 31] combining thermodynamics with economic analysis, process integration techniques and using optimization strategies to generate optimal process configurations. The objective is to assess the competition between hydrogen or electricity only production processes and polygeneration with and without CO₂ capture by studying the influence of the operating conditions and process configurations. The use of process integration techniques allows to focus on the heat recovery and the energy conversion performance and on polygeneration aspects of H₂, heat and power and captured CO₂.

2 Methodology

This paper follows a previously developed methodology for the optimal thermo-economic process design of liquid and gaseous fuel production from biomass [15, 12, 31, 13]. The conceptual design methodology is illustrated in Figure 1. First suitable process technologies are identified

and thermo-economic models of the process units are developed. The thermodynamic model computes the chemical and physical transformations and the associated heat transfer requirements. It is combined with a separate energy integration model representing the heat recovery system. Based on the pinch analysis methodology, the optimal thermal process integration is computed after defining the maximum heat recovery potential between hot and cold streams and considering a minimum approach temperature ΔT_{min} . The process needs are satisfied by different utilities such as, combustion of waste and producer gas (PG), steam Rankine cycle for power production, gas turbine and cogeneration by burning either H₂-rich fuel or almost pure H₂. The optimal utility integration is defined by maximizing the combined production of fuel, power and heat with regard to the minimal operating cost by solving a linear programming problem [12, 22, 23]. Using the data from the flowsheeting and process integration models, the costs are estimated based on equipment sizing and cost correlations from the literature [34, 35]. For the life cycle inventory model, the cradle-to-gate LCA approach described in [15] is applied with a functional unit of 1 kJ of biomass at the inlet of the installation. The impact assessment method developed by the Intergovernmental Panel on Climate Change (IPCC) considering a time horizon of 100 years for the global warming potential (GWP) is used as the environmental performance indicator. Finally a multi-objective optimization using an evolutionary algorithm [12, 25] is performed. The optimization allows to identify competing objectives with regard to environmental (i.e. thermodynamic, economic and environmental) criteria and to assess the different trade-offs. The main feature of this methodology is the use of flowsheeting and process integration models in a multi-objective optimization framework that takes into account simultaneously economic and environmental considerations. In conventional process evaluation approaches, the processes are first designed and then different process configurations are compared based on various criteria. In comparison with conventional methods where process configuration scenario are compared, the proposed method allows one to make a systematic generation of optimal process configurations and to compare these on the same consistent performance criteria.

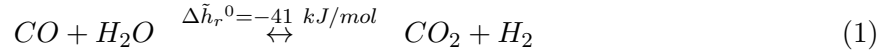
3 Process description

The thermochemical process converting biomass into H₂ fuel consists of wood handling, drying, gasification, gas cleaning and conditioning by reforming and shift conversion, and finally H₂ purification and/or H₂ burning for electricity generation. For each process step, several technology options can be proposed. This results into a process superstructure that is presented in Figure 2, where the investigated process layouts and the life cycle inventory (LCI) flows within the system's limits are highlighted. A separate torrefaction step is introduced before the gasification step to take advantage of the heat integration. The torrefaction operating at around 530[K] shifts part of the high temperature heat required for gasification to lower temperatures and yields torrefied gas which can be used for heat and power generation; leading to higher process performances. The products (i.e. H₂ or electricity) are defined by the options chosen at the cross points A and B. Depending on the production purpose and on the fuel which is burnt, the process either produces impure (80%mol) or pure H₂ (99%mol), imports or exports electricity, or is self-sufficient in terms of electricity.

3.1 Thermo-economic process model

The thermo-economic models for the drying, gasification and gas cleaning section have been developed in previous work [31, 13, 14] and the same specifications for the biomass and process units are considered in this work. The chemical conversion in the gasifier is modeled by equilibrium relationships with an artificial temperature difference as explained in [13, 14]. After the gasification the syngas is treated in two sequential water gas shift (WGS) reactors (Eq.1) one (HTS) operating at high [573-683K] and one (LTS) at lower [473-573K] temperature. This

increases the H₂ and CO₂ concentrations before CO₂ removal reducing the greenhouse gas emissions and producing higher LHV fuel.



For the H₂ separation and purification, CO₂ capture by chemical absorption with amines and by physical adsorption using pressure swing adsorption (PSA) are considered. A chemical absorption step is introduced before the PSA unit to produce pure H₂ and to capture high purity CO₂ which is compressed to over 100 [bar] for storage. For separating high partial pressure CO₂ from syngas methyldiethanolamine (MDEA) is commonly used, while for low partial pressure CO₂ (flue gas) monoethanolamine (MEA) is used. Here the chemical absorption with amines is modeled as a blackbox considering the average energy demands for the separation given in [27] and summarized in Table 3. In order to account for the influence of the solvent on the efficiency and cost, a more detailed model would be required, which was considered out of the scope of this study. In [32] a more sophisticated methodology for developing energy and cost correlations of CO₂ capture processes is proposed. For the PSA model, the approach outlined in [13] is adapted for H₂ / CO₂ separation based on data from [17]. PSA yields H₂ purities of over 99%mol. The nominal operating conditions of the main process units, as well as their ranges are summarized in Table 4.

4 Process performance

4.1 Performance indicators

The first law energy efficiency ϵ_{tot} defined by Eq.2 characterizes the chemical conversion and the quality of the process integration by taking into account the energy of the products and resources. In this definition thermal and mechanical energy are considered as being equivalent, however with regard to energy quality they are not equal. Therefore, the natural gas (NG) equivalent efficiency ϵ_{eq} (Eq. 3) is introduced to compare the value of the products with respect to the technical feasibility of their further conversion into final energy services. In ϵ_{eq} , the net electricity that is consumed is substituted by a NG equivalent calculated based on an energy efficiency of 55%. The H₂ productivity is defined by the H₂ yield Y_{H_2} (Eq.5). The conversion efficiency ϵ_{H_2} (Eq.6) expresses the production of the H₂ fuel with regard to the biomass resource consumption, without taking into account the electricity import or export. The CO₂ capture rate is given by the molar ratio between the captured carbon and the carbon entering the system (Eq.7). The exergy efficiency ϵ_{ex} (Eq.4) is also computed with $\Delta k_{dry,Biomass}^o = 20.9 \text{ MJ/kg}$. All the reported efficiencies are expressed on the basis of the lower heating value (Δh^0 , LHV) of dry biomass.

$$\epsilon_{tot} = \frac{\Delta h_{fuel,out}^0 \cdot \dot{m}_{fuel,out} + \dot{E}^-}{\Delta h_{Biomass,in}^0 \cdot \dot{m}_{Biomass,in} + \dot{E}^+} \quad (2)$$

$$\epsilon_{eq} = \frac{\Delta h_{Fuel,out}^0 \cdot \dot{m}_{Fuel,out} + \frac{1}{\eta_{NGCC}} \frac{\Delta h_{NG}^0}{\Delta k_{NG}^0} (\frac{1}{\eta_{HP}} \dot{Q}^- + \dot{E}^-)}{\Delta h_{Biomass,in}^0 \cdot \dot{m}_{Biomass,in}} \quad (3)$$

$$\epsilon_{ex} = \frac{\Delta k_{fuel,out}^o \cdot \dot{m}_{fuel,out} + \dot{E}^-}{\Delta k_{Biomass,in}^o \cdot \dot{m}_{Biomass,in} + \dot{E}^+} \quad (4)$$

$$Y_{H_2} = \frac{g_{H_2}}{kg_{biomass}} \quad (5)$$

$$\epsilon_{H_2} = \frac{\Delta h_{H_2fuel}^0 \cdot \dot{m}_{H_2fuel}}{\Delta h_{Biomass,in}^0 \cdot \dot{m}_{Biomass,in}} \quad (6)$$

$$\eta_{CO_2} = \frac{molC_{captured}}{molC_{in}} \cdot 100 \quad (7)$$

The economic performance is defined by the capital investment and the operating cost estimated according to [34, 35] with the assumptions given in Table 5. All the performance analyses are performed for a plant capacity of $380MW_{th,biomass}$ of dry biomass.

4.2 Energy integration

Heat integration and recovery are important with regard to the process performance since several parts of the system operate at high temperature. The minimum energy requirement is computed from the hot and cold process streams through the heat cascade method accounting for the potential heat recovery. Heat is required by the gasification, the endothermic reforming, the water evaporation for gasification and the CO₂ capture. The exothermic WGS and the process and offgas cooling release heat. The heat demands can be satisfied by different utilities. High temperature heat is delivered by the combustion of waste streams (i.e. unconverted char and gaseous residues of the separation and purification sections) and if necessary additional process streams (i.e. hot or cold syngas from the gasifier) and depending on the production scope also by the burning of H₂-rich gas or pure H₂ in a gas turbine to co-produce electricity. In general, the best choice is determined by assembling the potential fuels in a superstructure, integrating the different possibilities and computing the optimal solution by minimizing the operating cost using a linear programming model [23]. In the linear programming problem a cost is attributed to the electricity import/export (i.e. [50-270\$/MWh]) and to the CO₂ emissions (i.e. 36\$/to_{CO2}, [15-90\$/to_{CO2}]). Surplus heat can be recovered in a Rankine cycle with an extraction steam turbine/generator to generate additional electricity and supply steam for gasification, steam methane reforming and shift conversion. A cycle with two production, two usage and one condensation level is considered and adapted to the different process configurations with regard to the parameters given in Table 6. The remaining excess heat is removed by cooling water. The process integration including hot and cold utilities for two different configurations (Table 7:A&B) producing H₂ is represented in Figure 3 and discussed in detail in Section 4.3.

4.3 Process integration analysis

The influence of the heat recovery and the cogeneration systems including the introduction of a steam network, gas turbines (GT) or heat pumps (HP) is analyzed with regard to H₂ and electricity production and captured CO₂. Table 7 summarizes the different process configurations and the computed performances.

4.3.1 H₂ production processes

For the process configurations producing H₂ by biomass conversion, different options with H₂ purification and/or carbon capture are considered. The performances are reported as configurations A-F in Table 7. The energy integration of the process with H₂ separation by PSA and without or with carbon capture by chemical absorption with amines is illustrated in Figure 3. Since the pinch point is located at low temperature, there is no excess heat that can be used in a Rankine cycle. By introducing a heat pump, excess heat from below the pinch can be transferred to a higher temperature for valorization in a Rankine cycle and consequently the energy integration of the CO₂ capture is improved as shown in Figure 4 (Table 7:C).

The influence of CO₂ capture is studied by the comparison of configurations B and C (Table 7). CO₂ capture increases the power consumption considerably due to the energy requirement for the solvent regeneration and the CO₂ compression. The purchase of the capture unit equipments increases the capital investment and consequently the production cost are increased by around one third. Through H₂ purification, the H₂ yield is increased by over 10% and the environmental impact is decreased because of the CO₂ storage. By performing a multi-objective optimization, it is shown in Section 4.4 how the performance can be improved further to reach an overall energy efficiency around 60% with CO₂ capture (Table 7:C_{opt}) by changing the operating conditions.

For these configurations electricity is imported to satisfy the overall process demands. Alternatively, part of the H₂ rich gas and/or H₂ product can be burnt in a gas turbine to cover the power demand and yield a self-sufficient process in terms of heat and power (Table 7:D). The energy integration of such a configuration is represented in Figure 5. The self-sufficient H₂ process has a lower H₂ yield, since part of the product is used for electricity production which leads to an energy efficiency decrease of more than 10% points. The equivalent efficiency ϵ_{eq} is however increased by around 10% points which shows that the integrated electricity production is more efficient than a NGCC plant. Due to the reduced electricity cost, the production cost are slightly reduced, even if the H₂ yield is reduced and the capital investment increased. However, for keeping a higher level of CO₂ capture in the process, the production cost would be larger (Table 8). In Sections 4.4&4.5, it is shown that the process can become more attractive by changing the operating conditions and that the economic competitiveness of this option depends highly on the electricity and fuel market prices and the CO₂ taxes.

The H₂ purification by PSA (Table 7:C) increases the H₂ purity by around 2.5% compared to the process without PSA (Table 7:E). The electricity demand and the investment are increased, however the overall impact on the performance is relatively low since the H₂ yield is increased.

4.3.2 Electricity only production processes

Instead of generating H₂ as a final product, electricity can also be produced by burning the H₂ gas products in a gas turbine combined cycle. Different configurations producing electricity as a final product are assessed (Table 7:G-J): electricity generation from nearly pure H₂ and electricity generation by the combustion of the H₂-rich stream after WGS without (config. G&I) and with carbon capture (config. H&J). H₂ purification and carbon capture adding additional cost, the configuration burning impure H₂ (lower LHV fuel) without CO₂ capture (config. I) yields the lowest investment cost. Burning pure H₂ (higher LHV fuel) generates more electricity in the gas turbine which outweighs the additional power consumption for H₂ purification and consequently yields a higher energy efficiency. However, there are still some concerns with regard to flame stability which have to be addressed for high purity H₂ combustion. CO₂ capture leads to a negative CO₂ balance for biomass based processes. It reduces the efficiency by around 10% and increases the production cost considerably. The difference in the energy integration for the electricity generation without and with CO₂ capture is reported in Figure 6&7 respectively. In these configurations, the energy demand is satisfied by the heat generated from the gas turbine and by the combustion of waste streams. The computed efficiencies are in the range of the

IBGCC power plant efficiency with and without CO₂ reported in [6, 8, 19]. In [19] efficiencies in the range of 37-44% are reported and in [6] an efficiency of 33.9% is assessed with CO₂ capture.

4.4 Process optimization

To investigate the trade-off between several competing factors defining the process performance, multi-objective optimization is performed by applying an evolutionary algorithm. The decision variables and their variation range are given in Tables 4 and 6 for the H₂ process and the steam network, respectively. The advantage of using an evolutionary algorithm like the one described in [25], is that it allows to generate the Pareto set, even if some of the simulated points do not converge and if the problem is non-differentiable.

Four scenarios are optimized: H₂ production with electricity import (config. C), self-sufficient H₂ production (config. D), electricity generation from nearly pure H₂ (config. H) and electricity generation from H₂-rich gas (config. J). First, the maximization of the energy efficiency ϵ_{tot} and the minimization of the capital investment are chosen as objectives. The objective functions are obtained by solving the thermo-economic model described in section 3.1 for each set of decision variables. The heuristics embedded in the sizing and cost estimation model guarantee that industrial infeasible solutions are avoided. The optimal Pareto frontiers are presented in Figure 8. The energy efficiency increase is correlated with the investment increase. For each scenario, the performance of one optimal configuration yielding a high ϵ_{tot} is reported in Table 7 (C_{opt}/D_{opt}/H_{opt}/J_{opt}). The energy integration and the main operating conditions of the optimal H₂ process designs are represented in Figure 10&11, respectively. Looking at the equivalent efficiency instead of ϵ_{tot} , the self-sufficient scenario (D_{opt}) performs better than the one with electricity import (C_{opt}). The optimization leads to better energy efficiency and lower cost when compared to the base case configurations. However, the CO₂ capture rate is lower and consequently the environmental benefit is less important. The trade-off between the energy efficiency and the CO₂ capture rate is highlighted by the optimal Pareto frontier resulting from the maximization of the energy efficiency and the CO₂ capture rate as reported in Figure 9. With regard to competitiveness, a compromise between the different objectives has to be found. The performance of selected optimal configurations yielding relative high efficiency and capture rates are reported in Table 8. Depending on the biomass and the electricity import/export prices, production cost can become lower as shown in section 4.5. Compared to fossil power plants (Tables 1& 2) with carbon capture the biomass conversion into electricity and H₂ reveals to be competitive.

4.5 Economic evaluation

The H₂ production costs assessed previously depend strongly on the economic assumptions made in Table 5. The sensitivity analysis varying the wood cost [10-70\$/MWh_{BM}] and the green electricity cost [40-270\$/MWh_e] shows the influence of the resource price on the competitiveness of the H₂ production in Figure 12 for the configurations yielding a compromise between efficiency and CO₂ capture (Table 8) and for the base case without capture (Table 7:B). With the initial assumption of 50\$/MWh_{BM} for biomass from Switzerland [13] up to 60% of the production cost are attributed to wood purchase, whereas a decrease of the resource cost can reduce this fraction to around 20% and reduce the H₂ production cost by nearly 50%. According to [16], biomass prices as low as 7.2\$₂₀₀₂/MWh can be expected for Latin and North America. Consequently, the competitiveness is highly influenced by the resource price and location. The assessed costs are still slightly higher than the one reported in [16] because of the higher investment cost, especially for the gasifier purchase. Contrary to their approach, the investment estimation method applied here rates the equipment with conventional design heuristics that take the operating conditions into account. As pilot plant data are used as reference for the design parameters of the gasifier, it can be expected to yield realistic figures. The conservative cost estimation might however

overestimate the investment and consequently lead to higher production cost. Nevertheless, these biomass based H_2 processes yielding efficiencies of 40-60% and production costs in the range of 65-262\$/MWh $_{H_2}$ can become a competitive option with regard to fossil resource depletion and climate change compared to conventional processes using fossil resources (Table 1). Considering a new hydrogen plant based on fossil resources [24] producing H_2 with an efficiency of 60%, production costs of 28\$/MWh $_{H_2}$ and CO_2 emissions of 493kg/MWh $_{H_2}$ as a reference, CO_2 avoidance cost ¹ in the range of 45-220\$/t $_{CO_2}$ are assessed for the analyzed biomass based processes. In comparison, CO_2 avoidance cost in the range of 2-56\$/t $_{CO_2}$ are assessed for fossil H_2 production processes with CO_2 capture in [24] .

For the electricity production processes without CO_2 capture (Table 7:G/I) and with CO_2 capture (Table 8) a sensitivity analysis on the wood cost [10-70\$/MWh $_{BM}$] (Figure 13) yields electricity production cost in the range of 89-362\$/MWh $_e$. Considering the low wood cost, the costs are in the range of the one reported in [19] ($\approx 150 - 220$ \$/MWh). Compared to fossil power plants (Table 2) some scenarios with CO_2 capture are promising regarding the future energy market, especially when high CO_2 taxes are imposed. Considering a NGCC plant [24] with an efficiency of 57%, production costs of 40\$/MWh $_e$ and CO_2 emissions of 360kg/MWh $_e$ as a reference, CO_2 avoidance cost in the range of 98-254\$/t $_{CO_2}$ are assessed for the analyzed biomass based processes. In comparison, CO_2 avoidance cost in the range of 37-74\$/t $_{CO_2}$ are assessed for an NGCC with CO_2 capture in [24]. By the sensitivity analyses, it is shown that the variation of the resource price of a factor of 7 translates in a production cost increase of a factor of 4 due to the high weight of resource purchase in the total cost. Consequently, the resource price and location define the competitiveness of the biomass conversion processes. The analyses also highlight the importance of the process efficiency. The market price of electricity, fuel, biomass and CO_2 taxes will define if it is more advantageous to produce H_2 with or without electricity import as final product or to convert the H_2 fuel directly into electricity with or without CO_2 capture.

4.6 Environmental impacts

For the life cycle inventory, the method from the IPCC (IPCC07) is applied for the LCI flows identified in Figure 2. Following the approach of Gerber et al. [15], 1kJ of biomass entering the plant is considered as functional unit in order to make a consistent comparison of H_2 and electricity production scenarios. The data available from the ecoinvent database [9] are used for the different contributions. For the electricity impact contribution, the Swiss mix for medium voltage electricity production at grid is considered. The amount of CO_2 that is stored is accounted as a negative contribution of fossil CO_2 .

Regarding the climate change impact of H_2 processes, Figure 14 shows the advantage of CO_2 capture. CO_2 capture for storage has a negative contribution (n) to the climate change impact which outweighs all the other positive contributions (p).

For the electricity generation processes, the benefit of CO_2 capture on the climate change is highlighted in Figure 15. A large impact is attributed to the use of rape methyl ester (RME) produced from colza cultivated with insecticides and consumed for the cold gas cleaning, consequently alternative colza cultivation methods and the development of alternative cleaning technologies such as hot gas cleaning have to be analyzed.

With regard to CO_2 emissions mitigation, processes based on renewable biomass have a huge potential, especially if CO_2 capture and storage is implemented since this leads to a negative CO_2 balance.

¹ CO_2 avoidance cost expressed in \$/t $_{CO_2,avoided}$ are defined by the ratio of the difference of the production cost and the CO_2 emissions for a plant with capture and a reference plant without capture: $\frac{COE_{CC} - COE_{ref} [\$/MWh]}{CO_{2,emit,ref} - CO_{2,emit,CC} [kgCO_2/MWh]}$. For the biomass based plants, the CO_2 emissions equal to the negative value of the CO_2 captured since it is removed from the atmosphere.

5 Conclusion

A systematic methodology based on thermo-economic and LCI models coupled with a multi-objective optimization algorithm has been applied to the conceptual design of integrated plants for H₂ fuel, power and heat production. The competitiveness of H₂ and electricity production and co-production process options are evaluated consistently with respect to energy efficiency, cost and environmental impacts. It is highlighted in particular, how appropriate energy integration and operating conditions optimization improve the process performance by maximizing the combined production of fuel, heat and power. Based on multi-objective optimizations with regard to energy efficiency and capital investment or CO₂ capture rate, the trade-off between H₂ and electricity generation and CO₂ capture are assessed. Life cycle impact assessment underlined the climate change benefit of using renewable resources and capturing CO₂. Overall energy efficiencies in the range of 60% are reached for H₂ production and around 39% for electricity production with CO₂ capture. Depending on the biomass price evolution, H₂ production costs in the range of 65-262\$/MWh_{H₂} and electricity production costs in the range of 89-362\$/MWh_e are reported. With regard to conventional H₂ and electricity production processes based on fossil resources, CO₂ avoidance costs of 45-220\$/t_{CO₂,avoided} and 98-254\$/t_{CO₂,avoided} respectively, are computed. In comparison, the performances assessed in [24] for processes using fossil resources are for a NGCC plant 43-72\$/MWh_e and 37-74\$/t_{CO₂,avoided}, and for H₂ plants 27-48\$/MWh_{H₂} and 2-56\$/t_{CO₂,avoided}. The market price of electricity, fuel, biomass and CO₂ taxes will consequently define the competitiveness of biomass conversion into H₂ or electricity with or without CO₂ capture. With regard to a future energy system promoting renewable resources and reduced greenhouse gas emissions, biomass based H₂ and electricity production have to be considered as competitive alternatives.

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Table 1: Reference H₂ production plants performance.

Process	CO ₂ capture [%]	ϵ [%]	[\$/MWh _{H2}]	Ref.
Natural gas	0	83.9 _(HHV)	18.9	[18]
Natural gas	71	78.6 _(HHV)	20.2	[18]
Coal (Texaco gasif.)	0	63.7 _(HHV)	31.6	[3]
Coal (Texaco gasif.)	87	59 _(HHV)	37.8	[3]
Biomass (FICFB, CGC)	-	57.7	-	[33]
Biomass	-	51-60	29-40	[16]

Table 2: Reference power plants performance without and with CO₂ capture.

Type	CO ₂ capture [%]	ϵ [%]	gCO ₂ /kWh _e	COE [\$ /MWh _e]	\$/t CO _{2,avoided}	Ref.
PC	0	41-45	736-811	43-52	-	[24]
PC -CC	85-90	30-35	92-145	62-86	29-51	[24]
IGCC	0	38-47	682-846	41-61	-	[24]
IGCC -CC	85-90	31-40	65-152	54-79	13-37	[24]
NGCC	0	55-58	344-379	31-50	-	[24]
NGCC -CC	85-90	47-50	40-66	43-72	37-74	[24]
IBGCC	0	37 _(HHV)	-	67.5	-	[21]
IBGCC -CC	80	33.94	178	-	-	[6]

Table 3: Parameters for the H₂ purification.

Section	Specification	Value
Chem. abs.	Thermal \dot{Q} @ 150°C	3.7MJ/kg CO ₂
	Electric Power	1.0MJ/kg CO ₂
PSA	Adsorption P	10bar
	Purging P	0.1bar
	H ₂ recovery	90%

Table 4: Operating conditions of the process units and feasible range for optimization.

Operating parameter	Nominal	Range
Drying		
Temperature (in) [K]	473	-
$\theta_{wood,out}$ [wt%]	20	[5-35]
Gasification		
Pressure [bar]	1	[1-15]
Temperature [K]	1123	[1000-1200]
Steam/biomass [%wt]	50	-
SMR		
Temperature [K]	1138	[950-1200]
WGS		
Pressure [bar]	25	[1-25]
Temperature HTS [K]	623	[573-683]
Temperature LTS [K]	453	[473-573]
H ₂ O/CO molar ratio [-]	2	[0.2-4]

Table 5: Assumptions for the economic analysis.

Parameter	Value
Marshall and Swift Index	1473.3
Dollar exchange rate (€-US\$)	1.5 US\$/€
Expected lifetime	15 years
Interest rate	6%
Yearly operation	8000h/year
Operators [13]	4 p./shift
Operator's salary	91'070 \$ /year
Wood costs ($\theta_{wood}=50\%$ wt)	50 \$ /MWh
Electricity price (green)	270 \$ /MWh

Table 6: Feasible range for optimization of the steam network design and the gas turbine.

Operating parameter	Unit	Range
1st Production level	bar	[90-130]
2nd Production level	bar	[70-110]
Superheating temperature	K	[623-823]
1st Utilization level	K	[300-523]
2nd Utilization level	K	510
Condensation level	K	292
Combustion T	K	[700-900]
Turbine T	K	1500

Table 7: Investigated process configurations characteristics.

Configuration ^a	H ₂ Process						Electricity generation						
	A	B	C		D	E	F	G	H	I		J	J _{opt}
	Process Parameters						Process Parameters						
Products	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂	H ₂	E	E	E	E	E	E
Carbon capture [%]	y	n	y	y	y	y	y	n	y	y	n	y	y
PSA	y	y	y	y	y	y	n	n	y	y	n	n	n
PG burning	y	y	y	y	y	n	y	n	n	n	n	n	n
GT H ₂ impure	n	n	n	n	n	y	n	n	n	n	y	y	y
GT H ₂ pure	n	n	n	n	n	n	n	n	y	y	y	n	n
HP	n	n	387/478K	387/478K	387/478K	387/478K	n	387/478K	n	387/478K	387/478K	n	387/478K
Steam network:	n	n	y	y	y	y	y	y	y	y	y	y	y
Production levels [bar]	-	-	140	135&95	125	128	93	125&93	125&93	125&93	105&60	125&93	125&93
Consumption levels [K]	-	-	513&473	505&434	473&300	483&430	300	513&300	513&300	513&300	428&300	513&300	510&300
	Power Balance						Power Balance						
Consumption [kW/MW]	136.7	72.5	169.7	153.3	120.6	119.6	127.5	100.8	91.8	221.5	102.8	89.5	176.6
HP [kW/MW]	0	0	40.4	35.9	27.6	8.1	35.9	28.1	0	53.1	0.3	0	53.3
Steam network [kW/MW]	0	0	25.7	10.1	37.7	29.3	31.2	28.1	115.6	132.6	108.2	100.4	113.9
Gas turbine [kW/MW]	9.1	28.9	9.9	30.4	110.5	98.4	6.0	100.8	425.5	414.7	389.3	338.8	307.9
Net electricity [kW/MW]	-127.6	-43.6	-174.5	-148.7	0	0	-126.2	0	449.3	272.7	394.4	349.7	191.9
	Performance						Performance						
H ₂ yield [g/kg _{Biomass}]	79.8	92.4	102.9	107.1	68.7	83.8	96.7	72.2	0	0	0	0	0
H ₂ purity [mol%]	99.8	80.7	99.8	99.3	99.8	98.7	97.4	97.4	80.7	99.8	98.7	69.1	97.4
H ₂ production [t/d]	140.7	162.8	181.4	188.7	121.1	147.7	170.5	127.3	0	0	0	0	0
Energy H ₂ [kW/MW]	514.3	596.9	663	696.5	442.2	550.1	626.9	468.1	0	0	0	0	0
CO ₂ capture [%]	44.9	0	57.9	52.3	38.6	25	53.3	39.8	0	78.2	22.4	0	78.2
gCO ₂ captured/kWh	316	0	316	271	316	164	308	307	0	1037	205	0	1474
ϵ_{tot} [%]	45.6	57.2	56.4	60.6	44.2	55	55.6	46.8	44.9	27.3	39.4	34.9	19.2
ϵ_{eq} [%]	29.1	52.1	35.8	43.7	44.2	55	40.1	46.8	-	-	-	-	-
ϵ_{H_2} [%]	51.4	59.7	66.3	69.6	44.2	55	62.7	46.8	-	-	-	-	-
ϵ_{ex} [%]	39.9	49.7	49.6	53.1	38.2	47.5	48.7	40.4	39.9	24.3	35.1	31.1	17.1
	Economics						Economics						
Investment [M\$]	424	421	525	401	555	361	461	511	521	600	416	446	529
Annualized Inv. [\$/MWh]	27.9	23.9	26.8	19.5	42.5	22.2	24.9	36.9	39.2	74.5	35.7	43.2	93.4
Maintenance [\$/MWh]	21.7	18.7	19.4	15.5	30.2	18.5	18.8	26.9	28.5	51.6	28.0	33.1	67.4
Wood cost [\$/MWh]	95.7	82.4	74.2	70.6	111.3	89.4	78.5	105.1	109.5	180.4	124.8	140.7	256.3
Electricity cost [\$/MWh]	65.9	19.4	69.9	42.5	0	0	53.5	0	0	0	0	0	0
Production cost [\$/MWh]	211.2	144.4	190.3	148.1	184	130.1	175.7	169	177.2	306.5	188.5	217	417
	Environmental Impact (GWP 100)						Environmental Impact (GWP 100)						
10 ⁻⁹ kg CO _{2,eq} /kJ _{BM}	-3.5	0.98	-4.7	-4.2	-3.0	-1.5	-4.3	-3.1	0.8	-6.9	-1.44	0.8	-6.8

^a For the different technology options: n=not included, y=yes included. The net electricity output expressed in kW of electricity per MW of biomass is negative if it generates electricity. For H₂ processes the costs are expressed in \$/MWh_{H₂} while for electricity generation they are expressed in \$/MWh_e.

Table 8: Process performance of selected optimal configurations.

Process	H ₂ \dot{E} import	H ₂ self-sufficient	\dot{E} GT H2 pure	\dot{E} GT H2 impure
ϵ_{tot} [%]	60.6	40.2	32.5	27.5
CO ₂ capture [%]	65.1	74.4	75.2	56.4
gCO ₂ captured/kWh	324	668	835	741
10 ⁻⁵ kg CO _{2eq} /kJ _{BM}	-5.5	-6.4	-6.6	-4.8
Investment [M\$]	509	651	777	532
Production cost [\$/MWh]	129.7	214.1	284.3	291.3

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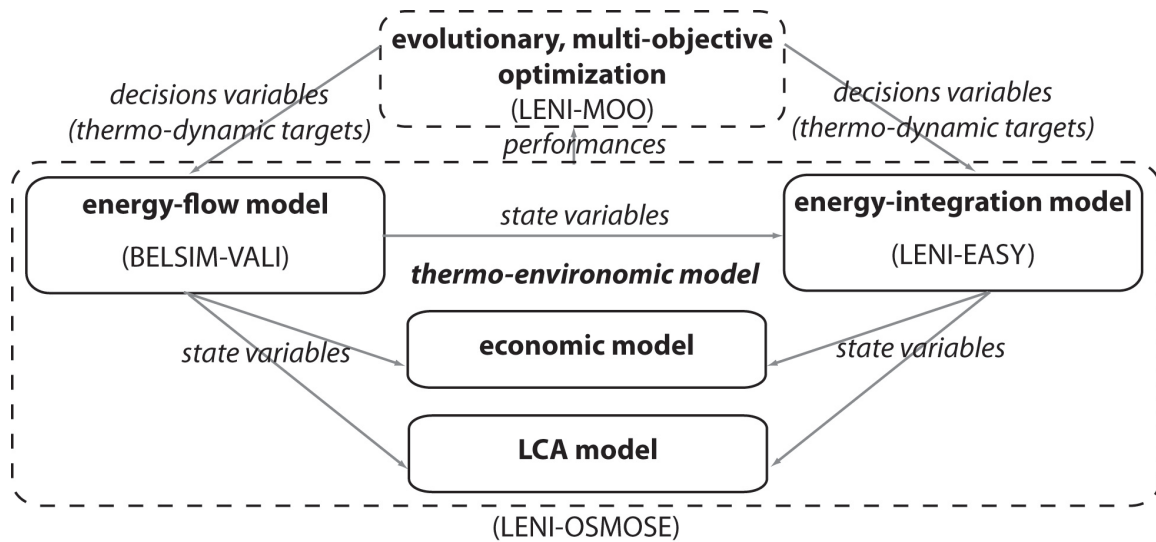


Figure 1: Design methodology: Thermo-vironomic optimization [15]

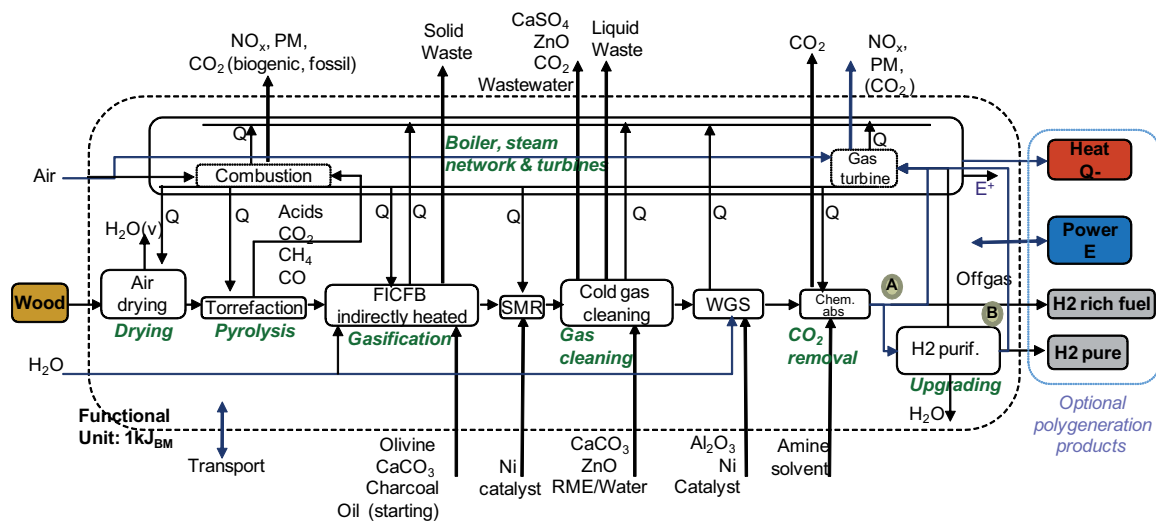


Figure 2: Flowsheet of the investigated biomass conversion processes with recycling options including thermo-economic and LCI model flows. The cross points A and B illustrate the different options with regard to H₂ and/or electricity generation.

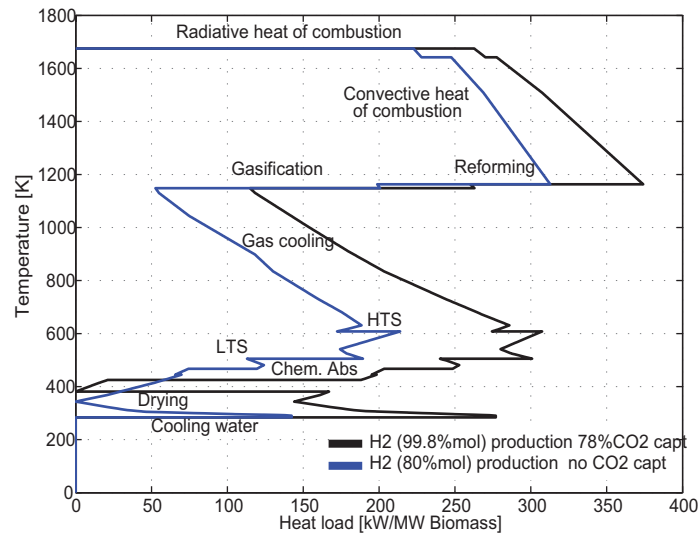


Figure 3: Integrated composite curves for H₂ process with and without CO₂ capture (Table 7:A& B).

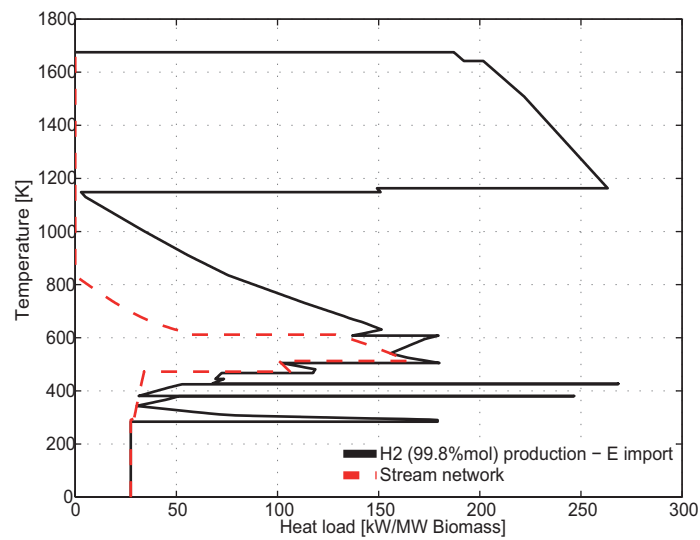


Figure 4: Integrated composite curve for a base case H₂ process with net \dot{E} import and with steam network integration (config. C).

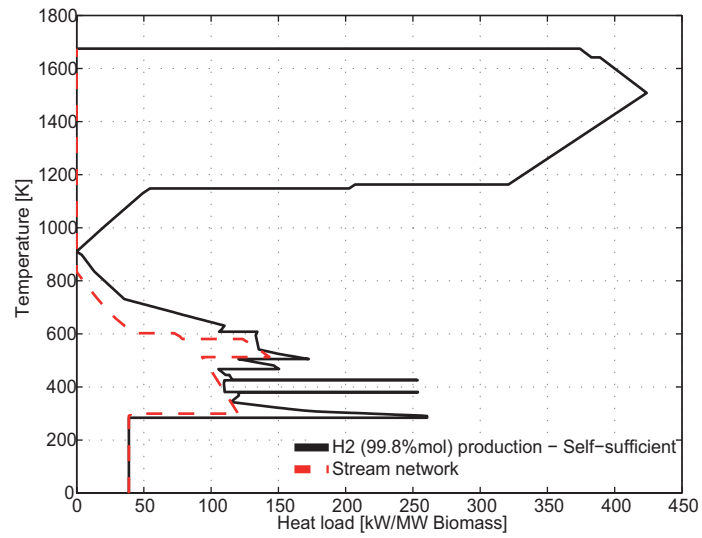


Figure 5: Integrated composite curve for a self-sufficient H_2 process with steam network integration (config. D).

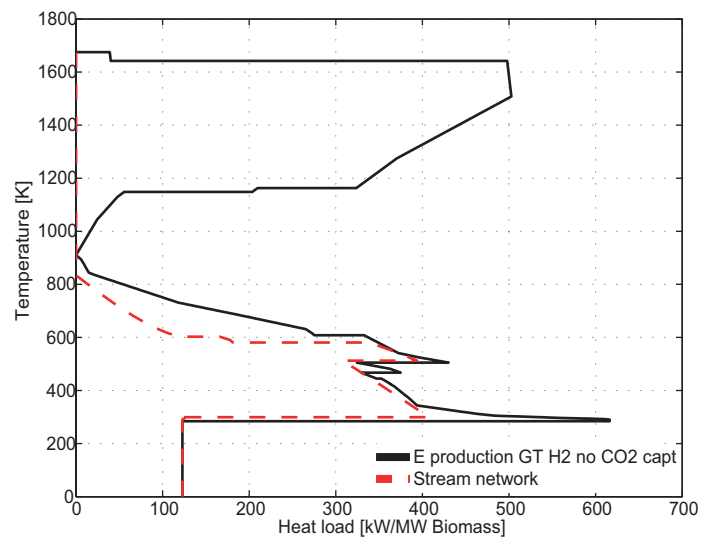


Figure 6: Integrated composite curve for the \dot{E} production without CO_2 capture and with steam network integration (config. G).

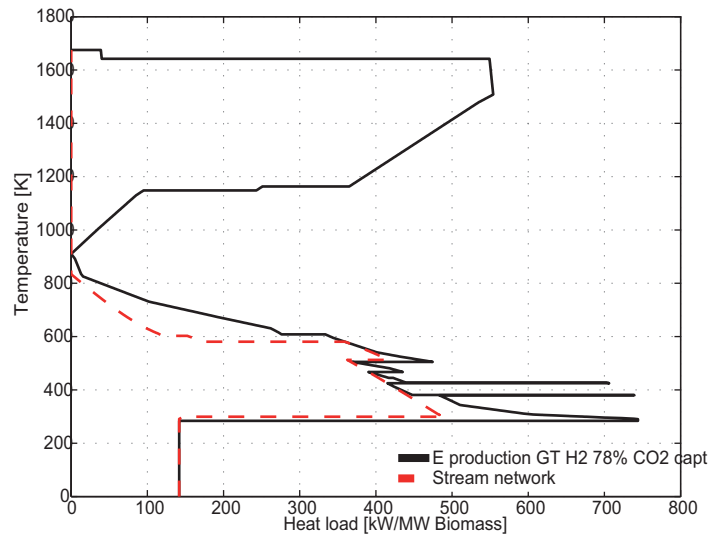


Figure 7: Integrated composite curve for the \dot{E} production with CO_2 capture and with steam network integration (config. H).

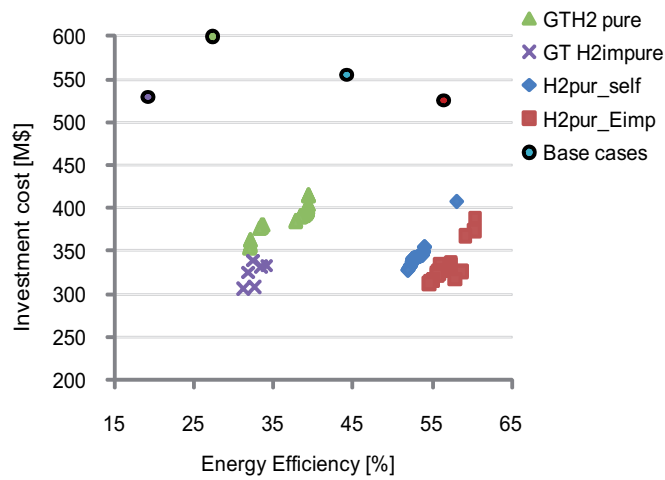


Figure 8: Optimal solutions in the Pareto domain for H_2 processes (config. C& D) and \dot{E} generation processes (config. I& H) with regard to efficiency and investment.

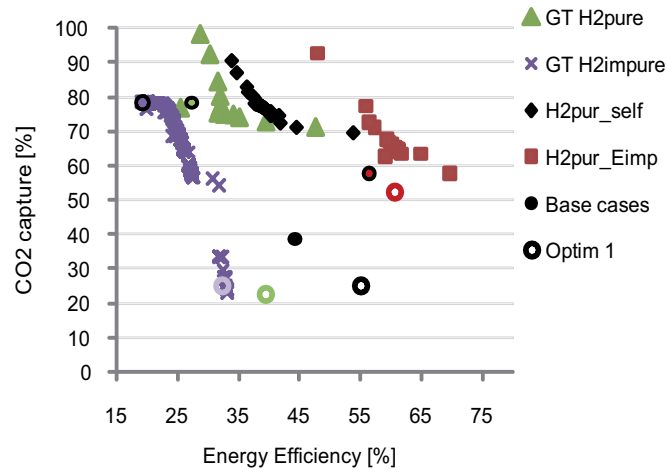


Figure 9: Optimal solutions in the Pareto domain for H₂ processes (config. C& D) and \dot{E} generation processes (config. I& H) with regard to efficiency and CO₂ capture rate.

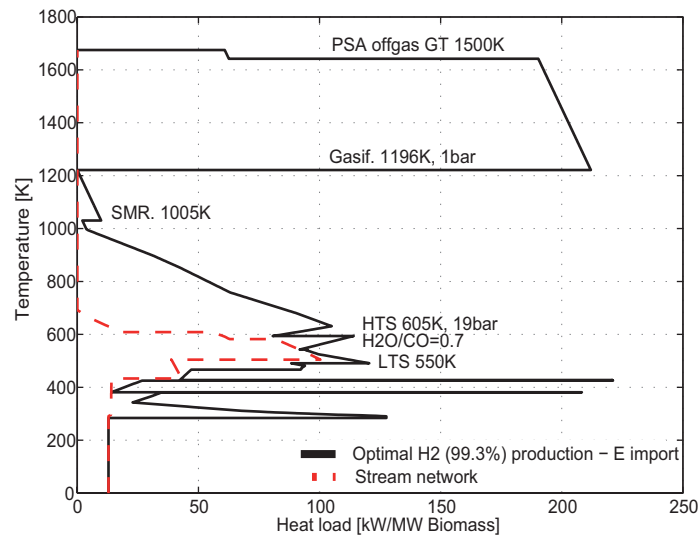


Figure 10: Integrated composite curve for an optimized H₂ process with net \dot{E} import and steam network integration (config. C_{opt}).

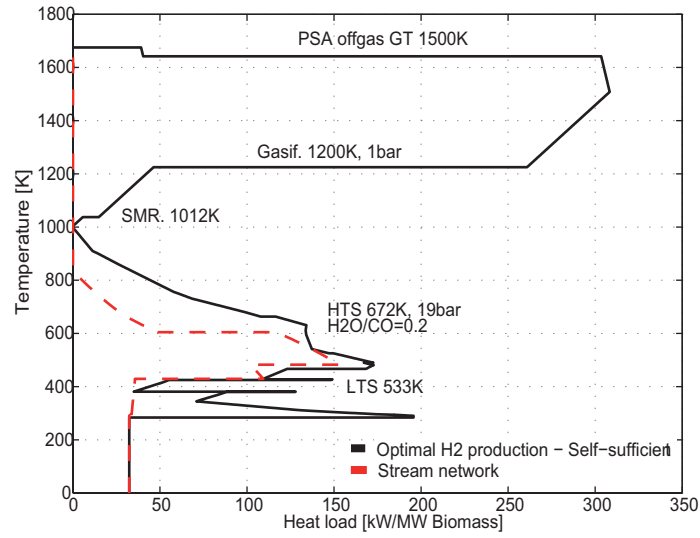


Figure 11: Integrated composite curve for an optimized self-sufficient H₂ process with steam network integration (config. D_{opt}).

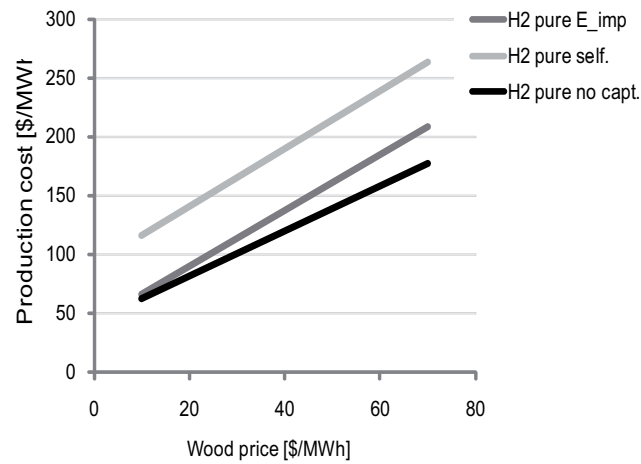


Figure 12: Sensitivity analysis of the wood cost on the hydrogen production cost [\$/MWh_{H₂}] for different H₂ scenarios (Table 7:B & Table 8).

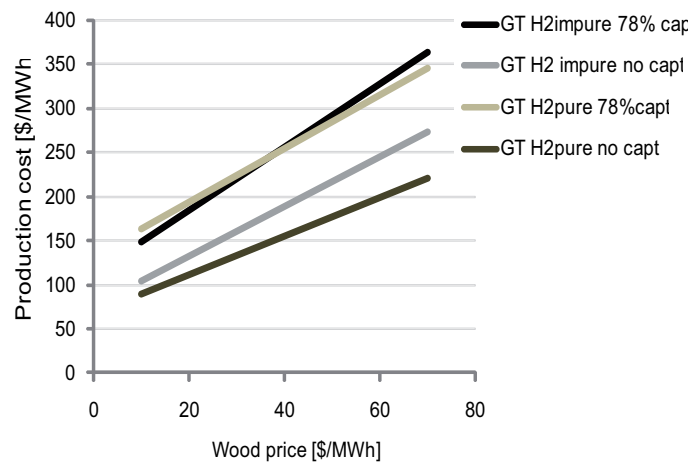


Figure 13: Sensitivity analysis of the wood cost on the electricity generation cost [\$/MWh_e] for different scenarios (Table 7:G/I & Table 8).

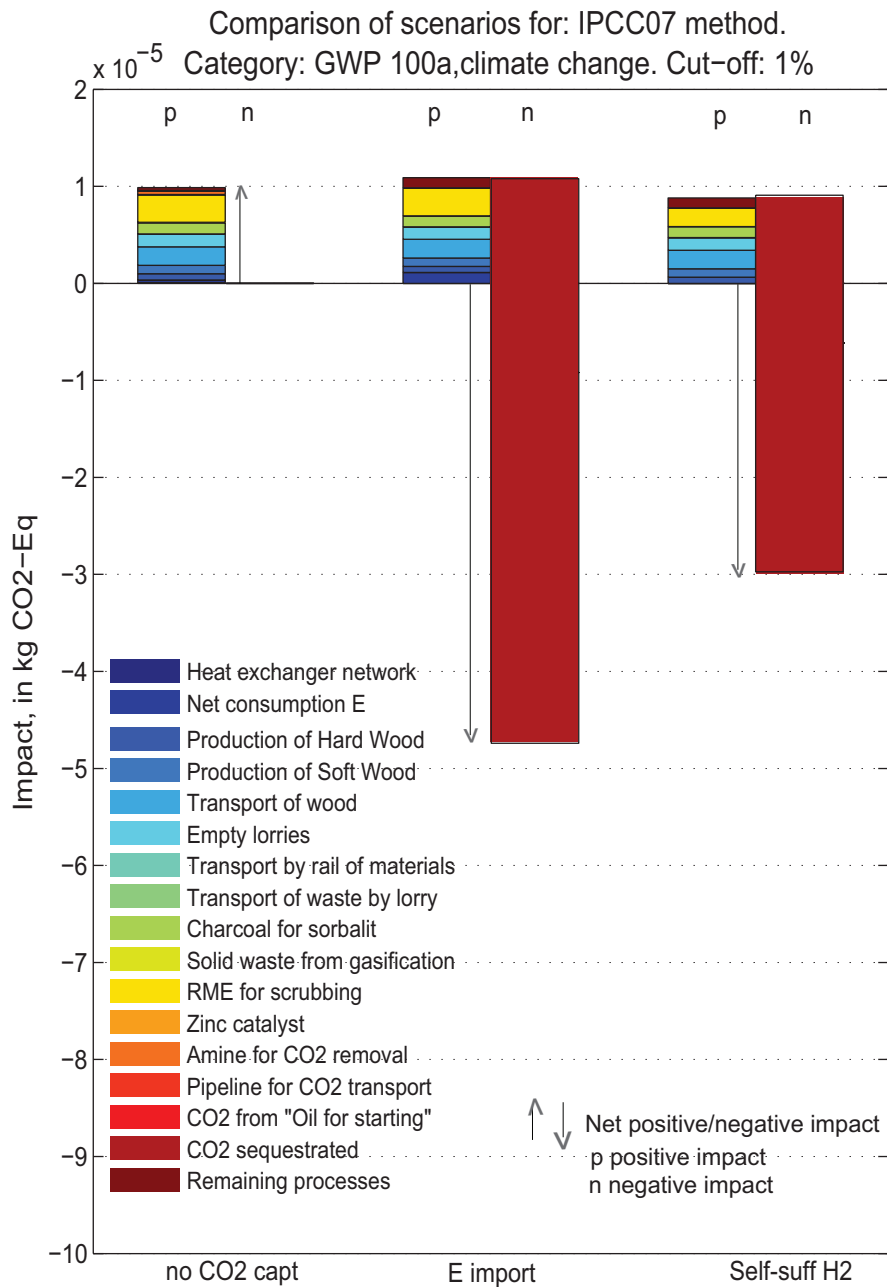


Figure 14: Comparison of the climate change impact of the H₂ generation processes (Table 7:B/C/D) based on impact method IPCC07 for 1kJ of biomass.

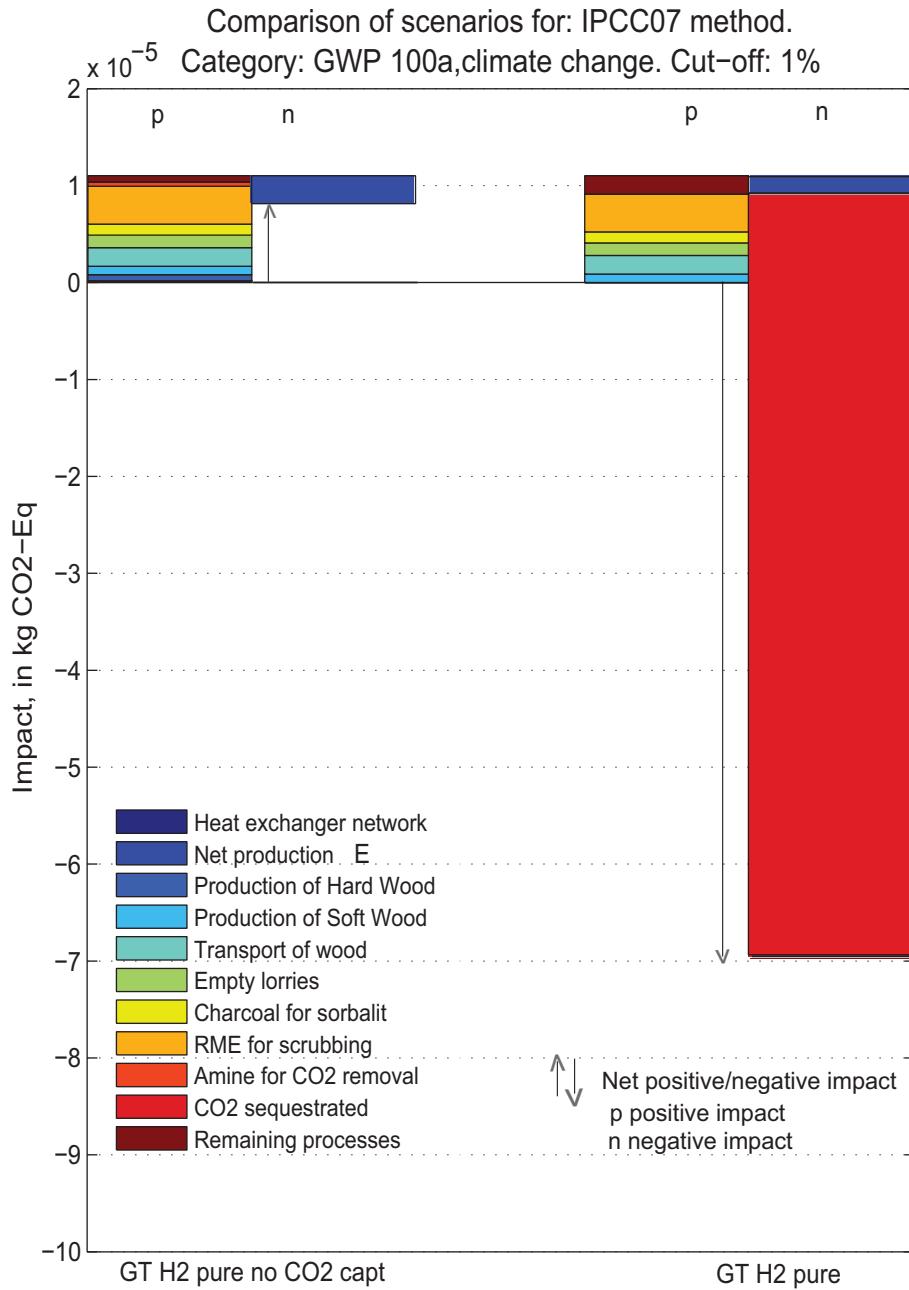


Figure 15: Comparison of the climate change impact of the electricity generation processes (Table 7:G-H) based on impact method IPCC07 for 1kJ of biomass.