Tritium permeation experiments using reduced activation ferritic/martensitic steel tube and erbium oxide coating

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HIGHLIGHTS

- Tritium permeation has been measured on uncoated and Er_2O_3-coated F82H tubes.
- Surface effects were observed in permeation tests with 1.2–40 ppm tritium.
- HTO was formed by isotope exchange reactions at Er_2O_3-coated surfaces.
- High-performance Er_2O_3 coatings could be fabricated on tubular F82H substrates.

ARTICLE INFO

ABSTRACT

Low concentration tritium permeation experiments have been performed on uncoated F82H and Er_2O_3-coated tubular samples in the framework of the Japan-US TITAN collaborative program. Tritium permeability of the uncoated sample with 1.2 ppm tritium showed one order of magnitude lower than that with 100% deuterium. The permeability of the sample with 40 ppm tritium was more than twice higher than that of 1.2 ppm, indicating a surface contribution at the lower tritium concentration. The Er_2O_3-coated sample showed two orders of magnitude lower permeability than the uncoated sample, and lower permeability than that of the coated plate sample with 100% deuterium. It was also indicated that the memory effect of ion chambers in the primary and secondary circuits was caused by absorption of tritiated water vapor that was generated by isotope exchange reactions between tritium and surface water on the coating.

1. Introduction

Understanding tritium migration behaviors in a fusion blanket is a key technology for the establishment of a fusion reactor fuel cycle. In a framework of the Tritium, Irradiation and Thermofluid for America and Nippon (TITAN) Japan-US collaborative program conducted in fiscal year 2007–2012, tritium permeation in fusion reactor materials including a tritium permeation barrier (TPB) has been investigated [1]. One of the remaining issues in tritium behaviors in the fusion materials is to clarify the permeation behavior at low partial pressure tritium for the interpolation to practical fusion blanket conditions. Regarding the TPB coatings, erbium oxide (Er_2O_3) thin films have recently shown remarkable permeation reduction factors using a physical vapor deposition method

[2]. Moreover, Er_2O_3 coatings have been fabricated on plate substrates by not only spin-coating but also dip-coating process with a metal-organic decomposition (MOD) method [3,4]. However, the establishment of plant-scale fabrication of the coating, for example on a tubular substrate, is challenging. In this study, low concentration tritium permeation experiments have been carried out using reducing activation ferritic/martensitic (RAFM) steel tubular substrates and Er_2O_3 coatings fabricated by the liquid-phase method.

2. Experimental

2.1. Sample preparation

Electrolytically polished RAFM steel F82H (8Cr-2W) tubes with dimensions of 150-mm length, 8-mm inner diameter, and 1.5-mm thickness were used as substrates. The MOD coating procedure is described in detail in the previous study [4]. First, the tube substrate was dipped into an Er_2O_3 coating precursor (Kojundo

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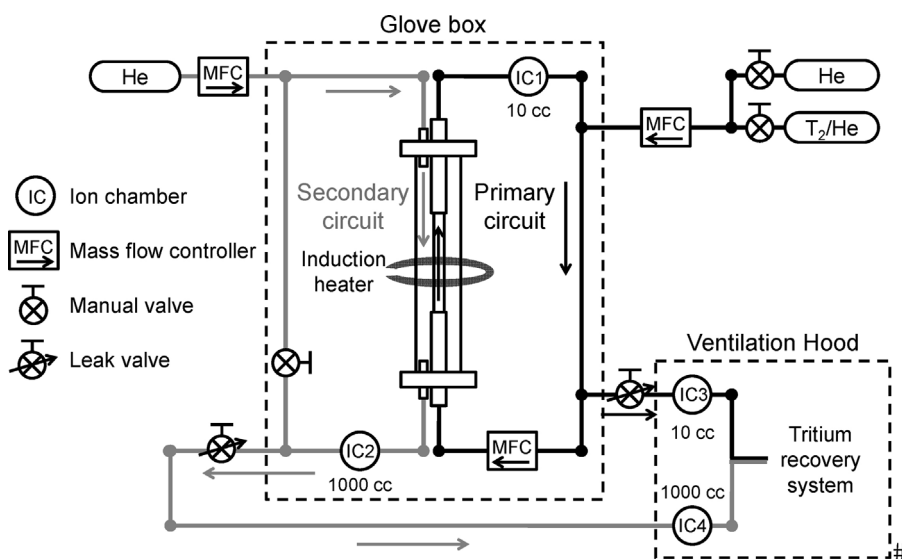E-mail address: chikada@nuclear.jp (T. Chikada).

Fig. 1. Schematic view of the tritium permeation system.

Chemical Laboratory Co., Ltd., Er-03®) without addition of thinner, and then withdrawn at a constant speed of $1.2\,mm\,s^{-1}$ using a dip-coater. Second, the sample was placed in a dry oven set at $120\,°C$ for 10 min to turn the solvent into a gel. Finally, the sample was heat treated in an infrared image furnace to decompose organic residues and to crystallize the Er_2O_3 coating. The heat-treatment condition was determined from the previous study [4]: at $700\,°C$ for 10 min in hydrogen (purity: 99.9999%, O_2: <0.02 ppm, H_2O: <0.5 ppm) with approximately 0.6% moisture. The flow rate is less than $10\,cm^3\,min^{-1}$, and the rate of temperature increase and decrease is 100 and $30\,°C\,min^{-1}$, respectively. Moisture was added to the hydrogen flow by passing it through ice-chilled water. The coating process was repeated three times in order to increase coating thickness and reduce unevenness of the coating. The thickness of the coating was estimated as $0.3\,\mu m$ on both inner and outer surfaces of the sample by the previous result [3]. Both ends of uncoated and coated samples were welded to prepare sample assemblies for the installation in a tritium permeation system which is describe in the next section.

2.2. Permeation setup

A conceptual diagram of the tritium permeation system is described in Fig. 1. The sample assembly was mounted in a quartz tube. Sample heating was conducted using an induction heater with a 1-turn, 5-inch-diameter coil made of copper. Tritium-helium mixture gas was introduced in a circuit (primary circuit) at a rate of 15 sccm (standard $cm^3\,min^{-1}$) and circulated at a rate of 1000 sccm using mass flow controllers to keep the tritium concentration in the circuit. Tritium that permeated outward through the sample was purged by a pure helium flow of 100 sccm in another circuit (secondary circuit). The tritium concentrations in the primary and secondary circuits were examined with 10 and $1000\,cm^3$ ion chambers which are called IC1 and IC2, respectively. After passing through the ion chambers, the circulating gas in the primary circuit was partly discharged at a rate of 15 sccm to a tritium recovery system placed in a ventilation hood, while the helium flow with purged tritium in the secondary circuit was once through. In the ventilation hood, tritium concentrations of flowing gases from both circuits were again examined by the ion chambers: a $10\,cm^3$ ion chamber is for the primary circuit (IC3) and a $1000\,cm^3$ one for the secondary circuit (IC4).

The procedure of a gas-driven tritium permeation measurement is described as follows. After setting a test temperature using the induction heater, the system was conditioned for 2 h until the signals of ion chambers became stable. Then the pure helium flow in the primary circuit was switched to the tritium-helium mixture. The tritium concentration in the mixture gas cylinders was originally prepared as 3 and 65 ppm; however, it was actually measured as 1.2 and 40 ppm using ion chambers before and after a series of experiments. Since pressures in both circuits were set at 105 kPa, the tritium concentration of 1.2 and 40 ppm was corresponding to the partial pressure of 0.13 and 4.2 Pa. It took approximately 2 h to reach the steady state tritium concentration after introducing the tritium-helium mixture. The length of the work day limited the duration of the permeation experiments to less than 6 h.

One of the important issues in the evaluation of tritium permeation through tubular samples is how to determine a temperature distribution. The induction heating power was controlled by a thermocouple inserted in the center of the sample with its head touching the inner wall. In this study, an infrared camera and a pyrometer were used to measure the temperature profile of the sample surface. The infrared camera could automatically store the temperature data only at more than $600\,°C$ in two-dimensional display. On the other hand, the uncoated sample was measured by the pyrometer because lower test temperatures were required to avoid surface oxidation. The temperature profile at the permeation section was manually measured with vertically moving the position. The temperature distribution was calculated by Gaussian fitting to the surface temperature data.

The permeation phenomenon of hydrogen isotopes from the primary circuit which contains a known concentration to the secondary circuit with a negligible small concentration are represented by following equation [5]:

$$J = P\frac{p^{0.5}}{d}, \tag{1}$$

where J is the permeation flux, P is named permeability which is intrinsic parameter for the sample, p is the partial pressure of the hydrogen isotope introduced into the primary circuit, and d is the thickness of the sample. The pressure exponent represents permeation regime: the value of 0.5 indicates the rate-limiting process is diffusion of hydrogen isotope atoms, and the value of 1 means the rate-limiting process is dominated by molecular reactions such as adsorption and recombination at the sample surface. For a simple