



A comprehensive review on environmental and economic impacts of hydrogen production from traditional and cleaner resources

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ARTICLE INFO

Editor: Y. Liu

Keywords:

Green hydrogen
Environmental sustainability
Renewable
Non-renewable
Global warming potential

ABSTRACT

This review paper considered the potential hydrogen (H₂) production methods using conventional fossil fuels and in a cleaner manner with biomass and water resources and evaluated them for economic sustainability, environmental impact, and energy efficiency. The study results revealed that the methods of biomass-based hydrogen production (e.g., photo-fermentation (PF), dark fermentation (DF), and microbial electrolysis cell (MEC)), by energy source, appear to more environmentally friendly than the other evaluated methods in terms of emissions since they offer the potential to significantly reduce CO₂ releases when their substrates are derived from renewable resources or wastes. Among the biomass-based processes, the PF is the most environmentally friendly H₂ production process, presenting a low global warming potential (GWP) value of 1.88 kgCO₂ eq./kgH₂ and acidification potential (AP) of 0.003 gSO₂/kgH₂, it is followed by DF and MEC processes. On the other hand, the highest GWP of 19.85 kgCO₂ eq./kgH₂ and AP 0.139 kgSO₂/kg H₂ were obtained for the fossil fuel-based gasification process related to coal mining and transportation operations. Although hydrogen production processes seem to consume high amounts of water sources, such as about 9 kg of water consumed for 1 kg of hydrogen produced during conventional electrolysis, the reality is that in the hydrogen ecosystem the water footprint of the process is reduced drastically where hydrogen is employed as fuel in fuel cell systems and converted back to water while generating electricity. So, the hydrogen ecosystem may diligently be recognized as the water conserving cycle. On the other hand, the study results showed that commercially available fossil fuel-based (e.g., coal) gasification and steam-methane reforming processes are more advantageous over other lab-scale technologies in terms of cost and process efficiency. Nevertheless, rising carbon costs may reduce the reasonable price of fossil-based H₂ and promote the cost-competitiveness of biomass-based renewable H₂. Overall ranking results also proved that biomass-based H₂ production processes are primarily promising options for H₂ production in an environmentally friendly and moderately cost-effective way.

1. Introduction

Energy consumption has continued to increase exponentially since the industrial revolution as a result of global population growth, introduction of new technologies, and rapid economic development. It is projected that the energy demand of world will increase by 48% within

the following 20 years as a result of the rapid increase in population [1]. Currently, 80% of the energy demands are provided by fossil fuels in worldwide. However, fossil fuel resources are reducing swiftly, strengthening the stress on available resources daily related to increasing energy demand. Moreover, one of the most significant environmental problems associated with using fossil fuels is the release of

Abbreviations: AE, alkaline electrolysis; AEM, anion exchange membrane; AP, acidification potential; CEM, cation exchange membrane; DF, dark fermentation; FE, freshwater ecotoxicity; FEP, freshwater eutrophication potential; FRS, Fossil resource use; GHGs, greenhouse gases; GWP, global warming potential; H₂, hydrogen; IR, ionizing radiation; LU, land use; MEC, microbial electrolysis cell; OD, ozone depletion; PEM, proton exchange membranes; PF, photo-fermentation; SOE, solid oxide electrolysis; WCP, water consumption potential; WSF, water scarcity footprint; ΔG, free energy.

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<https://doi.org/10.1016/j.jece.2023.111187>

Received 27 July 2023; Received in revised form 9 September 2023; Accepted 4 October 2023

Available online 5 October 2023

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greenhouse gases (GHGs), which contribute to global warming and concerns about climate change [2]. Namely, producing energy from carbon-containing fossil fuel resources damages the environment [3]. Therefore, a reliable and accessible energy supply is critical for the sustainability of modern communities. There is an essential need to quickly transition from conventional to renewable energy systems that are sustainable and able to fulfill the current and future global energy demand [4]. In this regard, various types of renewable energy resources may be utilized to prevent the problems of environmental sustainability and energy requirements. Recently, hydrogen (H_2) has gained significant interest as the best alternative among cleaner fuels, with its benefits over conventional fuels. As an energy vector, H_2 is believed to be the best option. It is an alternate type of energy that is produced by using three different energy-supply methods, such as nuclear reactors involving fission reactors and producers, renewable sources of energy (hydroelectric power, wind power systems, ocean thermal energy, biomass, etc.), and fossil fuels (coal, natural gas, petroleum, oil, etc.). Consequently, H_2 is widely accepted as an environmentally friendly secondary source of renewable energy and an attractive substitute for fossil fuels since it is the only carbon-free fuel with the largest energy content of any fuel currently in use [5].

Currently, the majority of H_2 production across the world is based on methods that extract H_2 from fossil fuel resources using commercialized gasification, steam reforming, and pyrolysis processes [6]. However, these conventional H_2 production technologies are not adequately sustainable since they heavily depend on fossil fuels that cause carbon emissions. Methods to produce H_2 without emitting CO_2 will be required as concern over potential climate change and reductions in GHG emissions in response to the Kyoto Protocol. Thus, it is crucial to consider the entire chain of activities for producing and using H_2 to determine if using H_2 instead of alternative energy sources will increase or decrease in GHG emissions. To tackle these challenges and provide promising solutions in terms of environmental sustainability, H_2 production from water using electrolysis and biomass (algae, agricultural residues, organic food wastes, and energy crops) using biological processes has attracted attention as sustainable and environmentally friendly technologies in recent years [7]. Moreover, when compared to the fossil fuel-based and water-based H_2 production methods, the H_2 production from biological processes such as dark fermentation (DF), photo-fermentation (PF), microbial electrolysis cells (MECs), and integration of these processes is much cleaner methods thanks to the simultaneous production of energy and the minimization of waste by using various biomass residues as a substrate [8].

In the literature, several review studies focus on various aspects of H_2 production processes and their resources [9]. However, assessing the environmental impacts of the H_2 production processes, considering their emissions into the environment and the associated effects of H_2 production, is essential for defining realistic objectives for emissions reduction [10]. Several previous investigations only concentrated on various production methods while considering the effects of catalysts or other operational parameters on process performance in order to highlight the environmental and economic aspects of H_2 production [11,12]. However, there is no detailed study on the economic and environmental evaluations of H_2 production processes using different resources, according to our humble opinion. For instance, most researchers considered global warming potential (GWP) and acidification potential (AP) as environmental impact parameters [13]. On the other hand, water utilization, fossil resource use (FRS), ionizing radiation (IR), ozone depletion (OD), freshwater ecotoxicity (FE), freshwater eutrophication potential (FEP), and land use (LU) are important environmental impact parameters to evaluate the performance of the H_2 production processes. Therefore, there still needs to be a knowledge gap in estimating these impacts of H_2 production processes for different resources as the power of the system, especially by means of direct environmental impact data. Overall, the assessment of H_2 production from fossil fuel, biomass, and water resources using different processes in terms of their environmental

impacts and economic sustainability is crucial to provide cleaner and sustainable pathways for H_2 production.

To our knowledge, this research is the first and most comprehensive review of the environmental and economic impacts of H_2 production methods from various resources. This review paper aims to present a comprehensive environmental impact and economic comparison of H_2 production processes utilizing different resources as substrates. In this regard, seven H_2 production technologies (gasification, steam reforming, DF, PF, MEC, DF-MEC hybrid, and electrolysis) are compared, considering their environmental impacts, including AP, FE, FRS, FEP, GWP, LU, OD, IR, water consumption potential (WCP), water scarcity footprint (WSF). Overall, this study presents insight into the industrial applicability of H_2 production processes from various resources by evaluating environmental sustainability. Overall purposes of this study were (i) to discuss the advantages, possible drawbacks, and future directions of specified thermochemical and biological H_2 production processes for different resources, (ii) to assess the economic and environmental impacts of processes, (iii) to compare the environmental impacts and economic sustainability of thermochemical and biological processes from biomass, fossil fuel, and water resources, and (iv) to determine the level of sustainability of the processes with a normalized ranking method. Consequently, this review paper provides insight into the real-scale applicability of processes for effective and sustainable H_2 production by assessing their environmental impacts and economic affordability.

2. Methods for H_2 production

H_2 can be produced from various resources, such as biomass, fossil fuels, and water [14]. The energy must be available in excess amounts and continuously accessible to extract H_2 from these sources [15]. Therefore, sustainable H_2 production would become possible by using the potential of renewable energy resources (solar, wind, hydroelectric power, geothermal, etc.) in H_2 production processes [16]. Fig. 1 presents the net energy production from coal, natural gas, petroleum, nuclear electric power, and renewable resources [17]. The results revealed that coal had been widely utilized for energy production and remained the dominant energy resource from 1995 to 2005 (Fig. 1a). On the other hand, the utilization of natural gas for energy purposes has increased significantly. The energy produced from natural gas in 2022 has been reported as almost 140,000 million kWh, while no significant change was observed in petroleum and nuclear power use.

Their results proved that a huge amount of the energy demand in 2022 was provided by using natural gas. Furthermore, considering the renewable resources in energy production, it is observed that wind energy is widely used in 2022, and biofuel, hydroelectric power, and wood resources follow the wind as renewable energy sources (Fig. 1b). Overall, this section considered the main H_2 production methods regarding raw material utilization: biomass, fossil fuels, or water, to understand the environmental and economic impacts of selected H_2 production processes. A brief description of the evaluated processes, their advantages and disadvantages, and current progress status with other significant remarks were also considered in this section.

2.1. Fossil fuels for H_2 production

Fossil fuels remain the main source of the H_2 supply because of the close correlation between production costs and fuel prices, which are currently maintained at reasonable levels. Hydrocarbon reforming, gasification, and pyrolysis are currently the most popular methods for producing H_2 from fossil fuels allowing the production of the actual demand [5]. The main remarks, type of utilized feedstock, and the maturity of fossil fuel-based H_2 production processes reported in the following subsection are summarized in Table 1.

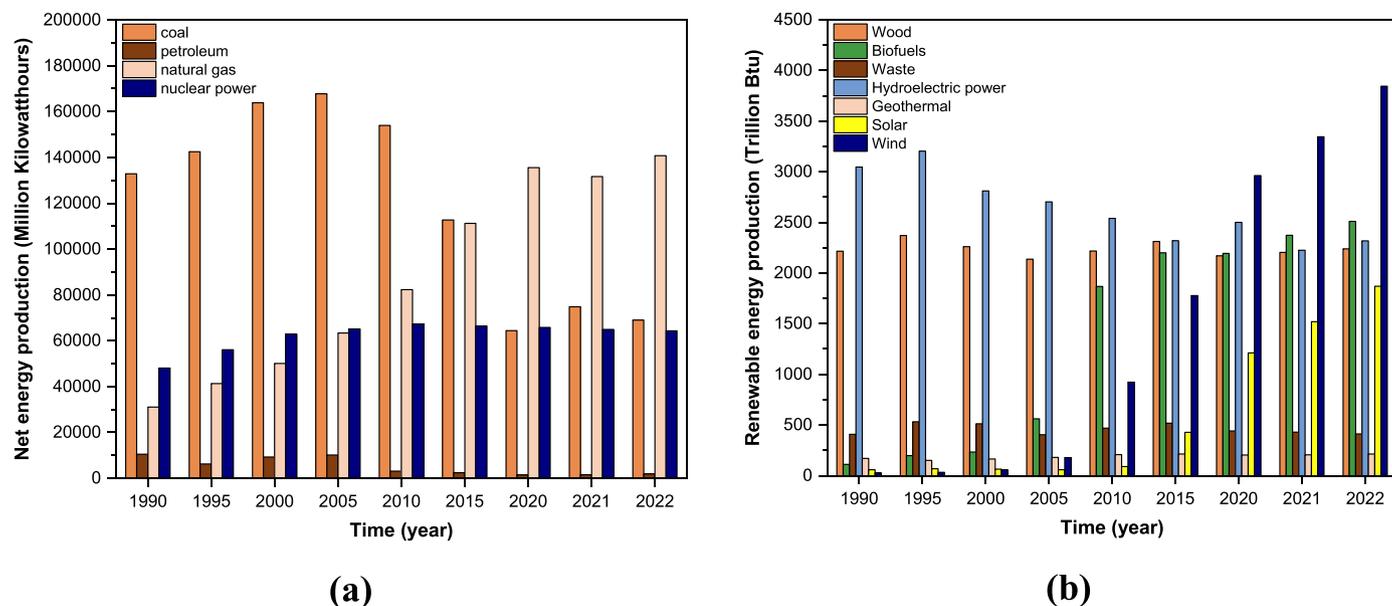
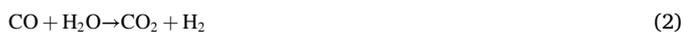


Fig. 1. Net energy generation from nonrenewable and renewable energy sources between 1990 and 2022 inclusive.

2.1.1. Reforming processes

Fundamentally, reforming processes consist of partial oxidation, autothermal, and steam-methane reforming (SMR) methods. In the steam reforming process, the reaction of a mixture of hydrocarbons and steam occurs to produce H_2 and carbon oxides at high temperatures. Natural gas is utilized in steam reforming to obtain H_2 , which is rarely performed with liquefied naphtha and petroleum. The SMR from natural gas or light hydrocarbons is the most often utilized hydrocarbon reformation technique. In this method, H_2 is first produced with carbon monoxide (Eq. (1)), providing an increase in synthesis gas. Then carbon monoxide is converted into carbon dioxide and H_2 with the water-gas shift reaction (Eq. (2)). Overall steam methane reforming reactions (Eq. (3)) are represented as follows:



In steam reforming, the H_2 production reactions are endothermic, and reactions need an effective heat source. Hence, commercial steam reformers usually perform in the range of 3.0–25 bars of pressure and temperatures of 800–1100 °C [20]. On the other hand, the high-temperature requirement is one of the most important drawbacks of the steam reforming process because high temperatures cause the requirement of expensive construction components for the reformer to prevent thermal stress [21]. Furthermore, developing temperature profiles in the catalyst field and coke formation may also need to be tackled [22].

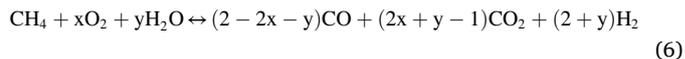
In recent years, partial oxidation processes have gained a lot of interest, which involves heating methane, natural gas, or another hydrocarbon in the presence of a stoichiometric quantity of pure oxygen. When less oxygen than required for stoichiometry is present, the reaction's byproducts are mostly H_2 and carbon monoxide (and nitrogen, if air is used instead of pure oxygen), with just a trace quantity of carbon dioxide and other molecules. The carbon monoxide then combines with water in a water-gas shift reaction to produce carbon dioxide and additional H_2 . Usually, the partial oxidation process is significantly faster than steam reforming and only needs a small reactor vessel [23]. This method initially generates less H_2 per unit of the input fuel than is generated by steam reforming of the same fuel, as can be demonstrated

by chemical reactions of partial oxidation. The overall partial oxidation reactions (Eqs. (4)–(5)) for H_2 production are presented as follows:



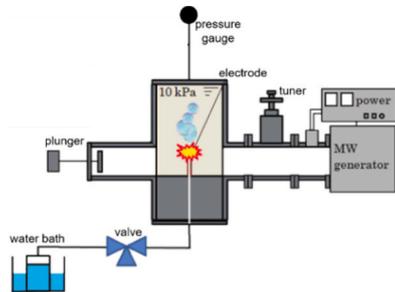
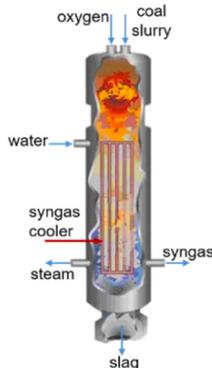
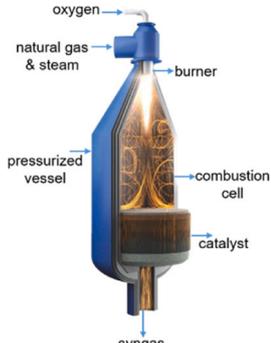
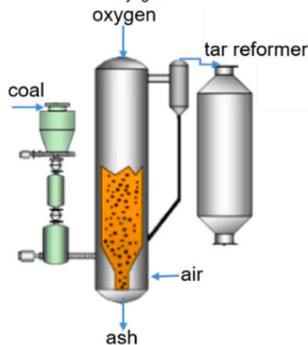
The method of partial oxidation of methane has several benefits over steam reforming, including quick response times, compact size, and less sensitivity to fuel variables. In the field of energy conversion, partial oxidation reactors, particularly in high-temperature fuel cells, are becoming increasingly popular. Moreover, without the use of a catalyst, the partial oxidation of hydrocarbon to CO and H_2 can take place at extremely high temperatures; however, the operating temperature will be greatly lowered with the use of catalysts. The most researched catalysts for the partial oxidation process include supported transition metals (Co, Fe, and Ni), noble metals, and perovskite oxides [24]. The metal species with various oxidation states play a crucial role in the methane conversion processes although transition metals are active catalyst components for the partial oxidation of methane. For instance, metallic nickel is used in a Ni-supported catalyst to promote syngas production, and Ni species with an oxidation number of ≥ 2 will stimulate methane burn completely [25]. It was found that perovskite-based catalyst materials for partial oxidation of methane showed strong catalytic activity, excellent stability, and the capacity to prevent carbon deposition. According to some reports, the perovskite catalyst can lower the threshold value required for generating carbon deposition [26].

Furthermore, the general form of the reaction may be represented as follows when it comes to the autothermal reforming of methane.



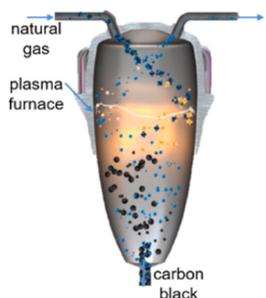
where x and y represent, the oxygen/methane (O/C) and steam/methane (S/C) ratios, respectively. The ratios are theoretically capable of control at $0 < x \leq 1$ and $0 < y \leq 2$. The thermo-neutral condition of the autothermal reforming of methane was reported to occur at $x = 0.025$ under the reaction temperature parameters of 400 °C and $2x + y = 1$, meaning that the production of CO_2 is neglected [27]. The thermo-neutral situation will occur at a lower value of x ($x < 0.025$) if the generation of CO_2 is taken into account. In other words, except for a small amount of oxygen, the autothermal reforming is an exothermic reaction. However, if the preceding reaction is triggered in the absence

Table 1
A list of common hydrogen production methods from fossil fuels (adapted from [5,18,19]).

Process		Advantages-Disadvantages	Maturity
Reforming	Steam methane reforming 	<ul style="list-style-type: none"> Utilizes light hydrocarbons Requires expensive high-temperature materials 	Mature
	Partial oxidation 	<ul style="list-style-type: none"> Uses hydrocarbons, oil, and coal Requires a smaller reactor vessel Provides low hydrogen production rate Causes fast reaction kinetics Requires costly high-temperature materials Depends upon fossil fuels Needs no catalyst 	Mature
Gasification	Autothermal 	<ul style="list-style-type: none"> Uses light hydrocarbons Requires low energy Requires expensive high-temperature materials Depends upon fossil fuels Results in lower heat requirement than partial oxidation Uses existing infrastructure 	Early market
		<ul style="list-style-type: none"> Utilizes coal Needs to supply heat Requires fossil fuels Causes carbon emissions Provides high H₂ production yield Achieves low-cost syngas production Holds feedstock impurities Requires carbon capture and storage 	Mature

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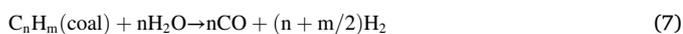
Table 1 (continued)

Process	Advantages-Disadvantages	Maturity
	<ul style="list-style-type: none"> • Utilizes hydrocarbons and natural gas • Prevents emissions with renewable energy • Reduces step procedure • Provides carbon-based materials • Depends upon fossil fuels 	Mature

of oxygen (i.e., $x = 0$), the exothermic autothermal reforming of methane becomes the endothermic steam methane reforming reaction. Reviewing previous research revealed that the O/C and S/C ratios were retained within the ranges of 0–1 and 0–6, respectively [28,29]. The reaction temperature was typically maintained below 900 °C. The H₂ yield (moles of H₂ produced from each mole of CH₄ reaction) may therefore reach 2.6. Overall, the autothermal reforming for methane has the advantage of being easy to use. Note that, reaction management is here somewhat challenging, though, because the S/C ratio and the O/C ratio are operating simultaneously. Additionally, the two impacted parameters significantly impact the reaction temperature of autothermal reforming process. In contrast, because of the two-stage response, the partial oxidation process in conjunction with water gas shift reaction is more complicated than the autothermal reforming technique. However, the methane conversion and reaction temperature in partial oxidation processes are often high. The heat released from partial oxidation may be used to start water gas shift reaction, and the amount of CO it converts likewise increases. As a result, in comparison to the one-stage reaction, the two-stage reaction operates with an acceptable degree of flexibility.

2.1.2. Gasification

Gasification is a technology that converts carbon-based raw materials, such as biomass, coal, and petroleum coke, into synthetic gas (carbon monoxide, H₂, and CH₄) using air, water vapor, or oxygen [30]. This section particularly aims to report the coal-utilized gasification processes for H₂ production since we compared biomass and fossil fuel-based resources regarding their environmental and economic impacts. It is reported that four various coal types, including anthracites, bituminous coals, sub-bituminous coal, and lignite, are widely used in the gasification process [31]. The published literature indicates that these materials are typically gasified at temperatures > 900 °C using the following techniques: (i) entrained flow gasification, (ii) fixed bed gasification, (iii) fluidized bed gasification, (iv) moving bed gasification, and (v) plasma gasification. In comparison to conventional coal combustion processes, coal gasification points to be a major technique for producing energy and other chemical products in a cleaner and more economical manner. Moreover, high ash content and moisture-containing coal is effectively converted into valuable products by coal gasification [32]. Moreover, as a result of coal gasification, energy may be provided with greater efficiency using H₂-rich gas [33]. Therefore, gasification-based methods are the most effective and ecologically benign methods for producing inexpensive energy and other products, which may also be easily modified for CO₂ storage and sequestration. The primary reaction in industrial H₂ production using coal gasification is presented as follows:



In coal gasification, synthesis gas is the term for the combination of CO and H₂ by-products. By applying the water gas shift reaction, CO is then transformed into H₂ and CO₂. Since coal gasification is

endothermic, the needed reaction rate can only be reached at a temperature of approximately 1273 K. On the other hand, the water gas shift process is exothermic and needs less heat for CO conversion. In the conventional method, gasification is initially carried out in a reactor with an operating temperature of over 1273 K, and the synthesis gas is then injected into a second reactor, which typically works at a temperature below 673 K for CO conversion. This conventional method also needs a CO₂ separation process after the CO conversion into H₂. Thermodynamic research shows that CO may be changed to H₂ at a very high temperature during the water gas shift process if CO₂ gas can be eliminated. This approach proposes that the three reactions (coal gasification, water shift process, and CO₂ separation) can be performed under identical operational conditions. For instance, several studies have been conducted on eliminating CO₂ during coal gasification using CaO adsorbent, and results proved that CO₂ was successfully fixed into the adsorbent and CO was completely converted to H₂ [34]. In this context, gasification allows companies to create unique combinations of cutting-edge technologies that provide affordable, dependable, and highly efficient solutions for various commercial applications.

2.1.3. Pyrolysis

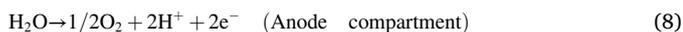
Some sources predict that blue and/or grey H₂ alternatives will continue to play important roles over the next decades, although green H₂ pathways are the preferred future scenario. The pyrolysis process, which thermally decomposes methane with or without a catalyst into H₂ and solid carbon, arises as a potentially viable natural gas-based hydrogen generation technique [35]. The technology readiness level and current developments on natural gas pyrolysis was evaluated by Schneider et al. [36]. The pyrolysis process has advantages and drawbacks compared to steam methane reforming and water-electrolysis processes. The authors reported that the pyrolysis process requires no water and uses 55% and 75% less energy than SMR and electrolysis when the reactions for the three methods were compared. Most crucially, direct pyrolysis generates no CO₂, contrary to SMR, which is GHG intensive. On the other hand, the pyrolysis of methane as a single molecule is often the only process discussed in the literature, and problems associated with using natural gas are not considered. The quality of produced H₂ is also rarely evaluated. The conversion of natural gas to H₂ and solid carbon via direct contact pyrolysis may be commercially appealing due to the abundant availability of natural gas and the ability to produce H₂ with free-of-contamination (CO and CO₂). However, in contrast to the steam reforming method, a continuous pyrolysis process for H₂ generation is not yet commercially viable and is still in an early stage of development. Recently, the Ekona Power and Aurora Hydrogen companies have been developing pyrolysis processes, while there have been critical drawbacks in the industrial-scale application of some pyrolysis projects [37]. Overall, there is an interest in researching natural gas pyrolysis to evaluate its possible strength based on techno-economic and environmental performance indicators considering the increased demand for H₂ and the requirement to mitigate climate change.

2.2. Water for H₂ production

Water as a resource for H₂ production is gained considerable interest as it is thought to be just the one that can produce great amounts of sustainable H₂ without emitting any pollutants or using any fossil or nuclear resources [38]. Moreover, water is the cleanest and most abundant resource on earth, and eventually, the combustion of H₂ will produce water again, which not only supplies energy but also is environmentally friendly. Water splitting to produce H₂ and oxygen has been extensively studied; its commercial applications began in the 1890 s [39]. The three types of water splitting are electrolysis, thermolysis, and photoelectrolysis, and their significant remarks and maturity of processes are summarized in Table 2.

2.2.1. Electrolysis

Water is the source in the electrolysis process, and under the force of the direct current, it is split into H₂ and oxygen. Overall water electrolysis reactions (Eqs. (8)–(10)) are represented as follows:



Several systems have been developed for effective H₂ production using water electrolysis, such as alkaline electrolysis (AE), proton exchange membranes (PEM), and solid oxide electrolysis (SOE). AE performs at temperatures of 60–80 °C, using NaOH or KOH solutions as the electrolyte, and the concentrations of electrolytes are almost 20–30%. The produced H₂ is 99% pure but must still be cleaned of an alkali fog, typically carried out by desorption. AE method has a maximum working current density of not more than 400 mA/cm², and the power needed to produce H₂ is around 4.5–5.5 kWh/Nm³ with an efficiency of almost 60% [42]. Furthermore, AEs require a long time to operate and thus struggle to adapt to the variable nature of renewable energy sources due to their prolonged startup procedures [43]. Hence, AEs are widely operated with constant power input. When we look into PEM-based electrolysis, it is based on the relatively novel PEM-based fuel cell process. Compared to AE, the PEM electrolysis systems are more environmentally friendly since the produced gas does not contain an alkaline fog. In addition, high H₂ production efficiency, simple design, fast response, and high-pressure output present PEM electrolysis as a promising H₂ production method. It has a substantially greater operational current density (10 A/cm²) than other AE methods. Furthermore, PEM electrolysis facilities are significantly easier to balance, making them more appealing for applications in the industry. On the other hand, PEM electrolysis is more expensive than AE due to the expense of the precious metals utilized as electrocatalysts. Therefore, studies should focus on reducing the cost of PEM electrolysis with cost-effective electrode and catalyst materials production. The SOE method has recently attracted attention due to its high energy efficiency of 90% with heat utilization. Namely, low voltage is needed for water electrolysis at high temperatures, which indicates less energy is consumed [44]. However, extreme temperatures create problems with material decomposition, and the period of SOE operation can be problematic. Furthermore, the produced H₂ mixture with water vapor using SOE needs extra treatment to achieve high-purity H₂, and therefore, the SOE process is still being developed.

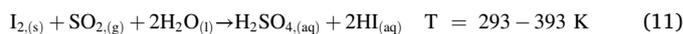
Although electrolysis has practically no direct emission, it is energy-intensive; hence the source of the system energy is directly linked to the total GHG emissions of the process [45]. Most energy utilized in large-scale electrolysis processes is obtained from grid power generated mostly by fossil fuels, which have significant GHG emissions [46]. In certain circumstances, electrolysis-related emissions may exceed those from the natural gas steam-reforming H₂ production process [47]. However, almost 90% of emissions from water electrolysis may be

reduced when fossil fuels in the grid electricity are switched with renewable energy resources like hydro and geothermal [48]. Despite its tremendous potential, water electrolysis only contributes to less than 0.1% of worldwide H₂ production [49]. Overall, integrating water electrolysis processes with renewable wind or solar power systems minimizes emissions and allows it to effectively valorize renewable resources.

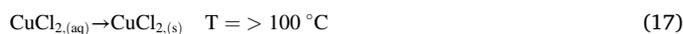
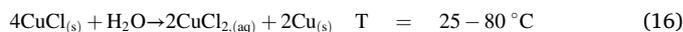
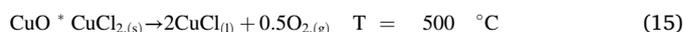
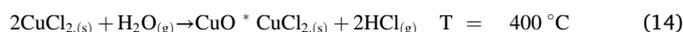
2.2.2. Thermolysis

In thermolysis, also called thermochemical water splitting process, heat is utilized to dissociate water into H₂ and oxygen [50]. In this process, water decomposes at 2500 °C, the effective and sustainable heat sources may, however, not easily be available. Hence, chemical reagents have been suggested to reduce the temperatures [51]. Since the 1970 s and 1980 s, researchers have investigated thermochemical cycles to develop less harmful fuels than traditional fuels [52]. A thermochemical cycle, in practice, is a set of low-temperature exothermic and high-temperature endothermic processes that use thermal energy to generate H₂ as an energy carrier [53]. Clean and sustainable H₂ production from water may be conducted using various processes like biochemical, photochemical, etc. However, thermochemical cycles have been widely performed because they can integrate with various renewable energy resources such as biomass, nuclear, solar, wind, and so on [54]. Although more than 300 cycles have been found, just a few were considered promising due to their numerous limits and drawbacks [55]. Three-step sulfur-iodine (S-I) cycle is the most promising and well-considered cycle in the studies up to now [56], and it can handle huge production rates owing to its ability to combine with high-temperature nuclear heat resources [57] and solar energies [58]. De Beni and Marchetti [59] also performed the first multi-step thermochemical cycle, Marc-1, utilizing bromine, calcium, and mercury with approximately 50% H₂ production efficiency. Recently, several lab-scale studies on magnesium-chlorine [60] and copper-chlorine [61] thermochemical cycles have been considered to improve the efficiency of the process by including an extra step to the cycle and examining the hybrid types of the cycles for sustainable H₂ production. Four examples of cycles are presented as follows:

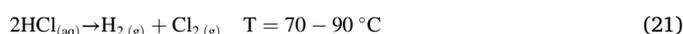
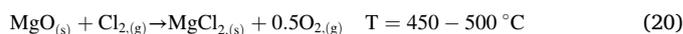
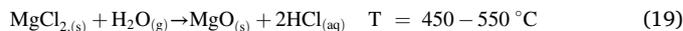
Sulfur-iodine cycle [62]:



Copper-chlorine cycle [63]:



Magnesium-chlorine cycle [64]:



Iron-chlorine cycle [65]:

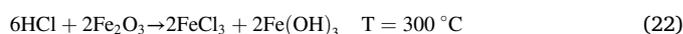


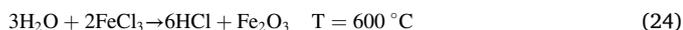
Table 2
A list of some hydrogen production methods from water dissociation (adapted from [40,41]).

Process		Advantages-Disadvantages	Maturity		
Electrolysis	<p>Alkaline electrolysis</p> <p> $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ (Cathode) $2OH^- \rightarrow 1/2 O_2 + H_2O + 2e^-$ (Anode) </p>	<ul style="list-style-type: none"> • Cost-effective operation • Long term durability • High production efficiency • Low degree of purity • Corrosive electrolyte type • Low dynamic operation • Low operational pressure • Prolonged startup procedure • Low current density 	Mature		
	<p>PEM electrolysis</p> <p> $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ (Cathode) $2OH^- \rightarrow 1/2 O_2 + H_2O + 2e^-$ (Anode) </p>			<ul style="list-style-type: none"> • Simple design • High gas purity • Rapid system response • High dynamic operation • High current density • High component cost • Acidic environment • Membrane scaling • Low durability 	Early market
	<p>Solid oxide electrolysis</p> <p> $2H_2O + 2e^- \rightarrow H_2 + O^{2-}$ (Cathode) $O^{2-} \rightarrow 1/2 O_2 + 2e^-$ (Anode) </p>				

(continued on next page)

Table 2 (continued)

Process		Advantages-Disadvantages	Maturity
Thermochemical water splitting		<ul style="list-style-type: none"> • Sustainable and clean • Abundant resource • Oxygen is the only by-product • Corrosive materials • Toxic chemicals • High capital cost 	Lab-scale
Photoelectrolysis		<ul style="list-style-type: none"> • Emission-free process • Abundant resource • Oxygen is the only by-product • Sunlight requirement • Low conversion efficiency • Effectiveness of photocatalytic material 	Lab-scale

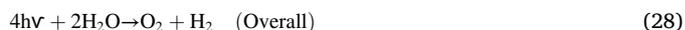
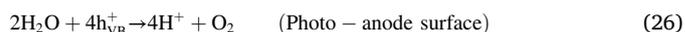


Overall, five crucial parameters should be considered to evaluate the performance and sustainability of cycles in the thermochemical water splitting processes: (i) the change in free energy (ΔG) of all reactions must be almost zero, (ii) the minimization of steps must be done, (iii) the reaction rates of the steps must be fast, (iv) the hazardous by-product formation must be prevented, and (v) Intermediate by-products need to be manageable. All these processes have been evaluated at a lab scale; however, scaling up the procedures is expected to help this technology overcome one of its main problems by enhancing thermal efficiency. Moreover, reduced costs for producing H_2 may also result from a better knowledge of the connection between capital expenses, thermodynamic losses, and thermal efficiency of the process. All present procedures involve three or more reactions; it is thought that an effective two-reaction mechanism would enable the practicality of this technology.

2.2.3. Photoelectrolysis

Photoelectrolysis is another method for H_2 production from water, which utilizes sunlight to dissociate water into H_2 and oxygen in the presence of semiconductor compounds to absorb electromagnetic radiation from the sun. The photoelectrolysis reactor comprises a sunlight-absorbing semiconductor electrode and a reference metal electrode immersed in an electrolyte. Basically, an electron-hole pair is produced when a photon that has more energy than the bandgap reaches the anode. Then, the holes dissociate water on the anode surface to form H_2 and oxygen while the electrons move through the anode to the cathode electrode. After moving through the electrolyte, the H_2 ions at the

cathode interact with the electrons to produce H_2 gas. Finally, for processing and storage, the oxygen and H_2 gases are separated using a semi-permeable membrane. The chemical equations for H_2 production in photoelectrolysis system is presented as follows [66]:



Overall, to decompose water molecules, the band gap of electrode materials, or potential, must be greater than the theoretically acceptable minimum value of 1.23 eV while also exceeding the electric resistance of the close circuit. Therefore, the effective band gap range is 1.6–2.2 eV in order to obtain maximum light absorption and high charge carrier mobility [67]. Hence, several studies have been focused on the production of effective and non-corrosive electrode materials including thin film Fe_2O_3 , TiO_2 , and WO_3 , CdS, and ZnS for the anode; and CIGS/Pt, p-SiC/Pt, and p-InP/Pt for the cathode electrode [68]. Furthermore, the energy of the electrochemical reaction must be in harmony with the solar radiation spectrum, which is a challenging problem, in order to enhance the efficiency of this process. Because, imbalance between solar radiation and the electrode materials can result in photo-generated holes, which can cause surface oxidations and either form an inhibitory layer on the semiconductor surface or destroy the electrode, leading to corrosion. Consequently, improvements on above mentioned drawbacks of photoelectrolysis method can reduce the surface over-potentials in relation to the water and speed up the reaction kinetics, which will reduce the electric losses of the system.

2.3. Biomass for H₂ production

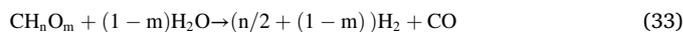
Biomass comes from growing plants, such as trees, crops, and algae, as well as from animal waste, industrial wood residues, food residues, and the organic content of municipal wastes. It is the most significant renewable energy source worldwide, with 55% of all renewable energy and over 6% of the world's energy supply coming from modern bio-energy [69]. Biomass has considerable capacity to accelerate the awareness of H₂ as a key fuel of the future [70]. Furthermore, compared to fossil fuels, biomass might have less net CO₂ effect since it is renewable and absorbs atmospheric CO₂ while producing. The use of biomass might also decrease the dependence on foreign energy resources, which is currently the case in many developing countries today. However, producing H₂ from biomass faces substantial challenges, such as no fulfilled technological demonstrations for biomass-based processes. Therefore, in recent years, studies on H₂ production from biomass resources have increased, considering their minimal environmental impact, and the focus is on making the necessary improvements so that biomass-based processes can be turned into commercially viable options. Thermochemical and biochemical processes are the two significant ways for biomass-based H₂ production. The critical remarks and maturity of the biomass-based processes, including gasification, steam reforming, DF, PF, and MEC processes, are summarized in Table 3.

2.3.1. Gasification

The conversion of waste biomass into useful energy products with greater energy density using a high-temperature process (500–1400 °C) is known as biomass gasification [72,73]. Both the product gas purity and the overall amount of energy used by the gasifier are influenced by the temperature of the process, with less favorable gas quality remaining the result of lower process temperature [74]. Gasification has been reported to have a higher H₂ yield per unit of biomass and a higher production efficiency, which makes gasification more appealing. The thermochemical conversion of biomass to H₂ can be carried out through a variety of pathways, including gasification, pyrolysis, and liquefaction [75]. The steam and supercritical water gasification methods for H₂ production from biomass have been widely considered. The biomass steam gasification method includes converting biomass feedstock into H₂, CO, CH₄, CO₂, and hydrocarbons at high temperatures, assisting in the introduction of water vapor as a gasification agent. In this process, temperature and biomass-to-steam ratio are the most important affected operational parameters on the H₂ content of produced syngas [76]. Moreover, it is widely recognized that steam is the best gasification agent to produce syngas with considerably high H₂ content. It may be improved by adding adsorbents and catalysts [77]. The carbon gasification, water-gas shift, methane, and hydrocarbon reforming reactions are the main processes in biomass steam gasification for H₂ production, which are reported as follows:



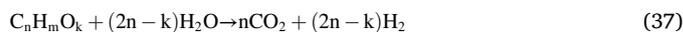
Supercritical water gasification is an improved form of traditional gasification that effectively converts biomass to H₂-rich syngas using water as the reaction media [78]. In the supercritical water gasification method, H₂, CO, CO₂, CH₄, and other gases can be formed via the pyrolysis, hydrolysis, condensation, and dehydrogenation of biomass. Steam reforming, water-gas conversion, and two-step methanation reactions are the key components of the process [79]. In this method, it is not essential to dry the biomass material before supercritical water gasification since water serves as both a reaction medium and a reactant, which considerably lowers energy utilization [80].



In addition, utilization of catalysts may reduce the reaction temperature and enhance the rate of carbon gasification and H₂ production from biomass for both steam and supercritical water gasification methods [81]. On the other hand, the potential of tar formation throughout the process is the major drawback of steam gasification compared to supercritical water gasification [82]. Furthermore, supercritical water gasification is more effective and can also handle a wider variety of biomass with higher moisture content [83]. However, scaling up supercritical water gasification may be difficult due to its higher production costs, safety concerns during operation due to high temperatures and pressures, and reactor corrosion triggered by solid deposition from salt and/or lignin-containing biomass. The operation of large-scale biomass gasification is still complex in many ways, including the temporal and geographical variability of biomass resources and supply chains. Nevertheless, literature studies revealed that the gasification process has the potential to be incorporated into several industrial processes. Therefore, the production of H₂ from biomass gasification continues to grow according to initiatives from France, Germany, the US, the UK, and Scandinavia, among other nations [84]. However, there is currently a lack of research on techno-economic analysis and environmental sustainability evaluation of the biomass gasification process for H₂ production. Future research on biomass gasification should thus focus on environmental sustainability, define the entire energy and carbon footprint, and evaluate the availability and effects of resource consumption in large-scale production applications.

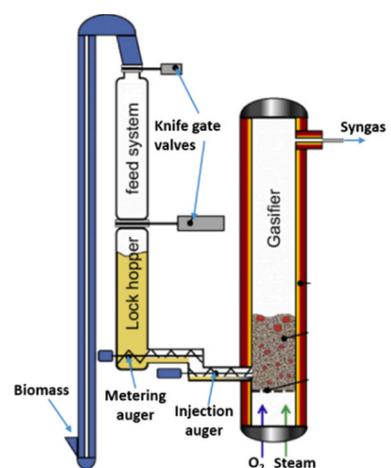
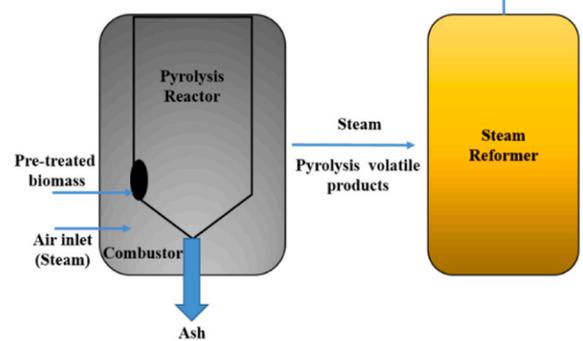
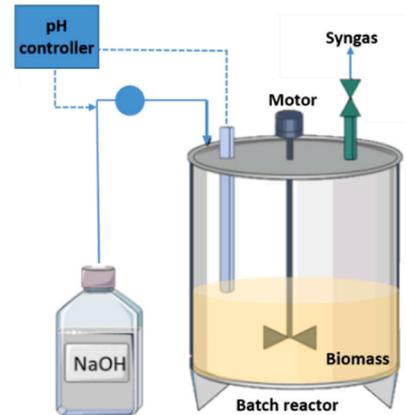
2.3.2. Biogas reformation

The biogas reformation is a common post-production method to produce H₂ from indirect biomass resources. Biomass resources are initially fed to a dryer to eliminate their moisture content and then transferred to a pyrolysis process to decompose into bio-oil, char, and gas. The by-products of pyrolysis are then put into a gasifier to generate syngas. After cooling and compressing, the produced syngas is transferred to a storage tank for further processing. In the biogas reformation process, the syngas reacts with hot steam to produce a gas that contains a substantial amount of H₂. Furthermore, this process produces H₂ through endothermic processes, which need a reliable heat supply. Hence, commercial steam reformers usually perform between 3.0 and 25 bars of pressure and temperatures at 700–1100 °C [20]. The following equation may be used to identify the potential paths in the steam reforming process to create H₂ from biomass feedstock while considering decomposition and water-shift processes [85].



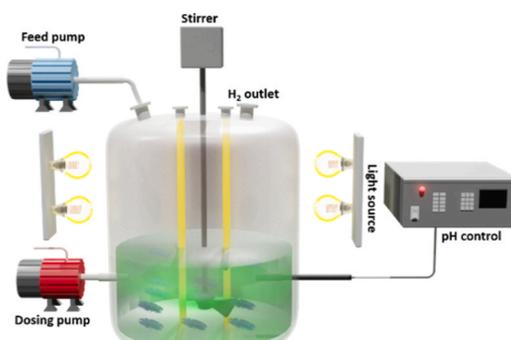
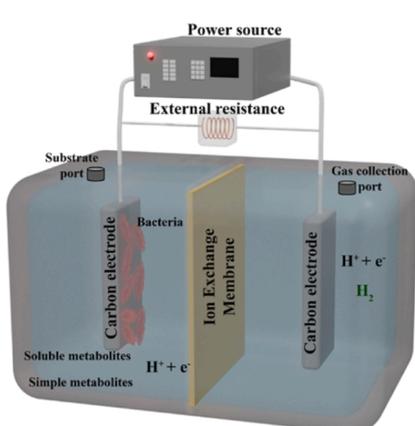
In steam reforming, the second stage involves lowering the CO level using high and low-temperature water-gas shift reactors. This lowers the level of CO since some H₂-powered applications, such as H₂ fuel cells, need further reforming procedures. Usually, a second reforming step is necessary because the generated gas contains significant quantities of unused hydrocarbons. H₂ production with steam reforming from biomass feedstock, such as agricultural residues, is a cost-effective and promising method. According to an economic and environmental analysis performed by [86], steam reforming to produce H₂ is a more ecologically friendly procedure than conventional options, which are widely utilized fossil fuels as a source. The authors reported that the average H₂ production cost of steam reforming of bio-gas was 0.27 US \$/kWh based on the economic model.

Table 3
A list of some hydrogen production methods from biomass (adapted from [71]).

Process	Advantages-Disadvantages	Maturity
<p>Gasification</p> 	<ul style="list-style-type: none"> • Impossibility of using biomass with a high moisture content • Renewable resource • Waste reduction • Space requirements • Some adverse environmental impact • Relatively long time required for start-up • High sensitivity to slag formation • Short life of system components, including gasifier vessel refractory • High level of sensible heat in product gas 	Early-market
<p>Biogas reformation</p> 	<ul style="list-style-type: none"> • Highest yield of H₂ • Relative stable during transition operation • CO₂ utilization potential • No oxygen need • Relatively lowest operating temperature • Continuous heat requirements for the reaction • Relatively high air emissions compared to other biomass-based technologies • High amount of carbon-based material formation • System complexity • Catalyst requirements 	Early-market
<p>Dark fermentation</p> 	<ul style="list-style-type: none"> • Utilization of a wide variety of different biomass resources • Simple reactor configuration • Light independence • High H₂ production rate • Considerably low emissions • Separation of H₂ required from H₂-CO₂ mixture • High BOD level in effluents • Requirement of pre-treatment • Development of an efficient reactor is required 	Lab-scale

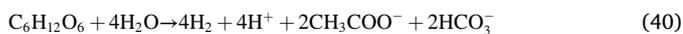
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Table 3 (continued)

Process	Advantages-Disadvantages	Maturity
<p>Photo-fermentation</p>  <p>Photo Fermentation</p>	<ul style="list-style-type: none"> • Use of a variety of substrates • Utilization of dark fermentation effluents • High H₂ yields • Environmentally friendly • Low light conversion efficiency • Slow H₂ production process • Inhibitory effect of O₂ on nitrogenase 	Lab-scale
<p>Microbial electrolysis cell</p> 	<ul style="list-style-type: none"> • Pollution free method • High COD removal performance • High H₂ recovery • Zero emissions • High degradation of biomass resources • High capital cost • Relatively low H₂ purity • Stability and scalability problems • External voltage requirement 	Lab-scale

2.3.3. Dark fermentation

BioH₂ production using DF to generate H₂-energy is a sustainable and environmental friendly option to fossil fuels to service the requirements of carbon emission reduction. Currently, significant efforts have been initiated to increase the production of H₂ using this attractive method. Namely, the DF has been reported as the most promising and effective bioH₂ production method with biomass conversion, with a net energy ratio of 1.9 [60]. Most importantly, DF is most well-understood process among biotechnological methods for H₂ production. In DF, biomass resources are converted into H₂ by anaerobic bacteria under a dark environment. Hydrogenases are the primary enzymes that control the metabolism of H₂ by microorganisms. The two fundamental hydrogenases are [FeFe] and [NiFe] hydrogenases, which vary phylogenetically and have various active sites. Compared to [NiFe]-hydrogenases, which predominantly facilitate the oxidation of molecular H₂, [FeFe]-hydrogenases are more active in the production of H₂. Though several organic compounds, including carbohydrates, sugars, proteins, and lipids, can be utilized as substrates, the reaction of glucose biotransformation to acetate is generally considered as a standard for estimating the theoretical yields of fermentative H₂. Overall, in the DF, the H₂ production reactions from acetic acid, butyric acid, and glucose are presented as follows:



Overall, it is widely accepted that the future power economy will be powered by H₂ since it is a clean, efficient energy carrier that emits no emissions when consumed and can be generated from renewable resources like biomass and waste materials. On the other hand, still, there are critical drawbacks in the DF process, such as the total amount of H₂ produced during fermentation being reduced by the formation of numerous final byproducts, including acetic, acetone, butyric acids, butanol, methanol, and propionic acid. It is reported that the mixture of different chemicals as a final by-product reduces the production yield to 1–2.5 mol of H₂ per mole of glucose [87]. Moreover, the yield of fermentation-based production of H₂ might vary depending on numerous operation parameters, including temperature, biomass type, type of bacterial species, mixing rate, pH, and reactor design. Therefore, several studies on the optimization of operational parameters of DF have been performed to obtain the H₂ yield close to a theoretical yield [88]. Numerous types of biomass and wastes have been studied as possible feedstock considering their origin and chemical constituents and reported promising results. Among biomass resources, even though energy crops are significantly successful, their utilization has recently come into doubt because of the growing food vs. fuel debate. However, the lignocellulosic residues that are remained from energy crops, mixed with other agricultural and forestry wastes, continue to be a rich source of biomass that may be used to produce second-generation H₂. Consequently, research efforts to (i) select the most efficient feedstock, (ii) develop more effective pre-treatment techniques for saccharification of lignocellulosic materials, (iii) discover and/or cultivate high H₂-producing strains through genetic engineering, and (iv) design the best reactor configurations and operating approaches through modeling and optimization may decide how efficiently fermentative H₂ can be produced in the future.

2.3.4. Photofermentation

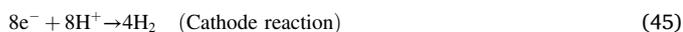
Photo-fermentative bioH₂ production is an attractive and environmentally sustainable method that performs quite effectively in ambient environments. Gest and Kaman first identified bioH₂ production by PF utilizing photosynthetic bacteria in 1949, and then this method demonstrated an efficient production of high-quality H₂ without the formation of oxygen [89]. Under anaerobic conditions with light, photosynthetic bacteria can convert the majority of organic acids to bioH₂ and CO₂ [90]. The primary enzyme involved in producing bioH₂ from photosynthetic bacteria (purple non-sulfur bacteria) is nitrogenase, and its synthesis is significantly promoted under light [91]. Fundamentally, the photo-fermentative H₂ production reactions by photosynthetic bacteria can be described as follows:



Optimization of feedstock source, reactor configuration, and operational parameters such as pH, temperature, light intensity and source, mixing rate, and bacterial species is essential for effective H₂ production in the PF process. Literature studies have revealed that effective H₂ production and bacterial growth obtains at initial pH and temperature of 5–7 and 30–40 °C, respectively [92]. Furthermore, the PF process can effectively transform organic compounds into H₂, presenting a higher substrate transformation rate and greater bioH₂ yields than the DF process. In the PF process, utilization of domestic and industrial wastewaters, organic wastes, and DF hydrolysate effluents may enhance the low H₂ production yield for commercial applications, low light conversion efficiency, and sensitivity of toxic substances [93]. On the other hand, this process has three major shortcomings: (i) the utilization of the nitrogenase enzyme, which has a high energy need; (ii) the low efficiency of solar energy conversion; and (iii) the requirement for complex anaerobic photo-bioreactors covering huge areas. Consequently, the photo-fermentation process is not yet an economically viable method to produce H₂. Therefore, the development of photo-fermentative H₂ production and cutting-edge technologies have gained significant interest.

2.3.5. Microbial electrolysis cell

MECs are consists of three individual compartment (anode, desalination, and cathode) and the anode-desalination and desalination-cathode compartments are separated by an anion exchange membrane (AEM) and a cation exchange membrane (CEM), respectively. In MEC, organic matter is converted into CO₂, electrons, and H⁺ by electrochemically active microorganisms that are forming on the surface of the anode. The H⁺ ions at the cathode, which diffuse from the anode to the cathode cell, typically through the CEM, recombine with the electrons as they pass through the electrical circuit to generate H₂ [94]. The microorganisms on the biocathode may reduce the H⁺ ions directly at the electrode surface, through hydrogenases, or by outer membrane cytochromes. Furthermore, the additional potential at optimized conditions (pressure of 1 bar, acetate concentration of 1 M, and initial pH of 7) is – 0.14 V, therefore, the additional energy is required to support the total reaction.



Moreover, the MEC process is more flexible than fermentative H₂ production processes because it may use various kinds of organic feedstock. For instance, DF cannot use biomass-containing other organic materials, such as proteins or organic acids, as a substrate because it needs a fermentable carbohydrate-rich feedstock. On the other hand, in MECs, bioH₂ production may occur using a broad range of materials, from simple combinations like pure alcohol, carboxylic acid, protein, and sugar to complicated mixtures like biomass hydrolysate and

household, animal, and food manufacturing wastewaters. Therefore, the MEC has attracted attention as a sustainable and economically feasible method due to its high H₂ yield and ability to utilize various biomass resources as a fuel of the system. Consequently, this process has several benefits over other methods for producing biofuel. However, it is considered too early to assess the total energy efficiency for producing realistic H₂ fuel of the MEC process.

Overall, due to their availability and high energy density, the thermochemical conversion of carbon-based fuels is the most significant technique for producing H₂. The thermochemical conversion of fossil fuels now produces most of the H₂ required for industrial uses. The thermochemical processes are good options for H₂ production considering their high energy efficiency, excellent H₂ production performance, and economic characteristics. Although water electrolysis has been acknowledged as a simple and effective method for producing pure H₂, its uses are frequently restricted to laboratory-scale units or small-scale industrial units as access to commercial H₂ production facilities is sometimes impossible owing to logistical and financial issues. In addition, renewable liquid reforming is a straightforward method for producing H₂. Still, it is not economically feasible since methanol and other renewable liquids are potential energy sources and are created through expensive processes like biomass gasification. Moreover, the thermochemical processes have several disadvantages related to their high carbon emissions, acidification potential, water consumption, and harmful impacts on the environment and humans. Recently biological processes have gained significant interest and presented as significantly environmentally friendly options considering their low carbon emissions, low contaminant release on soil and water media, relatively low water use, and low harmful impact on humans. Nevertheless, the high operational and maintenance costs, high operational times, relatively low H₂ production efficiency, requirement of critical pre-treatment steps, and lab-scale applicability are their major disadvantages compared to commercialized thermochemical H₂ production processes.

3. Comparative assessment of H₂ production methods

In this section, H₂ production processes from biomass, water, and fossil fuel resources considering their environmental impacts, H₂ production yield, energy efficiency, and total cost, were evaluated and discussed in more detail. Overall, evaluated parameters are normalized with the feature scaling normalization method to compare each H₂ production method to provide their sustainability levels. The environmental impact assessment of selected parameters is based on the LCA received from various studies and H₂ yield, energy efficiency, and cost parameters are based on the experimental studies, and all these corresponding references are listed in Table 4. The origin of the biomass and fossil fuels were also reported in Table SM1. In biomass-based processes, food waste, agricultural waste, wood waste, cornstalk, corn stover, and vegetal/algal wastes were widely utilized biomass resources. On the other hand, coal and natural gas were the main resources of the fossil fuel-based processes.

3.1. Environmental impact comparison

Fossil fuel sources are the most commonly utilized resources to produce H₂ among the potential H₂ production technologies. However, the shift to the H₂ economy demands that H₂ be created from abundant, clean, and renewable sources to create a sustainable energy system. Therefore, this paper considers seven primary methods to produce H₂: gasification and steam reforming of fossil resources, electrolysis of water, and DF, PF, MEC, and DF-MEC of biomass. Recently, there has been an increase in interest in producing H₂ from biomass resources as various industrial and agricultural operations produce a considerable amount of biomass waste. Moreover, H₂ production from biomass use, particularly biological processes, has currently gained significant interest in overcoming the drawbacks of conventional H₂ production

Table 4

A comparative table on experimental and LCA approaches used for H₂ production processes.

Technical aspects									LCA approach			Experimental approach			Ref.
Feedstock Resources									Impact categories			Impact categories			
Biomass						Fossil fuel		Water	GWP	AP	Others	Cost	H ₂ -yield	Energy efficiency	
DF	PF	MEC	DF-MEC	G	SR	G	SR								E
✓	✓	✓		✓					✓	✓		✓	✓		[95]
✓	✓	✓		✓					✓	✓	✓	✓	✓		[96]
✓	✓	✓		✓	✓				✓	✓		✓	✓		[97]
✓	✓	✓		✓	✓				✓	✓	✓	✓	✓		[98]
✓	✓	✓	✓	✓	✓	✓	✓		✓	✓	✓	✓	✓		[72]
✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[99]
✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[100]
✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[101]
✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[47]
✓	✓	✓	✓	✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[102]
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✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[105]
✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[106]
✓	✓	✓		✓	✓	✓	✓		✓	✓	✓	✓	✓	✓	[107]

*DF: Dark fermentation; PF: Photo-fermentation; MEC: Microbial electrolysis cell; G: Gasification; SR: Steam reforming; E: Electrolysis; GWP: Global warming potential; and AP: Acidification potential.

technologies. Overall, the environmental assessment of biological and thermochemical H₂ production processes was determined. The average values of the environmental impact parameters for biological and thermochemical H₂ production processes were reported in Table SM2.

This section compares the environmental impacts of 1 kg of H₂ produced from different processes. The H₂ production processes are compared regarding AP, FE, FRS, FEP, PMF, GWP, LU, OD, IR, WCP, and WSF. AP (kg SO₂ eq.) and GWP (kg CO₂ eq.) measure the amount of polluting acid deposited into water resources, soil, and organisms and CO₂ released into the atmosphere, respectively. IR (kBq Co-60 eq.) and OD (kg CFC-11 eq.) present the impact of radiation on materials and the amount of ozone destroyed by emission, respectively. FE (kg 1,4-DCB eq.) measures the ecotoxicity of freshwater, which occurred due to the air, land runoff, and chemical discharge into the water bodies. FRS (kg Cu eq.) represents the amount of utilized fossil resources. PMF (kg PM_{2.5} eq.) defines the particles or droplets in the air. WSF (m³) and WCP (m³ consumed) express the overall amount of freshwater consumed during the process and the amount of water used, respectively. The investigated environmental impact categories were selected according to the Operational Guide to the ISO Standards to define the LCA approaches [108]. Moreover, the investigated environmental impact variables were obtained from the LCA studies performed elsewhere. The average values of the selected environmental impact parameters are considered for assessment. However, some environmental impact data was not found in the literature. These results also revealed that there is limited research in the literature extensively investigating the environmental impacts of H₂ production processes, and there is a serious gap in this regard.

3.1.1. Global warming and acidification potentials

CO₂ releases are considered major GHG sources because of their harmful effects on the environment. Therefore, the shift to a carbon-neutral industry and economy with sustainable energy production is one of the literature's most thoroughly investigated research subjects. As reported in Section 3, H₂ can be produced from non-renewable and renewable resources. However, global H₂ production in 2020 completely depended on fossil fuel resources, with individual 0.7% of H₂ obtained from natural gas with enhanced carbon capture, storage, and utilization out of the overall production of 90 Mt. [109]. Natural gas remained the primary source of H₂ production in 2021, which resulted in 630 Mt. of direct CO₂ emissions from fossil fuel-based H₂ production. Despite the remarkable increase in low-emission H₂ production in 2021, it still accounted for a small amount of total production, with only 1 Mt

of low-emission production representing 94 Mt of global H₂ production in 2021 [109]. Therefore, replacing fossil fuel resources with renewables is important to minimize the emissions from H₂ production. Several low-emission H₂ production methods are presented, including biomass gasification and steam reforming, biomass dark and photo fermentation, and microbial electrolysis cell methods, which have been widely considered future energy opportunities. However, low-emission H₂ production methods, such as water electrolysis with biofuel power, water splitting with solar energy, and biomass fermentation, have not yet reached commercial production level due to low production efficiency, high production costs, and a lack of manufacturing and transportation facilities [11,110]. Alternative low-emission methods, including biomass gasification, biomethane reforming, and water electrolysis powered by renewable energy, are recognized to be technologically advanced and have the potential to be produced at a large scale either currently or throughout the coming years [111]. Therefore, particular evaluation of technologies from an environmental point of view is important in order to obviously forecast the applicability of the technologies. In this regard, the GWP and AP values of the renewable and non-renewable processes were evaluated and average values of the processes for different resources are presented in Fig. 2.

Among the thermochemical processes using fossil fuels, the highest

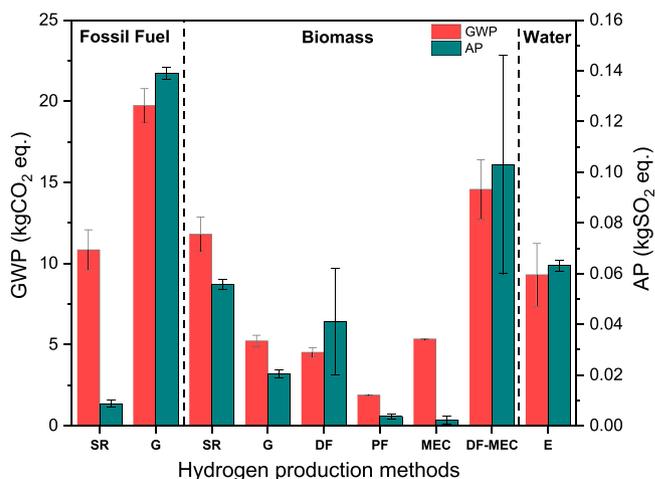


Fig. 2. Global warming and acidification potential values of H₂ production methods for different resources.

GWP of 19.85 kgCO₂ eq./kg H₂ and AP 0.139 kgSO₂/kg H₂ were obtained for the gasification process, while the relatively lowest GWP (10.85 kgCO₂ eq./kg H₂) and AP (0.0087 kgSO₂/kg H₂) values were observed for the steam reforming process. The highest GWP was obtained for gasification due to coal mining and transportation operations, even with renewable energy input [112]. Furthermore, the main reason for high AP values in the gasification process may be the SO₂ formation during the electricity production for different process steps like pre-processing raw materials and gasification plant. As expected, the GWP (5.22 kgCO₂ eq./kg H₂) of the biomass-based gasification process was significantly lower than that for the fossil fuel-based gasification owing to carbon fixation during biomass growth [113]. Namely, plants adsorb CO₂ from the atmosphere with photosynthesis; hence, this process positively impacts the emissions. Regarding AP (0.02 kgSO₂/kg H₂), biomass gasification presented a lower impact owing to the low sulfur content of the raw material. In the electrolysis process for H₂ production from water, relatively high GWP (9.29 kgCO₂ eq./kg H₂) and AP (0.06 kgSO₂/kg H₂) values were obtained, most probably due to the energy utilization during the water-splitting process [114]. Despite the fact that water electrolysis produces practically zero GHG emissions, the process is energy-intensive, and the supply of the process energy directly affects the total GHG emissions of the process. On the other hand, compared to thermochemical processes, biological H₂ production processes have the potential to considerably reduce CO₂ releases when their substrates are derived from renewable resources or wastes. The PF is the most environmentally friendly H₂ production process presenting low GWP (1.88 kgCO₂ eq./kg H₂) and AP (0.003 gSO₂/kg H₂) values, it is followed by DF and MEC processes. However, the integrated DF-MEC process presents considerably poor environmental performance with a GWP of 14.6 kgCO₂ eq./kg H₂ and AP of 0.103 gSO₂/kg H₂ compared to the other biological processes related to their superior energy and heat requirements by the production process. Hence, using renewable energy sources in production steps is crucial to preventing high GWP and AP values.

3.1.2. Ozone depletion and ionizing radiation

Ozone depletion is triggered by emissions that cause the stratospheric ozone layer to become thinner. Most of the UV radiation, which has been causing damage to animal and human health, natural ecosystems, and resources, reaches the Earth's surface due to this thinning. Methane and nitrogen oxide emissions are considered the main contributors to OD [47]. As provided in Figs. 2 and 3, surprisingly, the fossil fuel-based H₂ production processes presented a minimum contribution to the OD with a value of 3.35×10^{-6} kg CFC-11 eq./kg H₂ for gasification and 2.99×10^{-6} kg CFC-11 eq./kg H₂ for steam reforming. The

OD value of the gasification and steam reforming process was relatively low compared to other thermochemical and biological processes due to the utilization of renewable energy sources like wind and solar to power the process, and therefore, there was no emission in this process. On the other hand, the highest OD value of 1.9×10^{-4} kg CFC-11 eq./kg H₂ was obtained for the PF process, most probably because of the emissions from construction processes [115]. The OD value of H₂ production from the DF process is two orders lower than that for the PF process. Moreover, the biomass-based steam reforming process is found to have an OD of 1.47×10^{-4} kg CFC-11 eq./kg H₂. The relatively high values observed in DF and PF processes may also come from the oil processing and the energy utilized in H₂ production. This can be related to the higher amounts of nitrogen oxide and CH₄ emissions associated with the fossil-based source utilization for heating and energy purposes in the steam reforming process. Among the biological processes, the lowest OD value of 8×10^{-6} kg CFC-11 eq./kg H₂ was observed for the MEC process since almost zero emissions of this process and simple construction procedure. As expected, the OD (1.22×10^{-5} kg CFC-11 eq./kg H₂) potential of the water electrolysis process was considerably low since its relatively low emission potential.

The IR is another important environmental impact parameter to evaluate the effect of H₂ production processes on nature and humans. IR includes both indirect radiation exposure and the effects of releasing radioactive material [116]. The highest IR of 4.5 kgBqCo-60 eq./kg H₂ was observed for the DF-MEC hybrid process, probably due to the sludge pre-treatment using ionizing radiation [117]. The IR values of fossil fuel and biomass-based gasification, steam reforming, and electrolysis processes were relatively high compared to DF and MEC processes. The IR from these processes could be explained with the catalyst preparation process since this process includes the extraction and mining of zeolite and other substances [118]. Overall, the IR impact of the DF process was lower than the other processes, therefore, it could be concluded that the DF process is a more sustainable option considering its low IR value.

3.1.3. Freshwater ecotoxicity and eutrophication potential

Freshwater ecotoxicity and eutrophication potential are other environmental impacts that present an important remark. Eutrophication mainly occurs due to the release of nutrients like nitrogen and phosphorus into the environment and the impacts of these chemicals are defined as P equivalent for a better comparison. Furthermore, several substances used in the production of energy and H₂ processes have the potential to be harmful in a variety of media and these chemicals are also normalized as 1,4-DCB equivalent for a well evaluation. In this regard, the average FE and FEP values of processes are provided in Fig. 4. Results revealed that the fossil fuel-based H₂ production processes

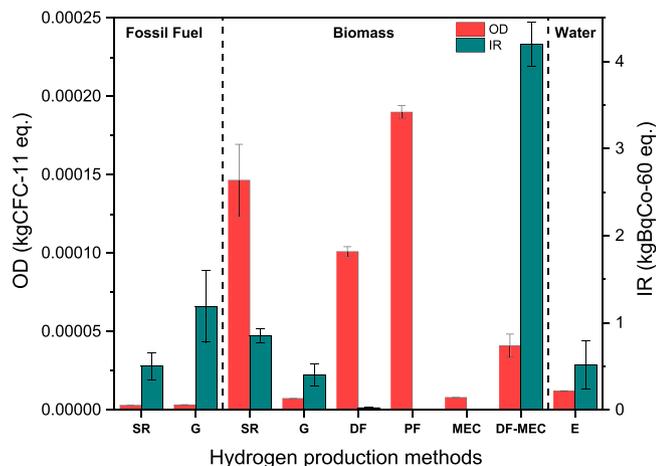


Fig. 3. Ozone depletion and ionizing radiation values of H₂ production methods for different resources.

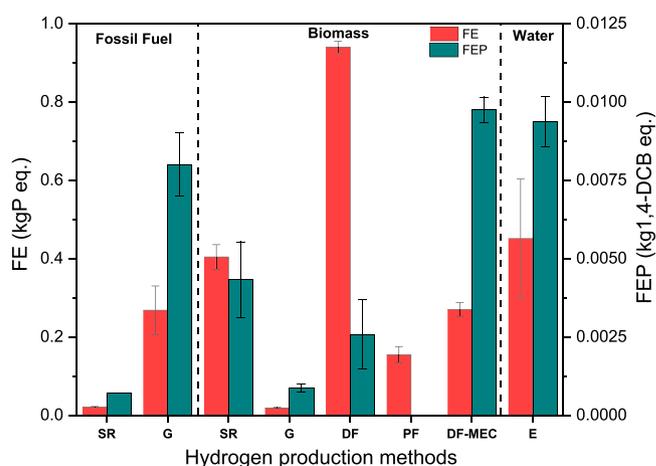


Fig. 4. Freshwater ecotoxicity and eutrophication potential values of H₂ production methods for different resources.

provided lower FE and FEP results than biomass and water-based processes. The chemical requirements of biological processes increased the impact of ecotoxicology indicators. All of these processes demand certain chemical types, which could release into terrestrial, freshwater, and marine bodies and result in ecotoxicity. Highest FE value of 0.94 kgP eq./kg H₂ was observed for DF process due to the great amount of chemical utilization during digestate handling and pre-treatment step of this method [119]. Similarly, the DF-MEC hybrid process has significantly the highest PFE (0.0097 kg1,4-DCB eq./kg H₂), mainly contributed by the biological and chemical oxygen demand of the biomass hydrolysate coming from the utilization of anaerobic sludge or/and wastewater and the formation of organic substances in the bioH₂ production method. This process also produces a considerable amount of wastewater that must be further treated to decrease its biological and chemical oxygen demand before its discharge into the environment. Moreover, eutrophication is harmfully affected by the utilization of ammonia, nitrate, and phosphate-containing fertilizers during crop growth. Therefore, in cases where biomass-based materials are used as the source, it is normal for all H₂ processes to cause eutrophication.

Furthermore, the FE value of biomass-based steam reforming was relatively high with 0.404 kgP eq./kg H₂, most probably due to the biomass treatment and the manufacture of solid oxide fuel cells [120]. Moreover, wastewater effluents with a great amount of nitrogen-based chemical (nitrates and nitrites) from catalyst production steps is another contributor of FE and FEP in the steam reforming process. In addition, the FE and FEP values were 0.45 kgP eq./kg H₂ and 0.0093 kg1,4-DCB eq./kg H₂ for the water electrolysis process. In this process, relatively high ecotoxicological outputs are initiated by the rare earth elements especially used in the electrolysis stacks, chemicals utilized in the process, reinforced steel electrodes, and some kind of epoxy applications used in the electrolysis reactor. Consequently, among the biological processes, the PF process is the most environmentally friendly option with its low FE value (0.15 kgP eq./kg H₂) due to the lack of direct phosphorous, nitrogen, and sulfur emissions of the system. Overall, results provided that the FE and FEP values obtained for the biomass-based gasification process were considerably low due to the use of renewable energy for heating and electricity in the reviewed studies. Moreover, based on these results, it could be concluded that in all bioH₂ production processes, pesticide and fertilizer emissions from agricultural land are the main causes of ecotoxicity [121].

3.1.4. Water consumption and water scarcity footprint

Recently, the LCA studies evaluated water-intensive H₂ production processes considering their water utilization potentials from source to end-point [122]. As expected, the highest WCP (119.90 m³) and WSF

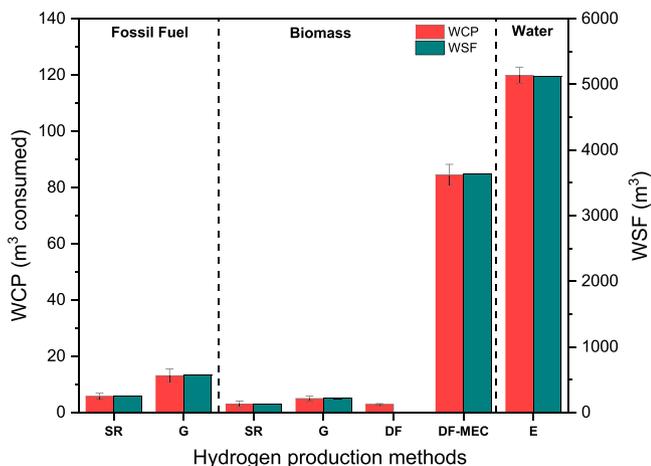


Fig. 5. Water consumption and water scarcity footprint values of H₂ production methods for different resources.

(5117.05 m³) values were observed for the electrolysis process, which directly utilizes water as a resource for H₂ production (Fig. 5). Furthermore, due to a more complicated supply chain and the connection between water use and agricultural production processes, biomass-based DF-MEC H₂ production faces some significant interconnected issues [123]. The WCP and WSF values were 84.5 and 3632.9 m³ for the DF-MEC process due to the huge amount of wastewater formation potential of this hybrid process. However, the WCP values of steam reforming, gasification, and DF processes were 3.1, 4.9, and 2.9 m³, respectively, 28, 18, and 30 times lower than that for the DF-MEC process. Electricity utilization could be the main contributor to water consumption in fossil fuel-based processes. On the other hand, the water utilizes during biomass production emerges as the main cause of water scarcity in biomass-based H₂ production methods. Overall, these results provided that biomass-based processes are environmentally friendly and sustainable processes related to their considerably low WCP and WSF impacts. It is possible to reach the conclusion that water consumption and the associated harm to the environment and to human health should be decreased through the development of new technologies or improving existing ones that use less or do not need water.

To date, questions concerning water utilization for hydrogen have been raised by numerous people, who claim that acquiring water for the economy will be extremely costly or demanding on both energy and water supplies. Therefore, we calculate the total volume of water that will be extracted and used for electrolysis in the projected hydrogen economy. The hydrogen production in 2023 is reported as 118 Mt, and the electrolysis process provides about 5% of this production. In this context, the sustainability of the annual water need of 5.9 Mt of hydrogen production using the electrolysis process in 2023 is evaluated. According to the reaction stoichiometry, 9 kg of water is required to be spent for every kg of hydrogen produced. Therefore, 5.9 Mt of hydrogen needs 53.1 million m³ of water based on the calculations. These results may reveal that although it is thought that water use in hydrogen production with the electrolysis process is high compared to other energy generation processes from fossil fuels, some quick calculations about water consumption of fossil fuel-based systems provided that the water used for power generation and energy production from fossil fuels was 251 billion m³. Comparatively, even if 53.1 million m³ of water must be utilized for electrolysis to produce hydrogen, this is still 99.9% less than the current consumption of fossil fuels for energy. When compared to other industries, such as the irrigated farming sector, which uses about 2700 billion m³ of water annually, or 70% of the world's total freshwater withdrawals, electrolysis uses a particularly low amount of water. However, despite the fact that adjacent businesses use significantly more water than even the most optimistic hydrogen production forecast, worries about freshwater shortage still demand reductions in water extraction from all possible perspectives. Therefore, proposing a solution that allows the utilization of the reuse of effluent water can further reduce the water footprint of the electrolysis process. In this context, the hydrogen produced using electrolysis process can then be utilized as fuel in fuel cell processes and converted into water with energy production as final valuable products. Therefore, the hydrogen ecosystem should be recognized as the water-conserving cycle where no water is lost. This really makes the forthcoming hydrogen economy an appealing choice.

3.1.5. Fossil resource scarcity and land use

Recently, a significant part of attention has been given to the ecological damage that burning fossil fuels causes as a result of increased energy consumption. Moreover, concern over the limited availability of fossil fuels is also increasing. In this regard, this review discusses and reports the FRS values of H₂ production processes (Fig. 6). The highest FRS value of 12 kg-oil eq./kg H₂ was obtained for the DF process related to its energy requirements. The utilization of huge amounts of mineral resources (like aluminum, copper, lead, iron, nickel, etc.) and fossil resources (coal, natural gas, and oil) in the DF process for system power are responsible for this significant FRS value. Similarly, high FRS values

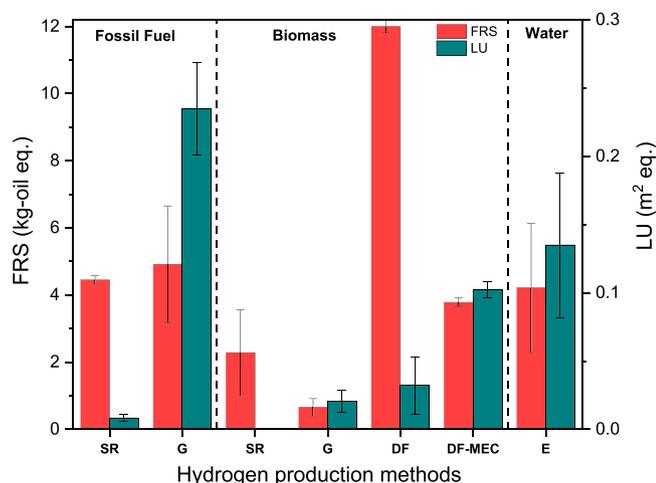


Fig. 6. Fossil resource scarcity and land use values of H₂ production methods for different resources.

were observed for the fossil fuel-based processes due to the utilization of fossil resources for both energy and feedstock purposes. On the other hand, relatively low FRS values were obtained since renewable energy sources, and feedstock are used in the biomass-based gasification and steam reforming processes. The FRS values of biomass-based steam reforming and gasification processes were 2.28 and 0.65 kg-oil eq./kg H₂, approximately 2 and 8 times lower than that for the fossil fuel-based steam reforming and gasification processes. As expected, the primary reason for the FRS (4.22 kg-oil eq./kg H₂) in the water electrolysis process is the utilization of fossil resources to power the system. Overall, these results proved that the significant FRS impact in biomass and water-based processes was caused by electricity consumption.

The requirement of LU is another environmental impact category shown in Fig. 6. This impact category considers the extent of land utilization for producing H₂. Among all H₂ production processes, biomass-based gasification has the least land area requirements with LU of 0.011 m² eq./kg H₂. On the other hand, the fossil fuel-based gasification and steam reforming processes seem to need the greatest area with LU values of 0.235 and 21.85 m² eq./kg H₂, respectively, because of the massive safety and management requirements like heating, pressure, and solid waste control. Furthermore, in biomass-based processes, the

main contributor of the large area requirements is most probably linked to the biomass cultivation steps [124].

3.2. Cost and energy efficiency comparison

In this section, the cost and efficiency of H₂ production processes are comparatively evaluated according to results in the literature. In cost performance evaluation, average H₂ production costs of processes are taken into account based on US\$/kg-H₂. Average production costs are presented in Fig. 7 for the different H₂ production processes. The average cost and energy efficiency values of the different processes were also reported in Table SM3. Results showed that the fossil-fuel based steam reforming process has the lowest production cost with 1.35 US \$/kg-H₂, followed by fossil fuel-based gasification (1.73 US\$/kg-H₂), biomass-based gasification (1.92 US\$/kg-H₂), and steam reforming processes (2.15 US\$/kg-H₂). Moreover, the most expensive process is water electrolysis, with a production cost of 1.99 US\$/kg-H₂, followed by DF, PF, and MEC processes. Namely, the H₂ production costs are projected based on the operational and capital costs of the processes. The capital cost comprises the majority of the production cost for higher-cost H₂ production methods including electrolysis, DF, PF, and MEC.

Therefore, the development of novel materials and their implementation into energy systems might reduce the associated capital costs, which may improve the cost-effectiveness of renewable H₂ production methods. In addition, these results demonstrate that large-scale, commercially available H₂ production technologies significantly surpass the relatively new, lab-scale alternatives economically. The price of energy supplies, capital expenses, advances in technology, and carbon taxation are some of the variables that affect the production cost of H₂. However, the research revealed that renewable resources like solar or wind could considerably decrease the H₂ production cost of the process related to engineering and technological enhancements. Furthermore, rising carbon costs would reduce the reasonable price of fossil-based H₂ and promote the cost-competitiveness of biomass-based renewable H₂.

As expected, the commercially available H₂ production processes presented promising energy efficiency compared to lab-scale systems. Fossil fuel and biomass-based steam reforming processes have the greatest energy efficiency (71.6–72.7%) among the evaluated production methods with different resources, followed by electrolysis and MEC processes. For instance, in most of the electrolysis processes, heat and electricity are utilized for water splitting, and therefore, high-temperature water electrolysis methods utilize less electricity and

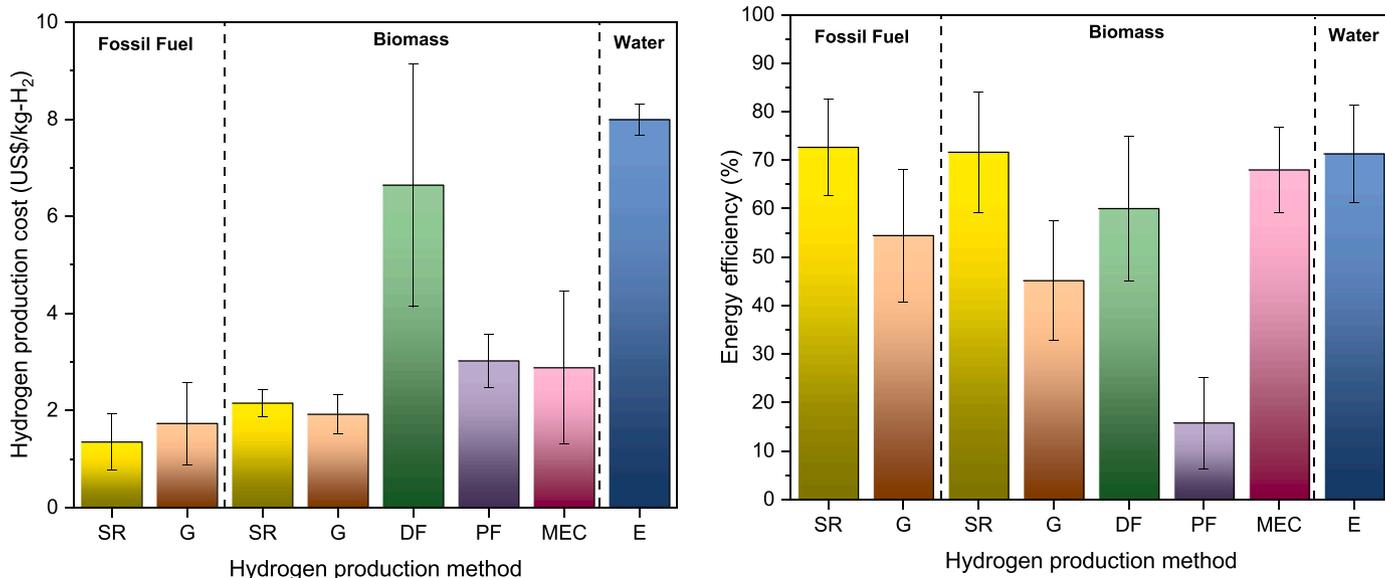


Fig. 7. Average cost and energy efficiency values of H₂ production methods for different resources.

have greater energy efficiency as thermal energy provides part of the needed energy for water splitting. On the other hand, the PF process provided the lowest energy efficiency among all processes. The most important parameter contributing to that poor energy efficiency is the lack of photoactive materials that effectively convert a considerable portion of the solar spectrum into H₂ [125]. Hence, developing effective, sustainable, robust, and reliable photoactive materials is necessary to improve the energy efficiency of the PF process.

3.3. Overall ranking

In this section, the selected environmental impact parameters, costs, and energy efficiencies are normalized with the feature scaling normalization method to compare each H₂ production method from different resources using the following equation:

$$\text{Rank}_{\text{method},i} = \left(\frac{\text{Rank}_{\text{method},i} - \text{Rank}_{\text{min}}}{\text{Rank}_{\text{max}} - \text{Rank}_{\text{min}}} \right) \times 10 \quad (46)$$

Here, the ranking ranges from 0 to 10, with 0 presenting poor performance and 10 representing the minimum impact reported. The lower values of selected environmental impact parameters are given higher rankings. In evaluated parameters, “0” is assigned to the highest environmental contaminations, highest cost, and lowest energy efficiency, while “10” defines the lowest contamination, lowest cost, and highest energy efficiency. The results of the environmental impact comparisons of H₂ production methods are presented in Fig. 8.

Regarding GWP (0/10) value, the gasification process presented the highest environmental contaminations. The GWP values were 3.96/10 for electrolysis and 4.98/10 for steam reforming, which have a significant potential to cause serious environmental effects due to CO₂ emissions. The water electrolysis process presented the highest environmental damage to water resources with WSF (0/10) and WCP

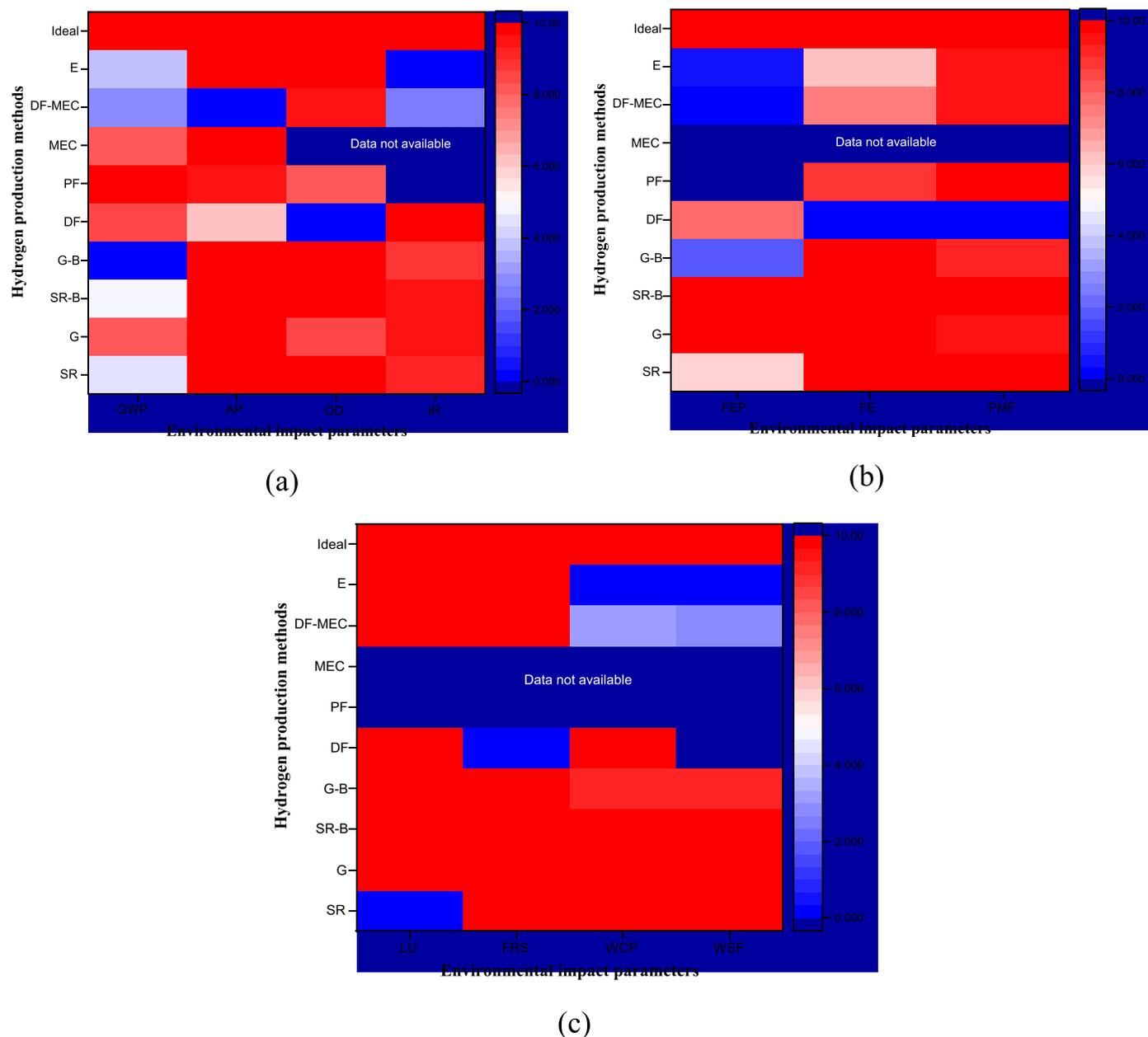


Fig. 8. Normalized ranking comparisons of environmental impact parameters of H₂ production methods: (a) for GWP, AP, OD, IR, (b) for FEP, FE, PMF, and (c) for LU, FRS, WCP, WSF.

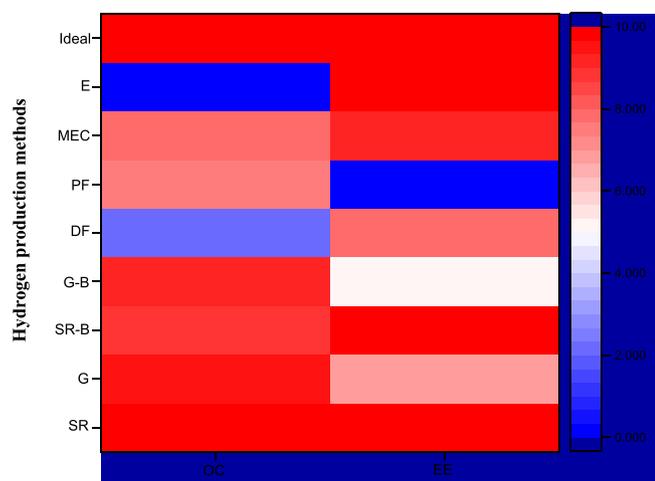


Fig. 9. Normalized ranking comparison of cost and energy efficiency values of H₂ production methods.

(0/10) values. On the other hand, among the biological processes MEC process achieves similar performance in the ideal case considering AP (9.78/10) and GWP (8.06/10) values. Moreover, the normalized ranking results proved that the PF method presented ideal results (10/10) for PMF and GWP, and close results to ideal for AP (9.65/10) and OD (8.14/10), which are the most important parameters in environmental sustainability assessment. Among the biological processes, the worst scenario was observed for the DF-MEC process with the particularly lowest GWP (2.98/10), AP and FEP (0/10), WSF and WCP (3/10), and IR (2.45/10), based on the ranking results. Overall, the normalized rankings showed that the biological processes (except the integrated DF-MEC process) provided closer-to-ideal results in view of environmental sustainability, providing less ecotoxicity, water-mineral-fossil utilizations, and emissions. The advantage of biological processes phenomena is to release less CO₂ and SO₂, and minimum utilization of renewable energy sources for heat or as well as low water consumption, respectively. Overall, replacing the energy sources used in the process with completely sustainable/renewable sources will make biological processes completely sustainable from an environmental point of view. Therefore, while existing biological processes need to be developed for commercial use, their environmental outputs should also be considered.

In addition, the normalized ranking results considering the cost and energy efficiencies of the processes, are illustrated in Fig. 9. The fossil-based steam reforming and gasification methods have the highest rankings considering their operating costs with 10/10 and 9.42/10, respectively, followed by biomass-based gasification (9.14/10) and steam reforming (8.79/10). On the other hand, water electrolysis provides the highest cost, with the lowest ranking value of 0/10 among the investigated processes. Moreover, the DF method is the most expensive process among biomass-based processes, with 2.03/10. Fig. 9 shows that in terms of energy efficiency, the high energy efficiencies were obtained for biomass (9.82/10) and fossil fuel-based (10/10) steam reforming processes. On the other hand, this process causes significant adverse environmental impacts with high GWP and AP values compared to other processes. Although the PF process has a high environmental impact parameter ranking, the worst scenario considering energy efficiency is observed for this process. The energy efficiency ranking of DF (7.77/10) and biomass-based gasification (5.16/10) processes are also relatively low, particularly compared to steam methane reforming and electrolysis. Overall, these results revealed that there is a significant trade-off between environmental impact and both the energy efficiency and cost of H₂ production processes. Namely, already commercialized H₂ production processes showed higher energy efficiencies and production costs with adverse environmental impacts than lab-scale or early-market processes. Therefore, recently, the key purpose of the successful

transition to H₂ becomes provide energy efficient and cost-effective H₂ production processes with minimum environmental adverse impacts.

4. Conclusions and future perspectives

This paper evaluates and discusses the environmental impacts of seven selected thermochemical and biological H₂ production methods to foresee the environmental sustainability levels of the processes and determine the measures to be taken in this regard. Results revealed that the fossil fuel-based gasification and steam reforming processes showed adverse environmental impacts, especially regarding air emissions. On the other hand, environmental sustainability assessment results showed that the GWP and AP of biological processes (except the DF-MEC process) are almost close to ideal conditions compared to fossil fuel gasification, steam reforming, and water electrolysis processes. However, due to energy requirements in the process operation, the integrated DF-MEC process has high AP (14.8 kgCO₂ eq./kg H₂) and GWP (0.103 kgSO₂/kg H₂). The present results showed that the most environmentally friendly method is PF, with the lowest environmental impact parameters, followed by the MEC process. Regarding the H₂ production cost, the lowest cost belonged to fossil fuel-based steam reforming at 1.35 US\$/kg-H₂ and gasification at 1.73 US\$/kg-H₂, followed by biomass-based gasification and steam reforming processes, MEC, DF, PF, and electrolysis. Moreover, the highest GWP values of 3.96/10 and 4.98/10 were observed for electrolysis and steam reforming based on the overall normalized ranking results, which have a significant potential to cause serious environmental effects due to CO₂ emissions. Regarding water use, the worst scenario was also obtained for the water electrolysis process with WSF (0/10) and WCP (0/10) values. However, among the biological processes MEC process achieves similar performance in the ideal case considering AP (9.78/10) and GWP (8.06/10) values. As expected, the commercialized H₂ production processes showed a closed-to-ideal case in terms of cost and energy efficiency. Overall, these results revealed that future studies need to focus on enhancing the H₂ production efficiency of the hybrid and biological systems in order to improve their Technology readiness level value and reduce their total costs. Moreover, producing H₂ from biomass using biological processes is not a common method, it is still a developing area. However, this paper revealed that focusing on producing H₂ from waste biomass will help minimize the use of fossil fuels, reduce the carbon footprint, produce clean and renewable H₂, and decrease the environmental effect of fossil fuels and human activities. Furthermore, it is believed that future studies would prioritize evaluating the sustainability aspects of diverse H₂ production systems using exergoeconomic and exergoenvironmental methods, two cutting-edge sustainability evaluation methodologies.

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.

Data availability

Data will be made available on request.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jece.2023.111187](https://doi.org/10.1016/j.jece.2023.111187).

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