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# Multi-criteria Comparative Review of Renewable Energy Driven Water-Splitting Hydrogen Production Technologies

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## Abstract

Hydrogen is key for zero emission scenarios and is producible using renewable components of water, plants, light, and renewable electricity and heat. Water-splitting Green or Clean Hydrogen Production (GCHP) technologies include electrolysis, photolysis, bio-photolysis, and thermolysis at different levels of [technology readiness and commercialization](#). They face challenges and also provide opportunities for GCHP. These technologies are reviewed and compared from energy, exergy, economic, environmental, technological, and social viewpoints (4E-TS). Each technology is studied specifically, and it is also compared with its counterparts. Opportunities and challenges of each technology are highlighted and important guidelines are also given to improve the market and [Social Acceptance](#) of hydrogen. While technological development and cost competitiveness are prerequisites for commercializing water-splitting technologies, several other factors greatly impact the uptake of the technologies. These factors include the availability and affordability of renewable energy and sustainable feedstocks, as well as existing electricity and hydrogen infrastructure, the [Capacity factor](#) of renewable electricity and heat, the cost of renewable electricity and heat, integrating different technologies, and the impact of off-grid and on-grid scenarios, and the existing energy networks. Furthermore, the uptake of GCHP technologies require proper governance, policies, and regulations to stimulate demand for clean hydrogen.

## Keywords

Electrolysis; Photolysis; Thermolysis; Bio-photolysis; Green hydrogen; Water-splitting; Renewable energy

## Nomenclature

| Symbols                         | Definition                                      | Symbols                            | Definition                                                     |
|---------------------------------|-------------------------------------------------|------------------------------------|----------------------------------------------------------------|
| ACME                            | American Company that Manufactures Everything   | $\dot{m}(kg.s^{-1})$               | Mass flow rate                                                 |
| AE                              | Alkaline electrolyzer                           | $\dot{n}_{H_2}(mol.s^{-1}.m^{-2})$ | Hydrogen production rate per unit area of photoelectrode       |
| AEME                            | Anion exchange membrane electrolyzer            | $N_i(-)$                           | Number of end-users experienced power disruption.              |
| $AVC_{CO_2}(USD.kg_{CO_2})$     | Avoided cost of CO <sub>2</sub>                 | $N_t(-)$                           | Total number of end-users receiving electricity from a utility |
| BP                              | Bio-photolysis                                  | OER                                | Oxygen evolution reaction                                      |
| CAPEX                           | Capital Expenditure                             | OPEX                               | Operational expenditure                                        |
| CCS                             | Carbon capturing and storage                    | P(W)                               | Power                                                          |
| CCUS                            | Carbon capturing, utilization, and storage      | PCCE                               | Proton-conducting ceramic electrolyzer                         |
| CF(%)                           | Capacity factor                                 | PDC                                | Power dish collector                                           |
| CGE                             | Carbon gasification efficiency                  | PEC                                | Photoelectrochemical                                           |
| CHP                             | Clean hydrogen production                       | PEME                               | Proton exchange membrane electrolyzer                          |
| CHPS                            | Clean hydrogen production standard              | PLQY                               | Photoluminescence quantum yield                                |
| DBP                             | Direct bio-photolysis                           | QGHPS                              | Quasi-green hydrogen production system                         |
| $d_t(min)$                      | Restoration time                                | PTC                                | Parabolic trough collector                                     |
| E(J)                            | Energy                                          | $P_t(W.m^{-2})$                    | Power density of illumination in photolysis                    |
| $\dot{E}(W)$                    | Energy rate                                     | PV                                 | Photovoltaic                                                   |
| $Em(kg_{CO_2}.kg_{H_2}^{-1})$   | Emission                                        | PV-E                               | Photovoltaic based electrolyzer                                |
| ESS                             | Energy storage system                           | RES                                | Renewable energy source                                        |
| FDP                             | Fossil fuel depletion potential                 | $S(h.year^{-1})$                   | Scheduled downtime                                             |
| $F_R(\%)$                       | Renewable fraction                              | $SAIDI(min)$                       | System average interruption duration index                     |
| Ge-E                            | Geothermal power-based electrolyzer             | $SAIFI(min)$                       | System average interruption frequency index                    |
| GEIF                            | Grid energy interaction factor                  | SCWG                               | Supercritical water gasification                               |
| GHP                             | Green hydrogen production                       | SOE                                | Solid oxide electrolyzer                                       |
| GHPS                            | Green hydrogen production system                | SOFC                               | Solid oxide fuel cell                                          |
| GHS                             | Green hydrogen standard                         | SPT                                | Solar power tower                                              |
| GWP                             | Global warming potential                        | $T(h.year^{-1})$                   | Time planned for operation                                     |
| HER                             | Hydrogen evolution reaction                     | TRL                                | Technology readiness level                                     |
| HGE                             | Hydrogen gasification efficiency                | TWS                                | Thermochemical water-splitting                                 |
| HP                              | Hydrogen production                             | TWSC                               | Thermochemical water-splitting cycles                          |
| HPS                             | Hydrogen production system                      | $U(h.year^{-1})$                   | Unscheduled downtime                                           |
| $HV(W.kg^{-1})$ or $(W.m^{-3})$ | Heating value                                   | UF                                 | Utilization factor                                             |
| Hy-E                            | Hydropower without reservoir-based electrolyzer | $\dot{V}(m^3.s^{-1})$              | Volume flow rate                                               |
| HyR-E                           | Hydropower with reservoir-based electrolyzer    | WACC                               | Weighted average cost of capital                               |
| GCHPSs                          | Green, and clean hydrogen production systems    | PEME                               | Proton exchange membrane electrolyzer                          |
| GTD                             | Generation-to-demand ratio                      | PLQY                               | Photoluminescence quantum yield                                |
| IBP                             | Indirect bio-photolysis                         | QGHPS                              | Quasi-green hydrogen production system                         |
| kh                              | Kilo hours                                      | PTC                                | Parabolic trough collector                                     |
| $LC(USD.product\ unit^{-1})$    | Levelized cost                                  | WDP                                | Water depletion potential                                      |
| $LCI(impact\ unit.kgH_2^{-1})$  | Life Cycle Impact                               | Wi-E                               | Wind power-based electrolyzer                                  |
| $LCOH(USD.kgH_2^{-1})$          | Levelized cost of hydrogen                      | WPV                                | Wind photovoltaic                                              |
| $LCOH\&E(USD.kgH_2^{-1})$       | Levelized cost of hydrogen and emission         | WST                                | Water splitting technology                                     |

|        |                                 |                                         |                                              |
|--------|---------------------------------|-----------------------------------------|----------------------------------------------|
| LFR    | Linear Fresnel reflector        | WSHP                                    | water-splitting hydrogen production          |
| LOHC   | Liquid organic hydrogen carrier | Subscripts and Greeks                   |                                              |
| LR (%) | Learning rate                   | nom                                     | Nominal                                      |
| LSC    | Luminescent solar concentrators | ele                                     | Electrolysis                                 |
| MAP    | Microwave-assisted pyrolysis    | $\eta(\%)$                              | Energy efficiency                            |
| MCFC   | Molten carbonate fuel cell      | $P(\mu\text{mol.g}^{-1}.\text{h}^{-1})$ | Performance of photocatalyst                 |
| MDP    | Mineral depletion potential     | $\psi(\%)$                              | Exergy efficiency                            |
| MLE    | Membrane-less electrolyzer      | $\epsilon(\%)$                          | Energy consumption ratio of the electrolyzer |

# 1- Introduction

## 1-1- Background

Transitioning to renewable and non/low-pollutant energies seems to be the concluding solution for energy-related concerns such as depleting fossil fuel resources, global warming, and air/water/soil pollution. However, renewable energies such as solar, wind, hydro, and geothermal are inherently changeable and their storage faces economic, technical, and environmental challenges. Using changeable renewable energies without backup or storage for providing the electricity demand of a region with fluctuating load profiles is highly unreliable. Although the simultaneous use of several renewable energies may improve the [availability](#) and [reliability](#) of the power system, the uncertainty and unpredictability of the future make it not a definitive solution [1]. D. Tong et al. [1] assessed the reliability of using a solar-wind mix with and without storage for 42 countries. In the best case with 12 h of storage the maximum achievable reliability was 94 %, while in industrialized countries, reliability is more than 99.95% [2] which is a big gap.

A. J. Harker Steele et al. [3] empirically investigated the impact of using diverse renewable energies on the reliability of power systems. Hence, there is still a need to develop and diversify Energy Storage Systems (ESSs) as well as renewable power generators. The main reason for renewable power generators' low reliability is their smaller [capacity factor](#). In 2020, the average capacity factor of power generators fed by solar, wind, coal, hydropower, natural gas, geothermal, and nuclear energies was 24.9%, 35.4%, 40.2%, 41.5%, 56.6%, 74.3%, and 92.5%, respectively [4]. While having highly diverse renewable energy sources in the power generation portfolio increases the reliability of the power system, due to climatic conditions, weather conditions, geography, technology development, economic situation, etc., we may not have access to diverse Renewable Energy Sources (RESs) . In such conditions, the lack of diversity can be compensated by using complementary power generators, converting and/or storing the extra renewable power. Al-Ghussain et al. [5] investigated combining solar photovoltaic, wind, and biomass sources with hybrid energy storage. They concluded that using biomass to provide the baseload increased reliability. The system's [Renewable Fraction](#) ( $F_R$ ) and [Demand–Supply Fraction](#) (DSF) reached 99.59%, and 98.86% respectively.

Some analysts predict that the surpluses of renewable electricity will be so excessive that it will be necessary to convert the electricity into hydrogen and then into synthetic hydrocarbons [6]. Over the centuries the tendency to use fuels with fewer carbon atoms resulted in replacing wood with coal, and coal with natural gas [7]. However, natural gas is still a problem and results in  $CO_2$  production. Hydrogen can be used as a fuel directly or indirectly while producing zero emissions. In other words, hydrogen is a key to decarbonization. Hydrogen can be directly used in fuel cells, converted to methane, ammonia, or other chemical derivatives, and used as fuel indirectly. For instance, the European Maritime Safety Agency (EMSA) identified the potential of ammonia as a long-term fuel with zero or near zero carbon emission in shipping and deep-sea cargo ships regardless of the hydrogen origin [8].

Hydrogen is essential for sustainability, it is one of the unique options with enough storage capacity, it provides seasonal storage, it is zero emission, and provides feedstock to many products [9]. Hydrogen is also utilized as a buffer to increase resilience of power systems or feedstock using captured carbon for Methanol, Urea, or Methane production [10]. ACME Group and Hydrogenious LOHC Technologies have decided to cooperatively investigate the development of large-scale hydrogen supply chains from Oman to Europe. This corporation will possibly expand to assess the hydrogen value chain from the USA to Europe [11]. Gasunie, a gas network operator in the Netherlands and northern Germany, evaluates the viability of an offshore hydrogen network in the Dutch North Sea to provide carbon-free energy for more than half of Europe by 2050 [12]. Advent Technologies integrates its 50 kW high-temperature PEM fuel cell modules with Siemens Energy's automation and electrification methods for electric and hybrid ships. The first module is scheduled to be accomplished in 2026 [13]. Anasol, a German-Spanish renewable hydrogen developer, has plans to construct an 800 MW factory to convert green hydrogen to green methanol by 2029 ([14] and [15]). In Denmark, Stiesdal Hydrogen and Danfoss Heat Exchangers have settled to introduce an electrolyzer to the global market [16]. Among the pioneer countries, France, Poland, Germany, USA, Swiss, Australia, England, Norway, Switzerland, Slovenia, Canada, China, Saudi Arabia, Kuwait, and Oman can be mentioned ([17], [18], [19], [20], [21], [22], [23], [24]).

## **1-2- Review of the review articles**

Considering the importance of green hydrogen for the energy transition, and decarbonization, it has attracted the attention of researchers, investors, industrialists, and energy providers for different sectors. Production of reliable green hydrogen at a competitive price compared to non-green hydrogen will be the most important challenge in utilizing green hydrogen in different sectors, particularly in power.

Review papers are a reliable source of data about the trends, advances and challenges of GHPSs. In this section, the most recent review articles published from 2020 to 2024 on WSHPs were studied in search of the trends, recommendations, identified knowledge gaps, forecasts, and challenges. Table 1 presents different performance indicators of WSHPs, and their definitions are presented in the Appendix. Review articles focusing on WSHP methods are scarce, which makes it hard to provide a comprehensive comparison between the WSHP methods. The main scope of some utilized review articles may not be WSHP, but they included important data on WSHP, hence they are used in this review. In addition, it can be seen that none of the reviews have addressed all aspects of WSHP methods simultaneously. This is partially due to the fundamental differences between the WSHP systems that make the full comparison hard. To draw a full picture of the current and future status of WSHP methods, they must be evaluated from energy, exergy, economic, environmental, technological, and social viewpoints (4E-TS). This review has provided a comprehensive collection of the 4E-TS performance indicators of the WSHP methods in one place. It can be used to draw a full picture of the WSHP methods, to make recommendations, and to identify knowledge gaps, challenges, and potentials of each WSHP method. Such comparative review is missing among the review articles. In the following, the “Review Articles” are reviewed and compared in Table 1.

Different GHPSs, their applications and innovations were reviewed by C. Anand et al. [25]. They briefly explained hydrogen types and production methods, and exclusively discussed how seawater and wastewater can be considered as a source for hydrogen production. M. S. Herdem et al. [26] presented a review of GHP systems that use solar and wind power by focusing on trends and standardization. They finally proposed performance indicators including the [Conversion Efficiency of RES to hydrogen](#), the [Exergy Efficiency](#) of the GHPS, the [Utilization Factor](#) of GHPS, the Grid Energy Interaction Factor ([GEIF](#)), the Levelized Cost of Hydrogen ([LCOH](#)), the Levelized Cost of Hydrogen and Emission ([LCOH&E](#)) [26]. Li X. et al. [27]. reviewed the thermochemical water splitting cycles (TWSCs) or thermolysis cycles that use solar heat and Thermal Energy Storage (TES) technologies for hydrogen production. The main TWSCs were classified as sulfur family, chlorine family, and metal oxide family. The operational temperature ranges of processes were classified as  $\leq 200\text{ }^{\circ}\text{C}$ ,  $200\text{-}500\text{ }^{\circ}\text{C}$ ,  $500\text{-}1000\text{ }^{\circ}\text{C}$ , and  $\geq 1000^{\circ}\text{C}$ . Correspondingly, they identified which process is endothermic, exothermic, or needs electricity as well (Hybrid-TWSCs). The concentrated solar systems with their corresponding capacity ranges integrated with TWSC include Parabolic Trough Collector (10-250 MW), Solar Tower (10-150 MW), Linear Fresnel Reflector (5-250 MW), and Power Dish Collector (0.01-1 MW). They also classified the heat storage systems according to their operating temperature range and their type as sensible heat (solid, liquid, and gaseous), latent heat (organic, inorganic, and eutectic), and thermochemical storage. They finally concluded that Copper-Chlorine (Cu-Cl) and

Sulfur-Iodine (S-I) cycles are the most suitable for solar heat-based hydrogen production. Hydrogen production is highly energy intensive and consumes about  $5 \text{ kWh} \cdot \text{m}^{-3}$  at  $80^\circ\text{C}$ . Furthermore, any accidental mixing of hydrogen with oxygen may cause an explosion [28]. Y. Chen, and G. Z. Chen [28] provided a critical review seeking the recent developments and alternate water electrolysis to solve these two challenges. They concluded that the explosion problem can be solved by decoupling hydrogen and oxygen evolution reactions in space or time. They reviewed the alternate water thermolysis and water electrolysis using liquid or solid redox intermediate and the alternate half-electrolysis of water. The solid redox intermediate and the alternate half-electrolysis methods omit the membrane. However, only the half-electrolysis option can reduce energy consumption without risking the hydrogen production rate and operation [28]. Hydrogen production by using different photocatalysts is overviewed by K. A. Sanoja-López et al. [29]. They aimed to collect the latest works on progress in photocatalysts for hydrogen production, with specific attention to photocatalyst type, [hydrogen production performance](#), and market trends. They concluded that although photocatalysts are under development their multi-capability to produce hydrogen, degrade emissions, and inhibit microorganisms can make the photolysis cost-effective. In addition, they predicted that photocatalytic hydrogen production will become an essential part of sustainable energy methods, for a cleaner environment in the future [29].

The electrolyzer technologies including PEM, SOE, AEME, and Alkaline were reviewed and their technical, economic, and environmental specifications and limitations were studied by S. S. Kumar and H. Lim [30]. They also collected information about the leading manufacturers of electrolyzers such as Nel, Cummins, McPhy, ITM Power, Plug Power, Elogen, and Green Hydrogen Systems with the technical data of the electrolyzers. This review [30] offers upcoming research instructions and a roadmap for the development and implementation of feasible GHP projects. They also provided some recommendations for future research such as reducing the membrane thickness of alkaline electrolyzers from  $460 \mu\text{m}$  to  $50 \mu\text{m}$  to reduce energy consumption and increase efficiency from 53% to 75% at  $1 \frac{\text{A}}{\text{cm}^2}$ . Another main challenge of Alkaline electrolyzers is their low current density. The main challenge of AEME is membrane durability, which needs to improve. For the PEM the main challenges are membrane cost and durability, electrocatalyst materials used, and stackability. The SOE, which is still under development its durability can be improved by increasing the conductivity of electrolytes and optimizing mechanical and chemical stability [30]. Another review on hybrid-TWSC was done by J. E. Lee et al. [31]. Hybrid-TWSC needs heat and electricity to convert water to hydrogen and oxygen. According to their study, multi-step thermochemical redox reactions are more efficient for water splitting because the maximum required temperature can be lowered. They reported that hybrid-TWSC is being used in large-scale hydrogen production due to their lower

[LCOH](#), furthermore integrating hybrid-TWSC with renewable energies such as solar can decrease the costs and increase the energy efficiency even more. They also briefly compared different HPSs including electrolysis, photoelectrochemical, reforming of hydrocarbons, plasma reforming, biomass gasifications, and thermochemical in terms of characteristics and processes. Their focus was mostly on comparing 2-, 3- and 4-step TWSCs ,and among them, the four-step TWSC by adding the recombination reaction has lowered the operating temperature. The four steps include hydrolysis of the chemical compound, Hydrogen Evolution Reaction (HER), Oxygen Evolution Reaction (OER), and recombination of reactants [31] .Their study has shown that amongst numerous metal chlorides, Iron(Fe), Copper(Cu), Cesium(Ce), Magnesium(Mg), and Vanadium (V) chloride, were in effect for the four-step TWSC for hydrogen production [31].

S. Singla et al. [32] reviewed different hydrogen production, separation, and storage technologies. They recommended hydrogen production through solar light, wind power, and water splitting to supply global energy demand. In addition, they concluded that researchers are focusing on direct hydrogen generation by using the photocatalytic method (Photolysis) and sunlight-responsive semiconductor materials. The membrane separation method was proposed due to its selectively permeable, and metal hydrides as the best choice for hydrogen separation and storage. M. Aravindan and K. G. Praveen [33] discussed the economic, environmental, and technical permanence of the WSHPs. The advantages, disadvantages, and limitations of each method are also presented. They have made recommendations to improve [hydrogen production performance](#), [conversion efficiency](#), [energy efficiency](#), and [LCOH](#), as well as policy proposals for the governments to improve investment terms while expanding the hydrogen market. They claimed Solar-based hydrogen production is five times costlier than biomass, three times more expensive than geothermal, and twice as costlier than wind-powered methods. Hydrogen research is still in its infancy due to the obstacles in its production, storage, LCOH, and transportation on a mass scale [34]. J. Jayaprabakar et al. [34] reviewed the effects of nanoparticles such as gold, silver, titanium, and iron on hydrogen production and storage. According to the results, metal nanoparticles achieved well, but other nanoparticle materials need to be investigated for hydrogen production and storage to reduce costs and increase the production rate [34]. Luminescent Solar Concentrators (LSC) photonic devices can use sunlight to produce electricity and hydrogen. Common LSC devices comprise an oxygenless, nitrogen-filled chamber with a high luminescent material that absorbs light and then reemits it with a longer wavelength depending on the luminescent material. The LSC devices are categorized into four main types of organic dyes, quantum dots, carbon dots, and rare-earth complexes [35]. Although the [conversion efficiency](#) of LSC-assisted electrolyzers is significantly lower (0.22%-1.5%) than polycrystalline silicon (17%), crystalline silicon (15.1%), and multi-

crystalline silicon solar (14.2%) the authors of [35] believe that LSC has a great potential for increasing its conversion efficiency. B. S. Zainal et al. [36] reviewed recent advances in GHP technologies globally. They discussed hydrogen color codes and reported advances in WSHPs and emphasized that electrolysis is the most mature technology for hydrogen production using RES. The green hydrogen cost is reducing rapidly, but the LCOH by RES is still high and can not be used in the market when it is supposed to compete with non-renewable-based hydrogen. Hydrogen production by water-splitting is also challenging for countries such as Pakistan and Turkey, which are already under water stress, because each kg of hydrogen production by electrolysis consumes 9 kg of water [36]. Non-noble metal-based photocathodes have attracted attention. Using non-noble metal-based photocathodes instead of costly noble-based photocathodes using titanium or gold can reduce the LCOH and scale-up of hydrogen production by photolysis [37]. M. Ji and J. Wang [38] reviewed the general renewable and nonrenewable-based HPSs based on their economic situation and environmental impacts. Among GHPSs, water-splitting by electrolysis is now the most viable for application, and PEME technology with more flexibility is taking the place of AE [38]. J. Webb et al. [39] compared the hydrogen market for producing blue and green hydrogen. They used the [learning rate \(LR\)](#) of different technologies, LCOH, transport cost of hydrogen in different scenarios, and [CO<sub>2</sub> avoidance cost](#). They also discussed the impact of electricity costs on the LCOH. The trend of LCOH, the LC of utility-scale wind and PV farms from 2015 to 2050 was discussed.

While hydrogen production must be acceptable from the energy, exergy, economic, environmental, and technological viewpoints, hydrogen itself and related technologies must also be socially accepted for a successful transition [40]. N. V. Emodi et al. systematically reviewed the [Societal Acceptance](#) of hydrogen technologies. They concluded that more than 60% of the countries analyzed in their investigation, had low awareness of hydrogen technology. In addition, despite the high acceptance of hydrogen technology, a gap between general and local acceptance of hydrogen technology was recognized, mostly due to the safety concerns [41]. The public perception on hydrogen infrastructure to be used for fuel cell vehicles in Japan showed that people have become a little more positive about hydrogen infrastructure but more cautious about the risks and benefits [42]. In another research E. B. Ates and E. Calik investigated the public awareness of hydrogen energy. They used a statistical approach, and the results showed that individual awareness changes depending on their knowledge level about hydrogen, educational level, and age. As a result, stakeholders, and policymakers must consider these correlations to construct the frameworks of policy [43]. The number of social acceptance investigations is scarce, and needs to design policies to improve public preception and awareness about green hydrogen benefits.

According to the study of review articles, the review articles that specifically work only on WSHPs are scarce. Therefore, this article considers only WSHPs. Biomass-based hydrogen production technologies are not included. The limited number of review articles published on WSHPs have examined this topic from a few limited and mostly economic aspects. In the present review, articles published between 2020 and 2024 were studied, and the extracted results were compared based on exergy, environmental, economic, technological and social criteria (4E-TS). It is worth mentioning that each criterion has some sub-criteria. For each technology, the challenges and opportunities ahead are stated, as well as areas that still require research, and investment. According to the extensive study of the sources, none of the published review articles have comprehensively compared the WSHPs by 4E-TS framework, which is proposed in this work.

**Table 1. Comparison of the review articles on GHP systems from RES used, parameters used in the studies, novelties and highlights**

| Ref. | WSHPs reviewed                      | RES used in HPSs                                        | Parameters used to compare HPSs                                                                                                       | Performance indicators HPSs                                                                                                                                                                                             | Selected recommendations /novelties/highlights                                                                                                                                                           |
|------|-------------------------------------|---------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| [26] | Electrolysis                        | Wind and solar-PV                                       | Power Source, Location of the System, Grid Connection, Hydrogen and Electricity Storage, Final utilization of hydrogen, Research type | <a href="#">Conversion efficiency</a> , <a href="#">Exergy efficiency</a> , <a href="#">UF</a> , <a href="#">GEIF</a> , <a href="#">LCOH</a> , <a href="#">LCOH&amp;E</a> , <a href="#">Avoided CO<sub>2</sub> cost</a> | Standardization of solar and wind-based GHP systems. Solar and wind-based GHPs are compared. More attention is needed on transient electrolyzer behavior, battery degradation, and grid stability.       |
| [27] | Thermolysis or TWSC                 | Concentrated solar heat by using PTC, SPT, LFR, and PDC | CSP technology, Non-solar (external) energy sources used, Electricity source<br>Heat recovery, TES                                    | <a href="#">Energy efficiency</a> , <a href="#">Overall exergy efficiency</a> ,<br>Economic, System composition, Multi-scenario applications                                                                            | TWSCs are more talented to be combined with solar thermal technologies. Copper chloride (Cu-Cl) and Selenium Iodine (Se-I) cycles are potentates for solar heat-driven hydrogen production systems.      |
| [28] | Electrolysis<br>Thermolysis or TWSC | Solar heat<br>Solar-PV                                  | Redox mediums, Oxygen evolution reaction electrode<br>The current density of the electrolyzer, State of charge                        | <a href="#">Ideal Energy consumption ratio</a> , <a href="#">Practical energy consumption ratio</a><br>Conversion efficiency of HER, and OER.                                                                           | The half-electrolysis of water can save 14% of the energy input. Alternate thermolysis and electrolysis of water with a metal oxide anode is possibly feasible in the industry.                          |
| [29] | Photolysis                          | Solar radiation                                         | The photocatalyst materials used                                                                                                      | <a href="#">Hydrogen production performance</a>                                                                                                                                                                         | Further studies on a larger scale and evaluate the effect of working with larger volumes . Photocatalysis can produce hydrogen, inhibit microorganisms, and degrade different pollutants simultaneously. |

|      |                                                                                                             |                                                     |                                                                                                                        |                                                                                                                                                                         |                                                                                                                                                                                                                                                                        |
|------|-------------------------------------------------------------------------------------------------------------|-----------------------------------------------------|------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| [30] | Electrolysis                                                                                                | Wind and solar-PV                                   | Type of electrolyzer including PEM, AEM, SO, and Alkaline.<br>Electrode materials used.                                | <a href="#">Energy efficiency of electrolyzer</a><br>Capital cost for minimum capacities of 1 MW and 10 MW<br>H <sub>2</sub> purity<br>Lifetime                         | Improving durability and stackability and reducing membrane thickness were recommended to improve performance and reduce costs. GHP from wind and solar using water electrolysis is likely the main key for the energy transition to solve the NZE challenges.         |
| [31] | Major:<br>Hybrid-TWSC<br>Minor:                                                                             | Solar-PV, Solar heat,                               | Energy source<br>Number of TWSC steps,<br>Chemical agents used (metal oxides, metal halide, and sulfur oxides)<br>HPSs | H <sub>2</sub> production rate (mol/s)<br><a href="#">Energy Efficiency LCOH</a><br>The maximum heating temperature of TWSC                                             | The TWSCs utilizing redox potentials of metal oxide pairs and solar energy increase hydrogen yield. More studies on efficient hybrid systems will be needed for GHP. TWSCs should be investigated to apply to utility-scale.                                           |
| [32] | Photolysis<br>Bio-photolysis<br>Electrolysis Photocatalytic<br>Photo-catalytic<br>Solar-thermolysis<br>TWSC | Solar light<br>Solar heat<br>Solar PV<br>Wind power | Material used for electrodes and catalysts,<br>Operating temperature of processes<br>Energy sources<br>Light source    | <a href="#">LCOH</a><br><a href="#">Hydrogen production performance (Activity)</a>                                                                                      | Focusing on GHPs, and developing low-cost efficient separation, purification, and storage systems for hydrogen are recommended. The development of high-efficiency sunlight-responsive photocatalysts is vital. Less-oxygen-sensitive organisms need to be engineered. |
| [33] | Electrolysis<br>Thermolysis<br>Photolysis<br>Thermochemical                                                 | Solar PV<br>Wind<br>Geothermal                      | HPSs<br>Energy source<br>Location of the study and H <sub>2</sub> plants and their operational status                  | <a href="#">LCOH</a><br>Hydrogen production rate (MW/day)<br><a href="#">Energy efficiency</a>                                                                          | Improving hydrogen supply chain is vital. Increasing the <a href="#">overall efficiencies</a> , commercialization of GHPs, market scale-up, feasible product expansion, infrastructure, and the market sustainability are highly recommended.                          |
| [34] | Electrolyzer<br>Photo-catalytic                                                                             | Solar-PV<br>Wind power<br>Hydropower                | The Nano-particle materials (gold, Silver, Iron, and Titanium oxide).<br>Production method of Nano-particles           | Hydrogen production<br>Hydrogen concentration.                                                                                                                          | The impact of using nano-particles on hydrogen production by using different GHPs is studied. Also, hydrogen storage in the solid state using Nano-particle is studied and they have shown a good potential for increasing production rate and storage capacity.       |
| [35] | Electrolysis                                                                                                | Luminescent solar concentrators (LSC) assisted, PV  | Different types of luminescent including organic dyes, quantum dots, carbon dots, and rare-earth complexes             | <a href="#">Conversion efficiency</a><br><a href="#">Energy efficiency (power to hydrogen conversion)</a><br><a href="#">PLQY</a><br><a href="#">Optical efficiency</a> | Increasing LSC efficiency by developing new materials and optimized designs. Light management improvement in LSCs, reducing losses, and maximizing catalyst efficiency for GHP are vital.                                                                              |
| [36] | Electrolysis (SOE, PEME, AEME, AE)<br>Photolysis, bio-photolysis<br>Thermolysis                             | Solar PV/thermal<br>Geothermal heat<br>Tidal power  | The energy source<br>The feedstock<br>Process requirements                                                             | <a href="#">LCOH</a><br>Hydrogen production<br>Hydrogen concentration.                                                                                                  | Green hydrogen is still expensive. Water splitting by electrolysis is the most mature GHP. The main challenges of GHP are high LCOH, extensive                                                                                                                         |

|      |                                                                                                          |                                  |                                                                                                                                                                                    |                                                                                                                                                                                                                                                         |                                                                                                                                                                                                                                                                                       |
|------|----------------------------------------------------------------------------------------------------------|----------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
|      | Biocatalysed electrolysis                                                                                |                                  | Hydrogen investment in different countries                                                                                                                                         |                                                                                                                                                                                                                                                         | infrastructure development, alternating hydrogen production due to dependence on RES, market growth, and international collaboration.                                                                                                                                                 |
| [37] | Photoelectrochemical (PEC) water splitting                                                               | Sunlight                         | Different non-noble metal-based photocathodes<br>Light source used<br>The preparation method of the photocathodes<br>Electrolyte type                                              | <a href="#">Conversion efficiency</a><br><a href="#">Stability of photocathode</a><br><a href="#">Applied bias photon to conversion efficiency (ABPE)</a> ,<br><a href="#">Incident photocurrent conversion efficiency (IPCE)</a> , <a href="#">STH</a> | Recent advancements in developing non-noble metal-based photocathodes and their efficiency in the HER during PEC water splitting are briefly reported. The progress in using non-noble metal-based photocathodes reduces the cost of GHP compared to noble metal-based photocathodes. |
| [38] | Direct and indirect bio photolysis, photo fermentation, Electrolysis, TWSCs (two- to five-step reaction) | Solar PV/Heat<br>Wind<br>Nuclear | Different GHPSS<br>The energy source<br>GHP method<br>Economic and<br>Environmental impacts<br>Chemical reactions                                                                  | <a href="#">LCOH</a><br><a href="#">Global warming potential (GWP)</a><br><a href="#">Acidification potential (AP)</a>                                                                                                                                  | GHP is continually being improved technologically. Among GHPSS, water-splitting by electrolysis is now the most viable for application, and PEME technology with more flexibility is taking the place of AE.                                                                          |
| [44] | Electrolysis<br>Thermochemical<br>Plasma arc decomposition<br>Coal-assisted Water<br>Electrolysis        | Solar-heat<br>Solar-PV           | The HPS and feedstock.<br>Temperature, pressure,<br>Products, Energy sources,<br>Maturity of technology,<br>Production scale, and cost,<br>Capital cost, and<br>bottlenecks of HPS | <a href="#">LCOH</a><br><a href="#">Technology Readiness Level (TRL)</a> , <a href="#">Efficiency</a> ,<br>Hydrogen Yield,<br><a href="#">Capital cost (CAPEX)</a> .                                                                                    | In the transition to a hydrogen economy, the invention of carbon-based byproducts or hydrolysis using electricity from renewable resources should be adopted to improve efficiency and reduce the emission of fossil fuel-based HPSs.                                                 |
| [39] | Electrolysis for GHP                                                                                     | Solar<br>Wind                    | GHPSS are compared economically and environmentally with blue hydrogen. Hydrogen transportation scenarios, Trend of LCOH and CAPEX of electrolyzers                                | <a href="#">LCOH</a><br><a href="#">Learning rate</a><br><a href="#">Avoided cost of CO<sub>2</sub></a> ,<br><a href="#">CAPEX</a><br>Transport cost of H <sub>2</sub> ,<br><a href="#">Emission abatement ratio (EAR)</a> .                            | The major parameters impacting LCOH are electricity feedstock cost, capital cost, and capacity factor. Land, labor, and water are minors. Directing green financing toward the GHP rather than blue hydrogen is recommended.                                                          |

## 2- Materials and methods

The trend of the number of published articles by ScienceDirect having the key expression of “Green Hydrogen” in their title was determined for the last 2 decades and shown in Fig.1. It shows that since 2020, research on green hydrogen has increased dramatically. The data are presented in three graphs by years of publication, by subject area, and the journal name. It shows that more than 93% of the articles have been published in the last 5 years (2020-2024), and energy is the focus of the green hydrogen research. The leading journals for publishing green hydrogen research are the International Journal of Hydrogen Energy,

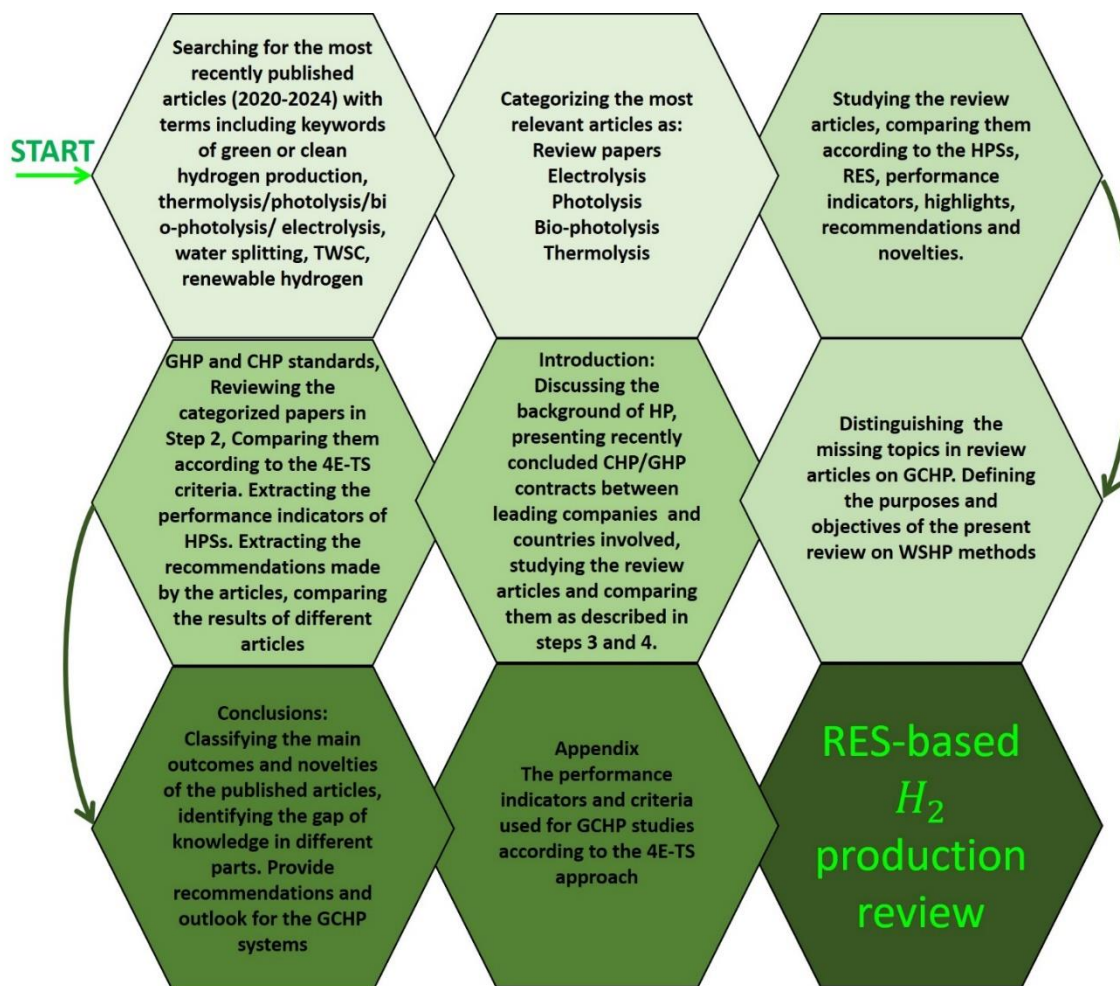
Fuel Cells Bulletin, and Energy Conversion and Management [45]. In addition, the orange column shows the number of review articles published in the same categories. The number of the review articles follows the same pattern during the last 5 years.



**Figure 1: The number of total publications, and review articles with the expression of “Green Hydrogen” in the title during 2004-2024 in different years, areas, and journals by ScienceDirect on March-1-2025 [45]**

The infographic of the material and method used for the present study is shown in Fig.2. To draw a full picture of the current and future status of the HPSs the most recent publications on HPSs based on RES from 2020 to 2024 were searched through the Scopus database by “Green Hydrogen Production”, “Clean” OR “Green”+ “Hydrogen Production Systems”, “Clean” OR “Green”+ “Hydrogen production + water

splitting + electrolysis”, “Clean” OR “Green”+ “Hydrogen production + water splitting+photolysis”, “Clean” OR “Green”+ “Hydrogen production+water splitting + thermolysis”, “Clean” OR “Green”+ “Hydrogen production+water splitting + TWSC”, “Clean” OR “Green”+ “Hydrogen + bio photolysis”. The terms were searched in the title and abstract of the articles.



**Figure 2.** The infographic of the method used for the present review on CHP and GHP systems

First, the “Review Articles” were chosen according to their scope. However, since review articles focusing on WSHPs were scarce, the review articles containing important data on WSHPs were included in the study. The most important data on RES-based WSHP methods were extracted and presented in Table 1. This table compares the review articles according to the WSHP systems, the RES used for hydrogen production, parameters used to compare different HPSs, performance indicators used to evaluate the HPSs, most important recommendations, novelties, and highlights of the articles. According to the outcomes of

the review papers, the missing topics in the review articles were distinguished and the scope of the present review was defined.

Then each category of the articles was studied to extract the main results, novelties, and performance indicators according to the 4E-TS approach and compare them. The technologies were comparatively studied from different viewpoints in a separate section, then the procepects are given. The conclusion comprises the identified trends and most important advances, knowledge gaps requiring more research and investment, and recommended topics for more in-depth research. In the end, the collection of the performance indicators and methods used in the 4E-TS comparative study is presented which can be used as a reference for future studies on RES-based HPSs.

### 3- Hydrogen production standards

According to the Green Hydrogen Organisation Standard (GHS):

*“Green hydrogen is hydrogen produced through the electrolysis of water with 100% or near 100% renewable energy with close to zero greenhouse gas emissions ( $\leq 1$  kg CO<sub>2</sub>e per kg H<sub>2</sub> taken as an average over a 12-month period).”* [46].

While the GHS is very strict about the feedstock, source of energy, and the emission criterion for GHP, the U.S. Department of Energy, Clean Hydrogen Production Standard (CHPS) that is defined under Bipartisan Infrastructure Law (BIL) Section 40315 puts no limit to the feedstock and source of energy. *“The CHPS establishes a target for well-to-gate lifecycle greenhouse gas emissions of  $\leq 4.0$  kgCO<sub>2</sub>e/kgH<sub>2</sub>”, “This target is likely achievable by facilities that achieve  $\leq 2$  kgCO<sub>2</sub>e/kgH<sub>2</sub> at the site of production, which potentially have additional emissions from upstream and/or downstream processes.”* [47].

The present article focuses only on WSHP by RESs and does not include biomass. Therefore, WSHPs other than electrolysis that meet the GHS target are considered **quasi-green hydrogen production systems** (QGHPS), and those satisfying the target defined by CHPS are called clean hydrogen. In this regard, the potential GHPs, QGHPSs, and CHPSs are categorized in the tree diagram shown in Fig. 3. Only water and biomass are considered as renewable feedstocks. WSHPs include electrolysis, thermolysis, biophotolysis, hybrid thermolysis, mechano-catalytic, plasmolysis, and magnetolysis. Different biological and thermochemical processes can do biomass conversion to hydrogen.

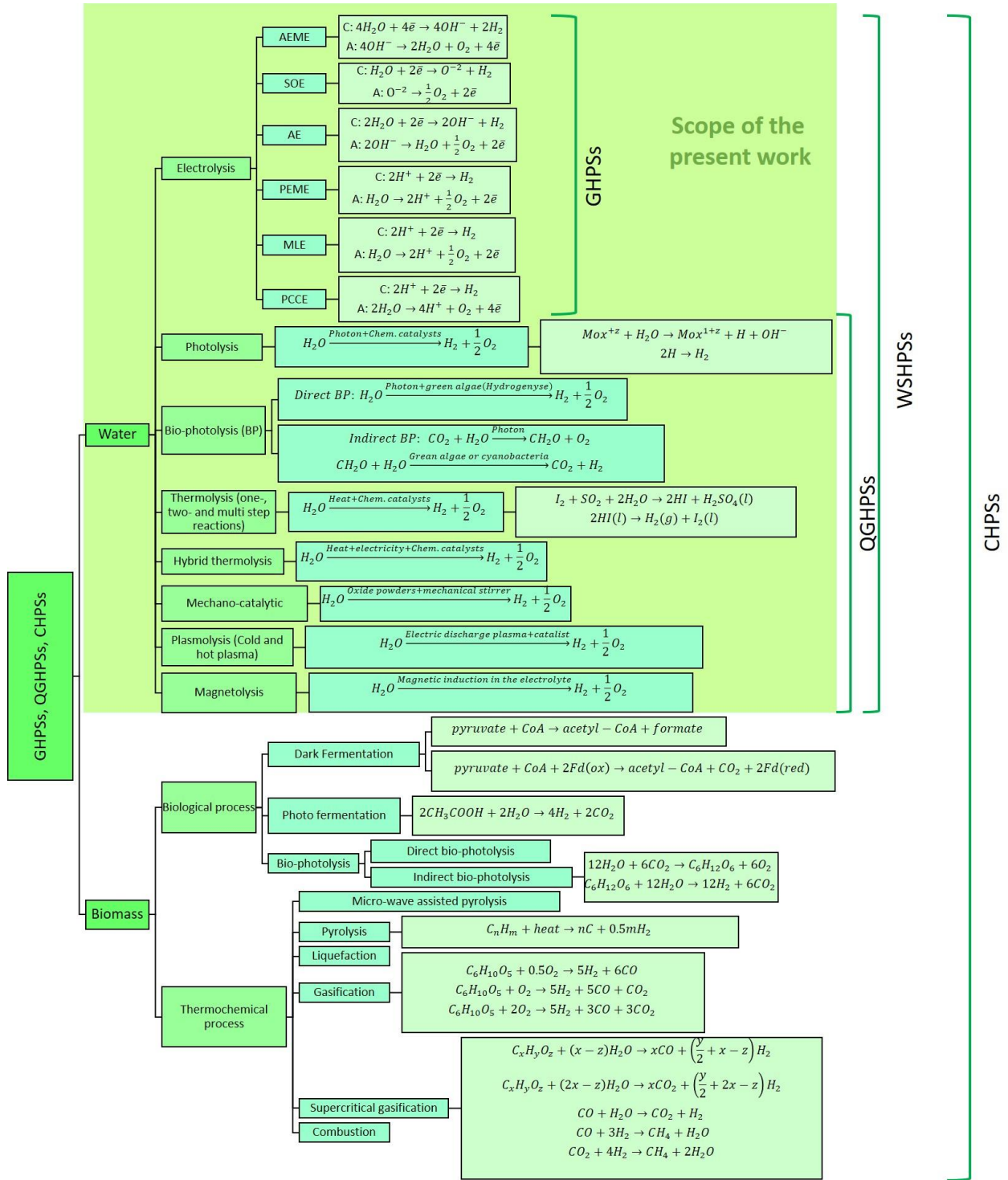


Figure 3. Tree diagram of the potential GHPs, QGHPs, and CHPs ( [31], [48], [32], [33], [36], [38], [49], [44], [50], [51], and [52])

#### 4- Water-splitting hydrogen production

Water-splitting is a chemical reaction to split water molecules into hydrogen and oxygen molecules. This can be done using electricity, heat, light, plasma, thermonuclear reactions, electromagnetic field, or a combination of these energy sources. The main water-splitting methods include electrolysis, TWSC or thermolysis, hybrid-TWSC, photolysis or photoelectrolysis, and bio-photolysis (direct and indirect). There are also miscellaneous WSTs such as Mechano-catalytic [53], water plasmolysis [54], water magnetolysis, magmalysis [55], and water radiolysis [51]. Among the miscellaneous WSTs except for radiolysis and magmalysis, the other methods have the potential to generate green hydrogen [51]. In the following, each of the main technologies is discussed and their potential for GHP is studied.

#### 4-1- Electrolysis

Water electrolysis returns to more than 200 years ago [51] and can be done with various technologies of proton exchange membrane (PEME), pressurized alkaline (AE), solid oxide (SOE), anion exchange membrane (AEME), membrane-less (MLE), and proton-conducting ceramic (PCCE) electrolyzers. The first two technologies (PEME, and AE) with [TRL](#) of 8 to 9 are ready for commercial deployment. SOE and AEME with lower TRLs of 5 to 6 and 2 to 3 have promising performance [56]. The Energy and Climate Change Directorate in a report for the Scottish Government, assessed the electrolyzers and reported a TRL of 9 for PEME, AE, and SOE, while 6 for AEME and MLE. MLE is an alternative to PEME technology that allows operation with changeable renewable electricity without losing too much efficiency [57]. Pressurized PCCE is an alternative to SOE and is another high-operating temperature technology with a TRL of about 3 [58]. IEA in its Tracking Electrolysers report [59] considers the TRL of 9 for PEME and AE (market uptake), 7 to 8 for SOE (demonstration level and approaching market uptake), and 6 for AEME (approaching demonstration level). SOE is commercializing rapidly. In April 2023, a 2.6 MW SOE was installed in a Neste refinery in the Netherlands, and after a short time, a 4 MW SOE was installed in a NASA research center in California [59]. Bloom Energy amplified its SOE manufacturing capacity in 2022 by moving towards GW-scale in the USA [59]. Topsoe is improving its manufacturing facility for a 500 MW/yr industrial-scale SOE in Denmark, for 2025 [59]. AEMEs are at very small-scale production and commercialization. Alhemr has an AEME at the kW scale, and Enapter targets to produce AEME at bigger scales from 2023 with the help of a new factory built in Germany [59].

The tendency to produce and utilize high-TRL electrolyzers such as AE and PEME for GHP on large-scale is high. For instance, QLS-H10 PEME and DQ-5000/3.2 AE produced by Shandong Sikesaisi Hydrogen Energy Co., Ltd., were combined with PV power for GHP in four climate zones of China and compared the potential of the zones by using  $\frac{kgH_2}{km^2}$  index. They combined 3% PEME with 97% AE to reduce the start-up

time. The two climates with higher solar radiation had higher daily [Hydrogen Production Forecast \(HPF\)](#) of  $32.5 \times 10^3$  and  $27.5 \times 10^3 \text{ kgH}_2/\text{km}^2$  [60]. In Malaysia, more than 140 offshore platforms will be decommissioned within 10 years from 2023, hence N. D. Alias and Y. I. Go [61] proposed a solution for reusing these platforms with three scenarios of installing solar PV panels for GHP by PEME. The three scenarios produced hydrogen with LCOH of 10.95, 27.45, and 14.53  $\left(\frac{\text{USD}}{\text{kgH}_2}\right)$ . For the best scenario, the annual STH was 74.5%. In another effort, the USA and EU aim to produce plentiful electrolyzers with large-scale capacities to reduce the LCOH for GHP. Cost reduction of GHP mainly depends on electrolyzer cost, electricity cost, electrolyzer efficiency, full load hours of operation, lifetime of electrolyzer, and Weighted Average Cost of Capital (WACC) [62]. As can be seen, among the impacting parameters except for the electricity price that depends on the Learning Rate (LR) of the renewable power technologies the other parameters directly or indirectly rely on the electrolyzer stack advancement technologies, the materials used in the production process of membrane, electrodes, interconnections, and improving the electrolyzer subsystems. Based on IRENA analysis for cost reduction of GHP from 6 USD/kgH<sub>2</sub> in 2020 to 1 USD/kgH<sub>2</sub> in the future the electrolyzer cost must decrease by 80%, the electricity tariff must be reduced from 53 to 20 USD/MWh, electrolyzer efficiency increase from 65% to 76%, full load operation hours increase from 3200 h to 4200 h, lifetime of electrolyzers increase from 10 to 20 years, and WACC decrease from 10% to 6% [62]. According to the guideline proposed by IRENA for cost reduction of GHP, there is a great opportunity for Research and Development (R&D) of electrolyzers. For instance, larger stacks manufacturing, quality control, improving efficiency, and reducing CAPEX and OPEX. A more efficient electrolyzer reduces the main OPEX and the electricity cost. However, there is always a trade-off between efficiency, durability, and costs. While manufacturing thinner membranes, more effective catalysts, and less critical raw materials increase the cell efficiency and reduces the CAPEX, the durability of the cell is reduced. In addition, when the operating pressure is increased, maintenance schedules are reduced and low-quality water is used the OPEX improves but durability reduces [62]. In other words, we should compromise between reducing durability and improving efficiency and costs.

In the following different electrolyzer technologies are studied and the 4E-TS criteria for each technology is reported in Table 2a. Table 2b shows the opportunities and potential challenges of each technology according to the study. This table provides the potential research and investment field for the future of GHPSs.

**Table 2. The 4E-TS performance indicator comparison of AE, PEME, SOE, AEME, MLE, and PCCE from the reviewed sources**

| Criteria                                                 | AE                                                                                                                                       | PEME                                                                                                                                                      | SOE                                                                                          | AEME                         | MLE                                  | PCCE                                                                        |
|----------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------|------------------------------|--------------------------------------|-----------------------------------------------------------------------------|
| $\eta_{ele}(\%)$                                         | $\geq 72.47^{**}$ [60], 62-82 [63], 60-70 [64], 85 [65], 60 [57]                                                                         | 71.02 <sup>u</sup> [60], 74.5 [61], 75.5 [66], 67-82 [63], 65-85 [64], 60 [57], 52 off-grid PV, 68 on-grid-PV [67], 85 [68], 86 [69], 88 [70]             | 84 [59], 90-95 [63], up to 97 [64], 80-90 [57], 84 [71]                                      | 70-80 [64]                   | System efficiency: 71, stack 90 [72] | 60 at 1.28 V for the free heat source and 600 °C, see Figs. 9c and 9d [52]  |
| Electricity consumption density ( $\frac{kWh}{Nm^3}$ )   | $\leq 4.9$ DC [60], 4.8 [73], 4.5-7 [63]                                                                                                 | 5 [60], 4.7 <sup>u</sup> [66], 4.7 [73], 4.5-7.5 [63]                                                                                                     | 3.4 [73], 2.5-3.7 [63]                                                                       |                              |                                      |                                                                             |
| Specific electricity consumption ( $\frac{kWh}{kgH_2}$ ) | $\leq 54.5$ DC [60], 54 [73], 59 [74], 51.2 [75], 50-78 [76]                                                                             | 55.6 [60], 52.2 <sup>u</sup> [66], 52 [73], 59 [74], 51.2 [75], 50-83 [76], [77]                                                                          | 37.5 [73], 38 with steam import, 48 without steam import [76], 40 <sup>u</sup> [71],         | 59, (54-65) [78], 57-69 [76] | Satck: 44.35 system: 55.9 [72]       | 54-66 at 550 °C to 700 °C [52]                                              |
| Hydrogen production ( $\frac{Nm^3}{h}$ )                 | 5 [60], 1-1500 [63]                                                                                                                      | 83 [60], 1-230 [63], 0.12 [79]                                                                                                                            | 365 [73], 1-40 [63], designed for 150167 equivalent to 13.5 t/h [71]                         |                              | 0.65 [72]                            | See Fig. 10b [52]                                                           |
| Hydrogen purity                                          | $\geq 99.8$ [60], 99.8 [63]                                                                                                              | $\geq 99.999$ [60], [66], 99.999 [63], 99.9 [77]                                                                                                          | 99.9 [63], 99.9 [71],                                                                        | $>99.9 \pm 0.05\%$ [80]      | 99.95 [72]                           |                                                                             |
| Operating pressure (MPa)                                 | 3.2 [60], $< 3$ [63], 0.3-20 [64], 2.5-3 [65], 0.1-3 [76]                                                                                | 0-3.2 [60], $< 70$ [63], 1-20 [64], 0.1-1.6 [79], 8-35 [76], $< 3$ [77]                                                                                   | Hydrogen pressure of 0.0025, steam pressure 0.45-0.55 [73], $< 3$ [63], 1-6 [64], 0-0.2 [76] | 0.1-1.5 [64], up to 3.5 [76] | 1 [72]                               | 0.21 to 1.26 [81]                                                           |
| Operating temperature (°C)                               | 60-80 [63], 20-80 [64], $< 100$ [57], 70-90 [76]                                                                                         | 50-80 [63], [76], [77] 20-200 [64], $< 100$ [57]                                                                                                          | $> 650$ [60], 650-1000 [63], 500-1000 [64], 500-850 [57], 500-900 [76], 600-850 [71]         | 20-200 [64], 40-80 [76]      | 145 [72]                             | 550 [58], 200-600 [76], 550 [81], 700 [82], 500-700 [52], 550, 400-600 [83] |
| Cell voltage (V)                                         | 1.8-2.4 [63]                                                                                                                             | 1.8-2.2 [63], 1.2-2.5 [77]                                                                                                                                | 0.95-1.3 [63], 1.3 [71]                                                                      |                              | 23.1 DC [72]                         | 1.7 [82], 1.28 [52]                                                         |
| Current density ( $\frac{A}{cm^2}$ )                     | 0.2-0.8 [63], 0.4-1 [76]                                                                                                                 | 0.6-2 [63], 0.2-4 [76], 1-2 [77]                                                                                                                          | 0.3-1 [63], 0.5-1.5 [76]                                                                     | 0.1-1 [78], 0.2-2 [76]       | 0.3 [84]                             | 0.5 [81], [82], as high as 0.8 [52]                                         |
| Charge carrier                                           | $OH^-$ [64]                                                                                                                              | $H^+$ [64]                                                                                                                                                | $H^+$ [64]                                                                                   | $O^{2-}$ [64]                | $H^+$ [85]                           | $H^+$ [86]                                                                  |
| $\psi_{ele}(\%)$                                         | 4 for solar-based, 30 for wind-based [26]                                                                                                | 4% for solar-based, 30 for wind-based [26], hydrokinetic-based 36.5 [70]                                                                                  |                                                                                              |                              |                                      |                                                                             |
| GWP ( $\frac{kgCO_{2,eq}}{kgH_2}$ )                      | PV-E: 3.24 <sup>u</sup> , Wi-E: 2.71 <sup>u</sup> , HyR-E: 0.53 <sup>u</sup> , Hy-E: 0.41 <sup>u</sup> [74], PV-E: 3.39, Wi-E: 0.85 [87] | PV-E: 3.24 <sup>u</sup> , Wi-E: 2.71 <sup>u</sup> , HyR-E: 0.53 <sup>u</sup> , Hy-E: 0.41 <sup>u</sup> [74], PV-E $< 3$ [67], PV-E: 3.13, Wi-E: 0.75 [87] |                                                                                              |                              |                                      |                                                                             |
| CAPEX ( $\frac{USD}{kW_e}$ ) for the entire system       | 500-1400 [59], 1180 [65], 500-1500 [76], normal 1180, low-cost high efficiency 628 [88],                                                 | 1100-1800 [59], 1189 [89], 1400-2100 [76], normal 1500, low-cost high efficiency 708 [88], 537 (stack only) [68], 700-1400, and 400 for stack only [77]   | 2800-5600 [59], 917-4000 [76]                                                                | 3333 [76], 400-800 [90]      | 15000 [72]                           |                                                                             |
| OPEX ( $\frac{USD}{kW_e \cdot year}$ )                   | 30 [65], 3% of CAPEX, 35.4 [88], 18.6 [90]                                                                                               | 17.5 [89], 3% of CAPEX, 45 [88], 21.5 [68]                                                                                                                |                                                                                              |                              |                                      |                                                                             |

|                                             |                                                                                                                                                                                                                                                                                   |                                                                                                                                                                                                                                        |                                                                                                                                            |                                                                           |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
|---------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------|----------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| LCOH ( $\frac{USD}{kgH_2}$ )                | 5.08**, 4.09*** [75], 5.042 [91], 5.35 [92], 4.3-4.8 [88], onshore wind 7.25, offshore wind 11.85, onshore-PV 11.77 [93]                                                                                                                                                          | 10.95 [61], 5.08**, 4.09*** [75], 4.20 [94], on-grid PV 3.7-6.5, off-grid PV 7.6 [67], 4.3-4.8 [88], 2.41 [68], onshore PV 13.44, onshore-wind 8.51 and offshore-wind 12.6 [93], offshore wind 6.71 [77]                               |                                                                                                                                            |                                                                           |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| Electrolyte type                            | Liquid [64], Liquid potassium or sodium hydroxide [57], 25-40% liquid KOH [76]                                                                                                                                                                                                    | Polymeric [64] solid polymer, perfluorosulfonic acid (PFSA) Membranes [57], Solid PEM (Nafion) [76]                                                                                                                                    | Solid ceramic [64], Solid ceramic, YSZ [57], Solid: ceramic – zirconia or ceria based [76].                                                | Polymeric [64], Liquid-solid hybrid: 1% KOH/anion exchange membrane [76]. | Liquid (acidic, alkaline, and neutral) [95].                   | Solid electrolyte, $BaCe_{0.4}Zr_{0.4}Y_{0.1}O_{3-\delta}$ (BCZYYb4411) [81], BCZY81–ZnO (5 mol%) [82], BZCY541 [52], $BaCe_{0.4}Zr_{0.4}Y_{0.1}O_{3-\delta}$ (BCZYYb) [96], [58]                                                                                                                                                                                                                                                                                                                                            |
| Anode and cathode material                  | A: Nickel-coated perforated stainless steel, C: Nickel-coated perforated stainless steel [57]                                                                                                                                                                                     | A: Iridium oxide, C: Platinum nanoparticles on carbon black [57]                                                                                                                                                                       | A: Nickel Oxide + Yttria-Stabilised Zirconia (YSZ), C: lanthanum strontium cobalt ferrite (LSCF)/lanthanum strontium manganite (LSM) [57]. |                                                                           |                                                                | A: (positrode): $BaCo_{0.4}Fe_{0.4}Zr_{0.1}Y_{0.1}O_{3-\delta}$ (BCFZY) [81], C: (negatrode): Ni–BZCYb4411Composite [81], A: $NiO - BaCe_{0.8}Zr_{0.1}Y_{0.1}O_{3-\delta}$ (BCZY81) [82], C: $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ (BSCF)-BCZY81) [82], A: positrode based on $BaGd_{0.8}La_{0.2}Co_2O_{6-\delta}$ (BGLC1082) and $BaZr_{0.5}Ce_{0.4}Y_{0.1}O_{3-\delta}$ (BZCY541) [52], C: Ni-BZCY541 [52], A: nickel-electrolyte composite [58], C: $BaCo_{0.4}Fe_{0.4}Zr_{0.1}Y_{0.1}O_{3-\delta}$ (BCFZY) [58] |
| Materials consumption for catalysts (kg/MW) | Nickel, Aluminium, Steel, Zirconium [57], Nickel: 800, Zirconium: 100, Aluminium: 500, Steel: 10000, cobalt, and copper [97], for the electrolyzer stack: Copper: 533.33, Unalloyed Steel: 53,333.33, Nickel: 5066.67, decarbonized water: 2933.33, Deionized water 22933.33 [87] | Platinum, Palladium, Iridium [57], Platinum: 0.3, Iridium: 0.7 [97], for the electrolyzer stack copper: 0.53, LoA-alloy steel: 29,113.04, Reinforced steel: 1293.91, Cast iron: 210.26, PGMs: 0.19, Titanium: 0.61, Silicon: 1.05 [87] | Nickel, Zirconium, Lanthanum, Yttrium [57], Nickel: 150-200, Zirconium: 40, Lanthanum: 20, Yttrium: <5 [97]                                |                                                                           |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| TRL                                         | 8-9 [56], 9 [57], [59], 9 [64]                                                                                                                                                                                                                                                    | 8-9 [56], 9 [57], [59], 7 [64]                                                                                                                                                                                                         | 5-6 [56], 9 [57], 7-8 [59], 6 [64]                                                                                                         | 2-3 [56], 6 [57], [59], 6 [64]                                            | 6 [57], laboratory test 3-5 [85], [84], demonstration 5-6 [72] | laboratory test, 3 [58],                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |
| Deployment situation                        | >1 GW deployed [76]                                                                                                                                                                                                                                                               | >100 MW deployed [76]                                                                                                                                                                                                                  | <10 MW deployed [76], 5-10 MW suggested, designed for 9 MW [71]                                                                            | <10 MW deployed [76]                                                      | 2.5 kW [72]                                                    | First stack integration [58]                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
| LR (%)                                      | 16-21% [62], 9% [98], [73]                                                                                                                                                                                                                                                        | 16-21% [62], 13% [98], [73]                                                                                                                                                                                                            | 28% [73]                                                                                                                                   |                                                                           |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| Availability (%)                            | 95 [89]                                                                                                                                                                                                                                                                           | 95 [89]                                                                                                                                                                                                                                | 95 [89], 93 [71]                                                                                                                           | 95 [89]                                                                   |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| Reliability (%)                             |                                                                                                                                                                                                                                                                                   |                                                                                                                                                                                                                                        | >90 [71]                                                                                                                                   |                                                                           |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| Capacity factor (%)                         | 95 [75]                                                                                                                                                                                                                                                                           | 95 [75]                                                                                                                                                                                                                                |                                                                                                                                            |                                                                           |                                                                |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |

|                                                                |                                                                                                                              |                                                                                                                                                                                                                                                                                                                                          |                                                                     |                                                                     |                                                       |                               |
|----------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------|---------------------------------------------------------------------|-------------------------------------------------------|-------------------------------|
| The capacity factor of the RES feeding the electrolyzer (%)    | Deep offshore wind turbine: 60-70 [78],                                                                                      | Solar-PV: 24.9, Wind: 35.5, Hydro: 41.5, Geothermal: 74.3 [99], solar-PV: 26.6 and 29.4 onshore Wind: 30.6 and 46.3 [89], Deep offshore wind: 60-70 [78], Wind turbine (3 and 6 blades), Solar-PV [69]                                                                                                                                   |                                                                     | Deep offshore wind turbine: 60-70 [78]                              |                                                       |                               |
| Water required electrical resistivity ( $\frac{M\Omega}{cm}$ ) |                                                                                                                              | $\geq 10 M\Omega/cm$ [60]                                                                                                                                                                                                                                                                                                                |                                                                     |                                                                     |                                                       |                               |
| Electrolytic water flow rate ( $\frac{L}{kgH_2}$ )             |                                                                                                                              | 10 [60],                                                                                                                                                                                                                                                                                                                                 | 10.5 [73]                                                           |                                                                     |                                                       |                               |
| Cold Start-up time (minute)                                    | <60 [63], 30-60 [76], 40 [88]                                                                                                | 1 [66], <20 [63], <5 [78], shutdown in seconds [78], 5 [76], 20 [88]                                                                                                                                                                                                                                                                     | Warm startup 10 min [73], >60 [63], >12 hours [57], >600 [76]       | 5-10 min, and 2-4 s for ramp-up from 20% to full load [78], 20 [76] |                                                       |                               |
| Ramp up time hot idle to full load (s)                         | 60 [76]                                                                                                                      | 10 [76]                                                                                                                                                                                                                                                                                                                                  | 600 [76]                                                            | 1800 [76]                                                           |                                                       |                               |
| Minimum operating load (%)                                     | 10-40 [76], 20 [88]                                                                                                          | 40 [66], 5-10 [76], 5 [88]                                                                                                                                                                                                                                                                                                               | >3 [76]                                                             | 5 [78], 10-20 [76]                                                  |                                                       |                               |
| Lifetime at full load (1000 h)                                 | 60-100 [76], 80 [88], 60 [90]                                                                                                | 50-90 [76], 60 [88], 15 years [68], 50-80 [90], [77]                                                                                                                                                                                                                                                                                     | 20-50 [76], <20 [90]                                                | 5-10 [78], 5-40 [76], >5 [90]                                       | >10 Years for the stack, 20 years for the system [72] | 3.5 [82], 1.68 at 600 °C [52] |
| Degradation (%) per 1000 hours (kh)                            | 0.13 [76]                                                                                                                    | 0.25 [76]                                                                                                                                                                                                                                                                                                                                | 0.55-1 [76], 10% V Assumption for replacement at end-of-life [71]   | 0.4 [76]                                                            |                                                       | <1 [58], 2 [82], 1.5 [96]     |
| RES used for GHP                                               | Solar-PV [60], geothermal [91], [92], mixed solar-PV and wind power [88], onshore/offshore wind turbine, and onshore-PV [93] | Solar-PV [60], Offshore Solar-PV [61], solar-PV, Wind, Hydroelectric [74], Offshore wind energy [63], solar-chimney electricity [100], mixed wind and solar-PV [65], geothermal [94], solar-PV [67], mixed solar-PV and wind power [88], hydro and solar-PV [68], onshore/offshore wind turbine, and onshore-PV [93], offshore wind [77] |                                                                     | offshore wind [78]                                                  |                                                       |                               |
| Leading developers and manufacturers                           | LONGI, John Cockerill, Clean power hydrogen (CPH2), NEL, Hydrogen pro., AUYAN, Sunfire [76]                                  | Plug, Ohmium, ITM POWER, Siemens Energy, SUNGROW, Clean power hydrogen (CPH2) [76]                                                                                                                                                                                                                                                       | Sunfire, TOPSOE, BloomEnergy, FuelCell Energy, Ceres, Elcogen, [76] | Enapter [76]                                                        | REMA [72]                                             |                               |

#The energy efficiency was calculated according to the data given by [60],  $\eta_{ele}(\%) = \frac{\rho_{H_2}(\frac{kg}{m^3})HHV_{H_2}(\frac{kWh}{kg})}{Power\ consumption(\frac{kWh}{m^3})} = \frac{0.0899 \times 39.5}{5} = 0.7102$ .

## The density of electricity consumption was calculated according to the data given by [66], Density of electricity consumption  $(\frac{kWh}{m^3}) = \frac{\rho_{H_2}(\frac{kg}{m^3})HHV_{H_2}(\frac{kWh}{kg})}{\eta_{ele}} = \frac{0.0899 \times 39.5}{0.755} = 4.7(\frac{kWh}{m^3})$ .

### Specific electricity consumption  $= \frac{Density\ of\ electricity\ consumption(\frac{kWh}{Nm^3})}{\rho_{H_2}(\frac{kg}{m^3})} = \frac{4.7}{0.0899} = 52.2(\frac{kWh}{kg})$ .

\* GWP  $(\frac{kgCO_{2,eq}}{kgH_2}) = 1.05GWP(\frac{kgCO_{2,eq}}{MWh}) \times Specific\ electricity\ consumption(\frac{MWh}{kgH_2})$ , the 1.05 is for including the GWP of the electrolyzer according to the recommendation of [74]. In the reference [74] the GWP is given in  $\frac{kgCO_{2,eq}}{MWh}$ , and specific electricity consumption is assumed  $59 \frac{kWh}{kgH_2}$ .

\*\* The LCOH in the [75] was given in Canadian dollars, here we assumed 1 Canadian Dollar equals 0.734 US Dollars.

\*\*\* A pathway with emission costs and output-based allocation of emissions credits reduces LCOH [75].

& Total specific electricity consumption in  $(\frac{kWh}{kgH_2})$  including stack (33.3), compression (2), e-heaters (3.2), AC/DC 97% eff. (1.5) [71], and when steam generation is included a total of  $47.7(\frac{kWh}{kgH_2})$  [71].

**Table 2b. The potential challenges and opportunities of different electrolyzers**

|      |                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |
|------|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| AE   | Opportunities: Mature, Commercialized, low-cost, Nickel catalyst is a non-noble metal and abundant, quick startup, stable operation for large-scale GHP, GW scale deployment [57], [76].                                                                                                                                                                                                                                                                                                                                                                                       |
|      | Potential challenges: Higher corrosion because of liquid electrolyte, low working pressure, medium pure hydrogen production, sensitivity to impurities, and require highly purified water [57], Low power density, larger footprint, high minimum load [76]                                                                                                                                                                                                                                                                                                                    |
| PEME | Opportunities: Relatively mature, Compact size, smallest footprint, high operation pressure, highly pure hydrogen production, solid electrolyte with lower risk of leakage and corrosion, flexible operation from small minimum load to slight overload, high power density, MW scale deployment [57], [76].                                                                                                                                                                                                                                                                   |
|      | Potential challenges: Expensive material in catalysts (i.e. PGMs, titanium), Sensitive to impurities, needs highly pure water, long-term stability should be confirmed in MW scale, and dependence on non-natural chemicals (i.e. PFAS) that hardly degrade in nature [57], [76].                                                                                                                                                                                                                                                                                              |
| SOE  | Opportunities: High efficiency, solid electrolyte, and less corrosion and leakage problems, less sensitive to impurities, hence less pure water can be handled, suitable for steady operation, high-quality waste heat, Potential for reversible operation as fuel cell, using inexpensive, abundant mineral materials [57], [76]                                                                                                                                                                                                                                              |
|      | Potential challenges: Long startup time, unsuitable for part load and changeable conditions, biggest footprint, fast aging, produced hydrogen must be separated from steam for purification, low working pressure, poor durability [57], [76]. O&M experience is only available on pilot and demo scale [71]                                                                                                                                                                                                                                                                   |
| AEME | Opportunities: Using abundant and non-expensive materials, it is compact in size and has a small footprint and is suitable for high operating pressure [76].                                                                                                                                                                                                                                                                                                                                                                                                                   |
|      | Potential challenges: Small-scale (kW), deployment only, has a shorter lifetime than PEME and AE due to the higher degradation rate of the membrane [76].                                                                                                                                                                                                                                                                                                                                                                                                                      |
| MLE  | Opportunities: Lower CAPEX and OPEX when up-scaled, smaller degradation, high efficiency, and the possibility of using low-quality water [85], [84].                                                                                                                                                                                                                                                                                                                                                                                                                           |
|      | Potential challenges: Lower Faradic efficiency at high working current densities, low hydrogen purity, oxygen and hydrogen crossover, electric arcing between anode and cathode due to applying high voltage across a small gap between electrodes, and upscaling limitations. [85], [84].                                                                                                                                                                                                                                                                                     |
| PCCE | Opportunities: Higher operating pressure reduces system complexity, footprint, and balance of plane costs, and improves efficiency. Intermediate operation temperature reduces aging due to thermal stress compared with SOE, and higher ionic conductivity due to the small size of protons reduces the cell size for the same capacity compared with SOE [58]. Solid state electrolyte reduces leakage, and make the cell more packageable to minimize footprint. This technology can be used as a reversible electrolyzer/fuel cell for hydrogen and power generation [83]. |
|      | Potential challenges: The technology is still immature and a lab-scale stack is presented [58]. Material optimization is required to reach an acceptable degradation rate, the degradation is still high compared with AE, PEME, and even SOE. The intermediate temperature increases the start-up time and ramp-up time compared with PEME and AE but it is smaller than SOE.                                                                                                                                                                                                 |

#### 4-1-1- PEME

PEME, a relatively mature technology with [TRL](#) of about 8 to 9 [56], [57], [CRI](#) of 2 to 3 [101] at the demonstration level, and deployed capacity of more than 100 MW [76] is an advanced technology for GHP with purity of more than 99.99%, GHG production of  $\leq 1 \frac{kgCO_{2,eq}}{kgH_2}$  [46] and ready for large-scale development. The USA and EU are pointing to manufacturing numerous electrolyzers with GW capacities. Siemens Energy [66] in 2023 started the industrial production of a multi-GW PEME facility in Berlin. The unit's capacity is supposed to increase to 3 GW by 2025 depending on demand. Silyzer 300 is an atmospheric PEME manufactured by Siemens Energy with plant efficiency of more than 75.5%, comprised of 24 stacks that can produce  $335 \frac{kgH_2}{h}$ . The hydrogen purity is 99.999%. Its start-up time is less than 1 minute, and it consumes less than 10 kg of demineralized water for production of 1 kgH<sub>2</sub>. The minimum part-load operation of Silyzer 300 is 40% [66]. Siemens proposed integrating the PEME with a heat pump to increase the energy utilization from 75 to 96% [102]. In another attempt to industrialize PEME in 2022, General Motors helped Norwegian manufacturer Nel to develop a novel PEME under a strategic collaboration. The Nel company has won a 56 million dollar electrolyzer order for a 290 MW USA green hydrogen project [103]. The main purpose of this collaboration is to produce more cost-competitive green hydrogen. The pivotal factor in improving the economics of PEME for large-scale implementation lies in the Platinum catalyst used on the membrane surface [104]. W. A. Hindson and S. James proved that using patterned membranes to increase the Platinum loading on the membrane surface increases current draw and GHP rate. The surface modification was done in four steps of roughening, single striations, double striations, and catalyst deposition. Patterned membranes produce 9% more green hydrogen than the only roughened membranes, they also increase current draw, and catalyst retention at a molecular level [104]. The vital materials for the electrolyzer chemistry include Platinum, Palladium, and Iridium [57]. Platinum Group Metals (PGMs) are 6 components with physical/chemical properties in common, including platinum, palladium, rhodium, ruthenium, iridium, and osmium. According to the Hydrogen Council [105], electrolyzer capacity rose 30% (equivalent to 170 MW) in 2022 and reached a total capacity of 700 MW. PEME has captured about 30% of this market. Looking forward to 2050, the IEA approximates that total electrolyzer capacity will reach 4,000 GW, involving 1,550 GW of PEME capacity [105]. Among the PGMs, iridium (*Ir*) is approximately 20 times rarer than platinum. Worldwide iridium supply is about 7.1 tons/year which is approximately equal to the present demand for all applications [105]. Currently,  $0.4 \frac{tonIr}{GW_{PEME}}$  is required and to reach 1550 GW of PEME in 2050 in the following 27 years, 620 tons of *Ir* should be supplied which corresponds to a mean value of about 23 tons/year, more than 3 times of the present

supply (even with excluding other applications demand for *Ir*). This shows a potential for a shortage of the iridium supply and development obstacle for the PEME. However, as mentioned PGMs have physical and chemical characteristics in common, and other PGMs can substitute a portion of demanded iridium. In addition, thrifting and recycling can supply some portion of the demand. According to the World Platinum Investment Council (WPIC) [105] research, the original supply, substituting, thrifting, and recycling together can supply the demand for iridium by 2050 and no shortage will happen for this rare material [105]. However, due to the scarcity of the PGMs, iridium has a very high potential to become a risk in the PEME's supply chain of the PEME. Hence decreasing dependence on PGMs is a wise approach that must be sought in future research and innovations [57].

Using the right source to provide renewable electricity for the electrolyzer can improve GHP's economic and environmental indicators. An Energy Ecology Efficiency (EEE) study was presented by L. B. B. Maciel et al. [74] to evaluate the impact of [GWP](#), electrolyzer efficiency, and efficiency of RES conversion to electricity on the GHP by renewable electricity originating from solar PV, hydroelectric (with and without reservoir), and wind power. They assumed that the share of the electrolyzer in the total GWP of the plant is 5% with electrolyzer efficiency of 75% with mean specific electrical energy consumption of  $59 \frac{kWh}{kgH_2}$ . The results showed that the GHP by hydroelectric power generation is the most efficient and solar PV is the most inefficient. Wind power lies between solar PV and hydroelectric power generation. The main reason is solar PV's low conversion efficiency compared to hydro- and wind powers [74]. The average GWP of different solar PV technologies, wind power, hydroelectric with and without reservoir are 52.3, 43.8, 8.6, and  $6.6, \frac{kgCO_{2,eq}}{MWh}$  which shows hydroelectric without reservoir has the least GWP [74]. Combining offshore wind turbines with PEME for GHP in Turkey, Edirne-Enez location resulted in an LCOH of 6.71 USD/kgH<sub>2</sub> (6.26 Euro/kgH<sub>2</sub>) and for the year 2050 it is estimated to become 1.21 USD/kgH<sub>2</sub> [77]. Different combinations of solar-PV and wind power coupled with PEME in 5 scenarios were examined theoretically, for this purpose they assumed a land with a specified area [106]. The outcomes showed that the case with only wind power achieves the lowest LCOH (2.5 USD/kgH<sub>2</sub>) and the scenario with PV power only has the highest LCOH of 4 USD/kgH<sub>2</sub> [106]. A feasibility study of oceanic energy for GHP was done by M. P'erez-Vigueras et al. [63] and PEME was chosen as the most suitable electrolyzer due to its fast startup and response to intermittent oceanic energy and offshore wind power. Salt caverns were the most economical large-scale hydrogen storage. For the short distances the most economical way to transport hydrogen was to transmit electrical power to the coast and convert it to hydrogen by PEME, for distances more than 1000 km, hydrogen transport by ship is the most economic option. Increasing the GHP rate by coupling a bifacial

PV (bPV) system with building cool roof paint and PEME was done experimentally by F. F. Ahad et al. [79]. They concluded that using cool roof paint with bPV and PEME increases the power generation and GHP by 33.02% compared to monofacial PV (mPV). GHP with grid-tied and off-grid PV-based battery and PEME system was investigated by A. L. Bukar et. al. [67] for Thailand. They used PEME with a PV-based battery due to its compatibility with variable solar power and wide modulation range. The PEME had an efficiency of 52% with maximum efficiency of 68% happening at 18% of its rated power. According to the results, the on-grid system can reduce the hydrogen capacity system. In addition, the hydrogen cost for on-grid PV-GHPS changes from 3.7 to 6.5 USD/kgH<sub>2</sub> depending on the electricity cost of the grid, while it is 7.6 USD/kgH<sub>2</sub> for the off-grid system. Which means an on-grid system is more profitable. The results also show the significant impact of electricity cost on the system design, and as a result, for high electricity price situations, a bigger GHPS is more cost-effective. In addition, they considered the carbon footprint limit of 3 kgCO<sub>2eq</sub>/kgH<sub>2</sub> for GHP, and for electricity prices of bigger than 120 USD/MWh, it was achievable [67]. The combination of wind and solar-PV for hydrogen production from distilled water by PEME and then converting the hydrogen to electricity by using PEMFC was investigated experimentally [69]. As the voltage increases the hydrogen generation increases as well, but the efficiency decreases because most of the electricity is lost due to different losses such as ohmic loss [69]. On-grid combined hydroelectric and solar PV power stations for GHP by PEME for green transportation in southern Oman are investigated [68]. The hydropower is generated by a water source having a head of 70 m and a flow rate of 1 m<sup>3</sup>.s<sup>-1</sup> They optimized the configuration by Homer-pro and achieved 2.41 USD/kgH<sub>2</sub> for the LCOH. The LCOH sensitivity to the 5 % OPEX and 10% CAPEX reduction of main components including PV panels, electrolyzers, hydrogen tanks, power converters, hydro turbines, and batteries was investigated and concluded that the LCOH is most and least sensitive to the costs of PV panels and batteries.

#### 4-1-2- AE

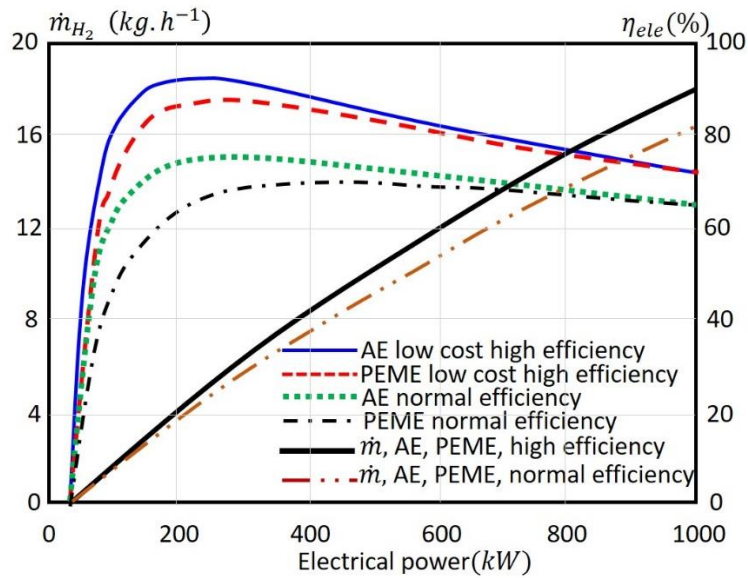
AE is a mature technology for GHP [51], commercially available with a TRL of 9 ( [56], [57], [59] and [64]), CRI of 4-6 [101], deployed capacity of more than 1GW [76] and reported electricity to hydrogen conversion efficiency ( $\eta_{ele}$ ) of 60-85% ( [60], [63], [64] and [65]). It has a learning rate (LR) of 9-21% ( [62], [98], and [73]) which means the scale-up of AE will further reduce the LC of GHP. AE has a moderately low CAPEX and stable technology. Still, it has slow kinetic, electrolyte corrosion, and gas permeation [64] and in comparison with PEME is less flexible when connected to an intermittent power supply [51]. Combining AE with PEME for GHP can improve its flexibility and reduce the startup time [60]. Assessment of GHP by AE, its compression, and then combusting it as a fuel in a cogeneration cycle

based on a GE 7HA.02 gas turbine [107] was carried out by D. Pashchenko [64]. It was concluded that the overall efficiency of renewable electricity conversion by AE to hydrogen, its compression, and the final utilization in the cogeneration cycle is about 38% [64]. The feasibility study of utilizing microgrids comprising AE, wind turbines, and solar-PV for GHP for the fuel cell buses was investigated in different scenarios of off-grid, on-grid, compressed hydrogen, and non-compressed hydrogen [65]. The LCOH for the on-grid scenario with compressed hydrogen to 40 bars and without compression was 9.08 USD/kgH<sub>2</sub>, and 8.37 USD/kgH<sub>2</sub> respectively. LCOH for the off-grid case with and without compression was 13 USD/kgH<sub>2</sub> and 12.1 USD/kgH<sub>2</sub> which is higher than the on-grid scenario in both cases. Hence the best scenario is the non-compressed hydrogen produced by the on-grid scenario [65]. Care must be taken that in the analysis they assumed the AE's efficiency is 85%, which is rather high according to the available AE electrolyzers in the market and literature. Geothermal as another RES resulted in LC of 5.042 USD/kgH<sub>2</sub> [91], and 5.35 USD/kgH<sub>2</sub> [92] for GHP by AE. Compared with [94] which used PEME with geothermal electricity for GHP, PEME results in a lower LCOH of 4.20 USD/kgH<sub>2</sub>. A reason for this difference is the higher efficiency of PEME, another reason is the higher temperature of the geothermal source, which resulted in higher exergy efficiency of the plant proposed by [94]. In another integrated system, S. Baral and J. Sebo evaluated hydrogen production by 8 different scenarios using solar thermal, geothermal, Organic Rankine cycle, solar-PV, onshore wind turbine, offshore wind turbine, hydropower, hydrogen tank, AE, and battery. The best scenario for 2023 was integrating solar photovoltaic (with storage) and onshore wind turbines with a LCOH of 4.7 USD/kgH<sub>2</sub>. The payback period was 8 years and the internal rate of return (IRR) was 5.06% [90] which is risky for unstable economics. For 2030, the best scenario was integrating solar-PV with offshore and onshore wind turbines and the LCOH was in the range of 1.46 USD/kgH<sub>2</sub> to 3.09 USD/kgH<sub>2</sub> depending on the capacity factor range. Using a hybrid system integrated with an ORC system was promising for 2050 when it is installed in industries such as glass, steel, and metal where the waste heat can be used for hydrogen production. According to the sensitivity analysis, the most impacting parameters for all 8 hybrid configurations studied are the LCOE and the [capacity factor \(CF\)](#) [90]. In another research AE and PEME were used with three RESs of onshore and offshore wind power and onshore-PV systems to reach the feasible configuration [93]. This study has considered the impacts of partial load operation of the components, hence the results are more realistic when compared with those data that consider operation at full load or rated power. The results show that the AE onshore wind power operated has the lowest LCOH (7.25 USD/kgH<sub>2</sub>) while the PEME onshore-PV operated has the highest LCOH (13.44 USD/kgH<sub>2</sub>) [93]. The most impacting parameters on the LCOH were the capacity factor of

the renewable energy used to supply the electrolyzer and the LCOE generated by the renewable power technology [93].

Applying policies for reducing GHP costs by two high-TRL technologies of AE and PEME was investigated and the LCOH was decreased from 5.08 USD/kgH<sub>2</sub> to 4.09 USD/kgH<sub>2</sub> by applying [Output-Based Allocations \(OBA\)](#), where emission credits are provided per Alberta's Technology Innovation and Emissions Reduction (TIER) regulation [75]. Applying this policy has reduced the LCOH by about 19.5% which is eye-catching and proves that over and above improving technology, using non-nobel materials in electrolyzers, increasing durability and efficiency of electrolyzers, reducing renewable electricity costs by scale-upping electricity production, and applying the innovative ideas to the membrane and electrodes of electrolyzers such as patterned membranes [104], applying right policies can boost the green hydrogen market soon [75]. This shows the importance of role of policymakers, parliaments, and governments. Related institutions and international organizations can play a critical role in shaping the minds of policymakers. Optimization of utility-scale hybrid solar-PV and wind power for GHP by AE and PEME is done in 16 scenarios for a location in Zaragoza (Spain) [88]. They minimized the LCOH using a genetic algorithm and two constraints of CAPEX and hydrogen production rate. They evaluated on-grid, and off-grid, with and without the possibility of purchasing electricity from grid scenarios. In addition to considering the part load efficiency of the electrolyzers (Fig. 4), they have considered the degradation of the renewable power generators (PV and wind turbines) during the cycle lifetime, which is assumed to be 20 years. Fig.4 shows the GHP and efficiency of the AE and PEME used in the analysis versus the electrical power consumption normalized for the 1 MW electrolyzer. The optimum LOCH achieved among 16 optimization scenarios was 4.3-4.8 USD/kgH<sub>2</sub>. [88].

It should be noted that operating temperature and pressure of AE can change the OPEX and CAPEX as well, this is studied by V. Kumar [108] using a simulation in Aspen.



**Figure 4. Comparison of GHP and efficiency of PEME and AE against electrical power consumption, in different cases, was normalized to 1 MW rated power. Graph regenerated by author [88]**

Four important [Life Cycle Impacts \(LCI\)](#), including Global Warming Potential (GWP), Water Depletion Potential (WDP), Mineral Depletion Potential (MDP), and Fossil fuel Depletion Potential (FDP) of AE and PEME depending on the renewable electricity source (solar PV or Wind), are compared, and the results are summarized in Table 3. As shown in all cases, wind-operated PEME has smaller LCIs, while solar-PV-operated AE has the biggest LCIs [87].

**Table 3. The comparison of AE with PEME operated with solar-PV or wind power for different LCIs [87]**

| LCIs                                         | AE-Wind | AE-Solar | PEME -Wind | PEME-Solar |
|----------------------------------------------|---------|----------|------------|------------|
| GWP(kgCO <sub>2eq</sub> /kgH <sub>2</sub> )  | 0.85    | 3.39     | 0.75       | 3.13       |
| WDP(litreH <sub>2</sub> O/kgH <sub>2</sub> ) | 16      | 28       | 15         | 26         |
| MDP (kgFe <sub>eq</sub> /kgH <sub>2</sub> )  | 0.64    | 0.86     | 0.55       | 0.74       |
| FDP (kgOil <sub>eq</sub> /kgH <sub>2</sub> ) | 0.23    | 0.91     | 0.21       | 0.85       |

#### 4-1-3- SOE

SOE is a novel technology for water splitting with a capacity deployment of less than 10 MW [76] and [TRL](#) reported in a wide range of 5 to 9 ( [56], [57], [59], and [64]), (Table 2). Depending on the leading manufacturers, and developers, the TRL of SOE is 6-8 [76], corresponding to the development of the systems and subsystems, and the transition from the demonstration level to the deployment level. Among the electrolyzers, it has the highest [LR](#) of 28% [73] meaning a very high potential for cost reduction in the scale-up. Out of 32.8 GW worldwide nameplate electrolyzer manufacturing capacity until 2023, only 2 GW (6%) is SOE, while PEME and AE shares are 11.1 GW (33.8%) and 19.7 GW (60%) [57]. Essential mineral materials and their consumption in building SOE include Nickel 150-200 kg/MW, Zirconium 40 kg/MW,

Lanthanum 20 kg/MW, and Yttrium: <5 kg/MW [97]. Fortunately, the required materials for SOE are abundant and inexpensive, and in comparison with the AE, it consumes less Nickel and Zirconium (Table 2). The high operating temperature of the SOE (500-1000 °C [64]) is a double-edged sword that causes a fast kinetic and improves the efficiency (80-97%) ([59], [63], [64], and [57]) on the one hand but causes high thermal stress on the internal components, aging and reducing durability on the other hand. Another reason for the higher efficiency of SOE is using steam as the feedstock that can be provided from different waste heat/steam sources in processes or renewable heat. Using waste steam (or steam import) as the SOE feedstock will increase its efficiency by at least 14% more than PEME and AE [76]. The high degradation rate of SOE (0.55-1% per kh of operation) that has reduced its lifetime to 20,000 to 50,000 h at full load operation [76] is an essential challenge that requires more investment, and investigation. Reducing the degradation rate of SOE with inexpensive and heat-resistant materials will help the SOE technology reduce the LCOH. SOE also takes up a bigger space when it is installed. Compared with AE and AEME for the same capacity, it needs 30% more than AE's footprint and 3.5 times more than PEME's footprint [76]. Hence, the packaging of SOE is another opportunity that can be investigated using more compact systems and auxiliary subsystems. The feasibility study of SOE upscaling potential and techno-economic aspects in three industries was carried out by the Institute for Sustainable Process Technology (ISPT) and the Netherlands Organization for Applied Scientific Research (IPT) in the Next Level SOE project [71]. According to the outcomes, more determinations are required to fully address dynamic behavior, reliability, hydrogen purity, and stack degradation for incorporating this in the design and O&M strategies. Current density is the key parameter for reducing CAPEX because less cell area is required for higher current densities for the same output ([71], and [76]). The scale-up potential depends on being able to increase the current density of SOE by 2 times (from 0.75 to 1.5 A/cm<sup>2</sup>), the cell area by 8 times (from 100 to 800 cm<sup>2</sup>), SOE stacks from 30-60 cells to 120-150 cells, and the capacity of the module up to 9 MW. For a cell voltage of 1.3 V, cell area of 800 cm<sup>2</sup>, current density of 1.5 A/cm<sup>2</sup>, and cell number per stack of 60, the stack current, voltage, and power will be 1200 A, 78 V, and 93.6 kW. They also concluded that steam generation can be done by recovering residual heat and not inevitably needing high or medium-pressure steam from the boilers that burn fuel in the plant. In addition, scale-up, mass production, and innovations in technology can potentially reduce the CAPEX by a factor of 4. This project has drawn a roadmap to develop and deploy SOE technology in this decade, and as the next step for upscaling it recommends developing a pilot plant for technology demonstration on the scale of 5 to 10 MW. The first project investigated ISPT [71] produces 13.5 t/h of hydrogen with 99.9% purity. The input feedstock to the SOE is 90/10 of steam/hydrogen on a molar basis, and the steam temperature and pressure are 380 °C and 42 barg at the turbine inlet. The air is

enriched with 30% vol oxygen. The schematic of the suggested GHP system for the first project is depicted in Fig.5 [71]. To reduce the LCOH to less than 3.23 USD/kgH<sub>2</sub> (3 Euro/kgH<sub>2</sub>), it is required to reach the goals defined for the current density, surface area, and also SOE plant scale bigger than 100 MW. The infographic of the plant is depicted in Fig.6 [71].

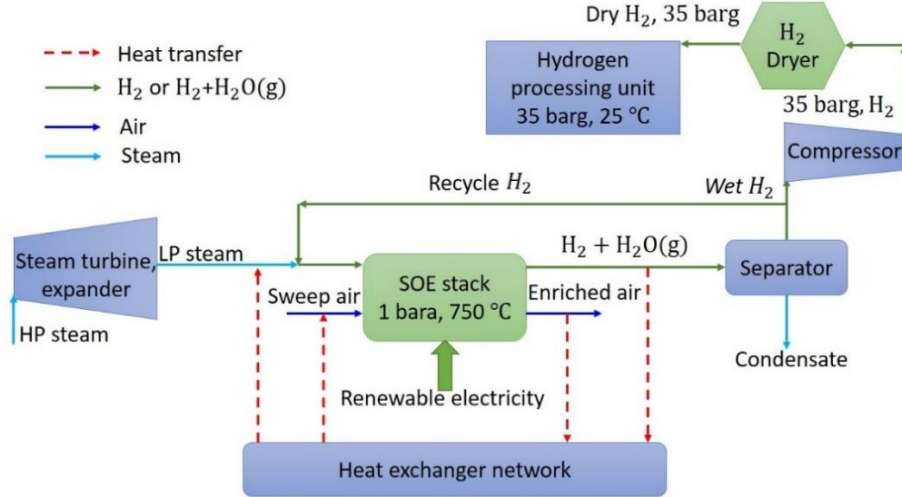


Figure 5. GHP system designed for the first project, in [71], the figure is regenerated by the author

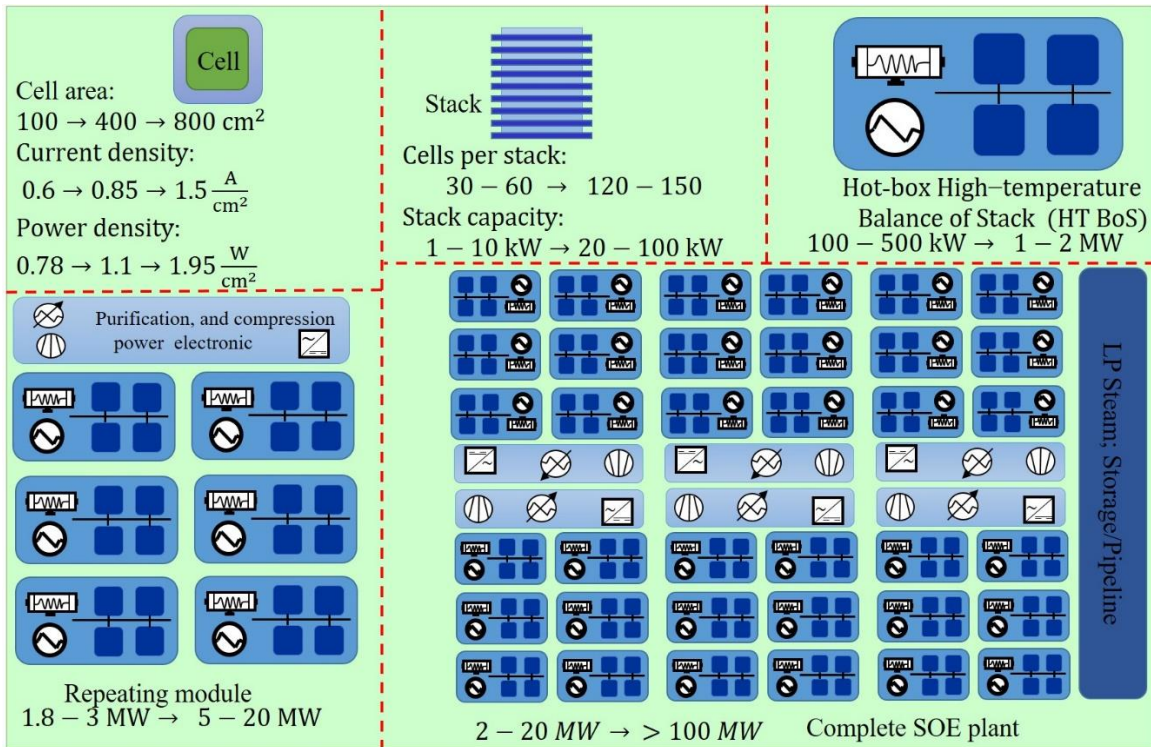


Figure 6. Infographic of SOE upscaling from cell to multi-MW capacity, figure regenerated by the author [71]

#### 4-1-4- r-SOE

Reversible solid oxide electrolyzer (r-SOE) is a technology that can be used for storing abundant renewable electricity as hydrogen and converting it back to electricity with the same device working in the fuel cell mode. The r-SOE has been demonstrated in the large-area stack by FueCell Energy (FCE) with a [TRL](#) of 5 and is expected to approach 6 due to the support of the Department of Energy's (DOE) National Energy Technology Laboratory (NETL). European firms such as Elcogen and VTT in Finland are testing r-SOE technology to reach a TRL of 6. Sunfire has also validated a 140 kW r-SOE unit with a TRL of 6. Currently, r-SOE produces hydrogen with a purity of 99.99%, minimum operational load of 10%, energy consumption density of 2.5-3.5 kWh/Nm<sup>3</sup>, power density of 0.65-2 W/cm<sup>2</sup>, voltage degradation rate of 1.5% per kh, stack lifetime 10,000 h and nominal stack efficiency of 80% [109]. The technical specification targets for r-SOE are durability of 40,000 h, power density of >1 W/cm<sup>2</sup>, roundtrip efficiency of 70% AC-AC, capacity size of >1 MW, and stack cost of 200 USD/kW [76].

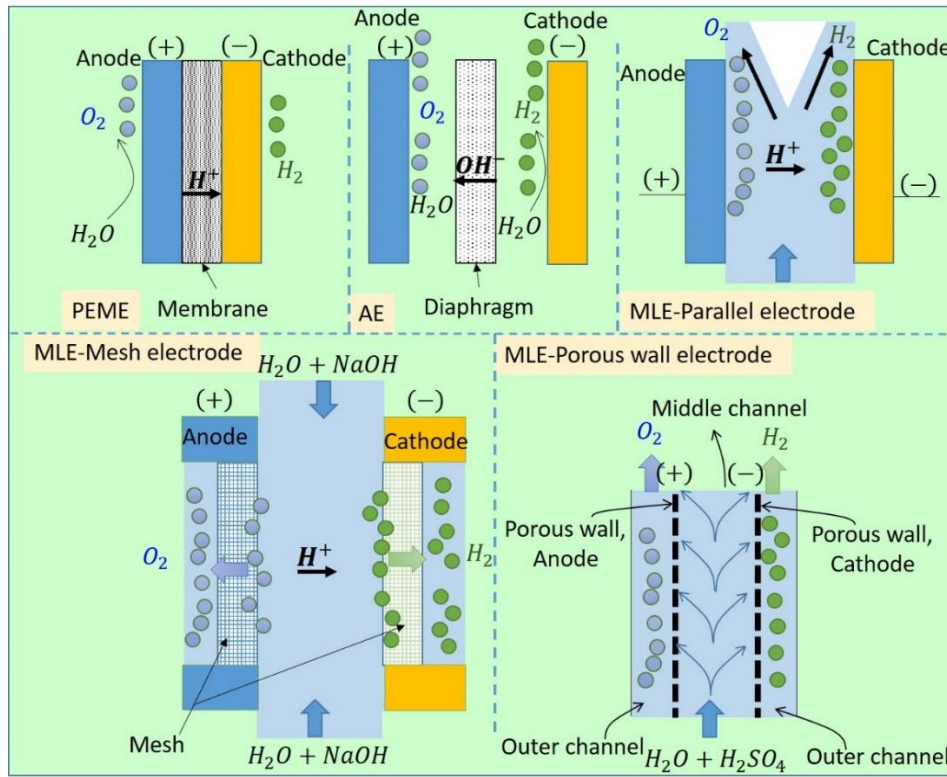
#### 4-1-5- AEME

AEME has a high efficiency ranging from 70%-80% [64], at the technology demonstration level with a [TRL](#) of 6 ( [56], [57], [59], and [64]) and operating temperature and pressure ranges of 20-200 °C and 0.1-1.5 MPa (Table 2). They still need more effort to develop systems and subsystems, test, launch, and commercialize. According to the International Renewable Energy Agency (IRENA) [56] report in 2021 the TRL of AEME was 2-3, now in 2024, it is 6, which shows this technology has attracted attention due to its high efficiency, polymeric membrane, and relatively low operating temperature, making it a promising option for fast startup applications. Replacing the cost-intensive components with non-noble metals is an approach for decreasing LCOH and scale-up purposes [37]. Although both PEME and AE have the potential to be used with offshore wind turbines with a separate dedicated platform, there are concerns about the cost and availability of iridium dioxide (IrO<sub>2</sub>) for PEME and also the need for additional facilities to compensate for the slow cold startup of AE that can increase the LCOH. AEME is highly talented in eliminating these concerns but still needs more research and development to reach the demanded lifetime, efficiency, LC, and scalability [78]. The main challenges of using direct seawater for electrolyzing are reported in [78], among them the fouling and corrosion problems due to seawater contents are major concerns. Some contents can be removed by filtration while some such as dissolved salt and ions can not be removed with filtration. According to the current situation of the electrolyzer technologies in the short to medium term it is advised instead of using seawater directly, to utilize a reverse osmosis desalination system with an ion exchanger to feed the electrolyzer [78]. In an effort, carbon-based electrodes were used for disinfection and GHP from natural seawater with the anion-exchange membrane (AEM; Type-I, Fujifilm, Tokyo, Japan)

between two electrodes, and the results were compared with conventional platinum-based electrodes. The results show that it has lower OPEX and CAPEX than platinum electrodes which can promote commercialization and demonstrate improved stability [80].

#### 4-1-6- MLE

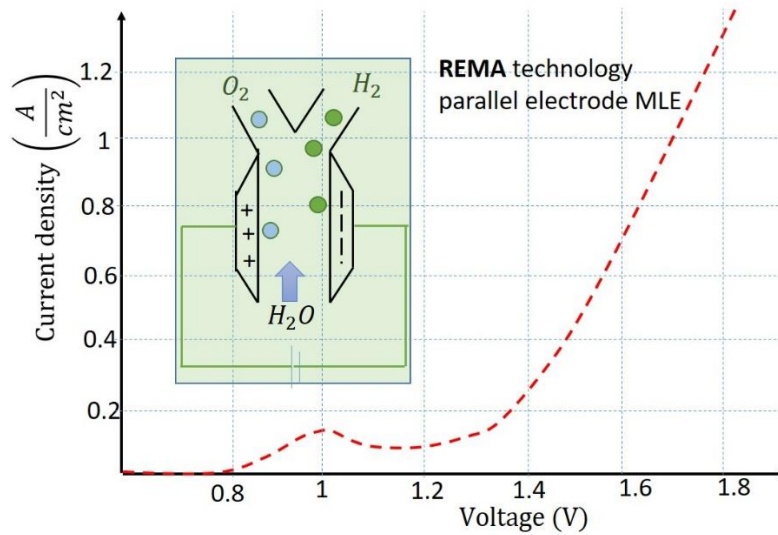
Membraneless Electrolyzer (MLE) with a low [TRL](#) of 3 to 6 ([57], [85], [84], and [72]) is a technology still under laboratory research and development and approaching the demonstration level in some cases. Currently, the economy of GHP is more dominant with the LCOE and capacity factor ([90] and [93]) of renewable electricity. Fortunately, the LCOE from RESs is decreasing rapidly, and the capacity factor can be improved by using hybrid renewable systems. For highly penetrated solar/wind power markets and regions, the price of electricity may even be free for some periods due to excessive production. The US SunShot goal is to reach 3 cents/kWh for renewable electricity in 2030 [95] in which the LCOH of 2 USD/kgH<sub>2</sub> is reachable. The next important impacting parameter is the CAPEX of the electrolyzer. Reducing part numbers, if possible, can reduce MDP, assembly costs, OPEX, and complexity of the system, and as a result, less CAPEX. An MLE uses neither a membrane (such as PEME) nor a diaphragm (such as AE) to avoid mixing the oxygen and hydrogen gases produced during the water-splitting process, it uses the flow of electrolyte solution as an external forced flow to guide the H<sub>2</sub> and O<sub>2</sub> gases out unmixed (Fig.7) [110]. MLE has several advantages over conventional PEME and AE. First, it has lower CAPEX due to eliminating the membrane. This reduces complexity, material costs, assembly cost, and O&M costs. Secondly, they are potent to have high durability and long lifetime, high tolerance due to impurity, and severe operational conditions that may degrade a membrane (such as corrosion, fouling, and blockage). As a result, MLE can operate with low-quality water such as tap water, and the costs for purification, deionizing, and decarbonizing will be saved. Thirdly, MLE can work with a wide range of sufficiently conductive aqueous electrolytes. MLE can operate with acidic, alkaline, and neutral solutions. In conventional electrolyzers, due to utilizing ion-selective membranes, the electrolyte type is limited. The MLE has faced some challenges, including the lower voltage efficiency at high working current densities (>0.5 A/cm<sup>2</sup>), low hydrogen purity (99.4%-99.8%), oxygen and hydrogen crossover, electric arcing between anode and cathode due to applying high voltage across a small gap between electrodes, material related challenges used for the electrodes, and upscaling limitations [85]. Fig.7 represents three MLEs including parallel electrodes, mesh electrodes, and porous electrodes which are compared with conventional PEME and AE. In all three technologies, the electrolyte flow is used to act as the membrane or diaphragm to separate hydrogen and oxygen ([110] and [85]).



**Figure 7. The presentation of hydrogen and oxygen in PEME, AE and MLEs, figures regenerated by the author ([110] and [85])**

In comparison with the parallel design the crossover in the porous wall electrodes is 58 times smaller ([85], and [84]). Several laboratory tests were done [85] for the MLE porous wall electrodes and the outcomes showed that product separation is not perfect, resulting in synthesizing products on the cathode, and a low Faradic efficiency of 43%. This problem is also reported by [84]. In addition, they concluded that steady electrolyte flow is very important, unsteady flow reduces the gas separation efficiency [85].

REMA a company from Swiss has patented a novel parallel electrode MLE with a capacity of 2.5 kW and has planned to produce it by 2025 (Fig.8). The current density versus voltage of this MLE is shown in Fig.8. The stack is supposed to reach an efficiency of 90%. By considering the subsystems, the system efficiency will be 71%. The hydrogen purity is 99.95% which shows low quality compared to PEME and AE and downstream purification is inevitable for applications that require high-purity hydrogen [72].



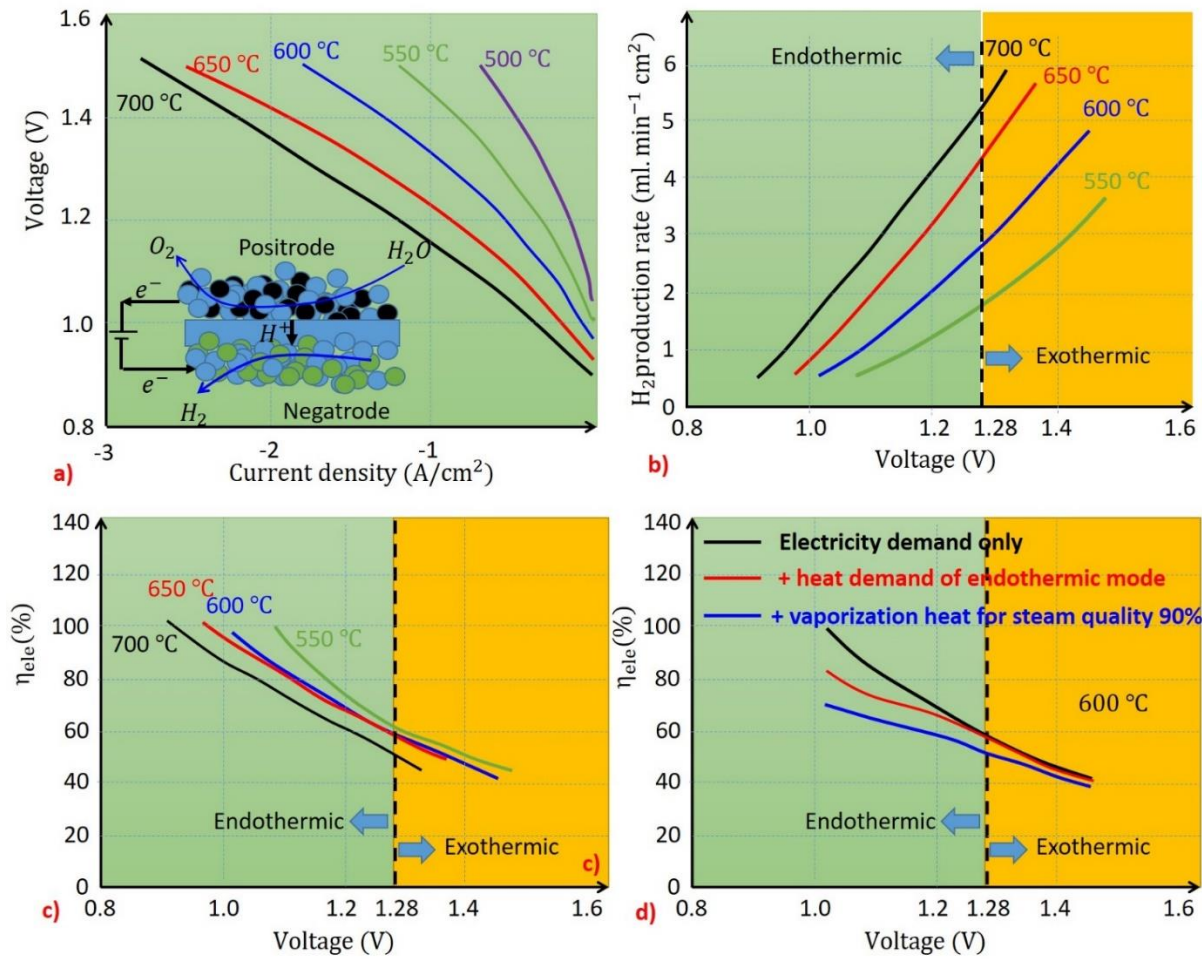
**Figure 8. The current density versus voltage of MLE-parallel electrode by REMA [72], the figure is regenerated by the author**

The potential of MLE to operate with low-quality water has attracted attention. HYLite, sHYp, and Fieldlab Industrial Electrification intend to develop an MLE that uses seawater. Their favorite design is the mesh electrode type in which gas products are generated merely on the outer surface of the electrodes. When the gas bubbles get large, their buoyancy force separates them from the electrodes and then moves upward to the collection containers. In the first step, they planned to develop a 1kW mid-scale prototype to provide technical, economic, and legal feasibility and test its efficiency. By doing this they will reach a [TRL](#) of 4 to 5. After that, a pilot-scale prototype with a [TRL](#) of 7 and a capacity range of about 5 to 10 kW will be developed [111].

#### 4-1-7- PCCE

With a [TRL](#) of about 3 [58], Protonic Conductive Ceramic Electrolyzer (PCCE) still is far from large-scale hydrogen production with high efficiency and purity. The impact of increasing operational pressure from 2.1 to 12.6 bars at a constant temperature of 550 °C on PCCE performance was investigated experimentally [81] and revealed that ohmic and polarization resistances decreased by 33% and 60% which means a 40% decrease in the overall over-potential to run the electrolyzer at current density of 0.5 A/cm<sup>2</sup>. Such improvement means enabling higher current densities at smaller driving voltages. In addition, the Faradic efficiency increased as the pressure was elevated and at a pressure of 5 bars, it reached 100% while at 1 bar it was 50-60% depending on the current density. The electrolyzer tested by [81] was a reversible PCCE (r-PCCE) that was also tested in the fuel cell mode. Ba-based r-PCCE was also studied by [82] under the operating temperature of 700 °C for the current density of 0.5 A/cm<sup>2</sup> and voltage of 1.3 V. The electrolyzer could operate reliably for more than 3500 h with an electrical degradation of 2% per kh. The materials used

in the electrolyte, anode, and cathode of the r-PCCE tested by [81] and [82] are reported in Table 2. A high degradation rate from 1 to 2% per kh and a short lifetime in comparison with other electrolysis technologies is a challenge that needs more research, innovations, and laboratory tests. Another important challenge is the electronic leakage that endangers the PCCE's performance for producing dry, and pure hydrogen with a high efficiency. The current leakage during electrolysis causes the reduction of the effective current for proton reduction at the negatrode ( [82] and [52]). A composite positrode based on  $\text{BaGd}_{0.8}\text{La}_{0.2}\text{Co}_2\text{O}_{6-\delta}$  (BGLC1082) and  $\text{BaZr}_{0.5}\text{Ce}_{0.4}\text{Y}_{0.1}\text{O}_{3-\delta}$  (BZCY541) was used by [52] and the experiments showed high reliability during 1.68 kh at a current density of  $0.8 \text{ A/cm}^2$  which is close to thermoneutral cell voltage at  $600 \text{ }^\circ\text{C}$ . The cell was tested for both working modes of electrolyzer and fuel cell. As an electrolyzer, the tests were conducted for an intermediate temperature range of  $500$  to  $700 \text{ }^\circ\text{C}$ . The specific electricity consumption was  $54$  to  $66 \text{ kWh/kgH}_2$  [52] which is close to the specific energy consumption of low-temperature operating electrolyzers such as PEME and AE ( [75], [76], and [77]). The results also showed that decreasing temperature will increase the Faradic efficiency at endothermic conditions. Other vital challenges to overcome for improving the Faradic efficiency are optimizing materials, cell design, and operating strategy [52]. The voltage versus current density for different temperatures is shown in Fig.9a [52]. It shows that as the temperature decreases the total current density increases while the share of current leakage from the electrolyte is mitigated and consequently, the power density reduces and the electrolyzer efficiency will increase. Furthermore, reducing temperature will decrease the hydrogen production rate per unit of surface area (Fig.9b). In Fig.9c and 9d the electrolyzer efficiency is compared for different temperatures and also when the heat demand of endothermic mode and vaporization heat of input steam are considered or neglected. It shows that if the required heat for the endothermic and vaporization heat are included as the input energies to the PCCE the efficiency will decrease. However, at higher voltages, since the reaction is exothermic and less vaporization heat is required, the efficiency for the three cases approaches a single value.



**Figure 9.** The specifications of PCCE, positrode are based on  $BaGd_{0.8}La_{0.2}Co_2O_{6-\delta}$  (BGLC1082) and  $BaZr_{0.5}Ce_{0.4}Y_{0.1}O_{3-\delta}$  (BZCY541), negatrode is Ni-BZCY541, and the electrolyte is BZCY541 a) Polarization curves for steam electrolysis mode of r-PCCE with 30% steam/air supplied to the positrode and 20%  $H_2/N_2$  to the negatrode b) Hydrogen production per minute per square centimeter versus voltage at different temperatures in the range of 550 to 700 °C in PCCE, c) Calculated PCCE efficiency, assuming free heat source for water vaporization (only electricity is considered as the input energy for water splitting). d) electrolyzer efficiency at a temperature of 600 °C based on three assumptions. [52], the figures are regenerated by the author.

A. V. Kasyanova et al. reviewed different materials used for protonic conductive ceramic cells used in the fuel cells and electrolyzers to seek the limitations and potential solutions [86]. They reviewed the research from 2013 to 2022 and concluded that PCCEs are suitable for both portable and stationary applications due to their efficient operation at a mid-temperature range of 400-700 °C and even lower [86]. PCCE has shown outstanding laboratory-scale electrochemical performance. However the full-scale operation of this technology is scarce [96], [58]. Scale-up and stack integration of r-PCCE was developed by [96] from button-cells to small-stack. The materials used for the electrolyte, cathode, and anode were

BaCe<sub>0.4</sub>Zr<sub>0.4</sub>Y<sub>0.1</sub>Yb<sub>0.1</sub>O<sub>3-δ</sub> (BCZYYb), BaCo<sub>0.4</sub>Fe<sub>0.4</sub>Zr<sub>0.1</sub>Y<sub>0.1</sub>O<sub>3-δ</sub> (BCFZY), and a nickel-electrolyte composite. The power density of the stack reached 0.69 W.cm<sup>-2</sup> in fuel cell mode for hydrogen input at 600 °C. To reduce the degradation rate in electrolyzer mode to 1.5 % per kh at the current density of 0.1 A.cm<sup>-2</sup> and temperature of 550 °C. and a gadolinium-doped ceria cathode-electrolyte interlayer was used. Later in 2022, the degradation of the r-PCCE was investigated and it became <1% per kh at 50% steam and 550 °C [58]. The PCCE can produce pure, dry hydrogen at high pressure, omitting the compression system from the downstream of the electrolyzer and thus reducing the system's complexity. PCCE with BCZYYb electrolyte shows higher ionic conductivity when transferring protons than O<sup>-2</sup>-ion conducting devices, as a result, they have higher efficiency at temperatures lower than SOE. The higher operating temperature of PCCE results in higher efficiency than PEME and lower LCOH [58]. The reversibility of PCCE was investigated by [83]. For this purpose, a triple proton conducting oxide of PrNi<sub>0.5</sub>Co<sub>0.5</sub>O<sub>3-δ</sub> perovskite was developed for operating in the temperature range of 400 to 600 °C. The hydrogen generated by the electrolyzer mode was converted to electricity in the fuel cell mode effectively without requiring any additional hydrogen and showed that this technology can be sustainable for power and hydrogen production [83].

#### 4-1-8- Miscellaneous electrolysis

*Reverse Electrodialysis (RED)*: GHP by using a RED and Assisted RED (ARED) electrolyzer was done by A. Pellegrino et al. [112] experimentally and a preliminary economic evaluation shows that the minimum LCOH by ARED is about  $3.2 \frac{\text{€}}{\text{kgH}_2} \cong 3.47 \frac{\text{USD}}{\text{kgH}_2}$  for the maximum salt concentration of  $300 \frac{\text{g}}{\text{L}}$ . The ARED stack comprises 20 cell pairs of commercial Fujifilm Type 10 ion-exchange membrane (AEM) and Tilburg, the Netherlands cation exchange membrane (CEM), and 300 μm thick woven spacers. An important advantage of ARED is its significantly lower current density ( $0.002\text{-}0.1 \frac{\text{A}}{\text{cm}^2}$ ) [112] in comparison with conventional electrolyzers such as PEME ( $0.6\text{-}2 \frac{\text{A}}{\text{cm}^2}$  [63]), AE ( $0.2\text{-}0.8 \frac{\text{A}}{\text{cm}^2}$  [63]), and SOE ( $0.95\text{-}1.3 \frac{\text{A}}{\text{cm}^2}$  [63]). In addition, the cell voltage and specific electricity consumption of the ARED depending on the salt concentration ( $35 \text{ to } 300 \frac{\text{g}}{\text{L}}$ ) changes from 0.02 V to 3.1 V and  $0.6 \frac{\text{kWh}}{\text{kgH}_2}$  to  $85.7 \frac{\text{kWh}}{\text{kgH}_2}$  [112]. According to the voltage and specific electricity consumption range, it is evident that there is a vast opportunity for optimization of this type of electrolyzer. The current status of this technology is in the first laboratory application with TRL of 1-2.

*Biodegradable electrolysis*: Recently, another research introduced an electrocatalyst for recycling dye wastewater while producing green hydrogen. They used an eco-friendly biodegradable, solvent-free, direct

method to fabricate cellulose paper-based electrodes enclosed with WSe<sub>2</sub>-CuO electrocatalyst for GHP from pure water and dye wastewater. The stability test was done for pure water for 20 h at a high current density of up to  $0.170 \frac{A}{cm^2}$ . The anode was made of an inexpensive and earth-plentiful graphite rod and achieved above 95% decontamination efficiency on the anode side [113].

## 4-2- Photolysis

Photolysis is the water-splitting by direct use of light absorbed by a photocatalyst. In other words, instead of converting sunlight to electricity in a photovoltaic system, and then using electricity to split water molecules in an electrolyzer, light is directly absorbed by a semiconductor electrode called photoelectrode, or photocatalyst particles dispersed in water or deposited on a substrate. When the photocatalyst is illuminated due to receiving enough photon energy ( $h\nu$ ) (the received energy must be equal to or greater than the semiconductor bandgap), an electronic charge carrier will form and make holes in the valence band, and electrons in the conduction band. The generated holes  $h^+$  react with water molecules on the interface between the electrolyte and photoelectrode [51] to form oxygen and hydrogen ions ( $H^+$ ), the  $H^+$  is transferred to the cathode through the electrolyte while the electrons pass to the cathode through the external circuit reacting with  $H^+$  and reducing it to  $H_2$  (Fig. 10). Photolysis using photocatalyst particles is called photocatalytic water-splitting (PC-WS); while water-splitting by photoelectrode is called photoelectrochemical water splitting (PEC-WS) [114].

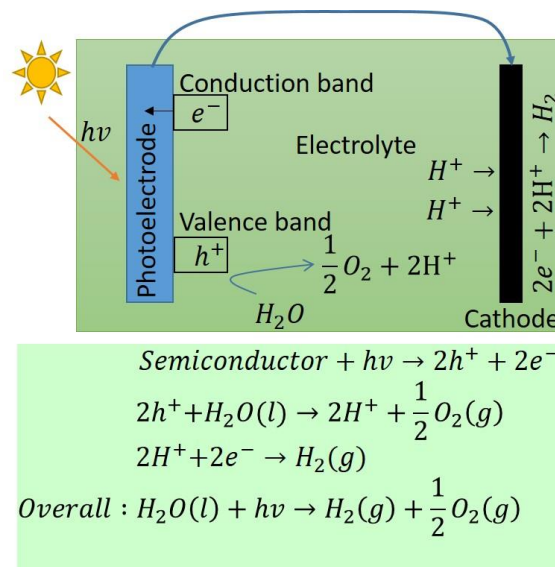


Figure 10. The schematic of the photoelectrolysis water-splitting system and the corresponding reactions

The reversible potential for water electrolysis under standard conditions is  $\Delta E^0 = -\frac{\Delta G^0}{nF} = 1.23 \text{ V}$  in which  $\Delta G^0$  is the change in standard Gibbs free energy,  $\Delta E^0$  is the standard electric potential of the reaction,  $n$  is the number of electrons, and  $F$  is the Faraday constant. The overall chemical reaction in Fig.10 happens when  $h\nu \geq 1.23 \text{ eV}$ . In practice, mostly the photovoltage created by the photons between the electrodes is less than 1.23 V which can be compensated by using an external electrical bias or internal chemical bias [51]. When determining the [STH](#) conversion efficiency or simply [photolysis efficiency](#), the bias voltage must be subtracted from the output power. For significant PEC-WS the valance and conduction band edges of semiconductor materials must overlap, the photoelectrode must remain stable while photolysing, and to avoid corrosion and create overpotential losses charge transfer from the semiconductor surface must be fast enough [51].

A sacrificial agent typically works as an electron donor to prohibit the reverse reactions. Since it has a small positive oxidation potential, reacting with the photo-generated holes and consuming them becomes easy, effectively preventing the fast recombination of electrons and holes. This allows the polymer surface to collect more photo-generated electrons. Sacrificial agents exist as reducing or oxidizing types. The most well-known reducing ones are triethanolamine (TEOA), ascorbic acid (AA), methanol, sulfide, and ethylene diamine tetraacetic acid (EDTA). Examples of oxidizing sacrificial agents are  $\text{Ag}^+$  and  $\text{Fe}^{3+}$  [115]. Consumption of sacrificial agents usually produces useless chemicals, bringing us new concerns about cost and environment. Due to this problem using sacrificial reagents remains a challenge [116]. Co-catalysts have a vital role in optimizing the catalytic characteristics of the main active materials. The primary purpose of a co-catalyst, particularly in photocatalysis, is to proficiently separate photo-generated charge carriers, such as electrons and holes, thus reducing the possibility of recombination. Remarkably, conductive inorganic nanosheets demonstrate potential as co-catalysts [117].

An economic assessment of hydrogen production by PC-WS, PEC-WS, direct bio-photolysis, and indirect bio-photolysis was done for the Netherlands and it was revealed that LCOH is 18.32 \$/kgH<sub>2</sub>, 18.98 \$/kgH<sub>2</sub>, 18.45 \$/kgH<sub>2</sub>, and 36.39 \$/kgH<sub>2</sub> respectively while it was 9.31 \$/kgH<sub>2</sub> for PV-based electrolysis. The reported STH efficiency of PC-WS methods is usually less than 2 %, while for the PEC-WS, it is 9 to 16 %. Although providing materials and production steps of a PC-WS (before hydrogen production) will emit some emissions, the LCA of this technology is scarce and mostly they are considered as zero emission. However, a techno-economic assessment was done for different scales of PEC-WS and estimated the pollution index of PEC-WS with an assumed STH efficiency of 6.2% for a silicon cell to be 1.64 kgCO<sub>2</sub>-eq/kgH<sub>2</sub> [118]. In addition, they are more compact in comparison with PV-based electrolyzers and need less

space but the risk of contact between photon absorber particles with the electrolyte remains a great risk for their stability [114]. The minimum test time for stable operation of PEC-WS and PC-WS is considered to be 50 h [114]. [TRL](#) of photolysis methods is 5 because it has not been implemented commercially yet. Most of the results are based on small-scale laboratory prototypes [114]. However, among the recent research, there are claims of the potential scale-up of some photocatalysts ([119] and [120]). Since photon-driven hydrogen production methods are not commercially available their LCOH is still high, and the economic assessments are just based on laboratory-scale experiments.

Metal-organic framework materials (MOFs) and their derivatives have become popular for hydrogen production by photocatalytic. Forming composite co-catalysts consisting of NiS and CdS is possible by the employment of Ni-MOF as a precursor in a thiourea solution containing  $Cd^{2+}$ . This process makes the formation of composite catalysts with homogeneously distributed metal ions possible. DUT-67 has proven its potential as a talented carrier to create an efficient catalyst [121]. Bi Y. et al. prepared a new ternary photocatalyst NiS/CdS@DUT-67 for hydrogen production and achieved  $9618 \mu mol_{H_2} \cdot g_{NiS/CdS}^{-1} \cdot h^{-1}$ . The hydrogen production by the ternary NiS/CdS@DUT-67 is 32 and 2.5 times that for unary CdS and binary NiS/CdS respectively. It is also more stable than CdS and NiS/CdS for long-term photocatalysis. They concluded that NiS and DUT-67 act as dual catalysts for CdS and enhance the charge separation during photocatalysis. They showed the advantages of using Ni-MOF derivatives for higher and more stable hydrogen production [121].

Due to the critical impact of expensive metals such as Pt on the cost of photocatalysts, researchers try to eliminate or reduce their share in the formation of photocatalysts without losing performance. Nanodot-carbon nitride ( $C_3N_4$ ) as a photocatalyst has attracted much attention. It is a metal-free visible light-responsive semiconductor material, it possesses exceptional chemical stability, proper band gap, and energy level position, it is also environmentally friendly, and easy to manufacture. Under the experiment, a high [apparent quantum efficiency \(AQY\)](#) of 16% at  $\lambda = 420 \pm 20$  nm and STH of 2% was achieved [122].

Vasilchenko D. et al. [119] utilized Graphitic Carbon Nitride ( $g-C_3N_4$ )-based nanostructures with a low content of Pt from 0.01 to 0.5 wt% to produce a highly active photocatalyst for hydrogen production. The process involved three main steps thermolysis of melamine cyanurate complex, chemisorption of platinum from the solution, and at the end reductive treatment with  $H_2$ . The hydrogen production performance depended on Pt content, the experiments showed that for 0.05, 0.1, and 0.5 wt % of Pt in the photocatalyst, [hydrogen yield](#) is 3000, 8500, and 11000  $\mu mol \cdot g^{-1} \cdot h^{-1}$ . They also used the hydrogen to run a 1 W PEMFC. They concluded that finding novel, robust, and clear synthetic approaches is the main part of the production

of highly active photocatalytic materials based on Carbon Nitride-based. Furthermore, they evaluated the scale-up potential of the reactor and according to the results, they found it convenient. The Pt content decreased significantly in the photocatalyst obtained by Vasilchenko D. et al. [119], because previous studies usually used 1 to 5 wt% Pt for (g-C<sub>3</sub>N<sub>4</sub>)-based photocatalysts ([123], and [124]). Qin Y. et al. [123] used a simplistic hydrothermal and photo-induced deposition method for forming photocatalysts for hydrogen production. They generated 1299.4  $\mu\text{mol}$  of hydrogen by exposing the 2D/2D WO<sub>3</sub>/Pt/g-C<sub>3</sub>N<sub>4</sub> (WPC) photocatalyst to visible light during 5 h, and the hydrogen yield remained stable for another 2 time periods (totally 15 h) but in the fourth cycle, the performance dropped significantly. The [hydrogen yield](#) was improved due to three parameters; the Schottky Ohmic junction can outstandingly accelerate 3-D charge separation and increase electron lifespan, the 2D/2D structure can reduce the charge travel distance, and Platinum with rich electron concentration can steadily adsorb H<sup>+</sup>. They used 0.5, 1, 1.75, and 2 wt% Pt for optimizing the experiments. 3D porous g-C<sub>3</sub>N<sub>4</sub> assembled by highly crystalline and ultrathin nanosheets (NS) and 3D g-C<sub>3</sub>N<sub>4</sub> NS was formed for direct hydrogen production from water splitting when exposed to visible light created by a 300 W xenon lamp equipped with a 420 nm cutoff filter [124]. They used 1 wt% Pt and 3 wt% IrO<sub>2</sub> as co-catalysts without using a sacrificial reagent. They used the photocatalyst for more than 100 h of hydrogen production during several 6 h working cycles. After each repeating cycle, the hydrogen yield dropped continuously, and after 102 h of water splitting, its hydrogen evolution rate was reduced by 44%, showing severe agglomeration of nanosheets on g-C<sub>3</sub>N<sub>4</sub> NS after the repeating cycles. Another important discussion states that the higher specific surface area is not the primary reason for the highly efficient activity of 3D g-C<sub>3</sub>N<sub>4</sub> NS, it is mainly related to its photogenerated charge carrier separation efficiency [124].

g-C<sub>3</sub>N<sub>4</sub> is a highly talented photocatalytic catalyst for water-splitting and hydrogen production. Still, in practice, it is limited by the rapid recombination of photogenerated carriers and inadequate use of surface charge. To resolve this weakness a succession of g-C<sub>3</sub>N<sub>4</sub>-NiO-NiS<sub>x</sub> photocatalysts were synthesized. NiO was loaded on g-C<sub>3</sub>N<sub>4</sub> and NiO was partially sulfided into NiS<sub>x</sub> through a hydrothermal process in situ [125]. They examined the g-C<sub>3</sub>N<sub>4</sub>-NiO-xT in which x denotes the magnitude of T (thiourea) added in mg. The best sample happened for x=0.4 mg with a hydrogen yield of 1248.1  $\mu\text{mol} \cdot \text{g}^{-1} \cdot \text{h}^{-1}$  which is 347 times bigger than that of the g-C<sub>3</sub>N<sub>4</sub>. According to the stability tests, after repeating the process in 4 cycles and a total of 20 hours, the hydrogen yield did not change significantly and they concluded that the stability was excellent [125], however, since the total test time was less than 50 h, the conclusion is not very solid. Another photocatalyst was prepared by boron/oxygen-codoped porous g-C<sub>3</sub>N<sub>4</sub> nanomesh (BO-C<sub>3</sub>N<sub>4</sub> nanomesh) in two steps of doping and etching approach [126]. The specific contact area of BO-C<sub>3</sub>N<sub>4</sub>

reached  $160.58 \text{ m}^2 \text{ g}^{-1}$  providing a high active area for light absorption and photocatalytic reactions. The codoping of Boron and Oxygen improved the electron transport capability and increased the lifespan of charge carriers. The hydrogen yield reached  $9751 \text{ } \mu\text{mol} \cdot \text{g}^{-1} \text{ h}^{-1}$  and was stable during 4 cycles and a total test time of 20 h. The AQY reached 8.1% at 420 nm. They believe that their method of preparing photocatalysts can be commercialized [126], however, they need to extend the stability test for a minimum of 50 h.

A critical obstacle in improving photocatalysis is the efficient transferring of photoelectrons ( $\text{e}^-$ ) and photoholes ( $\text{h}^+$ ) for Redox reactions on the same catalyst. To solve this problem during the last 20 years co-catalysts or sacrificial reagents have been used in most of the photocatalysts made. However, Majji M et al. [120] investigated an inexpensive, scalable, and new synthesis process for non-metallic single-element catalysts without adding Pt-based expensive co-catalysts, or sacrificial reagents. They introduced crystalline elemental Boron nanostructures through a scalable and typical synthesis method [120]. This photocatalyst was prepared in three different samples A, B, and C, in which sample B with the highest hydrogen yield of  $15120 \text{ } \mu\text{mol} \cdot \text{g}^{-1} \text{ h}^{-1}$  operated stably for 72 hours with a Solar-to-Hydrogen efficiency of 1.5%. In another work, two 2D metal-free materials of boron nanosheets (BNS) and  $\text{g-C}_3\text{N}_4$  nanosheets were coupled via tight heterojunction architecture (3.0B-CN) [127]. This photocatalyst enhanced the absorption of visible light, caused fast charge transfer, and increased carrier concentration resulting in a significant hydrogen yield of  $19910 \text{ } \mu\text{mol} \cdot \text{g}^{-1} \text{ h}^{-1}$  which is 35 times that for the  $\text{g-C}_3\text{N}_4$ . They examined the photocatalyst for different weight percentages of BCN from 0.5% to 5% and achieved the highest hydrogen yield at 3 wt% of BCN which is 35 times  $\text{g-C}_3\text{N}_4$ , and 226 times BNS alone. The stability test of the photocatalyst was examined for 4 cycles and a total of 12 h, in the last cycle hydrogen yield dropped slightly which may be due to photocatalyst loss during sample recovery [127]. In another experiment, an all-organic Z-scheme hydrogen-bonded organic frameworks (HOFs)/ $\text{g-C}_3\text{N}_4$  nanosheets (CNNS) heterojunction photocatalyst was synthesized via an in-situ electrostatic method and fabricated for WSHP [128]. They reported a hydrogen yield of  $4450 \text{ } \mu\text{mol} \cdot \text{g}^{-1} \text{ h}^{-1}$  with an [apparent quantum efficiency](#) of 22% at 450 nm which is about 11 times higher than CNNS.

Not using a sacrificial agent in photocatalyst formation for water splitting is challenging. Another attempt to form a 2D ultra-thin nanosheet of a Pt-loaded  $\text{g-C}_3\text{N}_4$  without sacrificial reagent for photocatalytic water splitting was done by Wu et al. [129]. The Pt loading as co-catalyst was 1.4 wt% and the hydrogen and oxygen yields were 42.6 and  $18.7 \text{ } \mu\text{mol} \cdot \text{g}^{-1} \text{ h}^{-1}$ . They used pulsed laser exfoliation 4 with a femtosecond pulsed laser (150 fs) centered at 805 nm with an average power of 340 mW instead of visible light. As can be seen, hydrogen yield and AQY are very low compared to other references presented in Table 4. Another

photocatalyst for hydrogen production under visible light is the ZnO-CuO-Au S-scheme heterojunction [130] integrated with the surface plasmon resonance effect. They used sol-gel to design the heterojunction. They designed different experiments to investigate the impact of different photocatalysts including ZnO, CuO-ZnO, Au-ZnO, and ZnO-CuO-Au and they achieved hydrogen yield of 24, 930, 1455, and 4655  $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$  respectively for 4 h exposure to visible light. In addition, they investigated the impact of sacrificial agents including Glycerol, TEOA, Ethanol, Methanol, and Ethylene glycol, and achieved hydrogen yields of 4655, 4544, 3990, 4183, 4360 respectively for ZnO- CuO-Au. They evaluated the impact of vol% of glycerol on the hydrogen yield as well and for 5, 10, 15, and 20 vol% of glycerol they gained 4130, 4655, 4924, 4567  $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$  respectively. They repeated the experiment for 5 cycles of 4 h, and the stability was 90% after the 5<sup>th</sup> cycle. The catalyst dose is another impacting parameter on the hydrogen yield. They examined 50, 100, and 15 mg of S- scheme ZnO- CuO-Au and gained hydrogen yields of 4320, 4655, and 4263  $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$  respectively. In conclusion, Au-bridged S-scheme ZnO-CuO nanoparticles are effectively synthesized through the sol-gel technique [130].

In another innovative GHPM b using PEC technology ZnO thin film was used under 150 W halogen light with 0.1 M phosphate buffer solution (PBS) and 0.1 M NaOH solution. The stability test was done for 4 cycles and the ZnO thin film with 0.1 M PBS achieved a performance of 75-80% with a good stability during the cycles. However, the ZnO thin film with NaOH solution achieved a performance of about 37% in the first cycle and dropped dramatically in the fourth cycle to about 5% [131].

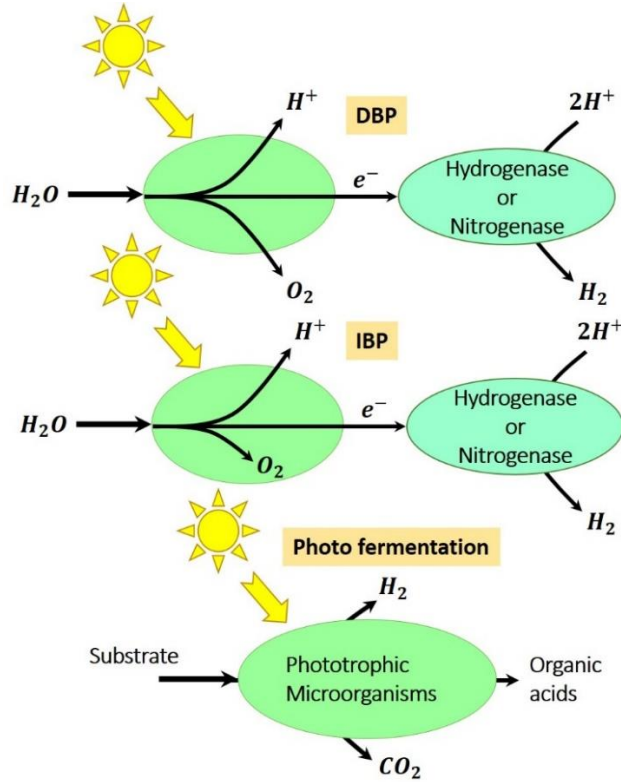
**Table 4. The comparison of different PEC-WS and PC-WS photocatalysts**

| Ref.  | Photocatalyst samples                                                                                                                                            | <a href="#">H<sub>2</sub> yield</a><br>( $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$ ) | Light source                               | Sacrificial agent | Solvent              | Co-catalyst                         | <a href="#">Photolysis (STH) efficiency /AQY</a> | <a href="#">Stability, run-time</a> | Scale-up potential |
|-------|------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------|--------------------------------------------|-------------------|----------------------|-------------------------------------|--------------------------------------------------|-------------------------------------|--------------------|
| [121] | NiS/CdS@DUT-67<br>NiS/CdS                                                                                                                                        | 9618<br>3847.2                                                                               | 300 W xenon lamp                           | MOF               | DI water and ethanol | CdS@DUT-67 (dual)<br>CdS (single)   | Not reported (NR)/NR<br>NR/NR                    | 5 h / 10 cycles                     | Not claimed        |
| [119] | Pt/g-C <sub>3</sub> N <sub>4</sub><br>0.5 wt%Pt<br>0.1 wt%Pt<br>0.05 wt%Pt<br>0.01 wt%Pt                                                                         | -<br>11000<br>8500<br>3000<br>Trace                                                          | 30 W LED, 380-nm                           | TEOA              | Ethanol and DI water | platinum nitrate-complexes          | NR/ 5% at 400 nm                                 | 2 h / NR                            | Claimed            |
| [123] | W <sub>5</sub> Pt <sub>1.75</sub> C(WPC)<br>W <sub>5</sub> CPt <sub>1.75</sub> (WCP)<br>Pt <sub>1.75</sub> /CN<br>CN<br>WO <sub>3</sub>                          | 5197.2<br>4477.6<br>3219.5<br>452.8<br>Trace                                                 | 300 W Xe-lamp with a 420 nm cutoff filter  | 20 vol% TEOA      | DI water and ethanol | 0.5-2 wt% Pt                        | NR / 7.8% at 420 nm                              | 5 h / 4 cycles                      | Not claimed        |
| [124] | 3D g-C <sub>3</sub> N <sub>4</sub> NS-2<br>3D g-C <sub>3</sub> N <sub>4</sub> NS-1<br>g-C <sub>3</sub> N <sub>4</sub> NS<br>bulk g-C <sub>3</sub> N <sub>4</sub> | 101.4<br>44<br>20<br>8.7                                                                     | 300 W Xenon lamp, ( $\lambda \geq 420$ nm) | Not added         | DI water             | 1 wt% Pt and 3 wt% IrO <sub>2</sub> | 0.06%/ 1.4% at 420 nm                            | 6 h / 17 cycles                     | Not claimed        |
| [120] | Boron NS<br>Samples B<br>Sample C<br>Sample A                                                                                                                    | -<br>15120<br>7812<br>5708                                                                   | 300 W Xenon lamp                           | Not added         | DI water             | Not added                           | 1.5%/ NR<br>1%/ NR<br>1%/ NR                     | 12 h / 6 cycles                     | Claimed            |

|       |                                                                                                                                                                               |                                                                       |                                                                                                                        |                  |          |                                                         |                     |                |                                                                                                    |
|-------|-------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------|------------------|----------|---------------------------------------------------------|---------------------|----------------|----------------------------------------------------------------------------------------------------|
| [127] | 2D/2D Boron / g-C <sub>3</sub> N <sub>4</sub><br>3 wt% B-CN<br>BNS<br>g-C <sub>3</sub> N <sub>4</sub><br>0.5 wt% B-CN<br>1 wt% B-CN<br>2 wt% B-CN<br>4 wt% B-CN<br>5 wt% B-CN | -<br>-<br>19910<br>88<br>570<br>1910<br>4125<br>7490<br>10731<br>4250 | 300 W commercial Xenon light source (PLS-SXE300)                                                                       | TEOA             | DI water | Chloroplatinic acid (H <sub>2</sub> PtCl <sub>6</sub> ) | NR/NR               | 3 h / 4 cycles | Not claimed, but it is mentioned as a pioneer for producing nonmetal photocatalysts in the future. |
| [128] | (HOFs)/g-C <sub>3</sub> N <sub>4</sub> nanosheets (CNNS)                                                                                                                      | 4450                                                                  | 300 W xenon lamp ( $\lambda > 420$ nm)                                                                                 | TEOA             | DI water | Not added                                               | NR / 22% at 450 nm  | 8 h / 8 cycles | Not claimed                                                                                        |
| [129] | g-C <sub>3</sub> N <sub>4</sub> ultra-thin nanosheets                                                                                                                         | 42.6                                                                  | Pulsed laser exfoliation 4 with a femtosecond pulsed laser (150 fs) centered at 805 nm with an average power of 340 mW | Not added        | Methanol | Divalent atomic Pt                                      | NR / 23% at 400 nm  | 9 h / 3 cycles | Not claimed                                                                                        |
| [130] | ZnO-CuO-Au<br>Au-ZnO<br>CuO-ZnO<br>ZnO                                                                                                                                        | 4924<br>1455<br>930<br>24                                             | 300 W xenon lamp ( $\lambda > 400$ nm)                                                                                 | 15 vol% Glycerol | DI water | Not added                                               | NR/NR               | 4 h / 5 cycles | Claimed                                                                                            |
| [125] | g-C <sub>3</sub> N <sub>4</sub> -NiONiSx                                                                                                                                      | 1248.1                                                                | 300 W xenon lamp ( $\lambda > 420$ nm)                                                                                 | TEOA             | DI water | Not added                                               | NR/NR               | 5 h / 4 cycles | Not claimed                                                                                        |
| [126] | BO-C <sub>3</sub> N <sub>4</sub><br>O-C <sub>3</sub> N <sub>4</sub><br>g-C <sub>3</sub> N <sub>4</sub>                                                                        | 9751<br>4720<br>345                                                   | 300 W xenon lamp ( $\lambda > 420$ nm)                                                                                 | TEOA             | DI water | Pt 3wt% (H <sub>2</sub> PtCl <sub>6</sub> )             | NR / 8.1% at 420 nm | 4 h / 5 cycles | Claimed                                                                                            |

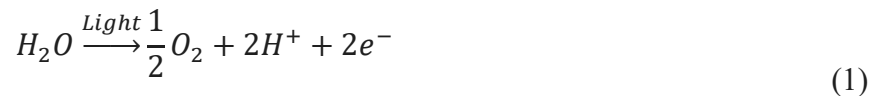
### 4-3- Bio-Photolysis (BP)

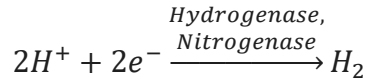
BP is an environmentally friendly water-splitting process using photon energy and biological activities. It involves renewable components such as plants, sunlight, and water without hazardous wastes. BP can be done directly (DBP), indirectly (IBP), or by photofermentation (Fig. 11) [132].



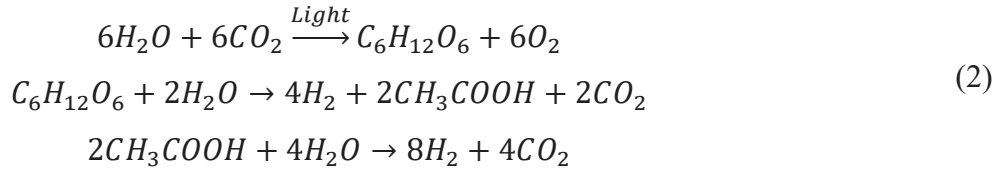
**Figure 11. Light-dependent DBP, IBP, and photo fermentation pathways for biohydrogen production by green algae or cyanobacteria, figures regenerated by the author [133]**

DBP is one-step water splitting with light energy and biological activities without the involvement of intermediates under aerobic conditions. Green algae and some specific unicellular cyanobacteria as the photoactivate materials evolve hydrogen through DBP because they contain reversible hydrogenase and nitrogenase enzyme as the photocatalyst. In this process, microorganisms use photosystems (PS II and PS I) to absorb light. Light excites an electron within the PS II and then is transferred to ferredoxin (FD) via PS I. FD passes the electron to the hydrogenase or nitrogenase enzymes within algae cells to reduce hydrogen ion ( $H^+$ ) and produce  $H_2$  (Eq.1) [134].





IBP on the other hand is the water splitting with two or more steps. IBP can be done using hydrogenase or nitrogenases in green algae and cyanobacteria. Carbohydrates such as glucose, starch, and glycogen are prepared via the photosynthesis of CO<sub>2</sub> in the presence of water and sunlight in the first step. In the second step, carbohydrates are consumed due to the occurrence of the process catalyzed by the hydrogenase or nitrogenase enzymes. Due to the consumption of carbohydrates, CO<sub>2</sub> and H<sub>2</sub> will be produced (Fig.3). That means CO<sub>2</sub> is only recycled and no accumulation of CO<sub>2</sub> happens [51]. IBP requires the least nutrients [134], but only a few microalgal species perform it [135].



DBP and IBP face serious challenges in becoming commercially available as hydrogen production methods. The main challenge is their small [STH conversion efficiency](#). The major reason for the low STH efficiency of BP is that microalgae can only use radiation in the wavelength range of 400 to 700 nm for hydrogen production which accounts for 43 % of the solar radiation. This wavelength region is called Photosynthetically Active Radiation (PAR) [51]. The average STH of DBP is about 10 % and for IBP is 9.5 % mostly in laboratory scales under AM 1.5 Spectrum which is significantly smaller than that for PEC-WS with a mean STH conversion efficiency of about 18% [114]. The TRL for DBP and IBP is 4 since these methods have just passed the “Concept” level and are being tested in the laboratory [114]. The LCOH produced by DBP and IBP is estimated to be 18.45 USD/kgH<sub>2</sub> and 36.39 USD/kgH<sub>2</sub> for the Netherlands [114]. The LCOH by DBP is close to the values reported for PC-WS (18.32 USD/kgH<sub>2</sub>) and PEC-WS (18.98 USD/kgH<sub>2</sub>) but approximately two times that of PV-Electrolysis (9.31 USD/kgH<sub>2</sub>) [114].

The high cost of biohydrogen production is still a challenge, a research [132] on hydrogen production by microalgae showed hydrogen yield and STH efficiency of biophotological and electrochemical processes are low, and further process development is required. In addition, they estimated the average cost of biohydrogen production by DBP, IBP, and photofermentation to be around 7.24 USD/kgH<sub>2</sub>, 7.54 USD/kgH<sub>2</sub>, and 7.61 USD/kgH<sub>2</sub> which is still high and they recommended more efforts more efforts to make it economical.

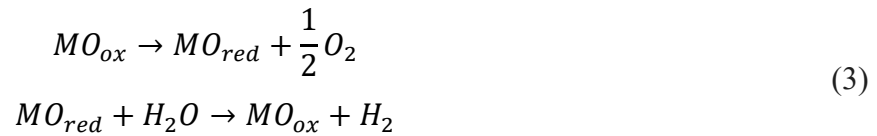
The purity of hydrogen production by DBP can reach 98 %. In comparison with IBP, it has lower LCOH. For efficient hydrogen production DBP needs a large-scale algae cultivation to absorb enough light. This means consuming more land [133]. Experiments have shown that co-culturing bacteria with green algae has improved hydrogen yields more than 7.5 times compared to a pure algae culture ( $150 \text{ mL}_{\text{H}_2} \cdot \text{L}^{-1}$ , compared to  $20 \text{ mL}_{\text{H}_2} \cdot \text{L}^{-1}$  in 17 days). In addition, it has slowed the reduction of chlorophyll, enhanced starch buildup, preserved protein content, and as a result improved hydrogen yields [136]. Oxygen scavenging, mineral deprivation, and co-culturing enhance hydrogen production. Optimizing process parameters such as PH, light absorption, temperature, and cell density can increase hydrogen and reduce oxygen concentrations. Nanoparticles and antioxidants effectively control process inhibitors, reduce substrate, and enhance hydrogen yields. Large-scale outdoor cultivation of BP hydrogen production is possible only for some seasons or regions. Furthermore, biohydrogen farm treatment and harvesting on a large scale is highly energy-intensive and complicated. Using oxygen regulation techniques (mineral deprivation, oxygen scavengers, and co-culturing), antioxidants, and nanoparticles are among the key methods to make BP hydrogen production economically viable [137].

To improve hydrogen production, activated sludge bacteria and *Chlorella vulgaris* are co-cultured to decrease oxygen concentration while increasing hydrogen production. In this process, activated sludge bacteria act as oxygen consumers while algae produce hydrogen [138]. According to the results, co-culturing has increased hydrogen production and oxygen consumption compared with the cases that used pure bacteria cultures. Hydrogen yield dramatically increased to  $1246 \text{ mL}_{\text{H}_2} \cdot \text{L}^{-1}$  in 6 days for algae to bacteria volumetric ratio of 1:1.5 while oxygen and methane content reached a minimum value of  $57 \text{ mL}_{\text{O}_2} \cdot \text{L}^{-1}$  and zero  $\text{mL}_{\text{CH}_4} \cdot \text{L}^{-1}$ . In addition, they reported that at PH of 4 hydrogen production has stopped [138].

Despite significant development in bio-photolysis-based HPSs, large-scale hydrogen production using these methods has faced critical challenges such as high LCOH and low hydrogen yield. Using promoters, and their working mechanisms as one of the main methods for improving BP-based hydrogen production methods needs more investigation. Their impact on biochemical and molecular levels and their influence on the metabolic processes must be explored. Compared to bulk usage nano-size promoters can significantly improve hydrogen yield due to their considerably bigger contact area, and change their energy level due to the quantum size effect. The nano-size promoters provide efficient electron exchange between promoters and hydrogenase enzymes [134].

#### 4-4- Thermolysis

Thermochemical Water Splitting Cycle (TWSC) known as water thermolysis or briefly thermolysis is splitting water molecules into hydrogen and oxygen molecules utilizing heat. The required heat may be from renewable energy sources such as solar heat [7]. TWSC needs a high-temperature heat source to run single or multi-step reaction cycles. TWSCs are closed-loop processes that recycle the utilized chemicals, and only water is split into hydrogen and oxygen as feedstock. TWSCs are categorized as direct or indirect. The direct methods are simple, compact, and involve a few steps. They need a very high-temperature heat source, usually above 1500 °C. A two-step direct TWSC based on metal oxide redox reactions includes an endothermic and an exothermic reaction. In the first reaction, metal oxide ( $MO_{ox}$ ) is converted to reduced-valence metal oxide and oxygen using concentrated solar heat. In the second step, the reduced metal oxide is oxidized with water exothermically, hydrogen is generated and the metal oxide is recycled [139].



Several metal oxides proposed for the redox reaction include ZnO, perovskites ( $ABO_3$ ),  $FeFe_2O_4$ ,  $CoFe_2O_4$ ,  $NiFe_2O_4$ ,  $FeAl_2O_4$ , and  $CeO_2$  [139].

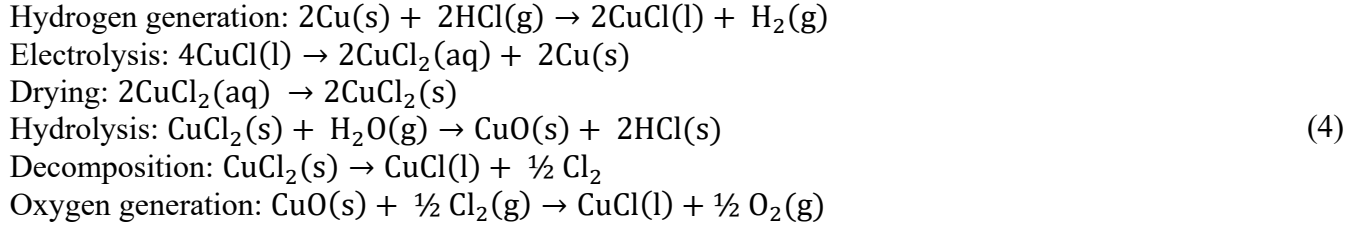
TWSCs still have a low TRL, which has hindered this technology's large-scale application and commercialization for CHP [140]. The direct TWSCs are still in the “research to prove feasibility” phase, which corresponds to a TRL of 2-4. The main obstacles to this technology are low hydrogen yield efficiency, low cost-effectiveness over the long term, and durability requirements of the materials used for the TWSC reactors against high temperatures and thermal cycling [139], [141]. Integration with a thermal energy storage system is vital for continuously operating solar-based TWSCs [139].

G. Barone et al. [142] evaluated a device's hydrogen production energy efficiency consisting of a parabolic dish collector equipped with an automatic tracking system and two cerium-based reactors to generate hydrogen experimentally and numerically. The yearly hydrogen production ranged from 1.19 m<sup>3</sup> to 1.64 m<sup>3</sup> depending on solar radiation, wind speed, and ambient temperature. They used a direct two-step redox cycle, using  $CeO_2$  as the redox material at an operating temperature of 1300-1500 °C. The results showed that cerium reduced at 1450 °C and oxidized at 900 °C. The dynamic model presented by G. Barone et al. [142] can be used and developed for similar research on direct TWSC coupled with solar heat. They have planned to use the model for iron-based thermochemical reactors in the future.

Indirect thermolysis on the other hand can be done with temperatures lower than 1200 °C while there is no need for packaging the solar heat receiver and reactor. As the number of steps increases, the operating temperature can be lower. Indirect methods have higher TRL due to their development and long history of using non-solar heat. Therefore, indirect methods can be integrated with solar heat energy more easily than direct methods. Indirect methods may have more than two steps, which allow hydrogen generation at lower temperatures, with higher efficiency. Designing a reactor also faces fewer challenges due to lower operational temperatures. The most known three-step TWSC is the Sulfur-Iodine (S-I) cycle with two endothermic and one exothermic reaction and a corrosive environment. It has a TRL of about 5-7, and efficiencies of 42%, 52%, and 60% at temperatures of 827 °C, 900 °C, and 1000 °C [139]. Solar-based TWSCs face technological challenges regarding the receiver's thermal energy storage and heating medium. The U.S. Department of Energy (DOE) SunShot has planned to develop the thermal storage and receiver of high-temperature solar concentrators with at least 12 hours of high-temperature heat storage by 2030 [143]. Cu-Cl, Ce-Cl, Mg-Cl, Fe-Cl, and V-Cl are among the four-step TWSCs. The Cu-Cl operating temperature is about 550 °C which is integratable with the existing solar tower technologies. The four-step Cu-Cl is less developed technologically than the three-step S-I therefore it has lower TRL [139]. In other words the new generation of solar towers with the ability to store temperatures as high as 800 °C is suitable for three-step TWSCs while the current technology of solar towers with a temperature range of about 550 °C is compatible with the four-step TWSCs. The direct methods with only two steps need further investigation and development to be compatible with solar towers generating and storing temperatures higher than 1500 °C.

R.B. Harikrishna et al. investigated using mild steel and iron-based metal scrap produced in many industrial processes for hydrogen production in a thermochemical reactor by injecting water steam [144]. They successfully produced  $500 \text{ mL}_{H_2}/g_{scrapmetal}$  at 1150 °C with a conversion efficiency of about 94%. The conversion efficiency is defined as the ratio of hydrogen volume generated by one gram of metal scrap to the volume of hydrogen production by one gram of pure iron. This corresponds to 0.5 million L of hydrogen for each ton of metal scrap. They introduced this opportunity as a carbon-free potential method for large-scale hydrogen production using metal waste in thermochemical reactions [144]. Later, they optimized hydrogen production by ferrous scrap metal with a particle size range of  $<250 \mu\text{m}$  to  $>1000 \mu\text{m}$  and a temperature range of 900 °C to 1200 °C experimentally. Maximum hydrogen yield was  $500 \text{ mL}_{H_2}/g_{scrapmetal}$  at 1100 °C. At medium size of scrap metal (355-600 $\mu\text{m}$ ), hydrogen production for all of the temperatures was higher than both small size ( $< 250 \mu\text{m}$ ) and large size ( $>1000 \mu\text{m}$ ) [145].

Ramdas S. Kadam and Ganapati D. Yadav [146] simulated a six-step Cu-Cl TWSC for hydrogen production by combining a pressure swing distillation unit with hydrolysis and hydrogen generation reactions to separate HCl and water. The six steps include hydrogen generation, electrolysis, drying, hydrolysis, decomposition, and oxygen generation as follows [146]:



The maximum operating temperature was 530 °C, which was required for the decomposition and oxygen generation reactions. The hydrolysis was done at 400 °C while the hydrogen generation reaction was carried out at 450 °C. The [LCA](#) reached a minimum [GWP](#) value of  $2.35 \frac{\text{kgCO}_{2,eq}}{(\frac{\text{kmol}}{\text{h}})_{\text{H}_2}}$  for a higher concentration of HCl

in the pressure swing distillation unit [146]. In another simulation [147] of the six-step Cu-Cl thermochemical cycle for hydrogen generation, the reactions and operating temperatures are hydrogen generation (450 °C), electrochemical (25 °C), drying (100 °C), hydrolysis (400 °C), decomposition (530 °C), and oxygen generation (530 °C). They also determined the energy and exergy efficiency of the six-step Cu-Cl cycle as 38.94 % and 52.74 % and the hydrogen production rate was also 0.002 kg/s [147].

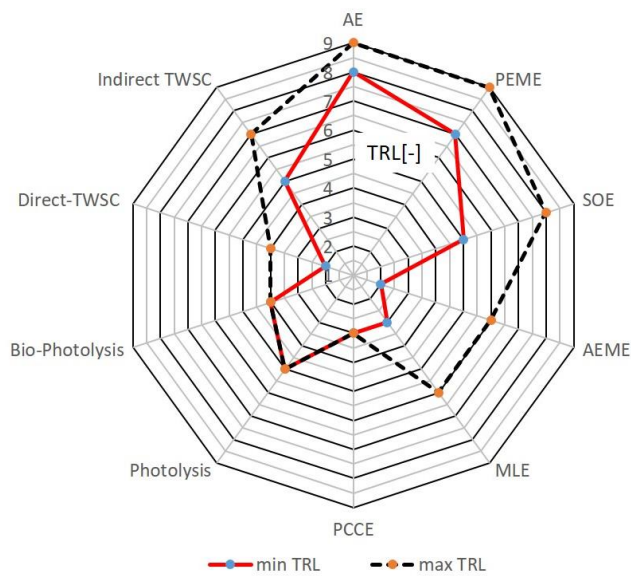
Designing an optimized heat exchanger with uniform temperature distribution in the thermolysis reactor is critical. Ehsan Armoudli and Ofelia A. Jianu [148] presented a molten salt heat exchanger design to reach a uniform temperature distribution in the reactor, for a four-step Cu-Cl cycle with an operating temperature distribution of 431.3 to 539.0 °C.

The six-step Cu-Cl cycle, a hybrid thermolysis that uses electricity and heat simultaneously, has attracted considerable attention since it is compatible with the current technology of solar towers, heat storage, and hot fluid in the receiver of the solar collector. In addition, designing the cycle components such as the reactor, heat exchangers, and receiver for this operating temperature range is feasible now. In contrast, high temperatures and metallurgical limitations are the main challenges for higher temperatures, which are required for direct two-step TWSCs thermal cycling.

## 5- Comparative results

The renewable-driven WSHP technologies including different electrolyzers (AE, PEME, SOE, AEME, MLE, and PCCE), photolysis (PC-WST, and PEC-WST), bio-photolysis (DBP, IBP), and thermolysis (direct and indirect TWSC) were studied and compared from energy, exergy, economic, environmental, technological, and social (4E-TS) criteria. To increase the readability of the data and make comparison possible, the data from numerous references were gathered in Tables 1 to 4. The comparative study is done by using both tables and some selected graphs. However, according to the availability, completeness, and importance of the data, 6 criteria including TRL, LCOH, CAPEX, degradation rate, lifetime, and specific electricity consumption (SEC) were chosen to be presented in Graphs. Presenting all of the data in graphs was impossible because the article would become too lengthy. However, the other criteria and data are used in the comparative study and discussions.

The TRL studies show that AE and PEME are the most mature technologies, and SOE, and indirect-TWSC come after it (Fig. 12). This shows a great potential for research and investment on the WSHPs with TRL of less than 9.

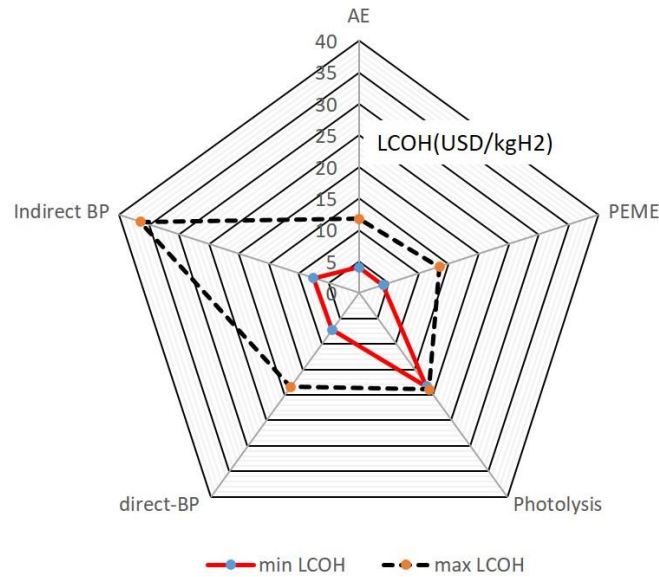


**Figure 12. The comparison of technology readiness levels for renewable-driven WSTs based on Table 2.**

The reduction of the levelized cost of green hydrogen, which is the most important factor for filling the gap between general and local social acceptance of hydrogen [41], mostly depends on a substantial cost reduction in the electrolyzer stack, improving electrolyzer efficiency, increasing yearly full load operation hours, increasing lifetime, reducing electricity tariff and right policies [75]. In addition, the capacity factor of the RES and technology degradation rate are other vital factors that need to be improved. Improving one

criterion may deteriorate another, hence a full understanding of impacting parameters and their behavior is essential. IRENA [62] has considered an LCOH of 1 USD/kgH<sub>2</sub> as a target, which may be achieved by improving 4E-TS criteria. However, in the current situation AE, and PEME are the most suitable options for investment and commercialization.

The LCOH by different technologies is shown in Fig.13. It shows the minimum and maximum reported LCOH for the technology. It shows AE and PEME respectively have the lowest LCOH while photolysis, direct-BP, and indirect-BP stay in the next levels. AE and PEME with [learning rate](#) ranges of 9-21% and 13-21% ([62], [98], and [73]) still have a high potential for cost reduction due to the scale-up of these electrolyzers. In addition, the LCOE used in these electrolyzers can be reduced further by scale-upping since learning rates of PV-based and wind turbine-based electricity generation are 24% and 15%, it means that with each doubling of the installed accumulative capacity, LCOE will reduce by 24% and 15% respectively ([149] and [150]).



**Figure 13. The maximum and minimum reported LCOH for different technologies**

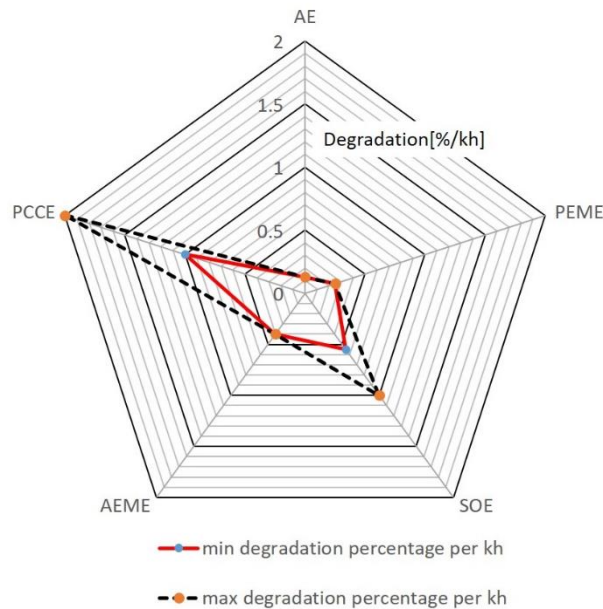
Among the significant parameters that have an impact on the cost of the PEME stack and its performance for large-scale implementation, the amount of Platinum and iridium catalyst used on the membrane surface is on top [104]. PGMs have similar physical and chemical characteristics and can substitute each other. Iridium is 20 times scarcer than Platinum and it is probably a weakness in the supply chain of PEME. In addition to substituting PGMs together, thrifting, and recycling are other methods to reduce the risk of

using PGMs in PEME [105]. However, there might be other technologies with increasing demand on PGMs, and the risk is still high. Therefore reliance on PGMs must be reduced, and other alternatives should be investigated [57].

Among RESs, hydropower with a [capacity factor \(CF\)](#) of 41.5% [4], is the most suitable RES for GHP, wind power, and solar electricity with capacity factors of 35.4% and 24.9% are next [74]. Geothermal is another RES that has a high CF of 74.3%, but it is highly dependent on geography. The RES type also determines the [Life Cycle Impacts \(LCI\)](#) of GHP. A comparison between AE and PEME coupled with wind power and PV power concluded that the combination of PEME with wind is the most environmentally friendly according to the magnitudes of GWP, WDP, MDP, and FDP [87].

On-grid renewable electricity-based hydrogen production systems are more profitable than off-grid systems because the electricity management strategies can be used for the import and export of electricity according to the costs during the peak hour and off-peak hours. For example, LCOH for an off-grid PV-based PEME is 1.17 to 2 times that for an on-grid PV-based PEME [67].

PEME is sensitive to impurities and needs highly pure water. This remains a challenge for improving its social acceptance, particularly in places under water stress. This sensitivity has caused the degradation rate of PEME to be 2 times AE [76] (Fig. 14). PEME's long-term stability should be confirmed on the MW scale, and its dependence on forever chemicals such as polyfluoroalkyl substances (PFAS) is another challenge that needs to be resolved [57].



**Figure 14. The comparison of maximum and minimum reported degradation rates of different technologies**

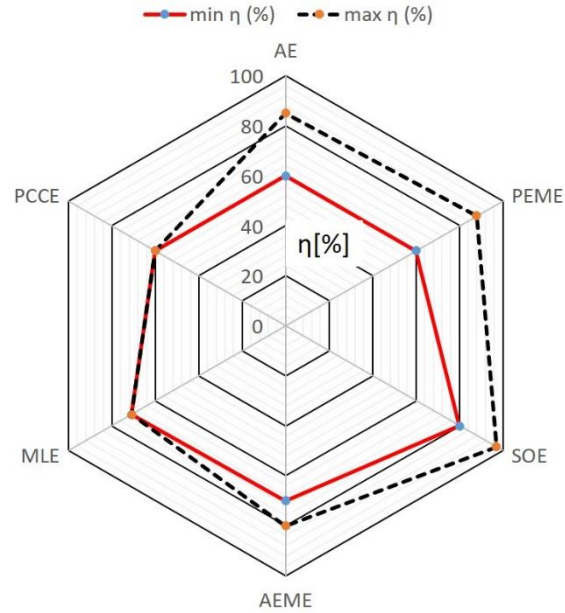
LCOH by using RES-based AE is mostly dependent on the LCOE and the [capacity factor \(CF\)](#) of the RES [90] and [93]. AE is the most mature GHP technology with the lowest degradation rate (Fig.14) and LCOH (Fig.13). The capacity factor can be improved by integrating different shares of RESs such as wind and solar-PV [1]. In addition, the simultaneous use of PEME and AE for GHP by RES can reduce the overall cold start-up [60] and degradation rate while keeping efficiency high during changeable load.

Apart from the different methods such as improving durability, reducing degradation rate, using inexpensive materials instead of PGMs, reducing LCOE, scale upping, etc. that can be used to reduce LCOH, applying the right energy governance [151], policies, regulations, and incentives can promote GHP technologies. Investigating the impact of policies on the GHP characteristics is scarce and this topic needs to be deeply investigated. For example, [Output-Based Allocations \(OBAs\)](#) is a decarbonization policy that compares industries based on the GHG produced per product unit. Using hydrogen as an energy carrier can improve decarbonization in industries and develop the usage of hydrogen in different sectors. An investigation [88] proved that using OBAs alone has reduced LCOH by about 20% and reached 4.09 USD/kgH<sub>2</sub>. IRENA [152] has published a guide for policymakers on green hydrogen production for industries and discussed twelve policies including 1-Industrial decarbonization strategies, 2-Targets for green hydrogen in industry, 3-Phase-out of fossil fuel technologies, 4-Emission trading systems, 5-Carbon taxation, 6-Grants and loans, 7-Fiscal incentives, 8-Carbon contracts for difference, 9-Green hydrogen auctions, 10-Quotas of green products, 11-Sustainable public procurement, and 12-Ecolabelling. The South Korean government has decided to become the leader in the hydrogen economy. For this purpose, they need to fill the gaps between institutional and technological perspectives. As a result, policymakers and experts have remained concerned. This case proposes practical policy implications for the co-assessment of socio-technical system for transition toward hydrogen economy [153].

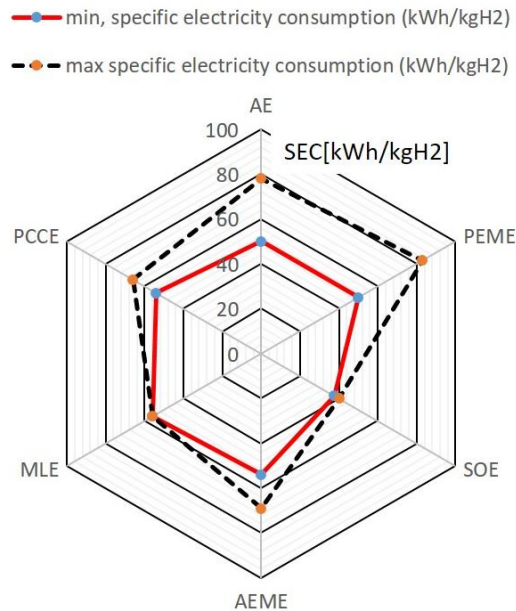
While PEME and AE suffer from depending on noble, scarce, and expensive materials such as PGMs, SOE benefits from abundant and inexpensive materials, and it consumes less nickel and zirconium per MW (Table 2). SOE has the highest efficiency (Fig.15) and lowest specific electricity consumption (Fig.16).

The high degradation rate of SOE (Fig. 14) has reduced its lifetime to 20-50 kh at full load operation (Fig.17). The impact of thermal stress due to the high operating temperature of SOE on the degradation rate, aging of internal components, and durability must be investigated with special attention. In addition, the dynamic behavior, reliability, and hydrogen purity of SOE need to be fully studied. In addition, data about the LCOH, and GWP by SOE is scarce and funds are required to support research and development

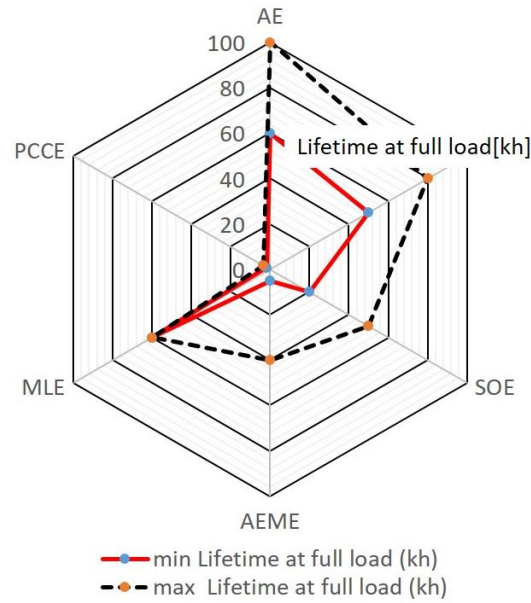
attempts in this field. The CAPEX of SOE is high and cost reduction depends on the possibility of scale-upping and increasing current density ([71], and [76]). SOE needs more auxiliary equipment due to its high operating temperature, hence it occupies more footprint than AE, and PEME [76]. This shows the need for packaging SOE using more compact heat exchangers and auxiliary equipment to be specifically designed.



**Figure 15. The comparison of maximum and minimum reported energy efficiency GHP technologies**

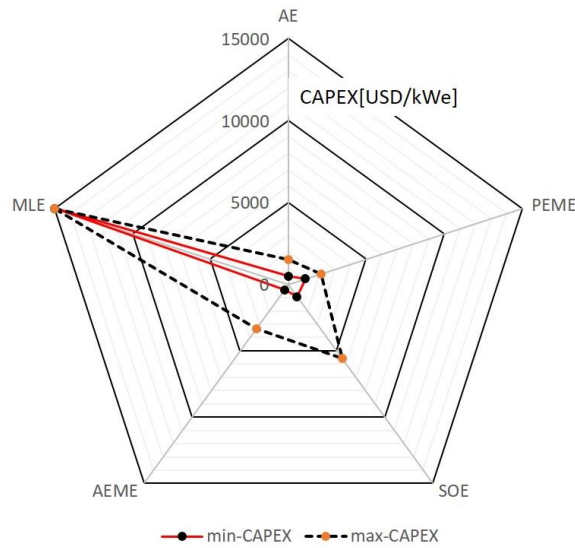


**Figure 16. The comparison of maximum and minimum reported specific electricity consumption (SEC) of GHP technologies**



**Figure 17. The comparison of maximum and minimum reported lifetime at full load operation of GHP technologies**

AEME with a reported TRL of 6 ([56], [57], [59], and [64]) is technologically at the technology demonstration level. In the last three years, it attracted too much attention and its TRL has improved from 2 to 6. This technology is supposed to be developed for using seawater directly. It has a relatively small degradation rate, however, at present it is recommended to use a reverse osmosis desalination system with AEME [78]. This characteristic is highly important for the regions under water stress, and also for hydrogen production by offshore electricity. Combining this technology with offshore wind turbines eliminates the need for electricity transfer to the shore. In other words, seawater after some initial filtrations and offshore electricity can be used directly to generate green hydrogen. Compared with PEME and AE it is less sensitive to the water source hence for offshore applications it has a high potential. With investment in research and development, using non-noble material on electrodes, following the learning rate curves, the TRL improves and its CAPEX and LCOH can decrease even more. It still has a short lifetime of about less than 20 kh at full load. Increasing the TRL depends on improving the lifetime, stability, degradation rate, and compatibility with seawater, therefore these fields need to be invested in and investigated. The CAPEX of AEME even with a lower TRL than SOE is smaller than SOE (Fig.18). It shows that developing this technology will reduce the CAPEX and OPEX even less than SOE, AE, and PEME [80]. The low operating temperature makes it a good option for fast startup applications. These characteristics have made AEME a potential option for the near future of GHP with a feasible cost.



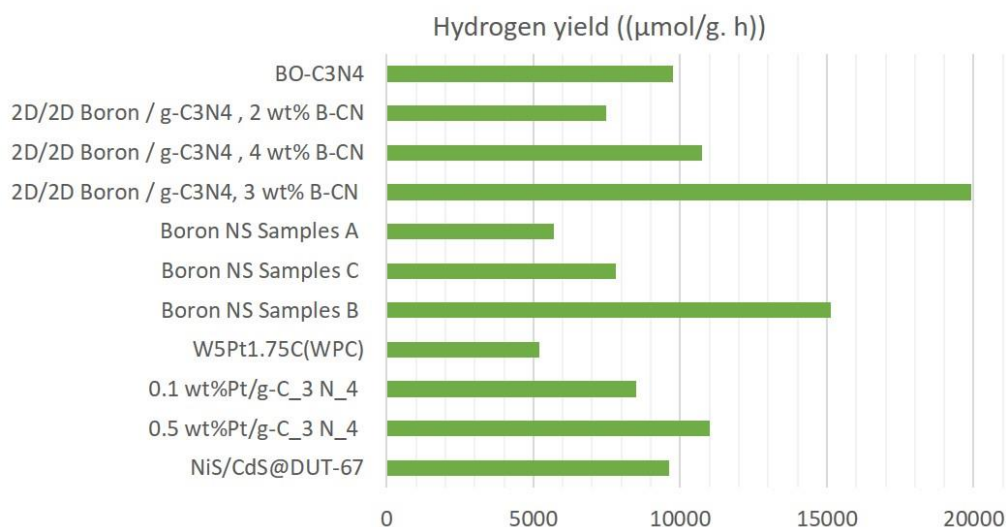
**Figure 18. The comparison of maximum and minimum reported CAPEX of GHP technologies**

Fewer part numbers mean less MDP, CAPEX OPEX, and complexity of the system. Unlike its counterparts, MLE does not use a membrane or a diaphragm to separate oxygen and hydrogen. Rather, it uses the momentum of the continuous flow of electrolyte solution ([110] and [85]). Technologically, MLE sometimes approaches the demonstration level with a TRL of 3 to 6 (Fig.12). While being membraneless is an opportunity to reduce MDP, CAPEX OPEX, and complexity, but successful product separation is not reported ([85] and [84]). A hydrogen purity of 99.95 is reported which is low compared with PEME [72], therefore hydrogen purity is still a challenge, and designing a perfect product separation mechanism needs to be investigated. Due to having no membrane, MLE is talented enough to be highly durable. In addition, it can have a long lifetime and high tolerance against impurity. Corrosion, fouling, and membrane blockage due to impurities are no longer problematic. This means that MLE like its counterpart AEME can use low-quality water, and the costs for purification, deionizing, and decarbonizing will be saved. MLE is also flexible in using different types of electrolytes such as acidic, alkaline, and neutral solutions. Currently, hydrogen purity, high CAPEX, and short lifetime at full load (Figs. 17 and 18) are the main challenges for the development of MLE and they need more investment and investigation.

PCCE is currently under laboratory testing with a [TRL](#) of 3 [58], and has the shortest lifetime under full-load operation (Fig.17) and the lowest energy efficiency among its counterparts. An important reason for the low efficiency is the current leakage during electrolysis ([82] and [52]). This technology still has a long way for large-scale hydrogen production with high efficiency and purity. It also has the highest degradation rate among the known electrolyzers (Fig. 14). Other vital challenges are optimizing materials, cell design, and operating strategy to improve Faradic efficiency [52]. The PCCE produces pure, dry

hydrogen at high pressure, omitting some subsystems from downstream and thus reducing complexity. The higher operating temperature of PCCE can reach a higher efficiency than PEME and lower LCOH [58]. This technology can be sustainable for power and hydrogen production [83]. In conclusion, improving degradation rate and energy efficiency, full-scale testing, increasing durability and material optimization are the main challenges for this technology and need to be invested in and investigated.

Photolysis in this review is introduced as a quasi-green hydrogen production system (QGHPS), in which light as renewable energy is used to produce hydrogen with GWP of less than 1 kgCO<sub>2, eq</sub>/kgH<sub>2</sub>. Technologically it is between technology development and technology demonstration steps with [TRL](#) of 5 (Fig.12). Most of the works on photolysis are small-scale and laboratory prototypes [114] but recent researches claim large-scale potentials ([119] and [120]). The LCOH by photolysis is higher than AE and PEME, however the difference is not too much. This difference may be compensated by large-scale production and improving TRL. Fig.19 shows the [hydrogen yield](#) using 11 talented photocatalysts (from Table 4) in which the full-scale potential is claimed or hydrogen yield is more than 5000  $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$ . It shows that 2D/2D Boron / g-C<sub>3</sub>N<sub>4</sub>, 3 wt% B-CN has the highest [hydrogen yield](#) of nearly 20000  $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$  and it is proposed as a pioneer for producing nonmetal photocatalysts [127], but it is restricted by the rapid recombination of photogenerated carriers and insufficient use of surface charge. Boron NS is another photocatalyst with a high potential with a hydrogen yield of more than 15000  $\mu\text{mol} \cdot \text{g}^{-1} \text{h}^{-1}$  [120]. In addition, Metal-organic framework materials (MOFs) such as Ni-MOF derivatives are recognized for more stable hydrogen production with a higher yield [121]. The minimum test time for stable operation of a photolysis system is 50 h [114]. However, by taking a look at Table 4 it can be seen that some of the full-scale claims have not passed this criteria ([126], [130], [127], [119], and [121]). This shows that the reader must be careful about the claims made in some of the references. The importance of the stability test must be fully addressed because after several cycles the hydrogen yield drops, which is highly important for full-scale tests, and scale-up. As a result, stability of photolysis is of high concern and needs research and investment.



**Figure 19. Comparison of photocatalysts with a hydrogen yield of more than 5000  $\mu\text{mol. g}^{-1}\text{h}^{-1}$  or full-scale claim in the reference**

Using sacrificial reagents is important to avoid reverse reactions and efficiently transfer photoelectrons  $e^-$  and photo holes  $h^+$  for Redox reactions [119], but it causes cost and environmental-related concerns [115]. The most well-known sacrificial reagent is TEOA ([118], [122], [126], [127], [124], and [125]); however, some photolysis experiments are performed without using sacrificial reagents ([128], [119], and [124]).

Crystalline elemental Boron nanostructures were introduced through a new synthesis process for non-metallic single-element catalysts without adding Pt, or sacrificial reagents [120]. Wu et al. [129] introduced a 2D ultra-thin nanosheet of a Pt-loaded g-C<sub>3</sub>N<sub>4</sub> without sacrificial reagent for photocatalytic hydrogen production. Most of the available studied experiments have been done under laboratory test conditions with a 300 W Xenon lamp light source ( $\lambda \geq 420$  nm). The STH efficiency is reported in the ranges of 0.06 % to 1.5 % ([120], and [124]) and [AQY](#) is in the range of 5 % to 23 % at 420 nm ([119], [123], [124], [129], and [126]). The efficiency is too low and needs to be improved using different methods. Using co-catalysts is a method to reduce recombination and separate photo-generated charge carriers, such as electrons and holes. Conductive inorganic nanosheets have shown potential as co-catalysts [117]. While using Pt in photocatalysts or co-catalysts has a direct impact on the amount of hydrogen yield [119] it has increased the cost of photocatalysts as well. In an attempt Nanodot-carbon nitride (C<sub>3</sub>N<sub>4</sub>) has been used as a photocatalyst. It is talented for scale-upping and synthetic approaches are the main part of production of the Carbon Nitride-based photocatalysts [122]. It is chemically stable and has an appropriate band gap and energy level under standard test conditions at  $\lambda = 420 \pm 20$  nm achieving STH of 2% and [AQY](#) of 16% [122]. In addition, the main reason for the highly efficient activity of 3D g-C<sub>3</sub>N<sub>4</sub> NS is the efficient separation of photogenerated charge carriers [124].

Bio-photolysis (DBP and IBP) is a quasi-green hydrogen producer technology. It includes renewable mechanisms such as plants, sunlight, and water. With a TRL of 4, it is less mature than photolysis and technologically is in the middle of the technology development step (Fig.12). BP has the highest LCOH among the reported values for WSHPs (Fig.13). Its commercializing status and [STH](#) of BP is limited by microalgae ability since they can only use radiation in PAR wavelength region [51]. The mean STH of BP is about 10 % in laboratory scales, meaningfully smaller than that for PEC-WS with a mean STH of 18% [114]. Therefore, improving STH and scale-up are highly challenging and further process development is required [132]. Efficient hydrogen production by DBP needs a vast land [133] for large-scale algae cultivation, which is possible only for some seasons or regions. In addition, co-culturing bacteria with green algae can boost hydrogen production compared with pure algae. Co-culturing activated sludge bacteria and *Chlorella vulgaris* is also used to increase hydrogen yield, in this co-culturing activated sludge bacteria consume oxygen, while algae produce hydrogen. Optimization of PH, light absorption, temperature, and cell density can improve hydrogen yield [138], and using oxygen regulation methods such as mineral deprivation, oxygen scavengers, co-culturing, antioxidants, and nanoparticles can make BP economical ([134], and [137]).

The direct TWSCs with a TRL of 2 to 4 are in the research to prove the feasibility step and in some cases, developing technology. The most important challenges facing this technology are low hydrogen yield, economic feasibility, and durability of reactors against high operating temperatures of above 1500 °C and thermal cycling [139], [141]. For solar-based direct-TWSCs having a high-temperature thermal energy storage system is vital for continuous operation [139], which has remained a challenge yet.

Indirect TWSCs have higher TRL (Fig.12) due to their lower operating temperature of less than 1200 °C, and their history of using non-renewable heat sources. Hence indirect four-step TWSCs can be integrated with existing solar concentrators more easily than direct TWSCs, and designing reactors and heat exchangers is less challenging. Having a uniform temperature distribution in the reactor is the challenging part of the design procedure. The three-step indirect TWSCs are more suitable to be used with a new generation of solar concentrators with the ability to store heat at 800 °C. The two-step direct TWSCs require more research and development to be well-matched with solar concentrators capable of generating and storing heat with temperatures as high as 1500 °C. The six-step Cu-Cl cycle is hybrid electro-thermolysis and is well-matched with the current technology of solar concentrators, heat energy storage, and hot fluid in the receiver. While designing the cycle components such as the reactor, heat exchangers, and receiver for the six-step TWSC is feasible, the metallurgical limitations are the main challenges for direct two-step TWSCs.

## 6- Prospects

Hydrogen is essential for sustainability [9] and RES-driven WSHPs play a critical role for supplying hydrogen demands in the future. However, there are still challenges that must be resolved by investment and investigation. The most important challenges include:

- 1- The social acceptance of hydrogen and its related technologies. More investigation and investment to increase the public awareness about the advantages of hydrogen technologies are required. Policymakers and governments play a critical role in this area. Hence, research institutes, organizations, and universities can shape the mind of policymakers regarding GHPs. Applying the right policies is as important as the technological improvements.
- 2- Research on non-PGMs is of great importance for the future of electrolyzers, while it reduces the cost, the dependency on rare materials will decrease.
- 3- The reliability of RES-driven WSHPs is still low due to the low capacity factor of RESs. It can be improved by combining different RESs and the grid. Finding more reliable electricity for the stable operation of WSHPs is of great importance and needs serious investment and investigations.
- 4- Except for AE and PEME with TRL of 9, other technologies are not commercialized for different reasons such as low hydrogen yield, high electricity price, expensive and rare materials used in electrodes and catalysts, low durability, etc.
- 5- The hydrogen production, storage and distribution network will be integrated with other energy networks such as the natural gas network, electricity grid, district heating and cooling networks. It is important to understand the interaction between these energy networks.
- 6- More detail on opportunities and potential challenges of different electrolyzers for possible future investment and investigation are presented in Table 2b.

In the conclusion section, the challenges, and opportunities for investment and investigation are presented in more detail.

## 7- Conclusions and outlooks

Since five years ago (2020-2024) investment in research and development on clean and green hydrogen production has increased dramatically. More than 93% of the journal articles published by ScienceDirect from 2004 to 2024 were published from 2020 to 2024, showing that green hydrogen has been recognized as a key for the future green planet. Using hydrogen as a fuel in gas turbines has been technologically

mature with a [TRL](#) of 9 and commercially is at the demonstration level with a [CRI](#) of 3. While different fuel cells are developing to consume hydrogen for electricity production, the existing fully commercialized and mature technologies such as gas turbines can also be upgraded to burn fuel mixtures with a high percentage of hydrogen. General Electric (GE) gas turbines such as 7HAs and 9HAs can burn as much as a 50/50 hydrogen/natural gas blend [154]. In another attempt, Siemens within the Zero Emission Hydrogen Turbine Center (ZEHTC) has set up a demonstration plant to show connecting gas turbines with hydrogen, RES, and ESS [155]. Considering these global trends toward research and developing hydrogen-related technologies, the possibility of storing excessive RESs as hydrogen, the reliable and sustainable transition to zero-emission is more feasible by integrating renewable energy technologies, water-splitting hydrogen production technologies, upgraded conventional infrastructures such as hydrogen-driven power plants, and fuel cells.

There is a gap between general and local social acceptance of hydrogen [41]. This can make a challenge for public use of hydrogen-driven appliances and technologies. This gap needs to be filled with proper decisions and policies made by policymakers, and research institutions. According to the review, investigation on social acceptance of hydrogen is scarce, and this topic needs to be addressed in the future. Most people, when they hear about hydrogen, the first thing that comes to their mind is a bomb, and explosion, not a clean energy carrier. This attitude needs to be corrected and for this purpose, more societal investigations are required.

Among water-splitting technologies used for green or clean hydrogen production only AE and PEME have reached a TRL of 9 with a mature technology and ready for scale-upping. The other technologies, including SOE, AEME, MLE, PCCE, photolysis, bio-photolysis, and TWSCs, are not mature yet and they are in different technology readiness levels from 2 to 7. Using expensive and rare materials such as PGMs in catalysts and co-catalysts has increased the LCOH. Research on non-PGMs is of great importance for the future of electrolyzers.

Another challenge is the low reliability of WSTs coupled with RESs which is mainly due to the low capacity factor of RESs, especially the solar-driven systems. Studies have shown that oversizing the plants, distributing plants in different sites instead of a single large plant, and integrating solar-PV with wind power, or hydropower can improve the capacity factor and also reliability of hydrogen production. This is highly important especially when high-temperature WSTs are used for hydrogen production and repetitive shutdown due to long startup time and thermal cycling is impossible. Therefore, improvement of the capacity factor of the energy source used for WSTs is of great importance and needs to be investigated and

invested in. In addition, coupling different WSTs together can improve reliability by reducing startup time, and increasing efficiency. The high temperature of some technologies such as SOE and hydrogen gas turbines can be used to run six-step TWSCs.

WSTs have different LCIs. Research has shown that using wind-power-based PEME is more environmentally friendly than the combination of AE with wind power or solar PV and PEME with solar PV. This is important to develop the most environmentally friendly integration of WSTs with RESs. In addition, using sacrificial reagents and co-catalysts to increase oxygen consumption and hydrogen yield by photolysis has brought environmental and cost concerns. Improving hydrogen yield without using sacrificial reagents and co-catalysts are challenges that need to be investigated.

Data for LCOH for large-scale water-splitting-based hydrogen production is scarce, apart from some limited data for large-scale GHP by AE and PEME, most of the other data are for laboratory and small-scale tests. As a result, most of the economic comparisons are done under different TRLs. The cost comparison can only make sense when the cost is determined for the same TRL.

It should be noted that achieving TRL 9 for hydrogen production based on water-splitting technologies is the first step towards large-scale production and reducing the LCOH. Other parameters that affect the LCOH and ultimately its social acceptance include reducing dependence on rare and expensive materials such as PGMs, using renewable electricity and heat sources with higher capacity factors, reducing the price of renewable electricity and heat through technological development and large-scale production, integrating different technologies and their operation under on-grid and off-grid scenarios, proper hydrogen-based and hydrogen-include national and international energy governance, policies and regulations, developing large-scale hydrogen storage technologies and upgrading hydrogen transmission and distribution networks.

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