

Hydrogen permeation through heat transfer pipes made of Hastelloy XR during the initial 950 °C operation of the HTTR

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Abstract

The permeation of hydrogen isotopes through the Hastelloy XR high-temperature alloy adopted for the heat transfer pipes of the intermediate heat exchanger in the HTTR, is one of the concerns in the hydrogen production system, which will be connected to the HTTR in the near future. The hydrogen permeation between the primary and secondary coolants through the Hastelloy XR was evaluated using the actual hydrogen concentrations observed during the initial 950 °C operation of the HTTR. The hydrogen permeability of the Hastelloy XR was estimated conservatively high as follows. The activation energy E_0 and pre-exponential factor F_0 of the permeability of hydrogen were $E_0 = 65.8$ kJ/mol and $F_0 = 7.8 \times 10^{-9} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$, respectively, in the temperature range from 707 to 900 K.

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1. Introduction

The HTTR (High Temperature Engineering Test Reactor) [1], which is the first high-temperature gas-cooled reactor in Japan, attained its maximum reactor-outlet coolant temperature of 950 °C in April 2004 [2]. Although Germany's AVR reached its reactor coolant temperature of 950 °C inside its

reactor vessel in the past [3], the HTTR was the first in the world to deliver 950 °C coolant helium outside the reactor vessel. The reactor-outlet coolant temperature of 950 °C makes it possible to extend high-temperature gas-cooled reactor use beyond the field of electric power. Also, highly effective power generation with a high-temperature gas turbine [4] becomes possible, as does hydrogen production from water [5] using high-temperature gas-cooled reactors. The achievement of 950 °C will be a major contribution to the actualization of producing hydrogen from water using high-temperature gas-cooled reactors.

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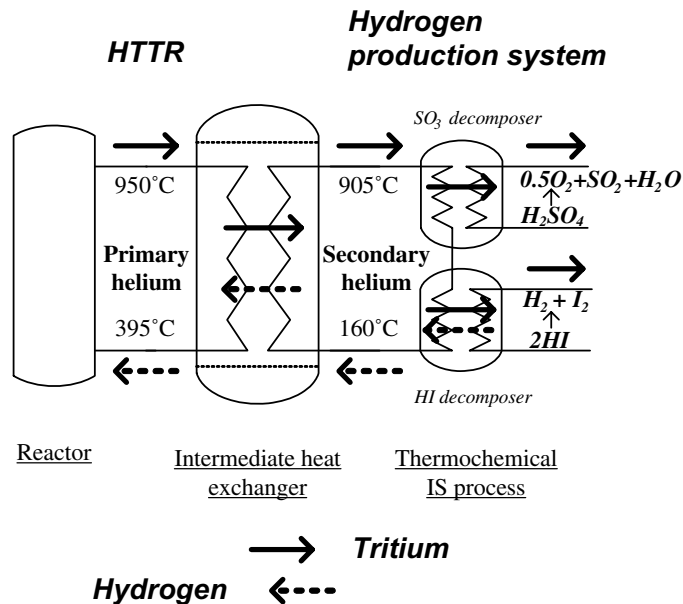
Nomenclature

C_{ps}	hydrogen concentration at the helium purification system [m^3/m^3 (STP)]	Q_{gen}	generated hydrogen in the cooling system [m^3/s (STP)]
d_o	outer diameter of pipe (m)	Q_{rem}	removed hydrogen by the helium purification system [m^3/s (STP)]
d_i	inner diameter of pipe (m)	R	gas constant (J/mol K)
E_0	activation energy (kJ/mol)	T	temperature (K)
F_0	pre-exponential factor [m^3 (STP)/($m \text{ s Pa}^{0.5}$)]	<i>Subscript</i>	
F_{ps}	flow rate of helium purification system [m^3/s (STP)]	1	primary
K_p	permeability [$m^3(\text{STP})/(m \text{ s Pa}^{0.5})$]	2	secondary
L	length of the pipe (m)	in	inlet
P	hydrogen partial pressure (Pa)	out	outlet
Q_p	permeated hydrogen through the heat transfer pipe [m^3/s (STP)]		

It is planned to connect a hydrogen production system based on the thermochemical water-splitting IS process [6] to the HTTR in the near future. Prior to coupling the reactor and chemical plant, out-of-pile tests [7] and design studies are being performed to confirm the controllability, operability, and safety under simulated operational conditions.

The permeation of tritium is one of the serious concerns with regard to release of the radioactive substance from the HTTR hydrogen production

system. Fig. 1 shows the tritium and hydrogen transport processes in the candidate HTTR hydrogen production system. The tritium is generated from several sources in the primary system of the HTTR [8]. Unlike other nuclear reactors, the HTTR has graphite components containing lithium as an impurity in the reactor core and its coolant gas of helium contains ^3He isotope, both of which generate tritium. The inventories of these sources have their maxima in the initial period of reactor operation



and consequently, due to burning-up, the amount of generated tritium decreases during reactor operation. Further sources are boron in the control-rods and uranium in the fuel-particles.

Initially, tritium, which is generated in the reactor, permeates a heat transfer tube of an intermediate heat exchanger (IHX), which is a helium/helium heat exchanger, and transfers from the primary cooling system to the secondary cooling system. Finally, with no countermeasures, e.g., chemistry control, oxide film control, it is possible to permeate catalyst pipes of an HI decomposer in the IS process and to mix with the hydrogen product.

The hydrogen produced in the IS process permeates the catalyst pipes and the heat transfer pipes of the IHX in the opposite direction to that of permeation of tritium. Therefore, it is expected that hydrogen transfers to the primary cooling system and mixes in the helium coolant. This extra source of hydrogen produced by the IS process makes it more complicated to control the hydrogen concentration within 3 volume-ppm, which is specified in the operation procedures of the HTTR [9]. In order to connect the hydrogen production system to the HTTR, it is necessary to evaluate quantitatively the amount of hydrogen that permeated through the IHX and the iodine cycle of the IS process.

Hastelloy XR [10], which is a nickel-based super alloy developed by JAEA and Mitsubishi Materials Co., is the material approved as a high-temperature structural material for use in the normal operation condition of 950 °C. The chemical composition of Hastelloy XR is shown in Table 1. It is used as the material of the heat transfer pipes of the IHX. Not only the amount of hydrogen isotopes that permeated through the IHX but also the amount of tritium mixing in the product hydrogen must be evaluated quantitatively in order to make a safety case for the HTTR hydrogen production system.

The permeation rate of hydrogen isotopes through a solid metal is an overall rate of transfer of the permeating species from a fluid on one side to the other side. Since this is a serial, multi-step

process, the overall rate is governed most strongly by the slowest step in the transport process. According to the previous studies [11–13], the overall permeation rate will depend upon the hydrogen partial pressure and the surface condition of the solid metal. The permeation process in a comparatively higher hydrogen partial pressure region is mostly governed by diffusion in the solid metal and the amount of hydrogen that permeated is in proportion to the difference of the square roots of the higher side and lower side hydrogen partial pressures. Also, the permeation process is governed by chemical adsorption in the case of very low hydrogen partial pressure. The amount of permeated hydrogen is in proportion to the difference of the first power of the hydrogen partial pressure in this case. When an oxide film is formed on the surface of the metal, hydrogen diffuses as molecule inside the oxidized film because the hydrogen molecule adsorbs on the surface without dissociation. The value of the index for hydrogen partial pressure is dependent on the existence and state of the oxide film, increasing as the oxide film gets thicker.

Prior to the construction of the HTTR hydrogen production system, some R&D had been performed to observe the permeation rate of the Hastelloy XR [14]. The results show that the rate-limiting step of the hydrogen transport process in the hydrogen partial pressure region of the HTTR hydrogen production system as well as the HTTR is diffusion in the solid metal and the amount of hydrogen that permeated can be estimated using the difference of the square roots of the higher side and lower side hydrogen partial pressures.

The permeability of hydrogen is calculated by use of the Arrhenius equation, and depends upon a pre-exponential factor and the activation energy. The permeability of hydrogen and its isotopes through high-temperature alloys was reported previously, i.e., Incolloy 800 [11,15–17], Hastelloy N [11], Hastelloy X [17,18], and Hastelloy XR [14]. Since those measurements performed previously were based on the laboratory-scale, we have evalu-

Table 1
Specification for the chemical composition of Hastelloy XR [10]

Range	Elements															
	C	Mn	Si	P	S	Cr	Co	Mo	W	Fe	Ni	B	Al	Ti	Cu	
<i>Chemical compositions (wt%)</i>																
Max.	0.15	1	0.5	0.04	0.03	23	2.5	10	1	20	Balance	0.01	0.05	0.03	0.5	
Min.	0.05	0.75	0.25	–	–	20.5	–	8	0.2	17	Balance	–	–	–	–	

ated the actual hydrogen permeation in a high-temperature gas-cooled reactor in this study.

The HTTR started its operation with a lower-temperature commissioning test in 1997. It is thought that some oxide layers were formed on the primary and secondary surfaces of the metal of the heat transfer tubes of the IHX. The concentrations of hydrogen in the primary and secondary helium coolants were measured continuously by using helium sampling systems. The permeability of hydrogen through the HTTR heat transfer tubes covered with the oxide layers was evaluated using hydrogen concentration data in the primary and secondary cooling systems and quantity of hydrogen removed from the primary and secondary helium coolants by the helium purification systems. This paper describes the preliminary evaluation results of the permeability of hydrogen through the Hastelloy XR, which is adopted for the heat transfer tubes of the IHX, by using hydrogen concentration data observed during the first 950 °C operation of the HTTR.

2. Summary of the HTTR

The detailed HTTR design has already been reported elsewhere [1] and so only the equipment

relevant to hydrogen permeation is described in this chapter.

2.1. Main cooling system

The cooling system of the HTTR, as shown in Fig. 2, consists of a main cooling system operating at normal operation; and an auxiliary cooling system and a vessel cooling system, which are the engineered safety features, operating after a reactor scram to remove residual heat from the core. The main cooling system, which consists of a primary cooling system, a secondary helium-cooling system, and a pressurized water-cooling system, removes heat generated in the core and dissipates it to the atmosphere by a pressurized water air-cooler. The main components of the primary cooling system are an IHX as shown in Fig. 3, a primary pressurized water cooler (PPWC), a secondary pressurized water cooler (SPWC), and a primary concentric hot-gas-duct. The helium gas primary coolant from the reactor at 950 °C maximum flows inside the inner pipe of the primary concentric hot-gas-duct to the IHX and PPWC. The primary helium is cooled to about 400 °C by the IHX and PPWC and returns to the reactor flowing through the annulus between the inner and outer pipes of the primary concentric hot-gas-duct.

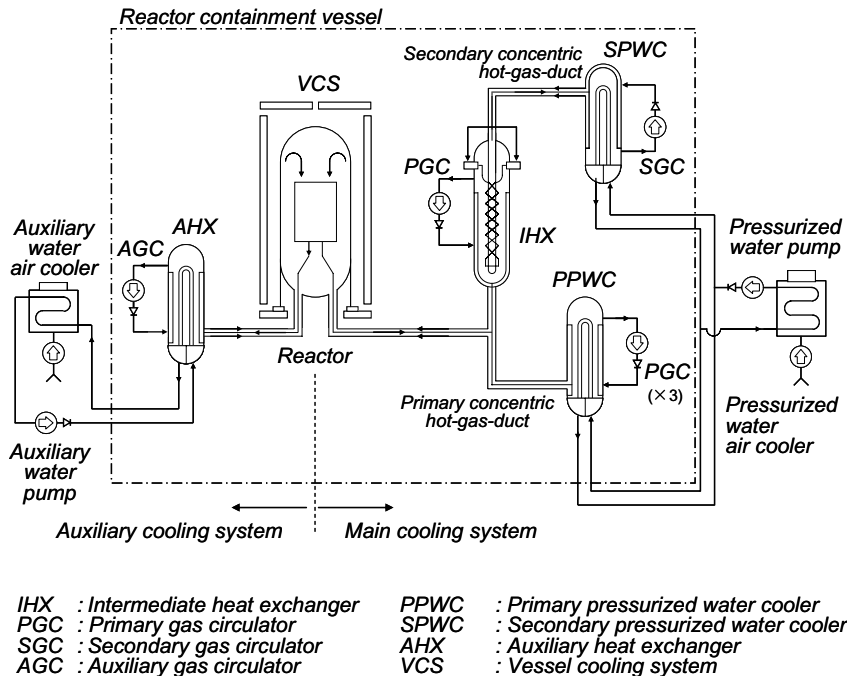


Fig. 2. Cooling system of the HTTR.

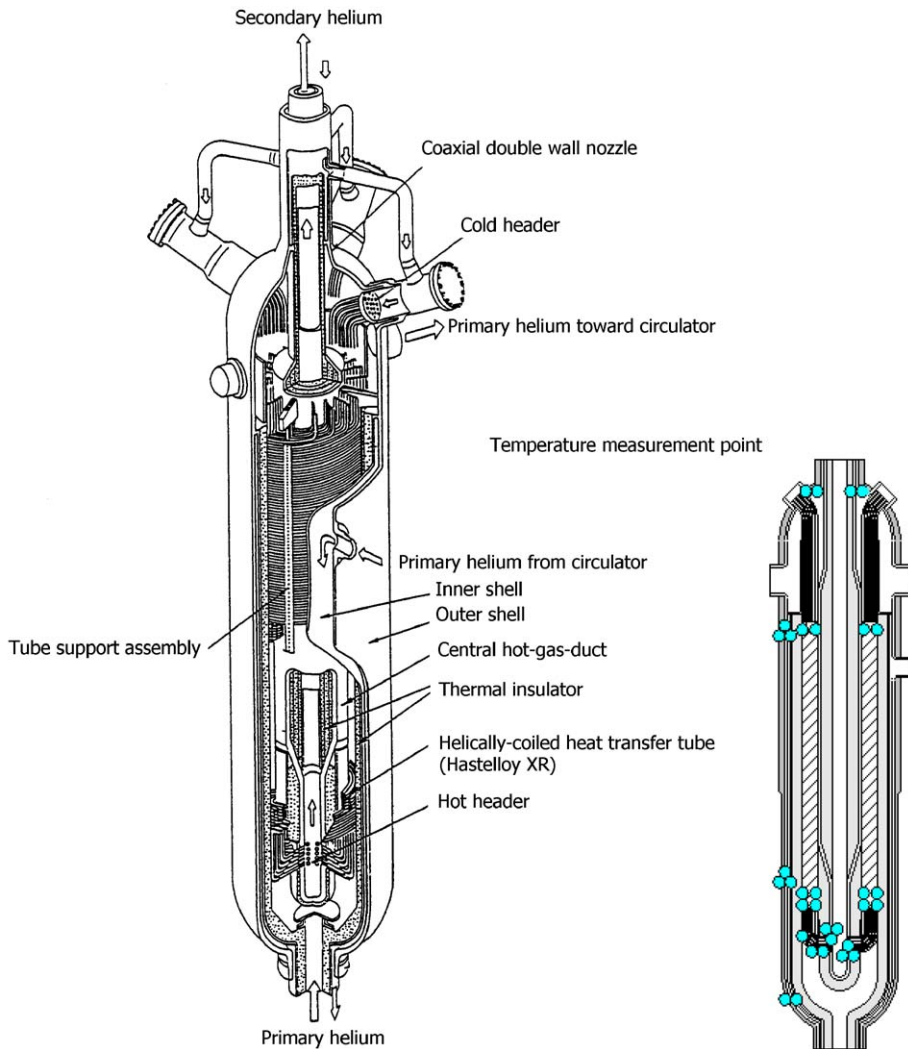


Fig. 3. Bird's eye view of the intermediate heat exchanger and its temperature measurement points.

The HTTR has two operation modes. One is the single-loaded operation mode using only the PPWC for the primary heat exchange. Almost all the basic performances of the HTTR system have been confirmed in the single-loaded operation mode. The other is the parallel-loaded operation mode using the PPWC and IHX. In the single-loaded operation mode the PPWC removes 30 MW of heat, while in the parallel-loaded operation mode the PPWC and IHX remove 20 and 10 MW, respectively. It is planned to connect the IS process to the secondary helium gas at the IHX. The auxiliary cooling system, whose main components are an auxiliary helium-cooling system, an auxiliary water-cooling system, and a concentric hot-gas-duct, is in stand-by during normal operation and starts up to remove

residual heat after a reactor scram. The vessel cooling system cools the biological concrete shield surrounding the reactor pressure vessel at normal operation, and removes heat from the core by natural convection and radiation outside the reactor pressure vessel under 'accident without forced cooling' conditions such as a rupture of the primary concentric hot-gas-duct, when neither the main cooling system nor the auxiliary cooling system can cool the core effectively.

2.2. Helium purification system

The helium purification systems are installed in the primary cooling system and the secondary in order to reduce the quantity of chemical impurities

such as hydrogen, carbon monoxide, water vapour, carbon dioxide, methane, oxygen, and nitrogen. The primary helium purification system is mainly composed of a pre-charcoal trap, an inlet heater, two copper oxide fixed beds, coolers, two molecular sieve traps, two cold charcoal traps and compressors. Fig. 4 shows the flow diagram of the primary helium purification system. The flow diagram of the secondary helium purification system is almost the same as that of the primary system except for the absence of a pre-charcoal trap.

The primary helium gas is introduced into the helium purification system through the auxiliary cooling system and the purified helium gas returns to the auxiliary cooling system and stand-pipes. The flow rate of the helium purification system is determined considering the following requirements:

- to satisfy the concentration limit of impurity to limit oxidation of core graphite structures (Limited impurity concentration values are shown in Table 2),
- to purify >10% of the helium inventory in the primary cooling system in 1 h, and,
- to purge the stand-pipes.

The helium purification system has three kinds of traps for reducing chemical impurities. Each trap has two identical systems for reliability of plant operation. The first trap is a copper oxide fixed bed where hydrogen and carbon monoxide are oxidized to water vapour and carbon dioxide, respectively.

Table 2

Upper limit of impurity concentrations in the primary coolant at a reactor-outlet coolant temperature above 800 °C

Impurity	Concentration (volume ppm)
H ₂	3.0
CO	3.0
H ₂ O	0.2
CO ₂	0.6
CH ₄	0.5
N ₂	0.2
O ₂	0.04

It is kept at a temperature of approximately 280 °C during its operation. The second trap is a molecular sieve trap where water vapour and carbon dioxide are removed by adsorption. The third trap is a cold charcoal trap where oxygen, nitrogen, methane and noble gases are removed by adsorption; it is kept at a temperature of approximately –195 °C. The flow rate in the primary helium purification system is 200 kg/h. The flow rate through the cold charcoal trap is 50 kg/h with a by-pass flow for the rest of the gas. When the efficiency of the traps decreases during operation, the traps are changed manually to stand-by traps. The deteriorated traps can be used repeatedly after regeneration.

The flow rate of the secondary helium purification system is 10 kg/h, which is determined in a similar manner to that of the primary helium purification system. The design of the helium purification system takes account of the experience obtained in the HENDEL loop of JAEA [19].

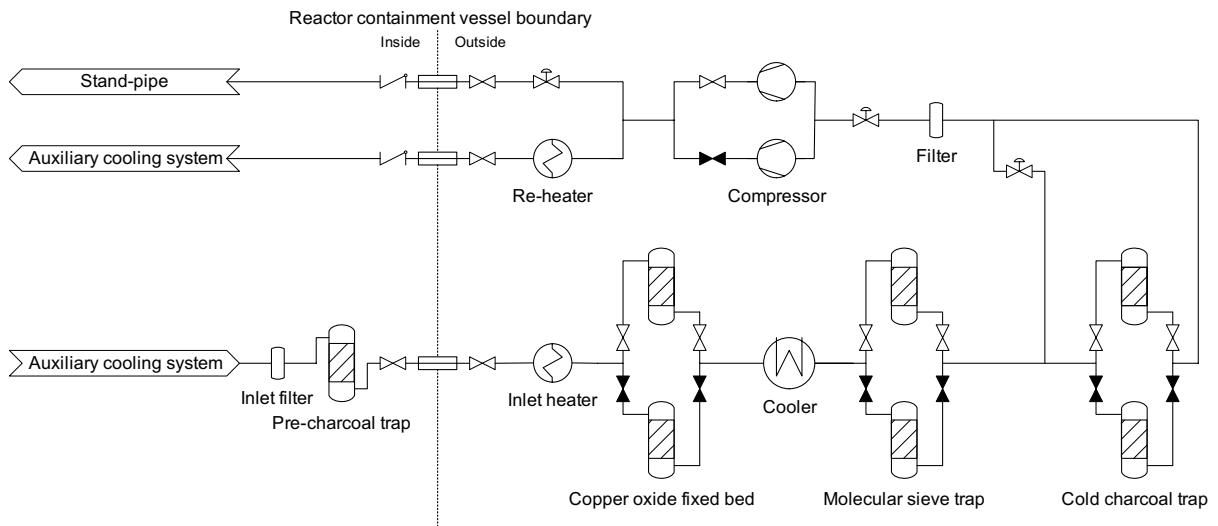


Fig. 4. Schematic diagram of the primary helium purification system of the HTTR.

2.3. Helium sampling system

The helium sampling systems detect chemical and radioactive impurities in the primary cooling system and secondary helium-cooling system. The concentration of chemical impurities: hydrogen, carbon monoxide, water vapour, carbon dioxide, methane, nitrogen, and oxygen, are measured with the gas chromatograph mass spectrometers. The primary helium sampling system, consisting of sampling equipment, a carrier-gas-supply system, and a standard-gas-supply system, automatically transmits the impurity concentration measurement to the main control room, as does the secondary helium sampling system.

The purpose of the primary helium sampling system is:

- to monitor the chemical impurity level for the purpose of avoiding core graphite oxidation and carbon deposits, as well as the carburizing and decarburizing of the Hastelloy XR in the intermediate heat exchanger,
- to detect the rupture of a heat exchanger tube in the primary pressurized water cooler and auxiliary heat exchanger, and,
- to monitor the performance of the traps in the primary helium purification system.

The sampling locations of impurities except water vapour for the primary circuit are the inlet and outlet of the reactor, the inlet and outlet of the primary helium purification system, and the inlet and outlet of the cold charcoal trap. Two detectors are installed for detecting water vapour. Sampling locations for the No. 1 detector are: the inlet to the reactor, the inlet and outlet of the primary purification system, and the inlet to the cold charcoal trap. No. 2 detector sampling locations are: the reactor outlet, the outlet of the primary pressurized water cooler, the outlet of the primary helium gas circulator for the primary pressurized water cooler and the intermediate heat exchanger.

3. Evaluation of the permeating hydrogen

As the preliminary evaluation of hydrogen permeation of the HTTR, the activation energy E_0 and pre-exponential factor F_0 were evaluated using hydrogen concentration data observed during the first 950 °C operation performed from March to June 2004.

The mass balance of hydrogen in the primary and secondary helium system is shown in Fig. 5. In this figure, the permeation of hydrogen through the Hastelloy XR, which is the material of the heat transfer pipes of the IHX, is considered.

The generated hydrogen in the primary cooling system can be traced back to a chemical reaction between water as an impurity emitted from the core and insulator at the hot-gas-duct, and graphite as a core material [20,21]. The chemical reaction equation of water and graphite is written as follows:



In the secondary helium-cooling system, hydrogen was emitted from an insulator used at the secondary hot-gas-duct. Since the concentration of hydrogen in the primary cooling system was smaller than that of the secondary helium-cooling system during the first 950 °C operation, hydrogen permeated from the secondary toward the primary in this case. Thus, the mass balance of hydrogen in both of the cooling systems can be written as Eqs. (2) and (3)

$$Q_{1\text{gen}} - Q_{1\text{rem}} + Q_p = 0, \quad (2)$$

where $Q_{1\text{gen}}$ is the amount of hydrogen generated in the primary cooling system, $Q_{1\text{rem}}$ is the amount of

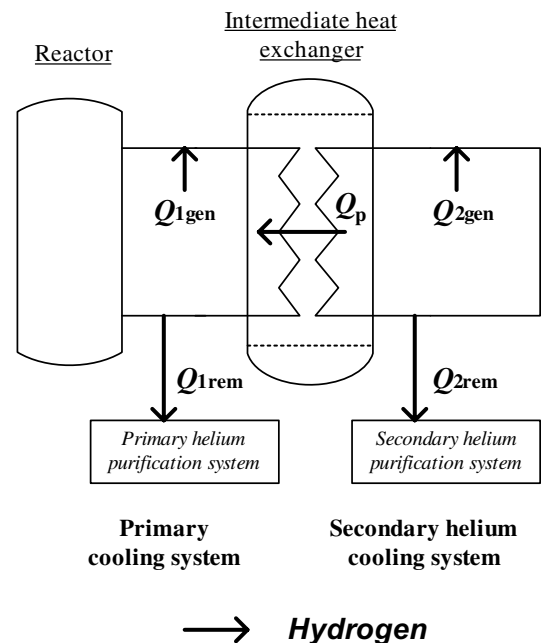


Fig. 5. Mass balance of hydrogen in the primary and secondary helium-cooling systems, considering hydrogen permeation through Hastelloy XR, which is the material of the heat transfer pipes of the intermediate heat exchanger of the HTTR.

hydrogen removed by the primary helium purification system, and Q_p is the amount of hydrogen that permeated through the heat transfer pipes of the IHX, and

$$Q_{2\text{gen}} - Q_{2\text{rem}} - Q_p = 0, \quad (3)$$

where $Q_{2\text{gen}}$ is the amount of hydrogen generated in the secondary helium-cooling system, $Q_{2\text{rem}}$ is hydrogen removed by secondary helium purification system. The amount of hydrogen generated in the primary cooling system $Q_{1\text{gen}}$ depends upon the Eq. (1).

The hydrogen is slightly removed by a copper oxide fixed bed in the primary helium purification system, while the hydrogen is supplied continuously by a reaction between water and graphite. Thus, the concentration of hydrogen depends upon that of water. During the first 950 °C operation in the HTTR, water concentration decreased slowly by adsorption at the molecular sieve bed in the primary helium purification system [20]. The behaviour of hydrogen and water in the primary cooling system observed during the initial 950 °C operation by a parallel-loaded operation-mode is shown in Fig. 6.

The amount of hydrogen removed in the primary cooling system $Q_{1\text{rem}}$ and in the secondary cooling system $Q_{2\text{rem}}$ can be calculated using the concentration data from the inlet and outlet of the helium purification system. During the parallel-loaded operation, there was permeation between the primary and secondary cooling systems, and generation and removal occurred in both of the cooling systems.

In order to estimate the permeation of hydrogen conservatively higher, the generated hydrogen is not considered in this evaluation (the value of $Q_{1\text{gen}}$ was set equal 0). Thus, the amount of hydrogen that per-

meated was equal to that of the hydrogen removed by the helium purification system, as shown in Eq. (4)

$$Q_p = F_{1\text{ps}}(C_{1\text{ps in}} - C_{1\text{ps out}}), \quad (4)$$

where $F_{1\text{ps}}$ is the flow rate of primary helium purification system, and $C_{1\text{ps in}}$ and $C_{1\text{ps out}}$ are the hydrogen concentrations at the inlet and outlet of the primary helium purification system, respectively.

The fluid flow through the primary helium purification system is measured with orifice plates, nozzles, and venture tubes which were produced in accordance with Japanese industrial standard. Its accuracy is $\pm 1.0\%$ of rated flow. The accuracy of $F_{1\text{ps}}$ is $\pm 2.1\%$ which is calculated from the accuracy not only of the orifice plates, nozzles, and venture tubes, but also of the instrumentation system of the primary helium purification system. The detection sensitivity of impurity concentrations measured with the gas chromatograph mass spectrometers of the helium sampling system depends upon the kind of impurity gas, and for hydrogen it is 0.08 volume ppm. The accuracy of this system is limited within $\pm 3\%$ and is confirmed by inspections before each reactor operation.

For the initial 950 °C operation in the HTTR, the temperature-rise was carried out in stages with the reactor-outlet coolant temperature held steady for more than 10 h at approximately 650, 700, 750, 800, 850, 890, 928, and 941 °C each. The average temperatures of the intermediate heat transfer pipes of the IHX were 707, 741, 774, 808, 840, 862, 890, and 900 K, respectively. These eight temperatures were utilized for the evaluation of hydrogen permeation. The amount of hydrogen that permeated through the heat transfer pipes of the IHX Q_p is calculated by Eq. (5) [22]

$$Q_p = \frac{2\pi L K_p}{\ln\left(\frac{d_o}{d_i}\right)} (\sqrt{P_2} - \sqrt{P_1}), \quad (5)$$

where L is the length of the heat transfer pipe, d_o and d_i are the outer and inner diameters of the heat transfer pipe, respectively, and P_1 and P_2 are the primary and secondary hydrogen partial pressures, respectively. The permeability of hydrogen K_p is calculated by the following Eq. (6) that was obtained from Eq. (5).

$$K_p = \frac{Q_p \ln\left(\frac{d_o}{d_i}\right)}{2\pi L (\sqrt{P_2} - \sqrt{P_1})}. \quad (6)$$

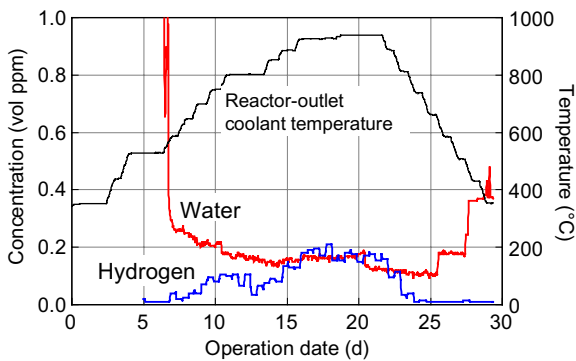


Fig. 6. Hydrogen and water behaviour in the primary cooling system during the first 950 °C operation in the parallel-loaded operation-mode in the HTTR.

The permeability of hydrogen was estimated using the hydrogen concentration and pressure of the primary and secondary helium; inner and outer diameters, and lengths of heat transfer pipes of the IHX. The flow velocities and temperatures of the primary and secondary helium purification systems were also utilized for the estimation of the amount of removed hydrogen.

The hydrogen permeability estimated in this evaluation is shown in Fig. 7. The activation energy E_0 and pre-exponential factor F_0 are estimated as follows:

$$K_p = F_0 \exp\left(\frac{-E_0}{RT}\right), \quad (7)$$

where R is the gas constant, and T is the absolute temperature of the pipe.

A solid line shows the average value of hydrogen permeability obtained from eight different hydrogen partial pressures. The average activation energy E_0 and pre-exponential factor F_0 were approximately 65.8 kJ/mol and $7.8 \times 10^{-9} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$, respectively.

Alternatively, it could be argued from the view point of chemical reaction in the HTTR core. While the reactor-outlet coolant temperature was above 800 °C, water concentration little decreased because water was continuously supplied from the emission traced back to the graphite core and insulator at concentric hot-gas-duct. Since the water concentration was kept steady in high-temperature circumstances, the helium purification system surely

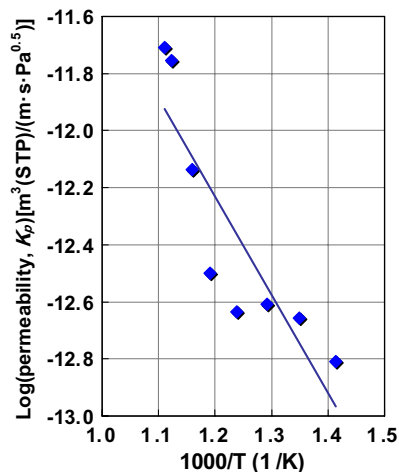


Fig. 7. Hydrogen permeability of the Hastelloy XR estimated conservatively using hydrogen concentration data observed during the first 950 °C operation of the HTTR.

removed water at molecular sieve traps by the adsorption. The fact that the water was removed by primary helium purification system suggests that the same volume of hydrogen was generated in the primary cooling system. Thus, it is reasonable to suppose that the sum of the amount of hydrogen that permeated from secondary helium cooling system and that of hydrogen that was generated by a chemical reaction at the reactor core is the amount of hydrogen existing in the primary circuit. In order to estimate the hydrogen permeability accurately, the volume of hydrogen that permeated was calculated by using the difference between the volume of removed water and that of removed hydrogen. The result is shown as a red line in Fig. 8. The estimated activation energy E_0 was 66.3 kJ/mol which was considerably similar to both of two results. The pre-exponential factor F_0 was $4.8 \times 10^{-9} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$.

Measurements of for hydrogen permeation of Hastelloy XR had been performed previously by JAEA [14]. One of the results, from an examination performed with no oxide film formation, is shown in Fig. 8. The previously estimated value of the activation energy E_0 of the Hastelloy XR for hydrogen permeation was 67.2 kJ/mol [14]. It is confirmed that the activation energies from the present work are considerably same comparing with that of previous work. It is thought that the difference may decrease after obtaining more data. On the other

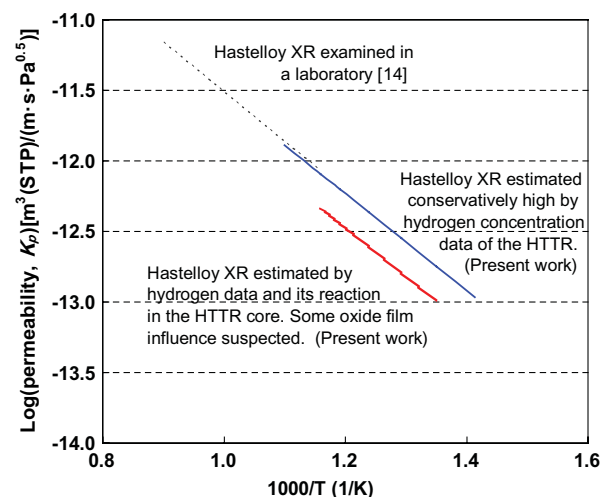


Fig. 8. Comparison of present work and previous work for hydrogen permeability of Hastelloy XR. (For interpretation of the references in colour in this figure legend, the reader is referred to the web version of this article.)

hand, the previous value of the pre-exponential factor F_0 was $(1.0 \pm 0.2) \times 10^{-8} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$ [14] and it decreased to $(4.7 \pm 0.4) \times 10^{-9} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$ due to the formation of oxidized film on the surface of the Hastelloy XR after about 140 h heating at 600–800 °C under the condition of helium gas atmosphere and water concentration of less than 1 ppm [14]. The previous value of the pre-exponential factor F_0 for the Hastelloy XR with oxidized film is very close to the present value ($4.8 \times 10^{-9} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$), which implies some oxidized film was formed at the surface of the heat transfer tubes of the IHX.

We are planning some research and development to prevent the hydrogen and tritium permeation through heat transfer tubes of the IHX by controlling the formation of oxidized film layer with adjusting the coolant chemistry. In the near future, the HTTR will be connected to the hydrogen production system and then the secondary helium purification system will be modified in order to control the hydrogen and tritium concentrations. The results of this study are expected to contribute to make some design criteria for the hydrogen and tritium permeations.

4. Concluding remarks

The permeation of hydrogen isotopes through Hastelloy XR, the high-temperature alloy adopted for the heat exchanger pipes of the intermediate heat exchanger in the HTTR, is one of the concerns for the hydrogen production system which will be connected to the HTTR in the near future. An evaluation of hydrogen permeation between the primary and secondary coolants through the Hastelloy XR was performed using the hydrogen concentration data observed during the initial 950 °C operation of the HTTR. The hydrogen permeability of the Hastelloy XR was estimated conservatively high as follows. The activation energy E_0 and pre-exponential factor F_0 of the permeability of hydrogen were $E_0 = 65.8 \text{ kJ/mol}$ and $F_0 = 7.8 \times 10^{-9} \text{ m}^3(\text{STP})/(\text{m s Pa}^{0.5})$, respectively, in a temperature range from 707 to 900 K.

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