Electrical conductivity of Wesgo AL995 alumina under fast electron irradiation in a high voltage electron microscope

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Electrical conductivity of a 295-μm-thick Wesgo AL995 alumina has been measured before and during 1 MeV electron irradiation in a dc electric field of 300 kV/m at temperatures up to 723 K. The difference between the activation energies before (0.49 ± 0.02 eV) and during (0.19 ± 0.06 eV) irradiation indicates a substantial impact of irradiation on the conductivity of Wesgo AL995 alumina. The electrical conductivity of Wesgo AL995 alumina is lower by approximately 2 orders of magnitude than its requirement for the magnetic coils in the international thermonuclear experimental reactor (ITER). Thermal disruption may not impact on Wesgo AL995 alumina insulating material in ITER because of the absence of thermally stimulated conductivity peaks in it. Although no substantial bulk degradation is observed under irradiation up to a fluence of 7.0×10^{22} e/m^2 (7.97×10^{-5} dpa) at 723 K, surface degradation is detected that could limit the application of Wesgo AL995 in ITER as a potential insulator. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1494845]

I. INTRODUCTION

Alumina is considered to be a potential insulating material for numerous applications in fusion devices for heating and current drive and diagnostics, because of its superior sustaining behavior against radiation among all ceramic insulators.[1] For insulator applications it is necessary to understand the basic electrical properties of alumina. Radiation induced conductivity (RIC), thermal stimulated conductivity (TSC), and radiation induced electrical degradation (RIED)[2] of alumina under irradiation are the critical issues for its application to fusion devices. Although many results for RIC, TSC, and RIED have been published, the RIED phenomenon is still controversial. Namely, several research groups failed to observe RIED in some grades of alumina where RIED should occur. Furthermore, severe surface degradation together with poor electrical contacts could cause an apparent bulk RIED.[3]

In order to confirm surface and bulk degradation, electrical conductivity measurements have been done on Wesgo AL995 alumina over a wide range of ionizing dose rates under various radiation sources and detected no bulk RIED to 3 dpa regardless of radiation sources.[3–10] This article gives additional insights not only into RIED but also into RIC and TSC of Wesgo AL995 alumina with a 1 MeV electron irradiation relevant to the international thermonuclear experimental reactor (ITER) operating condition.

II. EXPERIMENTAL PROCEDURE

In the experiments, the specimen was taken from a single bar of Wesgo AL995 alumina used in the round robin tests. The diameter and thickness of specimen was 5.5 mm and 295 μm, respectively. Titanium was deposited in vacuum to make the three-electrode system. A center electrode of 2 mm and a guard electrode of 3.5 mm inner and 4.5 mm outer diameter were prepared on the top and a base electrode of 4.5 mm was prepared on the bottom of the specimen. The contact resistance of both the guard and center electrodes was 30–40 Ω, being measured by a tester. The temperature dependence of electrical conductivity before irradiation was measured in a bell jar at 10^{-4} Pa from room temperature (RT) to 723 K. Temperature was increased at the rate of 3 K/min. The irradiation experiments were carried out in a high voltage electron microscope (HVEM) under 1 MeV electrons beam-on and -off conditions at temperatures ranging from RT to 723 K. The pressure of the HVEM was 10^{-5} Pa. Irradiation was done only to the center electrode of the specimen. Both bulk and surface conductivity were measured under irradiation with an electric field of 300 kV/m. Details of the specimen holders are given elsewhere.[11,12]

III. RESULTS AND DISCUSSION

A. Temperature dependence of conductivity

Figure 1 shows the electrical conductivity of Wesgo AL995 alumina against the reciprocal temperatures ranging from RT to 723 K with and without a 1 MeV electron irradiation dose rate of 8.7×10^4 Gy/s in the dc electric field of 300 kV/m. The electrical conductivity increases with increasing temperature, but with different activation processes. The estimated activation energy before irradiation is 0.49 ± 0.02 and 0.40 ± 0.01 eV during increasing and decreasing temperatures, respectively, and it reduces to 0.19 ± 0.06 eV during irradiation. In fact, the conductivity slowly changes up to an irradiation temperature of about 500 K and then is abruptly raised at higher temperatures. The preirradiation resistivity of Wesgo AL995 at RT is 3.6×10^{10} $\Omega\,$m (2.8

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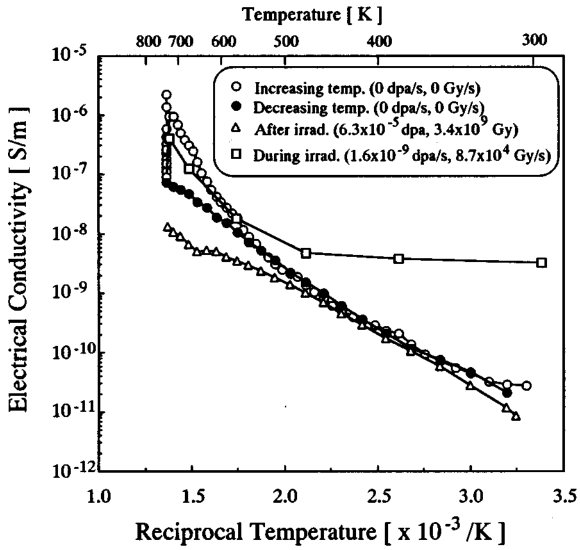

FIG. 1. Electrical conductivity as a function of reciprocal temperature for a 295-μm-thick Wesgo AL995 alumina specimen before, during, and after irradiation with a 1 MeV electron flux of $1.4\times10^{18} e/\mathrm{m}^2$ s in an electric field of 300 kV/m.

$\times10^{-11}$ S/m) in this experiment, being smaller than other experimental results. The amplitude of volume conductivity and the overall activation behavior of Wesgo AL995 alumina before and after a long period of irradiation in our experiment are consistent with those of Morono and Hodgson.[6] Unfortunately, the RIC versus temperature result is not present in their study. The agreement between the common data in both studies indicates the reliability and benefit of the round robin experiments on Wesgo AL995.

B. Dose rate dependence of conductivity

Figure 2 shows the electron dose rate dependence of a 295-μm-thick Wesgo AL995 alumina on RIC under 1 MeV electrons with increasing and decreasing beam intensity at various temperatures. The RIC is defined by $\sigma_{\mathrm{RIC}}=\sigma-\sigma_0$, where σ and σ_0 are the conductivity with beam-on and -off, respectively. The electrical conductivity proportionately increases with increasing dose rate and irradiation temperature. No substantial difference in the RIC during increasing and decreasing beam intensity is found at all temperature except at 723 K. Temperature sensitivity of the conductivity rather than irradiation flux is found in Wesgo AL995 and is comparatively weaker than that of the Kyocera alumina,[12] because of the thermal effect of the impurities present in the Wesgo AL995 alumina. Generally the electrical conductivity under irradiation is expressed by the equation $\sigma=\sigma_0+kR^{\delta}$, where σ_0 is the conductivity without irradiation, k is a material dependent constant, R is the ionizing dose rate, and δ is the ionizing dose rate exponent. The value of δ was estimated from the experimental curves shown in Fig. 2 by fitting the above equation. The estimated values of k vary from 3.3×10^{-14} to 1.8×10^{-12} s/(Gy Ω m) and δ vary from 0.91 to 0.59 for a 295-μm-thick specimen at RT and 723 K, respectively. Delta values of nearly unity are well understood to be related to charging and trapping of electrons in defect and impurity levels below the conduction band.[13,14] A decrease in the δ value with increasing temperature is observed, indicating changes in the number of trapping centers and/or levels.

The RIC results of Wesgo AL995 alumina of our work compared with those of other groups of round robin tests are shown in Fig. 3. Our results include the data from RT to 723 K at temperatures increased by about 100 K. The applied electric field and the ionizing dose rate are 300 kV/m and $\sim10^4-10^5$ Gy/s, respectively. The compiled data vary in the ionizing dose rate of 10^1-10^6 Gy/s. The irradiation temperature ranges between 673 and 773 K in most of the RIC measurements except for the HFIR tests,[9,15] where the temperatures are maintained at $\sim$314 and 440 K. The applied electric field ranges between 100 and 500 kV/m. However, the relative amplitude of the compiled works of Wesgo AL995 satisfy the requirements of conductivity of 10^{-6} S/m and dose rate of 2000 Gy/s (Ref. 9) for the magnetic coils in the ITER. The results presented here also indicate that the ITER requirements will be met.

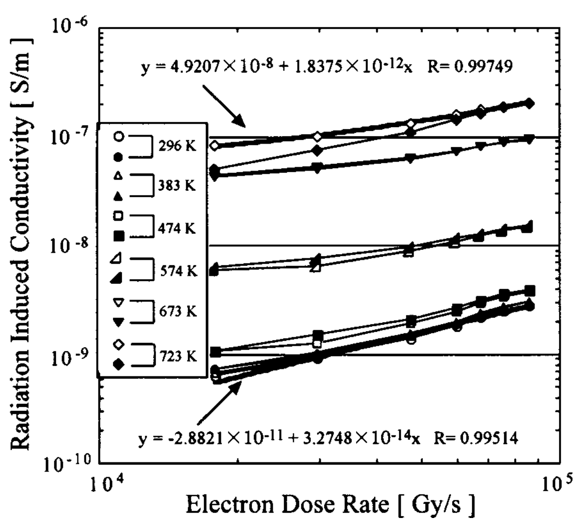

FIG. 2. Temperature dependence of RIC of a 295-μm-thick Wesgo AL995 alumina specimen under 1 MeV electrons irradiation with increasing (open symbol) and decreasing (filled symbol) beam intensity from 296 to 723 K.

C. Thermally stimulated conductivity (TSC)

Since the thermal disruption effect under the ITER condition is found in sapphire,[12] TSC measurements are also realized as being important in Wesgo AL995 alumina. For TSC measurements, the irradiations are composed of alternating times of irradiation for 1800 s followed by annealing at the temperature for 3600 s and then annealing at a higher temperature by about 100 °C for 3600 s as shown in Fig. 4. The conductivity measurements were done during isothermal periods at 5 min after the increase in annealing temperature to allow the specimen to reach thermal equilibrium. When turning on the beam at RT, the electrical conductivity is abruptly raised from 1.0×10^{-11} to 3.5×10^{-9} S/m and then it changes a little due to the accumulation of point defects acting as trapping centers. The difference between the beam-on and -off conductivity decreases with increasing

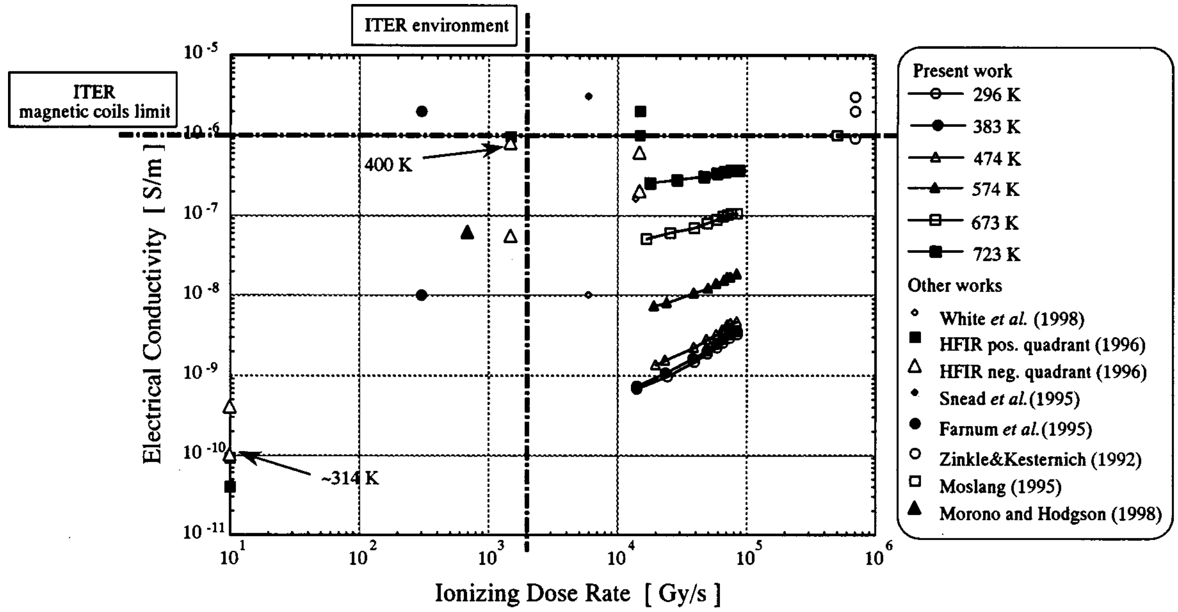

FIG. 3. Compilation of ionizing dose rate dependence of electrical conductivity of Wesgo AL995 alumina of different works at temperatures between 673 and 773 K except for the mentioned temperatures compared with those of our work. The present work includes data at RT, 383, 474, 574, 673, and 723 K.

temperature unlike Kyocera alumina,[4] indicating strong thermal dependence of conductivity in Wesgo AL995 alumina. No transient conductivity peak similar to that in single crystalline Kyocera alumina with Pt paste electrodes under beam-off at 623 K is found.[12] The possible reason is the difference in the type of specimen where Kyocera alumina used is single crystal and Wesgo AL995 alumina is polycrystalline alumina with grains of $\sim$30 μm. If any charge is released from the traps in Wesgo AL995 alumina, it might be circulated on the surface along the grain boundaries. The fact that there are no TSC peaks in polycrystalline Wesgo AL995 alumina indicates that any temperature disruption effects on the electrical conductivity of Wesgo AL995 alumina would not be a limiting factor to its application in ITER as magnetic coils insulating materials.

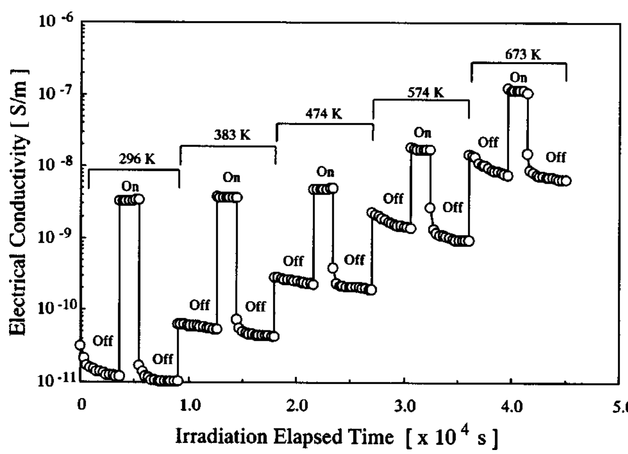

FIG. 4. Temperature dependence of electrical conductivity for a 295-μm-thick Wesgo AL995 alumina specimen under irradiation with a 1 MeV electron flux of $1.4\times10^{18} e/m^2$ s at beam-on and -off conditions.

D. Dose dependence of conductivity

1. Bulk conductivity

Bulk conductivity of a 295-μm-thick Wesgo AL995 alumina irradiated with a 1 MeV electron flux of 1.4×10^{18} e/m^2 s in an electric field of 300 kV/m of this study compared to the existing data of RIED experiments on Wesgo AL995 alumina is shown in Fig. 5.[3,5,6,8,10,16] A slight discrepancy in the absolute values of some data of compilation results may appear. No bulk RIED is found in this study. The postirradiation conductivity at 723 K is lower than that of the preirradiation. The specimen thickness, applied electric field, as well as temperature used in different experiments ranges from a few micrometers to 1 mm, $\sim$50–1000 kV/m, and 600–800 K, respectively. Moslang *et al.*[5] failed to find RIED in Wesgo AL995 alumina to 0.014 dpa using 104 MeV α particles in a dc electric field of 100 kV/m at 723 K. A decreasing trend of conductivity in Wesgo AL995 alumina was found by Kesternich *et al.*[3,17] to a total dose of 2.5×10^{-3} dpa under 28 MeV He ion irradiation with 350 kV/m at 723 K. Both the works of Farnum and Clinard[7,8] showed the decrease of conductivity by 1 order of magnitude throughout the total damage level under neutron irradiation. No RIED was observed to a dose of 0.02 dpa but surface and gas conduction was observed. Another research group at ORNL has done extensive irradiation measurements on Wesgo AL995 alumina with fission neutrons to 1.4 dpa at 610–640 K and found no evidence of RIED.[9] Morono and Hodgson[16] have found no bulk RIED detected in Wesgo AL995 to a dose of $\sim10^{-4}$ dpa with a 1.8 MeV electron irradiation for dc electric fields of 100 and 500 kV/m, but bulk degradation has been detected at 1000 kV/m. They explained the electric field threshold for RIED by a simple model based on electron acceleration in the conduction

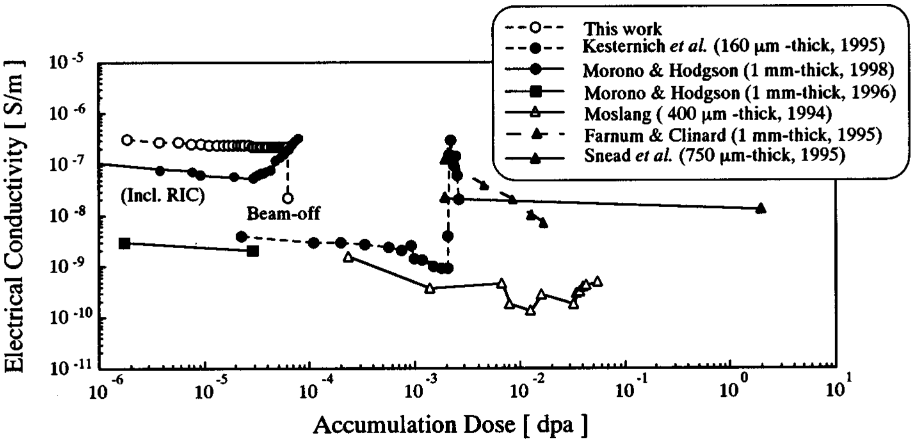

FIG. 5. Compilation of RIED measurements of different works on Wesgo AL995 alumina (see Refs. 3, 5, 6, 8, 10, and 16) irradiated at temperatures of ~623–830 K compared with this work. The damage rate of this work is 1.6×10^{-9} dpa/s.

band.[18] Inconsistency in the electrical conductivity about 1 order of magnitude among RIED results is observed in Fig. 5. We have found thickness dependent electrical conductivity of sapphire[19] and Wesgo AL995[4] alumina irradiated with 1 MeV electrons in a dc electric field of 300 kV/m at 723 K, however the thickness dependence of conductivity is not regarded in Fig. 5. Although no bulk RIED is detected in Wesgo AL995 alumina under various irradiation sources, more international round robin experiments should be performed not only on a single specimen, but also under similar experimental conditions for specimen thickness, applied electric field, etc.

2. Surface conductivity

Figure 6 shows the dose dependence of surface conductivity of a 295-μm-thick Wesgo AL995 alumina with a 1 MeV electron irradiation flux of 1.4×10^{18} e/m^2 s at 723 K. The surface conductivity increases very quickly after turning on the beam to a value higher than that of the unirradiation and then decreases with time until 2.0×10^{22} e/m^2. After that, the surface conductivity increases until the specimen shows overload current. Specimen overload current is found after irradiation to a damage level of 7.97×10^{-5} dpa. The overload surface current did not return even after keeping the specimen at 723 K for 60 min while turning off the applying voltage to the specimen. In addition, the surface current did not revert to that of the RT in the applied electric field condition. This evidence indicates that the surface of the specimen is degraded. As previously mentioned Wesgo AL995 was found to be very sensitive to surface electrical degradation irradiation with a 1.8 MeV electron to the top whole surfaces in vacuum.[6,16] Large grain size and/or low density of Wesgo AL995 alumina was thought to be responsible for this degradation. The surface of the degraded specimen between the center and guard electrodes and the surface of virgin Wesgo AL995 alumina specimen were examined at RT using a scanning electron microscope (SEM) under the same conditions. The SEM x-ray analysis of degraded and virgin surfaces has shown that impurity segregation along the grain boundaries is responsible for the apparent surface breakdown of Wesgo AL995 alumina at the electric field assisted conduction.[4] The surface degradation of Wesgo AL995 alumina may seriously limit its application in ITER.

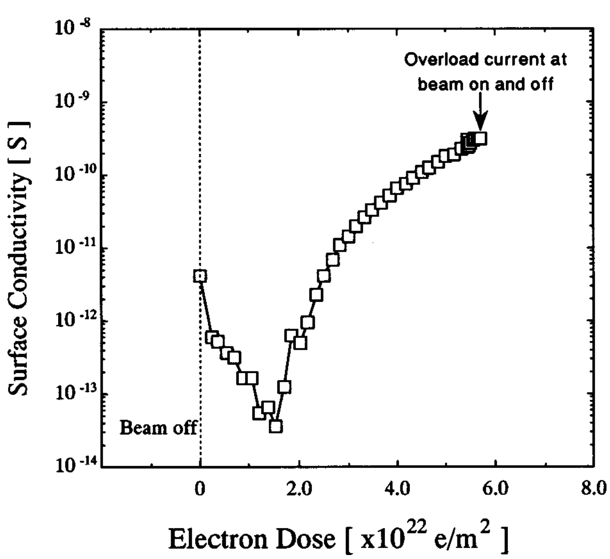

FIG. 6. Surface conductivity of a 295-μm-thick Wesgo AL995 alumina with a 1 MeV electron irradiation flux of $1.4\times10^{18}e/\mathrm{m}^2$ s in a dc electric field of 300 kV/m at 723 K.

IV. CONCLUSIONS

The difference between the activation energies before 0.49 ± 0.02 eV and during 0.19 ± 0.06 eV irradiation indicates a substantial impact of irradiation on the conductivity of Wesgo AL995 alumina. The electrical conductivity of Wesgo AL995 alumina is lower by approximately 2 orders of magnitude than the conductivity requirement for the magnetic coils in ITER. Thermal disruption may not impact on Wesgo AL995 alumina in ITER because of the absence of TSC peaks in it. Although no substantial bulk degradation is observed under irradiation up to a fluence of 7.0×10^{22} e/m^2 (7.97×10^{-5} dpa) at 723 K, the surface degradation of plasma facing insulating materials may be seriously affected in the ITER environment. Finally it is suggested that the international round robin experiments should be performed not only on a single specimen, but also under similar experimental conditions for specimen thickness, applied electric field, etc., unless any experimental constraint arises.

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