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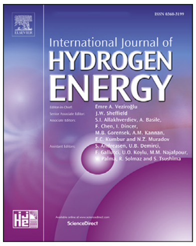

Selection criteria and ranking for sustainable hydrogen production options

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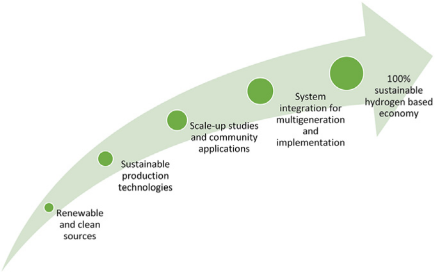
HIGHLIGHTS

- A holistic study of hydrogen production options for a sustainable and carbon-free future.
- It outlines the benefits and challenges of hydrogen production methods.
- Sixteen methods are selected for sustainability investigation based on seven different criteria.
- It covers economic, technical, environmental, and thermodynamic aspects of sustainability.
- It can help stakeholders deploy a hydrogen roadmap for a more sustainable future.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper aims to holistically study hydrogen production options essential for a sustainable and carbon-free future. This study also outlines the benefits and challenges of hydrogen production methods to provide sustainable alternatives to fossil fuels by meeting the global energy demand and net-zero targets. In this study, sixteen hydrogen production methods are selected for sustainability investigation based on seven different criteria. The criteria selected in the comparative evaluation cover various dimensions of hydrogen production in terms of economic, technical, environmental, and thermodynamic aspects for better sustainability. The current study results show that steam methane reforming

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with carbon capture could provide sustainable hydrogen in the near future while the other technologies' maturity levels increase and the costs decrease. In the medium- and long-terms, photonic and thermal-based hydrogen production methods can be the key to sustainable hydrogen production.

Introduction

We are in an era where climatic and environmental calamities have been more apparent and more devastating. There is a vital need to implement renewable energy technologies locally and globally to address the energy challenges and develop carbon-free fuel (primarily hydrogen) options.

The future role of hydrogen depends upon how effectively sustainable hydrogen production technologies will reach commercial maturity [1]. The path to sustainable hydrogen (Fig. 1) implies that there is a need to link clean and renewable sources to the end-users in the market via sustainable hydrogen production systems.

There are many examples in the open literature on the economic, environmental, and technical aspects of fossil and non-fossil-based hydrogen production methods [2–4]. For instance, Dawood et al. [5] have provided a pathway toward 100% renewable energy systems and investigated hydrogen's role in future energy systems. They have further assessed several hydrogen production options based on efficiencies, environmental impact, and technology maturity level rather than only focusing on cost-effectiveness.

Parra et al. [6] have presented a technoeconomic review of hydrogen energy systems, including power-to-power, power-to-gas, hydrogen refueling, and stationary fuel cells. They have focused on the capital and operating expenses and efficiencies and provided recommendations for policymakers. El-Emam and Ozcan [7] have extensively analyzed the literature on technological, economic, and environmental aspects of clean-hydrogen production. They have introduced hydrogen production routes compatible with renewable and nuclear sources by highlighting the recent advances and developments in the literature. In addition, El-Emam and Ozcan [7] have discussed the current and future expectations on cost aspects of the clean-hydrogen economy. They have also analyzed the recent literature on the environmental aspects of clean-hydrogen production technologies. They have concluded that clean and carbon-free hydrogen production can be economically feasible in the near future.

The technoeconomic assessment of hydrogen production methods [8] shows the current state of the hydrogen supply chain as a forwarding energy vector, comprising resources, generation and storage technologies, demand market, and economics. Ji and Wang [9] have reviewed the current status, recent advances, and challenges of fossil- and renewable-based hydrogen production methods. They have compared the life cycle cost and environmental impact of different hydrogen production methods. They have concluded that electrolysis and thermochemical cycles coupled with clean energy sources show considerable potential in terms of economics and environmental friendliness.

In another example of the sustainability assessment [10], the authors have reviewed various photocatalytic systems for hydrogen production via overall water splitting and photo-reforming of biomass-derived organic substances. They have also evaluated quantum efficiency and solar to the hydrogen conversion efficiency of existing photocatalysts. Ferraren-De

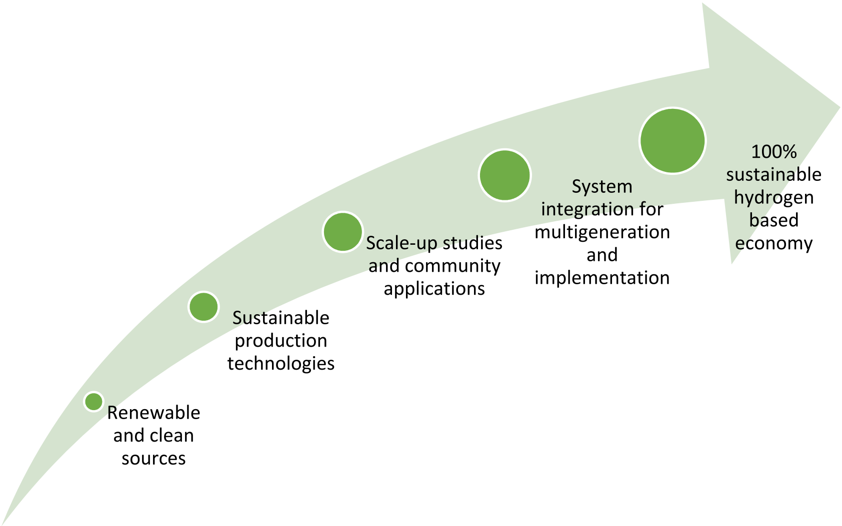


Fig. 1 – A potential pathway to reach 100% sustainable hydrogen in the energy market.

Cagalitan et al. [11] have provided an overview of a variety of hydrogen production methods via biochemical reactions (biohydrogen). They have also discussed the challenges of biohydrogen production and offered solutions to some issues.

With similar motivation, Vyas et al. [12] have reviewed photocatalytic hydrogen production via carbon quantum dots to make photonic and bio-based hydrogen affordable, reliable, clean, and the fuel of the future. Sun et al. [13] have evaluated graphitic carbon nitride heterojunction photocatalysts for solar hydrogen production. They have focused on solar-to-hydrogen and apparent quantum efficiency, hydrogen evolution reaction rate, charge transfer, and additional material properties for more sustainable photocatalytic hydrogen production.

In a roadmap for current and future exploration of carbon-free hydrogen production and exportation [14], the authors have assessed several available alternatives for Qatar. They have considered using natural gas as a feedstock for hydrogen production through steam methane reforming (SMR), solar integrated steam methane reforming with carbon capture, and electrolysis. They have identified the potential of each alternative based on selected technical, economic, and environmental criteria. They have concluded that green hydrogen has the potential as a sustainable fuel in Qatar in the near future. They have highlighted that green hydrogen will become quite competitive in the region as technologies associated with clean hydrogen production improve and the cost of renewable energy falls.

Dolle et al. [15] have reviewed the recent developments and trends concerning the electro-reforming of biomass. Similarly, Lepage et al. [16] have investigated biological and electrochemical hydrogen production processes with lower technology readiness levels. Both studies indicate that although electro-reforming processes are less mature, they have significant advantages. Some of these advantages are the mild operating conditions, lower energy consumption; clean hydrogen production without downstream purification processes; and the possibility of coproduction of value-added compounds at the anode.

Ma et al. [17] have provided insights into the feasibility of using hydrogen and bioethanol blends as energy carriers in the foreseeable future upon discussions on the advantages and the disadvantages. They have provided comprehensive overviews of hydrogen and bioethanol production, storage, and transportation. They have also summarized the current problems and potential solutions. According to the authors, the increasing research on bioethanol reforming to hydrogen and the emergence of solid-state storage methods for hydrogen could make it possible for hydrogen to be used as a carrier of energy sources in the near future.

As discussed above, numerous examples in the literature focus on evaluating and enhancing hydrogen production performance. Several studies also comparatively and quantitatively evaluate the sustainability performance of various hydrogen production options. However, there is a lack of studies combining energetic, exergetic, economic, and environmental performance with technology maturity. Also, there is a lack of studies comparatively assessing the sustainability performance of green, blue, gray, brown, orange, and turquoise hydrogen. The motivation behind this study is to address this gap, which can be a meaningful contribution to the literature.

This study develops selection criteria and ranking specifically for hydrogen production methods from renewable and non-renewable sources, and these methods are then holistically discussed, assessed, and compared with each other. These sixteen hydrogen production methods are comparatively assessed based on environmental, technical, economic, and thermodynamic performance. The study further aims to show the strengths and weaknesses of the selected hydrogen production methods, potentially providing valuable information to the industry, buildings, and transportation sectors. Such information will help provide a clear roadmap to 100% hydrogen use in all sectors and further guide researchers, policymakers, and industry while transitioning to a hydrogen economy for a sustainable and zero-carbon future.

Hydrogen production methods

This section introduces and discusses selected hydrogen production methods for sustainability evaluation. According to their color codes, the selected hydrogen methods are provided in Fig. 2. Green hydrogen is produced through processes, such as water electrolysis, by employing renewable energy sources. It is called green because there is no CO_2 emission during the production process. Blue hydrogen is sourced from fossil fuels. However, the CO_2 is captured and stored (carbon capture and sequestration). Gray hydrogen is produced from natural gas and commonly uses the steam methane reforming method. During this process, CO_2 is produced and eventually released into the atmosphere. Brown hydrogen is produced from coal. The gasification of coal is a method used to produce hydrogen. However, it is a very polluting process, and CO_2 and carbon monoxide are produced as by-products and released into the atmosphere. In this study, biomass gasification is classified as orange because it still releases CO_2 into the atmosphere, but biomass is not a fossil fuel. The hydrogen production options that rely on thermal energy are classified as turquoise hydrogen.

The selected hydrogen production methods can be categorized based on their primary energy and material (input) resources. These methods use either one or the combination of the following primary energy sources: electrical, thermal, photonic, and biochemical. Electrical and thermal energies can be obtained from renewables, biomass, nuclear, or fossil fuels. The electrical and thermal energy source significantly affects the corresponding methods' emissions.

Electrolysis

Water electrolysis powered by renewable energy sources is expected to enable the scale-up of hydrogen production. During the water electrolysis processes, there are no CO_2 emissions. Hence, storing surplus renewable energy as hydrogen shows excellent promise. Another advantage is that hydrogen from water electrolysis has high purity (99.9%) [18] and can be used in many industrial processes. Typical characteristics of leading electrolysis technologies are listed in Table 1.

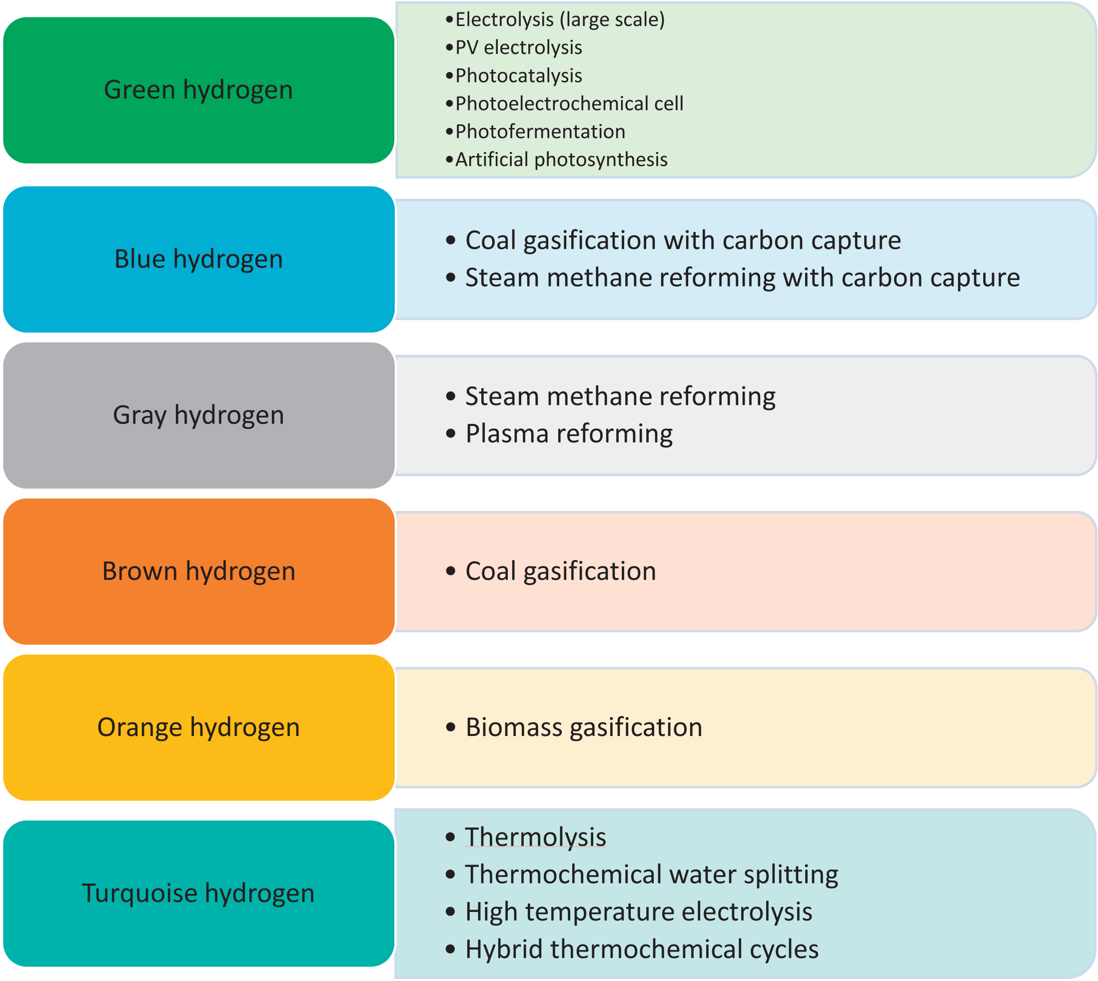


Fig. 2 – Specific hydrogen production methods selected for assessment and their color codes. (For interpretation of the references to color/colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1 – Typical characteristics of leading electrolyzer technologies (adapted from Ref. [19]).

Electrolyzer Type	Charge carrier	Temperature (°C)	Electrolyte	Application
Conventional alkaline	OH^-	20–80	Liquid	Commercial
Solid alkaline	OH^-	20–200	Polymeric	Lab-scale
PEM	H^+	20–200	Polymeric	Near-commercialization
H^+ SOE	H^+	500–1000	Ceramic	Lab-scale
O^{2-} SOE	O^{2-}	500–1000	Ceramic	Demonstration
Co-electrolysis	O^{2-}	750–900	Ceramic	Lab-scale

Note that the Perovskites are commonly used as anodes in conventional alkaline electrolyzers, and Ni-alloys are cathodes. Efficiency is reported to be between 59 and 70% [20]. The advantages are the low capital cost, relatively stable structure, and mature technology. However, the disadvantages are corrosive electrolytes, gas permeation, and slow dynamics. The challenges related to conventional alkaline electrolyzers are improving durability and reliability.

The anodes of solid alkaline electrolyzers are Ni-based, and the cathodes are also Ni alloys. Solid alkaline electrolyzers are in laboratory-scale operation, and therefore there is not sufficient information about their efficiency in the literature. Solid alkaline electrolyzers combine the advantages of alkaline and proton exchange membrane (PEM) electrolyzers. Then again, they have some drawbacks, including low OH^- conductivity in polymeric membranes. Improving the electrolyte conductivity is one of the main challenges related to solid alkaline electrolyzers [21].

PEM electrolyzers are in the near-term commercialization stage with 65–82% efficiencies. PEM electrolyzers have numerous advantages, including compact design, fast response and start-up, and highly pure hydrogen production. The disadvantages of the electrolyzers are the use of high-cost polymeric membranes and the requirement of noble metals due to their acidic medium. Reducing noble metal usage in PEM electrolyzers is one of the primary challenges [22].

In the literature, solid oxide electrolyzers (SOE) are grouped into two categories, as H^+ SOE and O^{2-} SOE. H^+ SOE is laboratory-scale, while O^{2-} SOE is in the demonstration stage [23]. Both SOEs have numerous advantages, including enhanced kinetics and thermodynamics, lower energy demand, and low capital costs. The challenges related to SOE are microstructural changes in the electrodes, delamination, and passivation [24].

Another electrolyzer technology, co-electrolysis, is in the laboratory-scale application. This technology shows great potential, especially in the direct production of syngas. Co-electrolyzers and SOE technologies have similar disadvantages, including mechanically unstable electrodes, which cause cracking, safety issues due to high temperatures, and sealing issues. The challenges associated with co-electrolysis are carbon deposition and microstructural changes on the electrodes to address these disadvantages [25].

Plasma reforming

The plasma reforming method can produce hydrogen from various feedstocks, including alcohols and fossil fuels, without wasting precious or rare resources [26]. In the literature, there are several examples of plasma reforming, such as corona discharge [26], dielectric barrier discharge [27], and gliding arc discharge (GAD) [28]. Gliding arc discharge has lower energy consumption and smaller reactor volume than corona discharge and dielectric barrier discharge [29].

In the open literature, some of the ideal sources to produce hydrogen via plasma reforming are methane, toluene, dodecane, high-octane gasoline, and diesel [30]. The need for fuels with high energy density is because of the high-temperature requirement of plasma reforming. In plasma reforming, the feedstock fuels need to be exposed to extremely high temperatures higher than 2000 °C in the presence of oxidants to produce syngas and hydrogen [31].

Thermolysis

Thermolysis is a thermal water decomposition reaction that occurs in a single step, following a straightforward chemical principle [32]. In thermal water decomposition reactions, the vast amount of energy needed to break water molecules is supplied by heat. For this reason, as a thermal water decomposition reaction, thermolysis requires large amounts of heat at high temperatures. In the literature [33], thermal water decomposition is reported to start at temperatures over 1700 °C. In order to achieve complete conversion of water via thermolysis, the temperature requirements are above 4000 °C. The heat requirement of the thermolysis reaction can be met by solar thermal, biomass, or geothermal heat [34]. The challenge with solar thermal as the energy source in thermolysis is the supply of uninterrupted heat to support continuous operation [35].

One major disadvantage related to thermolysis is high operating temperatures. It is not easy to find safe, durable, and high-temperature-resistant materials in most cases. Another challenge is the separation of hydrogen and oxygen because, in thermolysis, the product gases are not generated in two different outlets. Since the thermolysis reaction product gases are a mixture of oxygen and hydrogen gases, there is always a considerable risk of these two gases recombining into water. Besides, the recombination reaction is very explosive, which is a safety issue. One possible solution to prevent the recombination of oxygen and hydrogen is quenching. However, quenching is effective at lower temperatures. Preventing recombination of the product gases via quenching requires temperatures to be reduced dramatically by 1500–2000 °C in a few milliseconds after the thermolysis reaction [9]. There are several efforts to separate the product gases in the literature, such as membranes, different reactor centrifugations, and supersonic jets [8]. However, these technologies are pretty limited.

Several challenges, as discussed above, slow down the large-scale application, market introduction, and commercialization of thermolysis. However, despite the drawbacks, thermolysis technologies are continually improving. Thermolysis still has significant advantages, especially in small scales [35,36].

Thermochemical water splitting

As mentioned before, single-step water decomposition requires temperatures above 4000 °C for complete water dissociation [37]. Thermochemical cycles include a series of chemical reactions with a net reaction of water dissociation. By including multiple steps, the temperature requirement of thermal water dissociation is reduced, which is a significant advantage for integrating heat from renewable sources in thermal hydrogen production. However, among the current thermochemical cycles studied in the literature, only a few have promising results in the lab scale.

Mandal and Jana [38] have designed a reactive distillation column for the S–I cycle to tackle the reaction equilibrium limitations and overcome azeotrope formation in the HI dissociation reaction. HI decomposition and H_2 separation occur concurrently in the designed system, leading to lower energy consumption and higher efficiency.

Another example of the S–I cycle has been performed by Yilmaz and Selbas [39] by combining the cycle with solar energy. The authors have reported the overall energy and exergy efficiencies of around 32.76% and 34.56%, respectively. Park et al. [40] have presented an integrated system that combines the S–I cycle with a steam boiler. The steam boiler is used to supply the heat demand of the thermochemical cycle. The authors have eliminated the H_2SO_4 dissociation reaction in the S–I cycle, reducing the temperature requirements and associated costs.

Thermochemical S–I cycles require catalysts to enhance the HI dissociation and conversion rates. Since it is an expensive catalyst, Pt requirement increases the system costs and is a significant challenge for the deployment of thermochemical cycles. For this reason, there are many attempts in the literature to replace Pt with safer, more abundant, and more affordable catalysts. Some catalysts, such as Pd–CeO_2-300, show enhanced activity, stability, and HI conversion rates than Pt alone [41].

Another example of promising thermochemical cycles in the literature is Cu–Cl cycles, which have two to five steps. An example is the four-step Cu–Cl cycle, where HCl production and drying co-occur. The advantages are reduced sedimentation and clogging. However, there are several disadvantages, such as high-temperature requirements and low productivity. These challenges are addressed by combining oxychlorination and dissociation reactions in a single step called a three-step cycle. Using a single reactor for all thermochemical reactions is an essential advantage of the three-step cycle. Nevertheless, there are still disadvantages, such as high-temperature heat requirement and the corrosive medium, which increase the system costs [38].

Biomass gasification

Biomass gasification is considered the most convenient and economical method for hydrogen production. In this process, special chemical agents are used to convert biomass into a mixture of gaseous products called syngas or producer gas at relatively high temperatures between 900 °C and 1200 °C. Air, oxygen, steam, or their combinations can be utilized as chemical agents in biomass gasification [42,43]. In the gasification process, biomass quality is critical. For instance, biomass moisture content must be kept between 9 and 22% [44].

It is apparent that complex thermochemical reactions occur during the gasification process. As a result, gaseous and solid species are simultaneously interconverted. The critical stages in the process include drying, partial oxidation, pyrolysis, and gasification. There are two roles of partial gasification; one is to produce heat to be used in the following reaction stages, and the other role is to lower the moisture ratio in the biomass feedstock. Pyrolysis occurs between 200 and 700 C, where the partial oxidation reaction supplies heat. In this stage, oxidizing agents (O_2 or air) are used to produce a mixture of gases such as H_2, CO, CO_2, and CH_4 and release moisture. In the pyrolysis stage, char and tar are also formed, then they undergo thermal cracking and produce noncondensable gases and light hydrocarbons [45].

Compared to the biomass combustion and pyrolysis processes, gasification has significant advantages. Firstly, biomass gasification has a higher conversion efficiency. Because the product gases (CO, H_2, and CH_4) have a higher calorific value [46]. A wide range of biomass feedstocks can be used for gasification, including wood, agricultural waste, and the organic part of municipal solid waste [47]. Biomass gasification followed by combustion in cogeneration mode provides benefits [48,49]. This approach has higher efficiency and can be used for synthesizing different fuels as valuable products.

Biomass gasification effectively converts biomass into energy, suitable for distributed (decentralized) energy systems. Distributed systems have multiple benefits, such as reduced transmission, delivery, distribution losses, and lower transportation costs. Therefore, biomass gasification can play a crucial role in future hydrogen energy systems.

Photocatalysis

Photocatalysis is a prospective way to efficiently convert and store solar energy, which is beneficial for achieving sustainable hydrogen production [50]. There has been almost one century of research on photocatalysis, and there are still debates on the difference between photocatalysis and photosynthesis [51]. The International Union of Pure and Applied Chemistry (IUPAC) defines photocatalysis as “photocatalysis function as a change in the rate of a chemical reaction or its initiation under the action of ultraviolet, visible, or infrared radiation in the presence of a substance, the photocatalyst, that absorbs light and is involved in the chemical transformation of the reaction partners [52].”

Photocatalysis research can be grouped into five categories: water splitting for H_2 production [53], CO_2 reduction [54], nitrogen fixation [55], contaminant degradation [56], and organic synthesis [57]. In all categories, photocatalysts show the advantage of direct solar energy conversion into valuable products without generating electricity as an intermediate step [58].

Photocatalysis has several limitations, such as bandgap and thermodynamic spontaneity, hindering the reaction rate or yield [59]. Osterloh [60] has investigated several photocatalytic and photosynthetic water splitting methods to identify these limitations and provided several ways to tackle these challenges. Rajeshwar et al. [61] have investigated the free-energy change of photocatalytic and photosynthetic reactions to tackle the thermodynamic limitations. Photocatalysis is recognized as an ongoing and promising research topic in the literature, and many studies focus on the materials sciences and engineering design aspects [62,63].

Photoelectrochemical cells

The primary mechanism of photoelectrochemical (PEC) water splitting involves the direct conversion of sunlight into hydrogen fuel. The process occurs with the help of an external bias applied to the photoelectrodes immersed in an electrolyte [64].

Four basic steps are generally involved in PEC water splitting: First, sunlight falls on the photoactive working electrode (i.e., photoanode) and generates electron-hole pairs. Secondly, the photogenerated holes at the photoanode surface cause water oxidation. Thirdly, electrons generated from incident photons transfer through an external wire from the photoanode to the photocathode. Then reduction of H^+ at the photocathode surface takes place by these electrons to form hydrogen gas [65]. Efficient carrier separation is carried out using an external power supply between two electrodes. In the PEC process, to split water, the photoactive material must have an appropriate bandgap to generate the electron from the incoming photonic energy [66].

The band positions of the semiconductors are another critical factor affecting the PEC water splitting performance. The conduction band edge determines the reducing power of the photogenerated electrons. At the same time, the valence band edge determines the oxidation power of the photogenerated holes [67].

The PEC-based hydrogen production has gained considerable attention during the past years because of its several advantages, such as clean operation and direct sunlight conversion. These advantages give an outstanding opportunity to link solar energy to sustainable hydrogen production. PEC can also be linked to integrated systems for multigeneration, such as producing power, hydrogen, heat, cooling, and freshwater simultaneously [68]. It is also possible to produce hydrogen from wastewater with PEC [69].

The PEC-based hydrogen production technologies are still in the early research stages. Developing, designing, analyzing, building, evaluating, and enhancing PEC systems is crucial for affordable and reliable hydrogen production. For this reason, enhancing PEC performance requires intensive research focusing on the following areas: transport phenomena, thermodynamics, electrochemistry, engineering design and evaluation, materials science, system integration, and comprehensive performance evaluation.

Hybrid thermochemical cycles

Hybrid thermochemical cycles are primarily thermochemical cycles where heat and some electricity are used simultaneously, which gives two significant advantages. First, they have lower electricity consumption than electrolysis. Second, they have lower temperature and heat requirements than thermal water splitting. Low-temperature heat requirement allows for moderate-temperature heat sources, such as process waste heat [70]. Energy efficiencies up to 48–50% can be obtained from hybrid processes [71].

The Westinghouse's two-step hybrid sulfur (HyS) cycle is the most well-known hybrid thermochemical cycle for hydrogen production. Westinghouse originally proposed HyS as a thermochemical-electrochemical cycle to supply large-scale hydrogen in the 1970s. HyS is the first demonstrated hybrid thermochemical cycle and has just two reactions. These reactions are the thermal decomposition of sulfuric acid (heat consuming step) and the electrochemical oxidation of SO_2 with water (electricity consuming step) to yield sulfuric acid and hydrogen [72,73]. The voltage requirement of the electrochemical oxidation step is less than 0.20 V. This amount is significantly lower than the electrolysis voltage requirement, which is around 1.23 V [74].

The most critical challenges of the HyS cycle are reported to be SO_3 reduction temperature and corrosive chemicals [75,76]. The reaction rate can be enhanced by utilizing iron oxide-based catalysts. Silicon Carbide (SiC) can be used to make the system components corrosion-resistant [73]. Integrating the HyS cycle with concentrated solar energy is reported to have an overall system efficiency of over 25% and cost between 3 and 6 USD/kg H_2 [71].

Coal gasification

Hydrogen production via coal gasification is reported to be the most cost-efficient method [77]. The process also has another advantage of high calorific value syngas production [78]. Plasma gasification is comparatively new and promising among the existing coal gasification processes [79]. Plasma gasification has higher conversion efficiency than other gasification options because of its higher operating temperatures. For the same reason, it is the only gasification option that allows waste metal recovery. Some of the other advantages of plasma gasification are listed as follows [80–82]:

- ➢ Higher efficiency than combustion (<50%), pyrolysis (<43%), and other gasification technologies (<19%)
- ➢ Less tar output and a higher carbon conversion rate than other gasification methods
- ➢ Production of syngas from organic wastes with higher conversion efficiency
- ➢ Less toxic residues, such as ash, slag, et cetera
- ➢ Higher hydrogen content in the syngas output compared to other gasification methods

As a result, plasma gasification can be an advantageous coal gasification method to produce hydrogen. The process generates syngas that contains CO and H_2. After the gas cleaning process, pure hydrogen (about 99%) can be obtained. In the literature, coal gasification is reported to produce around 0.1 kg of hydrogen from 1 kg of coal [77].

Natural gas reforming

Natural gas is a mixture of gases, and its main constituent (up to 99%) is methane [83]. The primary use of natural gas is direct combustion for heat and electricity generation purposes. Other emerging uses of natural gas turn methane into several other fuels and industrial chemicals [84]. For example, methane can be converted to H_2 or syngas via steam methane reforming [85], dry methane reforming [86], and partial methane oxidation [87].

Steam methane reforming is the primary hydrogen and syngas production method used in the industry [88]. The process consists of an endothermic reaction between methane and steam at elevated temperatures (about 750–950 °C) and pressures (around 14–20 atm) [89]. In some cases, a water-gas shift reaction occurs during the process, further enhancing the production of H_2.

The research and development activities regarding natural gas reforming focus on developing affordable, clean, efficient, and reliable steam methane reforming technologies. Ni/Al_2O_3 is the most commonly used catalyst because of its high activity and low cost [90]. Nevertheless, it has some disadvantages, such as coking formation and sintering of Ni particles. For this reason, designing advanced catalysts is needed to develop catalysts with high sintering and coking resistance. Modifying Ni catalysts by promoters, novel metals, self supports, and solid solutions are some of the examples [91]. Other approaches are finding alternatives to Ni-based catalysts. It is also crucial to design catalysts that are not noble metal-based.

There are several approaches to reducing the energy requirement and enhancing the efficiency of steam methane reforming in the literature. Some examples are chemical looping, electro-catalytic reforming, oxidative reforming, photocatalytic reforming, plasma reforming, solid oxide fuel cell, sorbent enhancement, and thermo-photo hybrid reforming [92–96].

Photofermentation

In photofermentation, photosynthetic bacteria capture light energy and convert organic acids generated during anaerobic fermentation to H_2 and CO_2 in a nitrogen-deficient environment [97]. These photosynthetic microorganisms exist in the natural environment and can process a wide range of substrates over a broad light spectrum. There is no O_2 generation in photofermentation, which is advantageous because O_2 inhibits H_2 production [11].

In photofermentation, photofermenters capture the light from the sun or an artificial source. The sun is a cheaper light source, but it cannot produce hydrogen continuously. Artificial light can support uninterrupted biohydrogen production through cloudy, foggy days and during nights. On the other hand, artificial lights require energy input, and they have higher costs, such as capital, operating and maintenance, et cetera [98,99].

The hydrogen yield of photosynthesis depends on the light intensity, medium, microorganism, photofermenter design, substrate, and several other factors [97,98]. Light is the primary energy source of photofermentative hydrogen production [99,100]. Both type and intensity of light should be considered when designing photobioreactors. It is also essential for conducting the photofermentative biohydrogen production process.

The temperature range of photofermentative hydrogen production can be classified into four types: medium temperature (25–40 °C), elevated temperature (40–65 °C), extremely high temperature (65–80 °C), and ultrahigh temperature (above 80 °C). Most photofermentative bacteria produce hydrogen at medium temperature, typically 30–40 °C, although faster metabolism and higher hydrogen yield and production rate could be achieved at higher temperatures [101].

In addition, pH value is essential to maintain the intracellular dynamic equilibrium, hydrogenase activity, cellular redox potential, and many other metabolic activities. The optimal pH for photofermentative biohydrogen production is generally around 5.0–7.0. This interval may vary with different substrates, inoculum, or culture conditions. If the pH is not controlled, the final pH values after fermentation usually drop to 4.0 due to the production of volatile fatty acids. It is worthy to note that the decrease in pH would impact hydrogen production by inhibiting hydrogenase activity [102].

Artificial photosynthesis

In artificial photosynthesis, a biochemical reaction mimics natural photosynthesis. Titanium oxide nanoparticles imitate the role of chlorophyll and capture the incoming light [103]. Replicating the natural process can simultaneously provide electricity, food, and fuel (e.g., hydrogen, methane, methanol) [104].

Artificial photosynthesis has several challenges, such as low efficiency of light capture, electron transfer, water splitting, and CO_2 reduction [103]. The available catalysts (e.g., a blue dimmer, cobalt, iridium, and rhodium) have low efficiency and high cost. For this reason, there have been numerous studies on developing innovative catalysts to enhance the process [105–107].

Despite the disadvantages listed above, artificial photosynthesis has higher solar-to-hydrogen efficiency than PV-based electrolysis. One reason is that artificial photosynthesis uses light absorbers to use a more significant portion of the incoming photonic energy than the semiconductors used in PVs. However, the light absorbers (e.g., natural and synthetic dyes) increase the cost, and artificial photosynthesis has a shorter system lifetime. Therefore, PV electrolysis remains the preferred option due to its lower cost and longer life [106].

Sustainability performance assessment

This study investigates the strengths and weaknesses of the carefully chosen hydrogen production methods based on seven sustainability criteria. The first two criteria to be investigated are related to the hydrogen production efficiency from the first and second laws of thermodynamics. The first law of thermodynamics helps determine energy efficiency, while the second law brings exergy efficiency forefront. Since efficiency is generally described as the amount of desired output divided by the amount of required input, the energy efficiency equation becomes:

$$\eta = \frac{\dot{m}LHV}{\dot{E}_{in}} \tag{1}$$

Here, $\dot{m}$ is the hydrogen production rate in terms of kg/s, LHV is the lower heating value of hydrogen (taken as 121 MJ/kg) and $\dot{E}_{in}$ is the rate of energy use in the process in terms of MJ/kg. The energy efficiency equation can be modified into the exergy efficiency equation as

$$\psi = \frac{\dot{m}ex^{ch}_{H_2}}{\dot{E}x_{in}} \tag{2}$$

In the exergy efficiency equation, $ex^{ch}_{H_2}$ denotes the chemical exergy of hydrogen and $\dot{E}x_{in}$ is the rate of exergy input into the process. This study takes energy and exergy efficiencies of the selected hydrogen production methods from the literature [2,4,5].

The third criterion is the cost of hydrogen production, which is particularly important, especially during the commercialization and scaling up steps. An essential step towards sustainable hydrogen widely used in the market is making it more affordable. Currently, natural gas reforming and coal gasification have the lowest hydrogen production cost, but they also have the highest emissions. The emissions can be reduced by carbon capture (CC) technologies. However, CC increases the hydrogen production cost by about 10–20% [9].

Compared to traditional, fossil-based options, most renewable-based hydrogen production technologies are still in relatively early development stages. As a result, the life-cycle cost of renewable hydrogen is higher than the fossil-based one. However, there has been a significant decrease in the cost of renewable-based hydrogen. Further cost reduction is expected in most emerging renewable hydrogen production methods due to developments in materials sciences and system designs. Fossil-based hydrogen via carbon capture can be a transition step while renewable hydrogen becomes more affordable on large scales. In this study, the cost data of the selected hydrogen production methods are taken from Refs. [3,4,7,35,108].

The fourth and fifth criteria are the global warming and acidification potentials based on the life cycle assessment (LCA). LCA is a reliable method to investigate the actual environmental impact [109]. Global warming potential (GWP) shows kg CO_2 emissions per kg H_2 produced. Acidification potential (AP) indicates waste discharge into the soil and water in terms of grams SO_x released per kg H_2 produced. GWP and AP are the most commonly used environmental impact indicators [110]. The literature shows the clear advantage of renewable hydrogen over fossil-based hydrogen in terms of GWP and AP [111,112]. In addition to these resources, the GWP and AP data are gathered from Refs. [2,4].

The sixth criterion is the cost of carbon (CC). CC measures the marginal external cost of a unit of CO_2 emissions due to the damage to the environment and health. The estimation of CC is conducted based on different models, which can be found in detail in the literature [113–118]. The CC is estimated to be 160 USD per ton of CO_2 emissions in this study.

Technology maturity level (TML) which is treated as the seventh criterion, is a modified and consolidated rating scale between 1 and 10. The TML helps communicate the level of maturity of a particular technology. The rating 1 refers to the exceedingly early stages of research, primarily assigned to novel and small-scale options, and the rating 10 refers to the total market integration. The technology maturity level data are obtained from Ref. [5].

In the last step, all performance criteria are normalized and ranked between 0 and 10 to conduct the comparative assessment. 0 indicates the least desirable case, and 10 is the ideal option. There is no normalization applied to the technology maturity level since it is already on a 0–10 scale, with 0 and 10 indicating the least and most desirable options, respectively. For all criteria, the sustainability performance of the selected hydrogen production methods increases as their ranking increases from 0 to 10. Energy and exergy efficiencies are normalized by:

$$Rank(i) = efficiency(i) \times 10 \tag{3}$$

The remaining criteria (global warming and acidification potentials, production cost, and cost of carbon are normalized accordingly and ranked as

$$Rank(i) = \frac{max\ value - performance(i)}{max\ value} \times 10 \tag{4}$$

Here, (i) represents the selected method. The “max value” in equation (4) denotes the highest value in the corresponding performance category. For example, if a method has the highest emissions among the selected options, it ranks 0. Alternatively, if an option has the lowest cost, it would be assigned to the highest ranking. It should be noted that ranking 10 is assigned to zero cost and emissions. Therefore, none of the methods get the ideal ranking. This approach aims to show that each option still has improvement potential in all selected performance criteria.

Once all performance criteria for each of selected hydrogen production options are normalized and ranked between 0 and 10, their total sustainability scores are then calculated. The total scores are compared to the hypothetical ideal scenario where hydrogen is produced with 100% energy and exergy efficiencies, zero GWP, AP, and cost, and a TML of ten. Next, the average scores are calculated. There are eight cases to calculate the average scores, which are namely:

- EI: all criteria have equal importance
- EE: energy efficiency has a weight of 40%, and the rest of the criteria have a weight of 10% each
- ExE: exergy efficiency has a weight of 40%, and the rest of the criteria have a weight of 10% each
- Cost: production cost has a weight of 40%, and the rest of the criteria have a weight of 10% each
- CC: cost of carbon has a weight of 40%, and the rest of the criteria have a weight of 10% each
- GWP: GWP has a weight of 40%, and the rest of the criteria have a weight of 10% each
- AP: AP has a weight of 40%, and the rest of the criteria have a weight of 10% each
- TML: TML has a weight of 40%, and the rest of the criteria have a weight of 10% each

Results and discussion

This section presents and discusses the comparative performance evaluation results of the selected hydrogen production methods based on the selected performance criteria. The first criterion is energy efficiency (Fig. 3). High temperature electrolysis has the highest energy efficiency among the selected production options, followed by large-scale electrolysis and steam methane reforming. In high temperature electrolysis, electricity and heat are used together to split water. Since thermal energy supplies part of the required energy input for water dissociation, high temperature electrolysis uses less electricity and has higher energy efficiency. High temperature electrolysis is still in the research and development phase and is not commercialized, but it has many significant advantages. Some of its benefits support the reverse process (such as reversible solid oxide electrolyzer/fuel cell). The costs of high temperatures are expected to be competitive in a couple of decades [119,120].

On the contrary, photofermentation has the lowest energy efficiency. Photocatalysis and artificial photosynthesis have the second and third lowest energy efficiencies. One reason for that poor performance is the lack of photoactive materials that convert a more significant portion of the solar spectrum into hydrogen in an efficient manner. Therefore, more research and development activities are needed to develop more effective, reliable, durable, and clean photoactive materials.

As can be seen from Fig. 4, hybrid thermochemical cycles have the highest exergy efficiency, followed by steam methane reforming and coal gasification. Reforming and gasification with carbon capture have slightly low efficiency due to the energy requirement of the capture processes. On the other hand, photofermentation, photocatalysis, and photoelectrochemical cells have the lowest exergy efficiency. The exergy efficiency of the hydrogen production processes can be enhanced by eliminating the waste heat via energy recovery such as multigeneration. Similar to the energy efficiency results, advanced photoactive materials could significantly enhance the exergetic performance of photonic hydrogen production processes.

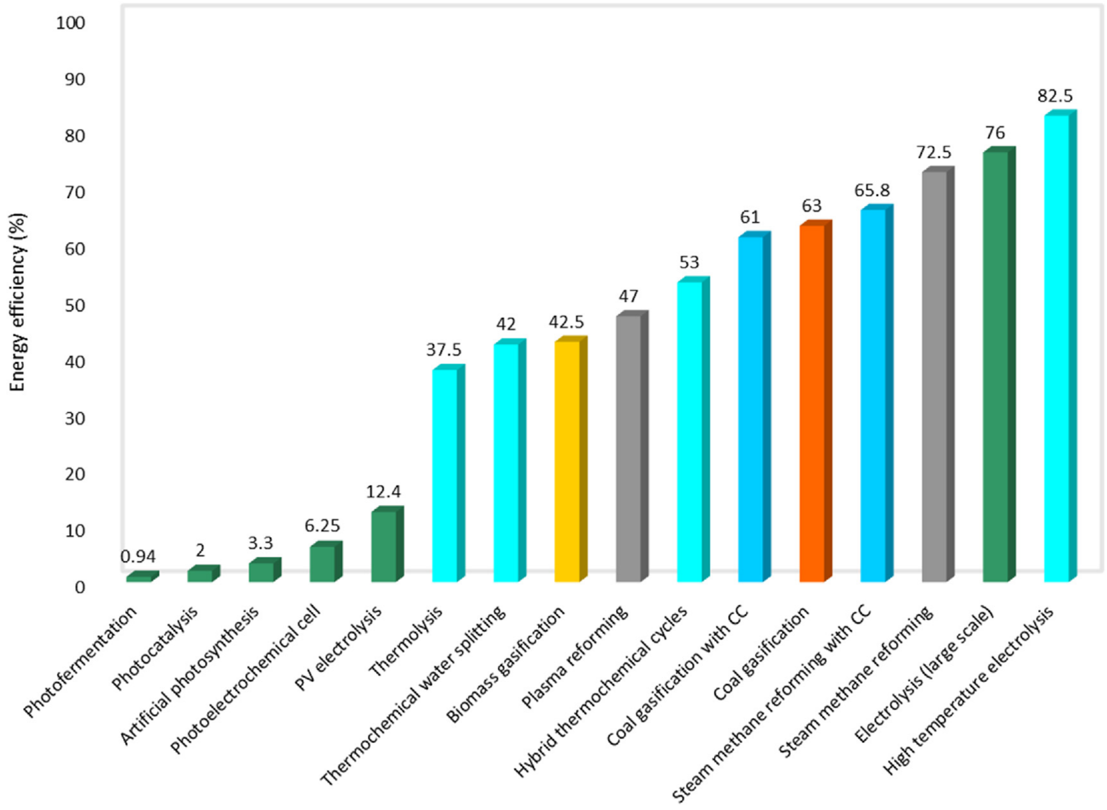


Fig. 3 – Energy efficiency comparison of the selected hydrogen production options.

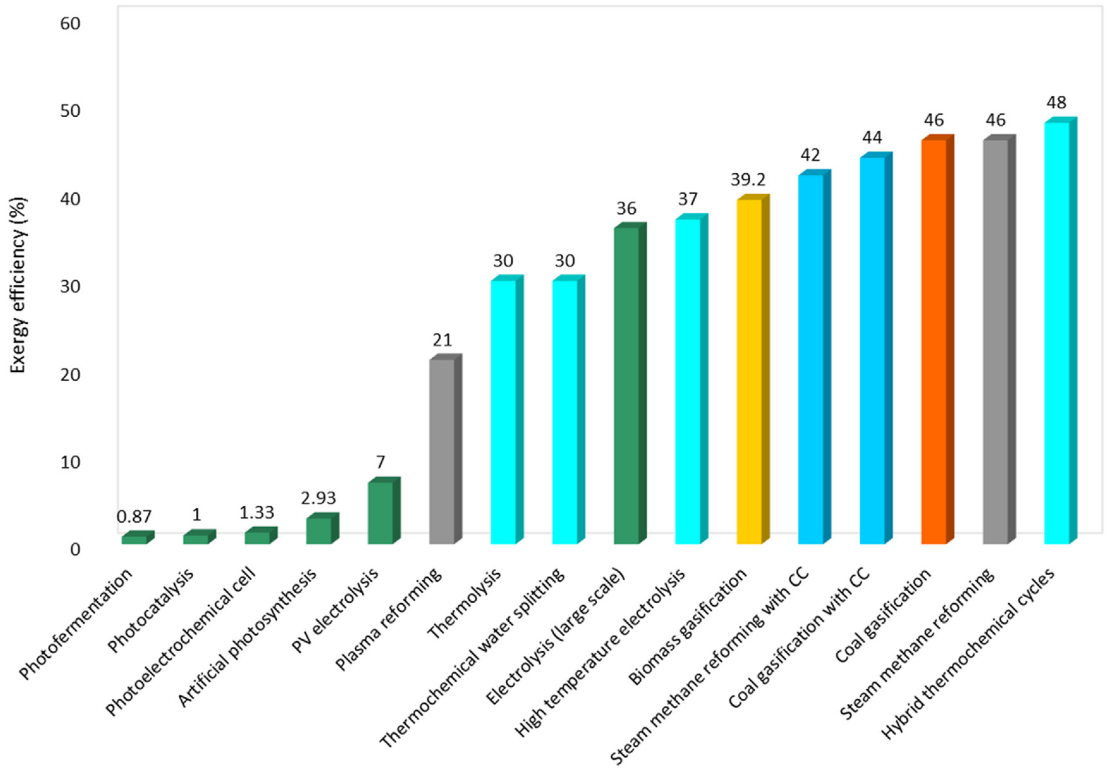


Fig. 4 – Exergy efficiency comparison of the selected hydrogen production options.

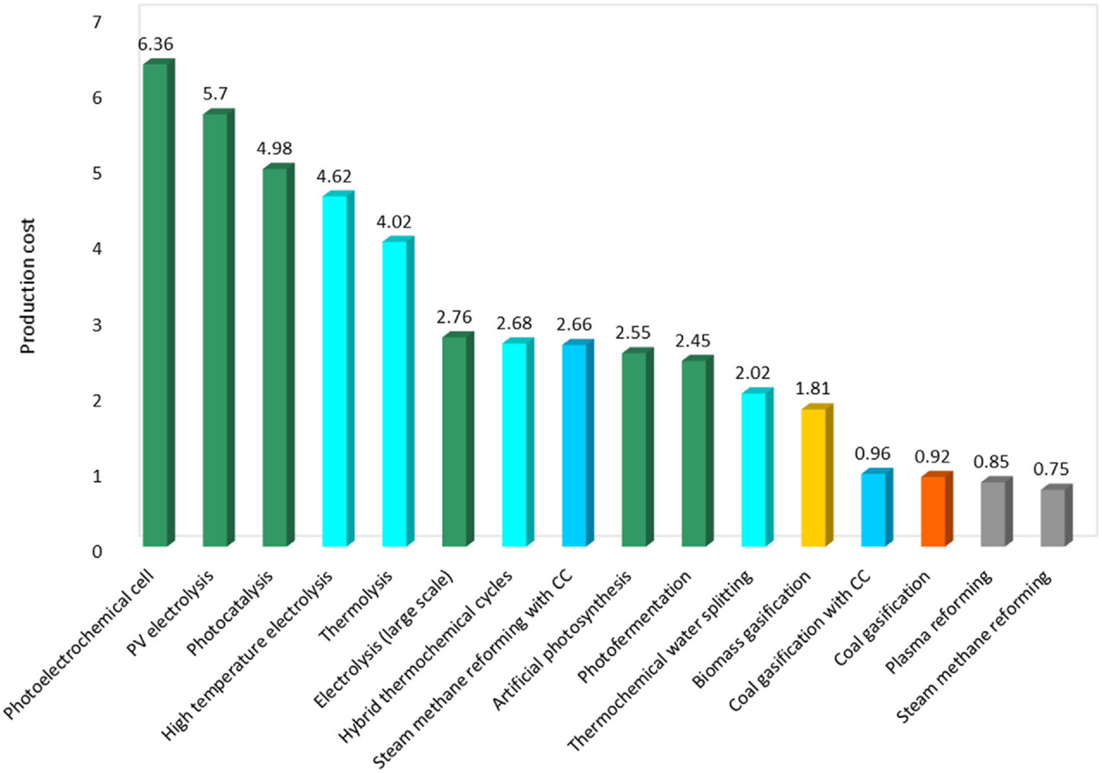


Fig. 5 – Production cost (US$/kg H_2) comparison of the selected hydrogen production options.

The production cost comparison (Fig. 5) shows that steam methane reforming has the lowest cost, followed by plasma reforming and coal gasification. Furthermore, the most expensive option is photoelectrochemical cells, followed by PV electrolysis and photocatalysis. The results show the clear economic advantage of large-scale, commercially developed hydrogen production methods over the relatively new, lab-scale options. The production cost of hydrogen production methods depends on various factors such as the energy input prices, capital or investment costs, technological maturity, and carbon prices. Undoubtedly, there are many uncertainties in the future costs of the selected hydrogen production options. The expectations show cost reduction in renewable, especially solar-based, hydrogen due to technological and engineering improvements, such as materials sciences towards highly efficient catalysts and more durability. Another expectation is that increasing carbon prices would make fossil-based hydrogen less affordable, increasing the cost-competitiveness of renewable hydrogen.

In the literature, the production costs of hydrogen are estimated based on the capital and operational costs. For the more expensive hydrogen production options such as PEC, PV electrolysis, and photocatalysis, the highest portion of the production cost belongs to the capital cost. Reducing the related capital costs through developing new materials and integrating them into the energy systems could potentially enhance the economic performance of the green hydrogen production methods.

The global warming potential, GWP, comparison (Fig. 6) shows coal gasification has the highest CO_2 emissions, followed by plasma and steam methane reforming. On the contrary, artificial photosynthesis, photofermentation, and photoelectrochemical cells have the lowest CO_2 emissions. As the results show, incorporating carbon capture technologies in fossil-based hydrogen production methods can significantly reduce their GWP. It is estimated that incorporating carbon capture can reduce the GWP of coal gasification by up to 80% and steam methane reforming by up to 70% [121]. Nevertheless, carbon capture technologies have not been demonstrated feasible in practical applications. Besides, even though carbon capture technologies could significantly reduce the global warming potential of fossil-based hydrogen production options, they may still not be considered as sustainable because of the depletion of non-renewable resources.

It should be noted that the results obtained from different literature sources have vastly different values for global warming and acidification potential values. The variation in the reported emissions is especially significant for smaller-scale hydrogen production options (e.g., green hydrogen). The variation may be due to the difficulty of obtaining normative parameters for methods not in large-scale production. The inventory data used for the evaluation are different or require more self-defined parameters. For this reason, in this study, average values of the reported data are used for CO_2 and SO_2 emissions.

The fifth criterion is acidification potential, AP (Fig. 7). The results show biomass gasification has the highest AP, followed by coal gasification. In contrast, photon-based options (artificial photosynthesis, photofermentation, photocatalysis, and photoelectrochemical cells) have the lowest AP. The results are pretty similar to the GWP because of the

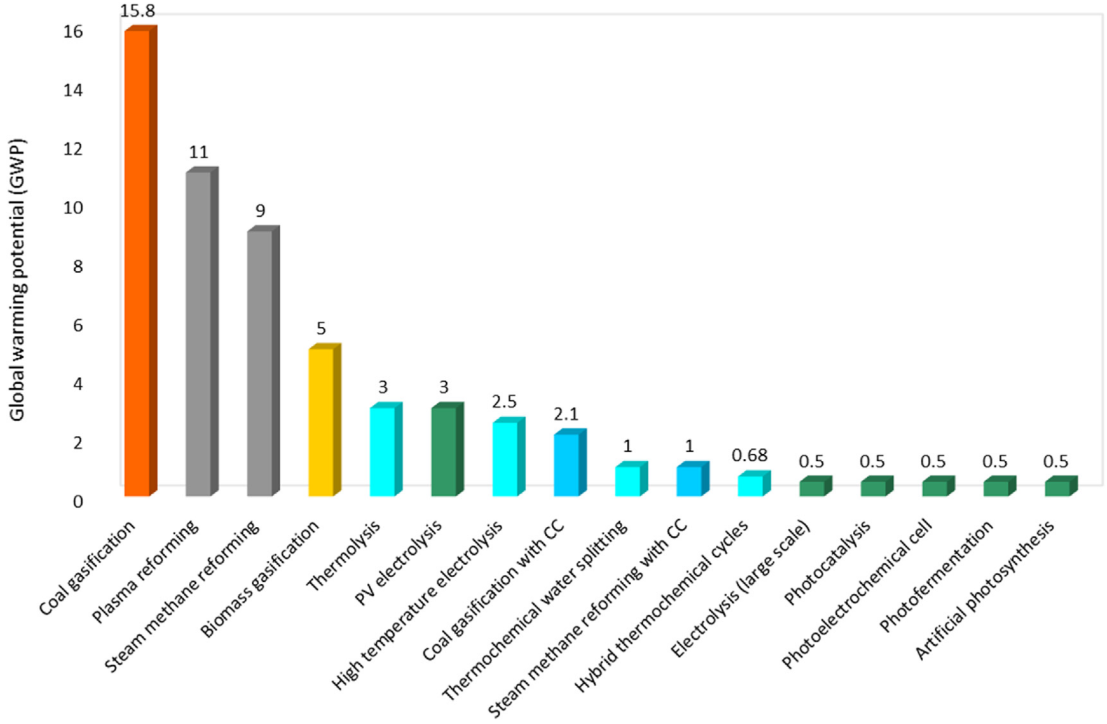


Fig. 6 – Global warming potential (kg CO_2/kg H_2) comparison of the selected hydrogen production options.

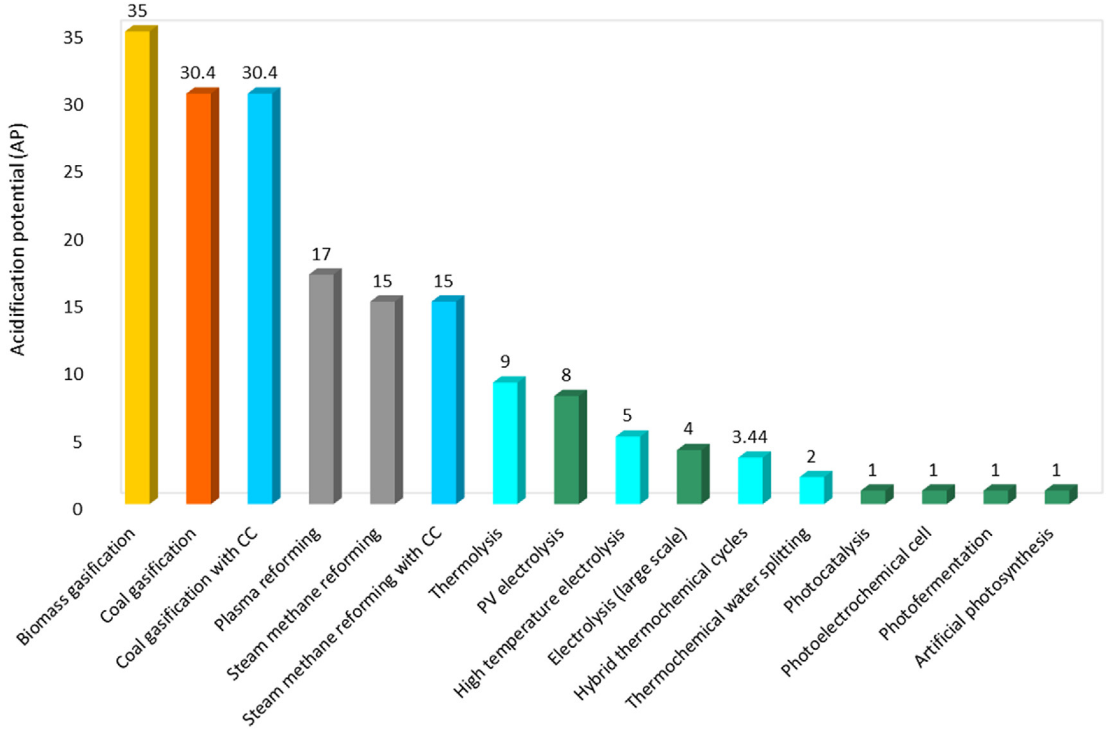


Fig. 7 – Acidification potential (g SO_x/kg H_2) comparison of the selected hydrogen production options.

high CO_2 and SO_x emissions of fossil fuels. The difference here is the biomass gasification performance. Although it can be considered carbon-neutral, biomass gasification has high SO_x emissions, which could endanger the water and land resources. The GWP and AP both depend on several factors, such as the primary energy and conversion process. In order to reduce the negative impact of hydrogen production on the environment, hydrogen should be produced from carbon-free sources via environmentally benign processes.

One remarkable outcome of the acidification potential assessment is biomass gasification performance. Biomass gasification has a relatively good performance in terms of global warming potential, but it has remarkably high acidification potential. Biomass gasification has lower CO_2 emissions than fossil fuel-based methods, but its CO_2 emissions are higher than green hydrogen production. When the current state of the art of green hydrogen production methods is taken into account, biomass gasification can offer an alternative option during the transition from fossil fuels to renewable sources.

The cost of carbon (CC) results (Fig. 8) show the same trends as the GWP data (Fig. 6). Coal gasification and plasma and steam reforming have the highest CC. On the other hand, photon-based options have the lowest SCC. This study takes CC as 160 USD per ton of CO_2 emissions. In the future, CC might increase, making photonic hydrogen more attractive in terms of its environmental, social, and economic advantages.

In the literature, it is anticipated that hydrogen production, just like electricity generation, could potentially face a carbon price for any emissions occurred onsite. Therefore, the cost of carbon could potentially be a significant criterion for estimating the sustainability performance of hydrogen production options. With the introduction of new carbon taxes and more strict carbon restrictions, fossil-based hydrogen production is estimated to become less favorable and sustainable, even for large-scale operations.

Fig. 9 shows the technology maturity level of the hydrogen production options. Large-scale and commercialized production options have the highest TML, such as steam methane reforming, coal gasification, and biomass gasification. Photonic hydrogen has the lowest TML. The biggest challenge with most sustainable hydrogen production methods includes the cost of hydrogen production, the maturity of the technology, and the scalability of the production process. Currently, a significant share of hydrogen production is from steam methane reforming, and it remains the most viable option for moving hydrogen into the energy market in the near term. Blue hydrogen is seen as the main route for low-carbon hydrogen production. The current cost of using this method is lower than green hydrogen production methods because of the existing high cost of capital investments required to transition to greener technologies.

Hydrogen production via steam methane reforming and coal gasification have higher technological maturity despite their significantly high global warming and acidification potentials. The dependence on natural gas and coal as finite resources and the necessity of long-term CO_2 storage in limited suitable geologic storage sites restricts the suitability of steam methane reforming and coal gasification as long-term solutions, even with carbon capture technologies. Nevertheless, hydrogen from steam methane reforming and coal gasification may be the bridging technologies that could facilitate the energy transition.

The results show that most of the better environmental performance methods have higher costs and lower TML. One significant challenge is to meet the environmental, social, technical, and thermodynamic requirements at once. Therefore, the following aspects should be considered in future research to conquer this challenge:

- Materials science and engineering research: Significant achievements are needed to accomplish substantial progress in theory. Equally, computer modeling and calculations are also helpful for predicting, evaluating, and

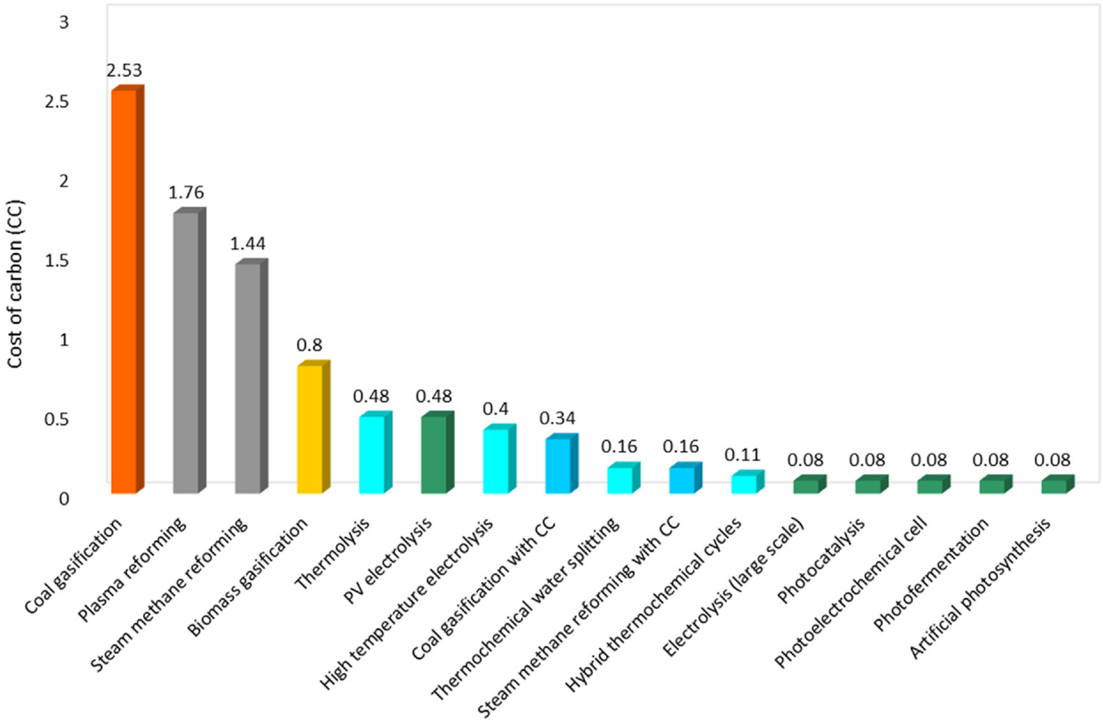


Fig. 8 – Cost of carbon (US$/kg H_2) comparison of the selected hydrogen production options.

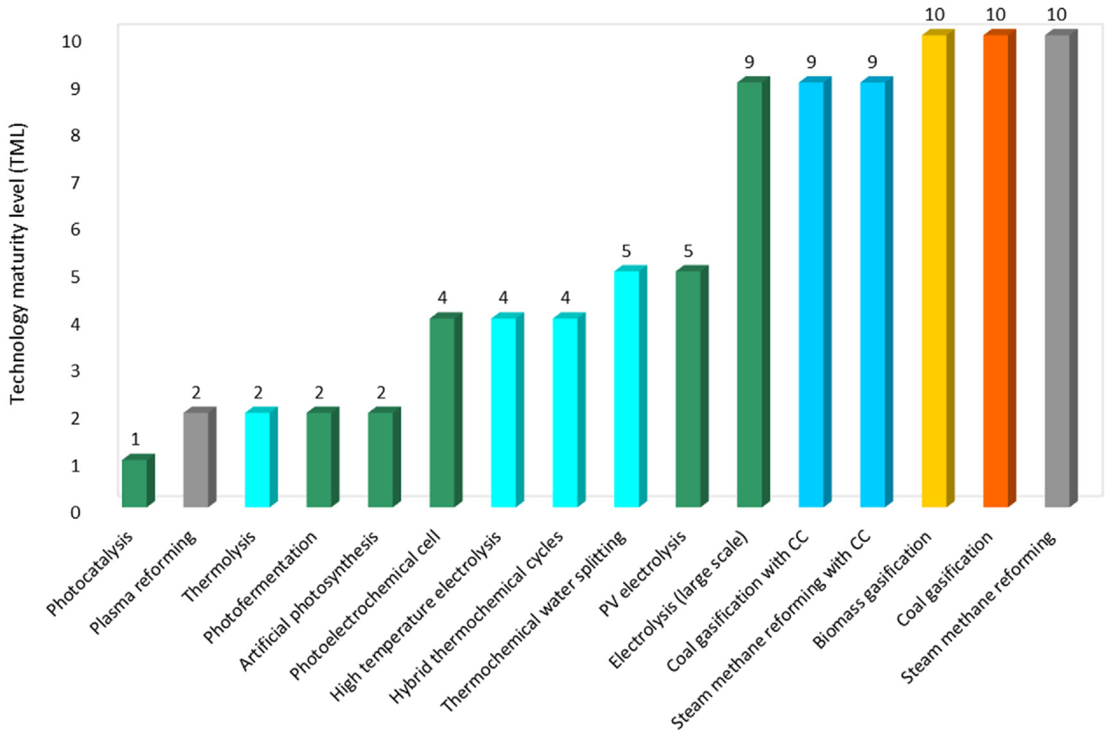


Fig. 9 – Technology maturity level (TML) comparison of the selected hydrogen production options.

optimizing different types of hydrogen-production methods. This aspect can guide the development of experimental routes and the optimization of process conditions, thereby improving the efficiency and reducing hydrogen production costs. The ultimate aim is to make large-scale green hydrogen more affordable, reliable, and accessible.

- Sustainable energy use and system integration: Current hydrogen production technologies are associated with complicated processes, high energy consumption, and costs. Therefore, the process coupling strategy is recommended for combining some essential processes to enhance the efficiency of hydrogen-production technologies, e.g., electrolysis in integrated energy systems. Moreover, the sustainability of the energy source and production process is critical. These measures could significantly reduce costs and the environmental impact of hydrogen production. Other advantages can be improving efficiency, operational flexibility, convenience, future market competition, and application prospects.

In addition to the TMLs, the technology readiness level (TRL) is considered a meaningful criterion to describe the extent of development necessary to reach the stage of commercialization. TRL ranges from theoretical principle, at TRL 1, to operational plant, at TRL 9 [33]. When evaluating the TRL, the minimum TRL of any technology component should be taken, as the limiting factor in development is the component with the lowest TRL. SMR, coal gasification, and large scale electrolysis are commercially available and have a TRL 9. More information on the TRL of the selected hydrogen production options can be found in Refs. [7,33].

The ultimate goal is to produce hydrogen in a clean, reliable, safe, affordable, and efficient manner. For this reason, the normalized rankings are comparatively presented in Fig. 10, which essentially indicates the ideal case.

In Fig. 10, the total ranking of each method is given as sustainability performance. For each criterion, the selected methods rank between 0 and 10. The ideal case has a total ranking, a sustainability performance, of 70, while the lowest possible performance would be 0. With this approach, the methods that have a total that is closer to 70 have a higher sustainability performance. The rankings are normalized based on the procedure explained in Equations (3) and (4). Therefore, there are no units in the ranking results.

Fig. 10 shows that large-scale electrolysis has the closest performance to the ideal case when all criteria are taken into account. Steam methane reforming with carbon capture has the second high score. It should be noted that this option has lower energy and exergy efficiencies, and it is more expensive than steam methane reforming without carbon capture. Developing more efficient, cheaper, and practical carbon capture technologies that support large-scale hydrogen generation via steam reforming could significantly accelerate the transition towards hydrogen energy. Thermochemical and hybrid thermochemical cycles also show promising performance. Their sustainability performance depends on the thermal and electrical energy sources and the catalyst performances. In Fig. 10, all performance criteria are assumed to be equally important.

In Fig. 11, several case studies are generated to identify the most and least desirable hydrogen production methods. There are eight different case studies in Fig. 11. All performance criteria are equally important in the first case study

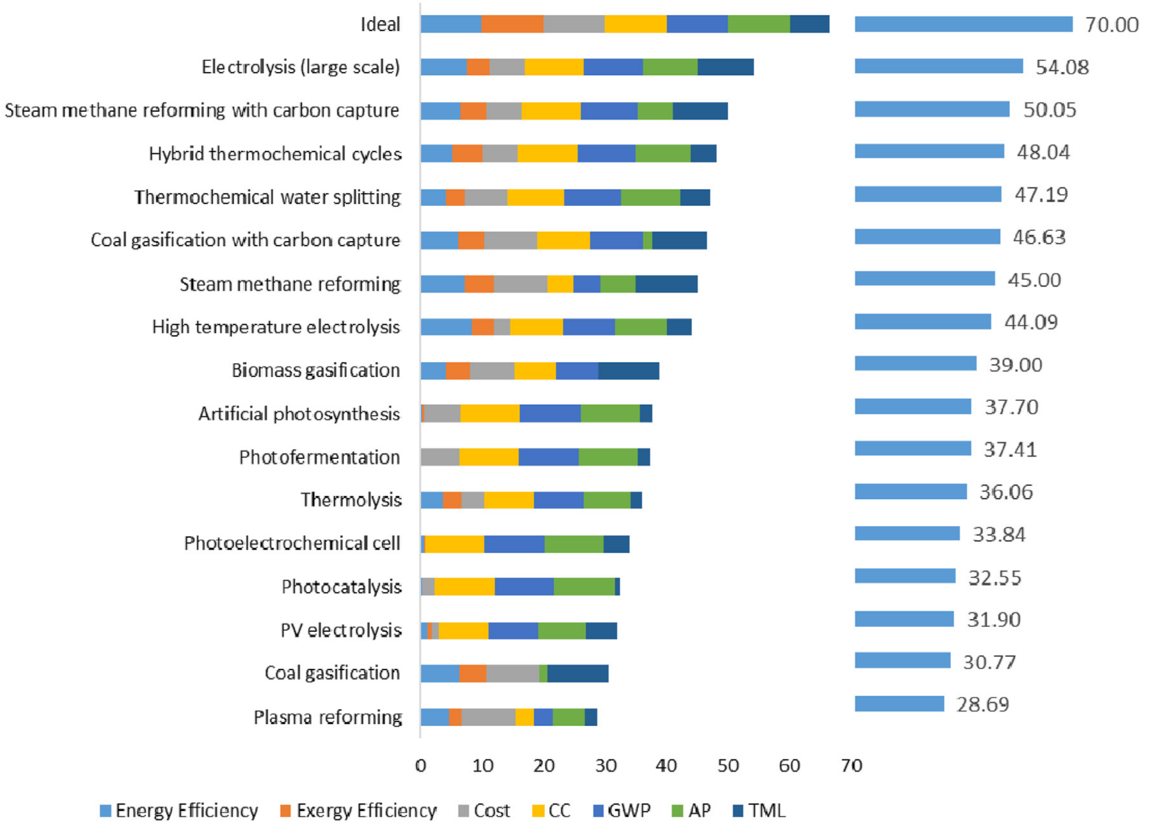
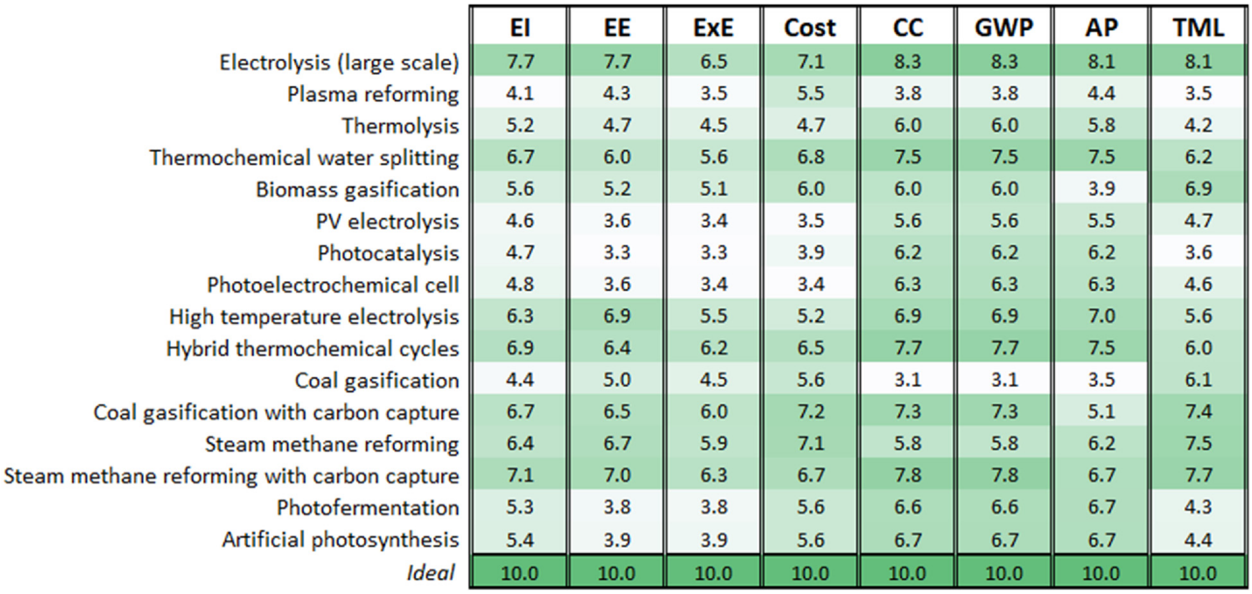


Fig. 10 — Overall sustainability performance comparison of the selected hydrogen production methods and the hypothetical ideal case.

(EI). In the other cases, one criterion is chosen to be the most critical, with a weight of 0.4 on the final grade, while the remaining six criteria are assigned the weight of 0.1. For instance, in the second case study (EE), energy efficiency has the highest weight (0.4), and the remaining criteria have equal weights (0.1). Exergy efficiency has the highest weight in the third case study (ExE) case study. Cost is the fourth case, where production cost has the highest importance. CC has the highest impact on the final decision in the fifth case.



	EI	EE	ExE	Cost	CC	GWP	AP	TML
Electrolysis (large scale)	7.7	7.7	6.5	7.1	8.3	8.3	8.1	8.1
Plasma reforming	4.1	4.3	3.5	5.5	3.8	3.8	4.4	3.5
Thermolysis	5.2	4.7	4.5	4.7	6.0	6.0	5.8	4.2
Thermochemical water splitting	6.7	6.0	5.6	6.8	7.5	7.5	7.5	6.2
Biomass gasification	5.6	5.2	5.1	6.0	6.0	6.0	3.9	6.9
PV electrolysis	4.6	3.6	3.4	3.5	5.6	5.6	5.5	4.7
Photocatalysis	4.7	3.3	3.3	3.9	6.2	6.2	6.2	3.6
Photoelectrochemical cell	4.8	3.6	3.4	3.4	6.3	6.3	6.3	4.6
High temperature electrolysis	6.3	6.9	5.5	5.2	6.9	6.9	7.0	5.6
Hybrid thermochemical cycles	6.9	6.4	6.2	6.5	7.7	7.7	7.5	6.0
Coal gasification	4.4	5.0	4.5	5.6	3.1	3.1	3.5	6.1
Coal gasification with carbon capture	6.7	6.5	6.0	7.2	7.3	7.3	5.1	7.4
Steam methane reforming	6.4	6.7	5.9	7.1	5.8	5.8	6.2	7.5
Steam methane reforming with carbon capture	7.1	7.0	6.3	6.7	7.8	7.8	6.7	7.7
Photofermentation	5.3	3.8	3.8	5.6	6.6	6.6	6.7	4.3
Artificial photosynthesis	5.4	3.9	3.9	5.6	6.7	6.7	6.7	4.4
Ideal	10.0	10.0	10.0	10.0	10.0	10.0	10.0	10.0

Fig. 11 — Detailed sustainability performance rankings of the selected hydrogen production in different cases and the hypothetical ideal case.

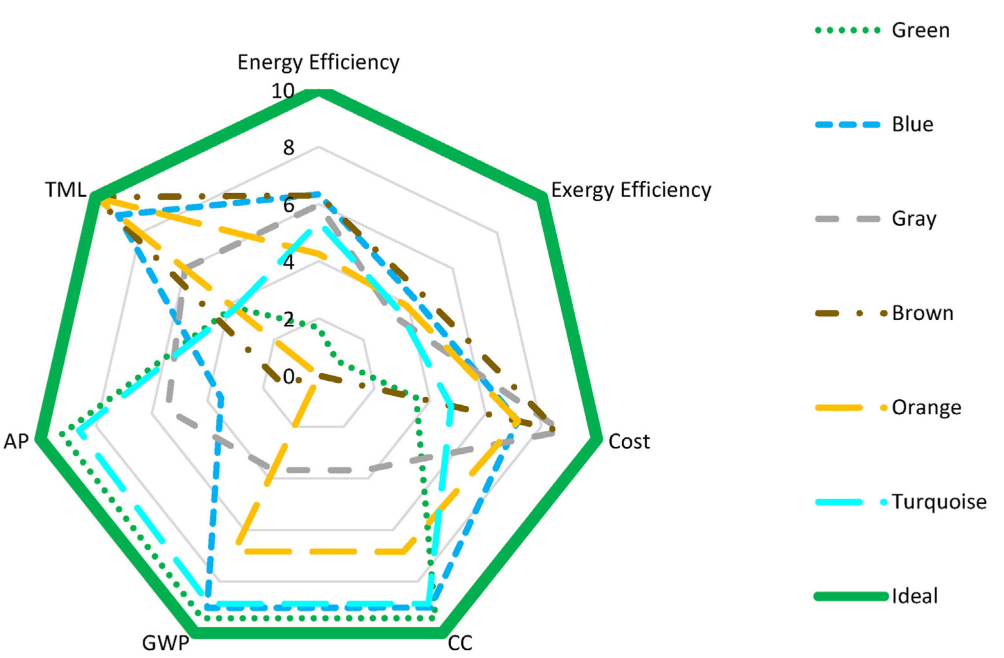


Fig. 12 – **Overall performance comparison of selected hydrogen production color groups. (For interpretation of the references to color/colour in this figure legend, the reader is referred to the Web version of this article.)**

Similarly, the parameters, namely GWP, AP and TML, have the highest importance in each one of the remaining cases. The results show that large-scale electrolysis has the highest sustainability performance in almost all cases. There is one exception, the fourth case. Production cost is the determining factor in the fourth case, and coal gasification with carbon capture has the highest score.

When the overall performance of green, blue, gray, brown, orange, and turquoise hydrogen production methods are compared (Fig. 12), it can be seen that green hydrogen production methods have higher performance in terms of environmental criteria. However, their energy and exergy efficiencies, cost, and technology maturity level performance are relatively low. On the contrary, blue hydrogen production methods have the highest energy efficiency performance. Gray hydrogen production methods have the highest economic performance. Brown hydrogen production methods have the highest exergy efficiency and the technology maturity performance; however, they also have the least desirable performance in terms of cost of carbon and the global warming potential. Similarly, orange hydrogen has high technology maturity performance but the least attractive performance in acidification potential. In terms of technology maturity level, turquoise hydrogen has the lowest performance, followed by green hydrogen.

Overall, when their average performances are taken into account, blue hydrogen has the highest sustainability score (6.91/10), followed by turquoise hydrogen (6.26/10), orange hydrogen (5.57/10), green hydrogen (5.42/10), gray hydrogen (5.26/10), and brown hydrogen (4.40/10). The results show the need to find alternatives to coal-based hydrogen. Green hydrogen production methods require research and development to enhance their technoeconomic performance. The short-term goal for green hydrogen should be to minimize the costs, and the long-term goal should include enhancing exergy efficiency, performance, and durability.

Research and development requirements for sustainable hydrogen production can be summarized as more comprehensive technoeconomic analyses, reliability enhancement, effective and feasible integration with renewables, advanced materials, innovation in system design and operation, and intelligent system control and integration. The technoeconomic analysis models should consider the entire lifecycle and economic impact of emissions, pollution, and nonrenewable resource degradation. Figs. 10–12 are generated not only to highlight the strengths and weaknesses of the selected hydrogen production options but also to underline the research and development opportunities related to each method. The different cases shown in Figs. 11 and 12 can help decision-makers, industrial professionals, and researchers identify research directions for different scenarios with different priorities.

Conclusions

This study comparatively assesses the sustainability performance of selected hydrogen production methods based on their technical, economic, thermodynamic, social, and environmental aspects. The main findings obtained from this study are sixfold as follows:

- The high temperature electrolysis and photofermentation have the highest and lowest energy efficiency, respectively.
- In terms of exergy efficiency, the hybrid thermochemical cycles and photofermentation are the most and least efficient ones.

- The global warming potential comparison indicates coal gasification has the highest while photofermentation has the lowest CO_2 emissions.
- Based on acidification potential, the artificial photosynthesis has the lowest, and biomass gasification has the highest SO_x-based emissions.
- The PEC has the highest hydrogen production cost, while steam methane reforming has the lowest.
- The cost of carbon comparison indicates coal gasification has the highest and photofermentation has the lowest cost.

Overall performance is ranked based on different case studies. In one case, all criteria have equal importance. In the other cases, in each case, one criterion has significantly higher importance than the rest, which have equal importance. In almost all cases, large-scale electrolysis shows the highest performance. There is one exception. When cost is the most crucial criterion, coal gasification with carbon capture has slightly better performance.

In this study, the sixteen hydrogen production methods are compared, and their study results point out the research-development-innovation needs, strengths, weaknesses, and opportunities related to each method. For future studies, additional sustainability criteria are recommended to be taken into account, such as scalability, rare material use, system lifetime, recyclability, response time, production rate, gross energy requirement, net utilizable energy product, and suitability for the circular economy (e.g., hydrogen production from waste streams). In addition, more biological hydrogen production processes are recommended to be considered in the sustainability evaluation, including dark fermentation.

In closing, this study further intends to showcase the strengths and weaknesses of several hydrogen production pathways for more sustainable operation. Another aim of this study is to highlight the research opportunities for renewable-based hydrogen production. In conclusion, this study shows that considerable efforts, new approaches, and innovative ideas are needed to decarbonize energy systems with hydrogen. The results can help governments, industry specialists, manufacturers, policymakers, researchers, scientists, and utility companies deploy a hydrogen roadmap for a more sustainable future.

Declaration of competing interest

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

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