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Journal of Nuclear Materials 329–333 (2004) 183–187
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Experimental evaluation of tritium permeation through stainless steel tubes of heat exchanger from primary to secondary water in ITER

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Abstract

Tritium permeation through heat exchanger from primary cooling water to secondary cooling water has been investigated experimentally with SS316L heat exchanger under simulated ITER (international thermonuclear experimental reactor) operation condition in order to establish the tritium permeation evaluation method through the heat exchanger. As the result, the permeation rate of aqueous tritium was found to be about three orders smaller than that of the gaseous tritium. Tritium permeation through the heat exchanger in ITER was then evaluated, and it was revealed that total tritium permeation amount based on obtained aqueous permeability was about one order less than that with the former method with the gaseous permeability and putting the permeation reduction factor as 1000. Evaluated tritium permeation amount into secondary water during 20 years was quite small, which could be considered as negligible from the safety viewpoint.

1. Introduction

Evaluation of tritium concentration not only in primary cooling water but also in secondary cooling water is one of the key issues for the safety of fusion reactors such as international thermonuclear experimental reactor (ITER), since the secondary water faces the environment directly.

Tritium concentration in the primary cooling water increases by the permeation of tritium implanted into the plasma facing components (PFC) or generated by the nuclear reaction in the PFC [1–4].

The secondary cooling water is parted from the primary cooling water by the heat exchanger with large surface area and thin wall thickness, through which tritium may permeate. It is expected that the tritium permeation through a metal from water to water is much smaller than that from gas to gas, and it has been proposed to evaluate tritium permeation from water to water with a combination of permeation from gas to gas and the proper permeation reduction factor [5]. Because there are only a few data on the tritium permeation through metal from water to water, permeation reduction factor should be selected conservatively for the estimation of the safety evaluation. Therefore, the current tritium permeation through the heat exchanger is evaluated conservatively in ITER.

In order to clarify the tritium permeation through metal from water to water and the conservativeness of the design of ITER, authors have built an experimental apparatus simulating the heat exchanger and have investigated the steady-state tritium permeation from high tritium concentration primary water to natural secondary water under simulated ITER conditions. This report presents the result of the experiment and also presents the result of the evaluation of tritium permeation into the secondary cooling water carried out under the ITER operation condition on the basis of the experimental results.

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doi:10.1016/j.jnucmat.2004.04.009

2. Experimental

Fig. 1 shows the flow diagram of the experimental apparatus for simulating the tritium permeation through the heat exchanger. This apparatus is designed as simulating the operation conditions of the heat exchanger of ITER (maximum temperature of 510 K). The experimental chamber involves 37 SS316L tubes of 6 mm outer diameter, 0.5 mm thickness, and 0.3 m length assembled in parallel, and the total surface area of heat exchange tubes for tritium permeation is 0.2 m^2. Those SS316L tubes are those pre-heated at 1353 K for 1.5 h and rapid cooled as the solution heat treatment without surface polishing. For the periodical measurement of the tritium concentration in the secondary water during the experiment, a bypass is prepared in the circulation path of the secondary water. The pressure of both primary and secondary water can be adjusted individually by pressure-controlled helium gas through bellows.

In this permeation experiment, tritiated water of 7.4 TBq/m^3 was filled inside of the tubes as the primary water (total hold up of the primary water was 600 cm^3), and tritium free pure water was filled in the experimental chamber outside of tubes as secondary water. The quality of tritiated and pure water used in this experiment is pH: 7, purity (electric conductivity): <0.1 μS/cm, and dissolved oxygen concentration: about 8.6 mg/L at room temperature, respectively. Whole experimental system, which includes experimental chamber, circulation pump and line for circulation, was kept at 423 K. Pressure of both primary and secondary water was controlled at 0.9 MPa. During the experiment, the secondary water was sampled periodically, and tritium concentration in it was measured by a liquid scintillation counter.

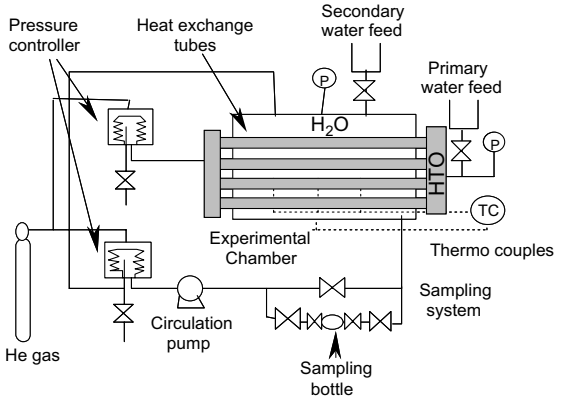

Fig. 1. Conceptual flow diagram of an apparatus simulating the tritium permeation through the heat exchanger.

3. Results and discussion

3.1. Tritium permeation rate through heat exchange tubes

Fig. 2 shows the increase of tritium amount in the secondary cooling water as a function of elapsed time at 423 K. Significant linear increase of tritium amount in the secondary cooling water was observed from 100-h exposure. The time dependence of the permeation flux ($J(t)$) can be expressed in Eq. (3) by solving simple diffusion equation (1) under the constant boundary conditions (2) [6].

$$\frac{\partial C(x,t)}{\partial t} = \frac{\partial^2 C(x,t)}{\partial x^2}, \quad J(t) = -D\frac{\partial C(d,t)}{\partial x}, \tag{1}$$

$$C(x,0) = 0, \quad C(0,t) = C_0, \quad C(d,t) = 0, \tag{2}$$

$$J(t) = -\frac{C_0 D}{d}\left(1 + 2\sum_{n=1}^{\infty}(-1)^n \exp\left(-D\left(\frac{n\pi}{d}\right)^2 t\right)\right), \tag{3}$$

here, $C(x,t)$, $J(t)$, C_0, D and d mean tritium concentration in the material, initial tritium concentration, permeation flux, diffusion coefficient and thickness of tubes, respectively.

Since averaged diffusion coefficient of tritium in SS316L at 423 K is estimated to be about 5×10^{-13} m^2/s [7–9], necessary time until permeation reaches steady state in this case (0.5 mm thick and 423 K) is estimated to be about 100 h, which agrees with experimentally obtained time mentioned above. Therefore, observed tritium permeation after 100 h can be considered as the steady-state behavior.

The tritium permeation rate derived from the observed slope shown in Fig. 2 is 4.2×10^{-3} Bq/m^2/s. In case of permeation through SS316L from gas to gas, the permeability (P_g) of hydrogen is reported as [8]

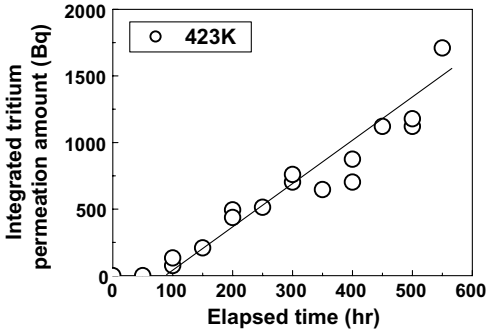

Fig. 2. Time evolution of tritium amount permeated into the secondary water at 423 K.

$$P_g = 2.6 \times 10^{-9} \exp\left(-\frac{62.7\ \text{kJ/mol}}{RT}\right) \quad (\text{m}^3(\text{STP})/\text{m}/\text{s}/\sqrt{\text{Pa}}). \tag{4}$$

The gaseous tritium permeation flux (J) at steady state can be expressed as follows, using P_g given in Eq. (4), partial pressure of gaseous tritium (P) and surface area (A) and thickness (d) of the heat exchanger tubes.

$$J = \frac{A}{d} P_g \sqrt{P}, \tag{5}$$

where A and d of the present experiment are 0.2 m^2 and 0.5 mm, respectively, as mentioned previously. P can be evaluated to be 7.7 Pa by converting the aqueous tritium concentration of 7.4 TBq/m^3 to the gaseous partial pressure of T_2. Inputting those parameters to Eq. (5), 3.6 Bq/m^2/s is derived as the tritium permeation rate in the gaseous case, which is about three orders magnitude larger than observed rate. This difference is attributed to the difference of solution process of tritium into the heat exchange tube. While the solution process of gaseous tritium into the metal is controlled by Sievert's law, which is proportional to the square root of partial pressure of gaseous tritium, that for aqueous tritium into the metal is considered to be controlled by the radicals in water such as T^+ or OT^-, which is proportional to the concentration of radicals in water. In the present case, permeability (P_a) of aqueous tritium can be evaluated to be 2.8×10^{-19} Bq/m/s/C from the obtained permeation rate and the geometrical conditions of experimental apparatus. Here, C is the tritium concentration difference (Bq/m^3) between primary and secondary water.

3.2. Evaluation of tritium permeation through ITER heat exchanger

Based on the permeability obtained in this study, tritium permeation amount from the primary cooling water to the secondary cooling water was evaluated in case of ITER. In ITER, there are three primary cooling water systems, those are the first wall/blanket (PFW/BLK) cooling system, the divertor and limiter (DIV/LIM) cooling system and the vacuum vessel (VV) cooling system. Here, since increase of tritium concentration in the primary cooling water was found to be about 1 GBq/m^3 or less in case of the DIV/LIM and the VV cooling system [4], permeation amount to their secondary cooling water could be negligible. On the other hand, tritium permeation amount into the PFW/BLK primary cooling system (total hold up of the primary cooling water is about 390 m^3 [10]) is estimated to be about 600 TBq through the beryllium first wall via copper cooling block after 20 years operation of ITER based on the permeation analysis using TMAP4 code [11] with the transport parameters for beryllium and copper and operational conditions of ITER shown in Table 1 [4,12–17]. The increase of tritium concentration in the PFW/BLK primary cooling water is evaluated to reach about 1.5 TBq/m^3. Therefore, tritium permeation evaluation from primary to secondary water for the PFW/BLK cooling system was carried out.

Tritium permeation amount into the secondary cooling water (Q_w) was evaluated as follows, using the permeability of water (P_a) obtained in this study

$$Q_w = \frac{A}{d} \int_0^{t_0} P_a (C_P(t) - C_S(t))\, dt, \tag{6}$$

here, the A and d mean surface area and thickness of heat exchange tubes. t_0, $C(t)$ mean evaluation period, tritium concentration in the cooling water. Subscript P and S mean the primary water and the secondary water, respectively.

On the other hand, the tritium permeation amount evaluation using gaseous permeability and the permeation reduction factor (PRF) proposed formerly is as follows [5]

$$Q_g = \frac{1}{\text{PRF}} \frac{A}{d} \int_0^{t_0} P_g \left(\sqrt{P_P(t)} - \sqrt{P_S(t)}\right) dt, \tag{7}$$

here, $P(t)$ means the partial pressure of gaseous tritium assuming all permeated tritium exists as the gaseous form in the cooling system.

In ITER, A and d for the PFW/BLK heat exchanger are designed to be 8130 m^2 and 2 mm, and normal operating temperature is about 420 K [10]. $C(t)$ and $P(t)$ in the primary cooling water of the PFW/BLK were determined by the result of the tritium transport analysis as mentioned above. The PRF is recommended to be 1000 in the proposed evaluation method by Tosti et al. [5].

Fig. 3 shows the evaluated tritium concentration in the primary cooling water of the PFW/BLK and evaluated tritium permeation amount from the primary cooling water to the secondary cooling water through the heat exchanger using Eq. (6). The result of tritium permeation amount evaluated with the gaseous permeability using Eq. (7) is also shown for comparison. As shown in Fig. 3, total tritium permeation amount into the secondary cooling water during 20 years of ITER operation is estimated to be about 0.4 GBq, which is nearly one order less than that estimated with the former method using Eq. (7). Present result assures that the formerly proposed evaluation method using Eq. (7) gives the conservative result. Anyway, the tritium permeation amount to the secondary water is estimated to be quite small which can be considered as negligible from the safety viewpoint.

Further investigation on the influence of dissolved gas such as oxygen or hydrogen in water on the tritium permeation is under considering for more detailed estimation.

Table 1
Tritium transport properties of Be and Cu and tritium source into Be used in the TMAP4 [11] analysis for tritium concentration increase in the primary cooling water through the Be first wall

Parameter	Be armor	Cu cooling block
A	900 m^2 [1]	
d	10 mm [1]	4 mm
D	D_0: 6.74×10^{-8} m^2/s E_D: 0.73 eV [12]	D_0: 6.2×10^{-7} m^2/s E_D: 0.378 eV [16]
K_S	K_{S0}: 2.2×10^{22} atom/m^3 $Pa^{0.5}$ E_S: 0.17 eV [13]	K_{S0}: 4.1×10^{23} atom/m^3 $Pa^{0.5}$ E_S: 0.37 eV [17]
C_t^0	500 ppm [14]	–
E_t	0.8 eV [14]	–
kr	kr_0: 1.4×10^{-29} m^4/s E_{kr}: 0.28 eV [14]	–
ϕ_i	10^{20} T/m^2/s (plasma) 0 T/m^2/s (waiting)	–
G	2.7×10^{11} T/cm^3/s[15] (plasma) 0 T/cm^3/s (waiting)	–
Rp	2.5 nm	–
Operational temperature	523 K at surface (plasma) 423 K (waiting)	423 K

Here, A: surface area, d: thickness, D: diffusion coefficient, K_S: Sievert's constant, C_t^0: trap site density, E_t: trap energy, kr: recombination coefficient, ϕ_i: implantation flux, G: tritium production rate in Be, Rp: mean range of tritium into Be. Plasma operation time (plasma) and waiting time (waiting) in ITER were assumed to be 3000 and 9000 s, respectively.

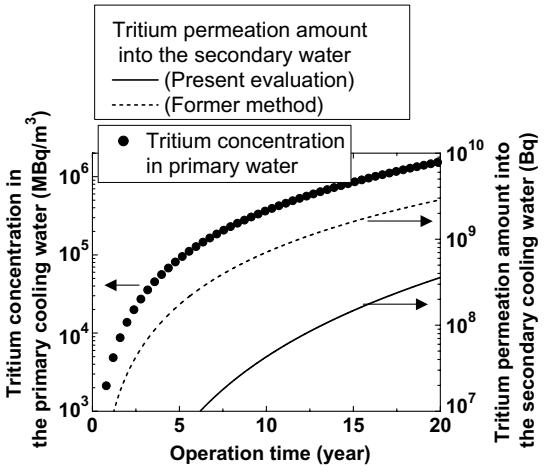

Fig. 3. Time evolution of tritium concentration in the primary water of the PFW/BLK cooling system and tritium permeation amount from the primary cooling water to the secondary cooling water through the heat exchanger. (present evaluation): based on obtained water-to-water permeability (former method): based on gas-to-gas permeability putting reduction factor as 1000.

4. Summary

Tritium permeation through SS316L heat exchanger from the primary cooling water to the secondary cooling water was investigated experimentally. As the result of experiment performed with the temperature of 423 K, the pressure of water of 0.9 MPa and the difference of tritium concentration of 7.4 TBq/m^3, the permeability of aqueous tritium was found to be 2.8×10^{-19} Bq/m/s/C (C: tritium concentration difference (Bq/m^3)), which leads the tritium permeation rate three orders smaller than that for gaseous tritium under the same tritium concentration. Tritium permeation through the heat exchanger from the primary cooling water to the secondary cooling water under the ITER operation condition was then evaluated. Evaluation of total tritium permeation amount based on the aqueous permeability obtained in this work was negligibly small in viewpoint of safety and was about almost one order less than that by the former method with the gaseous permeability putting the permeation reduction factor as 1000, which assures the conservativeness of the evaluation with the former method.

Acknowledgements

The authors would like to thank Drs S. Seki, H. Takatsu and H. Tsuji for their encouragement on this study, Drs T. Yamanishi and T. Hayashi for their suggestions and Safety Design Division of Department of ITER Project of JAERI for supporting this study.

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