DEVELOPMENT PROGRAMME ON HYDROGEN PRODUCTION IN HTTR

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Abstract

In Japan Atomic Energy Research Institute, a hydrogen production system is being designed to produce hydrogen by means of a steam reforming process of natural gas using nuclear heat (10MW, 905°C) supplied by the High Temperature Engineering Test Reactor (HTTR). The safety principle and criteria are also being investigated in the HTTR hydrogen production system. A facility for an out-of-pile test prior to the demonstration test with the HTTR hydrogen production system is under manufacturing. The out-of-pile test facility simulates key components downstream an intermediate heat exchanger of the HTTR hydrogen production system on a scale of 1 to 30. The test on safety, controllability and performance of the hydrogen production system will be started in 2001 and continued for 4 years or longer. In parallel to this, a hydrogen permeation test and a corrosion test of a catalyst tube of a steam reformer are being carried out to obtain data necessary for the design of the system.

As for the HTTR hydrogen production system, a conceptual design is in progress, and check and review for the demonstration program will be made in 2000 from a financial point of view as well as technical view.

Following a brief overview of the program, the design achievements including safety philosophy so far and technical issues to be resolved are to be summarized in the paper.

I. INTRODUCTION

Research and development (R&D) for clean, economical, stable, safe and abundant energy should be promoted from a viewpoint of technology as a potential measure to mitigate the global warming issue as well as for massive and stable energy supply and utilization. We have various options as alternative energy for fossil fuels: solar, geothermal, hydropower and nuclear energy and so on. While available natural energy is limited due to its stability, quality, quantity and density, it is sure that nuclear energy by high-temperature gas-cooled reactors (HTGRs) has the potential to come up with a share as regards a satiable energy supply and utilization. Nuclear energy has been exclusively utilized for electric power generation, but the direct utilization of nuclear thermal energy is necessary and indispensable so that the energy efficiency can be increased and energy savings can be promoted in the near future. The hydrogen production is one of the key technologies for direct utilization of nuclear thermal energy.
The Japan Atomic Energy Research Institute (JAERI) has carried out the R&D on the High Temperature Gas-cooled Reactors (HTGRs), and started the construction of the High Temperature Engineering Test Reactor (HTTR) in the Oarai Research Establishment in March 1991. The HTTR is a test reactor with thermal output of 30 MW and outlet coolant temperature of 950 °C, and has capability to demonstrate a nuclear heat utilization system. An intermediate heat exchanger (IHX) installed in a reactor containment vessel can supply thermal heat of 10 MW to the heat utilization system.

A hydrogen production system by steam reforming of natural gas, chemical reaction; \[ \text{CH}_4 + \text{H}_2\text{O} = 3\text{H}_2 + \text{CO} \], is to be the first heat utilization system of the HTTR. It is the reason that its technology matured in fossil-fired plant enables the coupling with the HTTR in the early 2000’s and technical solutions demonstrated by the coupling will contribute to other hydrogen production systems such as water splitting by a thermochemical method. From Science and Technology Agency (STA) of Japan, the JAERI has been entrusted with the R&D of the HTTR hydrogen production system in January 1997. The R&D of the HTTR hydrogen production system consists of studies of design and safety of the HTTR hydrogen production system, an out-of-pile test, and components tests, which are a hydrogen/tritium permeation test and a corrosion test, necessary for the construction of the HTTR hydrogen production system. The outline of the R&D and safety-related technology for the hydrogen production system in HTTR are reported in the following.

II. DEMONSTRATION PROGRAM OF HYDROGEN PRODUCTION IN HTTR

Figure 1 shows the development schedule of the demonstration program of the hydrogen production in the HTTR. The first half of the demonstration program boxed by solid lines in Fig.1 is from 1997 to 2004, and the last half boxed by dotted lines is planned until 2010. The execution of the last half is decided after taking the check and review (C&R) in 2000. The design and safety studies of the HTTR hydrogen production system are carried out until 2000. The facility of the out-of-pile test for the HTTR hydrogen production system is designed and constructed in 1997-2000, then the out-of-pile test is performed until 2004. In the component tests, two experimental apparatus were completed in 1997, and the hydrogen/tritium permeation test and the corrosion test are being carried out in 1998-2000. After the C&R of the HTTR hydrogen production system, the construction of the HTTR hydrogen production system will be started from 2001 and the demonstration test will be started in around 2005-2010.

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Fig.1 Schedule of demonstration program.
III. HTTR HYDROGEN PRODUCTION SYSTEM

1. Overview of HTTR hydrogen production system

The HTTR hydrogen production system is designed to utilize the nuclear heat effectively and achieve hydrogen productivity competitive to that of a fossil-fired plant with operability, controllability and safety acceptable enough to commercialization. Figure 2 shows an arrangement of the main components. The HTTR reactor supplies nuclear heat of 10MW with 950°C to the IHX in the reactor cooling loop, and then the nuclear heat is transferred from the IHX to the secondary helium loop to be utilized for the production of hydrogen. Due to heat loss along the secondary helium piping from the IHX to a steam reformer (SR), the secondary helium temperature is reduced to 880°C at the SR inlet, whereas the IHX outlet temperature is 905°C. Design specifications of the HTTR hydrogen production system is shown in Table I. The key components, such as the

![Flow scheme of HTTR hydrogen production system](image)

Fig. 2 Flow scheme of HTTR hydrogen production system.

<table>
<thead>
<tr>
<th>Table I</th>
<th>Design specifications of HTTR hydrogen production system and out-of-pile test facility.</th>
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SR and a steam generator (SG), and their arrangement were designed to achieve hydrogen productivity competitive to that of a fossil-fired system with operability, controllability and safety acceptable enough for commercialization.  

2. Safety-related technology to be developed

Although the hydrogen production system by steam reforming is matured in fossil-fired plants, some safety-related technology should be developed for coupling with HTGRs as well as the HTTR as shown in Fig. 3.

(1) Mitigation of thermal disturbance to reactor

The SG supplies steam to the SR, and can also stabilize the inlet temperature of the IHX in the secondary helium coolant loop. Even if the helium temperature at the SR outlet, that is, the SG inlet is increased by some thermal disturbance such a malfunction in the reforming process gas line, the helium temperature at the SG outlet can be kept constant at the saturation temperature of steam by controlling the pressure in the SG. It is possible that the nuclear reactor can be stopped according to a normal operation procedure but not with a reactor scram for some malfunction or accident at the heat utilization system by this performance of the SG working as an absorber of thermal disturbance. We aim to limit the temperature fluctuation of the secondary helium gas within 10°C at the SG outlet, because the temperature rise above 15°C compared with the normal temperature at the reactor inlet causes the HTTR reactor scram.

(2) Assurance of structural integrity of catalyst tube

(a) Control of pressure difference between helium and process gases at catalyst tube

The catalyst tube in the SR is a component important to safety, because it forms a pressure boundary between helium and process gases. In design of the catalyst tube, its wall thickness should be decided considering both outer pressure of helium gas at 4.1MPa and inner pressure of process gas at 4.5MPa to assure the structural integrity in all conditions such as not only normal start-up and shut-down but also malfunction and accident at the heat utilization system. This design, however, makes the wall thickness too heavy; for example, the wall thickness becomes about 130mm as against an inner diameter of 128mm using Alloy 800H. To realize the reasonable wall thickness, 10-mm level, it should be decided considering the pressure difference between helium and process gases, and a control system is required to keep the pressure difference within an allowable value.

Fig. 3 Safety related technology to be developed for HTTR hydrogen production system.
In concrete terms, the control system makes the process gas pressure follow pressure change of helium gas.

(b) Estimation of hydrogen embrittlement and corrosion of catalyst tube
The catalyst tube will be made of Hastelloy XR, which is a nickel-base, helium corrosion- and heat-resistance super alloy developed for the HTTR in the JAERI, and its strength in high temperature is nearly equal to that of Alloy 800H. In order to verify the validity of the structural design of the HTTR hydrogen production system, characteristics of corrosion due to metal dusting and oxidation and strength reduction due to hydrogen embrittlement should be estimated in the corrosive gases such as CH₄, CO, H₂O and H₂ simulating the SR condition.

(3) Estimation of tritium permeation
Tritium produced in the HTTR core flows with the primary helium coolant to the IHX, then permeates through the Hastelloy XR tube of the IHX to the secondary helium coolant and through the Hastelloy XR tube of the SR, at last, mixes with the process. Therefore tritium concentration in the process gas should be estimated because tritium can not be perfectly removed by a purification system in the HTTR.

IV. OUT-OF-PILE TEST

1. Objective and test facility
The main objectives are investigation of transient behavior and establishment of operation and control technology, focussing on establishment of the safety-related technology (1) and (2-a) described in section III.2, as well as design verification of performance of high temperature components, such as the SR and SG.

The test facility has an hydrogen production capacity of 110Nm³/h and simulates key components downstream the IHX of the HTTR hydrogen production system on a scale of 1 to 30⁴. Design specifications of this test system is also shown in Table I. Figure 4 shows a schematic flow diagram of the test facility. An electric heater with 380kW is used

![Schematic flow diagram of the out-of-pile test facility.](image_url)
as a heat source instead of the nuclear heat in order to heat helium gas up to 880°C at the SR inlet of the same conditions as the HTTR hydrogen production system. The process gas pressure is controlled by a control valve installed downstream from the SR, monitoring the pressure difference between helium and process gases.

Figure 5 shows the schematic view of the SR which has one bayonet-type catalyst tube made of Alloy 800H. Dimensions such as diameter and length is approximately the same as those of the catalyst tube of the HTTR hydrogen production system. By the way, the SR of the HTTR hydrogen production system has 30 bayonet-type catalyst tubes made of Hastelloy XR. Process gas flows in the catalyst tube at inlet temperature of 450°C and helium gas flows in a channel between catalyst and guide tubes at inlet temperature of 880°C. In the fossil-fired plant, the process gas receives heat from combustion air of about 1200°C by radiation, and the heat flux at the outer surface of the catalyst tube reaches 70,000-87,000 W/m². In order to achieve the same heat flux as that of the fossil-fired plant, it is very important to promote heat transfer of helium gas by forced convection because the temperature of helium gas, that is, the temperature of heat source is too low compared with that of the fossil-fired plant. So, disc-type fins, 2mm in height, 1mm in width and 3mm in pitch, are arranged around outer surface of the catalyst tube in the test facility in order to increase a heat transfer coefficient of helium gas by 2.7 times, 2150W/m²K with the fins, and a heat transfer area by 2.3 times larger than those of smooth surface, respectively. As the result, the heat transfer performance of the catalyst tube in the test facility becomes competitive to that of the fossil-fired plant.

2. Test plan

The tests are considered on three categories, that is, (i) normal start-up and shut-down test to establish the operation method, (ii) safety-related test dealing with malfunction and accident at process and helium gas lines, and (iii) high temperature components test to investigate the performance.

(1) Normal start-up and shut-down test

The normal start-up and shut-down of the hydrogen production system is carried out following in those of the HTTR reactor. The change of temperature and pressure of helium gas causes fluctuation of the reforming reaction in the SR attended with fluctuation of both helium gas temperature at the SR outlet and process gas pressure in the catalyst tube. The objective of the test is optimization of feed of natural gas and steam according to change of the temperature and pressure of helium gas supplied from the HTTR reactor in order to restrain the above fluctuation within allowable range.

Fig. 5 Schematic view of steam reformer.
(2) Safety-related test

(a) Malfunction and accident at process gas line

The objective is establishment of the safety-related technology (1) and (2-a) described in the section III.2, dealing with malfunction and accident at the process gas line. The test is carried out by step-change of feed amount of natural gas and steam and so on, and then the control system of the pressure difference is optimized and the performance of the SG for mitigation of fluctuation of helium gas is investigated. Even if the process gas feed is stopped completely, we aim to stop the HTTR reactor by the normal operation procedure but not with the reactor scram. At this time, heat of the helium gas can not be removed at the SR because the reforming reaction is curtailed. A cooling system of helium gas by the SG, whose detail is described in the section (3) in this chapter, is examined to investigate the cooling performance and controllability.

(b) Malfunction and accident at helium gas line

The reactor scram is conducted in this case. The emergency shut-down method of the hydrogen production system is established to assure the safety, especially structural integrity of the catalyst tube, by the experiment.

(3) High temperature components test

Thermal and hydraulic performance of the SR and SG is clarified. The SR is investigated focussing on reaction characteristics which is very important to predict transient behavior and hydrogen productivity of the hydrogen production system. The cooling system of helium gas at the trouble has been designed, using the SG and an air-cooled radiator which is installed above the SG to increase cooling power of the SG. Steam produced in the SG is condensed into water at the radiator, and steam and water circulate between the SG and radiator by natural convection. In order to limit the temperature fluctuation of helium gas at the SG outlet within an allowable value, it is important to control saturation temperature of steam in the SG, that is, to control the pressure in the SG. The pressure is controlled by a fan at the radiator adjusting flow rate of the cooling air as shown in Fig. 4. The pressure controllability, transient behavior of temperature of helium gas and steam, steam production rate and natural convection of steam and condensed water are investigated in detail.

V. COMPONENT TESTS

In parallel to the out-of-pile test described above, the following tests are planned out with other small testing apparatus to establish the safety-related technology (2-b) and (3) described in section III.2 and to obtain detailed data for a safety review of the HTTR hydrogen production system and development of calculation codes; (i) corrosion test and (ii) hydrogen/tritium permeation test.

The objective of the corrosion test is estimation of the effect of corrosion, oxidation and hydrogen embrittlement on strength reduction of Hastelloy XR. Metallography and material tests on strength and creep are in progress with test specimens exposed in the corrosive gases at temperature up to 900°C.

While tritium produced in the HTTR core permeates in to the hydrogen production system, hydrogen in the product gases also permeates in the opposite direction from the SR to the primary helium coolant loop. The aims of the hydrogen/tritium permeation test are to obtain the data of permeation coefficient in the very low tritium partial pressure less than 10 Pa, to examine the effect of an isotope of hydrogen simultaneously existing in the gas,
and the effect of protection for hydrogen/tritium permeation by the coating film on the reforming tube such as oxidation film, calorizing film and so on. In such a region of low tritium partial pressure, chemisorption phenomenon at the tube surface during dissociation and adsorption is more dominant than diffusion in the tube. In this region, it is known that tritium penetration rate is proportional to a square root of tritium partial pressure not to a linear of it, therefore hydrogen and deuterium are used instead of tritium in the test.

VI. ECONOMIC ASPECT

One of the biggest problems is absolutely economy for the commercial nuclear process heat utilization system. It is said that the economy of the total system depends on the capital cost for the HTGRs which supply heat to the heat application system via intermediate heat exchanger, because it is presumed that the fraction of the heat application system downstream the intermediate heat exchanger and the hot duct is relatively small in comparison with the HTGRs themselves. According to a private communication, a German simple estimation suggests the fraction is less than one-thirds, maybe one-fifth. Thus, the economy improvement of the reactor is inevitable for the success of the commercial plant.

It is obvious that the HTGR safety is achieved by a large core with low power density. In comparison to the current LWRs, the power density is less than one-tenth in the HTGRs. Such low power density yields the inherent safety aspect, whereas it requires more capital due to the scale demerit. For example, the size of the pressure vessel of the HTTR with 30 MW thermal output is as large as that of medium size of LWR with 500 MW electrical output. The HTGRs are apparently disadvantageous in economy in comparison with the current LWRs.

On the other hand, the inherent safety aspect in the HTGRs could make it possible that no or quite limited engineering safeguards of reactor grade quality are needed. The only safety elements in the entire system are the fuel element and graphite core components which can be checked in running operation, while the safety of LWRs with high power density is ensured by extensive, active and passive safeguards and the reactor grade quality of the components and materials. Sophistication and expensive reactor grade quality is particularly required for all components of LWRs, but, in the case of HTGRs, ultimately only for the fuel element and graphite core components. Thus, the HTGRs would provide a new, qualitatively different safety, resulting in decreasing the cost.

This safety aspect can also make the heat application system designed in a general industrial safety grade, not nuclear grade, resulting in the significant cost reduction of the system. JAERI is now under developing a new safety criteria applicable to the future commercial heat application system, including countermeasures against possible fire or explosion by combustion gasses like methane and hydrogen.

VII. CONCLUDING REMARKS

Under an understanding that HTGRs can play an important role to expand the nuclear heat application to chemical industries against the current environmental issue of the CO₂, JAERI proceeds with the development of the nuclear process heat application system coupling to the HTTR. Global eyes are kept by not only nuclear persons of interest but also the public upon the development of the HTTR heat application system,
since its successful achievement may enhance the possibility to solve the environmental issue of CO$_2$ emission as well as a possible energy crisis which might happen in the future.

Finally it should be emphasized that an overall support and understanding from the overseas countries of concern are needed and wished for the success of the Project. The Project is highly expected to contribute so much to promoting international cooperation on the development of HTGRs and its process heat application.

REFERENCES


