

Hydrogen energy production, storage methods, and applications for power generation

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Abstract. Nowadays, the transformation of energy structure has become an important task for society. The advantages of hydrogen energy also meet the criteria of people's choice of energy. This paper introduces hydrogen production, storage methods, and their application for the power generation. In hydrogen production part, POM is the most satisfactory of four methanol to hydrogen methods as this reaction does not require any energy and can be more than 50% efficient. In terms of photolytic water technology, Ti- and Zr-based Photocatalysts, Ta- and Nb-Based Photocatalysts, Ga and Ge-Based Photocatalysts have been applied to increase the reaction rate, in addition, the device has the advantage of a small footprint, ease of operation and high measurement accuracy. In the microbial hydrogen production section, experiments have shown that hydrogen production is highest in a thermally shocked environment, and that genetic modification of the organisms at the molecular level can also increase hydrogen production. For the hydrogen power generation, the PEMFC is by far the most used experimental product for grid-connected power generation. In the future, researchers should focus on solving various existing problems and developing more economical and efficient hydrogen power generation systems to realize the large-scale use of clean energy. Meanwhile, this paper expects to find better solutions to solve existing problems.

Keywords: hydrogen energy, hydrogen production, storage, grid-connected.

1. Introduction

Hydrogen energy as a clean and low-carbon green energy with high value, diverse sources, and flexible storage and transportation is honored as the "most ideal energy" [1]. Japan, United States, and some European countries were the first to develop and utilize hydrogen energy. Due to the early vigorous to develop heavy industry, these countries have serious environmental pollution [2]. To make up for the damage to the environment in the past, they made great efforts to study clean energy and reduce the environmental burden [2] [3]. Compared with these developed countries, China's development started relatively late and invested less in research and utilization of clean energy, especially hydrogen energy [4]. Nowadays, China has strong economic strength and capability, and people's awareness of environmental protection has gradually improved, no longer taking the route of "development first and treatment later", but achieving the transformation of structure to energy, building a low-carbon and environmentally friendly green society [5]. The hydropower station is the first project to realize the transformation of energy structure in China. Its principle is to generate electric energy by using the water level drop, and there is no pollution in the whole process. However, many years of investigation found that the construction of the hydropower station has seriously affected the downstream ecology, and the power supply is also affected by the season [6]. To solve these problems, the local government decided to dismantle some hydropower stations and advocate using other ways to generate power. Hydrogen energy as new clean energy is given priority in the scope of use.

In the whole hydrogen power generation system, hydrogen production is the first step. With the rapid progress of science and technology, hydrogen energy is produced in a variety of ways. Electrolytic water is the earliest green hydrogen production method. In 2013, Santos et al introduced the chemical principle of alkaline water electrolysis in detail in their paper and compared and analyzed other electrolytic water technologies, providing direction for the main research needs of subsequent technology development [7]. Hydrogen storage is the second step of the hydrogen power generation system. Hydrogen has the nature of extremely flammable, it is not easy to store. For better

application, researchers have evaluated several current hydrogen storage methods, but because each hydrogen storage method has its advantages in different situations, it is difficult to determine which method is the best. The last step is using hydrogen fuel cells to generate power, in 2016, 30 large-area proton exchange membrane fuel cells successfully worked for 2000 hours in a steady state, which is a tremendous advance [8].

This paper is to introduce the methods, performance indicators, advantages and disadvantages, and improvement measures of hydrogen production, hydrogen storage, and power generation, to help people develop a hydrogen power generation system with excellent performance, and make the clean fuel widely available.

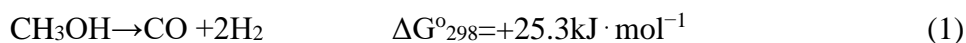
2. Hydrogen production technologies

2.1. Hydrogen production from methanol

In some of the early experiments, scientists evaluated four methanol-reforming reactions [9]. The following is a rough description of these four methods.

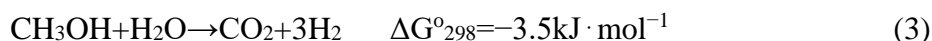
2.1.1. Decomposition of methanol.

The decomposition of methanol is the easiest of the above four reactions, as shown in Eq (1). According to the Gibb free energy, when ΔG is a positive value, it means this is a non-spontaneous and endothermic reaction that requires a lot of energy to push. Besides, the reaction produces a large amount of carbon monoxide, about 33% [10]. Some studies indicate that lots of carbon monoxide can damage the catalyst in the fuel cell and then affect hydrogen production. To improve efficiency and the amount of hydrogen, the industry uses a method called water gas shift Eq (2) to absorb the generated carbon monoxide and make it generate hydrogen and non-toxic carbon dioxide again [9]. When the content of carbon monoxide is more than 10%, people need to use the water-gas conversion method to absorb by-products many times [9].



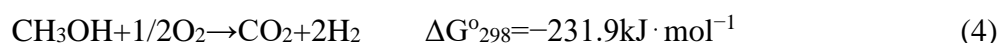
2.1.2. Steam reforming of methanol.

The method about SRM combined DM and water-gas shift methods, as shown in Eq (3). As early as 1970, scientists use the SRM method to produce hydrogen and use palladium membrane to divide carbon monoxide and carbon dioxide [11]. However, due to the price of Pd being too high, this experiment did not continue. Since then, researchers began to pay attention to catalysts and wanted to achieve better results by using catalysts. In 1992, copper-based catalysts were used in this field, achieving the goals of unidirectional conversion of methanol up to 99%, selectivity of hydrogen up to 99.5%, and low content by-products such as carbon monoxide and methane. SRM has become the most productive method of hydrogen quantity, only this process still needs massive heating to make the water become steam [9].



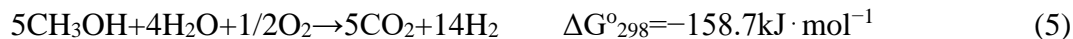
2.1.3. Partial oxidation of methanol.

Different from the reaction's first two methods, POM does not require an external heat supply. According to the Gibb free energy, when ΔG is a negative value, the reaction is spontaneous, and this also means the reaction does not need external energy, as shown in Eq (4). The bad is POM method produces hydrogen with a low content of only 40% [9].



2.1.4. Oxidative steam reforming of methanol.

Owing to POM method has a low hydrogen production, scientists decided to add water to improve the content of hydrogen, as shown in Eq (5). This reaction also is spontaneous, without the need for heat, the hydrogen content is increased to more than 50% [12].



2.2. Hydrogen production by photolysis of water

In the past, people generate electricity by burning fossil fuels, but it does a lot of damage to the environment. With the development of technology and the gradual improvement of people's awareness of environmental protection, scientists start to research clean energy and also hope the process of generating clean energy is green and pollution-free. The photolysis of aquatic hydrogen to generate electricity helps scientists achieve this vision.

Photo water converts solar energy into hydrogen energy using semiconductor photocatalysis. Fig. 1 shows a elementary diagram of the photolysis process. After absorbing a certain amount of photons, the photocatalyst generates electrons and holes through bandgap excitation [13]. Subsequently, the electrons and holes separate, they move toward the surface of the photocatalyst respectively [13]. The electrons migrate to the surface of the catalyst to convert hydrogen ions into hydrogen, and holes convert water into oxygen. Since 1972, when Fujishima and Honda first used photo water to produce hydrogen, scientists have been exploring what photocatalysis can be used in this technology and increase the reaction rate [14]. So far, there are several types of photocatalysis that have been tested: Ti- and Zr-based Photocatalysts, Ta- and Nb-Based Photocatalysts, Ga and Ge-Based Photocatalysts [15]. Combine these photocatalysis properties, the catalyst should include the three features: moderate band gap width, good light corrosion resistance and chemical stability; the photocatalysis has to have high crystallinity because higher crystallinity leads to fewer defects and better charge transfer; the active site of the photocatalyst should be sufficient [15].

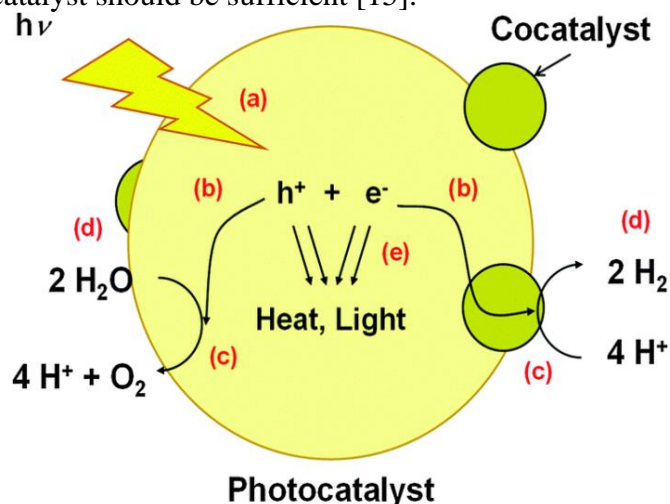


Figure 1. Elementary diagram of the photolysis process. (a) Photoabsorption (b) Transfer charge (c) Oxidation-reduction reactions (d) Product of reaction (e) Charge recombination [13].

This technology not only is a very green way to go, but it also solves the problem of the instability of direct solar power generation. Meanwhile, it also has the advantages of a small footprint, convenient operation, and high detection accuracy. Based on the related website, the total size of this system is only 680*450*980mm which saves a lot of space for the laboratories. Second, the operation is very simple, ready to use, sampling, and testing is a one-stop service. Third, it is suitable for all kinds of experimental requirements of extremely small to major gas collection and detection, because this system can measure accuracy up to 1ppm.

2.3. Microbial hydrogen production

As a whole, the method of microbial hydrogen production is not very mature, so this technology is not being used on a large scale. However, scientists still want to develop this new field to add away without pollution in the process. At present, the hydrogen-producing microorganisms divide into four kinds: green algae, cyanobacteria, photosynthetic bacteria, and fermenting bacteria [16]. This paper only chooses to ferment bacteria to introduce some studies about it. In 2019, Yang et al designed gamma-radiation, heat-shock, base, acid, and aeration five situation to measure fermenting bacteria's diversity and hydrogen production in every [17]. They found that all five conditions can increase the community of microorganisms [17]. Among them, the best hydrogen production is the thermal shock method, results showed the maximum hydrogen production rate is 37.65 ml / h and the maximum hydrogen production is 1.58 mol/mol glucose [17]. However, this result does not meet people's needs. It is necessary to use molecular-level methods to improve the content of target products. The research shows that the dynamic method with the highest hydrogen production is the engineering strain that knocks out the HUP gene [18]. Its principle is to inactivate HUP hydrogenase by using modern technology (the absorption of hydrogen by HUP hydrogenase will affect the hydrogen production effect of nitrogen-fixing bacteria), when the gene of hydrogenase is inactivated or mutated, the hydrogen absorption of nitrogen-fixing bacteria will be reduced, so more hydrogen will be produced [18].

3. Hydrogen storage technologies

Hydrogen energy conducts a very promising clean energy, it has more than a dozen storage methods, the most common methods are the following three.

3.1. High-pressure gaseous hydrogen storage

The HGHS can reach such a level of maturity, which is inseparable from a lot of research and improvement done in a very early period. At first, hydrogen was stored in seamless steel cylinders (type I) and steel tubes. Whereas these steel materials could not cope with hydrogen embrittlement [19]. In 1970, people use aluminum instead of steel. Although the aluminum tank will not face the problem of hydrogen embrittlement, compared with the steel cylinder, the aluminum tank has a higher cost and less water storage [19]. Nowadays, people pay more and more attention to the research of composites. Steel liner fiber circular winding cylinders (type II), aluminum liner fiber fully winding cylinders (type III), and plastic liner fiber winding cylinders (type IV) all belong to the composite hydrogen storage vessels [20]. These fiber composites are spirally wound on the outside of the energy storage tank to make the structure more stable [19].

3.1.1. Seamless steel cylinders (type I).

The first safe transportation of hydrogen under high pressure was a steel cylinder [19]. In 1885, a seamless steel cylinder came out (Fig. 2) [19]. The hydrogen storage of type I is a relatively direct and common way of hydrogen storage. The gas can be released through the regulation of a high-pressure valve. It can operate at room temperature and 300 bar pressure [19]. Since the heavy weight of all-metal steel cylinders, low hydrogen storage density, and it requires high cost to compress hydrogen, resulting in the limited application of type I cylinders in the industry [21].

3.1.2. Steel liner fiber circular winding cylinders (type II).

A type II vessel is a container that combines metal with a nonmetal, the structure is a metal lining with glass fiber wound around it (Fig. 2) [19]. This circular wrapping only covers the body of the vessel, the neck still made of steel. Using fiber as a pressure bearing layer can make hydrogen storage pressure up to 400 bar, compare with type I vessel, the tolerance to stress of type II improved [19]. And because the metal lining does not need to load the pressure, the weight of the vessel is low. As far as the current application scenario is concerned, type II vessels are mostly used for fixed energy supply.

3.1.3. Aluminum liner fiber fully winding cylinders (type III).

Hydrogen being a small molecule substance, is very easy to diffuse and permeate. The basic requirement of the lining of a high-pressure hydrogen storage vessel is strong hydrogen permeability resistance. Considering other general metal resistance to hydrogen penetration, weight, cost, and other factors, the aluminum alloy lining was a good choice at that time [22]. Compare with type II, the packaging area and method have been changed, from the original packaging only vessel body to all packaging, and the winding forms become polar winding and spiral winding (Fig. 2) [20]. This winding method can withstand higher compression stress, which greatly improves cylinder cycle life. In addition, the type III vessel can ensure that when rapidly releasing large amounts of hydrogen, this tank will not be affected by sudden temperature drops caused by pressure release.

3.1.4. Plastic liner fiber winding cylinders (type IV).

Different from type III, type IV vessel adopts a polyethylene plastic liner, which greatly reduces the weight of the hydrogen storage tank (Fig. 2) [19]. The periphery is wrapped in a new material - carbon fiber [22]. Carbon fiber is material scientists have been looking for. It has the characteristics of good flexibility and meets the stress bearing range required by the high-pressure hydrogen storage vessel. The tank measured to withstand the pressure of 700 bar, and 1000 bar is being developed [22].

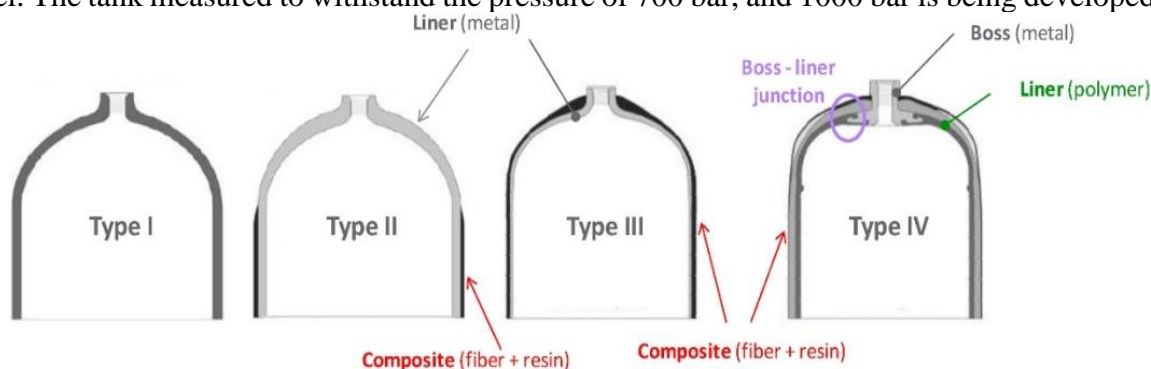


Figure 2. The style of hydrogen storage vessels and fiber winding mode [19].

3.2. Liquefied hydrogen storage

Hydrogen liquefaction technology has long been mature in some developed countries. According to statistics, more than 85% of the world's liquid hydrogen is produced and stored in North America [23]. However, in China, there are only three liquefaction plants, all of which serve space rocket launches [24].

The cryo-compressed hydrogen storage system mainly includes a compression unit, a refrigeration unit, and storage unit, this system working principle is shown in Fig. 3. In the compression stage, hydrogen enters several compressor cooling units for initial compression cooling [25]. Then the hydrogen will be cooled in refrigeration, for a instant, liquid hydrogen can make the temperature reach 80K, a special refrigeration allows the temperature zone to reach 30-80K [25]. The last stage is the need to store hydrogen in a vessel [25].

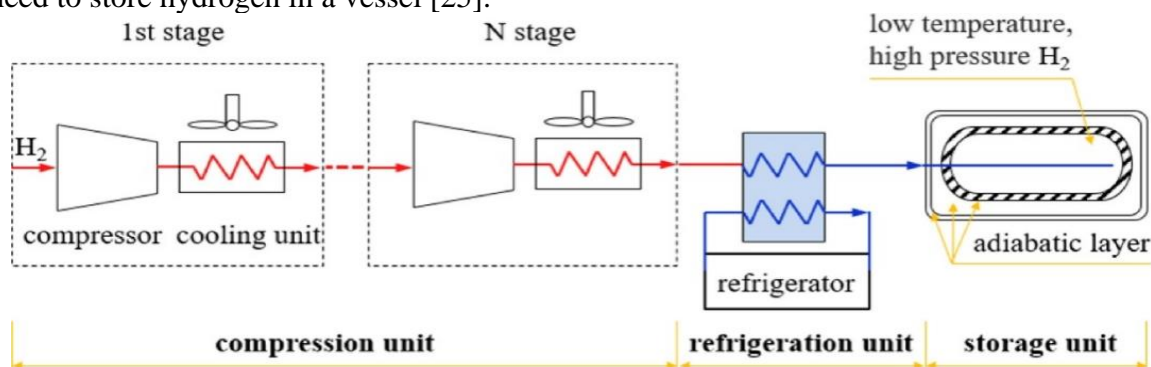


Figure 3. The working principle of cryo-compressed hydrogen storage [25].

This compact, high-capacity liquefied hydrogen storage technology saves cost because it requires less material to manufacture, to further help people achieve the goal of storage hydrogen with high efficiency [26]. However, some researchers think that liquefied hydrogen storage needs larger energy consumption. Because hydrogen needs to be liquefied under 21K temperature, this requires a low heating value of more than 30% hydrogen energy to be used in the process, compare with normal gas compression, the energy consumption of liquefaction is about twice that of gaseous compression [26]. Meanwhile, another problem is the vaporization of hydrogen. According to the report, there are 2%-3% of hydrogen energy will lose each day due to unavoidable heat entering the storage vessel [26]. To solve this problem, in recent years, scientists have developed a kind of storage tank suitable for storing liquefied hydrogen---cryogenic pressure vessels (CPV). A CPV consists of a inner container made of carbon-fiber coated metal which can bear high pressure, a vacuum space which has a large amount of highly reflective metalized plastic, and a metal casing [26]. In particular, a vacuum filled with a large amount of highly reflective metalized plastic can provide better thermal insulation, making CPV more heat resistant than conventional cryogenic hydrogen storage vessels [26]. Besides, the cryogenic pressure vessel can work at temperatures at 20K and withstand pressures up to 350bar [26].

3.3. Metal hydrogen storage

Metal hydrogen storage is a kind of advanced technology, that has a great development prospect. At present, scientists have been researching two kinds of methods of using metal to store hydrogen, respectively physical absorption and chemical absorption [27]. For physical absorption, the main working principle is to adsorb gas on porous materials by van der Waals force [27]. Materials that can be used for physical adsorption and storage mainly include metal-organic skeleton materials, covalent organic compound materials, carbon-based hydrogen storage materials, and inorganic porous materials [28]. While, Chemical absorption technology uses a chemical reaction between metal and hydrogen, making hydrogen atoms or molecules bind tightly with metal to store energy [26]. This paper just focuses on the chemical storage of hydrogen.

3.3.1. Metal hydride.

Common metals that can react with hydrogen include Na, Mg, Ca, and Li [26]. Among them, the maximum hydrogen capacity of LiH can reach 12.4% *wt*, followed by MgH₂, which can reach 7.6% *wt* [29]. However, judging from the current application volume, people tend to use more magnesium alloys [30]. In terms of the temperature required for decomposition, MgH₂ needs 600K can be decomposed, while, the temperature of LiH is around 400 K higher than the former [26]. Moreover, the price of Mg is relatively cheap, it is very suitable for large-scale applications [30]. The only disadvantage of MgH₂ is that the reaction is very slow when people use pure Mg and the decomposition temperature and pressure are still high [30]. Generally speaking, although this hydrogen storage technology is not as mature as the first two methods, by contrast, the hydrogen capacity has increased. According to some reports, the hydrogen storage capacity of this hydrogen storage alloy is very strong.

Hydrogen storage alloy except can store hydrogen, it can also be exothermic when hydrogen is absorbed, or endothermic when hydrogen is released [31]. Therefore, people use this principle to make refrigeration or heating equipment. The United States and Japan have used the exothermic - endothermic cycle to build solar and cooling-heating rooms. In China, this loop has been used in the situation of the industry and medical care.

3.3.2. Compound hydride.

Compound hydride can form strong bonds with hydrogen and generally have a high hydrogen capacity. For example, lighter metals such as lithium and sodium may have higher hydrogen capacities in the formation of complex metal hydrides due to their lightweight, which usually results in two atoms per metal atom [26]. At the moment, the most popular complex metal hydrides are NaAlH₄ and borohydrides. There is data showing, that the hydrogen capacity of NaAlH₄ is 5.6% *wt*,

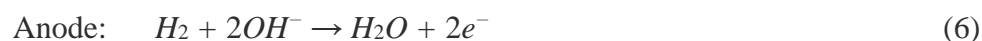
and its dehydrogenation temperature is about 450 °C [26]. Borohydrides are a very complex compound, also called LiBH_4 [26]. Most of them have a hydrogen storage capability which can achieve around 10% wt, some even more than 10% wt [26]. The range of dehydrogenation temperature is from 350-400 °C [26].

4. Hydrogen power generation

4.1. Fuel cell

4.1.1. Alkaline fuel cell (AFC).

The most mature fuel cell is AFC. AFC uses KOH as an electrolyte, uses hydrogen and oxygen as material [32]. Hydrogen and hydroxide in the electrolyte oxidation reaction under the action of the anode catalyst, to generate water, generated electrons through the external circuit to the cathode, under the action of the cathode catalyst, participate in the reduction of oxygen reaction to generate hydroxide [32]. The reaction is shown in Eq (6) and (7).



Different from acid fuel cells, the alkaline fuel cells can work in a wide range of temperatures which from 80-230 °C, and the efficiency can reach 60% [26]. The most important thing is that it does not need Pt as an expensive metal catalyst, it can realize the use of non-precious metals, which gives AFC the advantage of low cost [26].

4.1.2. Proton exchange membrane fuel cell (PEMFC).

The proton membrane of PEMFC is an electrolyte membrane with a solid polymer, in general, the membrane requires constant humidification to ensure good conductivity [26]. Another factor that affects fuel cell efficiency is the catalyst. Previously, scientists used platinum-based catalysts, due to the shortcoming of high cost and short life, it has not been widely used [33]. Later, people thought grinding Pt into granules would increase the reaction rate and durability. But that still doesn't solve the problem of high costs [33]. Now scientists want to develop non-precious metal catalysts, of which iron and cobalt-based catalysts are considered the most promising [33]. Most PEMFCs in the market can work under the temperature of 60-100 °C, efficiency can reach 55%, if the parameters of the vehicle fuel cell are taken as the standard, working temperature and efficiency stills need to be improved [26].

4.1.3. Molten carbonate fuel cell (MCFC).

The MCFC is suitable for working at very high temperatures (600-700 °C) [34]. In addition, this device does not accept highly purified hydrogen, so people generally use hydrogen-rich gas as the fuel which could omit the process of hydrogen purification [26]. Owing to MCFC's use non-platinum catalysts and simple ancillary systems, it also has a relatively low cost.

However, there are some drawbacks along with improved performance. For example, too high a temperature will influence the lifetime and power density, according to some reports, the lifetime of this fuel cell is just 5 years [35, 36]. Another point is due to the fuel cell system needs the circulation of carbon dioxide, so the carbon dioxide produced from the anode must transfer to the cathode which makes the system a little bit more complex [26].

4.1.4. Solid oxide fuel cell (SOFC).

At present, researchers have divided SOFC into two kinds based on the ions present in conducting electrolytes, containing oxygen ions conductive electrolyte is called SOFC (-), containing hydrogen proton conductive electrolyte is called SOFC (+) [26]. Similar to the MCFC, SOFC can work under high temperatures, according to the data show, the operation temperature of SOFC (-) can reach

around 1000 °C [26]. Compare with SOFC (-), the working temperature of SOFC (+) is relatively low from 200-700 °C. The higher temperature, the longer it takes to turn it on and off [38]. According to the investigation, turn on and off this system, people need to spend 12 hours or longer time to wait, so SOFC is always in the working situation [37]. To solve this problem, scientists are studying microtubule SOFC which could save time [26].

4.2. Application case

Among the above fuel cells, some have participated in the hydrogen energy power generation system and successfully realized grid-connected power generation [38]. In 2014, Dr. Zhang et al evaluated the overall performance of grid-connected microgrids [38]. The experiment was divided into two groups, one group was a smart grid with 300 PEMFC s in series, and another group is a traditional electricity grid device [38]. After working a few dozen hours at their suitable temperatures, comparing data found the service quality of the former was better than this of the latter [38].

5. Conclusion

The wide application of hydrogen power generation systems will reduce the degree of environmental pollution. This paper introduces each part of the power generation system one by one. In the section on hydrogen production, POM is the most satisfactory of the four methanol to hydrogen methods in terms of reaction energy consumption and hydrogen yield. In photolytic water technology, there are many new varieties of catalysts entering the experimental field, and in addition the device has the characteristics of small footprint, operate easily and high measurement accuracy. In the microbial hydrogen production part, it was experimentally demonstrated that microorganisms produce the highest hydrogen in a thermal shock environment, to increase the yield, scientists have modified the biological genes at the molecular level. In terms of hydrogen storage technology, the pressure tolerance value of the hydrogen storage vessel has been significantly improved after several times of improvement. The working principle of liquefied hydrogen storage is more technical and the performance index is better than the former. Metal hydrogen storage also gives a further improvement in hydrogen storage capacity. About hydrogen power generation, PEMFC is by far the most applied experiment, which has successfully achieved grid-connected power generation. In the future, researchers should focus on solving various existing problems, developing more economical and efficient hydrogen power generation systems, and realizing the large-scale use of clean energy.

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