24D08

Development of Tritium Permeation Barrier by Ceramic-Metal Multilayer Structures

金属－セラミック多層構造を用いたトリチウム透過防止膜の高性能化

Takumi Chikada[1], Akihiro Suzuki[1], Hans Maier[2], Takayuki Terai[1] and Takeo Muroga[3]
近田拓未[1]，鈴木晶大[1]，Hans Maier[2]，寺井隆幸[1]，室賀健夫[3]

[1] *School of Engineering, The University of Tokyo*
7-3-1, Hongo, Bunkyo-ku, Tokyo 113-8656, Japan
東京大学大学院工学系研究科　〒113-8656　東京都文京区本郷7-3-1

[2] *Max-Planck-Institut für Plasmaphysik, EURATOM Association*
Boltzmannstrasse 2, Garching, D-85748, Germany
マックスプランクプラズマ物理研究所

[3] *National Institute for Fusion Science*
322-6 Oroshi, Toki, Gifu 509-5292, Japan
核融合科学研究所　〒509-5292　岐阜県土岐市下石町322-6

In order to enhance tritium permeation barrier (TPB) efficiency and compatibility with blanket materials, Er_2O_3-Fe two-layer coatings have been fabricated by physical vapor deposition technique. Fe outer layers showed a good adhesion to the Er_2O_3 inner layer at elevated temperatures. Permeation barrier efficiency increased up to over 10^3 because of an additional contribution by oxidation of the iron layer after deuterium permeation measurements at 700 ºC, while it was not oxidized at up to 650 ºC. It is proved that multilayer structures have advantages on reliability as TPBs as well as a broad selection of coating materials.

1. Introduction

Tritium permeation through structural materials is one of the critical issues of fusion reactor blankets. Recent years, investigations on erbium oxide (Er_2O_3) thin films have been demonstrated as a promising candidate as a tritium permeation barrier (TPB) [1–3]. In our previous study, samples coated on both sides of reduced activation ferritic/martensitic (RAFM) steel substrates showed permeation reduction factors (PRFs) of 10^4–10^5 at 600 ºC [4]. Moreover, a high compatibility with liquid tritium breeder is expected by fabrication of metal coatings as an outer layer to protect corrosion of the ceramic coating [5]. In this study, two-layer coatings with Er_2O_3 and iron (Fe) have been fabricated and deuterium permeation experiments have been carried out in order to discuss permeation behaviors through the multilayer structure.

2. Experimental

Mirror-polished RAFM steel F82H (8Cr-2W, Heat No. 9753 42W-4) plates with dimensions of 25-mm length and 0.5-mm thickness were used as substrates. Er_2O_3 coatings were fabricated by filtered arc deposition described in Ref. [6]. The substrate temperature during deposition was at room temperature. The coating was deposited on one side of the substrate and the thickness of the coating was 1.3 μm. Thereafter metal coatings were deposited on the Er_2O_3 coatings by radio frequency (RF) magnetron sputtering. Fe was employed for the metal coating materials because it has a similar thermal expansion coefficient to Er_2O_3. The substrate temperature during deposition was 400 ºC. RF power was set to 200 W and deposition time was 15 min.

To verify the multilayer structure of the coatings, cross-sectional images of the coatings were observed by a scanning electron microscope (SEM). A chemical analysis can be carried out by energy-dispersive X-ray spectroscopy (EDX). The crystal structure of the coatings was analyzed using grazing incidence X-ray diffraction (GIXRD).

Deuterium permeation phenomena and evaluation methods are described in detail in [2,4]. Deuterium permeation flux through the samples was detected by quadrupole mass spectrometer in the temperature range of 500–700 ºC. The samples were mounted in the permeation apparatus with the coated side facing to the high deuterium pressure side. Driving pressure of deuterium was set at 10^4–10^5 Pa.

3. Results and Discussion

The cross-sectional image of the Er_2O_3-Fe coating is shown in Fig. 1. Although the surface of

the coating turned completely into a metallic one after 15 min-deposition, the thickness of the Fe layer was only approximately 30 nm. The iron layer was also confirmed using EDX and GIXRD. This slow deposition rate, approximately 2 nm/min, is caused by an interference with RF plasma because Fe is a magnetic material. However, the coating was smoothly deposited on the Er_2O_3 coating and no peelings or cracks were observed in the coatings.

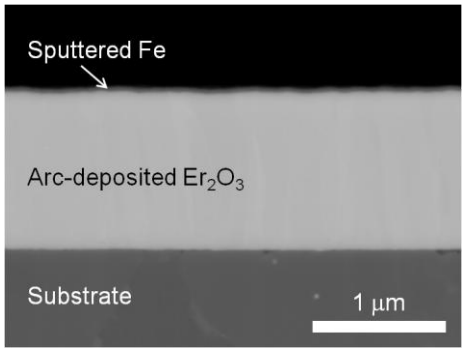

Fig. 1. Cross-sectional SEM image of the Er_2O_3-Fe coating on a F82H substrate. Deposition time of the Fe coating was 15 min.

In the deuterium permeation measurements, a spike of the permeation flux was seen in spite of the stable driving pressure at the beginning of the measurement at 700 ºC. The flux reached a steady-state value after decreasing by 60% from the top of the spike. The drop of the permeation flux was caused by crystallization of the inner Er_2O_3 layer, which had been discussed in our previous study [3]. Higher temperatures were required for the crystallization because of the protection by the Fe layer. After the permeation measurements at up to 700 ºC, the surface appearance of the coating turned rusty, while no change on the surface of the coating was confirmed when measured at up to 650 ºC. The rate limiting process of permeation, which can be evaluated by the driving pressure exponent, changed from diffusion of deuterium atoms into surface reactions after the measurements at up to 700 ºC, indicating that the Fe layer was oxidized and stabilized at 700 ºC. No peeling or cracks were observed after the permeation tests.

A series of deuterium permeability of the coating is shown in Fig. 2. The permeability clearly decreased as repeated the measurement. A PRF value finally reached over 10^3, which is a higher value than that of the Er_2O_3 single layer coating [2–3]. That means both the crystallization of Er_2O_3 and the oxidation of the Fe layer contributed to the enhancement of PRFs. By contrast, as for the coating measured at up to 650 ºC, PRF values showed up to 10^3, indicating that the Fe layer had small surface effects on the deuterium permeation if the Fe layer was not oxidized. These results show that the multilayer structure is a promising concept for the modification of tritium permeation barrier as well as compatibility with blanket materials. Moreover, the concept indicates a possibility that other ceramic materials would apply to inner layers. Investigations to confirm reliability to thermal stress and compatibility with liquid metal breeder should be the next step.

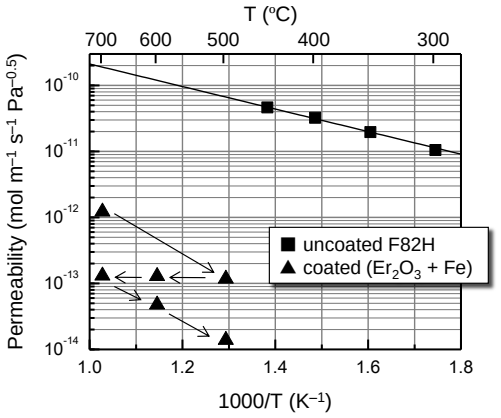

Fig.2. Arrhenius plots of the permeability of F82H substrate and the Er_2O_3-Fe coating. Arrows show the sequence of the permeation measurements.

4. Summary

Fabrication and deuterium permeation of Er_2O_3-Fe two-layer coatings have been carried out. The Fe outer layer deposited by magnetron sputtering had high adhesion to the Er_2O_3 coating at up to 700 ºC. The PRF value showed more than 10^3 after crystallization of the inner Er_2O_3 layer and oxidation of the outer Fe layer. The notion of the multilayer coatings can be extended to other combinations of ceramic-metal materials.

Acknowledgments

This study was supported by KAKENHI (23860017), Ministry of Education, Culture, Sports, Science and Technology, Japan and by the subsidy from Japan Atomic Energy Agency.

References

[1] D. Levchuk, S. Levchuk, H. Maier, H. Bolt and A. Suzuki: J. Nucl. Mater. **367–370** (2007) 1033.
[2] T. Chikada, A. Suzuki, Z. Yao, D. Levchuk, H. Maier, T. Terai and T. Muroga: Fusion Eng. Des. **84** (2009) 590.
[3] T. Chikada, A. Suzuki, T. Kobayashi, H. Maier, T. Terai and T. Muroga: J. Nucl. Mater. **417** (2011) 1241.
[4] T. Chikada, A. Suzuki, C. Adelhelm, T. Terai and T. Muroga: Nucl. Fusion **51** (2011) 063023.
[5] T. Muroga and B.A. Pint: Fusion Eng. Des. **85** (2010) 1301.
[6] F. Koch, R. Brill, H. Maier, D. Levchuk, A. Suzuki, T. Muroga and H. Bolt: J. Nucl. Mater. **329–333** (2004) 1403.