

Sustainability Analysis of Hydrogen Production Processes: a Comparison Based on Sustainability Indicators

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Hydrogen is a versatile energy carrier and storage medium that may be employed in a variety of applications. It may be produced using different processes. In this work, process simulation is used to obtain material and energy balances for each process investigated, as well as for the evaluation of capital and maintenance costs. Process simulation outcomes are then used to estimate three key performance indicators focusing on sustainability issues: the energy return of energy invested, the levelized cost of hydrogen and the life cycle assessment. We compared several hydrogen generation processes, each denoted by a unique colour code: (i) green hydrogen, produced by electrolysis of water using electricity from renewable sources, (ii) grid hydrogen, produced by electrolysis using grid electricity, (iii) grey hydrogen, produced from natural gas using steam reforming and (iv) blue hydrogen, like grey one, but coupled with carbon capture and storage. In conclusion, the most sustainable hydrogen production method is the green hydrogen, produced by water electrolysis.

1. Introduction

Human energy use has increased rapidly over the last few decades, putting growing strain on the energy industry. Recent reports of the Intergovernmental Panel for Climate Change (IPCC) and the International Energy Agency (IEA) indicate that a strong reduction in the usage of fossil fuels must be achieved in order to meet the 2030 greenhouse gases emission targets. Since energy production from renewable sources is not dispatchable due to their fluctuating nature, great attention is given to energy storage and energy carrier systems. Hydrogen is one of the most suitable energy carriers for several applications, mainly in heavy transportation and logistics. There are several indicators available in the literature that are commonly employed by decision-makers. One of them is the Energy Return on Energy Invested (EROEI, Hall et al. 2014), that relates the amount of net energy stored in the hydrogen produced to the total invested energy to produce it. It has recently been proposed as a benchmark tool by IEA in the guideline methodology for the net energy analysis.

Another key performance indicator, specific for hydrogen, is the Levelized Cost of Hydrogen (LCOH), which considers the cost of hydrogen production process and is calculated as the ratio between the net discounted costs over the amount of produced hydrogen (Minutillo et al, 2021). Inputs to LCOH include cost of capital, investment costs and plant lifetime. These two indicators focus on hydrogen productions in terms of energy consumption and economic analysis, however, they are insufficient for determining the overall environmental effect of the activities under consideration. We carried out a complete Life Cycle Assessment (LCA) study to achieve this goal (Barbera et al., 2022).

This paper aims at providing reliable data for different hydrogen production processes at an early process design stage. Process simulations are used for the estimation of the performance indicators of the following hydrogen production processes: (i) green hydrogen, produced by electrolysis of water using electricity from renewable source, (ii) grid hydrogen, produced by electrolysis using grid electricity, (iii) grey hydrogen, produced from

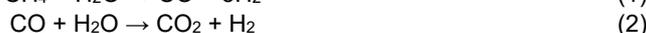
natural gas using steam reforming, and (iv) blue hydrogen, like grey one, but with carbon capture and storage (CCS). The processes considered are normalized for a hydrogen production of 1000 kg/h.

2. Materials and methods

Process simulation with Aspen Plus v.12 was used to perform material and energy balances, physical property estimations, design/rating calculations, process optimization, heat integration and economic analysis for a given hydrogen production process. Each simulation model has been implemented in terms of thermodynamics, chemistry, physical properties and unit operations and was finally optimized (Petrescu et al. 2021). The following chapters focus on the simulated processes, including evaluation of the performance indicators.

2.1 Grey hydrogen: methane steam reforming production process.

Grey hydrogen identifies H₂ derived from natural gas using steam methane reforming (SMR), which at present represents the main pathway used to fulfil the global H₂ demand. The corresponding Aspen Plus flowsheet is shown in Figure 1. The natural gas feed first undergoes a hydro-desulfurization treatment to remove sulfur, which is poisonous for the subsequent reformer catalyst. After sulfur removal, natural gas is reacted with steam in a primary reformer (PREF-T) and subsequently in a secondary reformer (SREF-R), where hot compressed air is also added. The main catalytic reactions occurring in the reformers, which operate at pressures around 30 bar and temperatures between 500°C and 1200°C, are the following:



After the reformers, the hydrogen yield is increased by means of water-gas shift (WGS), where reaction (2) is carried out. A higher-temperature WGS reactor (T= 380-460°C) is followed by a lower temperature (T = 210-270°C) one. Finally, the product hydrogen is purified in the unit H-PUR, which simulates a PSA unit (H₂ purity of 100% is assumed). The OUT-GAS stream is burnt in a furnace (modelled as RSTOICH reactor) with additional natural gas to provide the heat duty required by the reformers. Rigorous kinetics were implemented by means of Fortran subroutines for both reformers and WGS reactors, both of which were modelled as plug-flow reactors.

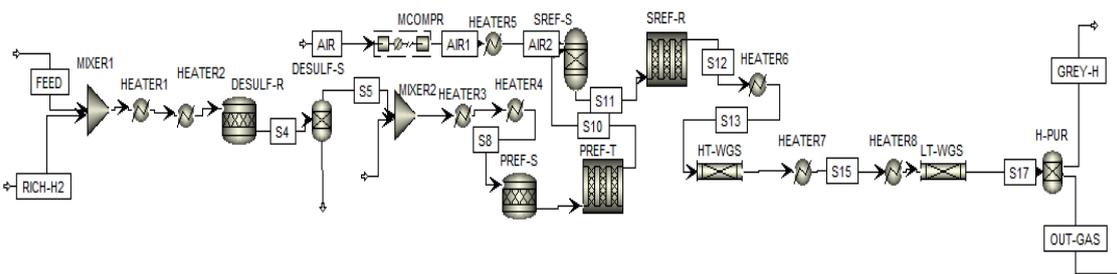


Figure 1. Aspen Plus flowsheet of the methane steam reforming process.

2.2 Blue Hydrogen: methane steam reforming with CCS

Blue hydrogen indicates H₂ produced by SMR coupled with Carbon Capture and Storage (CCS) to reduce the amount of associated CO₂ emissions. Several technologies are being developed for CCS from gaseous streams, however the most mature one today is represented by chemical absorption with amines. Accordingly, a CCS process was designed to capture CO₂ from the OUT-GAS stream derived from the SMR simulation, using a diglycolamine (DGA) solution. The process flowsheet developed in Aspen Plus is shown in Figure 2a. The OUT-GAS stream (45 mol% CO₂), at 55°C and 1 bar, is fed to the bottom of a packed absorption column, counter-current to the lean amine solution (LEANIN). To avoid excessive losses of DGA with the clean gases exiting from the top of the absorber, a washing section is added at the top of the column, where water is recirculated. The absorber (with height H = 20 m and diameter D = 4 m) captures 82% of the CO₂ from the inlet gases. The rich solution, after being heated up to a temperature T = 106°C, is regenerated by means of a reboiled stripper, operating at 2 bar, whose heat duty is satisfied exploiting the SMR flue gases. The CO₂ is recovered from the top of the column, after gas-liquid separation at a temperature of T=15°C (HX4+FLASH1), at 99 mol% purity.

2.3 Green Hydrogen: water electrolysis from renewables

Water electrolysis (WE) consists in the splitting of water into hydrogen and oxygen by applying an electrical voltage. Different types of electrolysis cells exist based on the type of electrolyte employed, namely Alkaline Electrolysis Cells (AEC), Polymer Electrolyte Membrane Electrolysis Cells (PEMEC), and Solid Oxide Electrolysis cells (SOEC) (Buttler and Spliethoff, 2018). Currently, AEC represents the most mature technology,

already employed at large scale. For this reason, it was chosen as representative for green hydrogen production in this work. The process flowsheet developed in Aspen Plus is shown in Figure 2b.

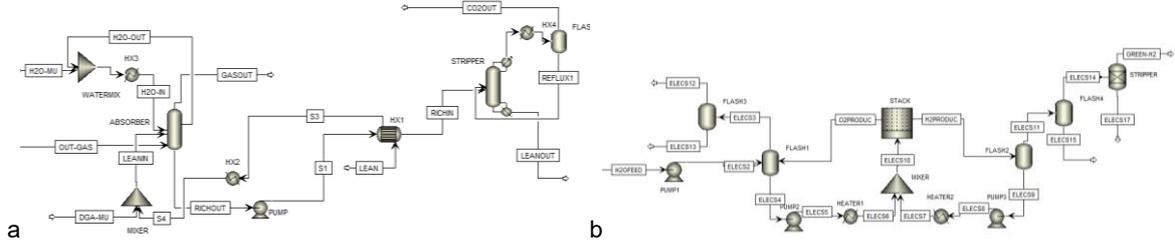


Figure 2. Aspen Plus flowsheet of the CCS unit (a) and of the alkaline water electrolysis process (b).

Since electrolytic cells are not implemented in Aspen Plus, a user defined unit model coupled to Excel calculator was used to insert the model equations for the stack. Specifically, material and energy balances were calculated according to the model proposed by Sánchez et al. (2020) which includes semi-empirical equations to describe the cell voltage, Faraday efficiency, and gas purity as a function of operating temperature, pressure, and current density. An aqueous solution of KOH (35 wt%) is fed to the cell stack. The oxygen produced at the anode and the hydrogen produced at the cathode are then led with the electrolyte to a series of flash vessels for gas-liquid separation. The electrolyte recovered in FLASH1 and FLASH2 is recycled, after make-up of water (stream H2OFEED), while the produced H₂ is further purified to remove moisture.

2.4 Grid hydrogen: water electrolysis using electricity from the grid

Grid hydrogen is produced through electrolysis using electricity from the electrical grid. For this study, the electricity mix of Italy is considered: renewables (45.04%), nuclear (3.22%), coal (6.34%) natural gas (42.28%) and others (2.64%).

2.5 Energy return on energy invested

Several methods and indices can be used to assess the efficiency of a production process involving the generation of energy carriers (electricity and/or hydrogen), but the best method for comparing different energy production industries is Net Energy Analysis (NEA). The goal of NEA is to calculate whether the energy produced by any production process is greater than the energy required to build, operate and maintain the infrastructure. Among the possible indexes derived from NEA, the most suitable indicator for the processes of interest is the EROEI defined as:

$$EROEI = E_{out}/E_{in} \quad (3)$$

where E_{out} is the available electrical energy that the process provides, which for hydrogen is the energy stored in a given quantity of hydrogen, and is defined as follows:

$$E_{out} = Q_{H_2} \cdot HHV \cdot (1 - Aux) \cdot cf \cdot L = P \cdot cf \cdot L \quad (4)$$

where Q_{H_2} (kg/h) is the mass flow rate of hydrogen produced, HHV (kWh/kg) is the higher heating value of hydrogen, Aux is the % of energy used for auxiliaries in the production process, cf is the capacity factor, L (years) is the plant life time and P (kW) is the net power output.

E_{in} is the total energy that is provided and consumed during the production and operations periods of the plant and is made up of three contributions: E_{cap} is the capital energy embodied in the materials and used for construction and decommissioning of the plant; $E_{o\&m}$ is the energy needed for operating and maintaining the power plant; E_f is the energy needed for procuring and distributing the fuels, which includes also the energy used for extracting, refining and transporting the fuels from the production well to the power plant.

$$E_{in} = E_{cap} + E_{o\&m} + E_f \quad (5)$$

In Eq. (3) All terms are expressed in GWh for consistency: the EROEI is thus dimensionless.

The capital energy embodied in the materials and used for construction and decommissioning of the plant E_{cap} is defined as follows:

$$E_{cap} = TPC/\varepsilon_c \quad (6)$$

where TPC is the total plant cost, related to CAPEX, and ε_c is the proportionality coefficient between the costs of energy and capital costs [€/kWh]. ε_c is evaluated from real plant data (Barbera et al. 2022).

The energy needed for operating and maintaining the power plant $E_{o\&m}$ is defined as:

$$E_{o\&m} = E_{cap} \cdot L \cdot s_{o\&m} \quad (7)$$

where $s_{o\&m}$ is the share of the investment costs dedicated to operation and maintenance, related to OPEX.

The energy needed for procuring and distributing the fuels, E_f , is defined as:

$$E_f = E_{out}/(\eta \cdot EROEI_{fuel}) \quad (8)$$

In Eq. (8) the term $EROEI_{fuel}$ refers to the energy used to extract, store, refine and transport the fuel (Hall et al., 2014), while the other terms are directly related to the plant dimensions and operations. Literature values of $EROEI_{fuel}$ from different sources are reported in Table 1. They consider all the boundaries of various types of EROEI analyses and the energy losses associated with the processing of fuel as it is transformed from “fuel at the wellhead” to consumer-ready fuels.

Table 1: Literature values of $EROEI_{fuel}$ for different resources

Resource	Rana et al. 2020	King et al. 2018	Hall et al. 2014
Wind	16.7 – 17.7	5 – 18	18
Solar photovoltaic	5 – 34	4 – 25	6 – 12
Hydro		59 – 84	>100
Nuclear		14	5 – 15
Coal		46	27 – 80
Oil		19	11 – 65
Natural gas		19	20 – 67

When a CCS plant is considered for treating the emission of the production plant, both E_{out} and E_{in} need to be modified to account for the energy consumed in the CCS process. E_{out} is reduced due to two effects: (i) the higher consumption of energy for auxiliary power, due to the electrical energy used for pumping and auxiliary work in the CCS plant and (ii) the thermal energy directly used in the reboiler of the stripping column. Information on both of these comes from the process simulation of the CCS plant. E_{out} and E_{th} are calculated according to Eq. (4) and (5). E_{in} and its components E_{cap} , $E_{o\&m}$ and E_f are calculated according to Eq. (6), (7) and (8) defined above, but the values of the independent variables are now estimated according to the results obtained from the simulation of the CCS processes.

2.6 Levelized Cost of Hydrogen

The levelized cost of hydrogen is calculated from Fan et al. (2022):

$$LCOH = \frac{COST_{Initial} + \sum_{t=1}^N \frac{COST_t}{(1+r)^t}}{\sum_{t=1}^N \frac{Q_{ht}}{(1+r)^t}} \quad (9)$$

Where $COST_{Initial}$ [€] is the initial capital investment, $COST_t$ [€] is the cost at year t , r [%] the discount rate, and Q_{ht} [t] the hydrogen production. $COST_{Initial}$ is equal to the TPC and $COST_t$ is evaluated from OPEX and $s_{o\&m}$.

2.7 Life Cycle Assessment

The Life Cycle Assessment (LCA) is a methodology for estimating the emissions of products during their whole service lifetime, from raw material extraction and refinement, to manufacturing processes, usage, transports and disposal. LCA framework, which is thoroughly described by ISO Standards (ISO, 2021), involves the development of four subsequent steps: Goal and scope, *i.e.*, the definition of the characteristics of the study, Life Cycle Inventory (LCI), *i.e.*, the collection of material and energy balances over the entire life cycle of the product system, Life Cycle Impact Assessment (LCIA), *i.e.*, the assessment of the environmental performance over various environmental compartments using several impact categories scores, and interpretation, which is when practitioners need to draw conclusions on the outcomes of the study. The findings of such investigations are frequently published using well-established impact methodologies, such as ReCiPe, Environmental Footprint (EF) or TRACI. In this paper, the EF impact categories have been employed: total climate change (CC-T); ecosystem quality, which includes freshwater and terrestrial acidification (EQ-FTA), freshwater ecotoxicity (EQ-FE), marine eutrophication (EQ-EM), eutrophication of freshwater (EQ-EF) and terrestrial eutrophication (EQ-ET); human health, which involves the evaluation of impacts related to carcinogenic (HH-CE) or non-carcinogenic effects (HH-NCE), ionizing radiation (HH-IR), ozone layer depletion (HH-OD), photochemical ozone creation (HH-PCOC), and respiratory effects (HH-RE); resources depletion concerning the impacts on available raw materials such as dissipated water (RD-W), fossil fuels (RD-F), land (RD-L) and minerals and metals (RD-M).

3. Results and Discussion

Material and energy balance data coupled with cost estimation obtained by process simulation software are used to calculate the performance indicators: the detail of the data transferred from the process simulation are

reported in Table 2. EROEI, LCOH and LCA are calculated using the methods explained above. Table 3 shows the most significant results obtained from the process simulation to be used for the indicators' estimation of the indicators.

Table 2: Summary of data for the estimation of EROEI, LCOH and LCA.

Data	Units	EROEI	LCOH	LCA
Hydrogen production (Q_{H_2})	kg/h	X	X	X
Higher heating value of hydrogen (HHV)	kWh/kg	X	X	X
Power for auxiliary (P_{aux})	%	X	X	X
Total plant cost (TPC)	€	X	X	
Proportion between TPC and cost of energy for construction (ϵ_c)	€/kWh	X		
Capacity factor (cf)	-	X		X
Plant life time (L)	years	X		X
Efficiency (η)	-	X		X
Share of investment costs for operation - maintenance ($s_{o\&m}$)	%	X	X	
EROEI fuels	-	X		
Total amount of solvent	kg			X
Discount rate	%		X	

Table 3: data output from process simulation and from literature used to estimate the performance indicators.

Quantity	SMR – Grey H ₂	SMR & CCS – Blue H ₂	Electrolysis – Green H ₂	Electrolysis – Grid H ₂
P [kW]	37178	32069	40293	40293
P_{aux} [%]	14.06	25.87	7.7	7.7
TPC [k€]	107828.6	139771.7	58524.1	58524.1
cf [€/kWh]	0.70	0.70	0.95	0.95
so&m [%]	0.047	0.030	0.020	0.020
η	0.76	0.69	0.80	0.80
EROEI _{fuel}	19	19	25	38

Calculation of EROEI and LCOH is done using the details reported in previous chapter and the data obtained from process simulations (see Table 3). The following common data were used for all the processes considered: L=20 years, $\epsilon_c = 0.656$ €/kWh, $Q_{H_2} = 1000$ kg/h, $HHV_{H_2} = 39.4$ kWh/kg, discount rate = 7.3%. The estimated indicators are reported in table 4.

Table 4: EROEI and LCOH values for the different hydrogen colours considered.

Quantity	SMR – Grey H ₂	SMR & CCS – Blue H ₂	Electrolysis – Green H ₂	Electrolysis – Grid H ₂
E_{out} [GWh]	4559.59	3933.01	6706.492	6706.49
E_{cap} [GWh]	1017.251	1318.601	552.114	552.114
$E_{o\&m}$ [GWh]	956.216	791.161	220.846	220.846
E_r [GWh]	315.761	300.001	335.325	220.608
EROEI	1.99	1.63	6.05	6.75
LCOH	2.15	3.23	5.12	5.49

Calculations reported in Table 4 are reasonable in terms of Energy in and energy out from the processes. The values of EROEI predicted are in line with the literature values reported by Camacho et al. (2018): SMR EROEI = 2.5 and PV supplied electrolyser EROEI = 5.2. The LCOH values are in line with those reported by the Fuel Cells and Hydrogen Observatory (2022) for PV supplied (5.3) and for grid supplied electrolyser (5.5). EROEI and LCOH shows that the best technology in terms of energy and cost impact is the hydrogen produced by electrolysis using energy from photovoltaic modules. Concerning LCA, Life Cycle Inventory of secondary data has been retrieved within ecoinvent database and LCIA has been performed using EF impact categories. Figure 3 reports the results normalized to the highest score for each impact category. Climate Change (CC) is strongly reduced using green hydrogen in comparison with the other ones. Water-related impact categories are mainly affected by raw-material extraction for construction of renewable technologies and electricity distribution network. Air-related impact categories are driven by emissions during fossil fuel combustion, therefore green hydrogen exhibits the best performances.

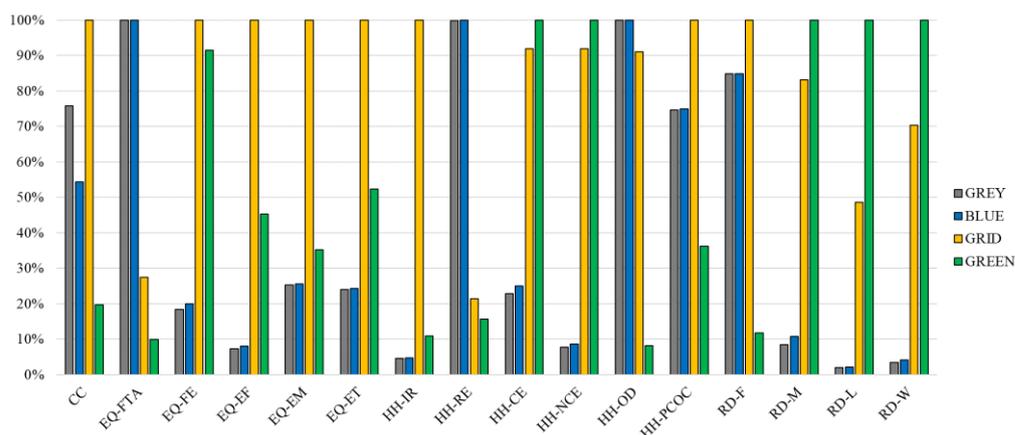


Figure 3. Normalized LCIA results for the considered hydrogen production routes.

4. Conclusions

Four hydrogen production processes have been simulated and the results of the simulations are used for the estimation of the indicators of interest; EROEI, LCOE, and LCA. The integration of indicators' evaluation with process simulation produces a double benefit: (i) we extended the scope of process simulation by considering up-to-date indicators of impact on energy, economy and environment and (ii) process evaluation may be performed at design time. The values of the key performance indicators shown in Table 4 and Figure 3, indicates that the best route for producing hydrogen in terms of global impact is the green hydrogen, based on electrolysis of water.

References

- Barbera, E., Mio, A., Massi Pavan, A., Bertucco, A., & Fermeglia, M. 2022, Fuelling power plants by natural gas: An analysis of energy efficiency, economical aspects and environmental footprint based on detailed process simulation of the whole carbon capture and storage system, *Energy Conversion and Management*, 252;115072;2022.
- Buttler A., Spliethoff H., 2018, Current status of water electrolysis for energy storage, grid balancing and sector coupling via power-to-gas and power-to-liquids: A review, *Renewable and Sustainable Energy Reviews*, 82, 2440-2454.
- Camacho C.E.G, Lombardelli G., Pirone R., Ruggeri B., 2018, Energy sustainability analysis of distributed H2 production, ISCRE25, Florence May2018.
- Fan, J. , Yu, P., Li, K., Xu, M., Zhang, X., 2022, A levelized cost of hydrogen (LCOH) comparison of coal-to-hydrogen with CCS and water electrolysis powered by renewable energy in China, *Energy*, 242, 123003.
- Fuel Cell and Hydrogen Observatory, 2022, Technology and Market, Levelized cost of hydrogen <https://www.fchobservatory.eu/observatory/>.
- Hall, C.A.S.; Lambert, J.G.; Balogh, S.B., 2014, EROI of different fuels and the implications for society, *Energy Policy*, 64, 141–152.
- ISO, The International Standards Organisation. ISO 14044, 2021, Environmental management - Life cycle assessment - Requirements and guidelines.
- King LC, Van Den Bergh J.C J.M., 2018, Implications of net energy-return-on-investment for a low-carbon energy transition. *Nature Energy*, 3, 334–340.
- Minutillo M., Perna A., Frcina A., Di Micco S., Jannelli E., 2021, Analyzing the levelized cost of hydrogen in refueling stations with on-site hydrogen production via water electrolysis in the Italian scenario, *International Journal of hydrogen production*, 46, 13667 – 13677.
- Petrescu L., Burca S., Fermeglia M., Mio A., Cormos C., 2021, Process simulation coupled with LCA for the evaluation of liquid - liquid extraction processes of phenol from aqueous streams, *Journal of water process engineering*, 41, 102077.
- Rana R.L., Lombardi M., Giungato P., Tricase C., 2020, Trends in Scientific Literature on Energy Return Ratio of Renewable Energy Sources for Supporting Policymakers, *Adm .Sci.*, 10,21.
- Sánchez M., Amores E., Abad D., Rodríguez L., Clemente-Jul C., 2020, Aspen Plus model of an alkaline electrolysis system for hydrogen production, *International Journal of Hydrogen Energy*, 45, 3916-3929.