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Deuterium permeation through metals under γ-ray irradiation

Hikari Fujita[a,*], Takumi Chikada[b], Jumpei Mochizuki[b], Seira Horikoshi[b], Moeki Matsunaga[b], Teruya Tanaka[c], Takayuki Terai[a]

[a] The University of Tokyo, Tokyo, Japan
[b] Shizuoka University, Shizuoka, Japan
[c] National Institute for Fusion Science, Toki, Japan

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ABSTRACT

Tritium handling in fusion reactors is important for economical fuel cycles and radiological safety; however, the extremely limited number of studies on tritium permeation through structural materials under irradiation conditions has been reported. In this study, γ-ray irradiation effect on tritium permeation has been investigated using three kinds of metals with different permeation properties including palladium, platinum, and austenitic stainless steel. An increase of deuterium signal on the quadrupole mass spectrometer during irradiation consisted of an increase of deuterium permeation flux and an electrical noise, and both were separately detected. The increase rate of permeation flux for all materials became lower as the test temperature increased. Dose rate dependence of the increase rate was confirmed. Oxidation of the austenitic stainless steel indicates a decrease of not only the deuterium permeation flux but also the irradiation effect.

1. Introduction

Tritium handling in fusion reactors is important for economical fuel cycles and radiological safety, and so a lot of knowledge about tritium permeation has been accumulated. However, the extremely limited number of studies on tritium permeation through structural materials under irradiation conditions has been reported [1–3]. Although hydrogen permeation through the Cr18Ni10Ti-type stainless steel increased under the neutron and γ-ray dose rate of $(1.4–1.6) \times 10^{18}\,m^{-2}\,s^{-1}$ at lower temperatures [3], no clue for the explanation of which neutron or photon makes more impact in permeation through a material has been found. In this study, γ-ray irradiation effect on tritium permeation has been investigated using fusion-relevant metals with different permeation properties. For example, palladium (Pd) is utilized for hydrogen separation membranes today because of its high solubility and diffusivity of hydrogen isotopes. Dissociation of hydrogen on Pd surface is much faster than diffusion in Pd; therefore, γ-ray irradiation effect on hydrogen diffusion can be detected using Pd. In addition, platinum (Pt) with larger reaction cross-section of γ-ray and 316-type austenitic stainless steel (SS316) widely utilized as structural material were also used in this research.

2. Experimental details

2.1. Sample

As-received Pd (The Nilaco Corporation, 99.95%), Pt (The Nilaco Corporation, 99.98%) and SS316 plates were used. The dimension of the samples was $30 \times 30 \times 0.5\,mm^3$, $20 \times 20 \times 0.5\,mm^3$ and $25 \times 25 \times 0.5\,mm^3$ respectively.

2.2. Deuterium permeation experiments under γ-ray irradiation

Deuterium permeation experiments were performed under γ-ray irradiation using a gas-driven deuterium permeation apparatus described in Ref. [4]. The apparatus was set up in a γ-ray irradiation chamber, namely HYPEγION (HYdrogen PErmeation system under γ-ray IrradiatiON), as shown in Fig. 1. A sample was sealed with two nickel C-rings with inconel coil spring inside (Usui Kokusai Sangyo Kaisha, Ltd. U-TIGHTSEAL®), whose inside diameter, outside diameter, and thickness were 12.8 mm, 17.6 mm and 2.4 mm, respectively. The permeation area was calculated to be $1.72 \times 10^{-4}\,m^2$ by measuring impressions of the seal materials on the sample after the permeation experiments. The sample was mounted in an electric furnace for temperature control after polishing the backside with abrasive papers to remove an oxide layer. In addition, a thermocouple was inserted through a hole made in the sample holder to directly touch the sample

* Corresponding author.
E-mail address: hfujita@g.ecc.u-tokyo.ac.jp (H. Fujita).

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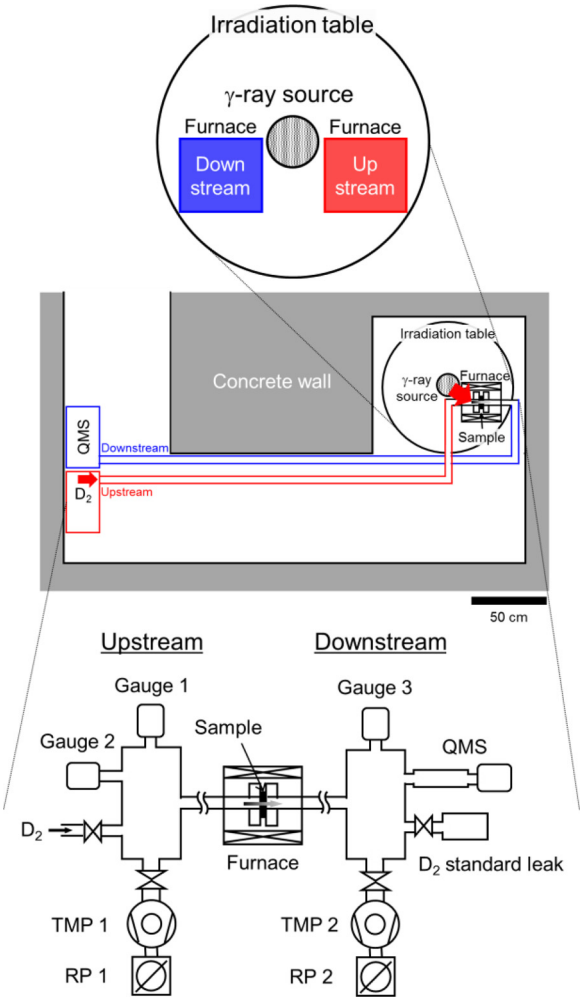

Fig. 1. Floor plan of HYPEγION with an enlarged layout of the irradiation table and conceptual diagram of gas-driven deuterium permeation apparatus.

surface. The pipework of which was made long (4–5 m) to avoid irradiation damage to the precision instruments. For example, the γ-ray radiation level at a quadrupole mass spectrometer (QMS, Canon ANELVA Corp. M-101QA-TDM) was several μGy s^{-1}. The tubing size was a quarter inch (0.635 mm) in outside diameter with Swagelok® VCR fittings. The volume on the downstream side was calibrated using a calibration volume. Deuterium permeation flux through the sample was detected as ion current by the QMS at the downstream. The ion current and the deuterium permeation flux was calibrated using a deuterium standard leak (Vacuum Technology Inc.).

The sample was irradiated from two directions, for example the deuterium high-pressure side (upstream) and the low-pressure side (downstream). ^{60}Co at Shizuoka University was used as a γ-ray source whose radioactivity was approximately 30 TBq. γ-ray flux at 10 cm from the source was estimated to be 2×10^{14} m^{-2} s^{-1}. Absorbed dose rate of water was calculated to be 0.15 Gy s^{-1} at 12 cm from the γ-ray source by dose rate measurements of Fricke dosimetry as shown in Fig. 2. Dose rate was proportional to the distance from the γ-ray source which was raised to the power of −2.1; therefore, the values obtained by the measurements would be reasonable in comparison with the theoretical power of −2. The measured dose rate exceeded the calculated one since a portion of γ-rays scattered in the electric furnace hit the sample, which is called build-up effect.

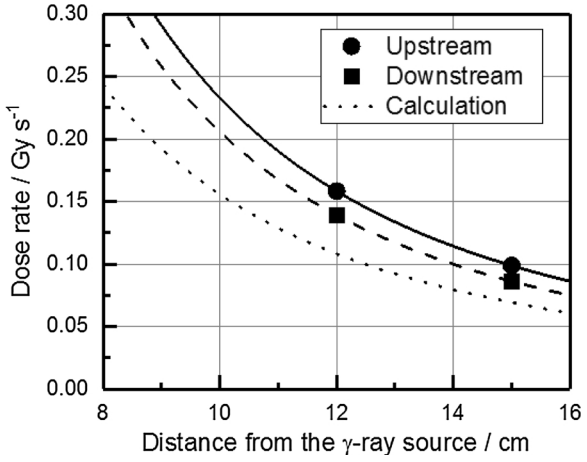

Fig. 2. Absorbed dose rate of water in relation to distance between the sample and the γ-ray source. The experimental values of upstream and downstream measured by Fricke dosimetry are plotted, and the fitted curves are also shown. The dotted line represents calculated dose rate from ^{60}Co radioactivity and distance.

2.3. Theory

Hydrogen isotope permeation through material is in principle dominated by two processes: solution and diffusion. First, the solid solution of hydrogen atom in material is represented by Sieverts' law:

$$S = K_S p^{0.5}, \tag{1}$$

where S is the hydrogen solubility, K_S is Sieverts' constant and p is the driving pressure introduced into the upstream. Second, the diffusion phenomenon of hydrogen is a thermally activated process expressed by the Arrhenius rate equation:

$$D = D_0 \exp\left(-\frac{E_D}{RT}\right), \tag{2}$$

where D is hydrogen diffusivity, E_D is the activation energy of diffusion, R is the gas constant and T is the temperature. Finally, the hydrogen permeation flux J at steady-state is expressed with the product of Eqs. (1) and (2):

$$J = K_S D \frac{p^{0.5}}{d}, \tag{3}$$

where $K_S D$ is named permeability P as the intrinsic parameter of the permeation, and d is the thickness of the sample. The pressure exponent represents permeation regime: the pressure exponent value of 0.5 indicates that the rate-limiting process is diffusion of hydrogen atoms in material, and that of 1.0 indicates that the rate-limiting process is dominated by molecular reactions such as adsorption and recombination at the sample surface.

2.4. Analysis

Effect of γ-ray irradiation on deuterium permeation was evaluated by analyzing temporal changes of deuterium permeation flux J. Fitted curve of deuterium permeation flux before γ-ray irradiation was extrapolated, and the difference between deuterium permeation flux during and before γ-ray irradiation was calculated. Magnitude of the difference and percentage of the difference to the original deuterium permeation flux were evaluated under various conditions.