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Characterization of Y_2O_3 coating under neutron irradiation

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Abstract

Ceramic coatings on the surface of structural materials such as 316SS have been considered for electrical insulators and tritium permeation barriers in fusion reactor designs. Y_2O_3 is one of the most promising materials as a coating from a point of high electrical resistivity, etc. In this report, the electrical conductivity of a Y_2O_3 coating was investigated under neutron irradiation with the Japan Materials Testing Reactor (JMTR). The specimen was 316SS with a Y_2O_3 coating, and was irradiated at 300°C in a He atmosphere.

From the results of in situ measurements on the electrical conductivity of a Y_2O_3 coating, the Radiation Induced Conductivity (RIC) was observed at JMTR power-up. The electrical conductivity of the coating before neutron irradiation and under neutron irradiation were about 3×10^{-13} and about 1×10^{-9} 1/Ω/cm, respectively. The electrical conductivity of the coating was constant under neutron irradiation but after neutron irradiation recovered up to that before neutron irradiation. The Radiation Induced Electrical Degradation (RIED) in the Y_2O_3 coating was not recognized up to a fluence of about 5×10^{20} n/cm^2 (>1 MeV). It was clear that the coating had good electrical resistivity under neutron irradiation corresponding to about 4×10^{10} Gy.

1. Introduction

In fusion reactor designs, ceramic coatings on the surface of structural materials such as 316SS have been considered for electrical insulators and tritium permeation barriers. Ceramic coating materials such as Y_2O_3 [1–6], Al_2O_3 [7–11], AlN [11], CaO [12], TiN and TiC [13] have previously been studied, with emphasis on fabrication and properties. On the other hand, in general industrial fields, ceramic and metal coatings are useful for heat resisting coating and anti-corrosion coating. In particular, plasma spraying coating is used widely, because this method is able to apply various materials as a coating and to make thick coatings. Y_2O_3 was selected as a coating, because it was one of the most promising materials as a coating from a point of high electrical resistivity, etc. However, concerning the neutron irradiation data of Y_2O_3, only Hurley et al. [14] have reported on swelling and thermal diffusivity changes.

In the case of ceramic coating fabricated on the surface of austenitic stainless steel (316SS) by the plasma spraying method, it has been observed that crack formation in the coating and a peeling of the coating occur due to the difference in thermal expansion between the coating layer and the substrate. In order to prevent these problems, a buffer layer of ferritic stainless steel (410SS) was fabricated by the plasma spraying method under the Y_2O_3 layer, because 410SS has a thermal expansion coefficient close to the ceramic coating [1–4]. Plasma sprayed coating has also a lot of pores. They were successfully filled by the chemically densification method, where the coated specimen was immersed in $Y(NO_3)_3$ solution and fired at 500°C [4].

In this report, the electrical conductivity of a Y_2O_3 coating was investigated under neutron irradiation with Japan Materials Testing Reactor (JMTR). The specimen was 316SS with a Y_2O_3 coating, and was irradiated at 300°C in a He atmosphere.

2. Specimen

The fabrication process for Y_2O_3 coating on the surface of 316SS developed by the authors [1–4] is shown in Fig. 1. Some properties were investigated in relation to some parameters for fabrication [4]. First of

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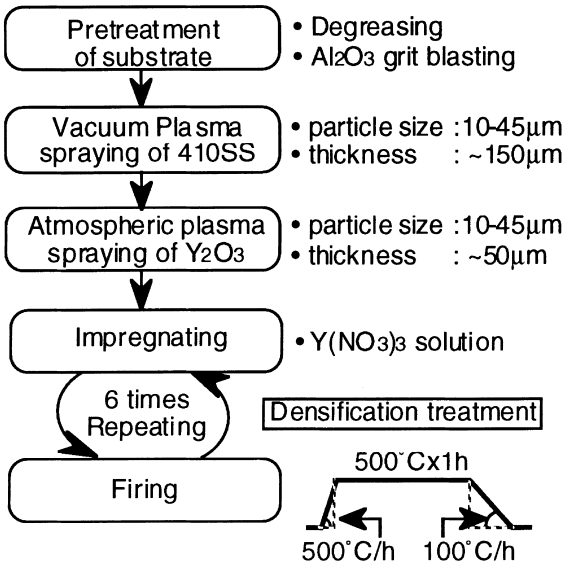

Fig. 1. Fabrication process of the Y_2O_3 coating.

all, the number of densification treatments was investigated by the hardness test in the cross section of the Y_2O_3 coating. Second, on the basis of the results on the thermal shock test of undercoatings performed by two plasma spraying methods (the atmospheric plasma spraying method and the vacuum plasma spraying method) and two kinds of 410SS powders with different particle sizes (10–45 μm and 10–75 μm), the fabrication condition was selected. Finally, the relation between

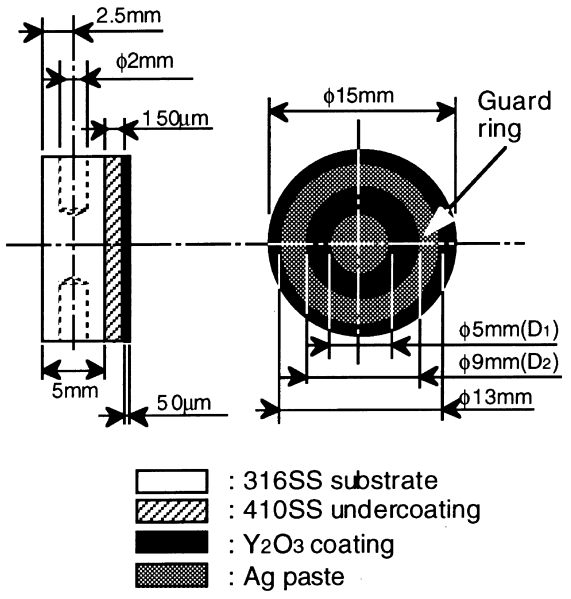

Fig. 2. The shape of 316SS with Y_2O_3 coating specimen for neutron irradiation.

Table 1
Spraying conditions of 410SS by the vacuum plasma spraying method

Spraying apparatus	PLASMA-TECHNIK AG A-2000V
Particle size	10–45 μm
Plasma gases	
Ar	7.5×10^{-4} m^3/s
H_2	1.3×10^{-4} m^3/s
Plasma current	685 A
Plasma voltage	64 V
Spray distance	275 mm
Pressure in chamber	5.9 kPa

undercoating condition and thermal shock resistivity of the Y_2O_3 coating was investigated. It was obvious from the results of the thermal shock test that the thermal shock resistivity of Y_2O_3 coatings could be improved on 410SS undercoating fabricated by the vacuum plasma spraying method using 10–45 μm 410SS particle size [4]. From the results of the hardness test in the cross section of Y_2O_3 coating, six repetitions of the densification peocesses were used.

The shape of specimen of 316SS with a Y_2O_3 coating used for the electrical conductivity measurement under neutron irradiation is shown in Fig. 2. The size of 316SS substrate was $\phi 15 \times {}^{t}5$ mm. The surface of the substrate was degreased by methyl alcohol and Al_2O_3 grit blasted to improve the adhesion between the 316SS substrate and the 410SS undercoating. The undercoating was coated about 150 μm thick by the vacuum plasma spraying method. The particle size of 410SS (Fe–12 wt% Cr) was from 10 to 45 μm. The spraying conditions of 410SS are shown in Table 1. The Y_2O_3 (purity: 99.95 wt%, particle size: 10–45 μm) was coated about 50 μm thick by the atmospheric plasma spraying method. The spraying conditions of Y_2O_3 are shown in Table 2. The Y_2O_3 coating was densified by impregnation with $Y(NO_3)_3$ solution and fired for one hour at 500°C in order to close open pores and cracks. The rate of heating and cooling were 500°C/h and 50°C/h, respectively. Furthermore, this densification treatment was repeated six times [4].

Table 2
Spraying conditions of Y_2O_3 by the atmospheric plasma spraying method

Spraying apparatus	PLASMA-TECHNIK AG A-3000S
Particle size	10–45 μm
Plasma gases	
Ar	7.0×10^{-4} m^3/s
H_2	2.0×10^{-4} m^3/s
Plasma current	550 A
Plasma voltage	74 V
Spray distance	120 mm

The guard electrode and the center electrode were formed by Ag paste on the surface of Y_2O_3 coating. The outer diameter and inner diameter (D_2) of the Ag guard electrode were 13 and 9 mm, respectively. The diameter of the Ag center electrode (D_1) was 5 mm (see Fig. 1). Ag paste was applied by heating for 20 min at 150°C.

3. In situ measurement

The electrical conductivity of the Y_2O_3 coating under neutron irradiation in JMTR was measured in-situ using a three-electrode guard-ring configuration of Ag electrodes as shown in Fig. 3.

The equations for electrical conductivity are as follows:

$$\frac{I}{\rho} = \frac{A}{d}R, \tag{1}$$

$$A = \frac{\pi(D_1 + g)^2}{4}, \tag{2}$$

$$g = \frac{D_2 - D_1}{2}, \tag{3}$$

where ρ: Electrical conductivity (1/Ω/cm), R: Resistance (Ω), A: Effective cross section of electrode (cm^2), g: Gap between guard and center electrode (cm), d: Thickness of coating (cm), D_1: Center electrode diameter (cm) (see Fig. 2), D_2: Guard electrode inner diameter (cm) (see Fig. 2).

The inner capsule configuration for neutron irradiation is shown in Fig. 4. Mineral insulated cables were used for electrical measurements and sintered Al_2O_3 (purity: 99 wt%) used as the specimen holder. The specimen was applied DC 100 V and irradiated at 300°C in He atmosphere. The thermal neutron flux (<0.6826 eV) and the fast neutron flux (>1 MeV) were 2×10^{14} and 4×10^{13} n/cm^2/s, respectively.

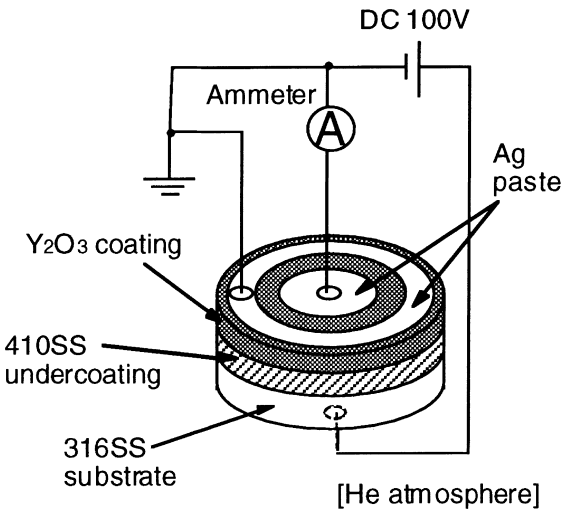

Fig. 3. The measurement method of electrical conductivity with a three-electrode guard-ring configuration.

4. Results and discussion

The result of electrical conductivity measurement with and without neutron irradiation is shown in Fig. 5. The electrical conductivity of Y_2O_3 coating increased with increasing reactor power at power-up. This phenomenon was named as the Radiation Induced Conductivity (RIC). This increasing of electrical conductivity of ceramic insulators under irradiation was understood to correspond to the photo-conductivity of the semiconductor [7]. The valence electrons in the insulator are excited by the ionization effect of the radiation and jump from the valence band to the conduction band. These excited electrons give rise to the increase of electrical conductivity. This effect is only observed under irradiation, and dose not occur without irradiation. That

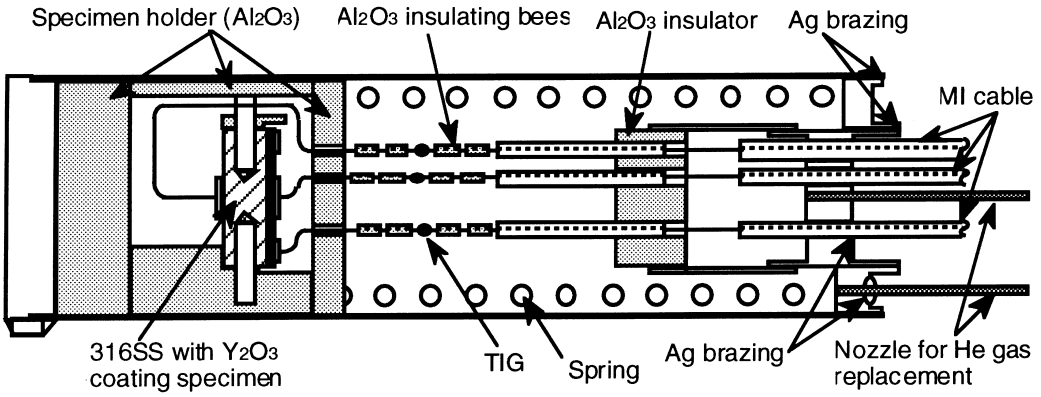

Fig. 4. Inner capsule configuration for neutron irradiation.

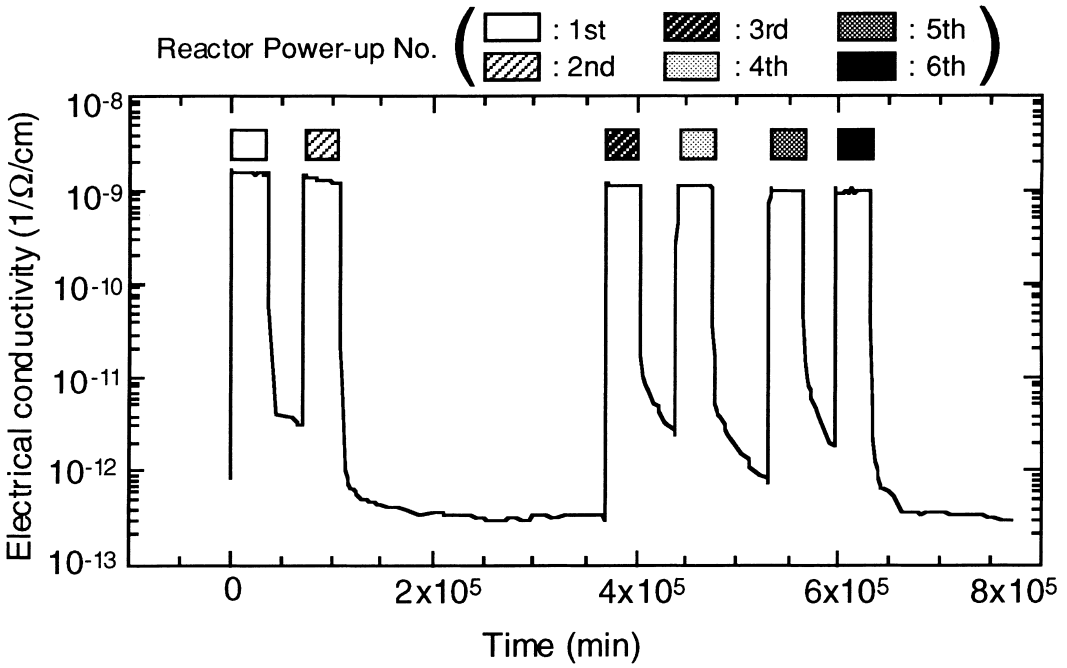

Fig. 5. The results of electrical conductivity measurement with and without neutron irradiation.

is why the lifetime of the conduction electrons excited by irradiation in the conduction band is very short.

The electrical conductivity of the Y_2O_3 coating before the 1st reactor power-up was about 3×10^{-13} 1/Ω/cm. The electrical conductivity of the coating after the 2nd and the 6th reactor power-up recovered up to the electrical conductivity before the 1st reactor power-up. However, the electrical conductivity after the 1st, the 3rd, the 4th and the 5th reactor power-up did not recover to the same level. After the 2nd and the 6th reactor power-up, fuel elements replaced all the dummy fuel elements so that the gamma-ray source in the JMTR core was very low. Therefore, this phenomenon occurred as a result of the gamma-ray irradiation from the used fuel elements used for JMTR operation.

The electrical conductivity of the Y_2O_3 coating under neutron irradiation was about 1×10^{-9} 1/Ω/cm. The electrical conductivity of the coating was constant under neutron irradiation at JMTR power operation. The Radiation Induced Electrical Degradation (RIED) in Y_2O_3 coating was not recognized up to about 5×10^{20} n/cm^2 (>1 MeV). It was clear that the Y_2O_3 coating had good electrical resistivity (about 1×10^{-9} 1/Ω/cm) under neutron irradiation corresponding to about 4×10^{10} Gy.

The results of the electrical conductivity measurement at reactor power-up in a log-log plot is shown in Fig. 6. The electrical conductivity at the 1st reactor power-up was higher than that in the later irradiations. Possible reasons were the elimination of the moisture on the specimen and the mitigation of the initial defect by the irradiation [8,9].

The RIC was recognized at reactor power-up. The RIC is described as below [7–9]

$$\mathrm{RIC} = KP^{\delta}, \tag{4}$$

where: *RIC*: Radiation induced conductivity, *K*: Constant of proportionality (ionizing efficiency), *P*: Ionizing dose rate, δ: Constant.

Parameters K and δ are constants strongly depending on materials (δ is usually equal to 0.5–1.0) [10]. Shikama et al. evaluated the electrical properties of single crystal alumina under neutron irradiation at JMTR [7–9] and found that δ was in the range of 1.0–1.2. Noda et al. evaluated the electrical properties of single crystal alumina under 14 MeV neutron irradiation using the Fusion Neutronics Source (FNS) [9]. The magnitude of δ was found to be in the range of 0.90–0.96. With regard to the RIC in the Y_2O_3 coating, it was found that the magnitude of δ was in the range of 0.76–0.87 at reactor power-up while the magnitude of K was in the range of 9.7×10^{-13}–3.4×10^{-12}.

5. Conclusion

The RIC on Y_2O_3 coating was recognized at each JMTR power up. The RIC on Y_2O_3 coating is described by the following relation

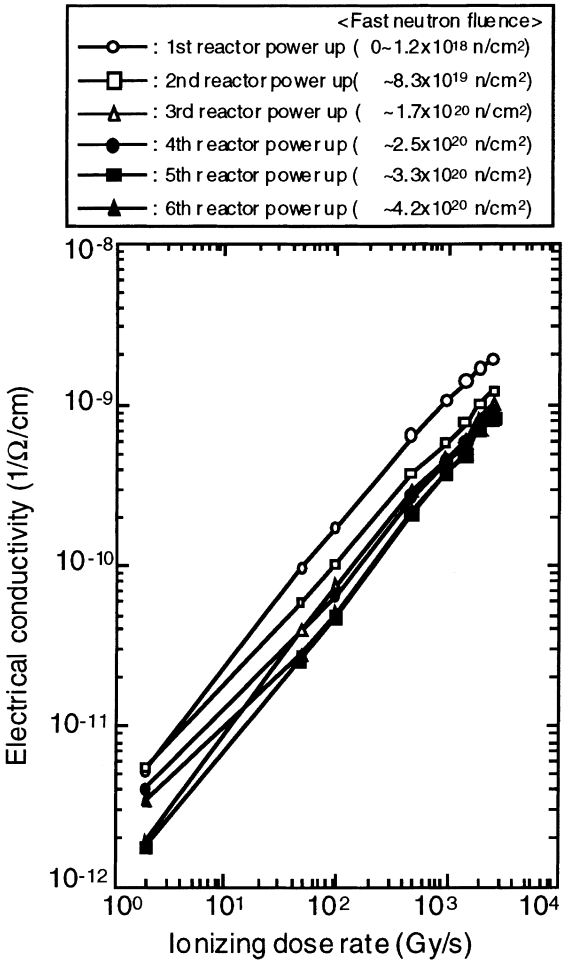

Fig. 6. The results of electrical conductivity measurements at reactor power-up.

$$\mathrm{RIC} = K \times P^{\delta} \quad (\delta\text{: } 0.76 - 0.87).$$

The parameter δ is a constant strongly depending on material. The magnitudes of δ for single crystal alumina under fission neutron irradiation and under 14 MeV neutron irradiation were in the range of 1.0–1.2 and 0.90–0.96, respectively. It was clear that the constant δ for the Y_2O_3 coating was in the range of 0.76–0.87 under fission neutron irradiation.

The electrical conductivity of the Y_2O_3 coating before neutron irradiation and under neutron irradiation was about 3×10^{-13} and about 1×10^{-9} 1/Ω/cm, respectively. The electrical conductivity of the coating was constant under neutron irradiation but was found to recover to that before neutron irradiation. The RIED in the Y_2O_3 coating was not recognized up to about 5×10^{20} n/cm^2 (>1 MeV). It was clear that the Y_2O_3 coating had good electrical resistivity under neutron irradiation corresponding to about 4×10^{10} Gy.

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