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Review

Hydrogen Production Using Modern Photocatalysts

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Abstract: Fossil fuels play a powerful role in the global economy and are therefore referred to as strategic raw materials. However, their massive use around the world is associated with concerns about the sufficiency of energy sources for future generations. Currently, fossil fuel resources are heavily depleted, with limited supplies. According to forecasts, the demand for energy will constantly increase, so it is necessary to find a solution that reconciles the ever-increasing demand for energy with the need to protect the environment. The main solution to this problem is to acquire energy from renewable resources, especially in the direction of obtaining alternative substitutes for transportation fuels. One of the main alternative fuels that can replace existing fossil fuels is hydrogen. An efficient way to obtain this compound is through the use of modern photocatalysts. Hence, the purpose of this paper is to review the recent literature on the effective use of catalysts in photocatalytic processes (e.g., glycerol conversion) that enable the synthesis of hydrogen.

Keywords: nanomaterials; photocatalysts; hydrogen; fossil fuels

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1. Introduction

Fossil fuels have long played a key role in the global economy, but extensive use has raised concerns about their future availability and environmental impact. As energy demand continues to grow, there is an urgent need to develop alternative energy sources that are sustainable and environmentally friendly. One such alternative is hydrogen, which can be produced by the use of modern photocatalysts [1–5]. This article gives the background for the emerging need of H_2 production and reviews recent developments in the synthesis and application of photocatalysts used for hydrogen production.

2. Photocatalysis as a Method for H_2 Production

Hydrogen can only be considered a renewable and clean fuel if the energy needed to produce it comes from a renewable source, such as wind or solar power. Steam reforming of methane and other fossil-fuel-based technologies do not meet the conditions of being green or sustainable. Therefore, hydrogen should be obtained from renewable sources using electrolysis, thermochemical cycles, plasmolysis, or photocatalysis. Electrolysis contributes 4% of the world's total energy demand, but it is still an energy-intensive process with some economic challenges [6]. On the other hand, photocatalysis, which has the potential to overcome these issues, can be considered one of the most promising ways to obtain clean hydrogen. Photocatalysis involves the excitation of semiconductor catalysts with an appropriate light exposure, leading to electrons and holes which can then participate in reduction and oxidation reactions such as water splitting, pollutant degradation, and CO_2 reduction [7]. The redox capability of the photocatalysts is determined by the positions of the conduction band (CB) and the valence band (VB). However, a significant challenge in the photocatalysis is the tendency of photogenerated electrons and holes to recombine. In addition, these electrons and holes can react with substances such as H_2O, O_2, and OH^-, producing reactive oxygen species (ROS) (Table 1), which are very effective in contaminant

degradation and disinfection [8]. Moreover, photocatalytic processes show the potential to convert pollutants into a clean energy carrier [9].

Table 1. Generation of various reactive oxygen species (ROS) and their corresponding redox potentials [8].

	Equation	E^0 (V)
1.	$e^- + O_2 \rightarrow \cdot O_2^-$	−0.33
2.	$h^+ + OH^- \rightarrow \cdot HO$	+1.99
3.	$h^+ + H_2O \rightarrow \cdot HO$	+2.33
4.	$\cdot O_2^-$ (ads) + $h^+ \rightarrow {}^1O_2$	+0.65
5.	$O_2 + 2H^+ + e^- \rightarrow H_2O_2$	+0.94
6.	$\cdot O_2^- + H^+ + e^- \rightarrow \cdot HO_2$	−0.46
7.	$\cdot HO_2 + H^+ \rightarrow H_2O_2$	+1.06
8.	$H_2O_2 + e^- \rightarrow \cdot OH + OH^-$	+0.32

Photocatalysts can be used to facilitate chemical reactions through exposure to light and are often present in eco-innovations, as they are capable of modifying surfaces, giving them new properties. The efficiency of the photocatalytic process can be increased by activating the material under visible light [10,11].

Heterogeneous semiconductor catalysts such as TiO_2, CdS, $BiVO_4$, Ta_3N_5, and g-C_3N_4 are commonly used as photocatalysts [12,13]. Photocatalysts activity can be enhanced by loading cocatalysts, such as metals, particularly Pt, Au, Pd, Rh, Ni, Cu, and Ag, which help prevent electron–hole recombination, which in turn leads to higher photocatalytic activity [14]. In addition, the efficiency and effectiveness of photocatalysts depends strongly on their composition and method of preparation. For example, it has been shown that the specific surface area of TiO_2-based photocatalysts determines the number of active centers available on their surface, which ultimately affects methane production [15].

Differences in the performance and efficiency of various photocatalysts are crucial in determining their suitability for various applications; thus, further research is needed to fully understand their potential. Photocatalysis can be used for/in the following: (i) removal of pharmaceutical residues from treated wastewater streams [16–18]; (ii) elimination of air pollutants [19,20]; (iii) in environmental remediation [21,22]; (iv) energy conversion [23–25]; (v) chemical synthesis [26,27]. Photocatalysis may also be involved in therapy and photodynamic treatment of cancer, sterilizing surgical instruments, and removal of unwanted fingerprints from sensitive electrical and optical components [28,29]. Moreover, the increasing demand of alternative energy sources and zero-emission economy makes the photocatalysis an emerging method for hydrogen production.

Photocatalysis is a promising technology for hydrogen production because it allows for conversion of solar energy into chemical energy through radiation-induced processes. Honda and Fujishima discovered the photoassisted electrochemical splitting of water into H_2 and O_2 in 1972, and since then, many approaches and photocatalysts have been developed to drive the catalytic production of H_2 under solar radiation. This process (Figure 1) can be achieved by using semiconductor materials as photocatalysts that absorb light with a wavelength shorter than 400 nm, or less than 5% of the solar spectrum [30].

The process of photocatalytic H_2 production from water splitting involves two chemical half-reactions: a proton reduction and a water oxidation (Equations (1)–(4)).

$$2\mathcal{Y} \rightarrow 2e_{CB\text{-}} + 2e_{h\nu} + \text{photon induced } e^- \text{ / } h^+ \text{ generation} \quad (1)$$

$$2H_2O + 4h^+ \rightarrow O_2 + 4H^+ \text{ water oxidation half reaction } E_{OX}{}^0 \quad (2)$$

$$4H^+ + 4e^- \rightarrow H_2 \text{ production reduction half reaction } E_{Red}{}^0 = 0 \text{ eV} \quad (3)$$

$$2H_2O + 2\mathcal{Y} \rightarrow 2H_2 + O_2 \text{ overall water splitting} \quad (4)$$

In this process, light energy is converted into chemical energy and electrons move from the valence band (VB) to the conduction band (CB), generating photogenerated electron–hole pairs. Photogenerated electron–hole pairs can drive photocatalytic half-reactions. The redox capability of a semiconductor depends on the highest energy state in the VB and the lowest energy state in the CB. The CB level must be more negative than the hydrogen production level (EH_2/H_2O) to catalyze the reduction of water to hydrogen, and VB must be more positive than the water oxidation level (EO_2/H_2O) to oxidize water in a relatively unfavorable four-hole process [31]. The minimum energy required to drive the reaction for two moles of incident photons is 1.23 eV. The efficiency of photocatalytic hydrogen generation from water splitting is relatively low due to the rapid recombination of photogenerated electron–hole pairs, the inability of many photocatalysts to utilize visible light, and the possibility of a reverse reaction that involves the rapid recombination of hydrogen and oxygen [32].

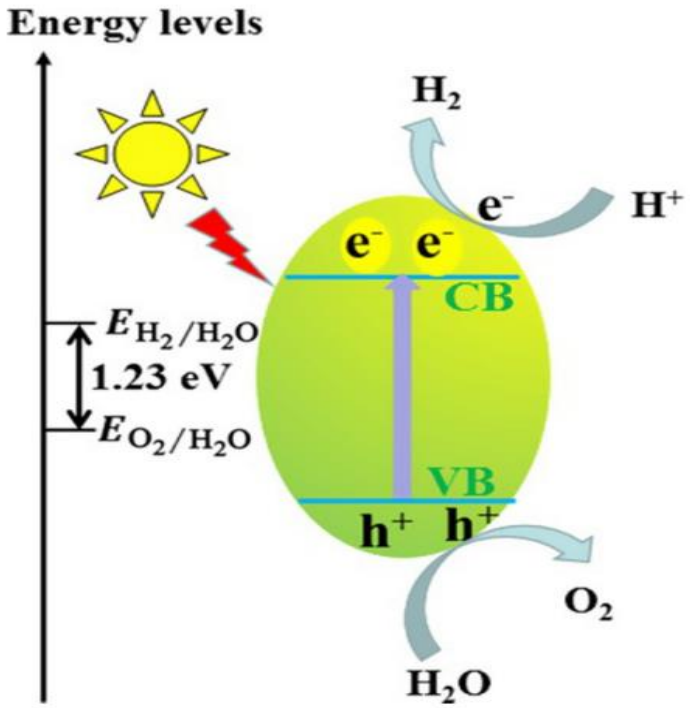

Figure 1. Photocatalytic hydrogen generation upon suitable semiconductor [30]. Reproduced with permission from Ref. [30], Elsevier, 2019.

In order to improve photocatalytic efficiency, sacrificial substances are usually added to existing photocatalytic systems. However, many sacrificial substances, such as methanol [33], lactic acid [34], triethanolamine [35], Na_2S-Na_2SO_3 [36], and ascorbic acid [37], are considered harmful and toxic. Research on photocatalytic hydrogen evolution without sacrificial agents is of great significance for environmental protection. However, due to the complexity of water oxidation kinetics and the low economic value of O_2, the development of photocatalytic hydrogen evolution from pure water without sacrificial agents could be challenging [38]. There are many difficulties and influencing factors in photocatalytic total water splitting, so it is quite challenging to achieve efficient photocatalytic total water splitting without sacrificial agents. In addition, the hydrogen and oxygen produced during the entire water decomposition process are difficult to separate, and the mixed gas produced during the reaction poses major safety hazards. Thus, photochemical reactions of sacrificial agents and their degradation products during H_2 production are still under systematic study (Table 2) [39].

Table 2. The most common sacrificial agents and their degradation products during photochemical H_2 production.

Sacrificial Agent	Expected Reaction Mechanism
Methanol	$H_2O + h+ \rightarrow \cdot OH + H^+$ $CH_3OH + \cdot OH \rightarrow \cdot CH_2OH + H_2O$ $\cdot CH_2OH \rightarrow HCHO + H^+ + e^-$ $2H^+ + 2e^- \rightarrow H_2$ $HCHO + H_2O \rightarrow HCOOH + H_2$ $HCOOH \rightarrow CO_2 + H_2$ Overall reaction: $CH_3OH + H_2O \rightarrow CO_2 + 3H_2$
Lactic acid	CH_3-CH(OH)-COOH + $H_2O \rightarrow CO_2 + H_2$+ CH_3-CO-COOH
Triethanolamine	$C_6H_{15}NO_3 \rightarrow C_6H_{15}NO_3^+ + e^-$ $C_6H_{15}NO_3^+ \rightarrow C_6H_{14}NO_3^{\cdot} + H^+$ $C_6H_{14}NO_3^{\cdot} \rightarrow C_6H_{14}NO_3^+ + e^-$ Overall reaction: $C_6H_{14}NO_3^+ + H_2O \rightarrow C_4H_{11}NO_3 + CH_3CHO + H^+$
Sodium sulfide	$Na_2S + H_2O \rightarrow 2Na^+ + S^{2-}$ $S^{2-} + H_2O \rightarrow HS^- + OH^-$ $HS^- + h\nu \rightarrow HS^{-*}$ $HS^{-*} + HS^- \rightarrow [(HS)_2]^{-*} \rightarrow H_2 + S_2^{2-}$
Sodium sulfite	Irradiation: $SO_3^{2-} \rightarrow SO_3^{2-*}$ Oxidation: $SO_3^{2-*} + 2OH^- \rightarrow SO_4^{2-} + H_2O + 2e^-$ Reduction: $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ Oxidation: $SO_3^{2-} \rightarrow S_2H_6^{2-} + 2e^-$ Reduction: $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$

3. Hydrogen as the Fuel of the Future

In recent years, there has been growing interest in the topics of fuels of the future with regard to photocatalysis, and nanomaterials. The fuels of the future, which are alternative energy sources, offer a number of advantages over traditional fuels, such as lower greenhouse gas emissions and less dependence on fossil fuels [40–42]. However, their development and deployment come with several challenges, among which are the following:

- Fuel economics, which can make it difficult to replace diesel in some applications [43].
- Technical feasibility—imposed requirements can effectively promote the adoption of future fuels. To scale up the adoption of future fuels, innovation and political support are needed, as building infrastructure requires significant investment [44,45].
- Sustainability issues related to biofuels are one of the obstacles in this transition process, and the industry faces unique key challenges in the transition process for developing and deploying future fuels [46].
- The economic export of clean energy—many countries do not have sufficient domestic energy supplies, neither renewable nor other, and rely on imported energy sources [47].
- Keeping up with modern technologies—older technologies often do not have a chance to take root on the market due to the rapid introduction of new fuels [48].

To overcome these challenges, cooperation and coordination is required between industry leaders, governments, research institutions, and technology providers.

In the last two decades of the 20th century, the use of energy from renewable sources began in earnest. Despite the emerging problems related to the need for backup sources based on conventional energy and the economics of the operation of such systems, this process is already global [49–51]. In the next dozen years, the demand for fossil energy may be reduced by future solutions for obtaining clean energy from hydrogen, fuel cells, and other sources. However, it is unlikely that over the next few decades new energy sources combined with renewable energy can meet the world's energy demand [52–54], as in 2020, fossil fuels still accounted for about 83.1% of the world's energy use, with only 12.6% and 4.3% from renewable sources and nuclear power, respectively [55].

Energy production in the European Union focuses on the use of various energy sources, such as solid fuels, natural gas, oil, nuclear power, and renewable energy (e.g., hydroelectric, wind, and solar power). Renewable sources account for the largest share of energy production [56,57]. In 2021, in the European Union, renewable energy was the largest source of primary energy (41% of total EU energy production). The second largest source was nuclear energy (31%), followed by solid fuels (18%), natural gas (6%), and oil (3%) [58].

For its own consumption, the EU also needs energy imports from third-world countries. In 2021, petroleum products (including crude oil, which is the main component) were the main energy imports, accounting for almost two-thirds of EU energy imports (64%), followed by natural gas (25%) and solid fossil fuels (6%) [59,60].

However, fuels of the future are an emerging area in the transportation sector, with a wide range of fuels being developed for subsequent use by various industries, e.g., the shipping industry is using bio and synthetic liquefied natural gas (LNG), ammonia, methanol, hydrogen, and biofuels [44,61–63]. Future fuels are being evaluated on the basis of their environmental friendliness, and the lifecycle approach considers the entire chain, namely, from the well to final application ("from well to wake") [64–66]. Sustainable fuels, such as hydrotreated vegetable oil, and synthetic fuels, such as ammonia or methanol, are also being developed, with a production capacity of 46 million tons by 2025 and a total planned investment of USD 40 to USD 50 billion. The benefits of using these future fuels include, among others, improved vehicle performance, increased energy security, and reduced emissions levels [67,68].

Alternative fuels also include hydrogen and ammonia, with hydrogen being typically produced from water using wind turbines, and ammonia produced from hydrogen and nitrogen extracted from the air [69–72]. Currently, more than a dozen alternative fuels are in production or under development. However, further research is needed to determine the impact of future fuels on human health, greenhouse gas emissions, and tailpipe emissions, as well as the necessary modifications in vehicles and required fuel infrastructure [73]. Overall, future fuels can help comply with regulations, save money, achieve decarbonization goals, and make transportation more sustainable [74].

At present, hydrogen is one of the most frequently identified energy sources that can be used on the way to a zero-emission economy (i.e., during combustion, virtually the only product is water (Table 3)). It can be used in transportation, industry, and energy production [75,76]. The European Union has set a goal of achieving total climate neutrality by 2050, i.e., capturing and storing the same amount of greenhouse gas emissions as will be released into the atmosphere [77,78]. The development of renewable energy sources provides space to stabilize the reliability of energy production. Hydrogen can be used as an energy carrier, which is particularly important for accumulation according to unexpected seasonal conditions, as well as for some mobility applications [79–82].

The use of hydrogen as a next-generation energy solution is gaining increasing attention because of its environmentally friendly nature and the potential to reduce dependence on fossil fuels. Hydrogen is a high-energy fuel (33 kWh/kg) that is currently in direct competition with battery technology [83].

Hydrogen could be produced through a variety of processes, namely (Figure 2), (i) steam reforming [84]; (ii) conversion of biomass [85]; (iii) electrolysis [86]; (iv) photocatalysis [87].

Table 3. Comparison of fossil fuels and hydrogen.

Raw Material	State of Concentration	Combustion Reaction	Description of Reaction	Presence of CO_2 in Exhaust Gases
Hard coal	Solid	$C + O_2 \rightarrow CO_2$	In addition to CO_2, harmful compounds (microdust) are produced.	YES
Gasoline	Liquid	$C_5H_{12} + 8O_2 \rightarrow 5CO_2 + 6H_2O$	Expensive platinum catalysts are required for complete combustion.	YES
Natural gas	Gas	$2CH_4 + 4O_2 \rightarrow 2CO_2 + 4H_2O$	Gas combustion eliminates microdust formation and reduces CO_2 emissions compared to coal.	YES
Hydrogen	Gas	$2H_2+O_2 \rightarrow 2H_2O$	Emission-free fuel.	NO

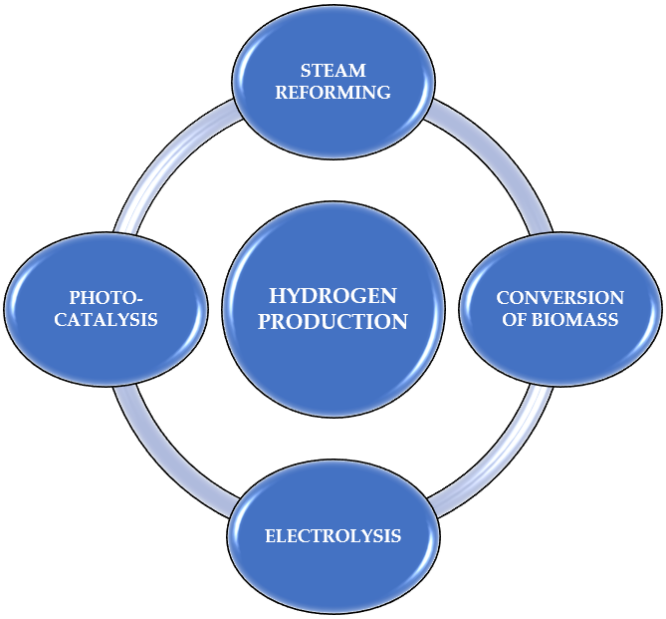

Figure 2. Main methods of obtaining hydrogen.

Steam reforming of methane is still the most widely used method due to the high hydrogen-to-carbon ratio in a CH_4 molecule. Fossil fuels such as coal can be used to produce hydrogen through a gasification process, which produces a mixture of hydrogen, CO, CO_2, methane, and other compounds [88,89]. However, the basis for these methods are fossil raw materials; hence, they have little future [90–92]. To make H_2 production environmentally friendly, renewable precursors must be used for hydrogen extraction [93–96].

Biomass is a renewable H_2 precursor and an attractive alternative to fossil feedstocks. Pyrolysis and gasification of materials such as grass, straw, or biological waste are possible thermochemical routes. Alternatively, biophotolysis, based on the biological gas conversion and fermentation reactions, may also be applied [97–99]. In water electrolysis, on the other hand, an electric current is used to separate water into hydrogen and oxygen. There are two main types of water electrolysis—alkaline electrolysis and membrane electrolysis. In alkaline water electrolysis (AWE), a solution of water and potassium hydroxide is used as the electrolyte. The anode and cathode chambers are separated by a porous membrane, so that the gases produced do not mix. This production process achieves efficiencies of up to 70% [83]. In proton exchange membrane (PEM) electrolysis, the proton-conducting membrane serves as the electrolyte, and the efficiency of this process is about 50% [100].

Another promising method to obtain hydrogen is photocatalysis, where H_2 evolution could be obtained via solar-driven photocatalytic water splitting (PWS). It is considered

an important strategy for enabling clean energy and overcoming global environmental challenges. As a result, many photocatalysts have been developed over the past several years [10].

Demand for clean hydrogen from industrial customers, particularly from ammonia, steel, and refining producers, is announced to be increasing each year. This reflects the positive tendency to decarbonize the existing industry, as well as the growing awareness of the need to switch to pure hydrogen as an energy source. In 2022, Europe's total hydrogen demand was 8.2 Mt, of which as much as 81% was consumed by the refining (4.7 Mt) and ammonia production (2.0 Mt) sectors. The remaining part was absorbed by the production of methanol and other chemicals, used as fuel for industrial heating, for semiconductor production, and in the automotive sector (Figure 3). The transportation sector currently accounts for only 0.04% of hydrogen demand. Nonetheless, hydrogen-based transportation is still being developed in Europe, embracing the drive to decarbonize maritime, road, and air transportation. However, the main obstacle to this development is still insufficient infrastructure, expensive vehicles, and the current cost of clean hydrogen production [101].

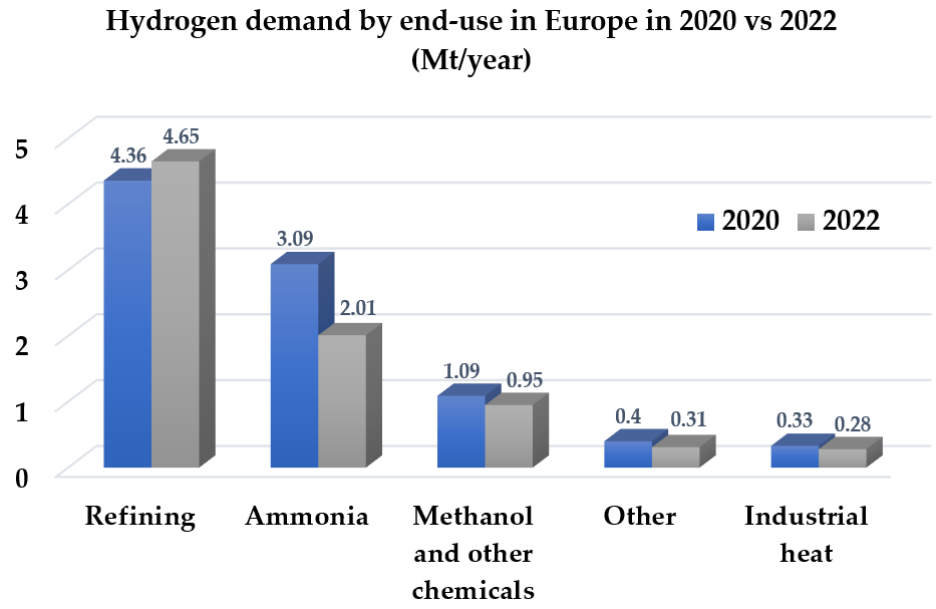

Figure 3. Hydrogen demand by end-use in Europe in 2020 vs. 2022 (Mt/year); based on [101].

The Renewable Energy Directive (RED) on renewable energy sources set a target for the use of hydrogen in the industry and transportation, and it should become one of the key drivers of demand for clean hydrogen. According to the RED, the industrial sector will have to increase its use of renewable energy by 1.6% per year. Moreover, by 2030, 42% of hydrogen used in the industry should come from nonbiological renewable fuels, reaching a level of 60% in 2035. According to the latest update to the Net-Zero Emissions Scenario (NZE) from the International Energy Agency (IEA), the demand for low-emissions hydrogen will grow particularly in heavy industry, transport, and the production of hydrogen-based fuels (Figure 4) [102].

Despite the fact that green hydrogen has enormous potential to decarbonize our energy system, currently planned projects would make it possible to produce only 44 Mt H_2eq by 2030, which is a quarter of projected global demand. Although more than 130 countries (representing 88% of global carbon emissions) have already published their national hydrogen strategies, accelerating the deployment of clean hydrogen will still require developing a viable business case, laying the groundwork for a climate-oriented market structure and focusing on building the right infrastructure [102].

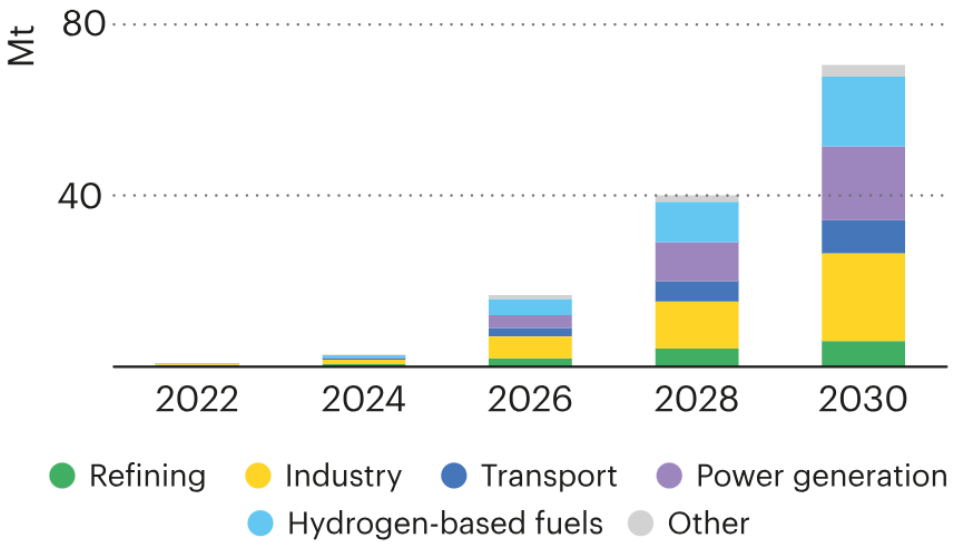

Figure 4. Demand for low-emissions hydrogen according to the 2023 update to NZE report from IEA [102].

4. The Role of Photocatalysts in Hydrogen Production

Modern photocatalysts play an important role in the development of sustainable technologies. They have different phase compositions and can also contain an amorphous phase. In addition, they have different specific surface areas and crystallite sizes, making them more suitable for different applications. They are used for water and air purification and for catalytic CO_2 reduction. In addition, photocatalysts are also used in energy conversion, such as converting solar energy to the chemical one. They are also used for hydrogen production and increased energy production from fossil fuels (Figure 5). Furthermore, they can be used to degrade hazardous pollutants, such as dyes and pharmaceuticals, as well as for biomedical applications, such as cancer therapy. Due to their unique characteristics, modern photocatalysts have become a promising technology for the development of sustainable technologies [103,104].

Photocatalysts face the problems of low selectivity and activity due to the complexity of water-splitting reactions. Photocorrosion and surface passivation also contribute to their stability problems [6]. Additional challenges include the use of unbalanced sacrificial reagents, limited photochemical stability, and insufficient knowledge of reaction mechanisms [105]. Meeting these challenges can lead to a highly efficient and stable photocatalyst for hydrogen production [106].

However, the development of efficient materials for this process remains a major problem, as there are many challenges that must be overcome to make this technology practical [107]. One of the key problems is improving the stability of photocatalysts to prevent their degradation over time [108]. In addition, the cost of photocatalysts needs to be reduced in order to lead to materials of wide practical use. Another challenge is the low light absorption and poor charge transfer of photocatalysts, as well as the poor separation of photogenerated electron/hole pairs [106,108]. Understanding the fundamental aspects and mechanisms of transfer is essential for the complete distribution of electron/hole pairs [108]. Photocatalytic processes proceed with low quantum efficiency due to the high recombination rate of electron/hole pairs, and the main efforts to eliminate these limitations involve modifying the electron structure of semiconductors by introducing special dopants, including dyes or noble metals, such as Pt, Au, Pd, or Ag, into the base material [109–111]. The introduction of platinum can increase the production of H_2 by up to about 25%, and the most active form of the photocatalyst can produce H_2 without water, paving the way for greener industrial hydrogen production [112]. Another promising material for hydrogen production is sodium-tantalate-based photocatalysts ($NaTaO_3$), which have shown high efficiency and stability during photocatalytic water decomposition reactions [113]. The

Cu/TiO_2 photocatalyst is unique in that the $Cu^+ \rightarrow Cu^0 \rightarrow Cu^+$ redox cycle is responsible for its behavior during hydrogen production. Bypassing the Cu^{2+} oxidation step during irradiation is crucial to keep the oxidation potential of photogenerated holes low enough to prevent CO_2 production and keep all carbon in the liquid phase [114]. Furthermore, TiO_2 nanoparticles doped with atomically dispersed copper are highly active and 100% selective for hydrogen generation in the photoreforming reaction of a methanol solution, as confirmed by synchrotron measurements that identified the degree of copper oxidation in samples before and after the reaction and confirmed the atomic dispersion of Cu clusters on the TiO_2 surface [114]. Hydrogen can also be separated from aqueous NaCl solutions, opening up new possibilities for using photocatalysis to produce hydrogen [115].

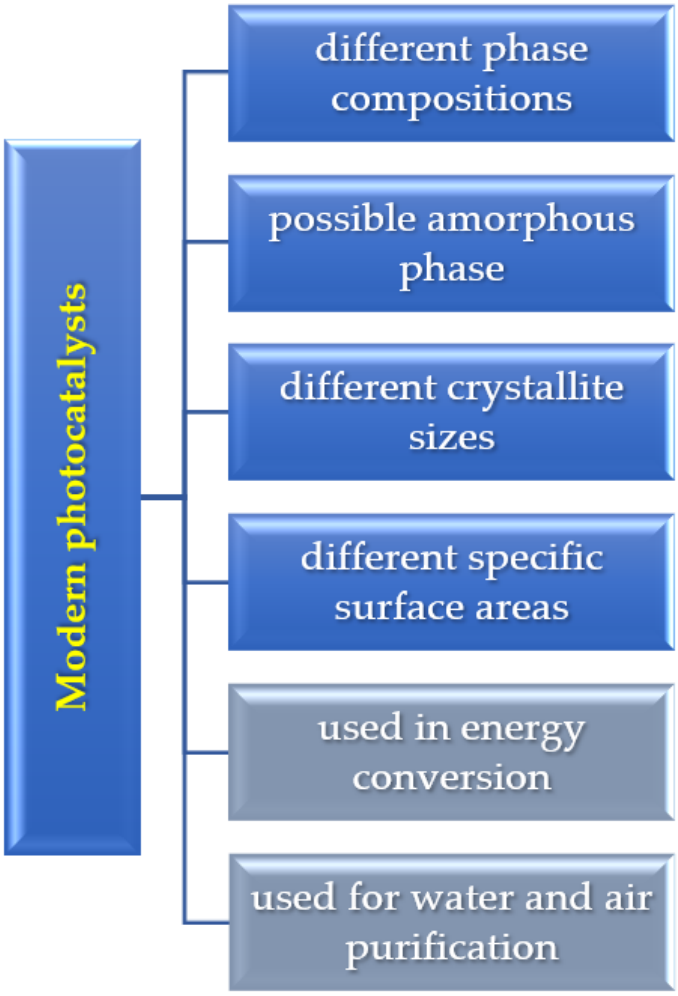

Figure 5. Features of modern photocatalysts.

TiO_2 is a commonly used semiconductor material for photocatalyst hydrogen production due to its advantages of biological and chemical inertness, easy production, and environmental friendliness [116,117]. Therefore, a lot of research has been conducted on this material for hydrogen production [118]. However, TiO_2 has a large band gap (about 3.2 eV), which limits its use in the visible electromagnetic spectrum [119]. Improving the photocatalytic activity of TiO_2 has become a challenge the scientific community. Various strategies have been developed to deal with this challenge.

This challenge includes methods such as metallic and nonmetal doping, incorporate organometallic structures, and use in polymers connect [120]. In studies including Perovi'c et al. [121], composite materials TiO_2 with different proportions of tin disulfide (SnS_2) and reduced graphene oxide was synthesized using an in situ solvothermal method.

$Ti_3C_2T_x$ MXene-derived TiO_2-based photocatalysts with broad light absorption and efficient charge separation have been used for antibiotic degradation, but the complementary effects of each component, especially oxygen vacancies (OV) and high O-terminated Ti_3C_2 work function (O-Ti_3C_2), affecting light absorption and photocatalytic activity, are controversial. $TiO_2/Ti_3C_2T_x$ photocatalysts prepared from $Ti_3C_2T_x$ are tuned by alkalization in a controlled KOH solution and calcination under different heating atmospheres to reveal the contribution of OV, Ti^{3+}, carbon species, and titanium carbide [122].

Li et al. [123–126] used colloidal polystyrene beads as templates to study and prepared three-dimensional ordered macroporous (3DOM) composites of strontium titanate ($SrTiO_3$) and TiO_2 through ordered three-dimensional decorated carbon quantum dots. The macroporous structure of these materials demonstrated the ability to convert long-wavelength light into short-wavelength light, thereby increasing their use in the visible range. This ternary structure is achieved by coating the surfaces of three photoanode tin dioxides doped with fluorine, and then the TiO_2/CdS is stacked on TiO_2 to form a binary structure, and finally a TiO_2/CdS/g-C_3N_4 binary structure is formed from a ternary structure [127].

The data in the literature have shown so far that more than 130 materials, including oxides, nitrides, and sulfides, may be successfully used as efficient photocatalysts for hydrogen evolution through water decomposition. Among them, $BaSr_2Nb_2O_7$- and NiO/$NaTaO_3$-doped materials were found to have the highest quantum yields. However, the maximum quantum efficiency compared to photocatalysts powered by visible light reaches only a few percent at wavelengths up to 500 nm, which is still far from the initial starting point for practical application. Nevertheless, it has been proven that many oxides, sulfides, oxynitrides, and oxosulfides are active in the process of H_2 and O_2 release under visible light irradiation [107].

Porous nanomaterials (photocatalysts) generally outperform nonporous counterparts, both in environmental and energy applications [128]. This is due to their large specific surface areas and the presence of numerous pores, which enable efficient mass capture and diffusion, leading to enhanced photocatalytic conversions. In addition, the presence of pores contributes to better light collection through reflection and scattering. Moreover, these types of materials show a higher density of active centers and shorter charge migration distance from the bulk to the surface [8]. The combination of adsorption and photocatalysis in these materials makes them highly desirable for the construction of multifunctional materials. Various types of porous photocatalysts, including porous carbons, metal oxides/sulfides, metal–organic frameworks (MOFs), and adsorbent–photocatalyst hybrids, have been designed and developed for their desirable properties. MOF photocatalysts have the main advantage of having a high level of design flexibility at the molecular level, which is achieved through targeted manipulation of metal ions and/or organic ligands. Synthesis or subsequent processing of these materials allows for the incorporation of different types of ligands and metals. The inherent structural arrangement of MOFs enables them to achieve enhanced levels of photocatalytic activity [129–131]. The combination of different materials within MOFs could significantly improve hydrogen production performance. Chi et al. [132] synthesized a series of zinc (Zn) compounds modified with MOF and CdS particles; this resulted in Zn(LFor)-MOF/CdS composites. These composite materials' hydrogen production was evaluated at different ratios, resulting in five combinations of Zn(LFor)-MOF and CdS particles (1:1, 2:1, 4:1, 8:1).

Preparation of these porous photocatalysts often involves the use of templates, both hard, such as silicas and carbons, and soft, such as surfactants and polymers. These templates direct the growth of porous structures, which can then be obtained by removing the templates. For example, porous TiO_2, C_3N_4, Fe_2O_3, and graphene have been successfully synthesized using the template approach and applied to various photocatalytic applications such as pollutant degradation and H_2 production [8,133–135].

An alternative approach to achieving synergistic adsorption and photocatalysis is to develop adsorbent–photocatalyst hybrid materials using conventional nonporous photocatalysts. In such systems, the porous adsorbent plays a key role in rapidly concentrating

the desired substances, which can then be degraded or transformed by the photocatalyst. Moreover, some adsorbents have the ability to modify the photoelectrochemical properties of photocatalysts by adjusting the bandgap, increasing the density of charge carriers and reducing the recombination rate of electron/hole pairs. The large surface area of the adsorbent prevents aggregation of photocatalyst particles, which is essential for an efficient photocatalysis process [8,136]. The potential of pores in photocatalysts influences the facilitation of efficient adsorption and mass diffusion, increasing light collection, providing numerous active sites and reducing the distance that charge carriers must travel from bulk to the catalysts' surface. In addition, the presence of pores in photocatalysts allows for the strategic incorporation of (nano)particles or photosensitizers, resulting in improved photocatalysis or multifunctionality. For example, gold nanoparticles (AuNPs) have been encapsulated in well-organized nanopores of cylindrical anodic alumina, as described by Nam and coworkers [137]. By increasing the amount of gold nanoparticles in the nanopores, plasmon-driven photocatalysis on AuNPs clusters was achieved.

To adapt to the principles of "green chemistry", researchers have also explored template-free methods for the synthesizing of porous photocatalysts. One common approach is a self-assembly strategy in which small photocatalyst molecules combine to form organized structures with cavities or pores. For example, Huang et al., by incorporating thiourea into a hydrothermal pretreatment process, obtained porous g-C_3N_4 nanosheets. These nanosheets exhibited a significantly higher surface area and led to an increased amount of photocatalytically produced hydrogen when compared to the nonporous bulk g-C_3N_4 [138]. In contrast, Yang et al. synthesized a porous Ag_2S-Ag photocatalyst by microwave-assisted surface sulfidation of Ag_2CO_3 nanowires [139]. This rapid process was completed in 15 min and involved the reaction of Ag_2CO_3 with thioacetamide in ethanol under low-intensity microwave radiation (400 W). The obtained hybrid structures manifested highly improved photocatalytic activity for the degradation of methyl orange and Cr(VI) reduction in the aqueous solution. Alternatively, Wang et al. used a microwave-assisted in situ method to produce porous hybrid N-TiO_2/g-C_3N_4 materials with a large surface area and better photocatalytic performance for the degradation of methylene blue [140]. On the other hand, carbon nitride nanostructures obtained by bioinspired supramolecular self-assembly have also been intensively studied as water-splitting photocatalysts [141].

Alternatively, the inspiration from nature may be taken and abundant materials may be used to develop porous photocatalysts. For example, biological materials such as pollen can serve as a template in the synthesis of efficient and porous photocatalysts [142]. In addition, naturally occurring minerals such as sphalerite and wolframite have been explored as low-cost photocatalysts that can be activated by visible light [143]. Another approach involves combining inorganic photocatalysts with organisms such as bacteria to create cost-effective hybrid photocatalysts. This has been demonstrated by coating cadmium sulfide nanoparticles on the surfaces of *Escherichia coli* and *Rhodopseudomonas palustris*, resulting in efficient hydrogen production and nitrogen fixation, respectively [144]. On the basis of these exciting findings, the integration of porous photocatalysts, especially at the nanoscale, with cell surfaces to enable cost-effective adsorption and photocatalytic processes seems to be a promising direction, which opens up a new field of biohybrid photocatalysts [8].

Basing on the literature reports, hydrogen generation processes by photocatalytic water splitting can be divided into two types: the reaction in photochemical cells and the reaction in photoelectrochemical cells. In a photochemical cell, a powder photocatalyst in the form of particles suspended in solution is used to carry out the water-splitting reaction [145]. Most of the photocatalytic water-splitting reactions introduced so far are examples of photochemical reactions [146,147]. In a photoelectrochemical cell, the photocatalyst is deposited as a thin film on a substrate, forming a photoanode to carry out the water-splitting reaction in solution [148].

An impressive visible-light-induced hydrogen production rate of 87.2 μmol h^{-1} and an apparent quantum efficiency (AQE) of 8.2% at 400 nm were observed in the $Ni(OH)_2$/g-

C_3N_4 hybrid photocatalyst. The enhanced photocatalytic activity of this hybrid can be attributed to two main factors. First, the Z-scheme effectively separates electron–hole pairs generated during the photocatalytic process. Second, the porous hollow 2D/3D structures of the hybrid enable efficient mass diffusion, provide a large surface area and enhance light collection through mechanisms such as light reflection [149].

The design of the Cu-In-Zn-S@$ZnCo_2O_4$[@In_2O_3] photocatalyst provided an effective method to improve the efficiency of photocatalytic hydrogen production. A tandem Cu-In-Zn-S@$ZnCo_2O_4$@In_2O_3 heterogeneous photocatalyst was designed and synthesized to improve the efficiency of hydrogen production [150]. Also, TiO_2/WO_3 nanocomposites exhibited enhanced photocatalytic activity, resulting in 101.88 µmol of hydrogen in 2 h per 100 mg of the catalyst. The introduction of oxygen vacancies decreased the bandgap and expanded the light absorption range, increasing the rate of photocatalytic hydrogen production [151].

Table 4 collects examples of modern materials used for photocatalytic hydrogen production.

Table 4. Examples of materials used for photocatalytic hydrogen production.

Materials	H_2 Production Rate	Reaction Conditions	Ref.
TiO_2-based photocatalysts			
Pt single atoms (SAs) on a defective TiO_2 (Pt_1/def-TiO_2).	52,720 µmol g^{-1} h^{-1}	CH_3OH as sacrificial electron donor.	[152]
Ru on the polygonal TiO_2 sphere.	7.2 mmol g^{-1} h^{-1}	300 W Xe lamp; CH_3OH (aq.).	[153]
Ru single atoms (SAs) into N-doped TiO_2/C carrier (Ru-SAs@N-TC) derived from a MOF of NH_2-MIL-125.	100.0 µmol g^{-1} h^{-1}	300 W Xe lamp (λ = 320–780 nm); 20 mg of catalyst dispersed in 100 mL of H_2O:MeOH solution (v/v = 4:1).	[154]
Mesoporous core−shell CdS@TiO_2 with Pt.	68,000 µmol g^{-1} h^{-1}	Sunlight irradiation. 10 mg photocatalyst dispersed in 50 mL of an aqueous solution of sacrificial reagent (0.1 M Na_2S + 0.02 M Na_2SO_3).	[155]
Co-, Ni-, and Cu-doped TiO_2.	8470 µmol $h^{-1}g^{-1}$ (Cu) 3390 µmol $h^{-1}g^{-1}$ (Ni)	450 W Hg lamp. 10 mg photocatalyst in solution with 50% MeOH as a sacrificial electron donor.	[156]
Ag/TiO_2.	470 µmol $h^{-1}g^{-1}$	254 nm wavelength of UV light catalyst concentration of 20 mg/L. 50 mL of solution without sacrificial agent.	[157]
Ga-doped TiO_2.	5722 µmol $h^{-1}g^{-1}$	Side-irradiation by a 150 W xenon arc lamp equipped with an aqueous $CuSO_4$ filter (310 nm < λ < 625 nm). 3 mg of the catalyst suspended in 3 mL of aqueous methanol solution (20 vol.%).	[158]
Spherical TiO_2 particles.	350 µmol $h^{-1}g^{-1}$	ABET 150 W Xe lamp. 20 mg of catalyst dispersed in 50 mL of aqueous methanol solution (50%).	[159]
g-C_3N_4-based (graphitic carbon nitride) photocatalysts			
Pd/g-CN. Pd single atoms in the space of adjacent g-CN layers and anchored Pd atoms on the surface of g-CN.	6688 µmol g^{-1} h^{-1}	Solar simulator as a light source; 50 mg of the photocatalyst dispersed in 80 mL of water and triethanolamine solution (v/v = 9:1).	[160]
Co SAs on carbonitride, and creating an active single Co_1–N_4 site on g-C_3N_4.	10.8 mmol h^{-1}	Simulated solar irradiation (λ ≥ 300 nm). Triethanolamine (TEOA) as the sacrificial electron donor.	[161]

Table 4. *Cont.*

Materials	H_2 Production Rate	Reaction Conditions	Ref.
Co_1–P_4 site confined on g-C_3N_4 nanosheets.	410.3 mmol $h^{-1}g^{-1}$	Simulated solar irradiation. 20 mg of the photocatalyst without sacrificial electron donor.	[162]
Co–N–C/g-C_3N_4. Isolated cobalt (Co) SAs synthesized and immobilized on a porous nitrogen-doped carbon support.	1180 mmol $h^{-1}g^{-1}$	LED light source (λ = 420 ± 10 nm). 2 mg of the catalyst suspended in an aqueous solution with 10 vol.% triethanolamine (TEOA) as the sacrificial electron donor.	[163]
NiAl-LDH/gC_3N_4. gC_3N_4 coupled NiAl layered double hydroxide (LDH) nanocomposite.	3170 μmol h^{-1} g^{-1}	Stimulated light irradiation (300 mW/cm^2). 15 mg of photocatalyst suspended in 50 mL of solution containing 45 mL H_2O and 5 mL TEOA as a sacrificial reagent.	[34]
UNiMOF/g-C_3N_4. Heterostructure with 2D nickel metal organic framework (UNiMOF) nanoflakes and 2D g-C_3N_4 nanoflakes.	20.03 μmol h^{-1}	300 W Xe lamp with a 420 nm filter. 50 mg of catalyst dispersed in 90 mL of H_2O mixed with 10 mL of TEOA.	[164]
Perovskite-based photocatalysts			
Ag/$La_{0.02}Na_{0.98}TaO_3$.	330 μmol $h^{-1}g^{-1}$	UV illumination by a 500 W xenon lamp (λ > 320 nm). 50 mg of the photocatalyst dispersed in 200 mL of a glycerol solution (10 vol.%).	[165]
Ag/$KTaO_3$.	2072 μmol $h^{-1}g^{-1}$	450W Xe-Hg UV lamp. 10 mg of the photocatalyst dispersed in 38 mL water and 12 mL methanol as a sacrificial agent.	[166]
Ag/$LaNaTaO_3$.	329.5 μmol $h^{-1}g^{-1}$	500 W high-pressure xenon lamp (λ > 320 nm). 50 mg of catalyst suspended in 200 mL of 10 vol.% glycerol.	[167]
$AgInS_2$ QDs/Bi_2WO_6 composite.	611 μmol $h^{-1}g^{-1}$	1000 W xenon lamp. 0.1 g of the photocatalyst dispersed in an 80 mL aqueous solution containing 1 M NaOH, 0.1 M Na_2S*$9H_2O$, and 0.5 M Na_2SO_3.	[168]
CdS/$NiWO_4$/CoP. Composite catalyst with CoP nanoparticles as a co-catalyst modifying the CdS/$NiWO_4$ p–n heterojunction.	47.7 mmol h^{-1} g^{-1}	Visible light irradiation (λ > 400 nm). 0.01 g of catalysts dispersed in 30 mL of 10% lactic acid solution.	[169]
MOFs-based (metal–organic frameworks) photocatalysts			
TiO_2–Ti_3C_2–CoS_x. TiO_2 nanocrystal photocatalyst confined by ZIF-67-templated porous CoS_x, with conductive Ti_3C_2.	9500 μmol $h^{-1}g^{-1}$	UV–visible light irradiation methanol as the sacrificial agent.	[170]
TiO_2/Co_3O_4/Ni. Highly porous ternary photocatalyst constructed from a heterometal–organic framework (H-MOF) template (ZIF-67@MIL-125).	27,000 μmol $h^{-1}g^{-1}$	Under UV–visible light methanol solution.	[171]
TiO_2/CuO heterostructure derived from mixed-phase MOFs based on Ti and Cu metal nodes (MIL-125, Cu-BDC, MIL-125_xCu).	19,036.2 μmol $h^{-1}g^{-1}$	450 nm LED lamp. 0.2 mg of metal oxide catalysts mixed with $[Ru(bpy)_3]Cl_2 \cdot 6H_2O$ (2 mg), acetonitrile (3.8 mL), and TEOA (0.2 mL).	[172]

Table 4. *Cont.*

Materials	H_2 Production Rate	Reaction Conditions	Ref.
COFs-based (covalent organic frameworks) photocatalysts			
Vinylene-linked 2D COFs containing benzobisthiazoles units.	15.1 mmol h^{-1} g^{-1}	300 W Xe lamp (λ > 420 nm). COFs materials suspended in 0.1 M ascorbic acid solution.	[173]
CYANO-COF—cyano-containing COF with ketene-cyano (D–A) pair.	1217 μmol $h^{-1}g^{-1}$	300 W Xe lamp (λ > 420 nm). 20 mg catalyst suspended in 100 mL water, 1 wt.% Pt (co-catalyst), and 10 mmol ascorbic acid (sacrificial agent).	[174]
Tp-nC/BPy^{2+}-COFs. Cyclic diquats (viologen-derived electron-transfer mediators) integrated into a 2,2′-bipyridine-based COF through a post-quaternization reaction.	34,600 μmol $h^{-1}g^{-1}$	Visible light irradiation (λ > 420 nm) in the presence of ascorbic acid (sacrificial donor) and Pt (co-catalyst).	[175]
Varia			
CdS/$Co_{1-x}S$ HHNSs. Sugar-gourd-shaped hollow hetero-nanostructure, $Co_{1-x}S$ hollow polyhedrons skewered on CdS nanowires.	13.48 mmol h^{-1} g^{-1}	300 W Xe lamp (λ > 420 nm). 20 mg of photocatalysts dispersed in 100 mL of aqueous solution containing 20% lactic acid as the sacrificial agent.	[33]
CuIn-CdS. quantum dot level Cu and In co-doped CdS.	105.44 mmol h^{-1} g^{-1}	300 W Xe arc lamp as simulated solar light source (320–780 nm). 10 mg of photocatalyst dispersed in 25 mL of aqueous solution with 0.35 M of sodium sulfide and 0.25 M of sodium sulfite as sacrificial agent, and with 0.1 mL of H_2PtCl_6 solution (1 mg/mL) as co-catalyst.	[35]
ZCS QDs. Ni atomically dispersed in zinc sphalerite cadmium-zinc sulfide quantum dots.	18.87 mmol $h^{-1}g^{-1}$	Xe arc lamp (300 W) with a UV-cutoff filter (λ ≥ 420 nm). 10 mg of photocatalyst suspended in a mixed solution of water and TEOA (20 vol.%) as a sacrificial reagent.	[176]
Ag/ZnO/CeO_2.	18,345 μmol $h^{-1}g^{-1}$	300 W xenon lamp irradiation. 5 mg of the catalyst dispersed in 40 mL of water and 10 mL of glycerol.	[177]

5. Summary

Hydrogen has the potential to replace traditional fossil fuels and become a clean, efficient, and sustainable energy carrier for power generation and transportation. Despite its many advantages, the use of hydrogen as a fuel source still poses many challenges, including high production, storage, and transportation costs. However, in recent years, significant progress has been made in the development of renewable energy technologies that can be used to produce hydrogen in a sustainable and carbon-free manner.

The development and deployment of fuels of the future, however, comes with significant challenges, which include the synthesis of high-quality nanomaterials for their large-scale production. Despite this, the transition to alternative fuels would allow for more sustainable and environmentally conscious operations. Investment and incentives are required to stimulate market demand and provide the necessary scale. Future research should focus on meeting the challenges of developing and implementing future fuels, including synthesizing high-quality nanomaterials with photocatalytic potential and assessing their environmental impact. Overall, the use of future fuels offers a promising solution for mitigating climate change and promoting sustainable energy use, but more research is needed to ensure their successful implementation.

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