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Review Article

Generation of green hydrogen using self-sustained regenerative fuel cells: Opportunities and challenges

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HIGHLIGHTS

- Green hydrogen generation using electrolyzers is the most sought-after technology.
- Integration of electrolyzer with PV systems makes the process fully sustainable.
- PEM electrolyzers directly coupled with PV arrays boosts system efficiency.
- Unitized Regenerative FCs possess cost and volume advantage due to single stack.

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ABSTRACT

The ever-increasing energy demand, depleting fossil fuel reserves, and rising temperatures due to greenhouse gas emissions have necessitated the transition towards the generation of green and clean energy through renewable energy sources. Solar energy is one such renewable energy source that has received significant attention owing to its abundance and inexhaustibility. However, solar energy alone cannot replace fossil fuels in the energy portfolio. There exists a need to develop another clean energy source that can potentially act as an alternative to conventional fuels. Hydrogen proves to be an ideal candidate in this domain and can be sustainably generated by water electrolysis by powering the electrolyzer using solar energy. The hydrogen thus synthesized has net zero carbon emissions and is a suitable asset for decarbonizing the environment. This review encompasses the generation of hydrogen using PV-Electrolyzer systems and addresses the challenges associated with the same. Overcoming these drawbacks can ensure a strong position for hydrogen as an alternative fuel in the energy infrastructure. By employing electrolyzers that are fueled by renewable energy and then using that hydrogen to feed a fuel cell, this study aims to clarify the potential and constraints of producing green hydrogen. Since this area of research has not yet been fully investigated, a review article that enables and encourages academics to develop original solutions is urgently needed.

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Contents

Introduction

The current global scenario poses the need for alternate sources of energy, due to environmental degradation associated with energy generation via fossil fuels utilizing conventional conversion technologies [1]. The world is undergoing an unprecedented climate crisis with global CO_2 emissions reaching unparalleled numbers. With 10.6 and 4.7 billion metric tons of CO_2 emissions in the year 2020 respectively, China and the United States are by far the two most extensive polluters in the world. U.S. emissions decreased by 16% since 2010, despite it being the second-largest polluter. China's CO_2 emissions, in contrast, rose by over a quarter [2]. They did, however, start to decline in 2022, falling by 1.4% in the first three months of the year, largely owing to the COVID-19 pandemic [3]. Despite this, there is still a long way to go, and significant progress can only be made by turning attention to alternative (and renewable) energy sources as a means of decarbonizing diverse sectors that result in CO_2 and greenhouse gas emissions. Due to the rapid depletion of traditional energy sources (such as coal, petroleum, and natural gas) as well as the environmental concerns they raise (as stated above), there is an ever-increasing demand for renewable energy sources worldwide. Over the past two decades, there has been a marked growth in the usage of renewable energy globally. In 2020, consumption totalled 32 EJ, as depicted in Fig. 1.

Among the various sources of renewable energy, solar photovoltaics has dominated capacity expansion in recent years, owing to a wide variety of characteristics solar energy possesses [5]. Firstly, it is available in abundance. The sun emits it at a rate of 3.8×10^{23} kW and the earth intercepts around 1.8×10^{14} kW of it [6]. Studies have revealed solar energy's ability to fulfil the global energy demand due to its cost-free and profuse nature [7]. Secondly, it is inexhaustible, having higher output efficiencies than other energy sources. In addition to this, solar energy does not have a negative impact on the ecosystem and solar systems can be used effectively for remote areas [8], industrial operations [9], and residences [10], thereby making it a viable option.

However, the use of solar energy can only partially address the issue at hand [11]. The dire need to reduce greenhouse gas emissions globally has put the policymakers in a situation of crisis to come up with sustainable alternatives to conventionally-used fossil fuels which are environmentally detrimental. Greenhouse gas emissions are significant contributors to this rise in temperature, with the amount of CO_2 in the atmosphere increasing day by day. Such emissions can only be reduced by finding an alternative to fossil fuels since

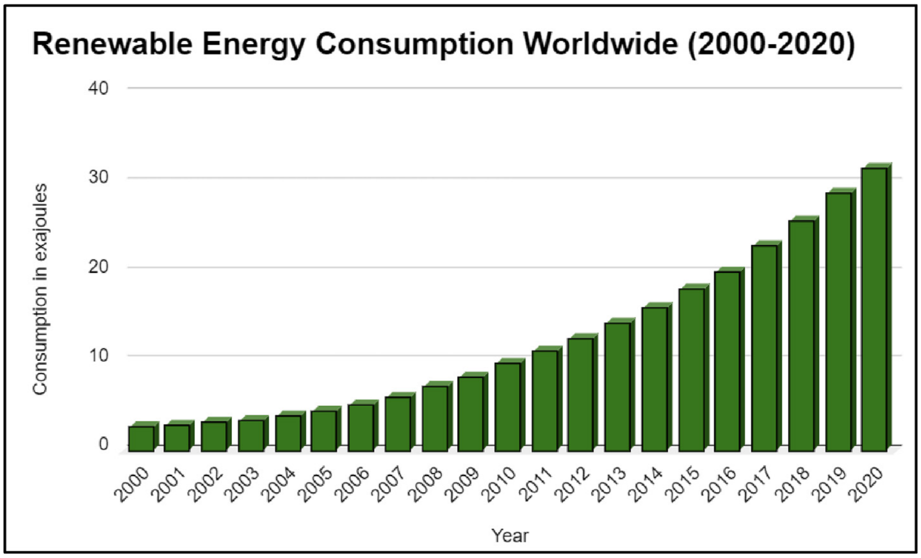


Fig. 1 – Renewable energy consumption worldwide (2000–2020) [4].

most of the carbon dioxide in the air is released from the combustion of oil, coal, and natural gas [8]. One of the most plausible approaches to replace the present carbon-based economy is to develop hydrogen as a potential fossil fuel substitute. Hydrogen can be thought of as a fuel that combines the benefits of fossil fuels (e.g., flexibility) with a low carbon impact [12]. Hydrogen is not only a strong candidate for the alternative energy source everyone is looking for but also has the potential to solve the global climate crisis [13]. Research has proven that hydrogen does not release any sort of emissions or any greenhouse gases in its efficient conversion into energy [14]. And hence, it has been under keen observation and research is being directed towards the establishment of hydrogen as an alternative in the energy infrastructure [15].

By 2050, the global population is expected to increase from 7 billion to nearly 9 billion people, resulting in an exponential increase in energy demand [16]. The transportation and energy sectors currently account for most of the rising greenhouse gas emissions and are far from being sustainable [17]. Today, the transportation industry, notably road transport, accounts for 37% of total CO_2 emissions [18], a figure that is only expected to rise in the coming years unless we find a sustainable energy source at the earliest. As a result, hydrogen has the potential to be a gamechanger in addressing energy security concerns and lowering reliance on oil [19].

Some believe hydrogen can be the primary energy storage technology, the primary heating fuel, and the primary transportation fuel for automobiles, trucks, planes, and other vehicles. Because of its intrinsic flexibility as a chemical and energy carrier, hydrogen has the potential to fill the gaps in the decarbonization endeavour [20]. A hydrogen economy with green hydrogen as a fuel is an appealing notion to mitigate climate change and energy security concerns. Several sources have identified the decreasing results in the discovery of new petroleum supplies [21], as well as ever-increasing global energy consumption [22], as major issues. Some sources even claim that we have only 47 years of oil reserves left [23]. To ensure a smooth transition to “greener and cleaner” options, effective mitigation strategies must be adopted at the earliest. The use of hydrogen as a fuel is one of the most promising of these alternative greener choices [24]. As a fuel, hydrogen has a gravimetric energy density that is 2.5–3 times that of today's most widely utilized fossil fuels [25]. In a three-phase approach to constructing a green hydrogen economy, a study presented a vision for integrating green hydrogen into the industrial, transportation, heating and buildings, and electricity sectors [13]. Considering all of the benefits of using hydrogen as a fuel in a future green economy, the goal of this study is to lay the groundwork for any future research in this area. An overview of the progress achieved in creating the technologies required to realize the green hydrogen economy is undertaken in this study.

Hydrogen is currently produced from a variety of sources, including fossil fuels [27,28], natural gas [29,30], water electrolysis [31], and even biological processes, such as biomass [32,33]. According to the “Global Hydrogen Review 2022” published by the International Energy Agency (IEA), the current global market share of each production source of hydrogen as of 2021, is illustrated in Fig. 2. The most popular technique for hydrogen production is catalytic reforming of methane, other hydrocarbons, or biomass, but it has several significant limitations, including high cost, low yield, and detrimental environmental impact [34]. Hydrogen synthesis via electrodissociation of water utilizing renewable energy sources, on the other hand, has garnered a lot of interest in the context of clean energy [35]. This is because the process produces ultrapure hydrogen and oxygen with a high density while also being environmentally benign. The electrolysis process and the half-cell reactions - Hydrogen evolution reaction (HER) at the cathode and Oxygen evolution reaction (OER) at the anode - are powered by electricity obtained from renewable energy sources such as solar [36], wind [37], and so on, and have a zero-carbon footprint, making this process environmentally friendly. Green hydrogen is hydrogen that is created by the electrolysis of water, powered by a sustainable renewable energy source. This is demonstrated in the equations below, which show the

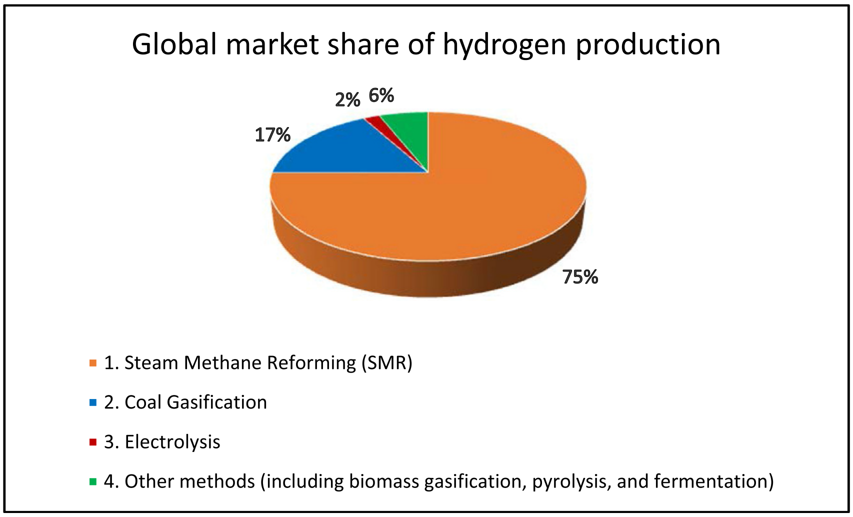


Fig. 2 – Global market share of hydrogen production based on each source [26].

electro-dissociation of H_2O into H_2 and O_2 [38,39]. The potential energy density of the hydrogen/oxygen reaction (3660 Wh/Kg-H_2O) is substantially higher than Li-ion batteries currently available [40,41]. And hence, extensive research is being carried out in this domain [42,43].

$$H_2O \rightarrow H_2\uparrow + \frac{1}{2}O_2\uparrow$$

$$2H^+ + 2e^- \rightarrow H_2\uparrow \text{ (HER at the cathode)}$$

$$H_2O \rightarrow \frac{1}{2}O_2\uparrow + 2H^+ + 2e^- \text{ (OER at the anode)}$$

Green hydrogen can be utilized for transportation, synthesis of green ammonia, heating applications, and in the natural gas industry. It can also prove to be a clean-energy carrier by providing an alternative to conventional fuels [44]. The applications of green hydrogen synthesized using an electrolyzer powered by a PV system have been depicted in Fig. 3.

As represented by Fig. 3, hydrogen is obtained from the electrolysis of water, powered by a combination of PV and grid systems. This hydrogen is further stored and sent to industries for utilization for numerous applications. Owing to such potential widespread applications of green hydrogen, it becomes essential to get familiarized with the concepts associated with its production using PV-Electrolyzer systems. This review focuses on the integration of electrolyzers with renewable energy sources, specifically solar PV systems to elucidate its merits and challenges associated with the same. The review also encompasses emerging electrolysis technologies and explores their potentiality in the synthesis of green hydrogen.

Solar photovoltaic systems

Photoelectric conversion using solar cells is the most efficient method of harnessing solar energy [46]. A solar array constitutes several solar panels, each of which is made up of multiple solar modules which in turn are made of discrete solar cells connected to one another [47,48]. Traditionally, solar cells are classified into Monocrystalline [49], Polycrystalline [50] and Thin-Film solar cells [51]. Among these, Monocrystalline Si cells have efficiencies of over 20% [52], while that of Polycrystalline Si cells and Thin-Film solar cells are 15–17% and 7–15% respectively. However, recently several contemporary solar cell technologies have emerged and have demonstrated promising results. Organic Solar cells (OSCs) are one such type of contemporary solar cell and prove to be advantageous due to their flexible and lightweight nature, along with the low cost of fabrication [53]. Initially, fullerenes were employed as electron-acceptors in OSCs, however, they faced certain disadvantages, some of which were large energy losses in the OSCs [54], and weak absorption in the visible regions of the spectrum [55]. Hence, these shortcomings paved the way for the development of non-fullerene acceptors and polymer donors in OSCs, having remarkable photovoltaic properties. In such fullerene-free cells, both the non-fullerene acceptors and electron donors provide a combined effort in achieving excellent efficiencies [56]. The energy losses in NF-acceptor OSCs can be reduced to below 0.5 eV due to the small driving force associated with the charge generation in this case. Hence, an efficiency of 20% or more can be surmised, as compared to efficiencies of hardly 10% that are achieved by fullerene-based OSCs [57]. Nevertheless, OSCs are still facing several challenges in terms of their practical usage, owing to low stability [58] and high cost in comparison to traditional inorganic solar cells. Attempts are being made to reduce the material cost of such cells worldwide. Although OSCs are not as stable as commercialized inorganic cells, their lifetime is sufficient for some applications with lower lifetime and cost requirements, and further research in the aforementioned fields can help in the advancement of this emerging PV technology.

Another next-generation PV technology constitutes organic/organic-inorganic Perovskite solar cells. These cells have seen massive growth in the PV sector over the years. Metal-halide perovskite-based solar cells are being researched extensively due to their inherent advantages including high efficiencies. In addition to this, these cells are advantageous over traditional organic and inorganic solar cells due to the low cost of fabrication and materials [59]. Metal halide-based Perovskite devices

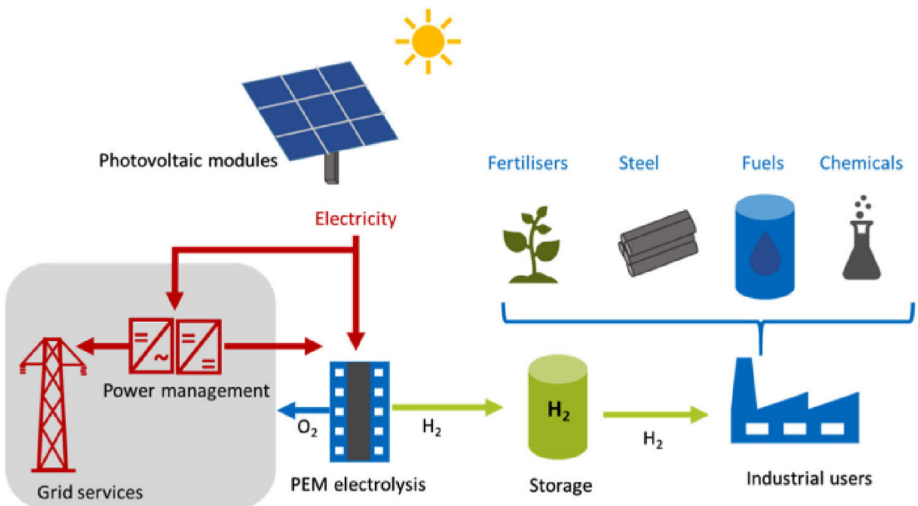


Fig. 3 – Schematic representation of synthesis and applications of Green Hydrogen [45]. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

exhibit high performance as a result of their excellent opto-electronic properties including long lifetimes of charge carriers, high absorption coefficient, and high defect tolerance. However, perovskite solar cells still face several issues with their durability and stability and additional research is necessary to bring about transformative changes in enhancing their performance [59]. Furthermore, Dye-Sensitized Solar cells (DSSCs), Quantum-dot sensitized solar cells (QDSSCs) and other tandem solar cell technologies are also undergoing extensive research. In DSSCs, efficiencies of around 14% have been obtained, whereas, in QDSSCs, they have been around 8–10% [60]. However, since DSSCs employ a liquid electrolyte, their production is currently limited. But it is expected to improve with further advancements in research and technology [61].

Several studies are still being carried out to increase the efficiencies of these cells and overcome the limitations associated with them. The PV energy market has seen exponential growth due to ambitions set by the government for the control and abatement of CO_2 emissions and the production of renewable energy. Also, the International Energy Agency has predicted that solar energy might become the world's largest source of electricity by 2050 [62].

Electrolyzer systems

The electrolysis process involves the splitting of water molecules into hydrogen and oxygen by passing a direct current through water. When the energy to carry out this process is acquired from a renewable energy source (per se solar energy), the hydrogen obtained is termed green hydrogen. An electrolyzer is used for this purpose and it consists of two electrodes separated by an electrolyte, which thereby increases the ionic conductivity [63]. The electrodes are required to possess the following characteristics:

- good electrical conductivity
- corrosion resistance
- structural integrity
- catalytic properties

The electrolyte employed must be inert to the electrodes so that it is not susceptible to any changes during the entire process. The electrolysis process also necessitates the use of a diaphragm (also known as a separator) to prevent the hydrogen and oxygen produced from recombining. The diaphragm also protects the electrodes from short-circuiting. It must have a strong ionic conductivity and should be physically and chemically stable. The electrodes, electrolyte and diaphragm, all constitute the electrolytic cell or electrolyzer.

To summarize, an electrolyzer is an electrochemical device that transforms electrical and thermal energy into chemical energy that is stored inside the fuel (as green hydrogen). Green hydrogen gives out zero emissions during its entire process of production and hence the research in this field is sought so much [64]. Electrons are captured or released by ions at the surface of the electrodes during this process, resulting in a multiphasic gas–liquid–solid system. At the cathode, the reduction-half reaction or the hydrogen evolution reaction (HER) occurs and hydrogen is generated, while at the anode the oxidation-half reaction occurs and oxygen is generated. The electrons flow from the anode to the cathode, to complete the electrical circuit [63].

Green hydrogen currently can be produced by electrolysis using three major types of electrolyzers: Alkaline water electrolyzer cell (AWEC), Solid oxide electrolyzer cell (SOEC) and Proton exchange membrane electrolyzer cell (PEMEC). A basic overview of the same has been demonstrated in Table 1.

We would be studying these electrolyzers in detail now [67].

Alkaline water electrolyzer cell (AWEC)

AWEC is one of the most mature technologies and has been conventionally in use worldwide. They are safe and reliable and possess a long stack lifetime [68]. The operating temperature typically varies from 40 to 90 °C and exhibits an efficiency of around 70–80%. KOH and NaOH-based electrolytes are the commonly used electrolytic solutions for AWECs with concentrations ranging from 20 to 30% [69]. For porous diaphragm separators, the most commonly used materials are asbestos, ceramic, and polyethylene. However, due to the health and environmental concerns associated with asbestos, its use has been largely phased out in recent years. Ceramic diaphragms are still used in some applications, but they are more expensive and fragile than polyethylene diaphragms, which are now the most widely used material for porous diaphragms in alkaline electrolyzers [70,180].

The operating principle and working of AWECs in the electro-dissociation of water into hydrogen have been demonstrated in a review [67]. It states that, at the cathode, 2 mol of water are reduced to produce 2 mol of hydroxyl ions and 1 mol of hydrogen gas. At the anode, 2 mol of hydroxyl ions are oxidized to form half a mole of oxygen gas by liberating two electrons. This is chemically demonstrated by the reactions given below:

$$4H_2O + 4e^- \rightarrow 2H_2\uparrow + 4OH^- \text{ (At cathode)}$$

$$4OH^- \rightarrow O_2\uparrow + 4e^- + 2H_2O \text{ (At anode)}$$

Water is fed and reduced at the cathode which yields hydrogen gas and hydroxyl ions. These hydroxyl ions move towards the anode circulating through the diaphragm membrane under the applied electrical field externally and recombining on the surface of the anode to produce oxygen gas. This process liberates electrons at the anode, thus completing the electrical circuit [38,67]. Table 2 compiles a list of the advantages and disadvantages of AWECs.

AWECs' potentiality has been explored in various sectors of the energy portfolio however, their large-scale adoption or application is yet to be enhanced owing to their low-rate capabilities, instability, susceptibility to corrosion and insufficient conductivity. In pursuit of resolving these issues while not compromising the capabilities of AWECs, attention is directed toward developing promising alternative technologies. Some studies have claimed that on using Anion exchange membranes [71] the rate capabilities have improved considerably while some have suggested the use of ion-solvating membranes to resolve these issues [72]. Nevertheless, there have been no studies that concretely claim that a particular technology counters the issue and can aid the large-scale adoption of AWECs. And hence, significant attention is to be directed towards achieving this and coming up with alternative technologies that support the application of AWECs in the energy sector.

Solid oxide electrolyzer cell (SOEC)

SOEC has received significant attention over the years owing to the highly efficient production of ultra-pure hydrogen. SOEC

Table 1 – Characteristic Overview of various Electrolyzer technologies [65,66].

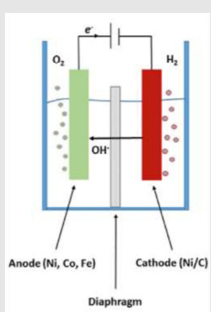
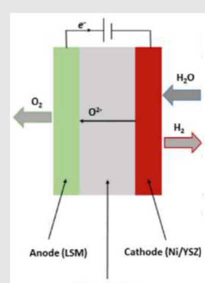
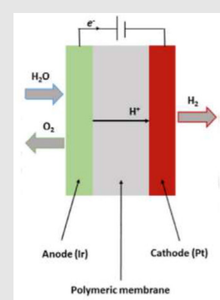
	Alkaline	SOECs	PEM
			
Cathode reaction	$2H_2O + 2e^- \rightarrow H_2 + 2OH^-$	$H_2O + 2e^- \rightarrow H_2 + O^{2-}$	$2H^+ + 2e^- \rightarrow H_2$
Anode reaction	$2OH^- \rightarrow 0.5O_2 + H_2O + 2e^-$	$O^{2-} \rightarrow 0.5O_2 + 2e^-$	$2H_2O \rightarrow O_2 + 2H^+ + 2e^-$
Electrolyte	KOH/NaOH (L)	Ceramic (S)	Polymer (S)
Anode	Ni	LSM-YSZ, $CaTiO_3$	Pt, Ir, Ru
Cathode	Ni alloys	Ni-cermets	Pt, Pt/C
Charge Carrier	OH^-	O^{2-}	H^+
Cell Voltage	1.8–2.4 V	0.95–1.3 V	1.8–2.2 V
Operating Pressure	2–10 bar	<30 bar	15–30 bar
Operating Temperature	60–90 °C (up to 200)	500–1000 °C	50–90 °C
Efficiency (HHV)	62–82%	~90%	67–84%
System Lifetime	20–30 yrs	–	10–20 yrs
Advantages	• Most mature • Long-term stability • Low capital cost	• High energy efficiency • Low capital cost • Reversible (can be used as a fuel cell)	• Dynamic operation • High current density • Rapid response
Disadvantages	• Corrosive electrolyte • Permeation of gas • Low current density	• Bulky design • Instability of electrodes • Sealing issues	• High electrode cost • Acidic environment • Low durability

Table 2 – Advantages and disadvantages of AWECs [38,67].

Advantages of AWECs:	Disadvantages of AWECs:
Conventionally used advanced technology.	Low current density (A/cm^2).
Produces hydrogen of highest purity with purity ranging from 99.7 to 99.9 vol%.	Susceptible to corrosion.
Electrocatalysts used are inexpensive.	Chances of carbonate depositions on the electrodes further affect the performance of the cell.
Extensively commercialized process.	Low operating temperature.
	Low operating pressure.
	Less dynamic operation.

performs steam electrolysis as compared to water electrolytic methods and operates at very high pressure (up to 3 MPa) and temperatures (typically ranging from 600 to 1000 °C) [73]. SOEC comprises a solid ceramic electrolyte that gives it a sleek design [38]. The working of SOEC and the conversion of electrical energy into chemical energy has been schematically demonstrated in a review [67]. It is stated that, at the cathode, water is reduced to produce hydrogen gas and oxide ions. These oxide ions circulate through the electrolyte and move towards the anode under the externally applied electric field. Oxide ions recombine at the anode to produce oxygen gas by liberation of electrons, thus completing the electrical circuit. Equations below chemically illustrate this process [74]:

$$H_2O + 2e^- \rightarrow H_2\uparrow + O^{2-} \text{ (At cathode)}$$

$$O^{2-} \rightarrow \frac{1}{2}O_2 + 2e^- \text{ (At anode)}$$

Strontium and Zirconia-doped lanthanum magnetite materials are widely in use as cathodic materials whereas, at the anode, zirconia is generally used [75]. In a SOEC cell, water consumption is roughly 80 times higher than hydrogen production. The primary reason for this is the cooling requirements due to the high-temperature operations of SOEC [76]. This technology is, however, yet to be commercialized due to some drawbacks that are discussed below. Table 3 compiles a list of the advantages and disadvantages of SOECs.

SOECs performances are often subjected to degradation due to the gradual delamination of the lanthanum strontium manganite (LSM) oxygen electrode by the electrolyte. This further results in the reduction of active sites and an increase in ohmic resistance all contributing to performance degradation. The delamination of LSM oxygen is attributed to several factors however, it is believed that the solid-solid two-phase interface that is subjected to a very high oxygen pressure build-up is the primary contributor [78]. Over the years several studies have been conducted to reduce this high-pressure build-up by several approaches. Some of which include the introduction of a composite electrode [79], nanoscale and nano-structured electrodes [80] and the use of several different combinations of electrodes [81]. However, most of these studies do not have experimental literature to support the hypothesis and this creates a massive knowledge gap. Efforts should be conducted to bridge this gap and find an amicable solution to prevent the degradation of SOECs' performance.

In addition to this, SOECs are not economically viable and not competitive with fossil fuels and large-scale adoption of SOECs raises concerns owing to their salient characteristics of high-temperature operation. Lowering the operating temperature, however, reduces the SOEC's performance. And, hence, extensive research is to be carried out to find new and low-cost electrolyte and electrode materials that can retain their conductivity and catalytic capabilities at lower temperatures while attaining similar or better efficiency than traditional high-temperature materials [82].

Proton exchange membrane electrolyzer cell (PEMEC)

PEMEC is the most promising method to convert electrical energy derived from renewable energy sources into chemical energy in the form of green hydrogen [83]. It is often also termed a Polymer electrolyte membrane electrolyzer as the cell makes use of a gas-tight thin electrolyte membrane made of polymer to separate the end products. Typically, PEMECs operate at low temperatures ranging around 20–100 °C and demand less energy due to their high-pressure operation (up to 40 MPa) which rules out the need for compressing. The electrolyte membrane is selectively permeable to only charge carriers, in this case-protons, thus preventing gases from intermixing [84]. The electrolysis of water by PEMEC has been demonstrated in a review [67]. It is stated that, at the anode, water is oxidized to produce oxygen gas and protons by liberating electrons. These protons travel through the membrane towards the cathodic side under the influence of the externally applied electrical field to recombine with the electrons and reduce to hydrogen gas and thereby, completing the electrical circuit. This is chemically illustrated in the reactions below:

$$H_2O \rightarrow \frac{1}{2}O_2\uparrow + 2H^+ + 2e^- \text{ (At anode)}$$

Table 3 – Advantages and disadvantages of SOECs [77].

Advantages of SOECs	Disadvantages of SOECs
High-efficiency production.	Large and heavy system design.
The catalyst used is non-noble.	Currently, the production is at a laboratory scale.
High-pressure operation.	Durability is low.
	The structure is brittle due to the use of ceramic materials.

$2H^{+} + 2e^{-} \rightarrow H_2\uparrow$ (At cathode)

For the HER at the cathode, precious metals such as Platinum (Pt) and its oxides are used whereas, at the anode for the OER, precious metals like Iridium (Ir) and its oxides are generally used [85]. The other metals employed as electrocatalysts are Gold (Au), Palladium (Pd), Rhodium (Rh) and Ruthenium (Ru). The amount of water required for cooling in this process is the least among other processes that further reduce the total water consumption roughly falling around 11 times that of total hydrogen produced which is far less than that of SOECs [38]. Some inherent advantages such as high current density, compact design, very high efficiency, low operational temperatures, very high operational pressures, fast response, and production of hydrogen gas of the highest purity (as high as 99.999 vol%) [86] make PEMECs the most sought-after technology for green hydrogen production [87]. Operation at very low current densities is also possible with PEMECs. This is because the polymer electrolyte membrane possesses very low gas permeability, thus, minimizing the risk of gas intermixing or the production of flammable gas mixtures. Another exceptional advantage of PEMECs is their ability to work under power fluctuations. There is a quick response in the proton transfer across the membrane and hence, it performs well under variable power supply as opposed to AWECs which perform adversely under power fluctuations [67,88].

Furthermore, the compact design structure of PEMECs facilitates faster cool-off time and heating-up also takes place at a quicker rate. The response time is also found to be shorter and the chances of gas intermixing are further minimized. The proton transport route is also found to be much shorter due to the sleek nature of the electrolyte membrane. This further lowers ohmic losses considerably. The ability of PEMECs to work at higher current densities facilitates in reduction of operational costs. This also proves beneficial in differential pressure operations. Under no circumstances is the oxygen side being pressurized, only the hydrogen side is under pressure. This further prevents accidents/hazards related to oxygen pressurizing and also prevents the risk of self-ignition [88,89]. As previously mentioned, the high-pressure operation reduces the cost and energy that would then otherwise be used for the storage and compression of hydrogen gas. It significantly improves the removal of product gas and shrinks the volume at the cathode. The high-pressure operations further bring advantages by maintaining the integrity of the electrolyte membrane as at high pressures, the membrane does not expand and the chances of dehydration are also low [88,90].

Table 4 comprehensively consolidates some recent advancements made in the electrolyzer sector.

PEMEC components

The major components of a PEMEC are current collectors (gas diffusion layer), membrane electrode assembly (MEA) and separator plates. A complete overview [178,179] of a typical PEMEC comprising its components: current collectors, electrocatalysts, separator plates and MEA is shown in Fig. 4.

Membrane electrode assembly (MEA). MEA is considered the heart of the PEMEC and consists of the membrane, electrocatalysts at the anode and cathode and ionomer solution. It accounts for 24% of the total cost of the cell [67,88]. Some of the most commonly used membranes are made up of Perfluorosulphonic acid polymers such as Fumapem®, Nafion®, Flemion®, etc [107]. These membranes possess some unique properties such as high thermal stability, high oxidative stability, high durability, great mechanical stability, stability at high current densities and majorly, excellent proton conductivity [108]. The ionomer solution is added primarily to promote proton transfer, thereby reducing ohmic losses and eventually increasing cell efficiency. In addition to that, the ionomer solution displays excellent characteristics as a binder that provides mechanical stability to the catalysts as well as electrodes. However, optimization of the ionomer layer is of utmost importance. This is due to the electron-resistant nature of ionomer solution which results in a decrease in electrical conductivity [109].

Current collectors. After the oxidation of water into oxygen and electrons, electrons travel through the current collectors to the anode where they recombine with the protons to form hydrogen gas. Hence, the role of current collectors is crucial in the smooth running of the entire PEM cell. Generally, porous titanium plates are used as current collectors because of their exceptional properties such as good resistance to an acidic environment, good mechanical strength, corrosion resistance and great electrical conductivity [110]. These titanium plates/disks act as current collectors and are attached to each side of the MEA. The plates also act as a gas diffusion layer and are encompassed by bipolar plates and carbon gaskets that act as cell sealants [111].

Separator plates. The separator plates or the bipolar plates are one of the most crucial components of the PEMECs and account for 80–85% of the total weight and volume of the cell. The primary functions of separator plates include the distribution of reactant gas across electrodes for diffusion, removal of water and excess heat, providing electrical connection between cell stacks and finally providing mechanical support to the cell [111]. Typically, these plates are made up of graphite, stainless steel, and titanium. Generally, titanium is preferred over others due to its mechanical strength, resistance to an acidic environment and good electrical conductivity [112].

Recent advancements in PV-electrolyzer systems

Photovoltaic (PV) electrolyzers systems have emerged as a promising technology for sustainable hydrogen production, utilizing renewable energy sources such as solar energy to power the electrolysis process. The principle behind PV-electrolysis systems is simple: photovoltaic cells convert solar energy into electricity, which is then used to power an electrolyzer that splits water into hydrogen and oxygen. This process produces high-purity hydrogen without any greenhouse gas emissions or pollution. The resulting hydrogen can

Table 4 – Recent advancements in the electrolyzer sector.

Sl. No.	Title of the Article	Key Findings	Ref.
1.	Unveiling Trifunctional Active Sites of a Heteronanosheet Electrocatalyst for Integrated Cascade Battery/Electrolyzer Systems	With high round-trip efficiency (61%) and high faraday efficiency (96%), power storage in the battery mode and H_2 generation in the electrolyzer mode were effectively and efficiently reversed with the incorporation of ReS_2/NiFe-LDH hetero-nanosheets as electrocatalysts. This system exploits the excellent trifunctional active sites' characteristics of ReS_2/NiFe-LDH hetero-nanosheets in the development of an integrated electrolyzer-battery system.	[91]
2.	Techno-economic Analysis of a More Efficient Hydrogen Generation System Prototype: A Case Study of PEM Electrolyzer with Cr–C Coated SS304 Bipolar Plates	Effective and economical production of hydrogen with the use of Cr–C coated SS304 bipolar plates and a septic mixture comprising urea, ammonia, and methyl alcohol as a chemical solution in the electrolysis cell along with mounting of super strong magnets on the outer surface of the electrolyzer cell.	[92]
3.	A membrane-free flow electrolyzer operating at high current density using earth-abundant catalysts for water splitting	H_2 generation with purity as high as 99.1% with the incorporation of FeP–CoP/NC bifunctional electrocatalyst with a sandwich-like structure in the membrane-free flow electrolyzer cell. The electrocatalyst possesses several advantages such as being highly effective, economical and a good resource in improving the current densities of the electrolyzers.	[93]
4.	Platinum-catalyzed Nb-doped TiO_2 and Nb-doped TiO_2 nanotubes for hydrogen generation in proton exchange membrane water electrolyzers	Nb-doped TNTs and Nb-doped TiO_2 were produced and studied as supports for Pt nanoparticles in water electrolysis for the HER electrocatalysis. TiO_2 and TNTs were found to have high specific surface areas due to the addition of Nb and thereby improving the performance of the electrocatalysis significantly.	[94]
5.	Development of a high-pressure membrane-less alkaline electrolyzer	The technology of membrane-less alkaline electrolysis is achieved with the introduction of a cyclic redox process resulting in the periodic generation of high-pressure hydrogen and oxygen in the electrolysis cell that is achieved by the use of active electrodes which reversibly react with oxygen. This system inherently avoids the simultaneous evolution of hydrogen and oxygen gas thereby eliminating the need for a separating mechanism further reducing the material and energy costs.	[95]
6.	Graphite/RGO coated paper *m*-electrolyzers for production and separation of hydrogen and oxygen	Combined with a photovoltaic (PV) cell, microfluidic electrolyzers made of graphite-covered paper electrodes were developed to electrolyze sea water into O_2 and H_2. The micro-electrolyzer fabricated is a cost-effective, metal-free and flexible device that performs electrolysis of water at a significantly lower applied voltage with a 1–2% efficiency.	[96]
7.	"Carbohydrate-Universal" electrolyzer for energy-saving hydrogen production with Co_3FeP_x@NF as bifunctional electrocatalysts	Bimetal-based Co_3FeP_x nanowires on Ni foam (Co_3FeP_x@NF) were produced and assessed as "carbohydrate-universal" earth-abundant, transition metal-based, bifunctional electrocatalysts that can catalyse COR and HER in a single electrolyzer in this study.	[97]
8.	Super hydrophilic porous transport layer enhances the efficiency of polymer electrolyte membrane electrolyzers	The findings show how the wettability of commercial porous transport layers (PTLs) affects mass transfer in PEM electrolyzers, allowing them to become super hydrophilic. At high current operation, super hydrophilic PTLs boost the PEM electrolyzers' efficiency by more than 11% (up to 20%).	[98]
9.	Bifunctional oxovanadate doped cobalt carbonate for high-efficient overall water splitting in alkaline-anion-exchange-membrane water-electrolyzer	In an alkaline-anion-exchange-membrane-water-electrolyzer, the self-supported oxovanadate-doped cobalt carbonate ($VCoCO_x$@NF) on nickel foam (NF) shows considerably strong activity for both oxygen and hydrogen evolution processes (HER and OER). It is a highly efficient catalyst and demonstrates great performance in alkaline media water electrolysis.	[99]
10.	Transition metal atom–doped monolayer MoS_2 in a proton-exchange membrane electrolyzer	Under typical circumstances of severe acidity, exposed basal planes of MoS_2 monolayer nanosheets with transitional metal dopants (Fe, Co, Ni, Cu) were employed as cathode catalysts for proton-exchange membrane (PEM) water splitting in an electrolyzer. In an electrolyzer, Co-$^{s}MoS_2$ produces the maximum current density when compared to others, with HER activity equivalent to that of commercially available cathode catalysts.	[100]
11.	Cellulose nanocrystals–blended zirconia/polysulfone composite separator for alkaline electrolyzer at low electrolyte contents	In Alkaline water electrolysis, a cellulose nanocrystal (CNCs) - blended Zirconia/Polysulfone composite porous separator demonstrates both low area resistance and gas permeability. Hydrophilic cellulose nanocrystals were effectively integrated into a hydrophobic polymer network, resulting in an excellent performance at low electrolyte concentrations.	[101]

(*continued on next page*)

Table 4 – (continued)

Sl. No.	Title of the Article	Key Findings	Ref.
12.	Ion exchange capacity controlled biphenol-based sulfonated poly(arylene ether sulfone) for polymer electrolyte membrane water electrolyzers: Comparison of random and multi-block copolymers	Hydrocarbon-based sulfonated poly(arylene ether sulfone) proton conducting polymers are created by changing their ion exchange capacity as alternatives to the pricey and extremely permeable hydrogen PFSA membranes. These membranes have a lower hydrogen permeability and a somewhat better proton-to-hydrogen selectivity than PFSA membranes.	[102]
13.	Polybenzimidazole-crosslinked-poly(vinyl benzyl chloride) as anion exchange membrane for alkaline electrolyzers	The synthesis and characterisation of polybenzimidazole membranes, which are one of the major components of the futuristic zero-gap alkaline water electrolyzers are described in this study. These membranes are viable candidates for anion exchange membranes, and they can be improved to match the performance of current AWECs.	[103]
14.	Rhodium-based cathodes with ultra-low metal loading to increase the sustainability in the hydrogen evolution reaction	The goal of this research was to create high-performance, low-metal-loading catalysts by immobilising an Rh-based organometallic complex (RhCp*Cl(phendiamine)Cl) on a carbon black substrate using a reliable synthesis process. The findings of this study pave the way for novel materials with very low metal loadings to replace traditional cathodes that are substantially loaded with metals in electrolytic HERs.	[104]
15.	Self-assembled $RuO_2@IrO_x$ core-shell nanocomposite as highly efficient anode catalyst for PEM water electrolyzer	The goal of this research was to develop and build a self-assembled $RuO_2@IrO_x$ core-shell heterostructure nanocomposite that can mediate OER activity while remaining durable. Some of the advantages include higher specific activity, larger mass activity, smaller overpotential and lower cell potential.	[105]

be used as a clean fuel for transportation, power generation, and various industrial applications.

Recent studies have focused on the performance evaluation of PV-electrolysis systems under various operational conditions. A study provides a comprehensive review of state-of-the-art PV-electrolysis systems and their performance under various operational conditions. The study emphasizes the importance of optimizing operational parameters, such as temperature, pressure, and electrolyte concentration, to improve the efficiency and reliability of these systems [114].

Another study highlights the key components and challenges associated with the development of PV-electrolysis systems. The article provides insights into the design, operation, and optimization of these systems, emphasizing the need for integrating with energy storage technologies to ensure uninterrupted hydrogen production and supply [113]. Fig. 5 demonstrates the components and their connections in a PV-Electrolyzer system. We will study the integration of Solar PV systems with individual electrolyzers in the subsequent sections.

Integration with alkaline electrolyzers

Alkaline Electrolyzers has been a frontrunner in PV-Electrolyzer systems and several different combinations have been studied rigorously. Hence, it becomes essential to compile all this research, and efforts have been made to achieve the same. Several approaches for direct coupling of PV and electrolyzers for direct solar hydrogen production, with varying levels of technical maturity and integration of components, were investigated in a study. The results have been summarized in Table 5.

In general, improving the PV module efficiency, and limiting the efficiency losses associated with integration and scale-up can enhance the STH efficiency of the systems mentioned in Table 5. Enhancing the durability of electrolyzers also requires additional research [45].

Generally, alkaline electrolyzers are coupled with PV systems by making use of an inverter. The voltage of the PV system must be in conjunction with the I–V curve of the electrolyzer for maximum optimization, as depicted in Fig. 6. The electric losses of the system are represented by any discrepancies between the PV module's maximum power point (MPP) curve and the alkaline electrolyzer's I–V curve. The MPP curve and the I–V curve should overlap in an ideal design [115].

Some studies have suggested that there is a rise in the temperature of the electrolyte when the system functions during peak hours of the day. This occurs due to an increase in the power production of photovoltaic modules. This trend favours hydrogen production since a higher rise in the electrolyte temperature results in a decline in power required for electrolysis [116].

The energy efficiency for this particular system is in the range of 8–16% [117], which is still very low when compared to other conventional energy production systems. Hence, a thorough study is to be conducted in pursuit of enhancing the efficiency and performance of the PV-alkaline electrolyzer system.

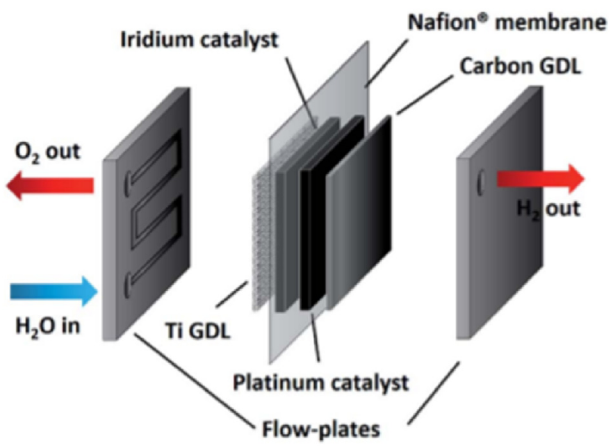


Fig. 4 – Overview of a PEM electrolyzer cell [106].

Integration with PEM electrolyzers

PEM electrolyzers have received extensive attention owing to their exceptional properties such as higher efficiencies, higher rates of production, and compact design. Hence, integrating PEM electrolyzers with PV systems proves to be beneficial. A study was conducted wherein a Bifacial Silicon Heterojunction Photovoltaic Module was coupled with a PEM electrolyzer for the production of hydrogen [45]. The study resulted in an STH efficiency of 17% with the average STH efficiency ranging around 14%. The system was stable and had a run-time of 55 h in the outdoor environment [118]. The second system devised in the same study consisted of commercial PV modules coupled with PEM electrolyzers. The system proved to be extremely flexible; depending on the number of modular units in an array, the sizing of the system could be varied. Over 9 months of operation, the amount of hydrogen produced was found to be 22 kgs and the system demonstrated an STH efficiency of 10% [45].

In another study, the PEM electrolyzer was directly coupled with concentrating photovoltaics without making use of power electronics. This system is advantageous in many aspects, some of which include the non-requirement of an electrical interconnection of multi-junction solar cells and various sun-tracking devices that would have been used otherwise. In addition to this, hydrogen production is not significantly affected by the failure of a single solar cell, while in the case of conventional systems, this breaks down the entire system. Furthermore, the water flowing through the electrolyzer aids the thermal management of the entire system significantly due to its capability of dissipating the heat of the solar cell. This particular system demonstrates a high conversion efficiency of solar energy to hydrogen production owing to its unique design. The sunlight is effectively focused on the multi-junction solar cell by making use of a fresnel lens and the anode side of the PEM electrolyzer is directly connected to these solar cells [119]. The system has been demonstrated in Fig. 7.

To the best of our knowledge, PV-PEM electrolyzer systems are yet to be extensively explored and to better exploit the features of this particular system it is recommended to conduct thorough efforts in this domain.

Integration with SOECs

SOECs can have a major role in the PV-electrolyzer domain majorly because of their high efficiencies, high robustness, and non-requirement of precious metals for manufacturing. Solid Oxide Electrolyzer Cells have been integrated with Solar-Thermal-Photovoltaic (STPV) technologies [120]. STPV technology has undergone massive leaps in recent years [121]. Out of the two available methods of utilizing solar energy, one being photovoltaic technology and the other being solar collectors, this combines the advantages of both while avoiding their probable drawbacks. STPV absorbs solar energy in its intermediate module and re-radiates photons at a higher temperature with a wavelength that is now suited for the photovoltaic cell [122]. PV efficiency can be significantly increased by employing this novel method. Many experimental and theoretical studies on improving conversion efficiency have also recently been conducted [123–125].

In a study conducted, an integrated system of SOEC and an STPV device is proposed and researched, with the main product being pure hydrogen and the byproduct being nearly pure oxygen. The schematic diagram of this process is depicted in Fig. 8.

On the STPV device, a complete system model analysis was performed. A sensitivity study was also performed to assess the effects of temperature and current density on the performance of high-temperature electrolyzer cells. The findings suggested that a tradeoff should be made between cell potential, power density, and efficiency in relation to current density. In the planned condition, the STPV and SOEC were roughly 17% and 54% efficient, respectively. Better spectrum control and the capacity to operate at greater temperatures can improve the efficiency of these devices. They still only achieve 30–50% of their thermodynamic efficiency limit [126] and have the potential to improve efficiency through further shape and material optimization.

However, SOECs are still in the pre-commercial stage and hence there is a requirement of conducting further research is PV-SOEC coupled technologies. A novel PV-SOE power plant for generating hydrogen [127] has been researched, yet it is still at the laboratory stage and the feasibility of the plant under practical conditions needs to be assessed.

Utilization of power electronics in the PV-electrolyzer system

Photovoltaic (PV) electrolyzer systems have been proposed as a promising technology for sustainable hydrogen production. These systems use solar energy to power an electrolyzer, which splits water into hydrogen and oxygen. The produced hydrogen can be stored, transported, and used as fuel for various applications, such as in fuel cells and combustion engines. However, the performance of PV-electrolyzer systems depends on several factors, such as the efficiency of the PV panels, the electrolyzer efficiency, and the power electronics used to connect the PV panels and the electrolyzer [128,129].

One approach to improve the performance of PV-electrolyzer systems is to use power electronics to regulate the current and voltage between the PV panels and the

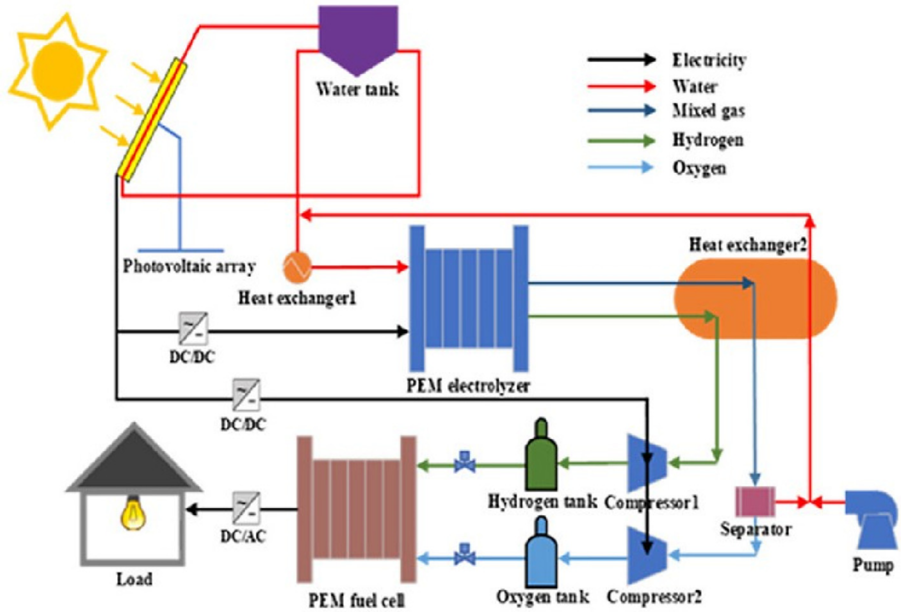


Fig. 5 – An illustration of a typical PV-Electrolyzer system [113].

electrolyzer. Power electronics are devices that can control and convert electrical power from one form to another, such as from DC to AC, or from high voltage to low voltage. In the context of PV-electrolyzer systems, power electronics can be used to optimize the power output of the PV panels and match it with the power demand of the electrolyzer [129,130].

Several studies have investigated the performance of PV-electrolyzer systems with and without power electronics. One study reviews different types of modelling used for proton exchange membrane electrolyzers and highlights the importance of using power converters to interface high DC bus voltage to low DC voltage required for PEM electrolyzers. The modelling of PEMEC is crucial for developing controllers for energy efficiency and hydrogen flow rate-based controls. The article discusses three types of PEMEC modelling - resistive, static, and dynamic load - and shows that the use of renewable energy sources implies different design constraints compared to stationary grid-supplied converters for electrolyzers. Dynamic load modelling is important to understand the behaviour of PEMEC in dynamic operating conditions due to weather changes when coupled with renewable energy sources. By considering these dynamics in designing the controller, the performance of the system could be greatly enhanced [131]. Fig. 9 illustrates how power electronics are integrated into the PEM electrolyzer systems.

Another study aimed to investigate the use of a load-managing PV system to reduce the cost of PV-driven hydrogen production. The proposed approach involves controlling the number of electrolyzer stacks connected to the PV array throughout the day without using power electronics, which reduces the cost and power losses. The study tested the approach on two Nel Hydrogen PEM electrolyzers, one with 4 stacks and the other with 8 stacks. The results showed that the load-management approach resulted in energy yields of 99.5% and 99.8%, respectively, which is a significant improvement over conventional PV systems that require inverters with peak efficiencies of 95%–98%. The approach also provided a greater energy yield than both power electronics-based PV systems and direct-coupled PV systems while maintaining the electrolyzer operating voltage within the acceptable range. Overall, the findings support the viability of implementing a load-management approach for driving hydrogen production with photovoltaics, which can significantly reduce the cost of producing hydrogen [132].

Furthermore, a study reports on the performance of PV-electrolysis systems using different types of PV modules and the use of power electronics. The findings show that direct integration of concentrated PV modules with electrolyzers leads to effective utilization of PV power, resulting in higher hydrogen production rates and solar-to-hydrogen conversion efficiencies compared to non-concentrated Si-based PV modules. Scaling up of the electrolyzers through direct coupling in series maximizes H_2 production and effectively utilizes PV power, while scaling up indirectly through power electronics resulted in voltage losses due to the mismatching of power points. Overall, the study highlights the importance of maximizing the utilization of PV power for efficient hydrogen production and suggests that the use of power electronics may not be as effective as direct integration in achieving this goal [133].

Additionally, a study also investigates the benefits of using power-optimizing electronic coupling between the PV array and the electrolyzer, which can increase annual gas yield by up to 5% even for optimally matched systems. The authors highlight the importance of optimal power matching of PVECs to maximize their gas yield and note that the quantitative benefit of electronic coupling is component-specific and requires a similar calculation of system output for accurate comparison among different coupling strategies [134].

In conclusion, PV-electrolyzer systems have the potential to provide a sustainable and renewable source of hydrogen

Table 5 – Technological advancements in PV-Alkaline electrolyzers systems [9].

PV Approach	Catalysts	PV Module Efficiency	Max. STH (Solar-To-Hydrogen) Efficiency	Challenges Associated
Triple-Junction Thin-Film Silicon	Bifunctional NiMoFe	–	4.7	Difficulty in replicating the design for a standalone electrolyzer
Silver-Doped $Cu(In,Ga)Se_2$	Bifunctional NiFe (LDHs)[b]	17.3	13.4	Optimization of the catalyst and the design layout is of utmost importance
Silicon-Based Heterojunction	NiMo (cathode)-NiFeO (anode)	17.6[a]	7[a]	The necessity of a heat exchanger between the electrolyzer and PV modules

[a] Calculated Using total PV area, b) layered double hydroxides.

fuel. However, the performance of these systems depends on several factors, such as the efficiency of the PV panels, the electrolyzer efficiency, and the power electronics used to connect the PV panels and the electrolyzer. The use of power electronics can improve the performance and efficiency of PV-electrolyzer systems by optimizing the power output of the PV panels and matching it with the power demand of the electrolyzer. However, further research is needed to optimize the sizing and configuration of the power electronics components and to develop new technologies that can further enhance the performance of these systems.

Challenges associated with the integration of PV systems with electrolyzers

PV-hydrogen production employing alkaline and PEM electrolyzers proves to be both advantageous and disadvantageous in their own aspects. This has been summarized in Table 6 and Table 7.

The PV-hydrogen energy system proves to be a viable approach to mitigate global warming concerns. For proper functioning, the system should be aimed to operate at its maximum output power levels [135]. Because PV output varies with solar irradiation, ambient temperature, and load current, all three elements must be taken into account while constructing a PV-Electrolyzer system [136]. A typical PV-Electrolysis system includes a solar generator, and an electrolyzer, along with other components (batteries, regulatory devices, MPPT, etc.). Batteries and electronic equipment are the most vulnerable components in this photovoltaic system and are prone to failure. Furthermore, they are extremely temperature sensitive. Hence, the way these systems are monitored has to be significantly improved [137].

The most significant issue with alkaline electrolysis is the delay in the cell stack's response when the input current is varied. The PEM electrolyzer, on the other hand, responds faster to changes in current density. Hence, it responds faster to fluctuations in PV energy. However, coupling PEM electrolyzers with solar PV modules is also subjected to certain limitations, one of them being the determination of the optimal configuration. This can be achieved by matching the MPP of the solar PV modules to the I–V characteristics of the PEM electrolyzer, which is carried out by an inverter constituting a DC-DC converter [138]. However, coupling with an inverter leads to a reduction in the overall system efficiency (due to an increase in the number of electrical devices).

The number of cells (including the electrolyzer assembly) in series and parallel can also be estimated for determining the optimal configuration. This system allows for maximum hydrogen production along with minimum energy losses [139]. Typically, this selection is made using a graphical approach, which involves intersecting the PV module's I–V curve with the PEM stack's load curve, as shown in Fig. 10.

Although effective, this approach proves to be intermittent, since variation in the radiation incident on the PV module leads to variation in the I–V curve of the module. Hence, hydrogen production varies as the incident radiation varies [140].

Solar Photovoltaics also pose certain disadvantages such as high initial costs of setting up solar PV arrays,

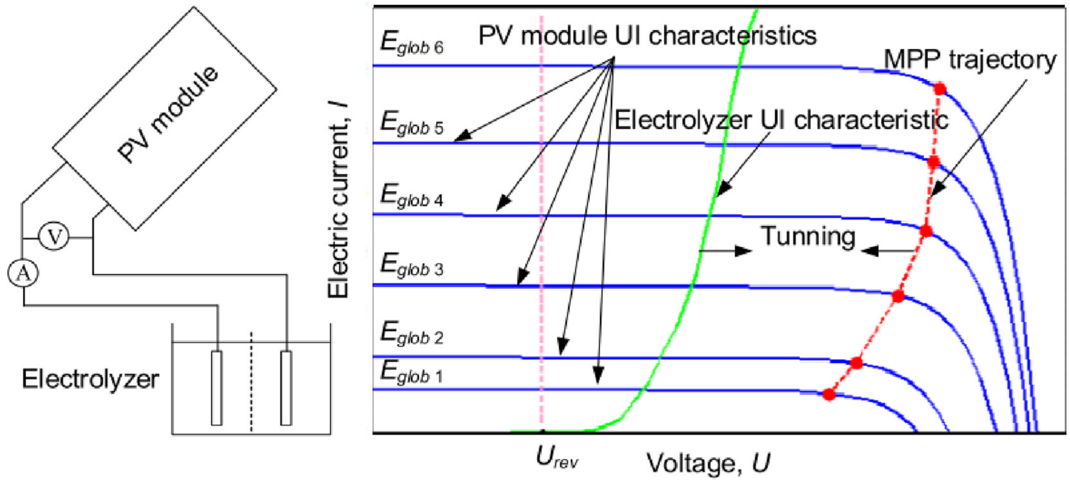


Fig. 6 – Optimization of the I–V characteristics of the PV system and electrolyzer [115]. E_{Glob}: Global Solar Irradiance (W/m2).

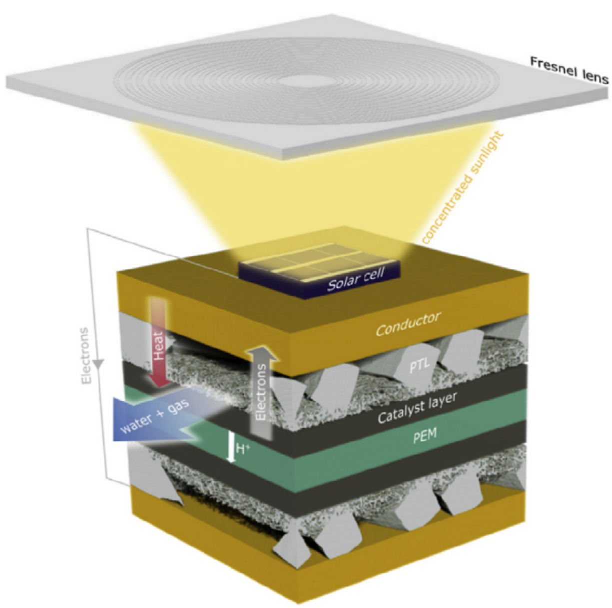


Fig. 7 – Schematic Representation of the direct coupling of the PV module and PEM electrolyzer [119].

unpredictable output and lack of economical-efficient energy storage. Moreover, solar energy is an intermittent resource, available only during the day. Hence to make these systems more viable, energy storage systems or battery banks are required to store energy and utilize it when sunlight is not available. However, such systems add to the life-cycle cost of the entire solar PV system [141]. Challenges like these, among many others, have to be catered to and solved, to integrate solar PV modules with electrolyzers successfully and generate green hydrogen.

Integration of an electrolyzer with a fuel cell

One attractive application of PEMECs is its integration with a PEM Fuel cell (PEMFC) into a regenerative fuel cell (RFC). The system is advantageous in many aspects and is one of the most effective ways of energy storage. Until now, AWECs have been a frontrunner in hydrogen energy storage and application due to their simple and conventional use [142]. However, several disadvantages of AWECs make it difficult to adapt them for large-scale energy operations and hence PEM is gaining extensive attention.

RFCs can be designed to have two different systems: a traditional system or a discrete RFC (DRFC) consisting of a discrete coupled stack and a Unitized RFC (URFC) [143]. When compared to URFCs, the traditional system (DRFC) is more efficient in operation since each cell stack is designed to execute a particular function; nevertheless, it is more expensive, large, heavy, and complex in design. The flexibility to develop the system based on the requirements is another advantage of classical DRFC. For example, if the RFCs are utilized as a power backup, the system will require a larger fuel cell and a smaller electrolyzer, however, if the RFCs are used as renewable energy storage, the electrolyzer and fuel cell sizes can be the same. Due to the usage of a single stack, URFCs have a cost and volume advantage, and they can reverse their functionality almost instantly. Furthermore, during the transition period, a DRFC generally requires battery backup. However, despite the advantages, there are no commercial applications for URFCs, this is due to efficiency concerns [144].

The discrete RFC system is highly efficient and has an energy conversion efficiency of around 40–60% [145]. Given the renewable nature of this system, it is an ideal replacement for applications only dependent on fluctuating and intermittent solar energy. A discrete RFC is a system that comprises a Fuel cell (FC) and an electrolyzer system. The FC makes use of hydrogen and oxygen as fuel which are obtained as a result of the electrolysis of water and uses it to produce energy. Such a device is often regarded as an H-ion battery. Heat and water are the only by-products produced in this process making it extremely environmentally friendly [146].

In general, RFCs find their applications in a plethora of fields: from use in several on-off grid systems as renewable energy storage to use as a backup system where an

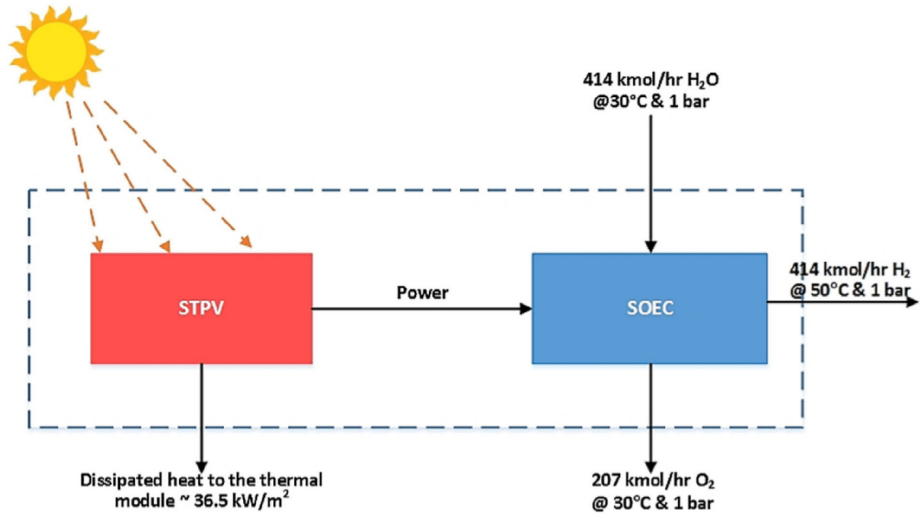


Fig. 8 – Schematic diagram of the SOEC-STPV integrated process [120].

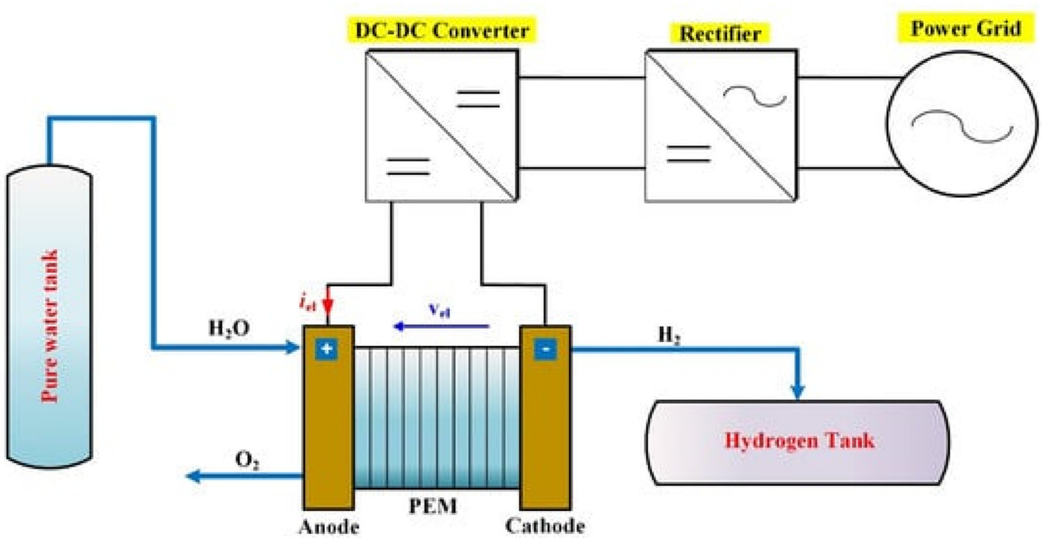


Fig. 9 – Integration of Power electronics in the PEMEC systems [131].

Table 6 – Advantages and Disadvantages of PV-Alkaline Hydrogen production [115].

PV-Alkaline Hydrogen Production	
Advantages	Disadvantages
Long-term stability	Low dynamics
Simple and scalable synthesis	Partial load
Low cost	Delay in cell stack responses
Mature and researched technology	

Table 7 – Advantages and Disadvantages of PV-PEM Hydrogen production [115].

PV-PEM Hydrogen Production	
Advantages	Disadvantages
Dynamic operation	High cost
Rapid response	Corrosive components
Good partial load	Noble metal catalyst employed
High voltage efficiencies	Low durability

uninterrupted power supply is required. The fact that it has no, or low, footprint makes it an ideal method for large-scale portable energy applications. It is a perfect fit for unmanned and remote applications because of its minimal maintenance requirement due to the lack of any moving parts, longer lifetimes, and silent operation [144]. Some other unique advantages of the RFC system include high specific energy density, longer lifecycle, and higher efficiencies for the charge/discharge cycle [146].

As illustrated in Fig. 11 a typical discrete RFC system consists of an electrolyzer, a fuel cell, and heat, gas, and water management systems.

Fig. 12 depicts typical discrete RFC and unitized RFC systems.

The power required for the electrolysis of water in the electrolyzer is supplied by a PV system and hence making it sustainable. Hydrogen gas evolved at the cathode and oxygen at the anode are fed to the FC as fuel with the assistance of their respective management systems. Since the electrolyzer and fuel cell are the core components of this system the efficiency and performance significantly depend on them [146].

Parameters to be considered

Oxygen vs air feed

It is determined by several experiments that using oxygen as feed during the fuel cell operation results in higher cell voltage, higher power density, higher efficiency and lower poisoning when compared to air [147]. And as oxygen is produced as a product of electrolysis it is always beneficial to store it and use it further as feed. This may be attributed to the high diffusion rates of oxygen when compared to air i.e., oxygen and nitrogen mixture. Another setback of using air as feed is the need for pumps. This further results in several other losses. However, the choice of using air or oxygen as feed entirely depends upon the application of the system and several other factors such as economic factors, size, environmental factors, etc [86].

Operating pressure

In a typical PEM fuel cell, there is a voltage gain with a rise in the operating pressure and generally, they work excellently at higher pressures [148]. And hence when air is used as feed it becomes necessary for the air to be pressurized and the use of pumps becomes inevitable. By using these pumps there is a negative effect on the voltage gain, thereby affecting the overall efficiency. Higher operating pressure also helps to avoid diffusion losses conveniently by operating at higher current densities [149]. However, the use of high pressure demands several system modifications such as the use of a thicker polymer membrane and other such parameters. And hence the decision of the selection of operating pressure is a difficult one [150].

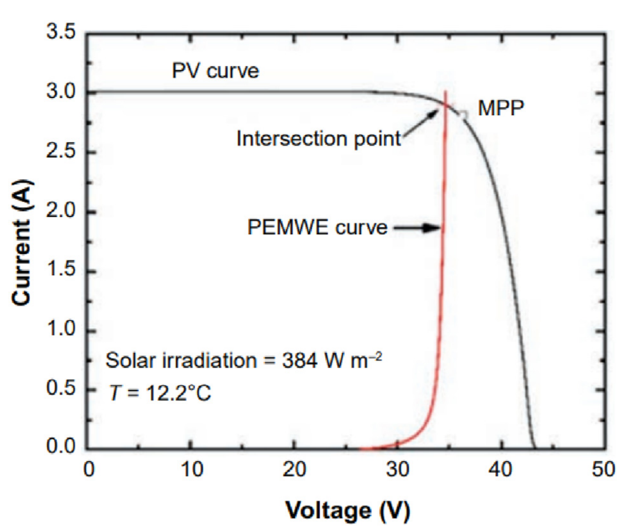


Fig. 10 – Graph depicting the intersection of the I–V curve of the PV module and PEM stack [139].

Nominal operating voltage

The nominal operating voltage of a fuel cell has a significant impact on the efficiency of the cell and the stack size. The stack size is somewhat proportional to the cell voltage. For a set of given conditions, a lower cell voltage always results in smaller stack sizes due to higher power densities [151]. A higher cell voltage also means lower fuel consumption i.e., better efficiency, which results in smaller storage and hence a smaller electrolyzer. However, a thorough techno-economic analysis is to be undertaken before determining the nominal cell voltage for a system [86,152].

Applications

RFCs have been looked upon as a strong contender for energy conversion and storage devices and extensive research is being carried out for the same. However, there are other tentative and potential applications of RFCs that intrigue many around the globe and are strongly studied. We will list some of those applications and study them briefly.

Aviation and aerospace industry

RFCs have demonstrated promising results in the aviation sector and have been applied in several areas over the years. The crucial factor of an aircraft is its mass and with the application of URFCs, this can potentially save a lot of volume and weight when compared to other energy systems employed for the same energy output. These systems can also be integrated with other renewable energy sources such as solar energy and wind energy. Considering the intermittent nature of these energy systems, RFCs can be a game-changer [153].

A study investigated the application of RFCs in high altitude long endurance unmanned aerial vehicles under practical assumptions for a continuous flight. It was found that for heights around the low-range stratosphere, RFCs-installed vehicles were successful for long flight duration. When compared with a PV system for the same conditions, the RFC system showed fewer fluctuations and was more stable throughout [154]. Similarly, in a research, a considerable study was performed on the feasibility of the implementation of RFCs in unmanned aerial vehicle propulsion. It was noted that it has high potential in low maneuverability and high endurance operations in an unmanned aerial vehicle. The consistency and quick start-up time of RFCs have great advantages and hence, in the coming years, they will be a part of the majority of applications in this industry [155].

Renewable energy

A particular research studied the application of RFCs in renewable energy storage systems. PEMRFCs typically those with oxygen (air) as cathode can be successfully optimized for power and energy independence. It was also noted that hydrogen RFCs will be an integral part of the energy infrastructure for long storage applications, especially in areas where volume and sizes are a constraint [156]. A study assessed the viability and analyzed the use of RFCs in

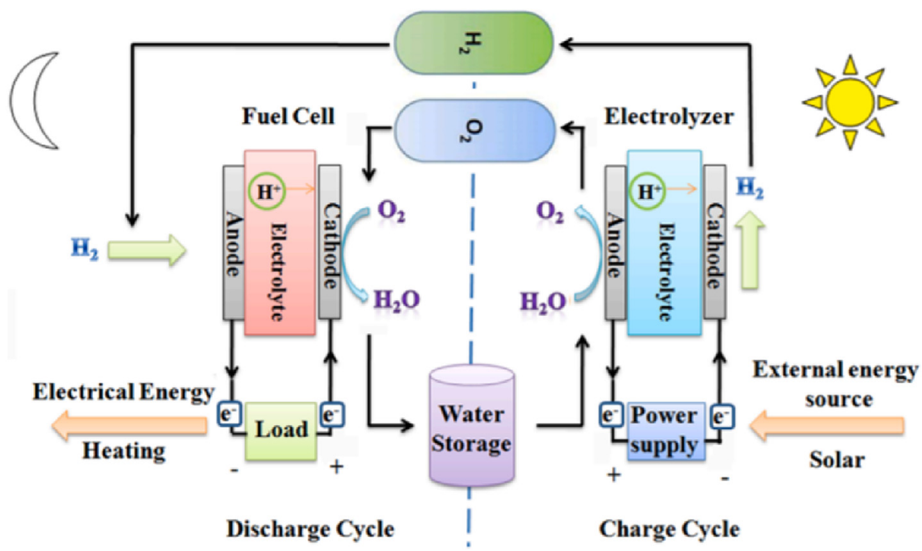


Fig. 11 – Schematic overview of an RFC system powered by PV energy systems [146].

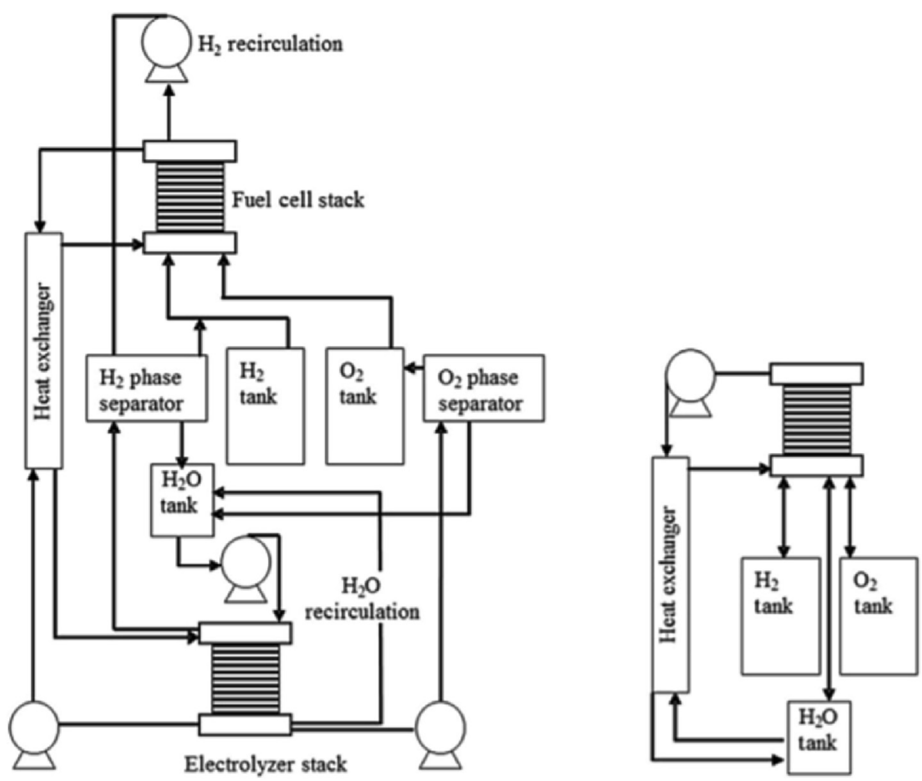


Fig. 12 – Schematic overview of a Discrete RFC (left) and a URFC (right) [144].

renewable residential power and energy applications under several operating parameters, different system configurations and strategies. RFCs were integrated with PV as a hybrid energy system and it was a complete utilization of power generated by PV and any extra power generated would produce hydrogen for the RFC system. The hydrogen generated would then act as a backup power supply in times with low irradiance and thereby, solving the problems of intermittency and the RFC system effectively increasing battery power efficiencies and density [157].

Automobile and transportation

One of the most promising and most studied applications of RFCs is in the automobile and transportation sector. The primary reason being the transportation sector accounts for a significant share of the total contribution towards GHG emissions and replacing it with a system that gives out zero emissions is always an ideal option. Hydrogen fuel can be produced during the off-peak period with the help of an electrolyzer and can be supplied to a fuel cell for power generation at the time of use. This system completely rules out the possibility of any emissions that are produced as a result of power generation using IC engines [158].

A study performed a differential economic analysis on the PV-diesel system vs. the PV-hydrogen system. The comparison stated that with exponentially rising fuel prices and with the rising concerns around the ecological impact of fossil fuels, RFCs offer a cost-effective option over PV-diesel configuration. As mentioned earlier, RFCs perfectly complement the PV system and eliminate the problem of intermittency [159]. A study compared and analyzed hydrogen and electricity as fuels for transportation. Several factors were rigorously analyzed such as economic viability, cost-effectiveness, durability, vehicle range and energy utilization. The main conclusion drawn from the article - there is no definite option for an alternative to conventionally used fuels. However, RFCs possess some tough advantages over other alternatives and can be superior fuel to electricity [160]. In addition to this, RFC-powered vehicles demonstrate strong potential in the transport sector in vehicles with high capacities such as busses, ships, trucks, etc. It is safe to assume RFCs are strong competitors of the traditional IC engine-powered vehicles considering the inherent disadvantages and detrimental environmental impact [161].

Challenges/research gaps associated with the application of unitized regenerative fuel cells and their solutions

Aside from the inherent benefits of URFCs, there are a few issues and limitations that must be addressed as soon as possible for URFCs to be used on a broad basis. The electrode design and catalysts employed in the URFC should be in the correct balance when developing it for a given application. The electrode is supposed to be flooded in the electrolyzer mode and dry in the fuel cell mode. This is owing to the very reversible nature of oxygen's redox processes, which results in a huge amount of polarization [40,162].

Secondly, in PEM URFCs, Pt has been the frontrunner as the choice for electrocatalyst owing to its excellent bifunctional properties which enhances the functioning and efficiency of the system [163]. Due to the expensive and rare nature of Pt, it cannot be considered a viable resource for electrocatalyst material [164]. To overcome this challenge many Pt-composites and structured materials are being investigated to produce a low-cost bifunctional electrocatalyst that works to a similar extent as Pt. Some of the most promising materials include Pt–Co nanoparticles [165], Pt–IrO_2 [166], Pt–Ni nano frames [167], Pt-Porous IrO_2 nanocomposites [168], Pt–Pb nanoplates [169], Pt nanowires [170], etc. However, the number of research studies in this domain is still very less considering the impact this technology possesses.

Thirdly, there is a high risk of corrosion of supportive materials. The performance of the electrocatalyst is strongly dependent on the supportive materials and hence they play a crucial role [171]. Generally, carbon materials such as carbon black with high electrical conductivity are used as support materials for Pt-based electrocatalysts [172]. Other supporting materials include carbon nanofibers [173], Pd/CNFs [174] and many more. High potential is applied when the URFC is in the electrolyzer mode which results in corrosion due to carbon in the supportive materials. Carbon corrosion obstructs the system's smooth operation, resulting in a significant reduction in efficiency. Several substitute supporting materials are being researched such as TiCN, TiC [175], carbon-free SiO_2–SO_3H [176], Sb-doped SnO_2 Aerogel-based material [177], etc. These materials can withstand high potential and are excellent supporting materials. In addition to this several other issues are to be resolved such as degradation of the polymer membrane and other components [68,69], corrosion of bipolar plates [181], catalyst layer composition [182], ionomer content [109,183], PTFE content [184,185] etc. Extensive research is being carried out to address these issues and find efficient alternatives.

In addition, the use of an ionomer solution (polymer solutions with ionic transport characteristics) has been discussed. It is considered that there are two conflicting impacts on the electrode in PEM electrolysis. One effect is that it encourages proton transport from the bulk of the catalyst layer to the membrane, enhancing overall efficiency by reducing ohmic losses. The ionomer also acts as a binder, providing mechanical stability and, as a result, longevity to the electrode [109,186]. On the other hand, the ionomer results in making the catalyst more hydrophilic, which could reduce mass movement out of the layer. To the best of our knowledge, only a handful of research has focused on optimization of the same to date.

For the successful large-scale application of URFCs, these are some of the prominent issues that are to be resolved. The issue of selecting the right catalyst is of significant importance. The catalyst should be compatible with oxygen and hydrogen production as well as consumption in the case of a URFC. Several catalysts that have bifunctional properties are sought after to tackle this issue and many have displayed promising results. The limitations are not only subjected to the choice of catalyst but also the electrode structure and the thickness of the catalyst. A thin catalyst helps to reduce ohmic losses as well as mass transport [187,188]. PEM URFC system demands a lot of technological advancements to present itself as a completely reliable and sustainable energy system. MEA is an integral part of the URFC system and protecting it from degradation while not compromising on other parameters is of utmost importance. Free radical scavengers are being applied and tested to prevent degradation of the membranes. These are primarily a series of compounds that react with detrimental chemicals thus, protecting the membrane [189].

Other improvements to the membrane include the introduction of a recombination layer [190] and internal reinforcement of a Polytetrafluoroethylene (PTFE) fabric [191] to make the membrane more durable and efficient. To ensure better operation and enhanced performance, the introduction of high-grade Ti-based porous transport layers (PTL) with microporous layers made up of Ti becomes imperative. PTLs facilitate good electrical and thermal conduction while enabling better gas and water transport. And hence, the performance of the system is highly dependent on the selection of these PTLs and the right selection could bring out transformative changes in the adoption of PEM RFCs [192].

Furthermore, to enhance the activity and utilization of precious metal catalysts, multicomponent nanowire alloys with a large surface area have shown promising results [193,194]. However, due to the difficulties in synthesizing these catalyst systems with these properties, the most prevalent strategies or approaches often demand the inclusion of sophisticated synthesis methods. And hence, studies are to be conducted in simplifying these intricate procedures and testing their potentiality. Incorporating these futuristic solutions will ensure a strong place for PEM URFCs in the energy portfolio.

Comparative analysis of solar PV-electrolyzers and wind power-electrolyzers systems for hydrogen production

Wind power-electrolyzers systems are starting to gain attention due to the increase in the production of wind energy. Wind energy is the fastest-growing renewable energy source in the world, with a global installed capacity of over 743 GW in 2020 [195]. Wind energy can be harnessed in different ways, such as onshore and offshore wind turbines. Wind turbines generate electricity, which can be used to power an electrolyzer to produce [196].

However, wind-powered electrolyzers have some limitations. One of the main challenges is the intermittent nature of wind energy. Wind energy is variable and dependent on weather conditions, which means that the electricity production from wind turbines is not constant. This variability makes it difficult to maintain the efficiency of the electrolysis process. Furthermore, the energy produced by wind turbines is usually transmitted through the grid, which results in transmission losses [197].

In contrast, solar PV-electrolyzers systems have some advantages over wind-powered electrolyzers. Solar PV-electrolyzers systems use solar energy, which is a reliable and abundant source of renewable energy. According to the International Renewable Energy Agency (IRENA), solar photovoltaic (PV) power is the most abundant renewable energy source globally, with the potential of generating 23,000 TW-hours (TWh) per year [198]. Furthermore, the use of solar PV-electrolyzers systems can reduce transmission losses, as the electricity produced by solar panels can be used directly for electrolysis without the need for transmission through the grid. This results in higher overall efficiency [199].

Several scientific papers and journals have highlighted the advantages of solar PV-electrolyzers systems over wind-powered electrolyzers. A recent study investigated the feasibility of using a solar photovoltaic (PV)-electrolyzer system for hydrogen production. The study found that a solar PV-electrolyzer system has higher energy efficiency and lower levelized cost of hydrogen (LCOH) compared to a wind-powered electrolyzer system. The authors attributed the higher efficiency of the solar PV-electrolyzer system to the direct conversion of solar energy into electricity without the need for mechanical energy conversion, which is required in a wind-powered electrolyzer system. Additionally, the study noted that a solar PV-electrolyzer system has a smaller footprint and can be easily integrated into existing buildings and infrastructure, making it a more versatile option for hydrogen production. These findings further support the advantages of solar PV-electrolyzer systems over wind-powered electrolyzer systems for hydrogen production [200].

Moreover, another study provides a comparative analysis of solar PV and wind energy systems for renewable hydrogen production. It highlights that solar PV-electrolyzer systems can achieve higher efficiency rates, which means they can produce more hydrogen per unit of energy input. Additionally, the cost of solar PV modules has been decreasing over the years, making these systems more affordable. On the other hand, wind energy systems require more space and are typically more expensive to install, but they can be more suitable for specific locations, such as offshore wind farms. The article concludes that both solar PV and wind energy systems have their advantages and limitations and can complement each other in achieving a sustainable hydrogen economy [201].

In conclusion, solar PV-electrolyzer systems seem to offer a more viable and efficient solution for renewable hydrogen production compared to wind-powered electrolyzers. Not only do they have a lower levelized cost of hydrogen (LCOH), but they also have a higher potential for cost savings and a lower carbon footprint. While wind energy systems have their advantages, the data suggests that solar PV-electrolyzer systems offer a more reliable and sustainable source of energy for hydrogen production. Nonetheless, it is important to consider that the suitability of each system will depend on specific locations and applications, and a combination of both systems might be necessary to achieve a sustainable hydrogen economy.

Conclusion

Current trends in energy demand and supply are not ecologically, economically, or socially sustainable. If not taken into immediate account, greenhouse gas emissions caused by humans would have more than doubled by 2050. There is a need to create a paradigm shift concerning the current energy outlook and renewable and sustainable energy sources have to be utilized widely. It is believed that the hydrogen economy is not a far-fetched notion and it is expected that the current hydrogen economy will expand to meet future demand. However, hydrogen today faces some fundamental issues in establishing itself in the energy sector, because of the energy losses occurring while converting electricity to hydrogen and the associated efficiency. On the other hand, hydrogen can be well adapted to decarbonization applications that cannot be decarbonized using renewable energy alone.

With the rising population and consequent rise in energy demand, hydrogen has announced itself as a strong contender, especially in the transport sector. Today, the transport sector accounts for a quarter share of greenhouse gas emissions and the numbers are set to increase if immediate reformations are not made. And hence, there is a huge market for the successful adoption of hydrogen however, there are some limitations pertaining to this particular approach that needs to be addressed at the earliest. Certain issues with this technology, limit its practical usage, such as component durability and the high cost of the rare elements used for its electrodes. SOECs were developed as an alternative, but require extensive research since they are still in their primitive stage. For the production of green hydrogen, the integration of solar pv system and electrolyzers is quite beneficial. The electrical matching between the pv systems and the electrolyzer stack is the key issue with the integration. Strategies to overcome this limitation have been put forth and are summarized in the review.

The current downward trend in solar electricity prices bodes well for the expansion of hydrogen generation. Solar hydrogen proves to be a clean alternative fuel that may replace conventional fuels and eliminate the need for oil and gaseous fuels, reducing CO_2 emissions and preventing global warming. In terms of environmental impact, solar PV-electrolyzer systems have a significantly lower carbon footprint (around 15 times) compared to fossil fuel-based energy generation sources.

Extensive research has been conducted to explore various aspects of PV-hydrogen coupling, specifically the integration of solar energy with hydrogen generation using electrolyzers. This integration has the potential to revolutionize energy systems, with applications such as hydrogen automobile refuelling stations and the creation of green cities. In addition, numerous economic studies have been conducted to assess the performance of PV-Hydrogen coupling. Through this review, an effort has been made to compile the research that has taken place and the challenges encountered while integrating PV-Electrolyzer-Fuel Cell systems. However, significant collective efforts between industry and academia are required to repress the current obstacles in establishing a full-fledged green hydrogen economy. With more research and technological advancements, hydrogen will own a large share of the energy infrastructure and portfolio.

Declaration of competing interest

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

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