Effects of specimen thickness and impurity on the conductivity of alumina under electron irradiation

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Abstract

The electrical conductivity of undoped (125, 332 and 545 μm-thick) and Cr³⁺ doped (332 μm-thick) α-Al₂O₃ single crystals was measured under electron irradiation at temperatures from 300 to 723 K. The conductivity under the ITER environment is found to be less than the limiting conductivity of 10⁻⁴ S/m required for insulators in ITER. Although the Cr³⁺ doping increases the electrical conductivity, it suppresses the radiation induced conductivity at higher ionization fields. The impurities in Cr³⁺ doped α-Al₂O₃ are acting as trapping sites or recombination centers for carriers, affecting the electrical resistivity. The radiation induced conductivity in undoped α-Al₂O₃ increases with increasing specimen thickness, which is explained in terms of the increase of energy absorption with increasing specimen thickness. No radiation induced electrical degradation was found in both undoped and Cr³⁺ doped α-Al₂O₃ in the present study.

1. Introduction

Ceramic materials are expected to be candidate materials for insulators and radiofrequency windows in the International Thermonuclear Experimental Reactor (ITER), where the electrical conductivity of insulators should be less than 10⁻⁶ and 10⁻⁴ S/m for magnetic coils and other uses, respectively. α-Al₂O₃ is one of the promising insulating ceramic materials for ITER and commercial reactors, because of its high electrical resistivity. However, it is well known that the electrical conductivity dramatically increases in radiation fields due to radiation induced conductivity (RIC) [1–5], thermally stimulated conductivity (TSC) and/or radiation induced electrical degradation (RIED) [6–9]. Those phenomena have been believed to be critical issues for the application of α-Al₂O₃ to insulators in fusion reactors. The RIC, RIED and TSC are influenced by impurities because they are caused by impurity levels in the forbidden band. Furthermore, RIC may change with the specimen thickness due to the difference of deposited electron charge and energy in various specimen thickness. However, few studies on the effect of impurity and specimen thickness in α-Al₂O₃ have been done. In the present study, in situ measurements of electrical conductivity for undoped and Cr³⁺ doped α-Al₂O₃ with different specimen thickness under electron irradiation have been performed in order to investigate the effect of specimen thickness and impurity on RIC, RIED and TSC, considering the application of α-Al₂O₃ to insulators in ITER.

2. Experimental

Specimens used in the present study were α-Al₂O₃ single crystals of 5.0 mm Ø and 125, 332 and 545 μm-thick (undoped, Kyocera and Union Carbide), and 5.0 mm Ø and 332 μm-thick (Cr³⁺ doped, Union Carbide). The measurement of the electrical conductivity was performed based on the three-terminal method using a
Hewlett-Packard 4339A high resistance meter. Center, guard and ground electrodes were made by using vapor deposited titanium on the specimens. The electrical conductivity was measured under 1 MeV electron irradiation with a flux of \(1.4 \times 10^{18} \text{ e/m}^2 \text{s} \) (\(1.6 \times 10^4 \text{ Gy/s} \) and \(8.7 \times 10^{-9} \text{ dpa/s} \)) and an electric field of 300 kV/m at temperatures from 300 to 723 K in a high voltage electron microscope (HVEM, JEM-1000) at the HVEM Laboratory, Kyushu University.

3. Results and discussion

The temperature dependence of the electrical conductivity of undoped and \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) (Union Carbide) without irradiation is shown in Fig. 1. The electrical conductivity of \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) is larger than that of undoped \(\alpha\)-Al\(_2\)O\(_3\) by one or two orders of magnitude in the temperature range of measurements. The apparent activation energies of undoped and \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\), as obtained from the slopes of the lines, vary between 0.62–0.77 and 0.65–0.81 eV, respectively. These apparent activation energies correspond to the energy from the bottom of the conduction band to the impurity levels which contribute to the electrical conductivity without irradiation, and their values are much less than 3.75 eV [10] for the \(\text{Cr}^{3+}\) level in \(\alpha\)-Al\(_2\)O\(_3\).

Fig. 2 shows the electrical conductivity of undoped and \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) as a function of the electron flux at 723 K. The electrical conductivity of undoped and \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) is enhanced through RIC, and it is evaluated to be less than the limiting conductivity of \(10^{-4} \text{ S/m} \) under the highest dose rate in ITER environments (\(3 \times 10^{3} \text{ Gy/s} \)) at 723 K. Although the electrical conductivity of \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) is higher than that of undoped one in the lower dose rate, this situation might be turned around at the higher dose rate. The RIC, \(\sigma_{\text{RIC}}\), is defined as the difference between the electrical conductivity under irradiation, \(\sigma\), and that without irradiation, \(\sigma_0\), and it is phenomenologically expressed by \(\sigma_{\text{RIC}} = \sigma - \sigma_0 = A \cdot \phi^\gamma\), where \(A\) is a constant, \(\phi\) the ionizing dose rate and \(\gamma\) the dose rate exponent. The RIC of undoped and \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) is shown in Fig. 3 as a function of electron flux. The RIC is linearly proportional to electron flux, indicating the RIC to be induced by electronic excitation. The RIC of \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) is less than that of undoped \(\alpha\)-Al\(_2\)O\(_3\) by one order of magnitude, implying that doped \(\text{Cr}^{3+}\) ions act as trapping and/or recombination sites for carriers. It is also speculated, based on the results shown in Fig. 3, that the trapping and/or recombination rates are proportional to the concentration of trapping and/or recombination sites and the ionizing dose rate. The lower electrical conductivity of \(\text{Cr}^{3+}\)-doped \(\alpha\)-Al\(_2\)O\(_3\) compared to that of undoped one at high dose rate can be understood on the basis of this speculation.

The specimen thickness dependence of undoped \(\alpha\)-Al\(_2\)O\(_3\) (Kyocera) was also measured at 273, 473 and 723 K. The RIC has been found linearly proportional to the electron flux at all temperatures and the proportional constant increases with increasing irradiation temperature. Fig. 4 shows the specimen thickness dependence of the RIC at an electron flux of \(1.4 \times 10^{18} \text{ e/m}^2 \text{s} \). The RIC super-linearly increases with increasing specimen thickness, in accordance with the specimen thickness dependence of the absorbed energy in aluminum, which is shown in Fig. 5 as a function of the specimen thickness.
in aluminum under 3 MeV electron irradiation [11]. The profile, in Fig. 5, of absorbed energy in $\alpha$-$\text{Al}_2\text{O}_3$ has little difference from that in aluminum. The mean range of 1 MeV electrons in $\alpha$-$\text{Al}_2\text{O}_3$ is 1.1 mm. The dotted lines indicate the specimen thickness of $\alpha$-$\text{Al}_2\text{O}_3$ used in the present study. As seen in Fig. 5, the absorbed energy super-linearly increases with increasing specimen thickness up to 545 $\mu$m. It is important that the electrical conductivity of the insulators is correctly estimated in ITER and commercial reactors, because the size of the insulators is designed in the rage from the mm region to the m region.

The electrical conductivity of undoped and $\text{Cr}^{3+}$ doped $\alpha$-$\text{Al}_2\text{O}_3$ was measured by rapid change of temperature during beam-off periods, with the results shown in Fig. 6(a) and (b), respectively. The increase of the electrical conductivity during increasing temperature, which is called TSC, is noted by the bold lines in Fig. 6. The intrinsic increase of electrical conductivity with increasing temperature is small, although the TSC is included. The TSC of undoped $\alpha$-$\text{Al}_2\text{O}_3$ increases monotonically with increasing temperature up to 673 K but decreases above 673–723 K, contrasting to a
monotonic increase up to 723 K for Cr$^{3+}$ doped $\alpha$-Al$_2$O$_3$ (note the log scale in Fig. 6). It is well known that the TSC is due to thermal excitation of the electrons trapped by the level of impurity or defect in forbidden band. The TSC is expressed as $\sigma_{TSC} = A \cdot \exp(-E/kT)$, where $A$ is a constant, $E$ activation energy, $k$ Boltzmann constant and $T$ temperature. The activation energy of undoped and Cr$^{3+}$ doped $\alpha$-Al$_2$O$_3$ is estimated from the temperature dependence of the TSC to be 0.51 and 0.63 eV, respectively. These values are comparable to the apparent activation energy obtained from the temperature dependence of the electrical conductivity.

The electrical conductivity was measured under 1 MeV electron irradiation with a flux of $1.4 \times 10^{19}$ e/m$^2$ s ($10^5$ Gy/s) at 723 K up to a high fluence level of about $6.0 \times 10^{22}$ e/m$^2$ ($4.3 \times 10^9$ Gy). The electrical conductivity increased quickly under electron irradiation and then kept constant up to $1.0 \times 10^{-4}$ dpa. This means that no RIED was found in both undoped and Cr$^{3+}$ doped $\alpha$-Al$_2$O$_3$ under electron irradiation up to $1.0 \times 10^{-4}$ dpa at 723 K.

4. Conclusions

The electrical conductivity of undoped and Cr$^{3+}$ doped $\alpha$-Al$_2$O$_3$ under ITER environment is less than $10^{-4}$ S/m which is the limiting conductivity for the insulators in ITER, except for magnetic coils use. The electrical conductivity of Cr$^{3+}$ doped $\alpha$-Al$_2$O$_3$ is less than that of undoped $\alpha$-Al$_2$O$_3$ in the high dose rate because RIC is reduced by doping Cr$^{3+}$. The RIC of undoped $\alpha$-Al$_2$O$_3$ increases with increasing specimen thickness. Therefore, the effects of thickness should be considered for the insulators under $\alpha$-particle environment. The shallow levels which dominate the electrical conductivity without irradiation may contribute to TSC. No RIED was found in the present study implying that Cr$^{3+}$ dose not influence the occurrence of RIED under 1 MeV electron irradiation up to $1.0 \times 10^{-4}$ dpa at 723 K.

References