Development of Hydrogen Production Technology Initiative to Create Decarbonized World

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Against the backdrop of accelerating energy transition across the world, there is an urgent need to achieve carbon neutrality in Gas Turbine Combined Cycle and steam power generation systems, which are the main products of Mitsubishi Heavy Industries, Ltd. Decarbonization of these thermal power generation systems necessitates developing not only decarbonization technologies for power generation facilities but also technologies to produce hydrogen, which is considered to be one of the fuels for such decarbonized power generation, economically and in large quantities.

MHI is developing both power generation facilities and hydrogen production systems, and the previous report (1) focused on hydrogen production technology and introduced the features and development status of the technology. This report presents the progress made since then.

1. Introduction

Solving global warming problems is critical to humanity. In October 2020, along with the growing momentum of international climate action such as the Conference of Parties (COP) on climate change, the Japanese government declared its intention of achieving "carbon neutrality" by reducing greenhouse gas emissions to net zero by 2050. To achieve such carbon neutrality, it is indispensable to substantially expand the use of renewable energy. Simultaneously, it is also important to maintain economic efficiency and stable energy supply. Mitsubishi Heavy Industries, Ltd. (hereinafter referred to as MHI) aims to achieve a carbon-neutral society in a realistic and speedy way, while minimizing social costs by promoting energy transition of existing thermal power generation facilities.

The expansion of the use of renewable energy requires the introduction of energy storage technology to deal with fluctuations in output. In general, lithium batteries are advantageous for storing energy for a short period of time, but for a relatively long period of time, such as days or weeks, it is advantageous to convert the energy into chemical energy such as hydrogen, which can be stored and transported. Therefore, hydrogen production technology will be an important energy storage technology.

The hydrogen production, an energy conversion technology, can be applied to energy storage for power generation and developed for the production of liquid synthetic fuels from carbon dioxide as well. In this manner, hydrogen can be converted to a liquid fuel to improve its transportability, so that it has the potential to promote carbon neutrality throughout society by being applied to the field of transportation, such as aircraft and ships. **Figure 1** shows an example of a value chain with hydrogen as a key.

Since the 1980s, MHI has been engaged in developing products based on chemical energy conversion technologies, such as Solid Oxide Fuel Cell (hereinafter referred to as SOFC), Polymer Electrolyte Fuel Cell (PEFC), hydrogen production by water electrolysis using a Proton Exchange Membrane (hereinafter referred to as PEM), and production of carbon nanotubes using a fluidized

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bed reactor. Taking advantage of these accumulated technologies, MHI has resumed development of hydrogen production systems. This paper presents the progress in technological development for producing hydrogen and synthetic fuel, both of which are indispensable for achieving a decarbonized society.

Figure 1 Hydrogen and ammonia / CCUS value chains

2. Summary of MHI's hydrogen production technology

Having declared "MISSION NET ZERO," MHI Group intends to achieve carbon neutrality Net Zero CO_2 emissions from the group's production activities and the entire value chain by 2040. MHI Group also aims to offer products and technologies that can make it viable for customers to achieve carbon neutrality by 2050. MHI Group's major undertakings include the energy transition for low-carbonization and decarbonization of businesses/products, and the expansion of carbon capture utilization and storage (hereinafter referred to as CCUS), including CO_2 capture, contributing to creating a carbon-neutral society.

Figure 2 gives the background of hydrogen and ammonia utilization. As mentioned earlier, the need to introduce energy storage technologies and the strengths of each technology are as follows. Lithium batteries are advantageous for short-time storage, while conversion to chemical energy such as hydrogen is advantageous and is necessary for storing energy for a relatively long period of time such as days and weeks. The right side of Figure 2 shows the regional characteristics of renewable energy resources. It is expected that the use of renewable energy will become more common across many regions of the world and that hydrogen products produced through water electrolysis by using surplus renewable electricity will become widely used. On the other hand, in regions that are not rich in renewable resources such as Japan and South Korea, the application of ammonia with high transportation efficiency will take precedence. There are also high expectations for turquoise hydrogen, which can be produced using existing natural gas infrastructure. Specifically, the production process is the pyrolysis of natural gas and is characterized by the by-product of solid carbon. As versatile as it may be, the term decarbonization can pertain to different technologies depending on the needs of each region, whose verification and social implementation are a matter of urgency.

Inexpensive hydrogen is needed to cut the social costs incurred by growing out of fossil fuels. As almost all of the cost of electrolytic hydrogen production is attributed to electricity, high-efficiency energy conversion technology is required. Moreover, many of the hydrogen applications involve pressures as high as several MPa. The power used for hydrogen compression considerably decreases the overall system efficiency. Because, generally, energy consumption can be reduced for liquid pressurization more than gas compression, it is desirable to have equipment that can electrolyze high-pressure water or steam.

To first focus on the utilization of hydrogen for power generation, MHI's Energy Systems Domain is working on the development of three types of hydrogen production technologies: high-pressure, high-efficiency and large-capacity Solid Oxide Electrolysis Cells (hereinafter referred to as SOEC), Anion Exchange Membrane (hereinafter referred to as AEM) water electrolysis, and production of turquoise hydrogen by methane pyrolysis. Synthetic fuel production technologies in which these electrolyzers are employed are also in development. The lower right of Figure 2 shows the technological development road map for decarbonized power generation. These core technologies are being tested comprehensively for long-term demonstration at Takasago Hydrogen Park [(2)] on the premises of MHI's Takasago District. On the other hand, Nagasaki Carbon Neutral Park [(3)] located at MHI's Nagasaki District is responsible for the development of core technologies.

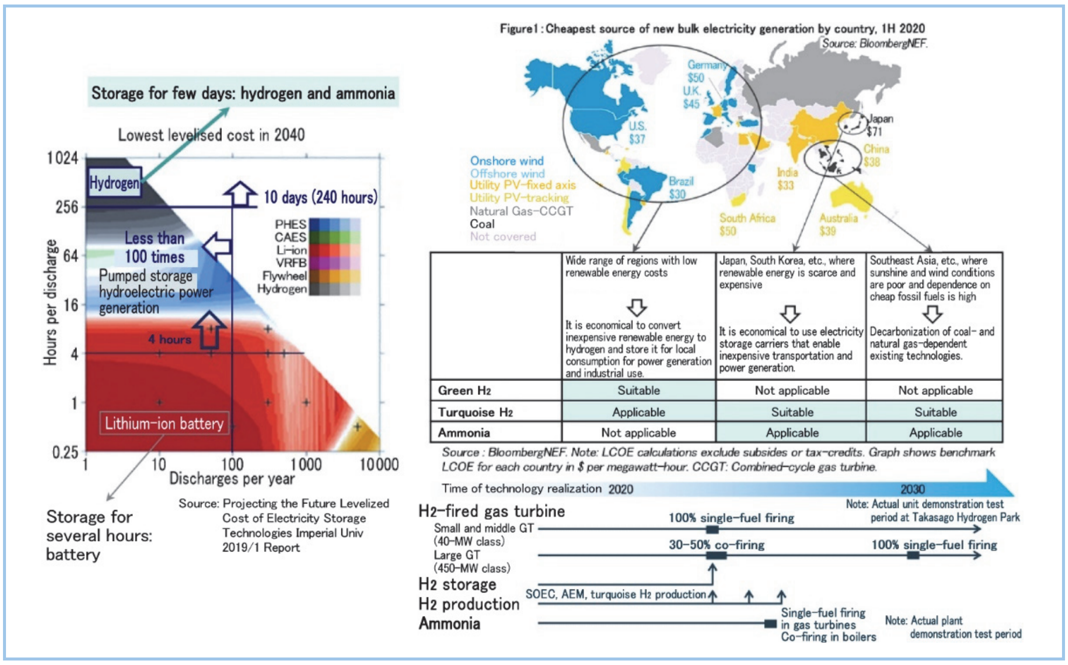

Figure 2 Background for hydrogen and ammonia utilization and technological development road map for decarbonization

3. SOEC development

SOEC is a process which produces hydrogen through electrolysis of high-temperature steam. This process uses less electricity than water electrolysis which means high efficiency (**Figure 3**). MHI applies a tubular-type SOEC cell stack (see the previous reports[(1), (4)] for details) of its own development.

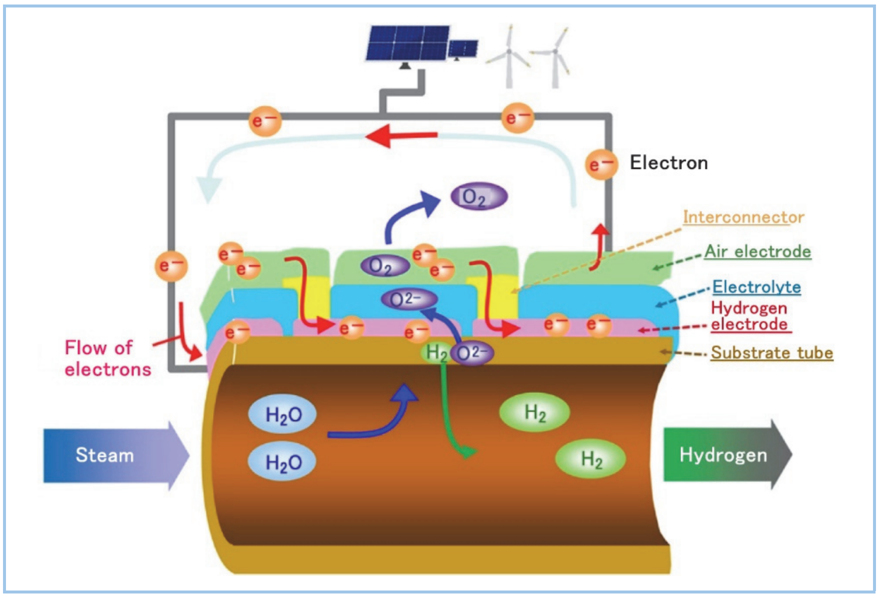

Figure 3 Operating principle of SOEC

As for the development status of core technologies for SOEC, MHI is aiming to increase hydrogen production with the cell by increasing the current flowing through the cell as presented in the previous report [(4)]. Through long-term durability tests of single cell stacks and electrolysis tests on single cartridge, the prospect of operating at higher currents compared to SOFC is shown (**Figure 4**).

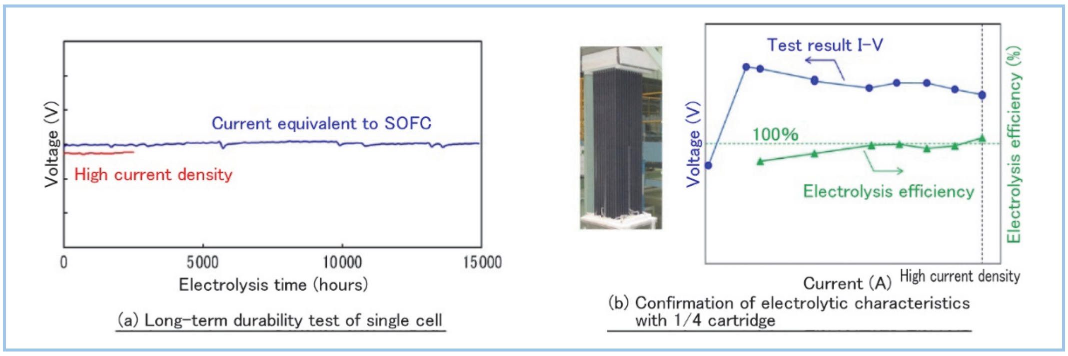

Figure 4　Element test results

Prior to configurate the system of high-current SOEC, MHI has verified the low-current SOEC module operated as 400-kW-class demonstration system at Takasago Hydrogen Park. The system configuration of this system is simple since the main purpose of this facility is to verify the module operation. MHI conducted a demonstration operation of the module which combined 4 cartridges for about 3,000 hours. Based on the verification results, MHI is seeing the prospect of establishing a high-current SOEC module (**Figure 5**).

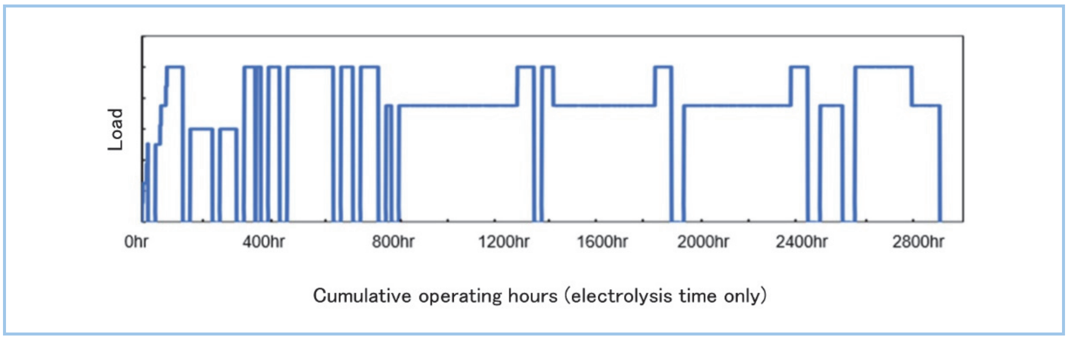

Figure 5　Operation results of 400-kW demonstration facility

Based on these results of the SOEC core technology development, MHI is currently studying a MW-class SOEC system configuration that can economically realize highly efficient operation of SOEC as a system.

As a highly efficient system that utilizes the characteristics of SOEC, MHI is studying a configuration characterized by "steam self-supplied system" (**Figure 6**), in which steam is generated using Joule heat generated during electrolysis, which is to say that hydrogen can be produced by just supplying water and electricity. Specifically, "steam self-supplied system" is possible by generating steam using the heat of hydrogen generated by SOEC and exhaust air heated by Joule heat during electrolysis.

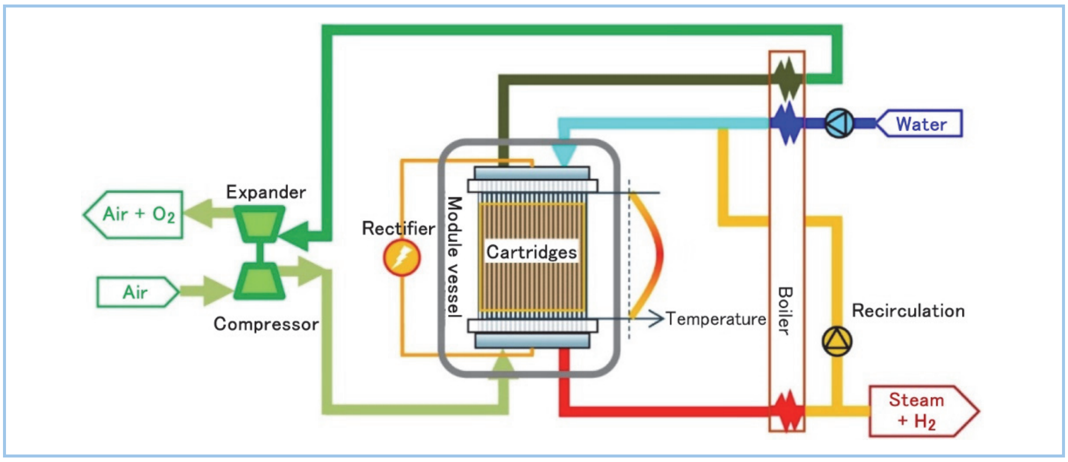

Figure 6　Steam self-sufficient SOEC system

In addition, a configuration that can utilize external steam such as factory waste heat (**Figure 7**) is also being studied. If steam can be supplied from existing facilities, a simple SOEC system can be made by effectively utilizing the steam.

Although the air supply system has a compressor, MHI aims to achieve a highly efficient system with an overall efficiency of more than 90%-HHV, including the efficiency improvement by recovering the exhaust air energy with a turbine. Since hydrogen is expected to be used at high pressure, MHI is aiming for a system in which the operating pressure of SOEC will be higher in the future, and even in such a case, the system will minimize energy losses. Finally, MHI aims to apply the system to a plant of several hundred MW class (100,000 Nm^3/h of hydrogen-class).

MHI will continue to develop these technologies and proceed with the SOEC development as shown in the roadmap in **Figure 8**.

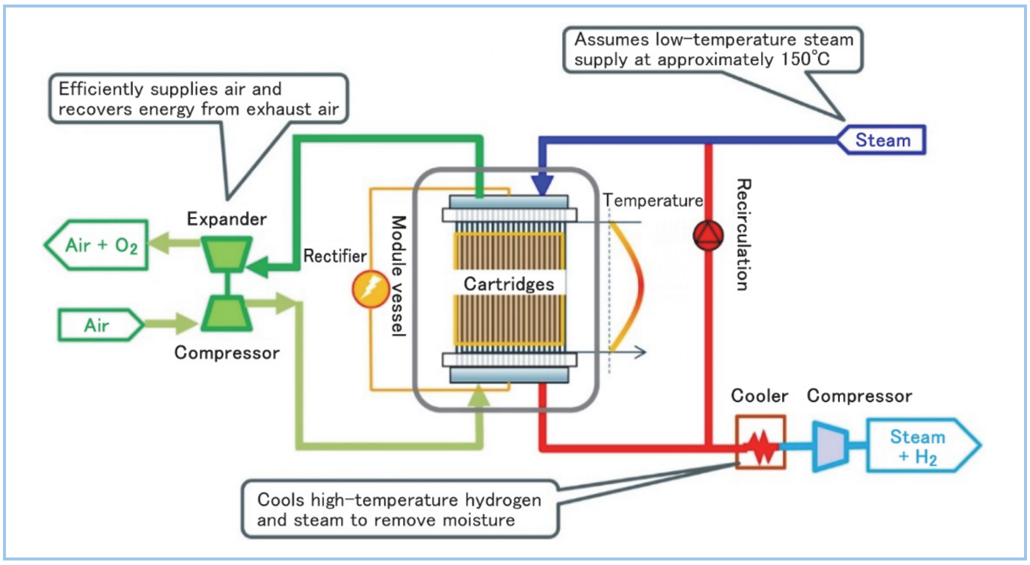

Figure 7 Configuration of SOEC system using external steam

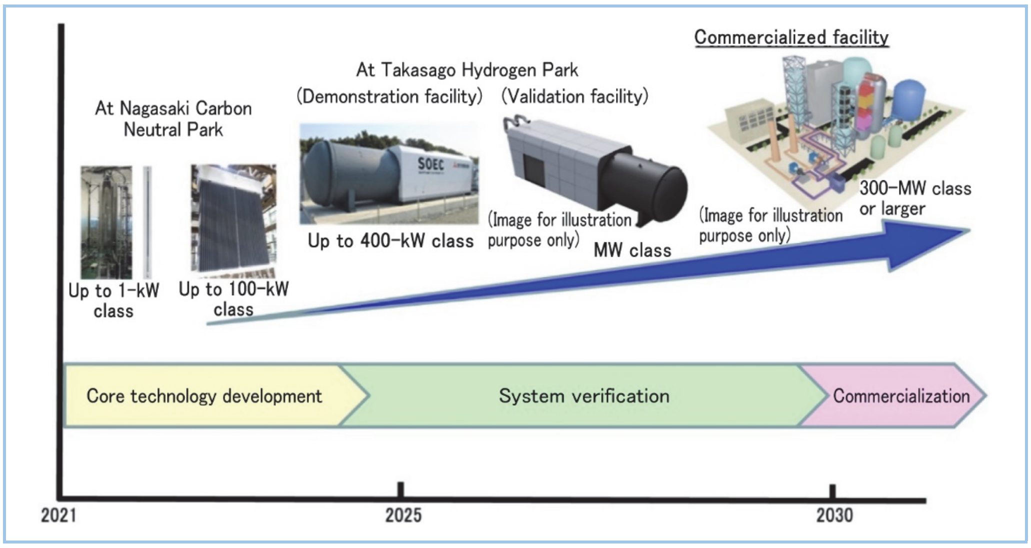

Figure 8 Road map for SOEC development

4. Current status in development of AEM water electrolysis

AEM water electrolysis, which is similar to PEM water electrolysis using hydrogen ion-conducting membranes, allows to downsize the electrolyzer. This technology is also expected to reduce costs due to the alkaline environment, which allows for the use of less expensive stainless steel materials. MHI has been developing AEM water electrolysis technology, and this chapter reports on the development status and findings to date.

MHI is continuously working on understanding the initial characteristics and durability of a small cell with an electrode area of several tens of cm^2; prototyping and evaluating a large cell stack of several hundreds of cm^2; determine an appropriate manufacturing method for stack materials and assembly; and optimizing the system configuration and operating conditions using a kW-class test facility.

The results of prototyping and evaluation of the high-pressure small cell shown in **Figure 9** confirmed that the IV characteristics are equivalent to those in the normal pressure test.

With regard to durability, verification of a single cell's voltage behavior in a long-term test is being conducted, and MHI plans to disassemble the cell after the test to elucidate the factors and mechanisms that contribute to changes in the voltage behavior.

MHI will continue to evaluate the durability of the membrane through single cell and long-term tests (**Figure 10**).

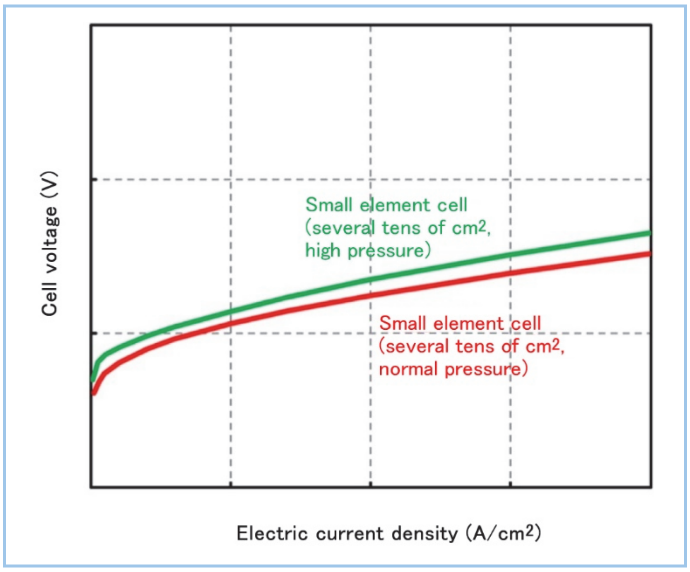

Figure 9 Evaluating results of prototyping high-pressure cell stack

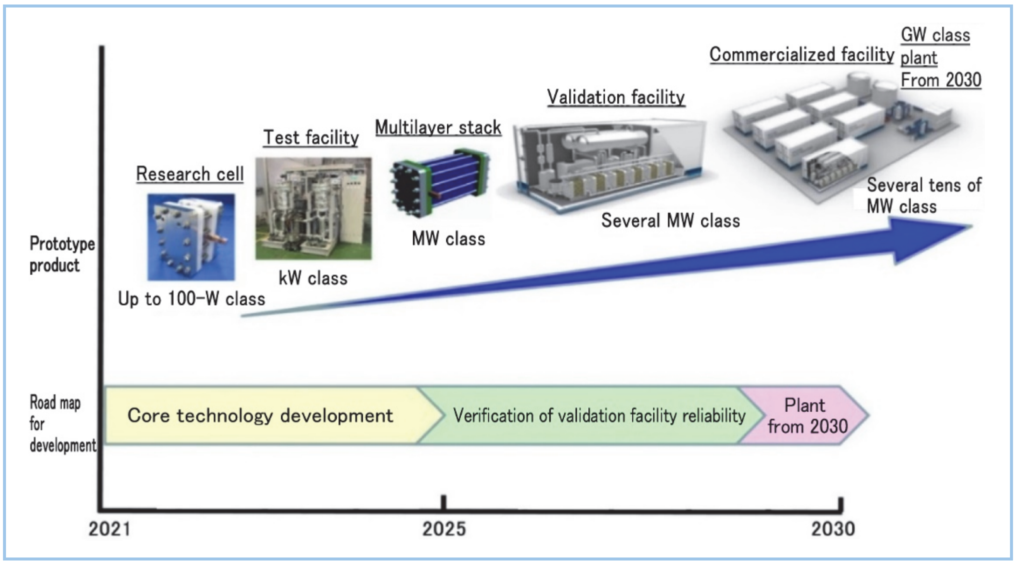

Figure 10 Road map for AEM water electrolysis

5. Current status in development of turquoise hydrogen (methane pyrolysis)

Turquoise hydrogen production by methane pyrolysis is a technology to decompose natural gas into solid carbon and hydrogen at high temperatures. Conventionally, this method has been used to produce industrial carbon materials such as carbon black. By focusing on the co-produced hydrogen, MHI identified the reaction mechanism to enhance hydrogen production efficiency.

Figure 11 illustrates the turquoise hydrogen production technology. The natural gas infrastructure has already been established. A turquoise hydrogen production plant will be installed either between the natural gas infrastructure supply line and the consumer, or upstream of the consumer's consumption equipment. This installation aims to achieve decarbonization. Taking a natural gas-fired power plant (Gas Turbine Combined Cycle, hereinafter referred to as GTCC) as an example, upgrading to hydrogen firing can be done if the main unit gas turbine's combustor is adapted to take hydrogen. Moreover, the solid by-product carbon is easier to fix or store than gaseous CO_2 at normal temperatures and pressures. When combined with turquoise hydrogen, existing thermal power plants can substantially reduce CO_2 emissions, eventually achieving decarbonization, meaning power generation without CO_2 emissions.

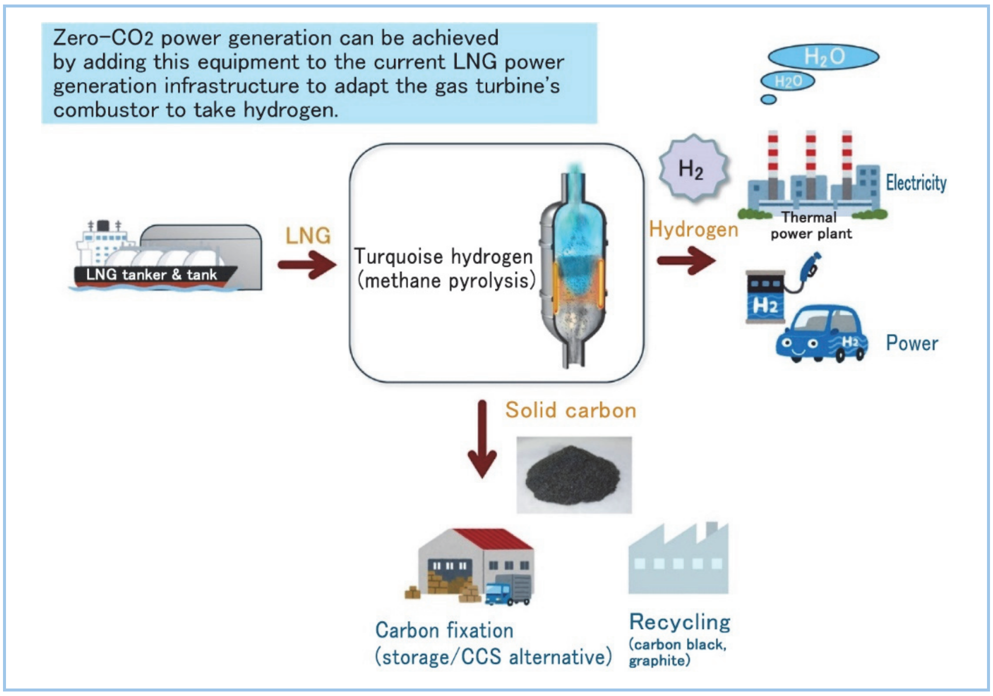

Figure 11 Overview of turquoise hydrogen production technology

Having selected a fluidized bed as the reactor for methane pyrolysis, MHI is examining how the reaction progresses and the screening for appropriate conditions using element test equipment. **Figure 12** shows the batch-type fluidized bed test equipment and the typical test results. In this equipment, a catalyst is placed in the reaction tube. While allowing methane to pass through, the reaction tube is heated by the heater to let methane pyrolysis occur.

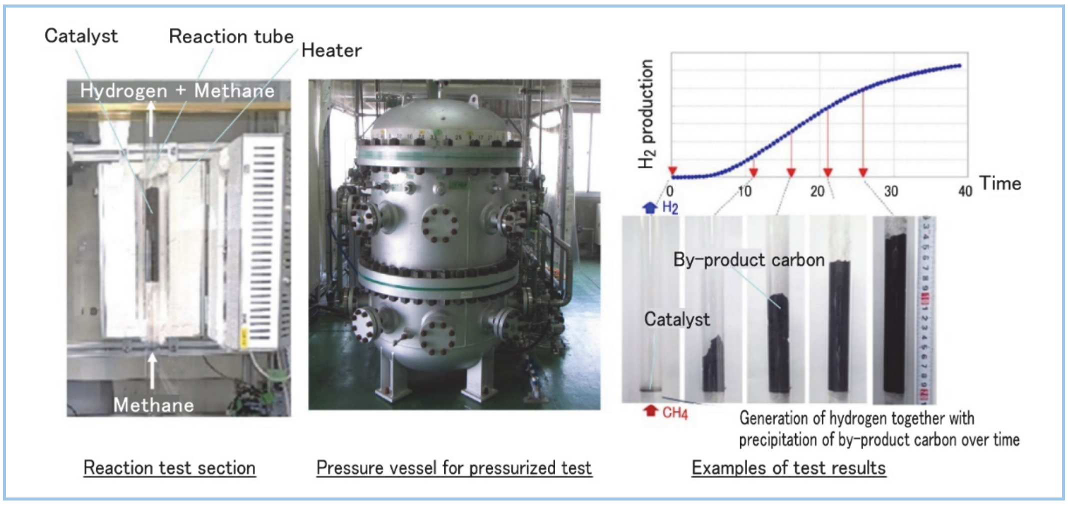

Figure 12 Batch-type fluidized bed test equipment and test results

Figure 13 shows the continuous pressurized fluidized bed test equipment. In addition to a fluidized bed reactor and heaters installed in the pressure vessel, the equipment also has a catalyst feeder and a by-product carbon removal system. Thus, the methane pyrolysis reaction test can be conducted continuously. Typical test results are given in **Figure 14**. Catalyst feeding and by-product carbon removal are carried out constantly under pressurized high-temperature conditions. The equipment is operated continuously while maintaining a constant height of the fluidized bed. By accumulating test data and optimizing operating conditions, the stability was greatly improved from 2024, and it was confirmed that continuous operation for more than 200 hours was possible. During this continuous test, conditions such as velocity and temperature were changed, and the conditions for stable operation and sensitivity of various parameters were obtained. Based on these data, a reaction model simulating continuous catalyst supply and continuous powder discharge was developed. By predicting reactions from scaling up with commercialization in mind using this model, optimizing the design through parameter studies, and reflecting future catalyst performance, MHI will improve the efficiency of the plant.

Through these stable and continuous tests, MHI obtained necessary data to study powder handling. And through the acquisition of the composition and particle size of the powder discharged under various conditions, MHI utilizes them in selecting powder transfer methods and collection equipment, as well as evaluating various life cycles, such as wear caused by the powder.

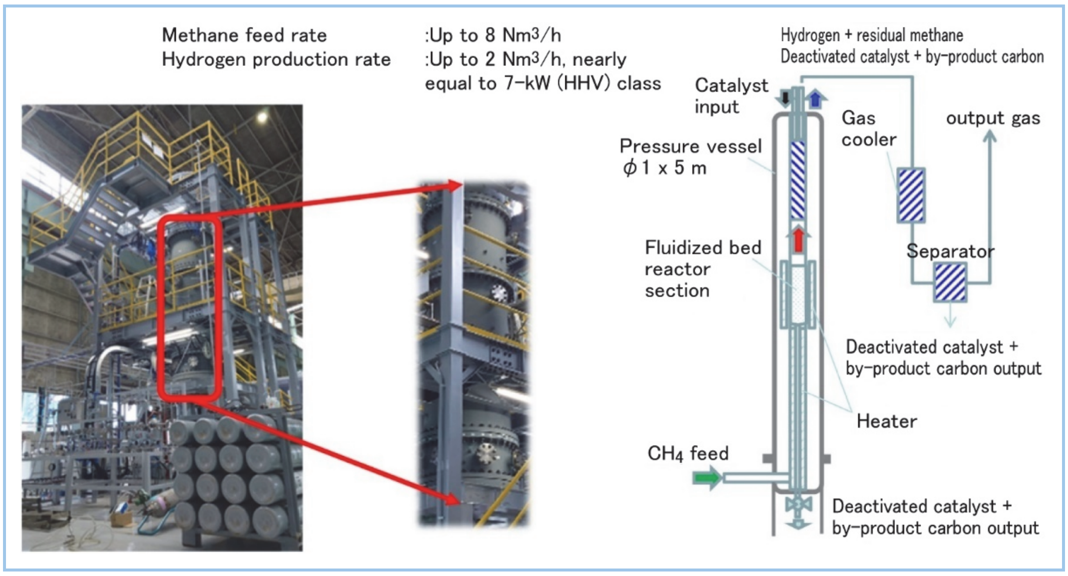

Figure 13 Continuous pressurized fluidized bed test equipment

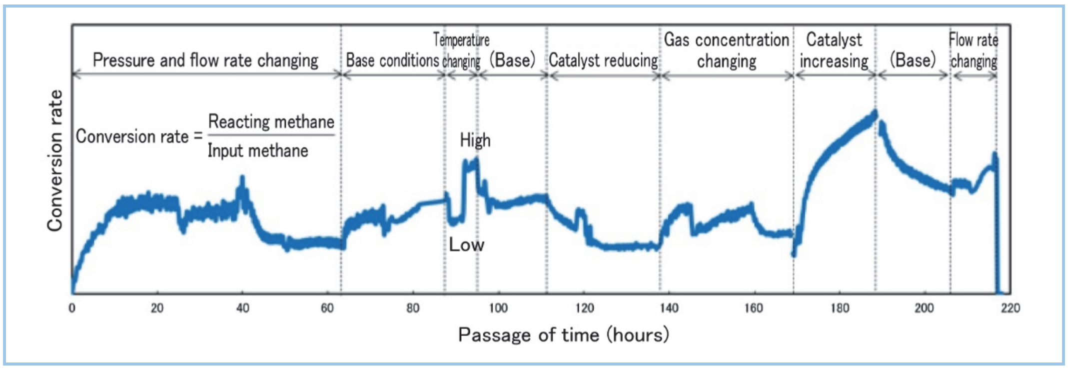

Figure 14 Methane pyrolysis test results using pressurized fluidized bed

Figure 15 shows the road map for development. Now, while the reactor test through the aforementioned batch-type and continuous reactor tests is in progress to investigate the characteristics, another demonstration test unit is being designed, which will be used to verify the whole process of the hydrogen production facility.

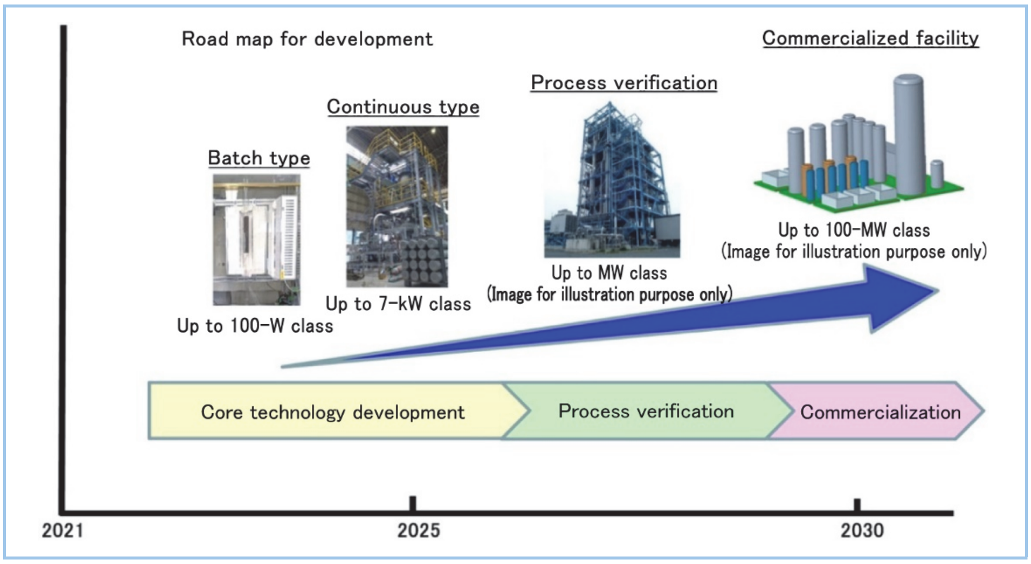

Figure 15 Road map for turquoise hydrogen development

6. Current status in development of synthetic fuel production technology

The previous chapters have provided an overview of MHI's hydrogen production technologies. As for examples of hydrogen usage, the previous report [1] focused on the position of carbon-neutral fuels (hereinafter referred to as CN fuels) and liquid synthetic fuels (synthetic fuels) production using electrolytic hydrogen. In the transportation sector, one of the world's major CO_2-emitting sectors, the CN fuels, the use of which does not increase CO_2 emissions, are expected to be a promising option for large aircraft and ships, which are difficult to decarbonize through electrification. This chapter reports on the status of MHI's development of synthetic fuel production technology.

SOEC can produce hydrogen by electrolysis of steam, and also can produce hydrogen and carbon monoxide, which are feedstocks for synthetic fuels, by mixing carbon dioxide with steam and performing electrolysis (co-electrolysis) [4]. This SOEC co-electrolysis enables the production of synthetic fuel feedstocks in a single unit of equipment, simplifying the process and achieving highly economical synthetic fuel production through the use of highly efficient SOEC. MHI is developing this technology in parallel with hydrogen production technology.

This co-electrolysis utilizes the internal reforming function, a feature of MHI's tubular cell stacks. When using MHI's cell stack as SOFC, natural gas or city gas can be supplied directly to the SOFC as fuel without a reformer. In other words, fuel gas is reformed into hydrogen and carbon monoxide inside the SOFC for power generation using catalytic components in the cell stack materials at the top/bottom parts of the cell stack and steam supplied by recirculation. Co-electrolysis is performed utilizing this internal reforming function as well. MHI completed a relatively short continuous co-electrolysis test of about 900 hours with no noticeable cell degradation observed [4].

Gas produced by co-electrolysis is required to have a ratio of hydrogen to carbon monoxide suitable for Fischer-Tropsch (FT) synthesis. In general, the suitable ratio of hydrogen to carbon monoxide is 2 to 1. It was confirmed in elemental tests that SOEC co-electrolysis can adjust this to a suitable ratio for FT synthesis by appropriately adjusting parameters such as the composition of the supplied gas and the recirculation rate (**Figure 16**(a)). It was also confirmed that the ratio of hydrogen to carbon monoxide can be adjusted by adjusting the ratio of steam and carbon dioxide in the feedstock gas even when the former ratio required in the synthesis process changes (Figure 16(b)).

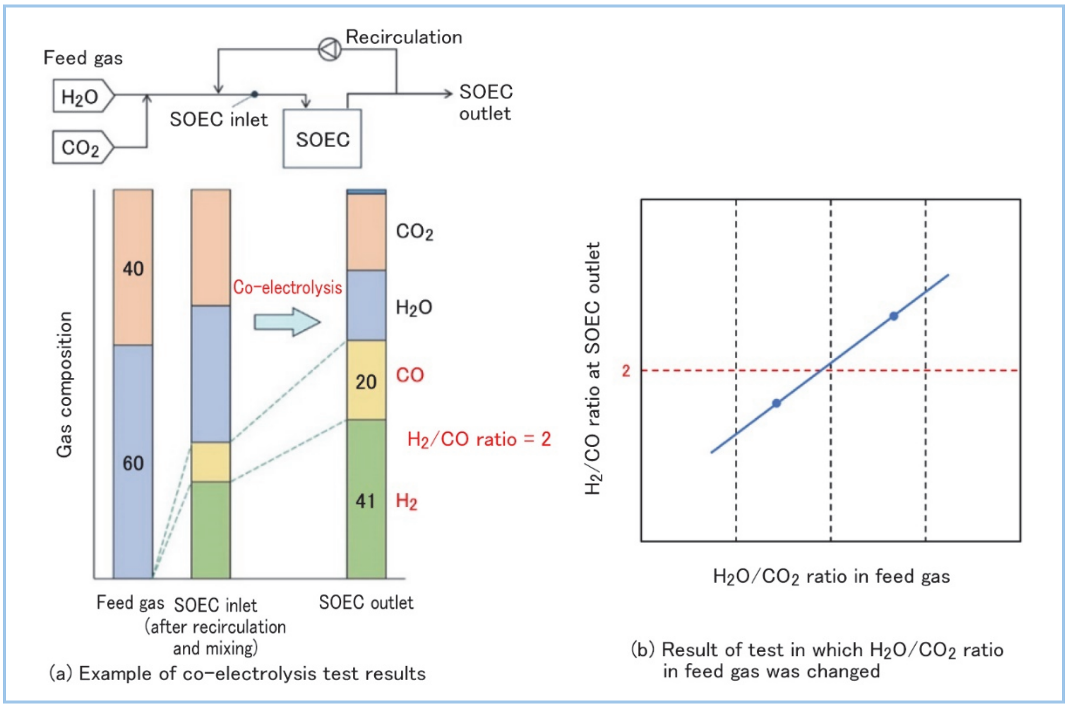

Figure 16 Co-electrolysis test results

In addition to FT synthesis, which is used for synthetic fuel production as described above, the hydrogen and carbon monoxide produced by co-electrolysis can be used as feedstock for methanol synthesis and other applications (**Figure 17**). Co-electrolysis is a promising technology with high versatility and can provide a variety of options for the realization of a decarbonized society.

Figure 18 shows a road map for development. MHI has a plan to proceed with the development by continuously working on elemental tests, simulations, and process studies of co-electrolysis, verifying an integrated synthetic fuel production system, and then deploying it for commercialized facilities.

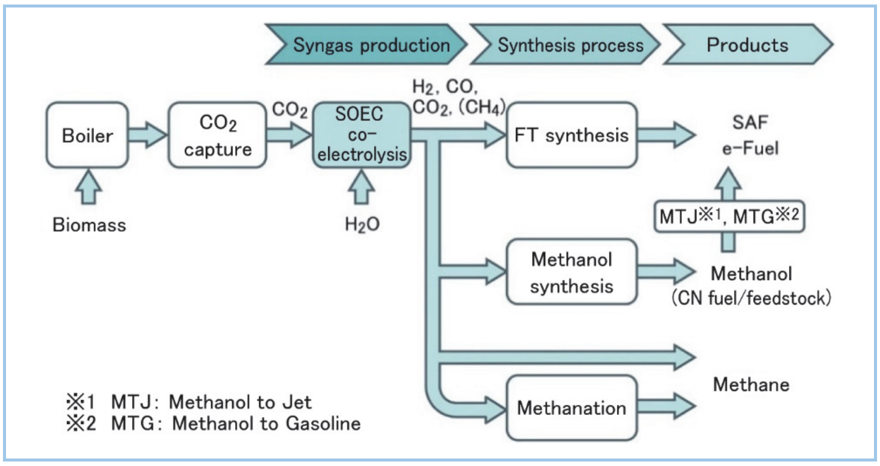

Figure 17 Processes and products derived from synthetic gas

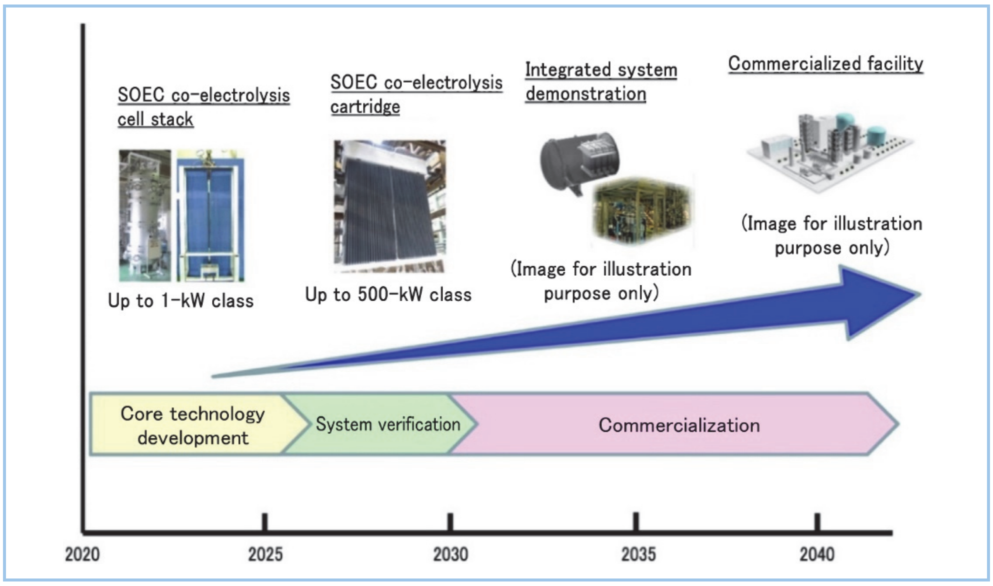

Figure 18 Road map for synthetic fuel production technology (SOEC co-electrolysis) development

7. Conclusion

This paper presented the progress in the development of three types of high-pressure, high-efficiency, and large-capacity hydrogen production technologies focusing on the use of hydrogen for power generation: SOEC, AEM water electrolysis, and methane pyrolysis, and the development of synthetic fuel production technologies derived therefrom.

MHI Group will use the energy transition technologies described in this report to achieve the declaration "MISSION NET ZERO" for the year 2040 and to contribute to the realization of a carbon-neutral society.

"Hydrogen Is Not the Future, This Is Real."

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