



Deuterium permeation through monoclinic erbium oxide coating

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ARTICLE INFO

Keywords:

Tritium
Permeation
Coating
Erbium oxide
Irradiation

ABSTRACT

Erbium oxide has been investigated as a tritium permeation barrier material in a D-T fusion reactor fuel system for more than a decade. Erbium oxide normally forms the cubic phase; however, the uncommon monoclinic phase was formed under ion irradiation or high pressure conditions. In this study, the monoclinic erbium oxide coatings were prepared to examine their microstructure and deuterium permeability with ion-irradiation effect. The monoclinic phase coatings with a preferred orientation showed 1–2 orders of magnitude higher permeability at 300–400 °C than the cubic phase coating reported previously due to the smaller grain with uncrystallized region. After the permeation measurement at 600 °C, the permeability drastically decreased due to the phase transformation to the cubic phase and a change in grain structure from columnar to granular. An Fe-ion irradiated coating with the damage concentration of 0.05 dpa showed the decrease in the permeability at 500 °C, indicating the irradiation damage to the grain structure may accelerate the grain growth at lower temperature, while the phase transformation would occur at 600 °C because the diffusivity did not change much at 500 °C and drastically decreased at 600 °C.

1. Introduction

In commercial fusion power stations, tritium permeation through structural materials must be strictly controlled to ensure efficient fuel cycles and radiological safety. Erbium oxide (Er₂O₃) coatings have been investigated as a promising tritium permeation barrier (TPB) for over a decade and shown permeation reduction factors of up to 10⁵ [1,2]. The cubic crystal structure has been basically used for the investigations on hydrogen isotope permeation behaviors in the Er₂O₃ coatings. However, two other crystal phases, monoclinic and hexagonal, can be formed for Er₂O₃ [3]. In particular, the phase transformation from the cubic to monoclinic was reported under ion irradiation or high pressure conditions [3,4]. Therefore, it is important to investigate the permeation behavior in the monoclinic phase Er₂O₃ coating which might be generated under an actual reactor environment. The monoclinic phase Er₂O₃ coatings are able to be prepared by a physical vapor deposition method with a bias voltage to accelerate Er ions [5]. In this study, microstructure analyses and gas-driven deuterium permeation measurements for the monoclinic phase Er₂O₃ coatings deposited on steels have been carried out in order to elucidate hydrogen isotope permeation mechanism in the Er₂O₃ coatings with different crystal structures. The ion irradiation effect on deuterium permeation through the coating

has been also investigated.

2. Experimental details

2.1. Preparation of Er₂O₃ coating samples

Reduced activation ferritic/martensitic steel F82H (Fe-8Cr-2W, F82H-BA07 heat) provided by National Institutes for Quantum and Radiological Science and Technology (QST), Japan were used as substrates. F82H plates (25 × 25 × 0.5 mm³) were mirror-polished before deposition. The Er₂O₃ coatings were prepared using a deflection magnetic field type vacuum arc vapor deposition device described in detail in Refs. [6,7]. The monoclinic phase Er₂O₃ were formed with the bias voltage of –150 V during deposition. The substrate temperature was at room temperature for the first minute followed by at 600 °C for 19 min to avoid substrate oxidation which causes degradation of coating adhesion, and the phase transformation during deuterium permeation measurements at lower temperatures. The thickness of the prepared coatings was 1.3–1.6 μm.

The monoclinic phase Er₂O₃ coating sample was damaged by iron (Fe²⁺) irradiation using a 3 MV tandem accelerator in Takasaki Ion Accelerators for Advanced Radiation Applications (TIARA) at QST. The

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ion energy of Fe^{2+} was 6 MeV, and the sample temperature was at room temperature during irradiation. The irradiation damage was introduced with the flux of $3.2 \times 10^{14} \text{ Fe}^{2+} \text{ m}^{-2} \text{ s}^{-1}$ and the fluence of $1.7 \times 10^{18} \text{ Fe}^{2+} \text{ m}^{-2}$. The displacement damage concentration calculated by SRIM-2008 code [8] was estimated to be 0.05 displacement per atom (dpa).

2.2. Characterization

Cross sections of the coatings were observed using a transmission electron microscope (TEM, JEM-2800, JEOL Ltd.). TEM specimens were fabricated using a focused ion beam system (FIB, NB5000, Hitachi High-Technologies Co.). Both are located at National Institute for Fusion Science (NIFS), Japan. Crystallographic information including crystal phase and grain orientation on the surfaces of the coatings were analyzed by grazing incidence X-ray diffraction (GIXRD, Smartlab, Rigaku Co. Ltd.) with a fixed incident angle of 0.5° and electron backscattering diffraction (EBSD) embedded in a field emission scanning electron microscope (FE-SEM, JSM-7000F, JEOL Ltd.) at The University of Tokyo.

Deuterium permeability of the samples was measured by a gas-driven deuterium permeation apparatus described elsewhere [9]. The procedure of the permeation experiments was followed by Ref. [10]. Before mounting to the sample holder, the backside of the sample was polished with abrasive papers to remove an oxide layer. The test temperature was set at 300–700 °C, and the measurements were conducted from lower temperature. The driving pressure of deuterium to the upstream was set to 1.00×10^4 – 8.00×10^4 Pa.

When the rate-determining process is atomic diffusion of deuterium through the solid (diffusion limited regime), the steady-state deuterium permeation flux per unit area and unit time J through a sample with the thickness of d is expressed by the following equation [11]:

$$J = K_S D \frac{p^{0.5}}{d} \quad (1)$$

where K_S is Sieverts' constant, D is the deuterium diffusivity, and p is the deuterium driving pressure. $K_S D$ is called the permeability P as a characteristic parameter of the sample. In this study, the diffusion limited regime was experimentally confirmed in both the monoclinic and cubic phase Er_2O_3 coating samples. In addition, the rate-determining step in deuterium permeation through the samples was diffusion in the coating because surface coverage of the coating is high enough, and the deuterium diffusion in the coating is much slower than that in the F82H substrate [9]. Note that when the rate-determining process is molecular processes such as molecular diffusion, deuterium adsorption or recombination at the surface (surface limited regime), the pressure exponent shows unity. In this study, the permeability of the samples was compared at the driving pressure of 8.00×10^4 Pa, which was the highest pressure and then had the smallest effect on surface reactions.

In addition, when the permeation flux is integrated with respect to time, the diffusivity can be obtained from an intercept t_{lag} on the t -axis with the following equation [11]:

$$t_{\text{lag}} = \frac{d^2}{6D}. \quad (2)$$

In this study, deuterium permeability and diffusivity were measured to discuss the permeation mechanism in the coated samples. In addition, since both are thermally activated processes, they can be expressed by

$$P = P_0 \exp\left(-\frac{E_P}{RT}\right) \quad (3)$$

$$D = D_0 \exp\left(-\frac{E_D}{RT}\right) \quad (4)$$

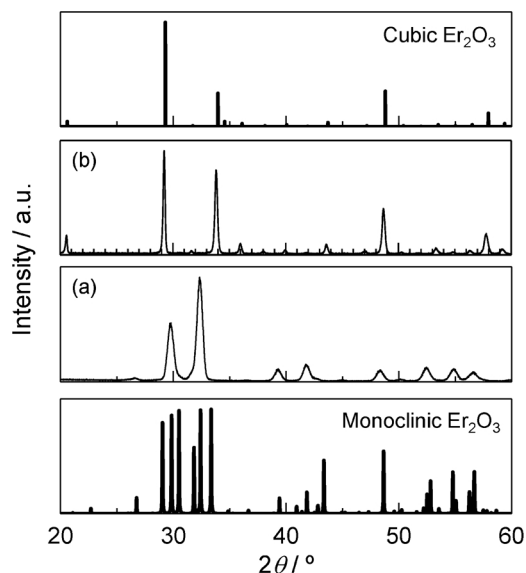


Fig. 1. GIXRD spectra of Er_2O_3 coatings: (a) as deposited and (b) after deuterium permeation measurements. Simulated peaks of monoclinic and cubic Er_2O_3 are also presented [12,13].

where P_0 and D_0 are the pre-exponential factors, E_P and E_D are the activation energy of permeation and diffusion, respectively, R is the gas constant, and T is the temperature. E_P and E_D can be experimentally obtained by temperature dependence of the permeability and diffusivity, and indicate energy barriers in each permeation process. Since the activation energy in permeation includes that of diffusion, the solution behavior can be indirectly discussed.

3. Results

Fig. 1 shows GIXRD spectra of the Er_2O_3 coating samples before and after deuterium permeation measurements. The spectrum of the coating deposited at 600 °C with the bias voltage was clearly different from that after the permeation measurements and powder diffraction file of the cubic Er_2O_3 [12], indicating the as-deposited coating did not have the cubic phase. From calculation of diffraction peaks using lattice information obtained by Ref. [4] and the program PowderCell [13], the peaks found in Fig. 1(a) correspond to the monoclinic phase Er_2O_3 : (310) at 32.4° , (401) at 29.8° , (511) at 41.8° , etc.

Fig. 2(a) and (b) show cross-sectional TEM images of the coatings before and after the permeation measurements. The grain structure is clearly different: the as-deposited coating showed a columnar structure with the column width of up to 100 nm, and the coating after permeation measurements showed a granular structure with larger crystal grains (approximately 520 nm in diameter). The TEM image of the coating irradiated by 6 MeV Fe^{2+} with a damage concentration of 0.05 dpa showed no clear difference from that of unirradiated as shown in Fig. 2(c). Fig. 3 shows grain orientation of the samples analyzed by EBSD. Fig. 3(a) indicates smaller grains in the coating with the monoclinic phase and partly unknown crystal structure or uncrystallized region which is shown as the black area. It was found that the coating had a strong preferred orientation. On the other hand, Fig. 3(b) shows that the coating after the permeation measurements had larger crystal grains of the cubic Er_2O_3 and was highly crystallized, which supports the results of peak positions and sharpness in XRD.

XRD and EBSD data for the irradiated sample were not shown because no clear difference had been observed in comparison with the unirradiated samples, as indicated in TEM observation.

Temperature dependence of deuterium permeability for the unirradiated and Fe-irradiated coating samples are shown in Fig. 4. In the first measurements at 300–500 °C, the unirradiated sample showed 1–2

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