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Nuclear Energy and Hydrogen Production
The Japanese Situation

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1. Design, Research and Development and Operation Experiences of HTGR in Japan

1.1 Introduction

The research and development on the high temperature gas-cooled reactor, HTGR was started in 1969 in Japan aiming to realize early application in industrial field, particularly, direct steel making. In this system, the reducing gases, namely hydrogen would be used to reduce iron ore. The design work of an experimental Very High Temperature Reactor, VHTR has been made at the Japan Atomic Energy Research Institute, JAERI in parallel with various R&D programmes. (Fig. 1) The joint work between JAERI and steel industry on nuclear heat application to the direct steel making had continued by 1980, until the situation in the industry changed remarkably.

On the other hand, the JAERI continued its work with the realization that the HTGR was one of the most promising nuclear reactors because of its many inherent safety characteristics and was expected to obtain high temperature heat around 1000^{o}C which surely not only widens the field of nuclear energy utilization but also rises considerably heat utilization efficiency.

Meantime, a European cooperative project, Dragon reactor started its operation, and two prismatic type HTGRs, Peach Bottom and Fort St.Vrain in USA, and two pebble bed type HTGRs, AVR and THTR in Germany had been constructed and operated. (Fig. 2) As any reactor of them, however, aimed at electricity generation by steam turbine, the reactor outlet helium gas temperature was around 750^{o}C.

1.2 Outline of the HTTR

Based on the research and development of HTGR and the HTTR (High-Temperature Gas-cooled Engineering Test Reactor) was built in JAERI. Table 1 summarizes the major specification of the HTTR. The HTTR produces 30 MW thermal output and outlet coolant temperature is 850^{o}C at the rated operation and 950^{o}C at the high temperature test operation. The fuel element is prismatic block type, which is shown later. The primary cooling circuit contained helium gas pressurized to 4 MPa is separated into two lines.

The bird's-eye view of the reactor building is shown in Fig. 3. The reactor containment vessel contains reactor pressure vessel and primary and secondary cooling systems. The reactor pressure vessel is 13.2 m high and 5.5 m in inner diameter. The core consists of 30 fuel columns piled up with 5 blocks each. The air cooler is located at the top level of the reactor building.

A coated fuel particle consists of a low enriched UO_2 kernel with TRISO coating, and about 20,000 fuel particles are combined with graphite powder to form a fuel compact. A fuel rod is composed with graphite sleeve in which fuel compacts are contained. (Fig. 4) A fuel assembly is a pin-in-block type hexagonal fuel element, that is, helium gas flows through the gap between a vertical hole and a fuel rod to remove the heat produced by fission and gamma heating. The reactor cooling system shown in Fig. 5 is composed of a main cooling system, an auxiliary cooling system and two reactor vessel cooling systems. The main cooling system is separated into two lines outside the reactor vessel. The heated helium gas is cooled by a He/He intermediate heat exchanger, IHX in one line or cooled directly by a pressurized water cooler, PWC in the other line. Another PWC is provided after the IHX in the first line and heat is finally removed by an air cooler in both lines. The high temperature helium gas around 900^{o}C from the IHX with heat capacity of 10 MW can be used as process heat for hydrogen production and others.

1.3 Key Technologies Developed for the HTTR

The JAERI had made many key researches and development for the HTTR. Figures 6 and 7 show just a couple of typical results. The Oarai Gas Loop No.1, OGL-1 was installed in the JMTR (Japan Materials Testing Reactor) in which fuel and graphite irradiation tests and fission product plate-out tests were made. The Very High Temperature Reactor Critical Assembly, VHTRC was practically used for reactor physics experiment. The HENDEL is helium engineering demonstration loop for full-scale demonstration test of high temperature components and thermal hydraulics test and so on. Various mechanical tests were made for heat resistant alloys. The high temperature fatigue test machine is shown in Fig. 6.

Excellent graphite was required for core and its surrounding components, which means specific type of graphite with less dimensional change due to neutron irradiation, large tensile strength and high corrosion resistance. Finally, the IG-110 was successfully developed, and the material is also used in the Chinese high-temperature test reactor HTR-10. As concerns coated fuel particle, great effort had been made to improve fabrication technologies to get very high quality one, even compared with coated particles developed by other countries. In addition, JAERI had developed heat resistant alloys and carried out various tests for them. The picture shows that heat resistant superalloy Hastelloy XR, in which some chemical compositions are tightened within specification of Hastelloy X, has very high corrosion resistance.

1.4 Operational Experiences

The first criticality of the HTTR was attained on November 10, 1998 at annular form of fuel loading which was specifically asked by USA. The minimum critical core was obtained by 19 fuel columns. (Fig. 8) After various tests at the rated power and reactor outlet coolant temperature of 850^{o}C, the HTTR was successfully achieved 950^{o}C at reactor outlet in April of this year as shown in a chart of a history of reactor power and reactor outlet coolant temperature in the test. (Fig. 9) Figure 10 shows fuel performance data in the HTTR. Improvement of fuel fabrication technique was continued based on the fuel irradiation data at the JMTR. It is reported that the fuel performance in the HTTR has been so good so far.

Several safety demonstration tests have been made in the HTTR to confirm the inherent safety features of HTGRs. Figure 11 shows transient behavior of reactor power, fuel and coolant temperatures without scram in case of sudden stop of two gas circulators out of three. The reactor power decreases due to negative reactivity feedback caused by temperature increases of fuel and coolant and converges to a certain level. This verified that HTGR has a salient inherent safety feature.

2. Possibilities of Application of Nuclear Technologies to Hydrogen Production

2.1 Present Status of Energy Consumption and Enlargement of Utilization of Nuclear Energy in Japan

The self-sufficiency of energy in Japan is only 4%, the lowest among the developed countries. It becomes somehow 20% if it includes electricity production by nuclear energy. Furthermore, it is not possible for Japan to import electric power, and oil and natural gas by pipe-line due to geopolitics situation. Japan, needless to say, has made and has to make continuously great efforts for reduction of CO_2 emission. Among the several measures to the issue, the following ones are executed, planned or can be expected. These are:

- Enlargement of utilization of nuclear energy for non-electricity generation as well as electricity generation, and renewable energy,
- Utilization of hydrogen.

Figure 12 shows a composition of primary energy supply in Japan. Electricity generation by nuclear energy contributes by 34.5% to the total electricity production which shares 40% of the total primary energy supply. Self-sufficiency of energy consists of nuclear energy, hydropower and renewable energy. On the other hand, half of the primary energy relies on oil. As concerns enlargement of utilization of nuclear energy, the total electricity generation capacity by nuclear energy will be increased from 45 GW at present to around 60 GW and utilities will make efforts to increase the capacity factor of plant up to 90%.

As for Non-electricity utilization, nuclear heat can be applied for various industrial fields depending on supplied heat temperature, such as hydrogen production, oil refinery, desalination, district heating, and so on, as shown in Fig. 13. In addition, if the temperature is enough high, electricity generation by gas turbine with higher heat efficiency is also possible. Furthermore, high temperature nuclear heat from HTGR can be applied for electricity and process heat cogeneration by cascade manner.

Figure 14 is a design example of HTGR cascade energy plant for production of hydrogen, electricity and freshwater with total heat utilization efficiency of 80%.

2.2 Hydrogen Utilization Programme in Japan

There is no so strong governmental initiative for hydrogen utilization in Japan at present. However, the fuel cell commercialization strategy committee of the Agency of Natural Resources and Energy under the Ministry of Economics, Trade and Industry proposes the target for introduction of fuel cells to market. The amount of annual hydrogen demand is currently 1.5 Gm^3. However, it will increase to more than 50 Gm^3 per year in 2030 for household and vehicles according to the estimation of the committee. As for vehicles, 15 million cars among 75 million cars will be operated with hydrogen. (Fig. 15)

The committee estimated amount of hydrogen demand at every 10 year's spreading step. The fossil fuel reforming, by-product hydrogen and water electrolysis are major means to supply hydrogen at the early stage. However, the committee indicated that the HTGR hydrogen, coal reforming hydrogen with CO_2 sequestration and renewable energy hydrogen would be possibly major sources in future. As for transportation of hydrogen, there are three options, and onsite supply and hydrogen tank lorry are available already although it is necessary to make effort for much cheaper ones. The pipe-line system might be the cheapest way to transport hydrogen for more than several hundreds km. (Fig. 16)

On the other hand, the Cabinet council decided the Energy Ground Plan last year, which was prepared, following the enactment of the National Energy Policy Master Law in 2002. The Ground Plan describes that hydrogen production processes with nuclear, solar and biomass energies, which are independent of fossil fuels, are expected as future technologies.

2.3 HTTR-Hydrogen Production Demonstration Programme

The JAERI has an HTTR hydrogen production demonstration programme. In this programme, high temperature helium gas from the outlet of intermediate heat exchanger of the HTTR is transferred to the hydrogen production plant through the hot gas duct. (Fig. 17) Although there are several processes to produce hydrogen using high temperature, both thermochemical iodine-sulfur process, IS process and steam reforming process are considered as the hydrogen production process to be connected to the HTTR. The IS process shown in Fig. 18 is a high temperature thermochemical decomposition of water with iodine and sulfur, and a really clean process which produces hydrogen from water. The high temperature process heat is required in hydrogen iodide decomposition and sulfuric acid decomposition. After the laboratory-scaled feasibility test, the test stepped to the bench-scaled test. Currently, the JAERI is preparing the pilot test in which hydrogen will be produced at the rate of $30m^3/h$. Finally, a hydrogen production plant with 1000 m^3/h will be demonstrated connecting with the HTTR.

Figure 19 shows the bench-scaled test apparatus of the IS process, most of them are made of glass. The continuous hydrogen production was successfully achieved with hydrogen production rate of 30 l/h for long time.

Table 2 shows major characteristics of bench-scaled, pilot and HTTR tests. The key differences between the bench-scaled test and the pilot test are material of chemical reactors and associated pressure of chemical process. The JAERI has used glasses as materials of the chemical reactors of hydrogen iodide decomposition and sulfuric acid decomposition which are highly corrosive. The development of new material for these chemical reactors has been a key issue which needs break-through for industrialization of IS process. Now, the JAERI succeeded in development of some industrial materials and is going to construct a pilot test plant.

The flow diagram of IS process in the pilot test plant is shown in Fig. 20. The heated helium gas from helium circulation loop is supplied to sulfuric acid decomposer and hydrogen iodide decomposer. Water is supplied to Bunsen reactor with the rate of 25 kg/h and hydrogen iodide flows into the HI decomposer through a separator to produce hydrogen at the rate of 30 Nm^3/h. On the other hand, sulfuric acid flows into the sulfuric acid decomposer to produce oxygen.

As for steam reforming process, the JAERI have made system integration simulation test with full-size facility. Figure 21 shows the flow diagram of steam reforming process facility, and the appearance of full-sized facility. Valuable data on static and dynamic behavior of the system was accumulated and also control technology was established which can restrain thermal disturbance anticipated to happen in the steam reforming vessel by using a steam generator.

There is no experience, nor safety guideline determined in the world on the coupled system with a nuclear reactor facility and a chemical plant, specifically with possibility of explosion. One of the important issues to realize nuclear hydrogen production system is to establish safety regulation rule on the coupled system. Several points to be discussed and ruled in the safety regulatory guideline, are:

(1) How should be maintained the safety of a nuclear reactor against an explosion accident in the chemical plant?
How large explosion should be postulated?
How much distance should be kept between a nuclear reactor and a chemical plant and between a chemical plant and site boundary?
(2) How should be maintained the safety of a chemical plant and employees in case of an accident of a nuclear reactor?
(3) How should make sure of a final heat sink in case of trouble or accident of a chemical plant?
(4) Should a chemical plant be in the reactor grade or not?

and so on.

The current schedule of HTTR-hydrogen production demonstration programme is shown in Table 3. According to JAERI, hydrogen production will be demonstrated using the heat from the HTTR in 2010 by IS process if it is possible, if not, by steam reforming process at first. It is quite worthwhile to demonstrate to produce hydrogen by using a real nuclear reactor as soon as possible solving technical and safety issues. Accumulation of operational experience and various data on the coupled system as well as the HTTR itself is also very useful for evaluation of economical competitiveness and other countries' programmes.

2.4 Toward Commercialization of Nuclear Hydrogen Production

As for verification of technological feasibilities, the HTTR hydrogen production test is ready for steam reforming process. As concerns IS process, it is necessary to obtain the result of the pilot test. (Table 4) The second issue is how and who, chemical industry, electric power company or other organization can commercialize nuclear hydrogen production and become a vender.

As concerns economical competitiveness, according to the estimation by JAERI, the cost of hydrogen produced by IS process with HTGR is about 30% lower than that by methane steam reforming with fossil fuel if CO_2 management cost is imposed and the cost by methane steam reforming with HTGR is about 20% lower than this case. (Fig. 22) If no CO_2 management cost nor carbon tax is imposed, the hydrogen cost by IS process with HTGR is rather high compared with that by the existing process and we need to make further effort to make the cost of HTGR much cheaper and to improve IS process. For this purpose, we need a medium sized, let say 600 MW, demonstration HTGR which can produce enough amount of hydrogen by IS process to be supplied to 1 million fuel cell vehicles. This project is desirable to be an internationally collaborated one based on various operational experiences of the HTTR and the results of hydrogen production demonstration test in the HTTR.

On the other hand, it is necessary to seek for an undertaking company and/or organization to build and operate a hydrogen production or cogeneration HTGRs. In response to liberalization of electric power market, electric power companies can make not only electric power business but also any other business, gas business, for example. In addition, only electric power companies have experienced to build and operate nuclear power reactors in business in Japan. Of course, there is possibility that an electric power company only sell high temperature process heat to chemical industries.

As for infrastructure, it will be considerably established by the time when nuclear hydrogen production system will become commercial. Currently in Japan, there are several tens of fuel cell cars and two-wheeled vehicles, and over ten hydrogen stations. (Figs. 23, 24)

3. Summary

A coolant outlet temperature of 950^{o}C was successfully achieved in the HTTR of JAERI, based on advanced technologies developed through long-term R&Ds in Japan. This opens a new era for nuclear heat applications. In particular, it will be greatly beneficial to produce hydrogen, a clean energy carrier in the 21st century to prevent environmental disruption such as global warming, abnormal weather, etc. Continuous hydrogen production was successfully achieved with a bench-scale apparatus of thermochemical IS process in JAERI. The HTTR-Hydrogen Production Demonstration Project by the institute is taking a role of the pioneer in this field. It is expected that valuable know-how, technologies and data can be obtained through the HTTR programme. Based on the result in the HTTR test, it would be necessary to build a medium-sized demonstration HTGR which will verify economical competitiveness of nuclear hydrogen production as well as a safe and reliable system. Under the limited resources, such a project would be desirably promoted by international collaboration.

Table 1 Major specification of HTTR of JAERI.

Thermal power	30MW
Inlet coolant temperature	395°C
Outlet coolant temperature	850°C/950°C
Fuel	Coated fuel particle / Low enriched UO_2
Fuel element type	Prismatic block
Pressure vessel	Steel
Number of cooling loop	1
Heat removal	IHX and PWC (parallel loaded)
Primary coolant pressure	4MPa
Containment type	Steel containment

Table 2 Major specifications of bench-scaled, pilot and HTTR tests by JAERI.

	Bench-scaled Test	Pilot Test	HTTR Test
Hydrogen production rate	~ 0.05 m³/h	~30 m³/h	~1000 m³/h
Heat supply	Electrical heater	Heat exchanger with helium gas (Electrical heater 0.4MW)	Heat exchanger with helium gas (Nuclear heat 10MW)
Material of chemical reactors	Glass	Industrial material	Industrial material
Height	About 0.3 m	About 1m	About 3m
Pressure of chemical process	Atmospheric pressure	High pressure	High pressure
Time	FY 1999 – 2004	FY 2005 - 2010	FY 2009 - 2014

Table 3 Schedule of HTTR hydrogen demonstration programme by JAERI.

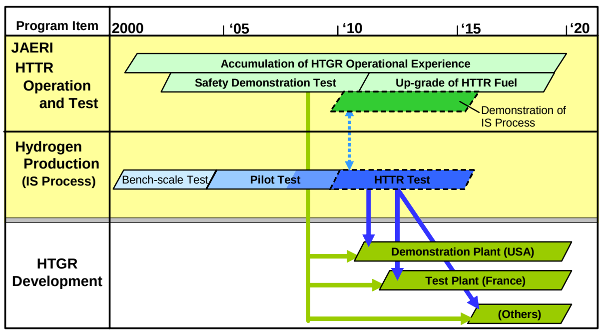


Table 4 Issues for commercialization of nuclear hydrogen production.

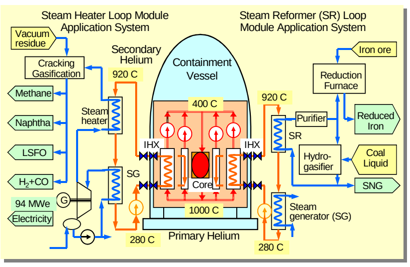
1. Verification of Technological Feasibilities
   - HTTR-hydrogen production test
     — ready for steam reforming process
     — wait the result of the pilot test for IS process (budgetary problem)
2. Verification of Economical Competitiveness
   - The cost of hydrogen produced by IS process with HTGR is lower than that by methane steam reforming with fossil fuel if CO_2 management cost is imposed.
   - We need a middle sized demonstration HTGR to improve technologies and surely verify economical competitiveness of nuclear hydrogen production.

   The international collaboration on the matter is desirable with the countries who have interest and can share technical and financial support.

Table 5 Issues for commercialization of nuclear hydrogen production.

3. Appearance of Undertaking Companies, Organizations
   - Electric Power Companies ?
     → to sell high temperature process heat to chemical industry ?
4. Establishment of Infrastructure
   - Infrastructure will be constructed as demand of hydrogen and, is estimated to increase in the following :
   - Tank lorries of { 2020 – 2030: Compressed hydrogen gas (350 → 700atm), Liquid hydrogen, Chemical hydride (2.2wt% → 6wt%) } ➡ 2050 - •Pipe-line of hydrogen
   - Transportation system will have been prepared to some extent as an infrastructure in by-product hydrogen-systems by around 2020s, when HTGR hydrogen systems will be commercialized.

- Design study of FM50 Experimental Reactor and Prototype Reactor of HTGR for Multipurpose Use, and Component Tests since 1960's in Japan



Prototype HTGR
In Nuclear Iron Manufacturing Project (1973-1980)
- Thermal Output; 500MW
- Outlet Temp. ; 1000C
- Hydrogen Production for Iron Manufacturing with Steam Reforming
- Production of Methane, Naphtha, Synthetic gas
- Electricity of 94MW

Fig. 1 Prototype reactor system of HTGR for multipurpose use designed in 1970s.

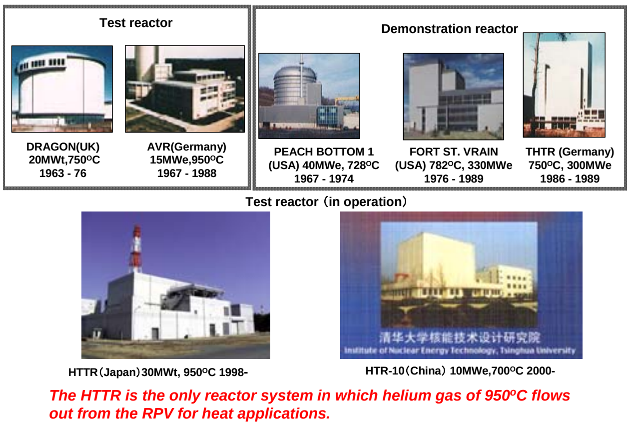


Fig. 2 HTGR history.

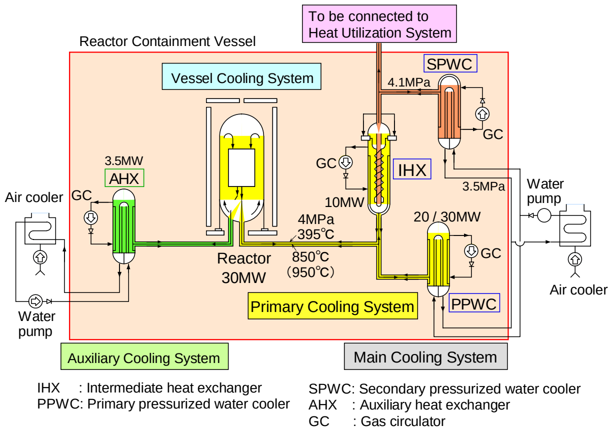


Fig. 5 Cooling system of HTTR of JAERI.

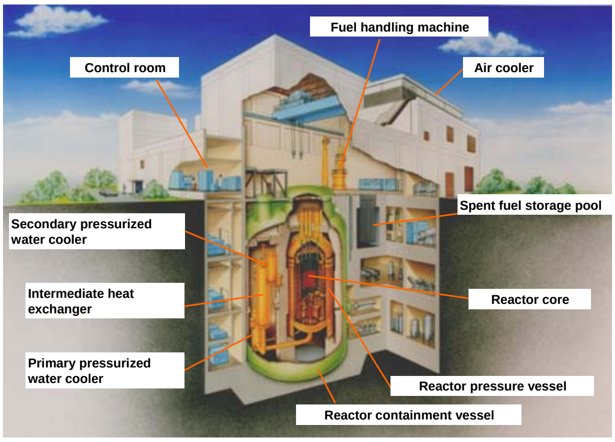


Fig. 3 Bird's-eye view of HTTR reactor building (by JAERI).

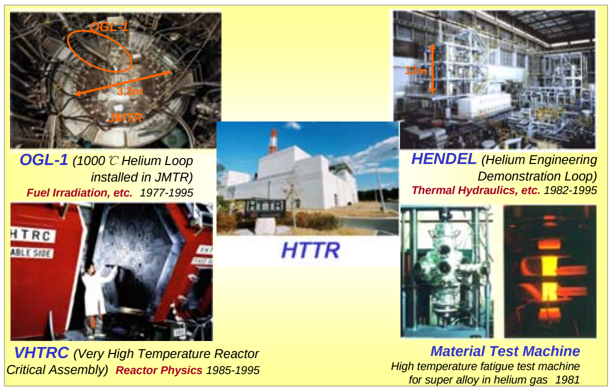


Fig. 6 Major technical R&D facilities in JAERI.

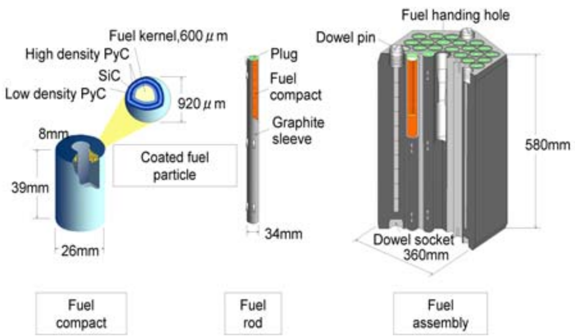


Fig. 4 Details of fuel structure of HTTR (by JAERI).

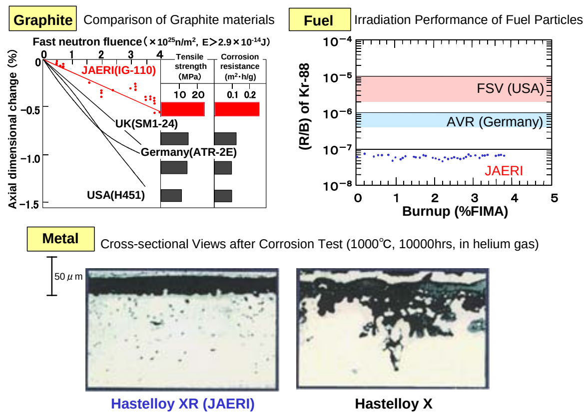


Fig. 7 Results of research and development on fuel and materials (by JAERI).

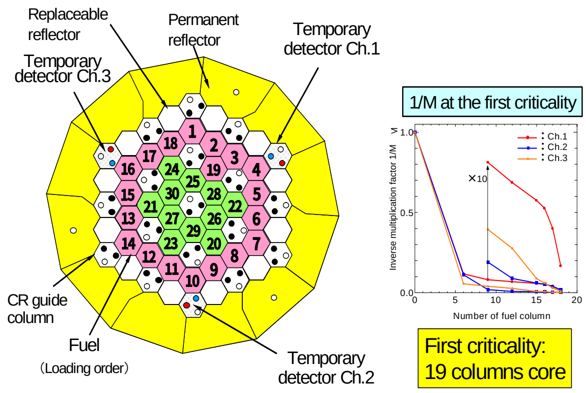


Fig. 8 First criticality at annular form of fuel loading in HTTR (horizontal cross section of core & fuel loading order).

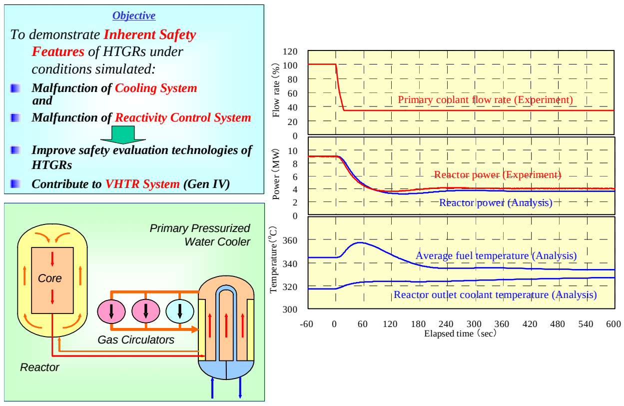


Fig. 11 Safety demonstration test using HTTR (Coolant flow reduction test).

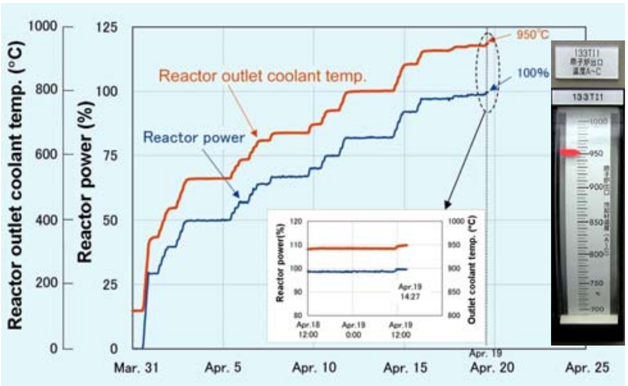


Fig. 9 Achievement of 950°C at reactor outlet of HTTR JAERI.

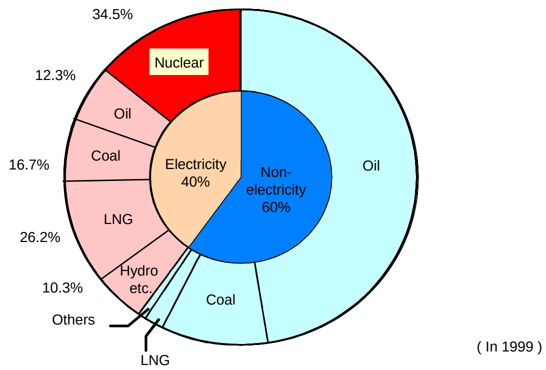


Fig. 12 Composition of primary energy supply in Japan.

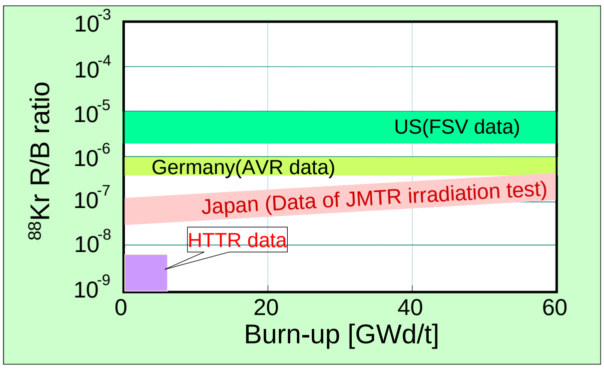


Fig. 10 Performance of coated particle fuel (by JAERI)

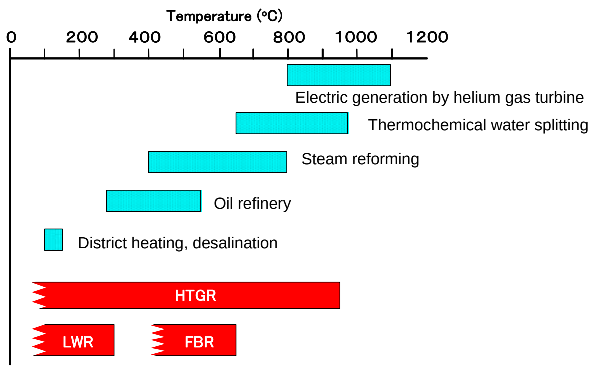


Fig. 13 Temperature ranges in various industries.

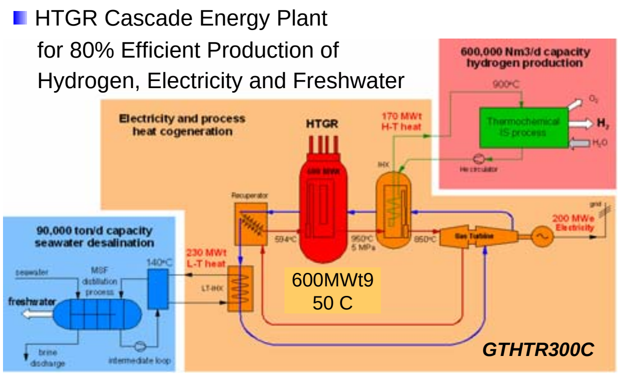


Fig. 14 GTHTR300C for multipurpose use by JAERI.

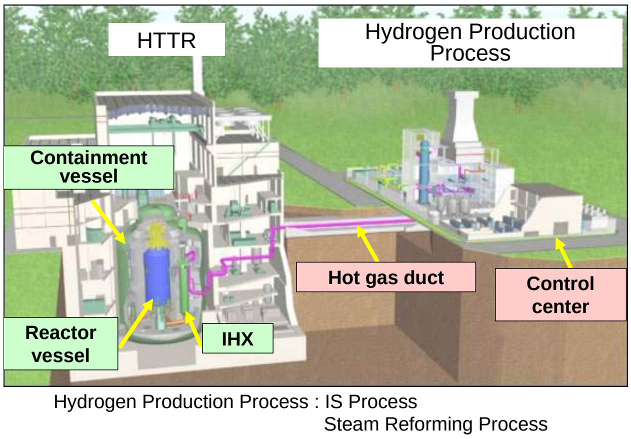


Fig. 17 HTTR hydrogen production demonstration programme.

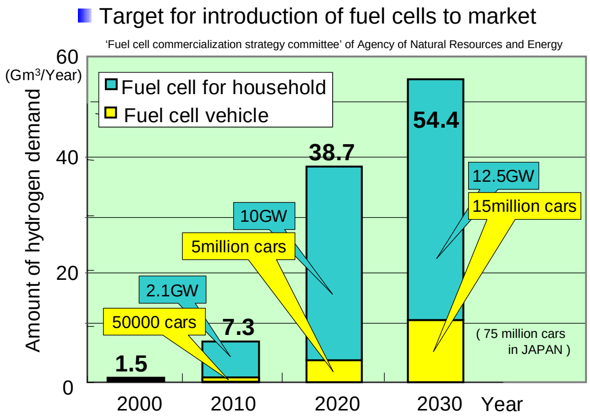


Fig. 15 Hydrogen utilization programme in Japan (by METI).

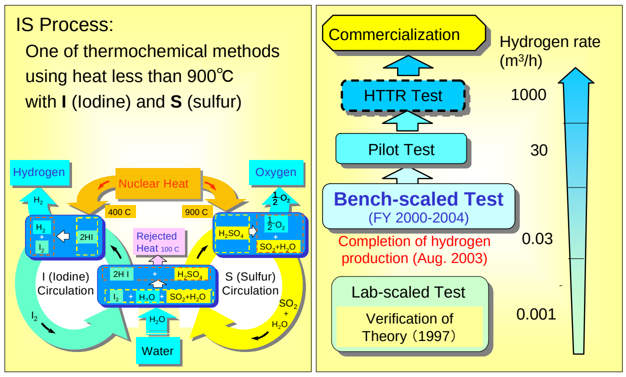


Fig. 18 Thermochemical hydrogen production process: IS process (JAERI).

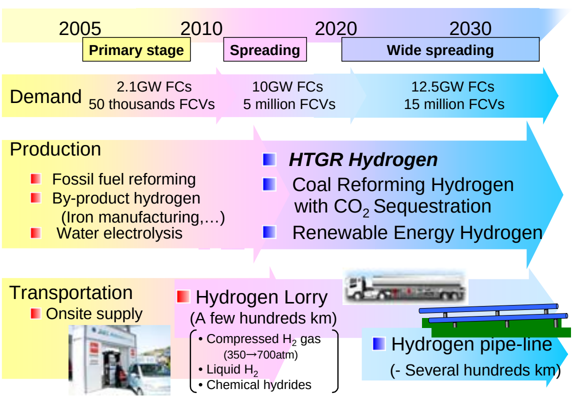


Fig. 16 Future plants for hydrogen production (METI).

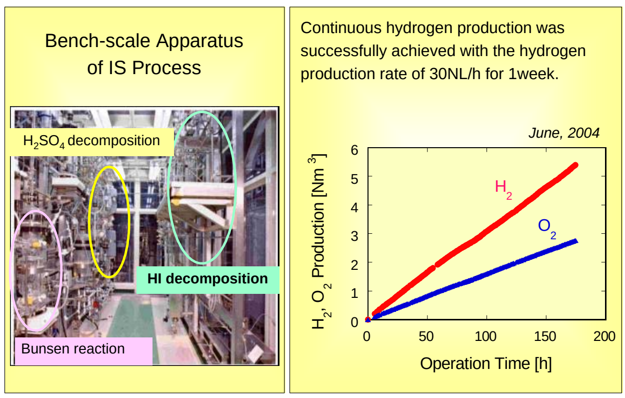


Fig. 19 Bench-scaled test result of IS process in JAERI.

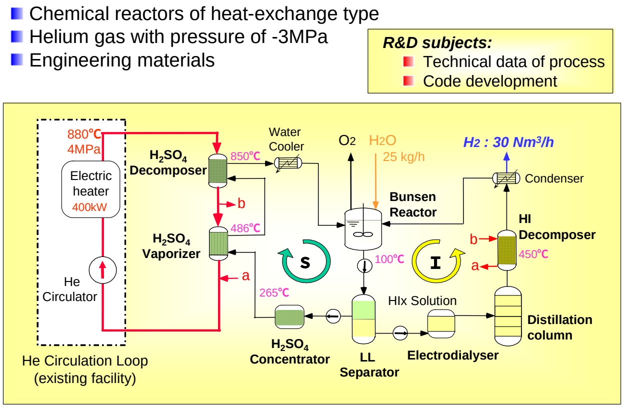


Fig. 20 IS process pilot test in JAERI.

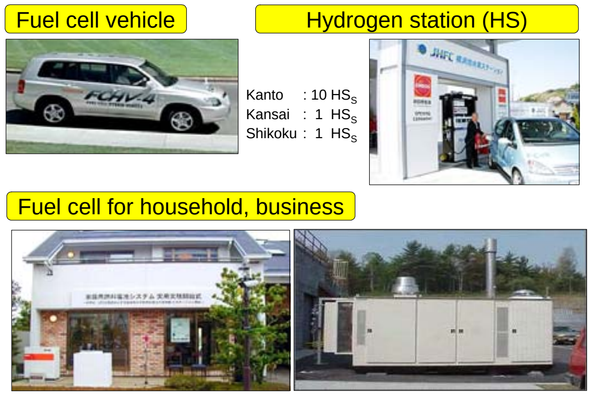


Fig. 23 Current status of hydrogen utilization and infrastructure.

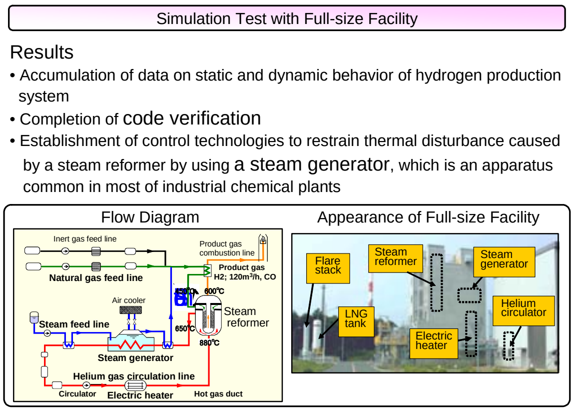


Fig. 21 System integration tests with steam reforming in JAERI.

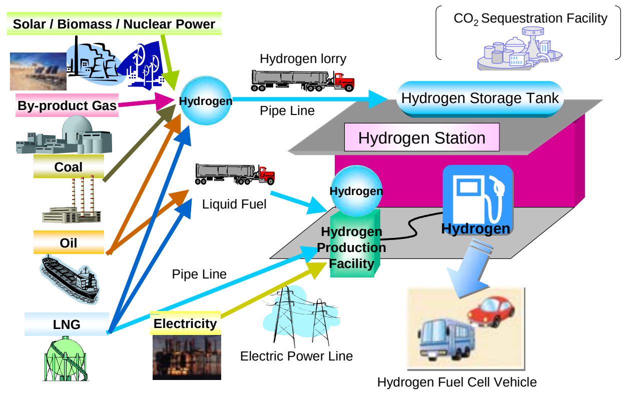


Fig. 24 Transportation of hydrogen.

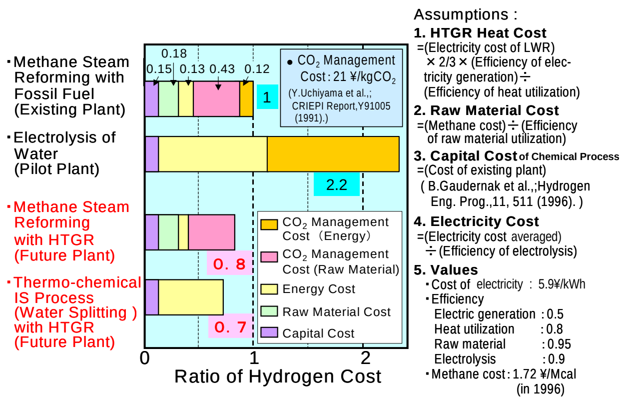


Fig. 22 Prospect of cost including CO_2 management cost (by JAERI).