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Impact of ceramic coating deposition on the tritium permeation in the Japanese ITER-TBM

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ABSTRACT

An effective coating for a tritium permeation barrier was developed by a chemical densified coating (CDC) method. This will help realize a reasonable design of the Japanese Test Blanket Module (TBM) of the International Thermonuclear Experiment Reactor (ITER). It was confirmed from results of tritium permeation experiments that the permeation reduction factor (PRF) of F82H steel with this coating reached about 300 at 600 °C. Furthermore, it was revealed by diffusion coefficient measurements that with this coating, diffusivity of deuterium is very low. With these measured tritium transport properties, a preliminary tritium permeation analysis was carried out for the Japanese ITER-TBM. It was indicated that tritium permeation through the F82H cooling pipe with the coating will be four orders of magnitude smaller than that without the coating.

1. Introduction

Development of a tritium permeation barrier coating on blanket structural materials is urgently required for the Japanese Test Blanket Module (TBM) of the International Thermonuclear Experiment Reactor (ITER). It is necessary to realize a reasonable design of a tritium recovery and processing system for fusion reactors [1,2]. In the course of designing the previous Japanese demonstration (DEMO) fusion reactor, it was revealed that of tritium permeation reduction by more than one order of magnitude is required to realize a reasonable fuel cycle design for the DEMO reactor [2]. Therefore, development of tritium permeation reduction coating has been carried out extensively. Ceramic coating materials such as Al_2O_3 [3–5] and TiN/TiC [6] have been studied in fabrication experiments and property measurements. Al_2O_3 coating has been developed using vacuum plasma spraying and pack-cementation methods, and TiN/TiC coating has been developed using chemical vapor deposition methods. These coating methods, however, have some disadvantages. It is difficult with a plasma spraying method to form a coating on the inner surface of a tube or a container. A pack-cementation process requires heating the substrate together with the coating at 800–1050 °C. It is difficult with a chemical vapor deposition method to form a thick coating on a large area of the substrate.

An effective coating was developed by the Japan Atomic Energy Agency (JAEA) in collaboration with Tocalo Co., Ltd. using a ceramic coating applied by a chemical densified coating (CDC) method [7,8]. Compared with other coating methods, the CDC method has some advantages, such as its capability to coat on the outer and/or the inner surface of a tube or a container at a low temperature (450 °C). While briefly described in Ref. [7], ceramic coating technology which increases these advantages is fully described in the present paper, along with discussion of modeling of tritium permeation through the coating. Preliminary results of tritium permeation analysis for the ITER-TBM are also presented.

2. Ceramic coating technology by chemical densified coating method

First, a fabrication technique for dense coating of Cr_2O_3–SiO_2 was developed based on the CDC method. The fabrication process is shown in Fig. 1. In this Cr_2O_3–SiO_2 coating fabrication technique, the substrate surface is coated with slurry consisting of SiO_2 granules and CrO_3 solution after pretreatment of the substrate. After this, the substrate thus coated with slurry is heated. In the final step, densification treatment was conducted by impregnation with CrO_3 solution and heating. This final step is carried out to fill the pores in the Cr_2O_3–SiO_2 coating. This densification treatment successfully fills pores in the coating [7]. However, it was found from results of a verification test for open pores that this coating still has some open pores. These open pores along grain boundaries from coating surface to substrate were considered defects because Cr_2O_3 in this coating is a crystalline ceramic.

Application of amorphous material was suggested for filling open pores in the coating of Cr_2O_3–SiO_2. This has led to

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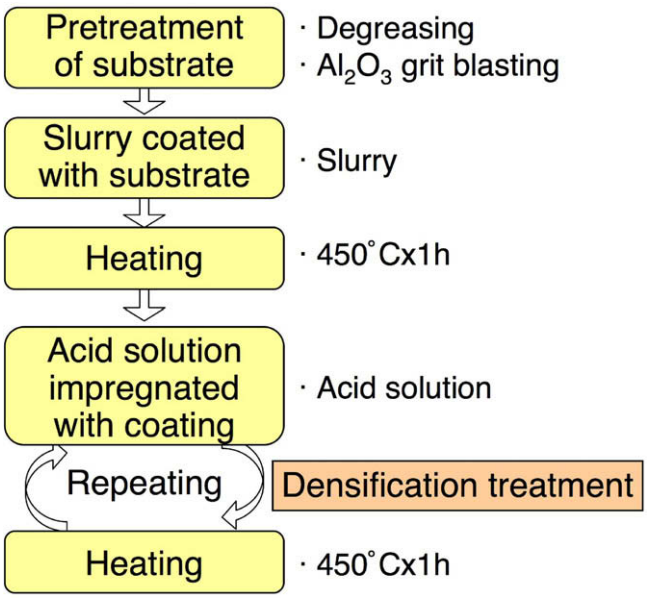

Fig. 1. Fabrication process of chemical densified coating method.

densification treatment using $CrPO_4$. The acid solution of $CrPO_4$ and H_2O was used for the densification treatment instead of CrO_3 solution. Fig. 2 shows a cross-sectional view of a coating fabricated only using the acid solution of $CrPO_4$ and H_2O. It is clear that pores remained in the coating. The acid solution did not soak through the coating due to formation of amorphous $CrPO_4$ on the coating surface. Therefore, use of a combination of different acid solutions in the densification treatment was examined. First, densification treatment using CrO_3 solution was carried out for filling pores in the coating. After that, densification treatment using the acid solution of $CrPO_4$ and H_2O was carried out for filling other defects causing passage of gas in the coating. This treatment using CrO_3 solution in the first of two densification treatments has successfully filled pores [7].

It was confirmed from results of in-pile tritium permeation experiments that the permeation reduction factor (PRF) of ferritic martensitic steel (F82H) with the coating of Cr_2O_3–SiO_2 including $CrPO_4$ reached about 300 at 600 °C [8], and it was also confirmed that this coating had a good adhesion property [7].

3. Deuterium diffusion measurement in the coating

Deuterium diffusion in the coating was studied by depth profile analysis of the nuclear reaction (NRA) [13]. Details of the experiment and analysis are given elsewhere [9]. To interpret the results, the diffusion coefficient of deuterium in this coating was calculated according to Eq. (1) [10]

$$D = 1.1 \times 10^{-10} \exp(-0.71eV/kT) \quad (1)$$

where D, k and T are diffusion coefficient [m^2/s], Boltzmann coefficient [eV/K] and temperature [K], respectively. The obtained diffusion coefficient was found to be smaller than that of SS304 [11,12] by five orders of magnitude. This means that an effective diffusion barrier can be expected if this coating is used. This diffusion coefficient was used for the following analysis of tritium permeation behavior in the ITER-TBM.

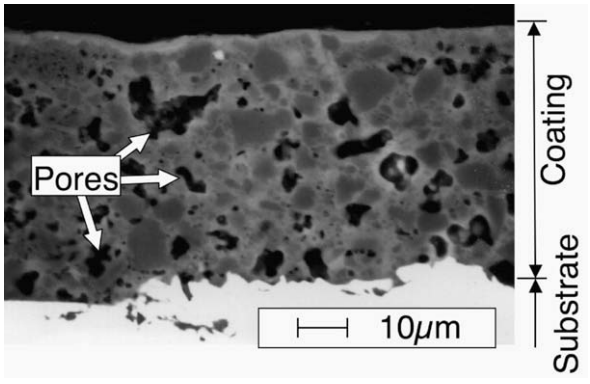

Fig. 2. Cross-sectional view of the coating fabricated only using the acid solution of $CrPO_4$ and H_2O.

4. Analysis model of tritium permeation behavior in the ITER-TBM

In the TBM program, various design concepts of tritium breeding blankets proposed by the ITER participant parties will be implemented. Among the blanket concepts, Japan proposes two kinds of DEMO-related TBM systems as solid breeder candidates for the blanket: a water-cooled solid breeder (WCSB) blanket and a helium-cooled solid breeder (HCSB), both with F82H steel structure. Both concepts are based on candidate designs for DEMO in Japan [13]. Both TBM systems are planned to be tested in ITER from the first day of the ITER operation.

In this preliminary analysis, tritium permeation from the tritium breeder region to the cooling water in the Japanese WCSB concept design was evaluated. The WCSB TBM and its analytical model are presented in Fig. 3. The cooling line of the first wall and the side wall as well as the manifold cooling pipe were considered as the permeability pathways. These pathways were simplified into one-dimensional analytical systems, shown schematically in Fig. 3. Design conditions of the WCSB TBM and calculation conditions are summarized in Table 1 [10,14].

Tritium permeation analysis was carried out using the Tritium Migration Analysis Program (TMAP) code [15,16]. Eqs. (2) and (3) representing tritium transport were used in the TMAP code:

$$\frac{\partial C(x,t)}{\partial t} = -\frac{\partial J(x,t)}{\partial x} + \gamma C(x,t) + S(x,t) \quad [\mathrm{Bq}], \quad (2)$$

$$J(x,t) = -D_{eff}(T)\left(\frac{\partial C(x,t)}{\partial x} + \frac{C(x,t)Q}{kT^2}\right) \quad [\mathrm{n/m^2/s}], \quad (3)$$

where $C(x,t)$, S, $J(x,t)$, γ, D_{eff} and Q denote tritium concentration [Bq], tritium source [Bq], diffusion flux [$n/m^2/s$], decay constant [s^{-1}], effective diffusivity [m^2/s] and heat of transport [W/m^3], respectively.

In an analysis modeling test, hydrogen concentration change in the sweep gas from the inlet to the outlet of the TBM sweep gas line was calculated for evaluation of the effect of sweep gas flow on hydrogen permeation. The analysis result of the change in hydrogen concentration at the sweep gas outlet from the start of sweep gas flow onward is shown in Fig. 4. The flow rate, a parameter in this analysis, was changed to be 0.1–2.0 times the flow rate of the design. Hydrogen concentration became steady within 100 s under the design flow rate. It also became clear that hydrogen permeated uniformly from the inlet to the outlet of the sweep gas line with all flow rates.

5. Result of tritium permeation analysis

Analysis of tritium permeation to the TBM cooling line by TMAP code was carried out using the measured deuterium coefficient described above in Section 3. The coating thickness was estimated to be 2 μm in this analysis, because the effective densification depth of Cr_2O_3–SiO_2 including $CrPO_4$ is considered to be on the order of

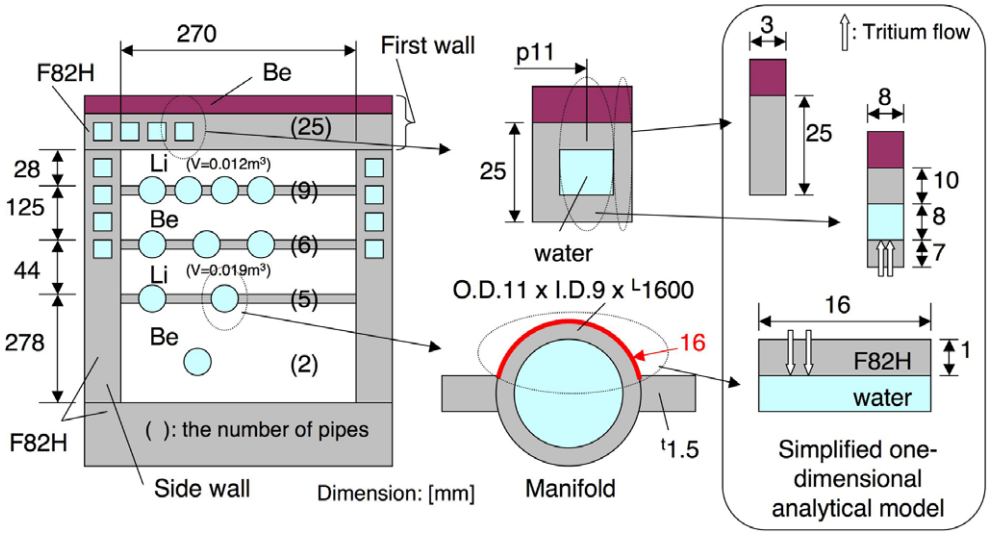

Fig. 3. Schematic drawing of the Japanese WCSB TBM and analytical model for TMAP analysis.

Table 1
Design and calculation conditions of WCSB TBM.

Temperature	598 K
Sweep gas component	He – 1000mpH_2
Sweep gas flow rate	2.59 Nm^3/h
Ratio of partial pressure (T_2/H_2)	1/100
Diffusion coefficient at 598 K	6.5×10^{-9} m^2/s (F82H steel) [14]
	1.1×10^{-16} m^2/s (Coating) [10]

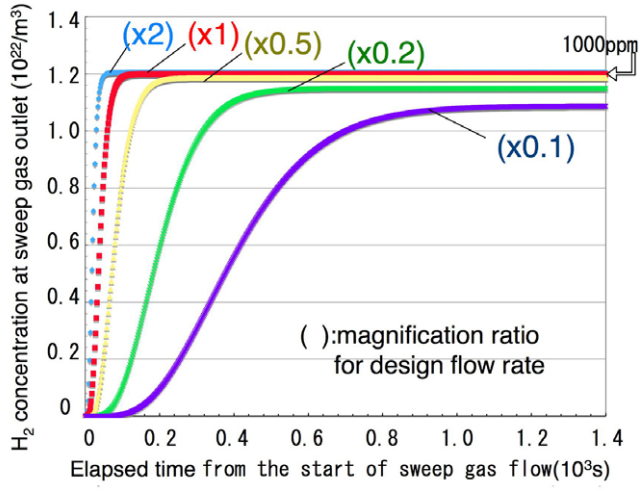

Fig. 4. Analysis result of hydrogen concentration at the sweep gas outlet from the start of sweep gas flow.

several μm and the effective analysis depth of the NRA is 2 μm [10]. The analysis result of the diffusion flux and the concentration of tritium are shown in Fig. 5 as a function of the elapsed time from the start of the sweep gas flow.

Tritium permeation flow reaches a steady state within 100 s in the case of F82H steel cooling pipes without the coating. On the other hand, it takes more than 10000 s to reach a steady state in the case of coated F82H cooling pipes. The increase of the tritium amount transported into the cooling water through F82H steel without and with the coating per full power day (FPD) was

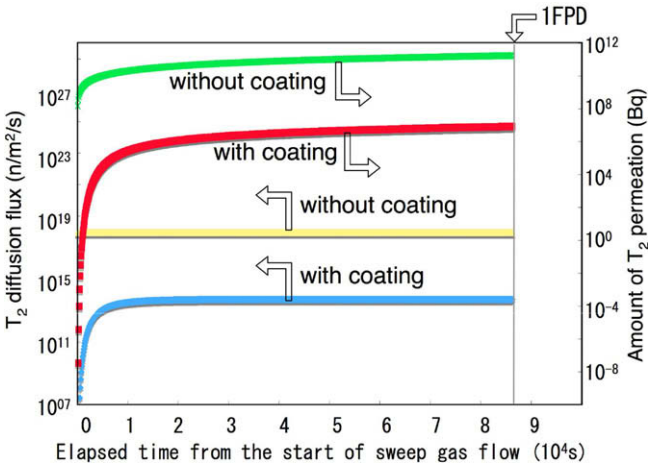

Fig. 5. Analysis result of diffusion fluxes and concentration of tritium from the start of sweep gas flow.

9×10^{10} and 1×10^{7} Bq/FPD, respectively. These results of the present preliminary analysis indicate that a reduction of tritium permeation in the ITER-TBM can be expected when this coating is used, as long as the integrity of the coating is maintained. Future research is desired, including tests of the integrity of the coating on the F82H cooling pipes, and a demonstration of tritium permeation reduction under simulated ITER-TBM conditions.

6. Conclusions

An effective coating for reduction of tritium permeation was developed by the chemical densified coating (CDC) method to impregnate amorphous material in the coating. This method has successfully filled pores when using CrO_3 solution in a first densification treatment, and also filled other defects allowing passage of gas by using acid solution of $CrPO_4$ and H_2O in the next densification treatment. Furthermore, it was clear from the results of diffusion coefficient measurements that with this coating, diffusivity of deuterium is very low.

Tritium permeation to the TBM cooling line was preliminarily analyzed by TMAP code using the deuterium diffusion coefficient

of the coating which was measured. The calculation results indicate that tritium permeation through the F82H cooling pipe with the coating will be four orders of magnitude smaller than that without the coating. In future research, we need to measure the tritium diffusion coefficient of this coating and to evaluate the effect of the different isotopes hydrogen, deuterium and tritium. This coating technology can be applied to the tritium permeation barrier of a tritium recovery and processing system for DEMO fusion reactors, which will contribute to realization of a reasonable design of the tritium system.

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