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An atmospheric impact assessment of water-based hydrogen production methods: Sustainability evaluation

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ABSTRACT

Population growth and urbanization have significantly affected the energy demand and environmental contaminant levels worldwide. Currently, global warming with greenhouse gas emissions, air pollution, acid rain, environmental degradation, and depletion of energy resources are all consequences of utilizing fossil fuel-powered energy infrastructure. Hence, renewable energy-powered alternative energy resources must be considered to minimize atmospheric emissions and environmental contaminants. Hydrogen (H_2) has become a viable fuel to satisfy energy needs, and in recent years, there has been a lot of interest in green H_2 production, particularly using electrolysis processes that produce no emissions. In this regard, this paper utilized the atmospheric emission assessment software to evaluate atmospheric contaminants from the alkaline electrolysis (AE), proton exchange membrane-based electrolysis (PEM), and solid oxide electrolysis (SOE) processes. Among these processes, the highest CO_2 emission comes from the PEM electrolysis process, accounting for 4.68 kg-CO_2/kg-H_2, while the AE process provides the minimum total CO_2 emissions of 3.28 kg-CO_2/kg-H_2. A similar trend was observed in the particulate matter (PM) emissions, and the $PM_{2.5}$ emissions were 1.36, 1.30, and 1.24 kg-$PM_{2.5}$/kg-H_2 for PEM, SOE, and AE processes, respectively. Moreover, the environmental impact parameters of the processes were assessed, and the lowest global warming potential (GWP) of 3.32 kgCO_2-eq./kg-H_2 was obtained for the AE process. Accordingly, these results demonstrated that energy production techniques may be completely environmentally sustainable by substituting fully sustainable resources for the energy sources employed in current H_2 production methods.

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1. Introduction

The global warming tragedy and its consequences have gained worldwide interest. Therefore, since fossil fuels are one of the main reasons for global warming, a greener energy source should replace traditional fossil fuels to attain carbon neutrality. In this regard, hydrogen (H_2) becomes a possible alternative since it burns without producing carbon dioxide (CO_2) and has extremely high efficiency. Moreover, H_2 production for energy storage via water electrolysis, biomass electrolysis, or other renewable energy source use has shown to be beneficial in several ways, including reducing toxic gas byproducts like CO_x, sulphur oxide (SO_x), and nitrogen oxide (NO_x), which are not good for the environment. In addition, H_2 has a heating value three times larger than any other fossil fuel, making it the ideal option for long-term and extensive use in the transition to green energy [1]. Nevertheless, electrolysis operations account for less than 4% of H_2 production, with H_2 serving as the primary by-product of chlorine synthesis. Therefore, most H_2 requirements depend on the fossil pathway via natural gas steam reforming [2]. Due to high electricity costs and conflicting rules, the production costs of electrolysis procedures are greater than those of conventional fossil sources, which is precisely what is causing this current situation [3].

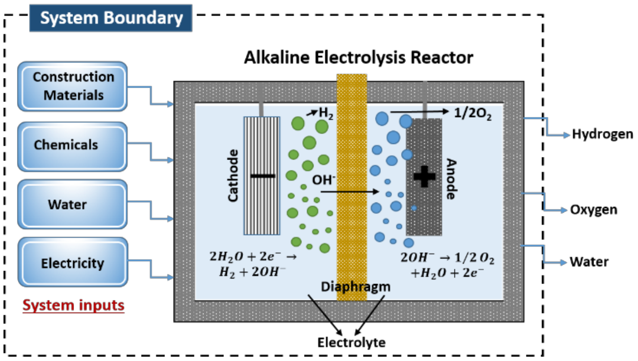
In the coming decades, there has to be a significant rise in the percentage of H_2 produced from renewable power sources to lower CO_2 emissions and achieve energy independence from fossil fuels. Water electrolysis is, therefore, a crucial technique for utilizing renewable energy to divide water into H_2 and oxygen. Combining water electrolysis with renewable energy sources is especially beneficial because extra electrical energy may be chemically stored as H_2 to close the gap between energy output and demand. Fundamentally, three methods are available for water electrolysis: solid oxide electrolysis (SOE), proton exchange membrane electrolysis (PEM), and alkaline water electrolysis (AE) [4]. Among these processes, high-temperature SOE technology is still in development, while low-temperature technologies, such as AE and PEM, provide high levels of technological readiness [5]. To date, several experimental studies have been performed on various electrolysis processes to improve their H_2 production performances and reduce environmental impacts [6,7]. Moreover, several theoretical environmental impact assessment studies have been conducted on electrolysis processes. For instance, Jolaoso et al. [8] performed a life cycle analysis of an SOE process for H_2, considering different wastewater and energy resources. They reported that the CO_2 emissions from the SOE process are 6.89 kg-CO_2eq./kg-H_2, and their results also proved that solar photovoltaic (PV) is a good energy source for SOE operations. In a separate study, the environmental impacts of the AE, PEM, and SOE processes are compared, and the life-cycle greenhouse gas (GHG) emissions of all electrolyzer technologies using low-carbon energy (biomass, solar, nuclear or wind) range from 0.3 to 2.4 kg-CO_2eq./kg-H_2 [9]. Kim et al. [10] investigated the greenhouse gas (GHG) emissions, particularly carbon dioxide emissions, from AE and PEM electrolysis processes, considering various renewable energy sources like solar, wind, and hydropower. Their results revealed that the carbon dioxide emissions for AE and PEM electrolysis processes were 8434 and 3695 kg-CO_2/kg-H_2, respectively, providing that an AE system emits about 2.3 times more greenhouse gasses than a PEM system. One potential reason for this significant difference may lie in excluding emissions associated with the membrane production stage in the assessment. Membrane production involves energy-intensive manufacturing processes and the utilization of specialized materials, both of which contribute to additional carbon emissions. By not accounting for these emissions, the environmental impact of the PEM system might appear underestimated in comparison to the AE system. Including these factors in the analysis could reveal a more comprehensive picture of the total greenhouse gas emissions, potentially reducing the observed gap between the two systems. Moreover, their study focused exclusively on carbon dioxide emissions, without considering other GHG emissions or environmental impacts that may also contribute to the overall footprint of the electrolysis processes. Zhang et al. [11] performed the life cycle assessment of onshore-offshore wind-powered H_2 production through water electrolysis, and it is clear from the study of the environmental impact proportion that the process of building infrastructure has a major influence on the total environmental effect. Wei et al. [12] also investigated the environmental impacts of four electrolyzer technologies SOE, PEM, anion-exchange membrane (AEM), and AE. The authors reported that significant CO_2 reductions are feasible with SOE and AEM electrolyzers, both of which are still in the development stage, whereas PEM and AE, being mature technologies, provide few possibilities for improvement.

Previous studies have primarily focused on process optimization and the evaluation of environmental impact parameters without providing a detailed analysis of the atmospheric pollutants influencing these parameters, both in type and magnitude. Moreover, they have predominantly focused on the operational phase of processes, overlooking the broader lifecycle impacts. This narrow scope has proven insufficient for generating realistic and comprehensive results. Since most of the literature studies only focused on the direct environmental impact parameters of these electrolysis processes. There is a critical need to evaluate the individual atmospheric emissions from electrolysis processes to understand the main contaminant types. Consequently, the novelty of our research lies in addressing this gap by incorporating emissions from all lifecycle stages, including material production, transportation, and operational phases. By identifying and quantifying the emissions that influence environmental parameters in detail, our study provides a more holistic and accurate environmental assessment. This comprehensive approach offers meaningful insights and practical relevance, making it better suited for real-world applications compared to prior research. Another novelty of this work also lies in its ability to address critical limitations of traditional approaches by offering significant advancements in cost-efficiency, reliability, and performance. By utilizing sustainable and readily available materials, the proposed system substantially reduces overall costs, making it a more economically viable solution. Furthermore, the design ensures enhanced reliability by integrating robust operational features that maintain efficiency under varying conditions. The optimization of key performance parameters further contributes to its superior functionality, ensuring consistent and high-quality outcomes. These advancements collectively position this work as a practical and scalable solution for real-world applications, bridging the gap between theoretical potential and operational feasibility.

In this context, the key purpose of this study is to evaluate the atmospheric emissions of different electrolysis-based H_2 production processes (alkaline, solid oxide, and proton exchange membrane-based electrolysis processes) using greenhouse gases, regulated emissions, and energy use in transportation (GREET) model, considering the feedstock utilization and extraction, energy production, transportation, and H_2 production stages. Overall, the critical goals of this paper are: (i) to review and discuss the benefits, limitations, and future perspectives of specified electrolysis processes for H_2 production, (ii) to assess and compare the environmental impacts and atmospheric emissions of the processes presenting their atmospheric contaminants for whole process chain, including feedstock utilization, transportation, and H_2 production stages, and (iii) to compare electrolysis processes to provide the best option in sustainability manner, considering their sustainability levels. Consequently, this study assesses the atmospheric emissions and environmental effects of electrolysis technologies for effective and sustainable H_2 production. Overall, this study proved easy to identify which steps in the electrolysis processes have the greatest environmental impacts and what kind of measures should be taken at which steps to prevent the potential emissions by measuring the atmospheric emission of the processes from feedstock processing to H_2 production.

2. System description and analysis

In this section, a description of the alkaline, solid oxide, and proton exchange membrane-based water electrolysis systems with boundaries for H_2 production are reported, and their environmental impacts are assessed, considering atmospheric emissions from these processes (Figure 1). A GREET study usually includes the following procedures: defining the objectives and scope, reviewing the inventory, assessing the environmental effect, and interpreting the results [13]. Furthermore, the total emissions from every stage are standardized to the preferred functional unit, which is established by the kg of H_2 produced for different H_2 production pathways. For the suggested system, the emissions of CO_2, CO, greenhouse gas emissions (GHG-100), particulate matter ($PM_{2.5}$ and PM_{10}), NO_x, volatile organic compounds (VOCs), etc., were calculated and addressed separately.



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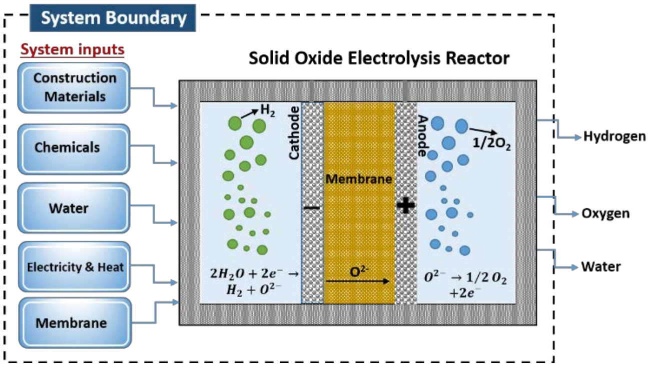

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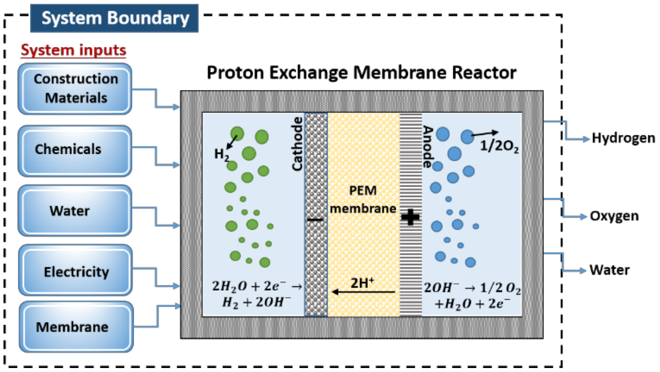

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Figure 1. *System boundaries and processes of investigated water electrolysis systems: (a) alkaline electrolysis, (b) solid oxide electrolysis, and (c) proton exchange membrane electrolysis.*

The GREET software, developed by Argonne National Laboratory, is a comprehensive and freely accessible life-cycle analysis tool widely used for assessing the energy consumption and atmospheric emissions of various technologies, including electrolysis processes such as alkaline, solid oxide, and proton exchange membrane systems. GREET models the entire life cycle of hydrogen production, from resource extraction to the end-use phase, enabling the quantification of GHG emissions, regulated air pollutants, and energy consumption. For electrolysis systems, the software requires inputs such as the type of system (alkaline, solid oxide, or PEM), energy source, and regional electricity grid mix (e.g., renewable, fossil-based, or nuclear), operating conditions like temperature and pressure, and material requirements for components like electrodes and electrolytes. Outputs generated by GREET include GHG emissions (e.g., CO_2, CH_4, N_2O), criteria air pollutants (e.g., NO_x, SO_x, $PM_{2.5}$, and PM_{10}), and energy consumption across the life cycle stages. Input values in Table 1 were entered into the model based on data from experimental studies on the processes examined, and emissions were calculated accordingly.

Table 1. *Typical specification of studied electrolyzers for 1 kg H_2 production*.

Specification	AE	PEM	SOE
Cell temperature	60 °C	50 °C	900 °C
Current density	0.4 A/cm^2	2 A/cm^2	1 A/cm^2
Voltage	2.4 V	2.2. V	1 V
Cell area	4 m^2	4 m^2	4 m^2
System life time	20 year	20 year	20 year
Hydrogen purity	99.9 %	99.9 %	99.9 %
Stack life time	90000 h	20000 h	30000 h

The software employs detailed formulations and mathematical models to evaluate emissions and energy use. These include energy balance equations to calculate electricity and heat requirements, material flow analysis to estimate emissions from raw material production and system operation, and electrochemical models to incorporate cell efficiency, hydrogen yield, and system degradation. For alkaline electrolysis, GREET accounts for emissions related to producing KOH electrolytes, steel electrodes, and operational energy use. For PEM electrolysis, it models the impacts of platinum group metals in the catalyst and the ion-conducting membrane. Similarly, solid oxide electrolysis inputs include high-temperature energy demands and ceramic material production. GREET assesses emissions by simulating the entire life cycle of each electrolysis system, identifying emission hotspots, and quantifying direct emissions, such as those from electricity generation, and indirect emissions from upstream activities like material production and transportation. It also allows scenario analysis, enabling emissions comparisons under different energy grid configurations (e.g., renewable vs. coal-based electricity) or optimization strategies, such as using waste heat or renewable electricity. In this study, a renewable-based energy grid is considered for all electrolysis processes. Overall, by integrating material and energy flows with life-cycle inventory data, GREET provides a robust framework for quantifying and comparing the environmental impacts of alkaline, solid oxide, and PEM electrolysis systems, offering actionable insights to advance sustainable hydrogen production. Consequently, all atmospheric emission analyses were conducted using the GREET software database, with inputs derived from the dataset of the project. This dataset was specifically developed to reflect the unique characteristics and parameters of the system under investigation, ensuring that the analyses accurately represent the study's context. By integrating this customized dataset into the GREET software, the evaluation of life-cycle emissions and energy use was carried out with precision and alignment to the objectives of the study. As a result, this section does not include additional analyses beyond those facilitated by the GREET database and the tailored dataset of the project.

2.1 Alkaline electrolysis

In particular, the AE is a scalable, adaptable, and widely utilized method for producing pure H_2 [14]. Fundamentally, an alkaline electrolyzer splits water into H_2 and HO^- at the cathode. The latter moves through the diaphragm and electrolyte before discharging at the anode and releasing the O_2. Low temperatures (60-80 °C) are used for AE, and the electrolyte used in the process is an aqueous solution of potassium hydroxide (KOH), sodium hydroxide (NaOH), or sodium chloride (NaCl), with a concentration of around 25–30% and applied voltage of 1.8 to 2.2 V. In contrast to traditional AEs, advanced AEs perform at a comparatively low voltage of 1.6 V and a current density of 2 A/cm^2. Moreover, AE produces H_2 at a purity of 99.9%, meeting fuel cell requirements for high-quality H_2 production [15]. Overall, although AE is an outdated technology, it remains one of the most straightforward and appropriate ways of producing H_2. However, AE has challenges related to energy consumption, durability, safety, and installation and maintenance costs [16]. Nevertheless, AE is a well-established method; industrial electrolyzers are available, and it is an environmentally friendly method of producing H_2 if the power is produced using CO_2-free methods like nuclear and renewable resources. For instance, together with innovative electrode technologies, the chlor-alkali electrolyzer's developed zero-gap technology offers improved performance [17].

2.2. Solid oxide electrolysis

Steam electrolysis utilizing an SOE is one method of water electrolysis currently being developed. Its high working temperatures of 600-1000 °C significantly reduce electrical energy usage [18]. Compared to other electrolysis processes, SOE technology with intermediate temperature has an electrical energy usage of around 3 kWh/Nm^3 of H_2. A SOE comprises a positive O_2 electrode, a negative fuel electrode, and an electron-insulating electrolyte with high ionic conductivity at high temperatures [19]. Namely, a subsequent rise in operating temperature results in a significant increase in thermal energy demand and a decrease in electrical energy demand in the SOE process, and the operating temperature does not affect the total energy requirement. High-temperature SOE is, therefore, beneficial as it offers more chances to use industrial waste heat to produce H_2. In SOE operation, high-temperature steam is supplied to the cathode side, where the catalyst site converts it to H_2 and O_2 gases. The phase potential then moves the O_2 ions across the electrolyte to the anode side, undergoing oxidation to produce O_2 molecules [20]. Nevertheless, in the SOE process, detailed phenomena often involve a complex interplay of material qualities, operating factors, reaction kinetics, and thermodynamics. Since SOE is still an emerging technology, modeling may be very important for both the design and management of SOEs. Overall, SOE and stacks operate at substantially higher temperatures, which accelerates reaction kinetics and may lead to improved electrical efficiency. Thermodynamically stated, waste heat from industrial operations or high-temperature heat from solar thermal power can supply a portion of the energy required for the endothermic water-splitting reaction, lowering the need for electric energy. Moreover, CO_2 and water molecules can be divided via high-temperature electrolysis, or a combination, to create synthesis gas or other energy carriers like methanol or methane through catalytic conversion.

2.3. Proton exchange membrane electrolysis

General Electric created the first water electrolyzer based on a solid polymer electrolyte concept in developing water electrolysis technology for effective H_2 production [21]. This novel technique was originally called a PEM or polymer electrolyte membrane. The PEM electrolyzer and the alkaline electrolyzer have a similar construction. The key difference is forming a zero-pole separation by employing a thin film-electrode assembly called a membrane electrode, and the Nafion-type membrane separator becomes very acidic after soaking in water. The anode and cathode catalysts are hot-pressed or electroless-plated onto both sides of the separator.

In the PEM electrolyzer, at 10000 to 20,000 A/m^2, the DC density is approximately 5 times higher than that of the AE process [22]. Moreover, the PEM electrolyzer can function at varying pressures because of its operating temperature range of 50 to 80 °C, pressure of less than 5 MPa, and lower volume than the AE. Fundamentally, in the PEM electrolyzer, the deionized water just needs to reach the anode side of the electrolyzer, in contrast to the alkaline electrolyzer. Following energization, the membrane electrode's anode side produces H_2 and O_2 ions, while the cathode side receives the H_2 ions in their hydrated form, which produces H_2 gas. Then, the O_2 and H_2 are separated by the membrane. The Nafion membrane separation and lack of solution voltage drop give this approach a high energy efficiency for producing H_2. In addition, compared to the AE process, PEM offers several benefits, including excellent safety, ease of handling and maintenance, low gas crossover, high proton conductivity, low power consumption, high purity of H_2, large size and mass, high-pressure operation, and control over electrical power variations.

3. Results and discussion

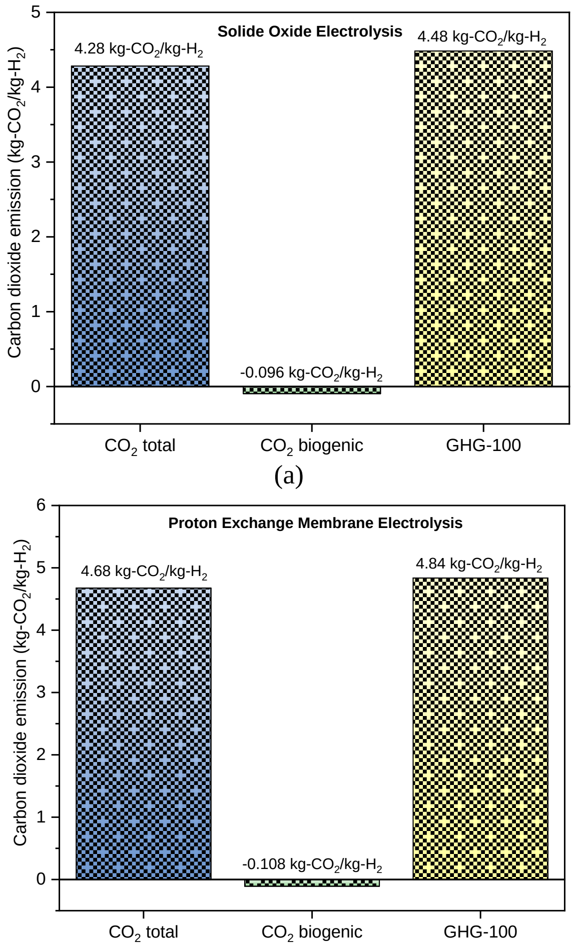
In this section, the atmospheric emissions and other environmental impact parameters from the three different water-based H_2 production methods (alkaline, solid oxide, and proton exchange-based electrolysis) are evaluated and discussed comprehensively. Furthermore, the normalized ranking method presents their sustainability potential based on impact parameters.

The evaluation of atmospheric emissions and environmental impact parameters for the three water-based hydrogen production methods (alkaline electrolysis, solid oxide electrolysis, and proton exchange membrane electrolysis) was conducted using distinct methodologies. Atmospheric emissions were calculated and compared using the GREET software, which provides a comprehensive life-cycle analysis framework. Environmental impact assessment, on the other hand, was performed by utilizing the coefficients of impact categories listed in Table 1. These coefficients facilitated a detailed quantification of environmental problems across various categories, enabling a comparative analysis of the three methods.

3.1. Atmospheric emissions

3.1.1. Carbon dioxide emission

The enormous increase in the concentration of anthropogenic CO_2 in the atmosphere from the mid-1800s is widely attributed to the increased usage of fossil fuels since the beginning of the Industrial Revolution [23]. Therefore, considering SDGs, there is a significant need to explore green energy resource alternatives and make our current energy systems more sustainable to overcome huge amounts of CO_2 emissions. In this context, the H_2 is one of the more favorable options for present-day and future energy carriers. Namely, commercialized gasification, steam reforming, and pyrolysis techniques are currently used to produce H_2 from fossil fuel resources, accounting for most H_2 production worldwide [4]. Nevertheless, a big disadvantage of fossil fuel-based H_2 production technologies is the large quantities of CO_2 emissions since they utilize fossil fuels in both heat source and manufacturing steps. Recently, to overcome these problems most of these systems combined with the different CO_2-capture processes. Moreover, the water-based electrolysis processes have significant advantages owing to their low CO_2 emissions. Overall, it is important to consider the total chain of activities from resource utilization to H_2 production to evaluate if using different resources and/or processes instead of the currently available options will decrease or increase GHG emissions. In this regard, the CO_2 and GHG emissions from different electrolysis processes are evaluated in this study (Figure 2).



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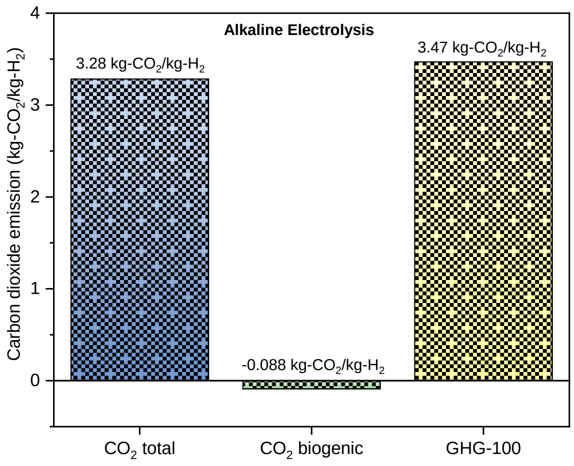

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Figure 2. *The CO_2 emissions from (a) solid oxide, (b) proton exchange membrane, and (c) alkaline electrolysis processes.*

The emissions from the transportation, feedstock use, and H_2 production phases contribute to the overall CO_2 emission values of the processes. For the SOE process, the CO_2 emission was 4.28 kg-CO_2/kg-H_2 with a GHG emission value of 4.48 kg-CO_2/kg-H_2 (Figure 2a). Namely, the CO_2 emissions associated with SOEs depend on several factors, primarily the source of electricity and heat. If renewable energy sources, such as wind or solar, are used to power the process, direct CO_2 emissions can be near zero. However, if fossil fuels provide electricity, emissions can rise significantly, with estimates suggesting that around 0.5 to 1 kg of CO_2 could be released per kg of H_2 produced. Additionally, high-temperature heat is often required, and if generated from natural gas or coal, this could add another 1 to 2 kg of CO_2 per kg of H_2, depending on the fuel efficiency. Another source of CO_2 emissions arises from the production and disposal of SOE components, such as ceramic electrolytes and metal interconnects, which may contribute between 5 and 10% of the overall carbon footprint, depending on material choice and manufacturing processes. Our findings agree with the literature studies, considering these possible sources of CO_2 emissions from electrolytes and materials production processes for the electrolysis processes. Therefore, these results revealed that achieving low-emission SOE operation requires integrating renewable electricity, efficient heat recovery, and sustainable materials, with each factor playing a crucial role in reducing overall emissions. For example, the CO_2 emission from the wind-powered electrolysis process was reported as 0.95 kg-CO_2/kg-H_2; however, the author neglected to account for the emissions from the transportation and extraction of materials [24].

Moreover, the CO_2 and GHG emissions from the PEM electrolysis process are evaluated, and the CO_2 and GHG emissions are 4.68 kg-CO_2/kg-H_2 and 4.84 kg-CO_2/kg-H_2, respectively. The results reveal that the membrane production procedure contributes the highest CO_2 emissions in the PEM process, considering the material extraction and chemical utilization steps. Therefore, selecting green and sustainable materials with low environmental emissions for membrane production can effectively decrease the CO_2 emission of the overall system. Similarly, Zhang et al. [25] evaluated the environmental impacts of the PEM process for solar energy-powered H_2 production, considering various lifetimes. The lowest CO_2 emission was found to be 4.63 kg-CO_2/kg-H_2 for a lifetime of 60 years, while it was 9.67 kg-CO2/kg-H_2 for a lifetime of 30 years. Overall, their results provided that the lifetime of the system is another important parameter to understand and minimize its environmental impacts. As estimated, the lowest total CO_2 emission of 3.28 kg-CO_2/kg-H_2 is obtained for the AE process, while its GHG emission was 3.47 kg-CO_2.eq./kg-H_2 (Figure 2c). For the AE process, the operational phase consumes primarily electricity and water which are the main two inputs of the life cycle of H_2 production, therefore, the electricity production step is the main contributor to the CO_2 emissions in the AE process. Overall, compared to the three electrolysis processes, the highest CO_2 emissions were obtained for the PEM process due to its complex membrane production procedure.

Furthermore, the biogenic CO_2 emissions of the electrolysis technologies are evaluated and presented in Figure 2. Fundamentally, the term "biogenic CO_2 emissions" mostly refers to CO_2 that comes from biological sources including plants, animals, and microbes, and is a component of the natural carbon cycle. Biogenic CO_2 is a component of very short-term carbon cycles, in contrast to fossil CO_2, which is produced by burning fossil fuels. Additionally, biogenic CO_2 emissions may be negative; in this case, the process sequesters more CO_2 than it releases. For example, because plants grow and store carbon in biomass and soils, some agricultural methods and reforestation may absorb CO_2 emissions from the atmosphere. As a result, the idea of biogenic CO_2 emission is crucial for carbon accounting and environmental impact analyses, particularly in fields like bioenergy, forestry, and agriculture. Namely, the formation of negative biogenic CO_2 emissions is possible when the input electricity is derived from renewable sources and the feedstock contains biomass or biogenic CO_2 sources in PEM, AE, and SOE processes. In this context, coupling the electrolysis with carbon capture technologies can lead to net-negative emissions. By integrating biogenic CO_2 into these processes, and utilizing renewable electricity, the system can contribute to a reduction in atmospheric CO_2 levels, resulting in net-negative biogenic CO_2 emissions. This approach aligns with carbon-negative strategies aimed at combating climate change by removing more CO_2 than is emitted, thereby establishing these processes as viable contributors to a sustainable carbon economy. Our results revealed that there was no significant change in the biogenic CO_2 emissions from the different electrolysis processes. The negative biogenic CO_2 emissions of -0.096, -0.108, and -0.088 kg-CO_2/kg-H_2 were observed for the SOE, PEM, and AE processes. The similarity in results among the PEM, AE, and SOE processes can be attributed to their shared goal of splitting water into H_2 and O_2, despite differences in electrolyte type, operational temperature, and design. Each method effectively achieves high efficiencies under optimized conditions, especially when powered by renewable energy sources. While SOE operates at higher temperatures, leading to slightly different thermodynamic efficiencies, all three methods benefit from advanced catalyst developments and enhanced system integration, reducing variations in overall H_2 production efficiency. Moreover, when these systems utilize biogenic CO_2 capture, they demonstrate comparable carbon-negative potentials, as they all facilitate CO_2 conversion and sequestration with similar effectiveness, resulting in closely associated outcomes across these electrolysis technologies.

3.1.2. Nitrogen oxide, carbon monoxide, and methane emissions

Nitrogen oxide (NO_x), also referred to as laughing gas, is another major greenhouse gas that has an impact 298 times higher than CO_2 [26]. Many chemical facilities produce nitrogen oxides as by-products, including nitric acid and adipic acid factories, and catalytic converters are needed to handle N_2O [27]. In this context, the NO_x emissions are also critical atmospheric emissions from the electrolyzers for H_2 production. The NO_x emissions from SOE, PEM, and AE processes are reported in Figure 3. These results showed that the NO_x emissions are highest in the PEM electrolysis system, with an emission concentration of 3.66 kg-NO_x/kg-H_2. The SOE system has a similar but slightly lower NO_x emission level at 3.16 kg-NO_x/kg-H_2, and the AE system has the lowest NO_x emissions, at 2.68 kg-NO_x/kg-H_2. Basically, these differences are influenced by the interaction of nitrogen and oxygen at various operational conditions and by the energy sources and materials used in these electrolysis systems. The high NO_x emissions in the PEM system could be related to its operating environment, where nitrogen can interact with oxygen species, forming NO_x compounds. Although SOE operates at high temperatures, which could theoretically encourage NO_x formation, the system's design typically minimizes direct nitrogen exposure, keeping NO_x levels slightly lower. On the other hand, AE has the lowest NO_x emissions, largely due to the alkaline environment (KOH solution) and the low likelihood of nitrogen oxidation at its operating temperature. Nickel and stainless steel materials used in alkaline systems also contribute minimally to NO_x formation. Moreover, PEM electrolysis operates with a membrane that allows for efficient H_2 separation but often relies on grid electricity, which, if sourced from fossil fuels, can lead to higher indirect NO_x emissions. A similar trend also can be observed for the other electrolysis processes without the utilization of renewable energy resources.

In terms of CO emissions, PEM electrolysis emits the highest concentration at 10.29 kg-CO/kg-H_2, followed by SOE at 9.99 kg-CO/kg-H_2, and AE at 9.69 kg-CO/kg-H_2. The higher CO emissions in PEM and SOE systems can be linked to the operating temperatures and the types of catalysts and materials used. PEM electrolysis relies on a polymer electrolyte membrane, typically made from materials like Nafion, and requires precious metal catalysts such as platinum. These catalysts can enhance reaction rates, but at moderate operating temperatures (50-80 °C), incomplete oxidation can occur, leading to CO as a by-product. Furthermore, SOE systems are often designed to integrate waste heat or renewable energy sources, like solar or geothermal, which can further reduce indirect CO emissions by lowering the need for external fossil-based electricity.

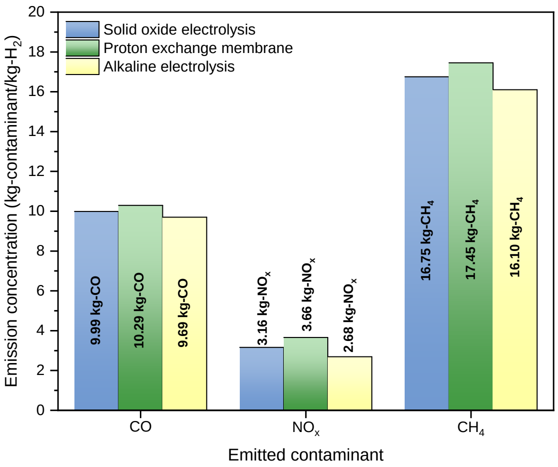


Figure 3. *The CO, NO_x, and CH_4 emissions from SOE, PEM, and AE processes.*

A similar trend was obtained for the CH_4 emissions. Namely, according to some reports, the CH_4 emissions from utilizing fossil fuels to produce H_2 would be comparable to those from using traditional natural gas sources to produce power [28]. Furthermore, the extraction and transportation of feedstocks can also result in CH_4 emissions from H_2 production activities; hence, incorporating these operations into the system boundaries results in a significantly greater contribution of CH_4 emissions to the impact of global warming [29]. Controlling CH_4 emissions from the extraction of feedstock and its transportation to the production of H_2 is therefore essential. Based on our results, the CH_4 emissions are most significant in PEM electrolysis with 17.45 kg-CH_4/ kg-H_2, followed by SOE at 16.75 kg-CH_4/ kg-H_2, and as expected, AE system with emission of 16.10 kg-CH_4/ kg-H_2. Overall, the materials and energy sources used in each electrolysis system significantly impact their emission profiles. In summary, the choice of electrolysis system should consider not only the direct emissions from the reaction process but also the environmental footprint of materials and the source of electricity used. Systems like AE, when powered by renewables or integrated with waste energy, offer lower overall emissions. In contrast, PEM and SOE systems may require cleaner electricity sources and careful management of catalyst and membrane materials to reduce indirect emissions.

3.1.3. Volatile organic carbon and sulfur oxide emissions

The SO_x emissions from the various H_2 production processes vary greatly; the environmental profiles of the gasification, electrolysis, and dark fermentation-microbial electrolysis cell (DF-MEC) processes are different. One notable source of SO_x emissions is gasification, especially when coal or other sulfur-containing feedstocks are used. During gasification, the sulfur in the feedstock is liberated as hydrogen sulfide (H_2S), which burns to produce SO_2, which causes acid rain and respiratory disorders. However, because electrolysis separates water into H_2 and O_2 without the need for any intermediates that contain sulfur, it produces very little SO_x emissions, particularly when it is fueled by renewable energy sources. In the same way, DF-MEC usually emits little SO_x. However, there may be very little SO_x emissions if the feedstocks include sulfur compounds. The SO_x emissions from the considered electrolysis processes are reported in Figure 4. When examining SO_x emissions, PEM Electrolysis has the highest emission rate at 4.65 kg SO_x/kg-H_2, whereas the SOE process emits 4.37 kg SO_x/kg-H_2, and the AE shows the lowest SO_x emissions at 4.11 kg SO_x/kg-H_2.

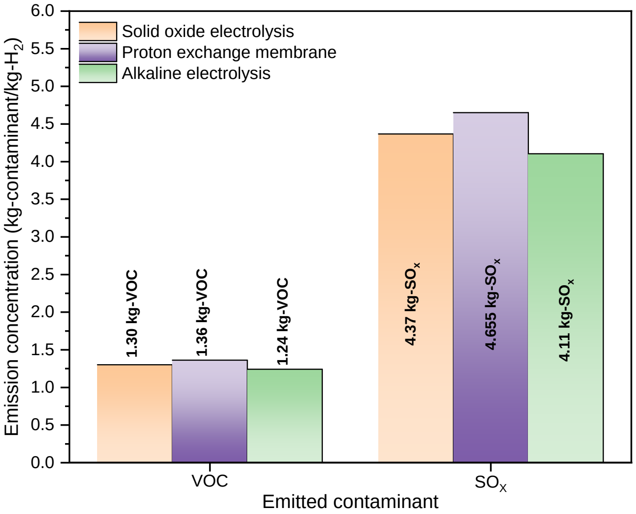


Figure 4. *The VOC and SO_x emissions from SOE, PEM, and AE processes.*

In addition, electrolysis processes present unique potential sources of VOC emissions, although generally low compared to fossil fuel-based processes. For instance, AE exhibits the lowest VOC emissions of 1.30 kg-VOC/kg-H_2, followed by SOE with 1.36 kg-VOC/kg-H_2, and PEM electrolysis with 1.42 kg kg-VOC/kg-H_2. The underlying reasons for these differences are primarily attributed to the distinct operating temperatures, electrolyte structures, and energy consumption patterns inherent to each electrolysis method. For instance, the SOE process typically operates at high temperatures, which enhances energy efficiency, but the elevated temperatures can partially promote the formation of VOC emissions. On the other hand, in PEM electrolyzers, VOC emissions may result from the degradation of polymeric materials within the membrane under prolonged exposure to high temperatures and operational stresses. The AE, which uses potassium hydroxide as the electrolyte, can also lead to VOC release if organic impurities present in the electrolyte or other system components degrade during operation. Overall, minimizing VOC emissions in these systems involves choosing durable materials, implementing effective sealing strategies, and controlling operating conditions to prevent thermal and chemical degradation.

3.1.4. Particulate matter emissions

Another significant problem is air pollution driven by particle matter (PMs), rather than SO_x and NO_x. Specifically, PMs can have a harmful influence on the environment, such as deteriorating air visibility, and on human health, such as cardiovascular disease and respiratory issues [30]. Additionally, the PMs are divided into two groups: $PM_{2.5}$, which is defined as particulate matter with a diameter of less than 2.5 µm, and PM_{10}, which is defined as particulate matter with a diameter of less than 10 µm. Because of its higher potential to cause health problems, $PM_{2.5}$ has been studied more than the other PMs [31]. Although they constitute a small percentage of all PM, $PM_{2.5}$ pollutants also have high surface-to-volume ratios, which enable them to concentrate sulfur compounds, nitrogen oxides, acids, heavy metals, bacteria, and more on their surfaces. Thus, it is essential to assess the PM emissions from the different H_2 production processes to comprehend their effects on the environment, create capturing methods, and/or implement stringent restrictions to reduce potential emissions. In this context, Figure 5 reports the $PM_{2.5}$ and PM_{10} emissions from the SOE, PEM, and AE H_2 production processes.

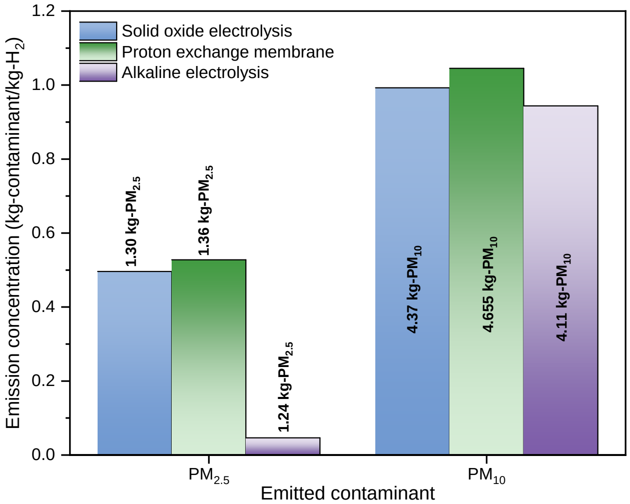


Figure 5. *The particulate matter emissions from SOE, PEM, and AE processes.*

Our results revealed that the highest PM emissions were observed for the PEM electolysis process with a $PM_{2.5}$ concentration of 1.36 kg-$PM_{2.5}$/kg-H_2 and a PM_{10} concentration of 4.65 kg-PM_{10}/kg-H_2. This result can be explaine with that in PEM electrolysis process for H_2 production, sources of $PM_{2.5}$ and PM_{10} emissions primarily arise from indirect stages related to the manufacturing, transportation, and disposal of materials and energy inputs. These particles can be formed during the production of high-purity metals like platinum, iridium, and titanium, used in catalysts and cell components, which often involve mining and refining processes that emit fine particulates. In addition, if sourced from fossil fuel-based energy, the electricity used to power the electrolyzer contributes to particulate emissions through combustion processes, particularly coal or natural gas plants. Manufacturing of the PEM membranes and the operation of machinery involved in the assembly and maintenance of PEM systems also release PMs. Thus, while the electrolysis process does not emit $PM_{2.5}$ or PM_{10} directly, upstream and downstream processes associated with materials and energy production may contribute to PM emissions in the H_2 production lifecycle. Similar PM emissions values were observed for the SOE process with a $PM_{2.5}$ concentration of 1.30 kg-$PM_{2.5}$/kg-H_2 and a PM_{10} concentration of 4.37 kg-PM_{10}/kg-H_2. On the other hand, the lowest PM emissions were obtained for the AE process. For the AE process, the $PM_{2.5}$ and PM_{10} emissions were 1.24 kg-$PM_{2.5}$/kg-H_2 and 4.11 kg-PM_{10}/kg-H_2. The manufacturing of electrodes and other cell components involves mining and metallurgical processes that generate PM due to the handling, crushing, and smelting of metals. Additionally, the production of the alkaline electrolyte solution requires chemicals like potassium hydroxide (KOH) and its synthesis and transportation can lead to PM emissions.

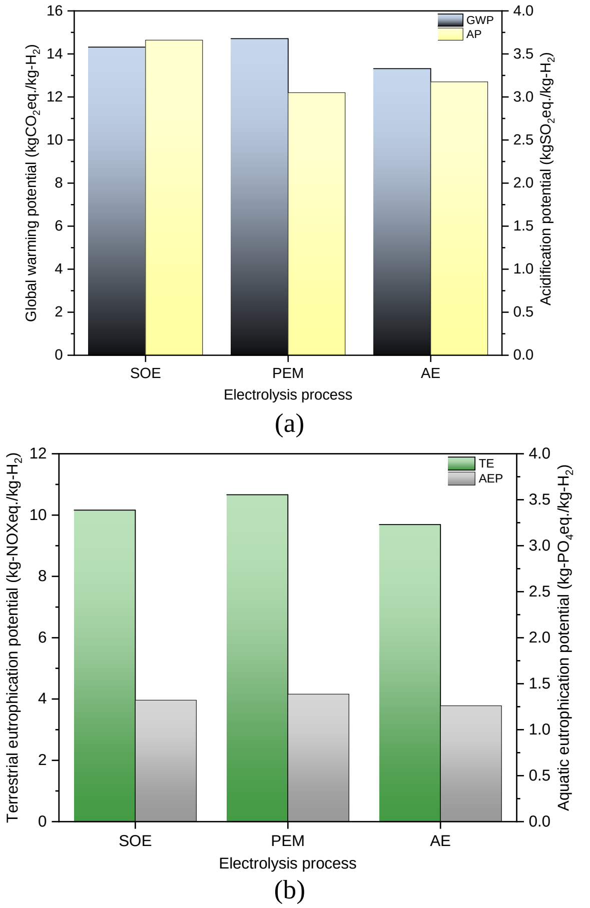
3.2. Environmental impact assessment

In this section, critical environmental impact parameters like acidification potential (AP), aquatic eutrophication (AEP), global warming potential (GWP), and terrestrial eutrophication (TE) potentials of the electrolysis processes are evaluated using the coefficients of impact categories (Table 2), which are reported by Fallahpour et al. [32].

Table 2. *Characterization values of emissions for various environmental impact categories.*

Substance (in kg)	Global warming potential (in kg-CO_2eq./kg)	Acidification potential (in kg-SO_2eq./kg)	Terrestrial eutrophication potential (in kg-NO_xeq./kg)	Aquatic eutrophication potential (in kg-PO_4eq./kg)
CO_2	1	-	-	-
CH_4	21	-	-	-
SO_2	-	1	-	-
NO_x	-	0.28	1	0.13

Figure 6a reports the GWP and AP values of the processes based on their emissions of CO_2, CH_4, SO_2, and NO_x. Since it converts GHG emissions into CO_2 equivalent, the GWP is a widely recognized indicator for comprehending the environmental consequences of the systems and can be used to determine its overall emissions, making it one of the most significant LCA parameters of importance in this study [33]. The amount of toxic acid accumulated in soil, groundwater, surface water, and organisms, as well as variations in acidity and SO_2 emissions, are all represented by the AP. Fuel or biomass burning for energy production and transportation are the primary sources of emissions that become acidic [34]. For instance, the GWP and AP values of 4.71 kgCO_2-eq./kg-H_2 and 3.05 kg-SO_2eq./kg-H_2 are observed for the PEM electrolysis process most probably related to the production and disposal of materials like Nafion in PEMs contributing to GWP and acidification due to perfluorinated compounds. Moreover, the TE and AEP values are other important impact parameters considered in this paper, and these parameters for different electrolysis processes are illustrated in Figure 6b. Terrestrial (10.66 kg-NO_xeq./kg-H_2) and aquatic eutrophication (1.38 kg-PO_4eq./kg-H_2) potentials in PEM electrolysis are also influenced by the emissions from raw material extraction and membrane manufacturing.



(a)

(b)

Figure 6. *Different electrolysis-based hydrogen production processes: (a) GWP and AP values and (b) TE and AEP values.*

For SOE processes, although the GWP value (4.31 kgCO_2-eq./kg-H_2) was similar to PEM, the AP value (3.66 kg-SO_2eq./kg-H_2) was found to be higher than PEM most probably related to its operating temperature. Namely, the high operating temperatures lead to material degradation, increasing the demand for replacement parts and, consequently, environmental burdens from material production. Furthermore, ceramic materials in the SOE process can release pollutants during their production, contributing to acidification and eutrophication (TE: 10.16 kg-NO_xeq./kg-H_2 and AEP: 1.32 kg-PO_4eq./kg-H_2). On the other hand, the lowest environmental impacts are observed for the conventional AE process. For instance, the lowest GWP of 3.32 kgCO_2-eq./kg-H_2 is observed for the AE process, while its AP value was 3.17 kg-SO_2eq./kg-H_2. Moreover, the TE and AEP values were 9.69 kg-NO_xeq./kg-H_2 and 1.26 kg-PO_4eq./kg-H_2 for the AE process, respectively. These results revealed that the AE process, while typically less energy-intensive than PEM, still contributes to GWP due to the electricity source. The potassium hydroxide electrolyte used in AE can lead to acidification and eutrophication if released into the environment. Material processing and disposal stages, particularly involving electrodes, also contribute to environmental impacts. Consequently, across all these processes, the impact on acidification and eutrophication potentials is strongly correlated with raw material extraction, chemical use, and disposal practices, underscoring the importance of sustainable energy and material sourcing in mitigating these environmental effects.

Overall, this study extensively evaluates the atmospheric emissions and environmental impacts of three widely used water-based H_2 production methods: AE, SOE, and PEM. Among these, AE emerged as the most environmentally favorable option, exhibiting the lowest CO_2 emissions (3.28 kg-CO_2/kg-H_2), global warming potential (3.32 kg-CO_2-eq./kg-H_2), and particulate matter emissions (1.24 kg-$PM_{2.5}$/kg-H_2). Its straightforward design and scalability make AE a robust choice for integration with renewable energy sources, potentially achieving a fully sustainable H_2 production cycle. However, AE is not without its limitations, such as the dependency on chemical-intensive electrolytes like KOH, which may contribute to localized environmental concerns if improperly managed. Conversely, the PEM process demonstrated the highest CO_2 (4.68 kg-CO_2/kg-H_2), methane (17.45 kg-CH_4/kg-H_2), and particulate emissions (1.36 kg-$PM_{2.5}$/kg-H_2), primarily due to the complex and energy-intensive membrane production process involving precious metals. While PEM offers advantages in terms of operational flexibility, high H_2 purity, and compact system design, its environmental footprint and cost remain critical drawbacks. On the other hand, the SOE process, characterized by its high operating temperatures, exhibited intermediate environmental performance. Although SOE benefits from reduced electrical energy demands and the potential to harness industrial waste heat, its reliance on ceramic materials and high thermal energy inputs presents challenges, particularly in terms of material degradation and acidification potential (3.66 kg-SO_2eq./kg-H_2).

One of the major contributions of this study lies in its comprehensive lifecycle approach, incorporating emissions from feedstock utilization, transportation, and operational phases. It highlights critical emission hotspots, such as membrane production in PEM and thermal energy requirements in SOE, and underscores the importance of renewable energy integration to mitigate environmental impacts. However, the study also reveals systemic challenges shared across all processes, including significant methane emissions (ranging from 16.1 to 17.45 kg-CH_4/kg-H_2) and indirect emissions tied to electricity generation. These findings stress the necessity of optimizing material selection, improving operational efficiencies, and transitioning to cleaner energy grids to enhance the sustainability of H_2 production.

Consequently, a key advantage of this study is its holistic approach to assessing the environmental impacts of H_2 production methods, integrating lifecycle emissions from feedstock utilization to operational phases using the GREET model. This comprehensive methodology allows for a detailed comparison of the processes. Additionally, its inclusion of a broad spectrum of atmospheric emissions, such as CO_2, CH_4, and PM, enhances its relevance and applicability for sustainability assessments. However, the study also has limitations such as while it evaluates environmental parameters comprehensively, it does not delve deeply into the economic viability or scalability challenges of the processes, particularly for emerging technologies like SOE. Moreover, the dependency on renewable energy sources for achieving optimal performance, while crucial, introduces variability that may not be practical in regions with limited access to clean energy. Nevertheless, this study provides a robust framework for identifying both the advantages and limitations of water-based H_2 production methods, offering valuable insights for policymakers and industry stakeholders aiming to achieve long-term environmental and energy sustainability.

Moreover, based on the current findings, future research should address both the technological and methodological gaps identified in this study to advance the sustainability of H_2 production. While the GREET software provides a robust framework for lifecycle analysis, certain limitations must be acknowledged. For instance, the model's reliance on standardized datasets may overlook regional variations in energy grids, feedstock availability, and material production methods, leading to discrepancies in real-world applicability. Future studies should aim to integrate region-specific data and incorporate more detailed assessments of upstream emissions. Additionally, the analysis would benefit from including more comprehensive economic evaluations. These could encompass the capital and operational costs of electrolysis technologies, including material sourcing, system maintenance, and infrastructure development, as well as long-term financial projections under various energy market scenarios. Moreover, the high dependence on renewable energy sources for achieving optimal emissions reductions highlights the need to evaluate the feasibility and cost of renewable energy integration across different regions. Future work should also explore the potential of coupling these electrolysis systems with carbon capture and utilization (CCU) technologies to mitigate methane and CO_2 emissions further and achieve net-negative emissions. Hybrid systems, combining the strengths of AE, PEM, and SOE, represent another promising area for exploration, potentially balancing the low emissions of AE with the efficiency and operational flexibility of PEM. Finally, expanding the scope of the environmental impact assessment to include social and economic dimensions, such as supply chain resilience, and public acceptance, will provide a more holistic understanding of the role of H_2 production in sustainable energy transitions. By addressing these aspects, future research can build on the solid foundation laid by this study to facilitate the practical implementation of environmentally and economically sustainable H_2 technologies.

4. Conclusions

This study offers a detailed environmental evaluation of AE, SOE, and PEM electrolysis processes for H_2 production, highlighting significant variations in their sustainability profiles. AE emerged as the most environmentally favorable method, with CO_2 emissions of 3.28 kg-CO_2/kg-H_2, methane emissions of 16.1 kg-CH_4/kg-H_2, and the lowest $PM_{2.5}$ emissions of 1.24 kg-$PM_{2.5}$/kg-H_2. These results confirm the potential of the AE process as a low-impact, renewable-compatible H_2 production process. On the other hand, PEM exhibited the highest CO_2 emissions at 4.68 kg-CO_2/kg-H_2 and the most significant methane emissions (17.45 kg-CH_4/kg-H_2), largely due to its reliance on energy-intensive membrane production and the use of precious metals. SOE, while showing some promise with reduced electrical energy demands, recorded CO_2 emissions of 4.28 kg-CO_2/kg-H_2 and the highest acidification potential at 3.66 kg-SO_2eq./kg-H_2, attributed to its high operational temperatures and material requirements. Notably, all systems demonstrated similar biogenic CO_2 emissions, ranging from -0.088 to -0.108 kg-CO_2/kg-H_2, indicating their potential for carbon-negative operations when integrated with renewable energy and carbon capture technologies.

Overall, these findings highlight the urgent need for innovation in material design, particularly for PEM and SOE, and the broader adoption of renewable energy grids to reduce lifecycle emissions. Future research should prioritize economic feasibility studies, hybrid system designs that combine the strengths of different electrolysis methods, and strategies to mitigate indirect emissions, such as methane and acidification precursors. By addressing these challenges, H_2 production can evolve into a cornerstone technology for achieving global energy sustainability and decarbonization goals.

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