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Review

Photovoltaic-Assisted Photo(electro)catalytic Hydrogen Production: A Review

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Abstract: The idea of supporting the Sustainable Development Goals (SDGs) has inspired researchers around the world to explore more environmentally friendly energy generation and production methods, especially those related to solar and hydrogen energy. Among the various available sustainable energy technologies, photo(electro)catalytic hydrogen production has been competitively explored, benefiting from its versatile platform to utilize solar energy for green hydrogen production. Nevertheless, the bottleneck of this photo(electro)catalytic system lies within its high voltage required for water electrolysis (>1.23 V), which affects the economic prospects of this sustainable technology. In this regard, coupling the photo(electro)catalytic system with a solar-powered photovoltaic (PV) system (PV-PEC) to unleash the fascinating properties and readiness of this system has heightened attention among the scientific community. In this context, this review begins by elucidating the basic principles of PV-PEC systems, followed by an exploration of various types of solar PV technology and the different types of semiconductors used as photocatalysts in the PEC system. Subsequently, the main challenges faced by the PV-PEC system are presented, covering areas such as efficiency, stability, and cost-effectiveness. Finally, this review delves into recent research related to PV-PEC systems, discussing the advancements and breakthroughs in this promising technology. Furthermore, this review provides a forecast for the future prospects of the PV-PEC system, highlighting the potential for its continued development and widespread implementation as a key player in sustainable hydrogen production.

Keywords: photovoltaic; photo(electro)catalytic; hydrogen; solar; renewable

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1. Introduction

Global energy demand is predominantly generated and supplied from various sources, either from renewable or non-renewable sources. Despite the surging increase in the renewable energy sources over the past few years, the International Energy Agency (IEA) reported that fossil fuels were still dominating the global energy supply in 2019, which accounted for up to 85%, whereas the rest is shared between other renewable energy sources [1–3]. Undoubtedly, the supply of clean energy via renewable resources will continue to accelerate with respect to the higher demand of this supply across the globe. Among the renewable energy sources that received considerable attention is solar energy [4,5]. Solar energy is a non-exhaustible resource and is readily accessible [6–8]. Nevertheless, the non-nocturnal and intermittent sunlight intensity depending on the weather behavior has been the major limiting factor of this solar energy [9,10].

Arising from these bottlenecks, the deployment of the solar energy harvesting technologies, particularly solar photovoltaic (PV) systems, emerged as an excellent alternative to facilitate and mitigate the aforesaid limitation within the solar energy technology [11–14]. Solar PV technology can be considered a mature technology that is capable of converting sunlight into electricity. The incorporation of semiconductor materials to fabricate the PV cells which subsequently form a solar PV panel are then connected with an inverter which converts the direct current (DC) generated from the panels into alternating current (AC) used in domestic or industry applications [15–17].

On the other hand, a photo(electro)catalytic (PEC) system is a process for extracting hydrogen and oxygen molecules from water sources using photocatalyst material consisting of semiconductor materials, either in the presence of an external bias voltage or without additional bias energy [18,19]. In general, the PEC system consists of three main components: (i) charge carriers generated by light absorption, (ii) the mobility of the photocharge carriers, and (iii) the photocatalytic reaction at the photocatalyst surfaces [20,21]. The hydrogen evolution reaction (HER) occurs when a positively charged hydrogen ion (H^+) is generated in the acidic state, while water is formed in a base condition. In the oxygen evolution reaction (OER), oxygen is generated from water in the acidic state, while a negatively charged hydroxide ion (OH^-) is generated in the basic state. Up to date, this system has only achieved a maximum Solar-to-Hydrogen (STH) efficiency of 30% with the help of a photovoltaic system and an electrolyzer [22,23].

There are two main advantages of this water splitting system, namely, (i) a low overpotential response leading to superior STH efficiency and (ii) a simple and unadorned system with less contrivance [12,20]. Thereinto, the PEC system can be considered as one of the cleanest and most sustainable approaches to generate clean hydrogen, because it utilizes two inexhaustible resources: (i) the solar energy needed to carry out the water splitting process and (ii) the water as a medium, which is abundant in nature. Because the PEC system is mainly influenced by the efficiency of the semiconductor photocatalyst, the development of a highly efficient photocatalyst that can satisfy the above three basic concepts of the aforesaid system is highly desirable. In recent years, hundreds of review articles have been published highlighting the critical development of semiconductor photocatalysts in binary or ternary heterostructures [24–28]. It is believed that this approach can unleash the versatile platform of a different combination of semiconductor photocatalysts, which yields better photocatalytic efficiency.

For example, Mahmoud and colleagues have summarized the advantages of porous photocatalysts that can be achieved by controlling the structures and properties of the photocatalyst particles [29]. A report by Yang et al. [30] focused on advanced strategies to improve photocatalytic efficiency through three different approaches, namely, (i) morphological regulation, (ii) ion doping modification, and (iii) composite modification. Despite the proposed advanced strategies, the authors concluded that there are still major limitations that need to be solved prior to this photocatalytic technology being readily practical. In another related study published in 2023, Ahmad and his team provided further insight into the most efficient strategies and methods to mitigate the limitations of this system to make it readily available for large-scale applications [20]. Figure 1 demonstrates several strategies presented in their work, including improving the spatial separation of charges through various approaches such as co-catalyst loading, doping engineering, vacancy engineering, crystal facet engineering, and phase engineering approaches. Another approach that can be considered to improve the photocatalytic activities is regulating the active sites which can be achieved by basal engineering and quantum confinement approaches [31]. On the other hand, Saraswat et al. [32] provide a deep insight into the recent progress of the PEC system for hydrogen production using visible light. According to Saraswat, different approaches can be used to produce hydrogen energy as shown in Figure 2. In addition, Saraswat's review highlighted the critical elements that require a centralized and recognized standard methodology to study and report photocorrosion and photocatalytic stability. With such recognized and standardized methodologies, it is easy to study the type of photocatalyst that is suitable for practical large-scale applications.

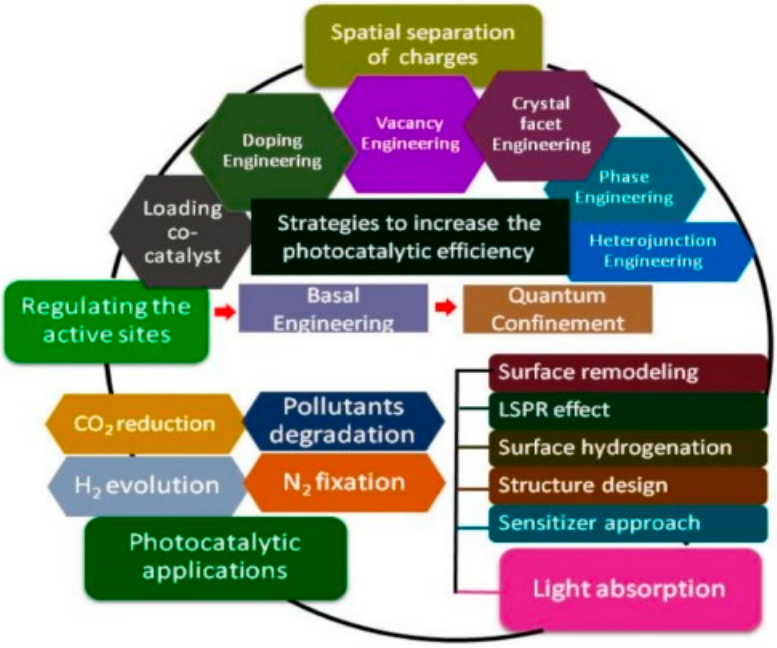

Figure 1. Multi-approach strategies to alleviate the photocatalytic efficiencies (adopted with permission from [20]).

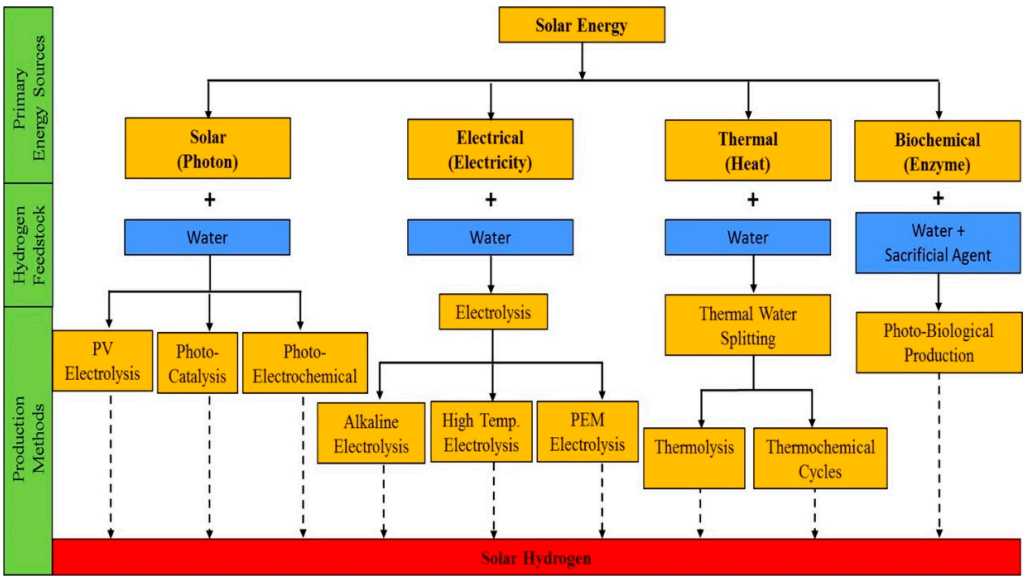

Figure 2. Various approaches to produce hydrogen through the utilization of solar energy (adopted with permission from Ref. [32]).

It is worth noting that, in recent years, a considerable number of reports have been published on PEC applications, especially on synthesis techniques, physicochemical analysis, and potential applications [33–35]. Notwithstanding, there are hardly any articles in the current literature dealing with the recent development and achievements of the integrated PV-PEC system.

In view of the noted limitation in the literature, this review aims to provide an overview of the PV-PEC integrated system for sustainable hydrogen production. This overview starts with the basic principle of PV-PEC systems. In this section, a concise explanation of the underlying principle of the PV system is provided. Subsequently, this review delves into the concept of the photo(electro)catalytic (PEC) system, which forms the basis for hydrogen production via the PV-PEC system. Next, this review further examined the

different types of solar PV technologies from the first, second, and third generations of solar PV technologies. In each generation of solar PV technology, several examples of PV technologies that were developed within each generation were provided to ensure that the readers have a broad understanding of the types of PV technologies that are mature and currently developing. Additionally, this review outlines several archetypal semiconductor photocatalysts utilized in the PEC system. Each photocatalyst's advantages and disadvantages are summarized, providing valuable information on their suitability and potential for enhancing the efficiency and effectiveness of the PV-PEC system. Next, the main challenges of this PV-PEC system are presented. Lastly, this review highlights recent research endeavors related to PV-PEC systems, showcasing advancements and breakthroughs in this field.

2. Basic Principle of the PV-PEC Integrated System

2.1. Basic Principle of the PV System

The concept of the photovoltaics (PV) system lies within the conversion of direct sunlight energy into electricity in the presence of the PV cell. The term "photovoltaic" stems from the word "photo" meaning light and "voltaic" meaning electricity [4,36]. When the cell is illuminated by light, the PV cell absorbs the photon generated from the sunlight which causes the electrons to be excited and move from the atoms of the cell, resulting in the creation of holes in the system. As the PV cell is connected to the load as depicted in Figure 3, the voltage differences cause the related charges to move from the n-side of the cell to the p-side of the cell and thus generate electricity.

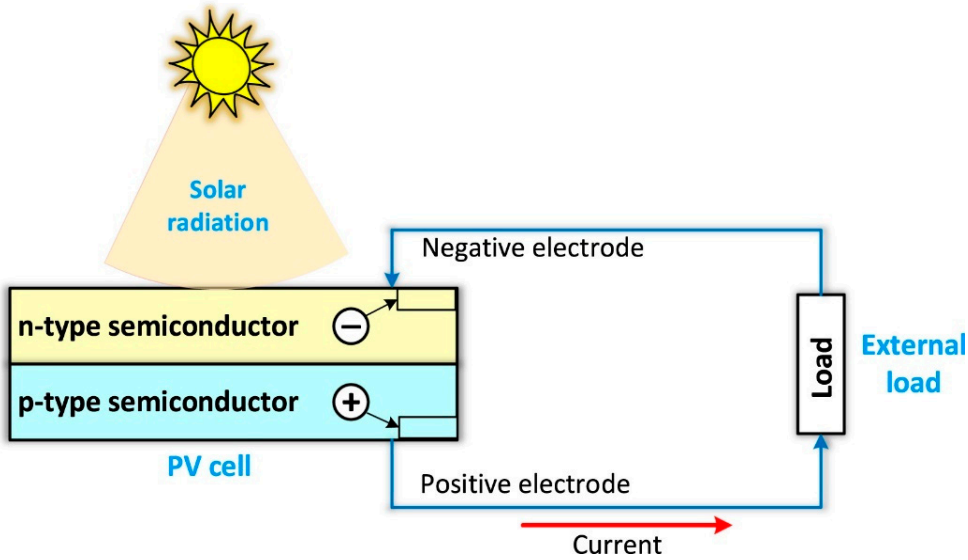

Figure 3. Basic principle of solar PV cell (adapted with permission from Ref. [36]).

Typically, a single PV cell is only capable of producing around 0.5 V, whereas the current generation depends on several factors such as weather, the intensity of the sunlight, and the surface of the cell [37–39]. Thus, to enhance the capability of the PV cell, it is common to see that the PV cell is usually designed in a big module or panel in which the individual cell is either connected in a series arrangement or parallel arrangement. It is worth noting that different arrangements of the cell result in different enhancements. For example, the PV cell that is connected in series would result in the enhancement of its voltage, whereas the PV cell that is connected in parallel would result in the enhancement of the current. Notwithstanding, depending on the desired or targeted application, the PV cell can be connected in a hybrid design to form a PV array which is often used for a specific targeted application.

2.2. Basic Principle of the PEC System

The photoelectrochemical (PEC) system employs photoelectrodes, comprising semiconductor photocatalysts deposited on a substrate, usually fluorine tin oxide (FTO) or indium tin oxide (ITO). The PEC system requires a source of energy to initiate the water splitting process. The photoelectrode is typically composed of a photoactive semiconductor that separates photogenerated charge carriers through the space-charge field [19,24]. The minority of these carriers then migrate to the interface between the semiconductor and liquid for the reaction to take place.

Figure 4 illustrates the typical photocatalytic process occurring on the surface of a semiconductor photocatalyst loaded with hydrogen evolution cocatalyst (HEC) and oxygen evolution cocatalyst (OEC) [40]. The presence of this HEC and OEC is responsible for further facilitating the water splitting process. There are three main parts governing the water splitting process, which are (i) light absorption capacity of the photocatalysts, (ii) photocharge carrier mobility and transport, and (iii) redox reaction [19]. Upon light illumination, the semiconductor photocatalyst will absorb the photon energy that is greater or equivalent to their band gap energy. This process will stimulate the generation of the photocharge carriers, where the excited electron in the valence band is transported to the conduction band, consequently leaving a hole in the valence band. Notably, the band edge location of the semiconductor photocatalyst should be more positive and negative than the O_2/H_2O redox potential of 1.23 V vs. the normal hydrogen electrode (NHE) and $H+/H_2$ redox potential of 0 V vs. NHE, respectively, in order to initiate the water reduction reaction. The overall water splitting reaction can be summarized in the following equations:

$$\text{Hydrogen Half Reaction: } 2H^+ \text{ (aq)} + 2\,e^- \rightarrow H_2 \text{ (g)} \quad (1)$$

$$\text{Oxygen Half Reaction: } H_2O \text{ (l)} + 2\,h^+ \text{ (aq)} \rightarrow 2\,H^+ \text{ (aq)} + 0.5\,O_2 \text{ (g)} \quad (2)$$

$$\text{Overall Water Splitting Reaction: } H_2O \text{ (l)} \rightarrow H_2 \text{ (g)} + 0.5\,O_2 \text{ (g)} \quad (3)$$

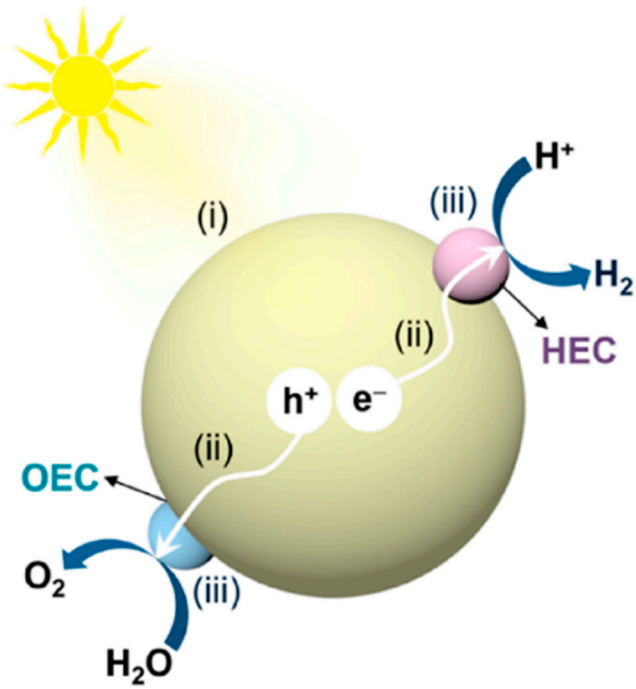

Figure 4. The basic principle of the water splitting reaction in which (i) light absorption, (ii) photocharge carriers' separation and transport, (iii) redox reactions (adopted with permission from Ref. [40]).

3. Types of Solar PV Technology

The development of solar PV technology has witnessed a surge of interest among the community and society as one of the most promising renewable energies. The use of solar PV technology to produce electricity has attracted multifarious attention across the globe due to its versatile offer which includes less reliance on fossil fuels, less emission of greenhouse gases, and better energy independence. As solar PV technology progressively becomes affordable, the yearning to cut the price for solar PV technology has seen significant efforts. In this regard, there are currently three generations of solar PV technologies that have been developed as illustrated in Figure 5. The listed generation of solar PV technology is important as it can be viewed as a way for tracking the historical development of this system over time.

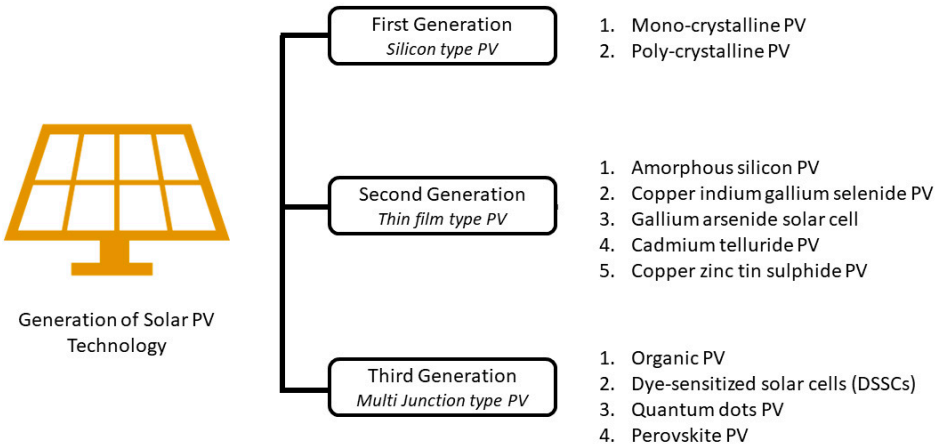

Figure 5. Generations of solar PV technologies that have been developed.

3.1. First Generation of PV Technology

The first generation of solar PV cells that debuted into the market is made up of silicon-based cells. At present, more than 80% of the global installation of solar PV technology comes from this first generation of silicon-based PV technologies [16,17]. This is attributed to its mature efficiency that is suitable for market needs. Typically, the first generation of PV technology, which is also known as wafer-based technology, is composed of a thick crystalline layer of silicon either in the form of mono- or polycrystalline silicon as illustrated in Figure 6. The advantages of this wafer-based PV technology are that the source of the material is abundantly available in the Earth's crust and the non-toxic properties of the silicon allow delays in the contamination process and durability loss.

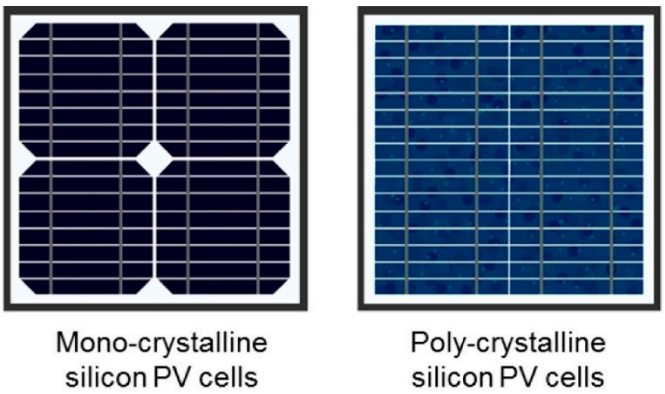

Figure 6. First generation of PV technology (adapted with permission from Ref. [36]).

In the case of monocrystalline silicon PV technology, the entire set of the PV cells is made up of a single crystal of silicon that was extracted from the sand. Nevertheless,

the process of purifying the silicon from the unwanted impurities required a multi-step process of extraction, purification, and heating at high temperatures before the Czochralski process [41,42]. As per the brief description of the manufacturing process, the complicated manufacturing process of this monocrystalline silicon has greatly impacted the production cost of solar PV technology despite its advantages of high efficiency around 26–27% [2,8].

As the manufacturing cost for monocrystalline PV systems is highly unfavorable, polycrystalline silicon PV technology has come into the market to mitigate the cost limitation in monocrystalline silicon PV systems [2,41]. Polycrystalline PV technology has the same principle as monocrystalline PV technology with a slight deviation in terms of its crystalline silicon grains. The crystalline silicon grains in the polycrystalline PV system consist of an additional recombination source which actually impacted the overall efficiency of the PV cell. It is reported that the efficiency of the polycrystalline PV technology typically ranges from 15 to 20% depending on the technology usage and environmental conditions in which the PV system is operated [17,42].

3.2. Second Generation of PV Technology

The second generation of PV technology is known as thin film solar PV technology, which as the name says, is composed of either single or multiple thin layers of PV elements. These single or multiple layers are usually printed on a glass or a metal substrate depending on the desired thickness ranging from several nanometers to tens of micrometers. As such, the aforesaid second generation of PV technology has a good advantage in terms of a thinner PV cell in comparison to the first generation of solar PV technology. There are several types of the second generation of solar PV technologies that have been developed which include amorphous silicon PV, Copper indium gallium selenide PV, Gallium arsenide PV, Cadmium telluride PV, and Copper zinc tin sulfide PV systems [43–45].

On account of the second generation of PV technology, amorphous silicon PV systems can be considered one of the most matured thin-film PV technologies in this generation. In the amorphous silicon PV system, the structure is composed of either a p-i-n or n-i-p type junction in which each p-layer and n-layer are responsible for creating an internal electric field (i-layer). The amorphous silicon PV system is also known to have a high absorption capacity which ranges from 1.1 to 1.7 eV. This absorption capacity range is higher than the first generation of solar PV technology which only accounted for up to 1.1 eV [42,46].

It is worth noting that the amorphous silicon is composed of a low structural homogeneity which impacts the electron and hole movement and separation, consequently affecting the absorption capacity. In addition, the unsaturated silicon atoms in the amorphous silicon PV system are observed to have structural changes at different light intensity exposures. Thus, overcoming this limitation in amorphous silicon PV systems has been of great interest to the scientific community. Among the recent approaches to mitigate this issue is forming a heterojunction of amorphous silicon with crystalline silicon, commonly referred to as HJT cells, as shown in Figure 7. According to Zeng et al. [47], this approach has been observed to significantly improve the overall efficiency of the PV technology by having a high open-circuit voltage, superior efficiency in terms of energy conversion, and minimum thermal degradation coefficient.

On the other hand, gallium arsenide (GaAs) and cadmium telluride (CdTe) PV systems are other examples of the second generation of PV technologies. Typically, the GaAs PV system is used in space applications owing to its promising properties for having strong resistance to the radiation source in space. Meanwhile, the CdTe PV system has been considered as one of the promising PV absorbing materials concerning its bandgap energy of 1.4 eV. The promising bandgap energy of the CdTe PV system allows the system to be capable of absorbing a wide range of light intensities, which later has urged the scientific community to escalate the maturity of this type of PV technology. It was reported that the first CdTe PV system was developed by Kodak in 1982 which is capable of achieving >10% efficiency [48]. In recent years, the efficiency of this CdTe PV system has exceeded more than 15% [42,48].

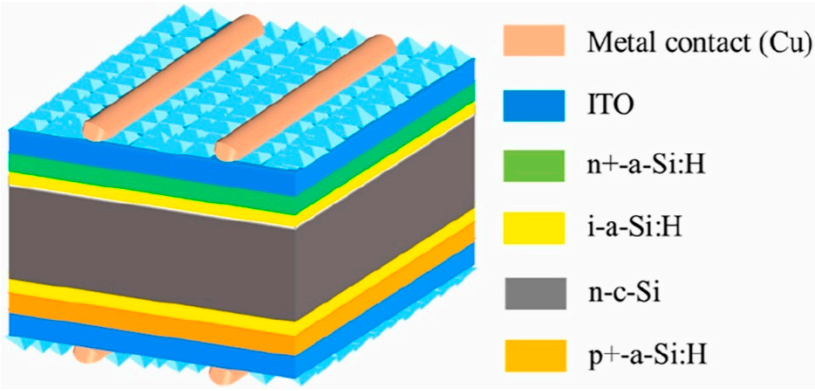

Figure 7. An example of HJT cells which combine the amorphous silicon and crystalline silicon (adapted with permission from Ref. [47]).

3.3. Third Generation of PV Technology

The third generation of PV technology can be classified as a frontier and progressive solution developed by the scientific community to further heighten the efficiency and versatility of solar PV systems in comparison with the second generation of PV technologies that has been studied since the late 1970s [11,17]. Among the current third generation PV technologies that received significant attention and have been progressively optimized are organic PV, dye-sensitized solar cells (DSSCs), quantum dots PV, and perovskite PV systems. The emergence of this latest generation of PV technology aimed to alleviate the inherent limitations possessed by the previous generation of PV technology such as the high manufacturing cost, sluggish solar conversion efficiency, and limited applicability in certain environments.

Due to the aforementioned virtues of this third generation of PV technology, the scientific community has engaged in utilizing novel materials that can be used for capturing a broader range of solar radiation such as organic dyes, quantum dots, and perovskite-based materials in comparison to the old generation of PV technology that focused on silicon-based materials. In this approach, the selection of materials plays an important role in improving the light absorption capacity and thus improving the overall efficiency of the solar PV system and ultimately reducing the cost per watt of the new solar PV technology. Figure 8 illustrates the performance efficiency of the current generation of solar PV technology in which the significant improvement that has been made by the third generation of PV technology as a potential alternative to the current commercially available solar PV technology is highlighted. Furthermore, the advantages of this third-generation PV technology allow the design of PV systems to be printed in more flexible substrates which mitigate the issue related to limited applicability in certain environments. An interesting review of the flexible properties of the solar PV substrate has been written by Li and colleagues, and the details of the flexible solar PV system will not be revisited in this review [8]. As the world is moving toward a more sustainable approach, this third generation of PV technology offers lower environmental impacts due to the capability of utilizing biodegradable materials and thus having greater stability under a broad range of operating conditions. Notably, this third-generation PV technology can be considered an emerging technology as most of the PV systems that fall under this generation are yet to be commercialized due to extensive exploration and optimization in the R&D stages.

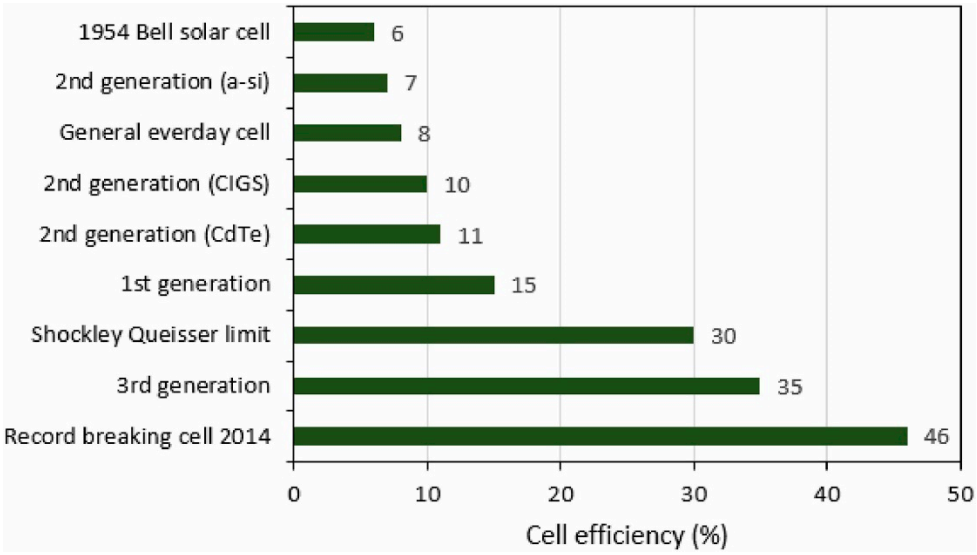

Figure 8. The performance efficiency of the available solar PV systems across various generations of PV technology (adapted with permission from Ref. [8]).

4. Recent Research on PV-PEC Systems

Since the emergence of solar PV technology, this technology has been explored for the production of hydrogen energy as the next frontier of sustainable technology in replacing conventional fossil fuels [24,49]. The growing interest in these two technologies, either solar PV technology or solar PEC technology, has resulted in hundreds of studies published related to both technologies [19,50]. This section aims to recapitulate the recent related progress on the PV-PEC systems reported by various researchers across the globe. Table 1 summarizes the recently selected research studies on various types of solar PV technologies ranging from the first generation to the latest generation of solar PV technology, which is incorporated into the PEC system for the study of photoelectrocatalytic hydrogen production.

The work of Landman and co-workers reported the new conceptual idea of cell separation in the PEC system for decoupling the PEC system into individual oxygen and hydrogen cells as illustrated in Figure 9 [51]. Figure 9 illustrates the redox reaction that occurred in a separate compartment, where Figure 9a is dedicated for the O_2 evolution reaction and Figure 9b is designed for the H_2 evolution reaction. In this decoupled PEC system, the authors used the first generation of silicon PV modules for assisting the photoelectrocatalytic hydrogen production in a separate cell system. The Si PV module has a capacity of 164.5 mA and 5.4 V of a short-circuit current and open-circuit voltage, respectively. It is worth noting that the charge carrier separation in both individual cells was obtained by integrating a battery-grade nickel hydroxide into both cells' compartments. This indicated that nickel hydroxide plays an important role in mediating the ion exchange in both individual hydrogen and oxygen compartments if a decoupled PEC was designed. Apart from that, the STH efficiency of this decoupled PV-PEC system is 0.68%, taking into account that the Faradaic efficiency is 100% and short-circuit current is 55.2 mA. Nevertheless, the idea of decoupling the PEC system into individual H_2 and O_2 evolution reactions has its own drawback, because a decrease of 5% of the overall STH efficiency was observed.

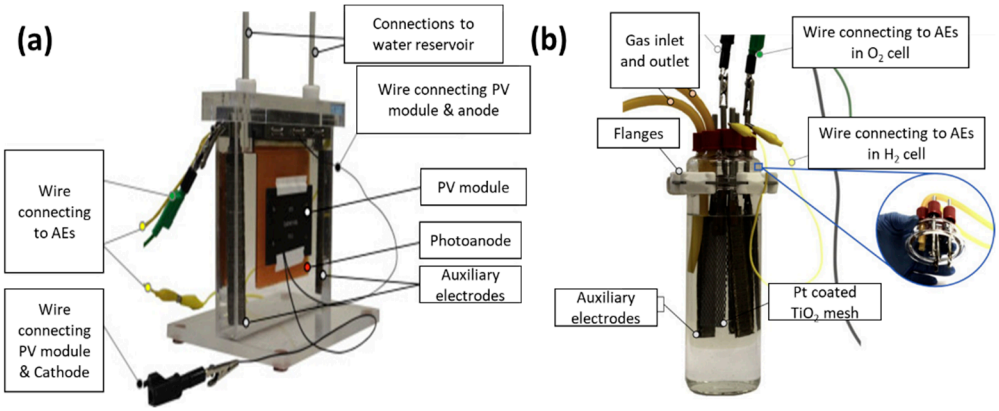

Figure 9. Decoupled PV-PEC system with an (**a**) individual O_2 cell compartment and (**b**) individual H_2 cell compartments (adapted with permission form Ref. [51]).

An example of PV-PEC technology that employed first-generation solar PV technology is reported by Mikolasek and colleagues [46]. In this work, the authors used an amorphous silicon PV system for initiating the PEC reaction. The PV system used in this work is a simple PV cell that exhibits a low open-circuit voltage (V_{OC}) of 0.5 V. The authors point out the recombination of the photocharge carriers at the interface of a-SiC:H(n)/c-Si(p). The low V_{OC} observed in the presented work is due to the fabrication of the aforesaid solar PV system not integrating with an intrinsic passivation layer and back surface recombination fields, which impacts the V_{OC}. Nevertheless, the designed PV-PEC system was capable of achieving a photocurrent of 50 $\mu A \cdot cm^{-2}$ at 0 V vs. RHE and the STH efficiency was observed as 0.76%. An example of a second-generation solar PV-PEC system is reported by Welter and colleagues [52]. In this combination of the PV-PEC system, the authors employed a NiMO/$NiFeO_x$ catalyst for the PEC system, whereas the PV cell used in this work was an a-Si:H/a-Si:H/mc-Si: H triple junction cell. It was found that such a combination is capable of achieving a better STH efficiency of 5.1% in comparison with the STH efficiency of 4.8% when utilizing a conventional Pt/IrO_x catalyst.

A report by Luo et al. [53] examined the potential of combining the perovskite PV system with a NiFE layered double hydroxide foam PEC system for photoelectrocatalytic hydrogen production as illustrated in Figure 10A,B. In this study, the authors used a perovskite PV system made up of $CH_3NH_3PbI_3$ that was fabricated using a two-step spin coating technique. The aforesaid perovskite PV system has a capacity of 21.3 $mA \cdot cm^{-2}$ for the short-circuit photocurrent density, 1.06 V for the open-circuit voltage, and a fill factor of 0.76. On the other hand, the NiFE LDH foam photoelectrode was designed to overcome the inherent limitations of the PEC systems as described in Section 2.2 earlier. As a result, the incorporation of the NiFE LDH foam catalyst was capable of reducing the overpotential requirement to less than 240 mV in order to reach a current density of 10 $mA \cdot cm^{-2}$, which is almost a 30% reduction in the required overpotential when compared with a conventional Pt/Ni foam electrode. As their designed PV-PEC system was arranged in a series as a tandem cell, the system is capable of achieving a high PCE of 15.7% as illustrated in Figure 10C. The measured operating current density at 10.0 $mA \cdot cm^{-2}$ obtained by this system highlights that the solar-to-hydrogen (STH) efficiency of this system is 12.3%. It was noted by the authors that the STH efficiency of this system was measured in an unbiased light illumination, which is the correct way of measuring and reporting the STH efficiency as depicted in Figure 10D.

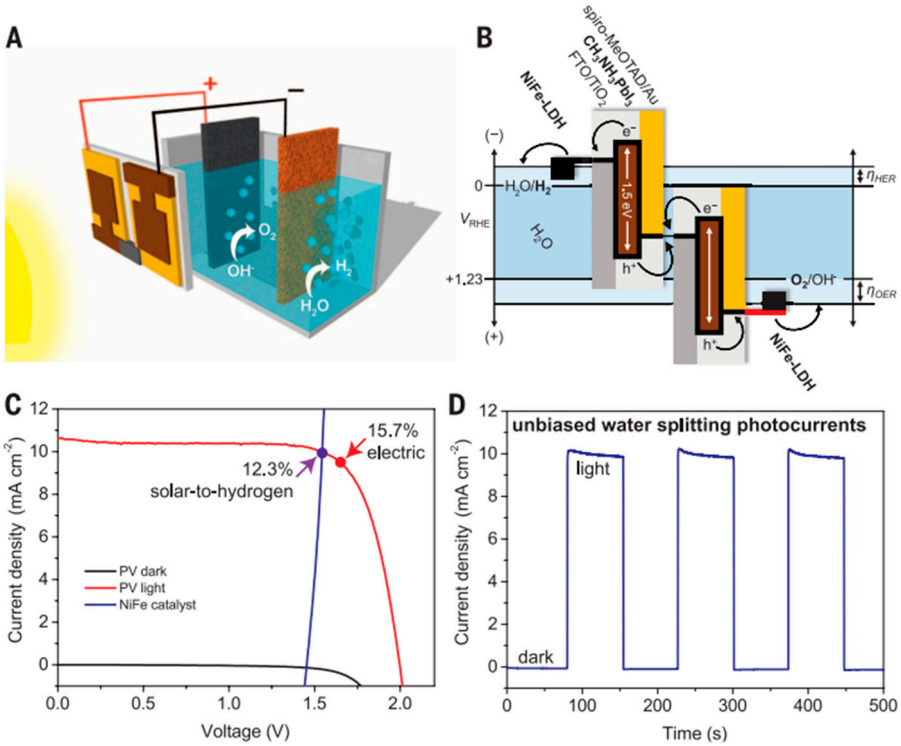

Figure 10. Combination of perovskite PV system with NiFE LDH/Ni foam photoelectrode. (**A**) Schematic illustrations of the perovskite PV-NiFE LDH/Ni foam photoelectrode, (**B**) schematic illustration of the PV and photoelectrode components, (**C**) J-V curve under dark and simulated AM 1.5 G 100 mW·cm^{-2} illumination, and (**D**) current density–time curve for the integrated perovskite PV-PEC system (adapted with permission from Ref. [53]).

A notable study of a successful integration of DSSCs with a bimetallic Cu-Ni/TiO_2 PEC system for hydrogen production was published by Bashiri et al. [54]. The authors recorded a heightened hydrogen evolution production of 694.84 µmol in the presence of glycerol as an intermediate, which undergoes photooxidation in the PEC system. The enhanced performance of the reported materials was attributed to the optimal particle size and appropriate ratio of Ti^{3+}:Ti^{4+} and Cu^{+}:Cu^{2+} in their bimetallic PEC system, resulting in minimized photocharge carrier recombination problems. This allows more available photocharge carriers to partake in the reaction and yield greater hydrogen production.

Another exploration of DSSCs has been performed by Aravinthkumar and colleagues [55], in which the authors explored the potential of utilizing $SrTiO_3$ as a material for the fabrication of both DSSCs and PEC. The authors suggested that the utilization of $SrTiO_3$ as a material for fabrication of DSSCs is capable of achieving a short-circuit current (JSC) at a rate of 3.36 mA·cm^{-2}, an open-circuit voltage (V_{OC}) of 0.63 V, and an efficiency (η) of 1.12%. Furthermore, it was found that the annealing temperature of $SrTiO_3$ is a key determining factor in enhancing the efficiency of the DSSCs. As such, this study highlights the importance of a proper selection of catalyst materials in constructing solar PV system as well as its operating conditions which will have a greater impact on the performance of the system as a whole. On the other hand, the utilization of the third generation of solar PV technology is also reported by Pan et al. [56]. In their report, Pan and colleagues employed a $NiMo_4/MnO_{3-X}$ catalyst as a HER catalyst with a combination of monolithic perovskite/Si tandem PV cells for photoelectrocatalytic hydrogen production. The combination of the aforesaid PV-PEC system is capable of achieving an STH efficiency of 21.32% under AM 1.5 G 1 sun illumination.

Table 1. Progress on the different types of PV-PEC systems for the photoelectrocatalytic hydrogen production.

Type of PV System	PEC Materials	Source of Light	Performance Activity	Ref./Year
Silicon PV	Hematite photocatalyst	Visible Light	• Short-circuit current (J_{SC}) of PV: 200 mA • Open-circuit voltage (V_{OC}) of PV: 5.04 V • STH efficiency: 0.68%	[51]/2020
Amorphous silicon carbide	Light intensity of 1000 W/m^2	Visible Light	• Photocurrent density: 50 μA·cm^{-2} • STH efficiency: 0.76%	[46]/2019
a-Si:H/a-Si:H/mc-Si:H triple junction cell	NiMo/NiFeO$_X$ catalyst	Visible Light	• STH efficiency: 5.1%	[52]/2018
Monolithic perovskite/Si tandem solar cell	NiMo$_4$/MnO$_{3-X}$ catalyst	Visible Light	• STH efficiency: 21.32% • Power conversion efficiency (PCE): 19.68% • Fill factor: 84% • Short-circuit current (J_{SC}): 20.38 mA·cm^{-2} • Open-circuit voltage (V_{OC}) of PV: 1.15 V	[56]/2021
Perovskite PV	NiFe DLH/Ni foam electrodes	Visible Light	• STH efficiency: 12.3% • Photocurrent: 10 mA·cm^{-2} • Short-circuit current (J_{SC}) of PV: 21.3 mA·cm^{-2} • Solar-to-electric power conversion efficiency (PCE) of 17.3%	[53]/2014
DSSCs	Bimetallic Cu-Ni/TiO$_2$	Visible Light	H$_2$ production: 694.84 μmol	[54]/2017
DSSCs	RGO@g-C$_3$N$_4$/BiVO$_4$	Visible Light	• Photocurrent density: 14.44 mA·cm^{-2} • H$_2$ production: 63.5 mmol·h^{-1}	[57]/2020
DSSCs	Cu-Ni/TiO$_2$	Visible Light	• H$_2$ production: 338.4 μmol·cm^{-2}	[58]/2020
DSSCs	BiVO$_4$	Visible Light	• H$_2$ production: 9.52 mmol·h^{-1} • ABPE: 0.056%	[23]/2020
DSSCs	SrTiO$_3$	UV illumination	Short-circuit current (J_{SC}): 3.36 mA·cm^{-2}, efficiency: 1.12%	[55]/2022

5. Different Types of Semiconductor Materials Used in PEC System

As mentioned earlier, the photoelectrochemical (PEC) system utilizes photoelectrodes, which consist of semiconductor photocatalysts deposited on a substrate, typically fluorine tin oxide (FTO) or indium tin oxide (ITO). The photoelectrode is a critical component in achieving high efficiency of hydrogen production, and it must meet specific criteria to function effectively. Firstly, the semiconductor within the photoelectrode must possess certain characteristics. The conduction band (CB) of the semiconductor should be located at a more negative potential than the hydrogen evolution reaction (HER) at pH 7, which is approximately 0.41 V vs. NHE (Normal Hydrogen Electrode). This positioning ensures that the electrons generated during the photoexcitation process have sufficient energy to drive the hydrogen evolution reaction effectively [2,4].

Secondly, the valence band of the semiconductor should be positioned more positively than the oxidation evolution reaction, which occurs at approximately 0.82 V vs. NHE. This band edge position enables the efficient transfer of holes generated in the photoexcitation process to facilitate the oxidation reaction [2,4]. Moreover, the designed semiconductor photoelectrode should be capable of achieving an electrode potential higher than 1.23 V, which is the thermodynamic potential required for water electrolysis. Thus, the band gap energy of the semiconductors should align within the visible light range of the solar spectrum, as this portion of the spectrum carries sufficient photon energy to promote the necessary electron–hole pair generation.

Figure 11a illustrates the position of the conduction band and valence band of a semiconductor photocatalyst under light illumination. In the context of photocatalytic systems, two types of light irradiation are of particular significance: visible light and UV light illumination. UV light encompasses wavelengths between 200 and 400 nm, while visible light covers wavelengths from 400 to 800 nm. Notably, the band gap energy of a semiconductor photocatalyst plays a critical role in its light absorption capacity. A larger band gap energy leads to a reduced capability to absorb light, subsequently impacting the overall PEC efficiency. For instance, if a semiconductor photocatalyst possesses a band gap energy of 3.0 eV, the PEC system can only be activated by UV light irradiation, which constitutes merely ~4% of the total solar light intensity. On the other hand, semiconductor photocatalysts with band gap energies lower than 3.0 eV, as depicted in Figure 11b, are responsive to visible light energy. This allows such photocatalysts to harness a more substantial portion of the solar spectrum, specifically up to 48% of the total solar light intensity [59]. As such, it is imperative to ensure that the type of semiconductor photocatalyst that is used in designing the PEC system is one that is responsive to visible light energy.

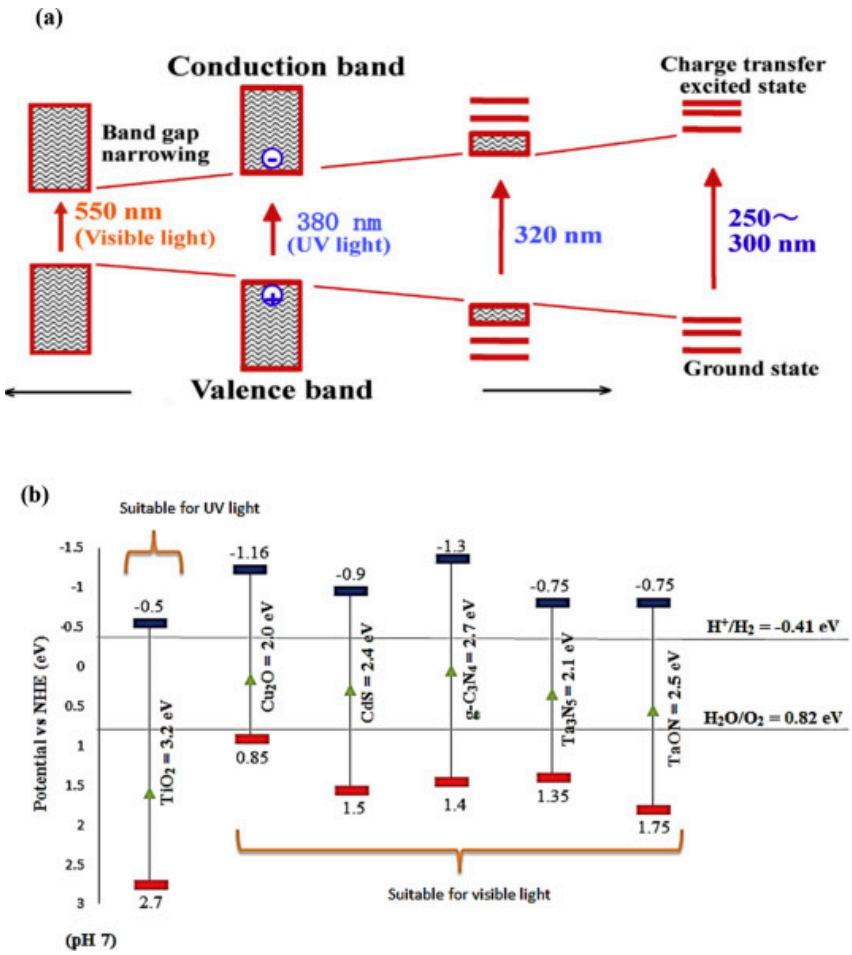

Figure 11. Schematic illustration of (**a**) the expansion of the valence band and conduction band during absorption of light, (**b**) example of the band gap energy structure for different types of photocatalysts (adapted with permission from Ref. [60]).

In addition, Figure 11b also depicts some of the well-known semiconductor photocatalysts that have been widely explored in the PEC system such as titanium dioxide (TiO_2), copper oxide (Cu_2O), cadmium sulfide (CdS), graphitic carbon nitride (g-C_3N_4), tantalum nitride (Ta_3N_5), and tantalum oxide (TaON). It is well known that TiO_2 possessed a band gap energy of 3.2 eV, which indicates that such photocatalysts can only be activated using UV light irradiation. Therefore, to utilize this copious TiO_2 in the PEC system, a modifica-

tion towards the pure TiO_2 needs to be performed by combining the TiO_2 with different types of semiconductor photocatalysts to construct a heterostructure system, adding metal doping, or photosensitizing the TiO_2 with organic or inorganic dyes [61]. By performing such strategies of modification, the valence band and conduction band of TiO_2 located at 2.7 eV vs. NHE at pH 7 and −0.5 eV vs. NHE at pH 7 can be narrowed further within the visible light region. For example, Lee et al. [62] modified the TiO_2 with gadolinium (Gd) and Lanthanum (La) through a liquid phase plasma. The authors observed that the band gap energy of TiO_2 was narrowed from 3.15 to 3.00 eV upon successful synthesis of the Gd-La-codoped TiO_2. The improvement in the band gap energy of the modified TiO_2 showed a significant increase in photocatalytic activities. Similarly, Zhuang et al. [63] fabricated a heterostructure of TiO_2 with a MXene (Ti_3C_2) nanocomposite. The authors observed that the TiO_2/Ti_3C_2 photocatalysts exhibited stronger light absorption capacity in the range of visible light illumination in comparison to pure TiO_2. Furthermore, the authors suggest that the conduction band of the synthesized photocatalyst is −0.31 eV vs. NHE at pH = 0 which is superior for the photocatalytic hydrogen production compared to the pure TiO_2.

On the other hand, g-C_3N_4 is another widely explored photocatalyst in the PEC system. This is due to its excellent stability, suitable band edge location for hydrogen evolution reaction, and Earth-abundant sources [64–66]. Nevertheless, the performance of this photocatalyst was still hindered by its small specific surface area and fast recombination rate of the electron–hole pairs. Thus, a lot of studies explored the potential of a heterostructure system for the g-C_3N_4 with several types of semiconductor photocatalysts. For instance, Wang et al. [67] incorporated B-doped g-C_3N_4 quantum dots (BCNQDs) into the g-C_3N_4 using the hydrothermal method. Interestingly, the incorporation of BCNQDs into g-C_3N_4 shows little improvement in the light absorption capacity of the pure g-C_3N_4, which indicates that the BCNQDs are only anchored on the surface of materials. However, the photoluminescence analysis showed a quenching of behavior which highlights that the composite structure suppressed the recombination of electron–hole pairs and thus heightened the photocatalytic efficiency. Meanwhile, Nasri et al. [49] explored the benefits of incorporating MXene with g-C_3N_4 by investigating various loading amounts of MXene in their published work. It is known that MXene is currently an emerging 2D transition type of metal carbide that is synthesized by etching the Ti_3AlC_2. MXene has an excellent number of hydrophilic functional groups on its surface and unyielding redox reactivity due to its surface terminal Ti sites which helped to further boost the photocatalytic activities. Based on the UV-Vis diffuse reflectance spectra analysis, the loading of MXene onto g-C_3N_4 further narrowed the light absorption capacity of the pure g-C_3N_4, in which the band gap energy was improved from 2.7 to 2.09 eV.

Another interesting semiconductor photocatalyst is cadmium sulfide (CdS), which possesses a band gap energy of 2.4 eV which corresponds to the visible light region. Although CdS possesses a narrow band gap energy, its low photocatalytic stability and rapid recombination of the electron–hole pairs greatly impacted its photocatalytic efficiency. In this regard, Cui et al. [68] performed Z-scheme heterostructure strategies by combining the CdS with cadmium tungstate ($CdWO_4$) owing to its excellent band edge location for hydrogen evolution reaction and better photocatalytic stability. It was found that the $CdS/CdWO_4$ exhibited a threefold higher photocurrent density compared to pure CdS with 2400 μ/h/g of hydrogen produced. In a similar strategy performed by Zhang et al. [69], the authors created a Z-scheme heterostructure system by combining the CdS with silver bromide (AgBr) and reduced graphene oxide (rGO) to synthesize the CdS/AgBr-rGO composite. In this work, the photocurrent response of the synthesized composite was 3.28 times higher than the pure CdS and 11.5 times higher than the pure AgBr, which further highlights the superior separation efficiency of the photogenerated charge carriers. This finding was further solidified with the photoluminescence analysis (PL), in which the CdS/AgBr-rGO composite exhibited the lowest PL intensity compared to the pure samples

and thus signified the minimum recombination of the photocharge carriers which yield an excellent photocatalytic activity.

6. Challenges in the PV-PEC System

On account of the huge potential of the PV-PEC system as the next alternative for sustainable energy technologies, there are several challenges that need to be taken into consideration prior to debuting this PV-PEC system into a commercial and large-scale application. Firstly, as discussed in Section 4, the STH efficiency reported by certain published studies is relatively low. This requires further improvement of the reported efficiency in order to ensure that such PV-PEC systems become more competitive for practical usage. The low efficiency monitored can be due to the resistance losses in the series arrangement of the PV-PEC system, which critically needs to be mitigated to further heighten the STH efficiency of this system.

Secondly, the stability of the catalyst used in both PV and PEC cells is often unstable and prone to have a shortened lifetime over time. This leads to a high maintenance cost of this system if the cells need to be replaced frequently. Moreover, the stability of the catalyst in converting the light energy during cloudy weather remains a huge challenge in this field. Thus, designing a stable catalyst that can withstand harsh conditions of the environment while retaining its efficiency is highly desirable.

Next, although the price for the first generation of solar cells has declined over the years, there is still room for improvement in the market price particularly for the latest generation of the solar PV and PEC systems. This is due to the potential of achieving greater efficiency in comparison to those that are available in the market via the latest generation of solar PV and PEC systems. It is recommended that, apart from excavating the full-fledged potential of the system, the scientific community also needs to embark on finding a way to minimize the cost of the PV-PEC system. Finally, the scale of study for this PV-PEV system is still limited to a small scale, whereas a pilot scale of this system is yet to be proven. Thus, it is timely for the scientific community to start exploring the pilot scale of this PV-PEC system, which can further escalate the readiness of this technology.

7. Conclusions

The growing interest in clean energy supply has spurred the escalation of the development of solar energy technologies, with a particular focus on PV-PEC systems. The PV-PEC systems highlight the futuristic advantages of combining two advanced technologies, which are photovoltaic and photo(electro)catalytic systems which allow a direct conversion and transformation of solar energy into sustainable hydrogen fuel. Hence, in this review, the current state-of-the-art PV-PEC systems and the challenges faced in this intriguing field have been summarized. Despite the progress achieved so far, several important aspects and issues remain to be addressed and mitigated to facilitate the widespread implementation of this PV-PEC system.

One of the notable challenges that has been summarized is the development of stable and commercially attractive catalysts for the PEC system. Catalysts play an important role in the efficient conversion of solar energy into hydrogen, and finding cost-effective catalysts and catalysts with good stability remains a key focus of research and development efforts. Another critical aspect is the superficial energy conversion efficiency of PV-PEC systems. Enhancing the efficiency of energy conversion is essential to ensure that a larger proportion of the incoming solar energy is effectively converted into usable hydrogen fuel. Moreover, standardizing the testing protocols for PV-PEC systems is also among the important challenges remaining to be solved. It should be noted that consistent and well-defined testing procedures are required to evaluate and compare the performance of various PV-PEC designs accurately. Gratifyingly, while laboratory-scale demonstrations have shown great results, upscaling these systems to meet the demands of large-scale hydrogen production requires complex engineering and a multidisciplinary approach, which is necessary for further exploration and commercialization.

Nevertheless, the readiness of this PV-PEC system can be further escalated with the following recommendations. Firstly, the levelized cost of hydrogen production from PV-PEC systems needs thorough investigation from various angles, such as the location and space of installation. Selecting the optimal geographical location with high light intensity and sufficient water supply is crucial to maximize the efficiency and cost-effectiveness of these systems. Identifying cost-effective land areas for installation could significantly improve the overall project cost. Secondly, strategically placing PV-PEC systems in areas with ample sunlight intensity is essential for the successful deployment of this technology.

Despite the challenges, hydrogen generation from PV-PEC systems offers a promising pathway toward a sustainable energy future. By addressing the highlighted challenges through rigorous research and innovation, these approaches will unlock the full potential of this PV-PEC technology. Subsequently, it can pave the way for its large-scale application and thus contribute to a greener and more sustainable energy landscape.

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