Analysis and Prospect of Key Technologies for CCUS Coupling Hydrogen Production

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Abstract. Systematically studied the process principle, system construction, supporting equipment and application scenarios of CCUS key technologies, and deeply analyzed the carbon dioxide capture purification and comprehensive utilization technology; At the same time, the process principle and development status of various key technologies for hydrogen production in the field of hydrogen energy development are presented in detail, mainly including hydrogen production from fossil fuels, hydrogen production from electrolytic water, hydrogen production from high-temperature cracking of chemical raw materials, hydrogen production from industrial by-product gas and other key hydrogen production technologies. The advantages and disadvantages of various hydrogen energy production technologies are compared and analyzed. Finally, based on the low-carbon, clean and sustainable development strategy, the development route and supporting process system of low-carbon hydrogen production key technologies coupled with CCUS technology are deeply analyzed, and innovative proposal of CCUS coupled hydrogen production process system. At the same time, in-depth thinking and practical prospect are carried out to realize the sustainable and large-scale development direction of hydrogen energy. The analysis shows that with the implementation of the global carbon-neutralization strategy, the development potential of green, low-carbon and renewable new energy represented by hydrogen energy is huge. However, as the main source of hydrogen energy, the use of fossil fuels for hydrogen production seriously restricts the large-scale utilization of hydrogen energy due to the high carbon emissions in the hydrogen production process. Therefore, actively promoting the research and project construction

J. Lin (Ed.): IFEDC 2023, SSGG, pp. 676–697, 2024.
https://doi.org/10.1007/978-981-97-0268-8_53

of CCUS coupled hydrogen production key technologies is of great significance for promoting global energy structure transformation, as well as deep emission reduction and comprehensive utilization of carbon dioxide.

Keywords: Hydrogen production · Carbon dioxide capture · CCUS technology · Low carbon

1 Introduction

In recent years, with the high-quality and accelerated development of the economy and society of all countries in the world, the growth rate of global energy consumption has surged year by year, resulting in a sharp increase in the consumption of traditional fossil energy, mainly oil, coal, natural gas, and the sharp decrease in the available reserves. At the same time, the traditional use of fossil energy has exacerbated the global carbon emissions greenhouse effect and environmental pollution and other problems. Therefore, the global demand for clean, high energy storage density Low-carbon development and utilization of sustainable new alternative energy is imminent [1, 2].

Among them, hydrogen energy, as a new type of high-quality clean energy and energy storage carrier with no pollution, high energy storage density and renewable energy, can not only directly supply energy through electrochemical reaction or combustion reaction, but also can couple solar energy, wind energy and other volatile renewable energy for energy storage and supply. Hydrogen energy has many advantages, such as zero carbon, clean, high energy storage density, recyclable, extensive sources and various forms of utilization, and it can also react with carbon dioxide to obtain important chemical products such as methanol and methane, which is of great significance for promoting the green and low-carbon transformation of the global energy structure, promoting the large-scale utilization of renewable energy and achieving the goal of carbon neutrality in an all-round way.

However, in order to realize the clean, low-carbon, large-scale and commercial application of hydrogen energy, there are still a series of key technical problems to be solved. Among them, the development of safe, economic, energy-saving and environmentally friendly low-carbon hydrogen production technology is the key technical guarantee for the large-scale development and utilization of hydrogen energy in the future [3, 4]. At present, the development of hydrogen energy preparation technology is diverse, but the problems of high carbon emissions and poor economy in the process of hydrogen production seriously restrict the large-scale clean utilization of hydrogen energy. At the same time, carbon capture, utilization and storage (CCUS) technology, as the most promising large-scale industrial decarburization and emission reduction technology at present, is an important cornerstone to achieve the net zero emission of fossil energy. The research on key technologies of CCUS coupling hydrogen production and the realization of multi-energy complementation of the two technologies will effectively promote the large-scale low-carbon clean utilization of hydrogen energy and the large-scale consumption of renewable energy. Therefore, this paper systematically analyzes the process principle, system architecture, key supporting equipment and main adaptation scenarios of CCUS technology, and deeply analyzes the carbon dioxide capture purification and comprehensive utilization technology. The characteristics, application status and research focus of various key technologies for hydrogen production in the field of hydrogen energy development are presented in detail. The advantages and disadvantages of existing technologies and the bottleneck problems are summarized. Finally, based on the low-carbon, clean and sustainable development strategy, the development route and supporting process system of low-carbon hydrogen production key technologies coupled with CCUS technology are deeply analyzed, and innovative proposal of CCUS coupled hydrogen production process system. At the same time, the research direction is proposed for realizing the sustainable and large-scale development of hydrogen energy, which is of great significance for promoting the environmental protection and low-carbon transformation of energy structure, promoting the large-scale utilization of clean energy, and promoting the deep emission reduction and comprehensive utilization of carbon dioxide.

2 CCUS Key Technology

Recently, with the gradual implementation of the global carbon emissions trading and the supporting driving policies of the "peak carbon dioxide emission and carbon neutrality" target, the research on the deep carbon dioxide emission reduction technology has been deepened. At present, the absorption and neutralization of carbon dioxide can't be completely realized only by improving energy efficiency, afforestation and other traditional methods. Therefore, we must rely on large-scale deep decarburization and emission reduction technology represented by CCUS technology to achieve large-scale "negative emission" and resource utilization of carbon dioxide. The key technology of CCUS is to separate and utilize the carbon dioxide generated in the process of industrial processing and energy utilization, and seal the excess carbon dioxide from the atmosphere, which is an important way to achieve large-scale and industrialized carbon dioxide emission reduction [5, 6].

According to the process flow, CCUS key technologies can be subdivided into four technical links: carbon dioxide capture, transportation, utilization and storage, as shown in Fig. 1 [7]. Among them, carbon capture technology can be divided into pre-combustion capture, post-combustion capture and oxygen-enriched combustion capture according to different stages of carbon dioxide separation [8]. Carbon dioxide transportation technology is to realize the storage and transportation of gaseous or liquid carbon dioxide, which is mainly transported by high-pressure carbon dioxide gas tank car or pipeline. In addition, carbon dioxide utilization technology is mainly used for food-grade utilization, chemical product conversion, oil displacement and production increase, biological breeding, etc. Carbon dioxide storage technologies mainly include deep-sea storage, geological storage, biological storage, mineralization storage, etc.

Pre-combustion capture technology mainly means that after the gasification reaction of fossil fuels such as coal gasification and methane steam reforming, the carbon monoxide in the multi-component synthesis gas is separated, and then the hydrogen and carbon dioxide are obtained through the conversion reaction with high-temperature steam and catalyst, and then the carbon dioxide is separated and captured. This technology has low operating cost and high carbon capture purity, but it is mainly used for fossil fuel gasification technology. Post-combustion capture technology is mainly to install carbon dioxide

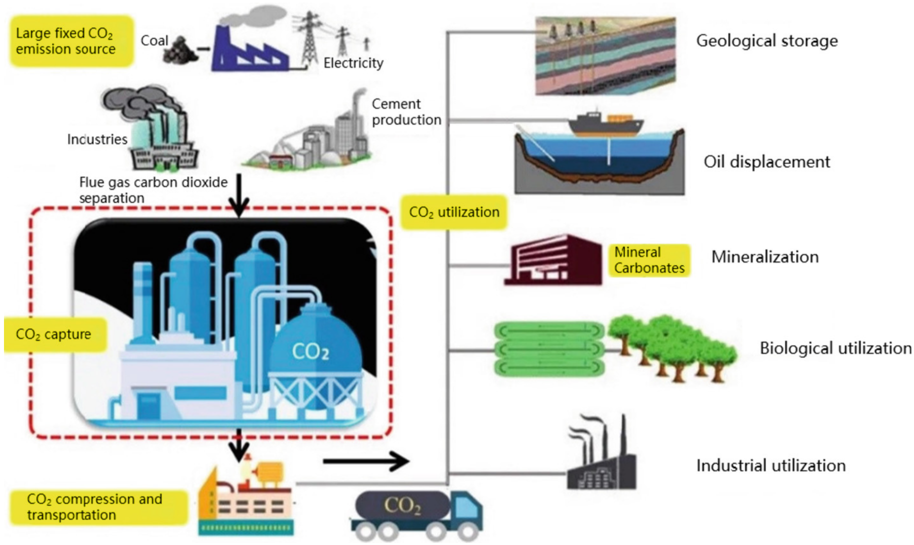


Fig. 1. CCUS process flow diagram.

separation and collection device after the original combustion system. This technology has high maturity, wide application range, simple process, but high energy consumption cost. The oxygen-enriched combustion capture technology mainly uses the mixture of oxygen and carbon dioxide as the combustion reaction medium. After combustion, the carbon dioxide in the production gas will be separated and collected, and some of the carbon dioxide will be mixed with oxygen again to participate in the cycle combustion reaction. This technology has low energy consumption cost and high carbon capture purity, but it needs to add an air separation oxygen generation device.

3 Key Technologies of Hydrogen Energy Preparation

Hydrogen is the most abundant element in nature and widely distributed, but it mainly exists in the form of compounds. Therefore, hydrogen energy preparation technology is the process of separating hydrogen from various hydrogen-containing compounds or multi-component gases [9]. In recent years, with the continuous development of the field of hydrogen energy development, hydrogen energy preparation technology has also made significant progress. At present, hydrogen production technologies at home and abroad mainly include: hydrogen production from fossil energy, hydrogen production from industrial by-product gas, hydrogen production from high-temperature cracking of chemical raw materials, hydrogen production from electrolytic water and other hydrogen production technologies.

Among them, fossil energy hydrogen production technology can be subdivided into coal, natural gas, petroleum hydrogen production, etc. The technology of hydrogen production from industrial by-product gas mainly includes the separation and purification of by-product gas for hydrogen production from electrolytic salt and alkali production industry, petroleum refining industry, coal coke chemical industry, synthetic ammonia fertilizer industry, etc. High temperature cracking hydrogen production technology of chemical raw materials mainly includes methanol reforming hydrogen production and ammonia decomposition hydrogen production; The technology of hydrogen production by electrolysis of water includes the use of traditional electric energy to hydrolyze hydrogen and the use of solar energy, wind energy, tidal energy and other renewable energy to produce hydrogen by electrolysis of water. In addition, other hydrogen energy preparation technologies such as hydrogen production from photolysis of water and biomass are in the experimental research stage, but have not yet been industrialized (as shown in Fig. 2) [10].

China is the world's largest hydrogen producer, with a hydrogen production capacity of approximately 41 million tons per year and a production capacity of 37.81 million tons per year in 2022. It is predicted that under the 2030 carbon peak vision, China's hydrogen production is expected to exceed 50 million tons/year. At present, almost all hydrogen production in China comes from fossil energy and industrial by-product hydrogen. These two hydrogen production paths have mature technology, large production capacity, wide distribution of production capacity, and low costs. However, most of them are gray hydrogen produced from carbon based energy, and their carbon emissions are relatively high. Coal based hydrogen production has the lowest cost, but its emissions are high. Renewable energy based hydrogen production has no carbon emissions, and its cost is three times that of coal based hydrogen production.

In China, coal is the main hydrogen source, and renewable energy electrolysis of water to produce hydrogen is limited by high cost, accounting for a small proportion. Currently, there is a significant gap between China's hydrogen source structure and the world's hydrogen source structure. From the perspective of global hydrogen source structure, 48% of hydrogen comes from natural gas, 30% comes from by-product hydrogen, and 18% comes from coal. At present, China is still dominated by hydrogen production from coal, accounting for 62%, 19% from natural gas, 18% from petroleum and industrial by-products, and only 1% from electrolysis of water. China's hydrogen source structure is related to the resource endowment of "rich coal, lack of oil, and lack of gas", but the proportion of renewable energy electrolysis of water to produce hydrogen is small, and the main limiting factor is the high cost, of which electricity price accounts for 60%–70% of the total cost. Although in recent years, the cost of solar cost of electricity by source in China has declined significantly, and some regions have achieved parity online, the comprehensive cost of renewable energy electrolysis of water to produce hydrogen is still about three times that of coal hydrogen production, and twice that of coal hydrogen production + CCS. Therefore, electrolysis of water to produce hydrogen cannot completely replace fossil fuel hydrogen production in the short term.

3.1 Hydrogen Production from Fossil Energy

Hydrogen production from fossil fuels relies on a series of complex reactions such as oxidation, reduction, catalysis, transformation and steam reforming of fossil fuels to produce multi-component products mainly composed of carbon monoxide, hydrogen and carbon dioxide. After separation and purification, high-purity hydrogen is obtained.

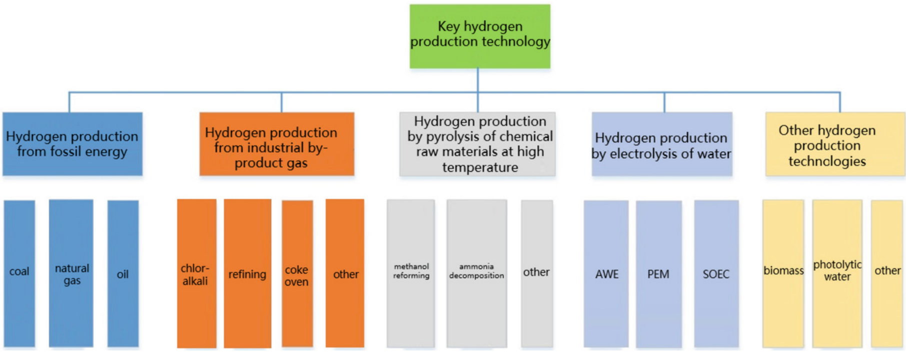


Fig. 2. Classification of key hydrogen production technologies.

According to the classification of fossil fuels involved in the main reaction, the technology of hydrogen production from fossil energy can be subdivided into coal, natural gas and petroleum [11, 12]. According to statistics, more than 95% of the world's existing industrial hydrogen production mainly comes from fossil energy, including about 22% from coal, about 41% from natural gas and about 32% from oil.

Hydrogen Production from Coal

According to different reaction conditions, coal hydrogen production technology can be divided into coal coking and coal gasification [13, 14]. Among them, the coal coking hydrogen production process is to isolate the coal from the air and raise the temperature to about 1000 °C, so that the coal can be decomposed at high temperature to obtain multi-component products such as coal coke, semi-coke, coke oven gas and tar, and then separate and purify the coke oven gas mainly composed of hydrogen, methane and carbon monoxide to produce high-purity hydrogen. Coal gasification hydrogen production process is to place coal in a high-temperature gasifier and conduct a series of physical and chemical reactions with gasification agents (oxygen-enriched air, water vapor), such as drying, oxidation, reduction, methanation, pyrolysis, etc., to generate multi-component products mainly composed of carbon monoxide, hydrogen and methane, and then obtain high-purity hydrogen through carbon monoxide transformation, purification, separation and purification. Compared with coal coking, coal gasification reaction conditions are easy to meet and the process is easy to control. Therefore, coal gasification hydrogen production technology is mainly used to prepare hydrogen from coal in industrial production. The process flow is shown in Fig. 3.

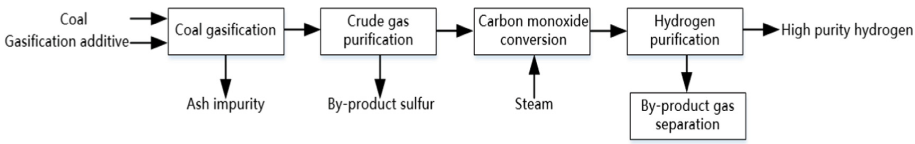


Fig. 3. Process flow of hydrogen production from coal gasification.

Because China's fossil energy structure has the characteristics of "rich coal, less gas, and poor oil", coal resources account for about 70%, and coal is rich in hydrogen production raw material resources and low in cost [15, 16]. Therefore, hydrogen production from coal has the advantages of economic efficiency, mature technology and equipment, and is widely used in the field of hydrogen production in China. However, due to the large amount of carbon dioxide produced by the process of hydrogen production from coal, if it is discharged directly without treatment, it will aggravate the greenhouse effect and pollute the environment. In the important period of achieving the "peak carbon dioxide emission and carbon neutrality" target, its environmental cost is obviously high.

Hydrogen Production from Natural Gas

Methane (CH4), the main component of natural gas, is the compound containing the largest proportion of hydrogen by mass, with a hydrogen content of up to 25%. Meanwhile, as one of the three major fossil energy sources in the world, natural gas has abundant reserves, low cost and mature relevant utilization technology. According to statistics, hydrogen production from natural gas accounts for about 40% of the world's existing industrial hydrogen production. Therefore, natural gas hydrogen production technology has become the most mainstream hydrogen energy preparation technology in the world [17, 18].

According to different reaction principles, natural gas hydrogen production technology can be divided into three ways: methane steam reforming hydrogen production, natural gas aromatization and natural gas cracking hydrogen production. Among them, the steam reforming process of methane hydrogen production is the most mature. The main principle is that natural gas is placed in a high temperature reformer to undergo steam conversion reaction and oxidation reaction with water vapor and oxygen-rich air to obtain hydrogen, carbon monoxide and other multi-component gases. Then it enters the catalyst fixed bed reactor, where the carbon monoxide conversion reaction occurs under the action of high temperature water vapor and catalyst, and the carbon monoxide is converted into hydrogen and carbon dioxide. Finally, the multi-component product was purified, separated and purified to obtain high purity hydrogen. This hydrogen production process is mature and economical, and is the most widely used natural gas hydrogen production process at present. However, carbon dioxide production accounts for about 80% of by-product gas. The process flow is shown in Fig. 4, and the main reactions are as follows:

$$CH_4 + H_2O \xrightarrow{\text{high temperature}} 3H_2 + CO \text{, Steam reforming reaction} \tag{1}$$

$$CH_4 + \frac{1}{2}O_2 \xrightarrow{\text{high temperature}} 2H_2 + CO \text{, Oxidation reaction} \tag{2}$$

$$CO + H_2O \xrightarrow[\text{catalyzer}]{\text{high temperature}} CO_2 + H_2 \text{, Carbon monoxide shift reaction} \tag{3}$$

Oxygen-free aromatization hydrogen production process of natural gas is a new technology for hydrogen production from natural gas developed in recent years. Compared with methane steam reforming hydrogen production process, it can directly obtain pure

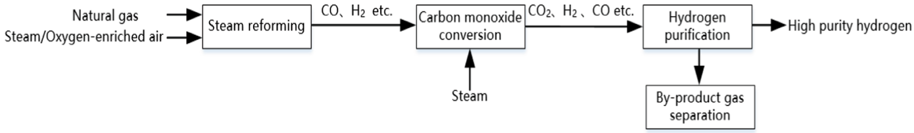


Fig. 4. Methane steam reforming hydrogen production process.

hydrogen without by-products such as carbon monoxide and carbon dioxide and aromatic products with high added value, which is easy to separate and produce high-purity hydrogen, but limited by the low conversion efficiency of methane, its main reactions are as follows:

$$6CH_4 \xrightarrow{\text{high temperature}} 9H_2 + C_6H_6, \quad \text{Oxygen-free aromatization reaction} \tag{4}$$

In addition, the process of hydrogen production from natural gas cracking is another kind of new technology of hydrogen production from natural gas developed recently, which enables methane to be directly cracked under high temperature and catalyst to obtain high-purity hydrogen and solid carbon nanomaterials. This technology can obtain high-quality carbon nanomaterials through chemical vapor deposition, which is a hot spot in the field of hydrogen energy development and nano research in the future. Its main reactions are as follows:

$$CH_4 \xrightarrow[\text{catalyzer}]{\text{high temperature}} C + 2H_2, \text{ Methane cracking reaction} \tag{5}$$

Hydrogen Production from Petroleum

Due to the huge energy utility and economic value of crude oil, high value-added fuel, chemical raw materials, asphalt and other derivative products can be obtained through refining and extraction [19, 20]. Therefore, in order to realize resource conservation, economy and reliability, petroleum hydrogen production technology usually uses crude oil refining by-products to produce hydrogen, specifically including partial oxidation of petroleum coke, naphtha oxidation and partial oxidation of heavy oil to produce hydrogen.

Among them, the process of producing hydrogen from naphtha and heavy oil is similar to that of natural gas. The main process is to obtain hydrogen, carbon monoxide and other multi-component gases after desulphurization and conversion with high-temperature steam. Then it enters the catalyst fixed bed reactor, where the carbon monoxide conversion reaction occurs under the action of high temperature water vapor and catalyst, and the carbon monoxide is converted into hydrogen and carbon dioxide. Finally, the multi-component product was purified, separated and purified to obtain high purity hydrogen. The process of hydrogen production from petroleum coke is similar to that of coal. The main process is partial oxidation with oxygen-rich air and gasification of water coke slurry to generate hydrogen, carbon monoxide, hydrogen sulfide and other hydrogen-rich synthetic gases. After carbon monoxide conversion, low temperature methanol desulfurization, separation and purification, high purity hydrogen and sulfur, carbon dioxide and other by-products are obtained.

3.2 Hydrogen Production from Industrial By-Product Gas

At present, hydrogen production from industrial by-product gas is one of the important sources of hydrogen energy. Its main process principle is to prepare high-purity hydrogen from industrial hydrogen-rich synthesis tail gas, mainly from refining by-product gas, chlor-alkali by-product gas, chemical coke oven gas, etc., as raw materials, combined with separation and purification technology. According to statistics, China produces tens of millions of tons of industrial by-product hydrogen in coal, petroleum, chemical, steel and other fields every year. Industrial by-product gas is rich in resources and cheap in raw materials. If fully combined with multi-component gas separation and purification technology, it will not only help industrial energy conservation and emission reduction, reduce environmental pressure, but also obtain cheap and high-quality high-purity hydrogen and other important chemical raw materials.

The key of hydrogen production process from industrial by-product gas is the separation and purification technology of by-product hydrogen. At present, the main applied separation and purification technologies of industrial by-product hydrogen include pressure swing adsorption (PSA), cryogenic separation and membrane separation. Among them, pressure swing adsorption (PSA) method is to make the adsorbent realize selective adsorption of each component gas under different pressures by periodically changing the pressure of the adsorption bed, and adjust the pressure of the adsorbent to realize gas dissociation and regeneration, and finally purify hydrogen. The cryogenic separation method is to heat the liquid mixture to different temperatures to selectively distill and condense the gas components according to the boiling point difference of the gas components after cooling and liquefying the multi-component gas, which can effectively separate the multi-component gas streams and finally obtain high-purity hydrogen. The membrane separation method is to make multi-component gas pass through a specific molecular membrane (organic membrane or inorganic membrane), and realize gas separation by using the difference of membrane permeability of each component gas. After pressurization, the hydrogen molecule can have better permeability, so it can quickly realize hydrogen separation without phase change, and obtain high-purity hydrogen.

Among the separation and purification technologies of industrial by-product hydrogen, pressure swing adsorption (PSA) is the most widely used in industry because of its mature technology, simple process, low production and operation cost, high purity of separated hydrogen, but the separation process will lose hydrogen and low recovery rate; The cryogenic separation method has large capacity, high hydrogen recovery rate, and can adapt to low hydrogen content gas sources, but high energy consumption and large equipment investment; The membrane separation method has low equipment cost, can achieve phase-free separation and high purity of hydrogen separation, but the molecular membrane and supporting materials have high cost and have not yet been applied in a large scale. With the continuous development of hydrogen production technology from industrial by-product gas, the development of separation and purification technology of industrial by-product hydrogen gradually tends to the joint development and application of multiple processes.

3.3 Hydrogen Production by Pyrolysis at High Temperature

Hydrogen Production from Methanol Reforming

Liquid methanol is an important basic chemical raw material widely used in industry. It has a wide range of sources, high hydrogen storage capacity, easy storage and transportation, and high safety. It is an important raw material for the production of organic chemical products such as formaldehyde, aromatics, olefins, and dimethyl ether. At the same time, with the rapid development of hydrogen energy fuel cell technology, the chemical industry has developed the technology of hydrogen production by methanol cracking and reforming. The main process principle is to directly decompose methanol into carbon monoxide and hydrogen under about 300 °C and catalyst fixed bed reaction, and then produce hydrogen by steam conversion reaction of carbon monoxide and high temperature water vapor, and finally obtain high-purity hydrogen through multi-component gas purification and separation. The process flow is shown in Fig. 5, the general contractor responded as follows:

$$CH_3OH + H_2O \underset{\text{catalyzer}}{\overset{\text{high temperatur}}{\rightleftarrows}} CO_2 + 3H_2, \quad \text{Methanol cracking and reforming reaction} \tag{6}$$

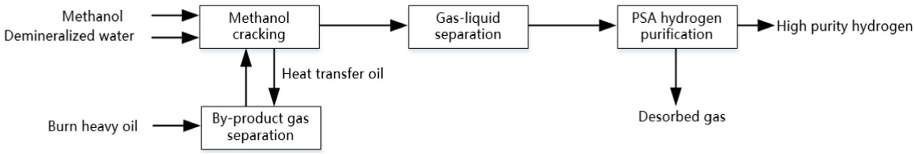


Fig. 5. Process flow of methanol cracking and reforming for hydrogen production.

Hydrogen Production by Ammonia Decomposition

Ammonia (NH3) is a nitrogen-hydrogen compound, with high hydrogen atom mass fraction, easy liquefaction, storage and transportation, high safety, simple process, low production and operation costs, high conversion efficiency, and the decomposition products of ammonia are only hydrogen and nitrogen, without carbon-containing by-products. Therefore, the use of ammonia decomposition hydrogen production technology can achieve zero carbon emissions in the hydrogen production reaction process, high hydrogen purity, and is conducive to improving the electrode activity of hydrogen fuel cells. The technical principle of ammonia decomposition for hydrogen production is the reverse reaction of synthetic ammonia. The main process flow is to make ammonia (or liquid ammonia) directly catalytic cracking at about 800 °C and catalyst fixed bed reaction to obtain 75% hydrogen and 25% nitrogen, and then obtain high-purity hydrogen after gas purification and separation. The main reactions are as follows:

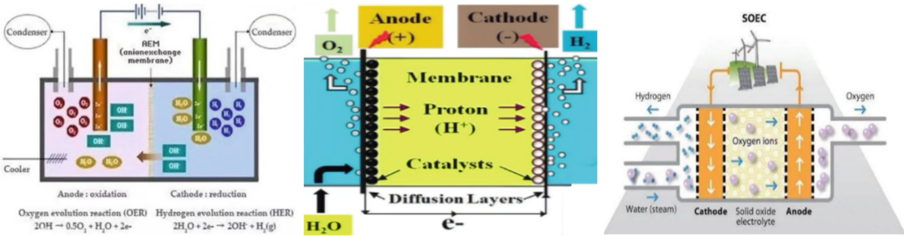
$$2NH_3 \underset{\text{catalyzer}}{\overset{\text{high temperatur}}{\rightleftarrows}} N_2 + 3H_2, \quad \text{Ammonia decomposition reaction} \tag{7}$$

3.4 Hydrogen Production by Electrolysis of Water

At present, with the rapid expansion of the installed scale of renewable energy such as solar photovoltaic power generation, wind power generation and so on in the world, hydrogen production by electrolysis of water after the use of renewable energy power generation can not only solve the outstanding problems such as poor grid stability and difficulty in peak shaving caused by the discontinuous and unstable power generation of renewable energy, but also achieve zero carbon emission in the hydrogen energy preparation process and high environmental protection value. However, due to the high power consumption of hydrogen production from electrolytic water and the high cost of electrochemical reaction devices and materials, the production of hydrogen from electrolytic water only accounts for about 4% of the existing industrial hydrogen production in the world.

The basic principle of electrolytic water hydrogen production technology is to use direct current to ionize water molecules in the electrolytic cell and then obtain hydrogen and oxygen at the cathode and anode respectively. According to the different types of electrolytes, the electrolytic water hydrogen production process can be mainly divided into alkaline water electrolysis (AWE), proton exchange membrane pure water electrolysis (PEM) and solid oxide high-temperature steam electrolysis (SOEC), as shown in Fig. 6.

Among them, the alkaline water electrolysis hydrogen production process is the most mature, the equipment and operating costs are the lowest, and the device is simple and easy to operate. However, because the electrolyte usually uses strong alkaline solutions such as NaOH and KOH with a mass ratio of about 30%, it is easy to cause corrosion of the reaction device and loss of product gas flow, so the purity of hydrogen produced is relatively low, about 99.8% [21, 22]. Proton exchange membrane pure water electrolysis hydrogen production process is relatively mature, the device structure is compact and flexible, the hydrogen purity is high, and can adapt to large current density changes. It is particularly suitable for renewable energy generation and energy storage, but the cost of related materials is high. Therefore, through research to reduce equipment and operating costs, this process will have great application prospects. The solid oxide high-temperature steam electrolysis hydrogen production process has high conversion efficiency and high purity of hydrogen. However, due to the high operating temperature, high requirements on the thermal stability of various components and materials of the device, and high equipment operating costs, the process is still in the research and development demonstration stage.



(a) AWE (b) PEM (c) SOEC

Fig. 6. Hydrogen production process from electrolytic water.

4 Analysis of Key Technologies for Coupled CCUS Low Carbon Hydrogen Production

According to statistics, at present, more than 95% of hydrogen energy in the world comes from fossil energy, mainly from coal and natural gas. Due to the high maturity of fossil energy hydrogen production technology and low operating costs, experts infer that in the future, countries around the world will still use fossil energy hydrogen production as the main source of hydrogen energy. At the same time, CCUS technology is highly integrated with the fossil energy system. Therefore, the development of coupled CCUS technology for hydrogen production from fossil energy will be the key to realize low-carbon hydrogen production technology [23, 24].

In order to ensure global energy security and achieve the "peak carbon dioxide emission and carbon neutrality" target, it is necessary to actively promote the large-scale application of green and low-carbon renewable energy represented by hydrogen energy [25, 26]. At present, hydrogen energy itself can be used to achieve zero carbonization, but its preparation process is often accompanied by a large amount of CO_2 emissions. Therefore, in order to realize the green and low-carbon process of hydrogen energy development and utilization, it is necessary to develop low-carbon hydrogen production technology coupled with CCUS technology. In addition, through the analysis of the above key technologies of hydrogen production, it can be found that the current process of hydrogen production from fossil energy is the most widely used, mature, economic and reliable, and suitable for large-scale industrial hydrogen production. However, the process of hydrogen production has a large carbon emission, a large amount of gas impurities, and does not meet the development needs of low carbon and environmental protection. Therefore, with the implementation and promotion of carbon emission trading and supporting policies of the "peak carbon dioxide emission and carbon neutrality" target, hydrogen production technology from fossil energy must be coupled with CCUS technology to achieve effective carbon dioxide emission reduction. The development of low-carbon hydrogen production key technology coupled with CCUS technology is a key entry point to realize planned clean and green hydrogen utilization in the future.

By comparing the raw material price, technical maturity and other characteristics of various key technologies for hydrogen production, the fossil energy hydrogen production technology, mainly represented by coal hydrogen production, is relatively mature and has large-scale application scenarios, and the economic and cost advantages of system operation are prominent; Although the technology of hydrogen production from industrial by-product gas is relatively mature, its scale is limited. The overall technology maturity of hydrogen production from renewable energy electrolysis water and biomass is low, and it is difficult to promote large-scale commercialization in the short term. Although the overall cost of hydrogen production from fossil energy will increase after coupling the CCUS system, the economic benefits can be improved through the resource utilization after carbon dioxide capture, which can be used to synthesize high-purity carbon monoxide, process food additives, oil displacement and fracturing in oil fields, improve agricultural production, improve saline-alkali water quality, synthesize biodegradable materials, etc., thus promoting the large-scale application of CCUS technology and hydrogen energy in the short term.

Currently, hydrogen can be extracted from fossil fuels, biomass, water, or their mixtures. Natural gas is the main raw material for hydrogen production, accounting for approximately 47% of global hydrogen production annually. Secondly, hydrogen production from coal accounts for 13% of the global total hydrogen production, and the rest of hydrogen comes from biomass hydrogen production and electrolysis of water to produce hydrogen. For most regions of the world, fossil fuel hydrogen production will still have a certain cost advantage in the short term (before 2030).

Hydrogen production from Coal gasification is to convert coal into synthetic gas under certain temperature and pressure, and then extract high-purity hydrogen through Water gas shift separation. The Technology roadmap of hydrogen production from coal is mature and efficient, which can be stably and massively produced, and is the lowest cost hydrogen production method at present. According to the data of China Hydrogen Energy Alliance, taking the 100000 m^3/h (Standard state) coal to hydrogen project as an example, raw coal is the most consumed raw material in coal to hydrogen production, accounting for about 50% of the total production cost. When the price of raw coal is \$86/t, the cost of hydrogen production is approximately \$1.2/kg. At present, the cost of CCUS in China (calculated in CO_2) is approximately \$50 ~ 57/t. Adding the average ratio of CO_2 generated per unit of hydrogen in the coal to hydrogen production route, coupled with CCUS, the cost of coal to hydrogen production will increase by approximately \$1.7/kg. Therefore, it is necessary to further develop this technology in the future, promote the reduction of energy consumption and costs, and expand the utilization channels of CO_2. Natural gas is one of the main hydrogen production raw materials in developed countries in Europe and America. Among natural gas hydrogen production technologies, Steam reforming technology is relatively mature. Taking the scale of the 100000 m^3/h natural gas hydrogen production project as an example, natural gas is the most important raw material for natural gas hydrogen production, accounting for approximately 82% of the total cost of hydrogen production. When the price of raw natural gas is \$0.23/(m^3/h), the cost of hydrogen production from natural gas is approximately \$1.11/kg. The cost of raw materials in different countries and regions accounts for different costs of hydrogen production. The prices of natural gas in the Middle East, Russia, and North America are relatively low, and these countries only account for 45% to 75% of the total cost of hydrogen production. Therefore, the total cost is also relatively low. In addition to directly producing hydrogen products from fossil fuels, some chemical plants also produce a large amount of by-product hydrogen during this process. About 50% of it is used for furnace combustion, and the remaining industrial by-product gas is recovered and purified through pressure swing adsorption. Industrial by-product gas is one of the main hydrogen sources in China for a certain period of time, and its cost mainly depends on the price of raw gas. Taking the 10000 m^3/h scale coke oven gas hydrogen production as an example, when the coke oven gas is \$0.07/(m^3/h), the cost of hydrogen production is approximately \$2/kg [27].

The carbon emissions footprint of different hydrogen production processes varies significantly. The cost of coal based hydrogen production is relatively low, with abundant sources of raw materials, but it also faces the problem of high carbon emissions. The carbon emissions from the coal hydrogen production process (calculated as CO_2 emissions per kilogram of hydrogen, the same below) range from 25.00 to 35.00 kg. In order to control carbon emissions, coal based hydrogen production must be combined with CCUS technology. Without CCUS technology, the carbon emission of natural gas hydrogen production process is lower than that of coal hydrogen production process, about 4.95–15.00 kg. The carbon emission of electrolysis of water to produce hydrogen process mainly depends on the carbon emission of power generation process. In order to reduce the carbon emissions of the electrolysis hydrogen production process compared to the natural gas hydrogen production process without CCUS, the carbon emissions of the power generation process should be less than 185 g CO_2/kW. For hydrogen production from by-product gas, the typical coke oven gas hydrogen production process is relatively simple, and the pressure swing adsorption method can be approximated as purifying hydrogen. Therefore, the carbon emissions of the by-product gas hydrogen production process are much lower than those of coal hydrogen production and natural gas hydrogen production. Considering the superimposed mass distribution of by-product hydrogen process, its carbon emissions are approximately 1.75–5.00 kg. The carbon emissions from hydrogen production from coke oven gas using typical domestic routes and processes are approximately 2.00 kg. See Table 1 for comparison of hydrogen production costs and corresponding carbon emissions of different Technology roadmap in China. From Table 1, it can be seen that fuel prices are the largest single component of the cost of producing hydrogen from coal and natural gas. Therefore, the cost of hydrogen is greatly influenced by the prices of electricity, coal, and natural gas. In the next decade, coal based hydrogen production is likely to be the cheapest source of pure hydrogen in most regions of China, with a cost in the range of \$1.19/kg. Meanwhile, the coal hydrogen production process coupled with CCUS will be the lowest source of low-carbon hydrogen production in many places. The application of CCUS will increase the production of hydrogen. If CO_2 is injected underground for geological storage, the cost will be \$1.16–1.76/kg. From the perspective of short and medium term hydrogen energy industry planning, China is rich in coal resources. We should promote CCUS coupled coal hydrogen production technology, and gradually realize the low-carbon development of China's coal hydrogen production under the premise of low cost. In the long term, China should gradually convert coal hydrogen production to renewable energy hydrogen production, and promote the low-cost development of "green hydrogen".

Table 1. Cost comparison of different hydrogen production processes.

Hydrogen production process	Hydrogen production cost/(dollars/kg)	Carbon emission/kg	Characteristic
Hydrogen production from coal	1.2	22.00 ~ 35.00	Large scale, low cost, highest CO_2 emissions, and equipment prone to poisoning
Hydrogen production from coal + CCUS	2.9	13.99	Large scale, equipment prone to poisoning
Hydrogen production from natural gas	1.11	4.95 ~ 15.00	High energy conversion and high raw material cost
Hydrogen production from natural gas + CCUS	1.97	—	Low carbon emissions and high costs
Hydrogen production by electrolysis of water	2.64 ~ 4.12	0.10 ~ 43.78	High cost, environmentally friendly
Hydrogen production from industrial by-products	2.0 ~ 2.5	1.75 ~ 5.00	Low capital investment and small scale

As one of the current cutting-edge coal gasification technologies, underground coal gasification (UCG) hydrogen production process is to form a natural gasifier by burning underground coal in the early stage, and put the coal into the underground high-temperature gasifier through the injection well drilled into the coal seam, where the coal and the gasifier (oxygen enriched air, water vapor) undergo a series of physicochemical reactions such as drying, oxidation, reduction, methanation reaction, pyrolysis, etc. Thus, a multi-component product mainly composed of carbon monoxide, hydrogen, and methane is generated and extracted through production wells. Afterwards, high-purity hydrogen gas is obtained through carbon monoxide transformation, purification, separation, and purification. Compared with coal coking, Coal gasification reaction conditions are easy to meet and the process is easy to control. Therefore, Coal gasification hydrogen production technology is mainly used to prepare hydrogen from coal in industrial production, and the main reactions are as follows (process flow is shown in Fig. 7):

$$C + \frac{1}{2}O_2 \xrightarrow{\text{high temperature}} CO\text{, Partial oxidation reaction} \quad (8)$$

$$C + O_2 \xrightarrow{\text{light}} CO_2\text{, Complete oxidation reaction} \quad (9)$$

$$C + H_2O \xrightarrow{\text{high temperature}} CO + H_2\text{, Steam reforming reaction} \quad (10)$$

$$CO + H_2O \xrightarrow[\text{catalyzer}]{\text{high temperature}} CO_2 + H_2, \text{ Carbon monoxide shift reaction} \quad (11)$$

$$CO_2 + 4H_2 \xrightarrow[\text{high pressure}]{\text{high temperature}} CH_4 + 2H_2O, \text{ Methanation reaction} \quad (12)$$

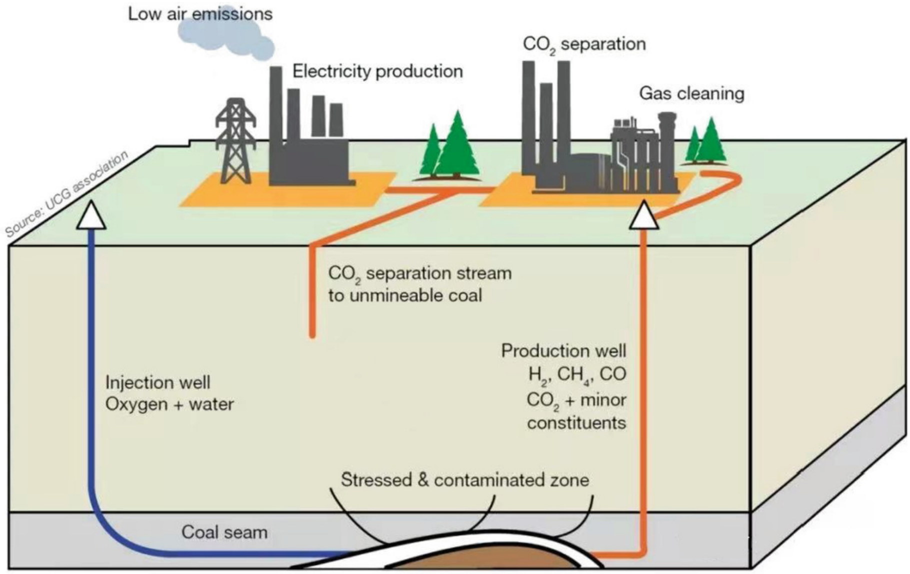


Fig. 7. UCG hydrogen production process flow diagram.

From the above reaction principles of underground coal gasification for gas production, it can be seen that hydrogen gas in the underground coal gasification process mainly comes from the decomposition of water vapor, the conversion reaction of dry distillation gas and CO. The chemical decomposition of water vapor is mainly the reaction of high-temperature carbon and water vapor to generate CO and H_2. The reaction speed of chemical decomposition of water vapor increases with the increase of temperature, but decreases with the increase of pressure. The temperature of the reduction zone of underground gasifier is generally between 600–1000 °C, its length is 1.5–2 times of the oxidation zone, and the pressure is between 0.01–0.2 MPa. Therefore, the reduction zone is conducive to the increase of the concentration of products. The CO generated in the water gas shift reaction will react with water vapor to further generate H_2. This reaction can occur at temperatures above 400 °C. The rate of chemical decomposition is equivalent to that of water vapor at 900 °C, and it is very fast at temperatures above 1480 °C. In the underground gasifier, it can be considered that the CO shift reaction can reach the Thermodynamic equilibrium state, but the actual degree of reaching the equilibrium is related to the temperature, steam decomposition rate and the length of the gasification channel, as well as the reactivity and catalytic activity of the gasified coal seam.

From the perspective of resource endowment and low-carbon environmental protection, the coupling of coal hydrogen production and CCUS technology will be promoted and applied globally as an emerging integrated technology for a long period of time in the future. Among them, underground coal gasification (UCG) hydrogen production coupled with CCUS technology is a clean coal low-carbon hydrogen production technology vigorously promoted in recent years in China. The process principle is to drill injection wells and gas production wells to the depth of underground coal seams, complete well construction, form a gasification chamber, and then inject gasification agent to conduct controlled combustion with underground coal. After a series of complex thermochemistry reactions such as pyrolysis and oxidation, steam conversion, carbon monoxide conversion, etc. Hydrogen High purity hydrogen gas is obtained through the recovery and purification of multi-component gases such as carbon monoxide, methane, and carbon dioxide. Underground coal gasification can avoid many environmental problems caused by surface mine and underground coal mining. The ash, oxide and radiation after underground gasification are all left underground, avoiding the accumulation of ground coal slag, reducing the space of the combustion area, and avoiding the surface subsidence. The sulfur in coal is basically converted into hydrogen sulfide, which can be centrally purified on the ground. After purification, the coal can be separated to obtain cheap clean fuel hydrogen. Moreover, using CCUS technology to capture carbon dioxide and utilize it as a resource for geological storage has significant advantages such as high CO_2 concentration, low capture cost, and short transportation distance in the recovery of production components. The composition of the process flow system is shown in Fig. 8. At present, the development potential of CCUS low carbon hydrogen production technology coupled with underground coal gasification is huge, but further research on ground centralized transportation and treatment process is needed to improve the gas

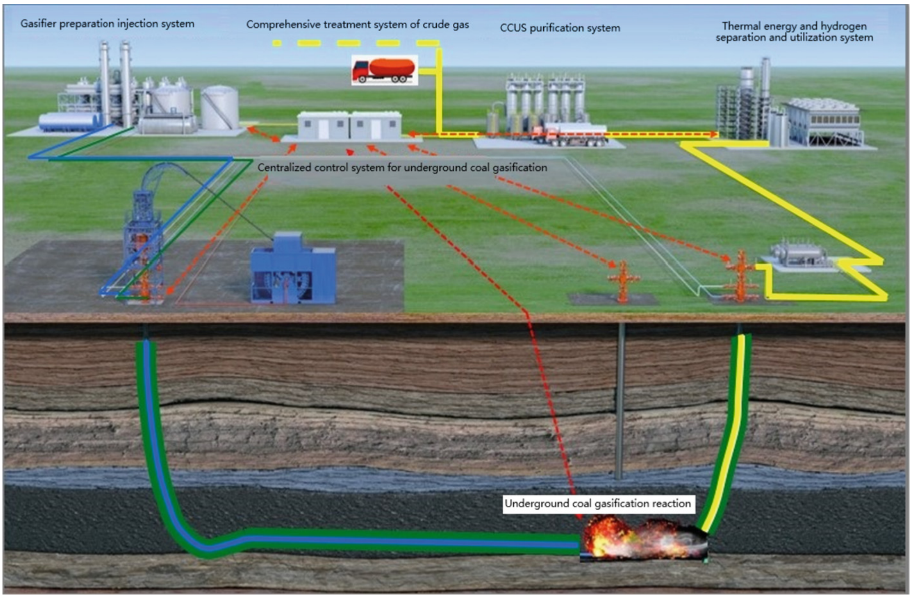


Fig. 8. UCG coupled CCUS hydrogen production process diagram.

production rate of underground coal gasification. In addition, the development of integrated energy utilization system of CCUS technology coupled with Coal gasification hydrogen production, hydrogen turbine combined cycle power generation and fuel cell power generation will be the development direction of clean utilization of hydrogen energy in the future.

Furthermore, the carbon dioxide captured by CCUS technology coupled with low-carbon hydrogen can generate a large number of high-value-added chemical raw materials and synthetic energy, which can not only help reduce carbon dioxide emissions, but also realize large-scale resource utilization of carbon dioxide. Take the carbon dioxide hydrogenation catalytic conversion process to methanol (as shown in Fig. 9) [28] as an example. The carbon dioxide catalytic conversion pathway includes thermal catalysis, photocatalysis, electrocatalysis and biocatalysis, etc. The technical core is to develop efficient carbon dioxide conversion catalysts. Chemical utilization can not only reduce carbon dioxide emissions, but also create additional income, which plays an important role in the transformation and upgrading of traditional industries. In recent years, various carbon dioxide chemical utilization technologies have made great progress, with good product added value and economic benefits, and bright prospects for commercial application. In the future, based on the integrated utilization technology of carbon dioxide capture and hydrogen coupled conversion of methane, methanol and other chemical products, it will have great potential for promoting the development of CCUS coupled low-carbon hydrogen production process and ensuring the low-carbon sustainable development of global energy.

In addition, CCUS coupled with fossil energy hydrogen production technology and renewable energy can also help develop new energy storage methods [29, 30]. The CCUS technology is used to capture carbon dioxide from the process of hydrogen production from fossil energy and produce hydrogen by electrolysis of water from renewable energy. The reaction of carbon dioxide and hydrogen can be made into methanol, and the fluctuating renewable energy can be converted into chemical energy storage. In January 2020, the world's first 1000-ton solar fuel synthesis demonstration project was successfully tested in the Green Chemical Park of Lanzhou New Area, and the first step was taken in the industrial production of converting renewable energy such as solar energy into liquid fuel.

At present, as an important technological link in the entire industrial chain of hydrogen energy utilization, hydrogen energy storage and transportation technology can be divided into high-pressure gas hydrogen storage and transportation, low-temperature liquid hydrogen storage and transportation, solid material hydrogen storage and transportation, organic liquid hydrogen storage and transportation, and natural gas hydrogen blending pipeline transportation. Through comparative analysis, it can be seen that high-pressure gas hydrogen storage and transportation has low operating costs, relatively low energy consumption, and fast response speed for hydrogen charging and discharging. It is suitable for short distances and scattered user situations, and is currently the most commonly used storage and transportation method. However, it has high pressure requirements for equipment, low hydrogen storage density per unit volume, and low safety. Low temperature liquid hydrogen storage and transportation has high energy density (about

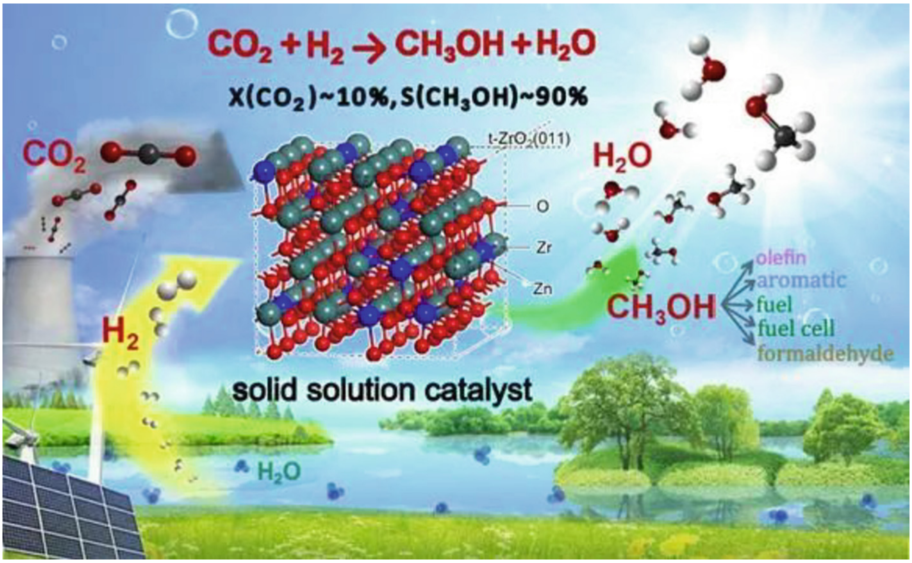


Fig. 9. Catalytic hydrogenation of carbon dioxide to methanol.

845 times the density of gaseous hydrogen), high transportation efficiency, and is suitable for medium and long distance transportation. It is mainly used as the fuel of aviation carrier rocket propellant. It has high requirements for vacuum insulation, vibration damping, shock resistance, leakage prevention performance of hydrogen storage devices, and deep cold liquefaction has large consumption and high cost. Solid hydrogen and organic liquid hydrogen storage and transportation are generally relatively safe, efficient, with high hydrogen storage density and good recyclability. However, there are high requirements for the performance of hydrogen storage materials, which is an important research direction for future hydrogen energy storage and transportation. The transportation cost of natural gas mixed with hydrogen through pipelines is low, and energy consumption is low. It can achieve continuous, large-scale, and long-distance transportation of hydrogen energy, which is an inevitable trend for the large-scale utilization of hydrogen energy in the future.

In the near and medium term, hydrogen production from fossil fuels is an inevitable choice to promote the rapid and mature implementation of hydrogen energy technology and supporting systems. However, since hydrogen production from fossil energy (coal or natural gas) will be accompanied by a large amount of carbon dioxide, the coupling of CCUS and fossil fuel hydrogen production process is an important and necessary technology to achieve low-carbon hydrogen production [31, 32]. At the same time, the conversion of carbon dioxide into chemical chemicals or fuels by hydrogen produced through green and low-carbon ways will help solve the problems of CO_2 emissions, over-dependence on fossil fuels and storage of renewable energy.

5 Conclusion

As a renewable green energy, hydrogen energy has advantages such as clean, zero carbon, wide sources, recyclable and various application forms, and can act as a high-quality energy storage carrier of fluctuating renewable energy. It can be converted into important chemical products through catalytic reaction with CO_2, which is of great significance for promoting the transformation of energy structure and the deep emission reduction and comprehensive utilization of carbon dioxide.

At present, more than 95% of the world's hydrogen energy comes from fossil energy. Due to the high maturity of hydrogen production technology from fossil energy, low operating cost and suitable for large-scale industrial application, experts conclude that countries around the world will continue to use fossil energy as the main source of hydrogen energy in the future for a long time [33, 34]. However, the process of hydrogen production from fossil energy will release a large amount of CO_2, which does not meet the requirements of low-carbon and environmental protection development. Therefore, in order to realize the green and low-carbon development and utilization of hydrogen energy, it is necessary to vigorously develop the key low-carbon hydrogen production technology of fossil energy coupled with CCUS technology. At the same time, efforts should be made to break through the bottleneck of high energy consumption and high equipment operation cost of hydrogen production by electrolytic water, so as to promote the low-carbon and sustainable development of hydrogen energy and help countries realize the "peak carbon dioxide emission and carbon neutrality" target as soon as possible. At present, the overall development of the key technology of hydrogen production coupled with CCUS low carbon hydrogen production from fossil energy sources is still in its infancy. The main difficulties are process coupling and operation cost control. It is suggested to take clean coal low carbon hydrogen production technology of UCG coupled CCUS, which has been vigorously promoted in recent years, as a starting point. We will develop low-carbon hydrogen production coupled with carbon dioxide to produce high-value-added derivative products, and jointly promote the rapid development of key technologies for large-scale low-carbon hydrogen production around the world.

This study analyzed and sorted out the global development status of CCUS coupling hydrogen production technology in recent years, as well as the main factors affecting the global deployment of CCUS technology. And focused on analyzing the UCG coupled CCUS hydrogen production process and hydrogen energy storage and transportation technology, and proposed development suggestions. Through the comparative analysis of the costs of different hydrogen production paths, it is found that, compared with renewable energy hydrogen production, the CCUS coupled low-carbon hydrogen production technology has obvious cost competitive advantages in promoting the low-carbon transformation of the energy system and implementing the energy revolution. The development of coupled CCUS technology for hydrogen production from fossil fuels is an important way to achieve low-carbon hydrogen production from fossil fuels. At the same time, coupling low-carbon hydrogen production with CO_2 to prepare high value-added chemicals is beneficial for CO_2 emission reduction and carbon resource utilization.

Acknowledgments. The project is supported by National Key R&D Program of China (Number: 2021YFB3401400); Scientific research and technology development project of CNPC (Number: 2019E-2504).

Fund Projects:. National Key R&D Program of China (Number: 2021YFB3401400); Scientific research and technology development project of CNPC (Number: 2019E-2504).

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