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The effect of γ-ray irradiation on deuterium permeation through reduced activation ferritic steel and erbium oxide coating

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ABSTRACT

Deuterium permeation through a fusion-relevant ferritic steel F82H with and without erbium oxide coatings under γ-ray irradiation has been investigated in a temperature range from 300 to 700 °C. The deuterium permeation flux through F82H sample increased by γ-ray irradiation at lower temperatures below 450 °C. The irradiation effect increased with dose rate, and the percentage of the permeation flux gain might be several percent under the dose rate of a few Gy s^{-1}. Temperature of the F82H sample surface rose by about 0.5 °C depending on the dose rate, and so the γ-ray irradiation effect is mainly attributed to γ-heating. On the other hand, at higher temperature above 500 °C, no appreciable change of the deuterium permeation was observed. Similarly, the deuterium permeation flux through erbium oxide coated samples increased under γ-ray irradiation at lower temperatures (350–450 °C), but no appreciable change of permeation flux through coatings was observed at higher temperatures (600–700 °C). The coating surface temperature increased at lower sample temperatures by γ-heating.

1. Introduction

Hydrogen solution and diffusion in metals make characteristic effects on structural materials of nuclear fusion/fission reactors and hydrogen energy systems. In the fusion reactors, tritium loss by permeation through structural materials is particularly a serious issue in terms of fuel cycle and radiological safety. Tritium permeation barrier (TPB) has been developed with ceramic coatings since 1970s [1,2]. In our previous studies, precise hydrogen isotope permeation behaviors with the highest permeation reduction factor at elevated temperatures have been achieved using erbium oxide (Er_2O_3) coatings prepared by gas and liquid-phase methods [3–5]. Although a lot of studies on hydrogen permeation through the structural materials were performed, an extremely limited number of studies addressing the hydrogen isotope permeation behavior under irradiation environment has been published. The only literature reported that hydrogen permeability through 316 stainless steel sample showed no significant change with the presence of 1 mCi γ-ray source [6]. However, γ-ray dose rate is estimated to be up to 0.89 Gy s^{-1} in ITER [7] and 2.4 kGy s^{-1} in the demonstration power reactor [8], which is several orders of magnitude higher values than estimated in the reference. Another report showed an increase of hydrogen permeability through the Cr18Ni10Ti-type stainless steel under irradiation of both neutron and γ-ray [9]. As for irradiation effects on TPB coatings, in-pile tritium permeation experiments were conducted using research fission reactors and fusion-relevant ferritic steel samples coated by Fe-Al/Al_2O_3 or Cr_2O_3-SiO_2 including $CrPO_4$, but in both cases, the irradiation effect on permeation was unclear [10,11]. Therefore, this study addresses an effect of γ-ray with secondary electrons and bremsstrahlung emitted by the γ-ray-material interactions on hydrogen isotope permeation through a ferritic steel as a candidate structural material for fusion reactors with and without Er_2O_3 coatings.

2. Experimental

2.1. Sample preparation

Reduced activation ferritic/martensitic (RAFM) steel F82H (Fe-8Cr-2 W) disk substrates with dimensions of 20-mm diameter and 0.5-mm thickness were mirror-polished. Palladium (Pd) coatings were deposited on both sides of F82H substrates by vacuum deposition to avoid surface oxidation during deuterium permeation measurements at elevated temperature.

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Er_2O_3 coatings were fabricated on one side of the substrates by filtered vacuum arc deposition (VAD) [12] or metal organic decomposition (MOD) [5]. The thickness of the coatings fabricated by VAD and MOD was approximately 1.2 μm and 0.25 μm, respectively. The crystal structure of the coatings was confirmed to be the cubic phase Er_2O_3 by grazing incidence X-ray diffraction.

2.2. Deuterium permeation experiments under γ-ray irradiation

Deuterium permeation experiments were performed using a gas-driven deuterium permeation device described in Ref. [12] in the temperature range of 300–700 °C. The driving pressure of deuterium to the upstream was set to 1.00×10^4–8.00×10^4 Pa. The deuterium permeation apparatus was set up in a γ-ray irradiation chamber, namely HYPEγION (HYdrogen PErmeation system under γ-ray IrradiatiON) which was recently presented in Ref. [13]. The pipework from the sample to the measurement system was extended to be 4–5 m so as to reduce irradiation damage to the precision instruments such as vacuum gauges and a quadrupole mass spectrometer (QMS). A sample was irradiated from the deuterium high-pressure side (upstream). ^{60}Co at Shizuoka University was used as a γ-ray source whose radioactivity was approximately up to 33 TBq. γ-ray flux at 10 cm from the source was estimated to be 2×10^{14} m^{-2} s^{-1}, and absorbed dose rate of water was calculated to be 0.13 Gy s^{-1} at 10 cm by Fricke dosimetry as shown in Fig. 1. The measured dose rate was inversely proportional to the square of the distance from the γ-ray source, which was theoretically reasonable. Note that this dose rate included interactions of not only γ-rays but also secondary electrons and bremsstrahlung. The dose rate changes due to the existence of an electric furnace positioned between the γ-ray source and sample since electromagnetic waves such as γ-rays produce scattered rays due to interaction with substances. It was confirmed by Fricke dosimetry that the build-up factor in this experimental system was about 2, which indicated that the real dose rate at 10 cm from the radiation source was 0.26 Gy s^{-1}.

Deuterium permeation flux through sample was detected as ion current by a QMS installed at the low-pressure side (downstream). The QMS ion current was calibrated using a deuterium standard leak (Vacuum Technology Inc.). The Er_2O_3-coated sample was mounted in the permeation apparatus with the coated side facing to the upstream after polishing the backside of the sample with abrasive papers to remove an oxide layer.

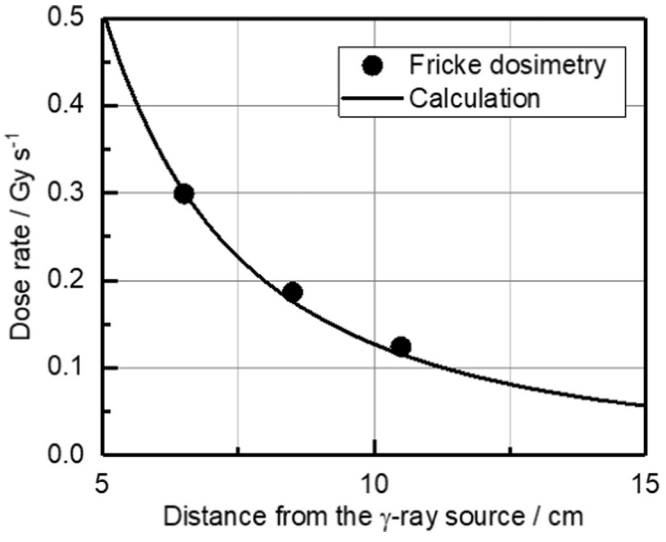

Fig. 1. Dose rate against distance between the sample and the γ-ray source.

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 of permeation data

γ-ray irradiation effect on deuterium permeation was evaluated by analyzing a temporal change of deuterium permeation flux J converted from the QMS ion current. 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. The difference of both actual values and its percentage to the original deuterium permeation flux were evaluated under various conditions.

3. Results and discussion

3.1. Permeability

Temperature dependence of deuterium permeability for the samples is shown in Fig. 2. Because the permeability of the Pd-coated F82H sample agreed well with the reference data of uncoated F82H [14], a rate-determining process of the permeation through the sample was diffusion in F82H. The permeability of the Er_2O_3 coated F82H sample was 2 − 3 orders of magnitude lower than the data of F82H; thus, a rate-determining process of permeation was deuterium migration in the Er_2O_3 coating [3].

3.2. F82H sample coated with Pd

Fig. 3 shows a typical temporal change of deuterium permeation flux through the Pd-coated F82H sample at a set temperature of 300 °C with deuterium introduction from the upstream before, during and after γ-ray irradiation. The permeation flux gently increased after starting γ-ray irradiation, and slowly decreased after finishing irradiation. Immediately after finishing irradiation, the sample surface temperature measured by a thermocouple directly contacting to the sample rose by 0.2–0.5 °C depending on the γ-ray dose rate as compared to that before starting irradiation. There was a 2-h time lag until the permeation flux reached a steady state. Judging from the previous runs with other materials [13], the time lag would be caused not by a fluctuation of the