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In situ measurement of electrical conductivity of alumina under electron irradiation in a high voltage electron microscope

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Abstract

Radiation induced conductivity (RIC), thermally stimulated conductivity (TSC) and radiation induced electrical degradation (RIED) are major concerns of insulating ceramic materials under the effects of flux, electric field and temperature and may lessen their performance in fusion reactors. In situ measurements of the electrical conductivity of single crystal α-Al_2O_3 (alumina) using the standard electric guarding technique has been performed under 1 MeV electron irradiation with an applied electric field of 93 kV/m at temperatures ranging from room temperature to 723 K. Experimental results imply that electronic excitation associated with radiation induced defects controls RIC of α-Al_2O_3 and show that TSC, especially the transient peak resulting from excess charges stored in defects, may affect the performance of α-Al_2O_3 in fusion reactors. A significant surface conductivity is confirmed from a 1 MeV electron dose of 3×10^{22} e/m^2 (3.3×10^{-5} dpa) but no substantial bulk degradation is found under irradiation up to a dose of 7.1×10^{22} e/m^2 (7.7×10^{-5} dpa) at 723 K. In conclusion, it is emphasized that RIC and RIED of α-Al_2O_3 are not severe for insulators in the International Thermonuclear Experimental Reactor (ITER) but TSC could limit their applications even to ITER.

1. Introduction

Ceramic materials are often recommended in a variety of components in fusion reactors because of their low activation capability and high electric performance under radiation environments. Included are first wall components as well as blanket, radio frequency (rf) system, heating system and neutral beam injector materials [1,2]. Alpha-alumina (α-Al_2O_3) is one of the best candidate materials for fusion reactors and has an electrical conductivity of $\sim 10^{-15}$ S/m at room temperature and $\sim 10^{-7}$ S/m at 1000 K which may highly satisfy the conductivity requirement (less than 10^{-4} S/m) for all the insulators and (less than 10^{-6} S/m) for magnetic coils in the International Thermonuclear Experimental Reactor (ITER). The ceramic insulators of fusion reactors, however, will be subjected not only to high energetic particles but also to high voltage and temperature and will deteriorate in the fusion environments.

The ionizing radiation field can excite electrons from the valence to the conduction band and those electrons contribute to the conduction process thereby producing radiation induced conductivity (RIC). Thus principally, RIC is the direct result of the creation of free electron–hole pairs in the ionizing radiation field and proportional to the dose rate [3]. It is well known that RIC occurs transiently, the typical value is the order of 10^{-9} s and that the greatest part of RIC of ceramic insulators returns back near to zero immediately after turning off the radiation source. Therefore, in situ measurement is indispensable for criticizing the performance of insulators in fusion environments.

Under the operation of ITER and any other proposed fusion reactors, any kinds of disruption may happen. Further, even after reaching the stable operation of reactors, the temperature may fluctuate in a limited range. Thus, it is reasonable to postulate any uncontrollable fluctuations of temperatures of first wall representatives especially of magnetic coils. For these reasons, it is important to examine the effects of step-wise temperature changes of candi-

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date insulators with and without electric and radiation fields. Under such temperature changes, the kinetics of trapped and detrapped electrons stimulate the electrical conductivity. This phenomenon is well known as thermally stimulated conductivity (TSC).

Hodgson [4,5] first found a permanent increase in the electrical conductivity of sapphire under certain ionizing and displacive radiation fields under electric fields at a moderate temperature range. This phenomenon of insulating materials was originally called radiation induced electrical degradation (RIED). The conductivity corresponding to RIED, on contrary to RIC, does not revert to its pre-irradiation value after the removal of the radiation source. Subsequent irradiation studies [15–20,23] found the RIED effect in sapphire, where some studies indicated that RIED is a bulk effect [15,19,20,23] and the rest showed that it is not a substantial effect [18] or it is a surface degradation effect [13,16,17,23]. In fact, recently Pells and Hodgson [20] found RIED in sapphire and examined the cause of RIED by optical and transmission electron microscopy. They found γ-alumina in the bulk of the irradiated materials along with the measured electrical degradation. However, even though there are several studies explaining the cause of RIED [13,15–20,23], the exact cause for RIED has not yet been identified. So, further experiments are needed to resolve the RIED controversial fact.

Consequently, RIC, TSC and RIED are major concerns of insulating ceramic materials in fusion reactors and require in situ measurement under the effects of flux, electric field and irradiation temperature. The objectives of this study are to find the controlling factors of RIC and TSC of α-Al_2O_3 other than the number of electron–hole pairs and trapping centers, to understand how α-Al_2O_3 performs with variation of the temperature in fusion reactors and to get insight into a possible mechanism responsible for RIED of α-Al_2O_3.

2. Experimental procedure

Single crystal α-Al_2O_3 (Kyocera SA 100 with 99.99% purity and [1$\bar{1}$02] orientation) was used in this study to observe RIC, TSC and RIED. Sample disks of 300 μm to 500 μm in thickness and 10 mm in diameter were cut from the single crystal using a low-speed diamond saw and then specimens of 5 mm in diameter were taken from the 10 mm disks by an ultrasonic cutter. The specimen disks were put on a copper stub with low melting wax and polished both surfaces with different grades of polishing papers until the surfaces become mirror. The final thickness of specimens was reduced to 228–320 μm. The specimens were cleaned by soaking in acetone followed by annealing in air at 1573 K for 1 h and then cooled slowly to room temperature.

The configuration of three electrodes on the specimen surfaces is shown in Fig. 1. A schematic diagram of a specimen holder including top and side views of the specimen cell after placing the specimen in the cell is shown in Fig. 2. More details are illustrated elsewhere [18]. The center electrode of 2 mm and the guard electrode of 4.5 mm outer and 3.5 mm inner diameter were made on the top side of the specimen by platinum paste. On the other hand, the ground electrode of 4.5 mm in diameter was pasted on the backside of each specimen. A gap was left between the outside of the guard ring and the ground electrode including edge only for the prevention of provable leakage current from the edge. Twenty four hours of pasting later, the specimens were heated at 373 K for 1 h in vacuum to make a good adhesion of the electrode paste with the surfaces. The specimen was placed for irradiation in the specimen cell at the front gap of a ready-made holder onto the macor insulator bar which was covered by a thin copper plate acting as a ground electrode contacting through the back electrode of the specimen. The heater was positioned inside the macor insulator just beneath the copper plate and a thermocouple was connected to the copper plate. The guard and center electrodes were pressed by a circular copper ring of 4.5 mm in diameter, 0.2 mm in thickness and 0.5 mm in height and an L shaped copper wire of 0.1 mm in diameter, respectively. The other side of the ring and the wire were screwed to the macor insulator. An alumina insulated tri-axial copper cable was used as the outlets of the guard, center and ground electrodes through such screws in order to minimize the unexpected irradiation loss of outlets.

The experiments were performed in a high voltage electron microscope (HVEM) under irradiation with a 1 MeV electrons under beam-on and-off conditions at temperatures ranging from room temperature to 723 K with an electric field of 93 kV/m. Irradiation was done only to the center electrode of the specimen. The vacuum pressure of the microscope was ~ 10^{-5} Pa. The measured temperature error of the specimen was ~ 5°C mainly due to the placement constraint of the thermocouple near the back electrode. During irradiation, the specimen temperature

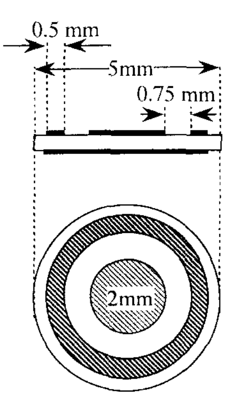


Fig. 1. Configuration of three electrodes on the specimen surfaces.

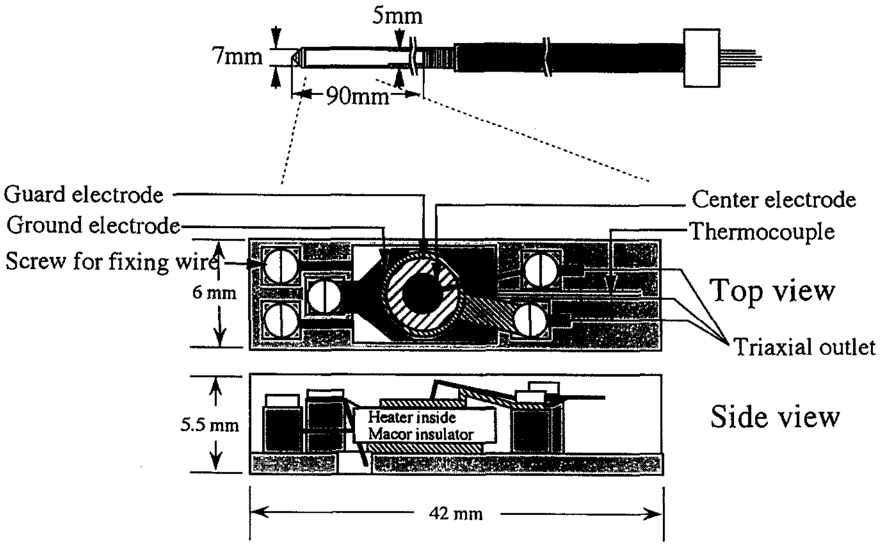


Fig. 2. A schematic diagram of the specimen holder including top and side views of the specimen cell after placing the specimen in the cell.

was raised by ~ 10°C higher than the heater temperature due to the beam heating. A dc potential was continuously applied during beam-on and -off conditions to the back electrode with a Hewlett-Packard 4339A power supply unit. The specimen current was recorded every two minutes using a Hewlett-Packard 4339A high resistance meter. The ohmic nature of specimens was examined at different stages with varying applied voltages in the forward and backward directions.

3. Results and discussion

A series of experiments were carried out in order to investigate RIC, TSC and RIED of single crystal α-Al_2O_3. Fig. 3 shows a time sequence of the electrical conductivity for a 270 μm thick specimen under irradiation with a 1 MeV electron dose rate of 1.41×10^{18} e/m^{2}s in a dc field of 93 kV/m at 723 K. The electrical conductivity raises immediately to a certain value after starting irradiation, increases gradually with time and decreases after attaining the maximum value. After turning off the electron beam, a large portion of the conductivity is recovered and remained a certain value of conductivity. The conductivity before and after irradiation is 4.9×10^{-8} S/m and 5.0×10^{-8} S/m, respectively. Ivanov and his co-workers [6] originally found the analogous increase and decrease in radiation induced conductivity of alumina under neutron irradiation and suggested that RIC may be due to the dynamic effect between the formation and recombination of radiation induced defects in the structure of the material. On the other hand, spallation neutron irradiation [11] on alumina at 928 K showed the initial rise followed by the gradual decrease in conductivity with irradiation and implied that the accumulation of damage increased the concentration of trapping and recombination centers resulting in a decrease of conductivity. Gamma irradiation [7] also showed the same behavior and it was argued that the free carrier generation rate and the production of defect trapping sites during irradiation affect RIC. Thus, the abrupt raising and the following monotonous decrease of the conductivity in the present study is supposed to be the effect of electronic excitation and the interaction between radiation induced defects (trapping centers) and free electrons in the crystal, respectively.

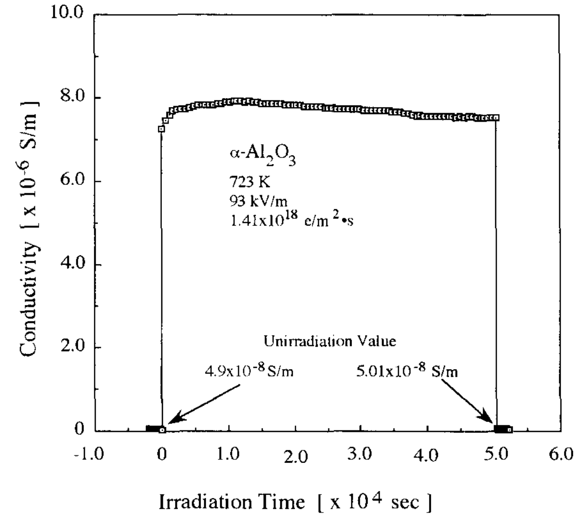


Fig. 3. Time dependence of electrical conductivity for a 270 μm thick α-Al_2O_3 specimen under irradiation with a 1 MeV electron dose rate of 1.41×10^{18} e/m^2 s in a dc field of 93 kV/m at 723 K.

Fundamentally, fusion reactors will be operated in a pulse mode with ignition and termination phases which will be controlled by toroidal and poloidal fields. So the surroundings of plasma will be subjected to lower to higher radiation fields. This fact leads us to perform measurements of conductivity with increasing and decreasing electron beam intensity over a wide range of temperatures. The measurement was done with increasing and decreasing temperature by steps of 50°C between 296 K and 723 K, as shown in Fig. 4. In both cases, the conductivity is almost proportional to the beam intensity or dose rate as well as to the temperature. The result also implies that the conductivity increases with increasing beam intensity and rarely depends on beam history but depends on temperature history.

It is generally accepted that the electrical conductivity (σ) during irradiation is expressed by the equation $\sigma = \sigma_0 + kR^\delta$, where σ_0 is the conductivity in the absence of radiation, k a material dependent constant, R the ionizing dose rate and δ the ionizing dose rate exponent. The temperature dependence of the δ values for the 288 μm thick α-Al_2O_3 specimen was obtained from Fig. 4 and it is shown in Fig. 5 both for increasing and decreasing temperatures. The values of δ are nearly unity, indicating the formation of point defects which act as trapping centers. Even though the values of δ are fairly scattered, the tendency of δ to decrease with increasing temperature, except at 623 K may, also indicate a correspondence to the density and/or the structure of point defects.

A comparison between the RIC of alumina of the present study irradiated at room temperature with that of other studies is shown in Fig. 6. The present results also include data at 365, 435, 638 and 723 K. It should be pointed out here that the operating temperature of ITER is 623 K and the maximum ionizing dose rate at lithium cooled blanket is ~ 2000 Gy/s [21]. Even though the present study is done near the maximum ionizing dose rate and the maximum temperature of ITER, the value of RIC is still less by several orders than the critical requirements for insulators in ITER.

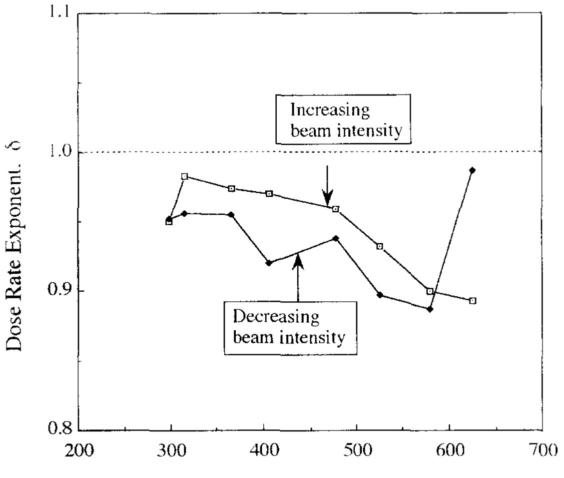
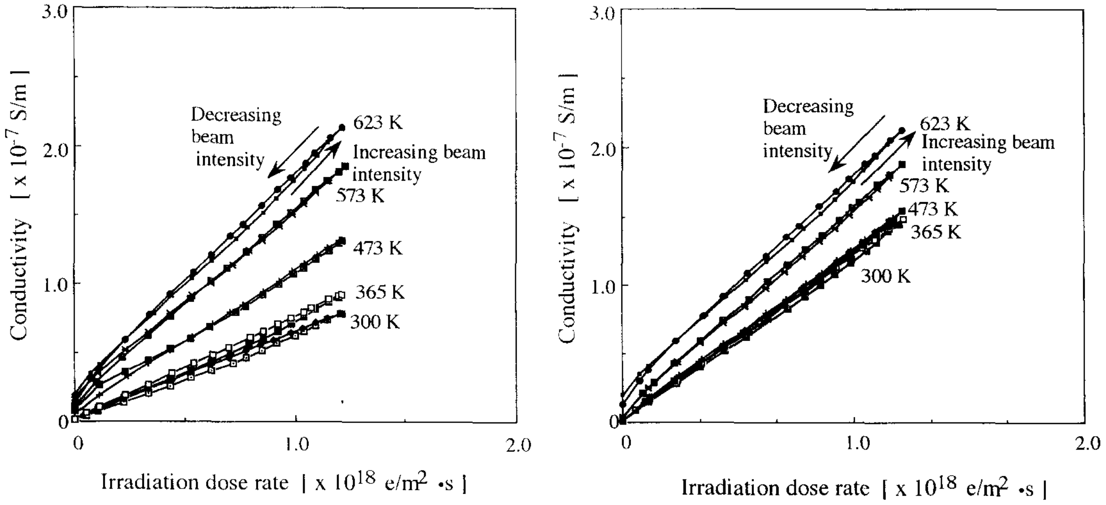


Fig. 5. Temperature dependence of dose rate exponent (δ) of electrical conductivity (σ) based on $\sigma = \sigma_0 + kR^\delta$ with the experimental data shown in Fig. 4, where the significance of parameters is written in the text.



(a)

(b)

Fig. 4. Temperature dependence of RIC of a 288 μm thick α-Al_2O_3 specimen under 1 MeV electron irradiation with increasing and decreasing beam intensity. (a) Increasing temperature from room temperature to 623 K. (b) Decreasing temperature from 623 K to room temperature.

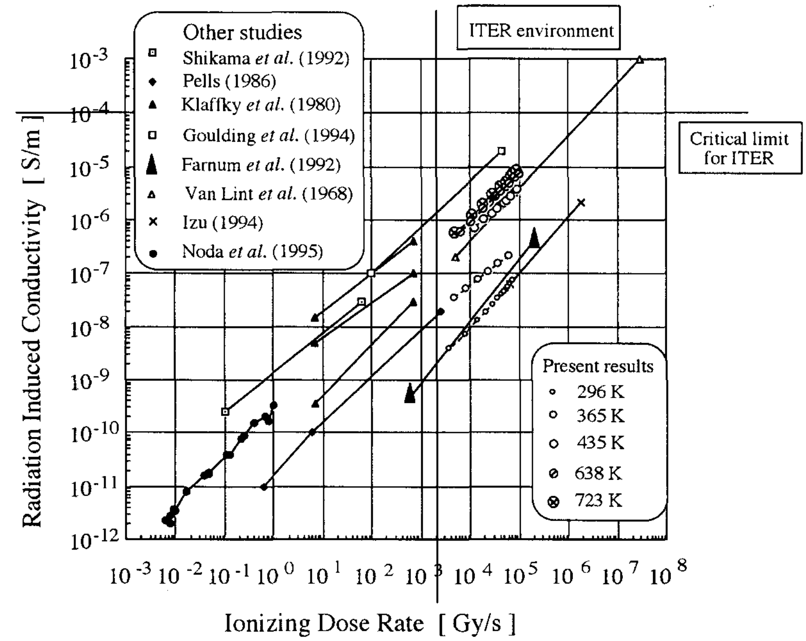


Fig. 6. Ionizing dose rate dependence of RIC of α-Al_2O_3 irradiated at room temperature compared with that of other studies. The present results include data at 365, 435, 638 and 723 K.

Our next attempt was to perform the TSC measurement. The specimen was irradiated with a 1 MeV electron dose rate of 1.4×10^{18} e/m^2 s and its electrical conductivity was measured under beam-on and -off conditions to observe how the inactive carriers vigorously participate in the conduction process through thermal excitation. The specimen was irradiated by stair stepped temperatures from 296 K to 673 K, composed of alternating times of

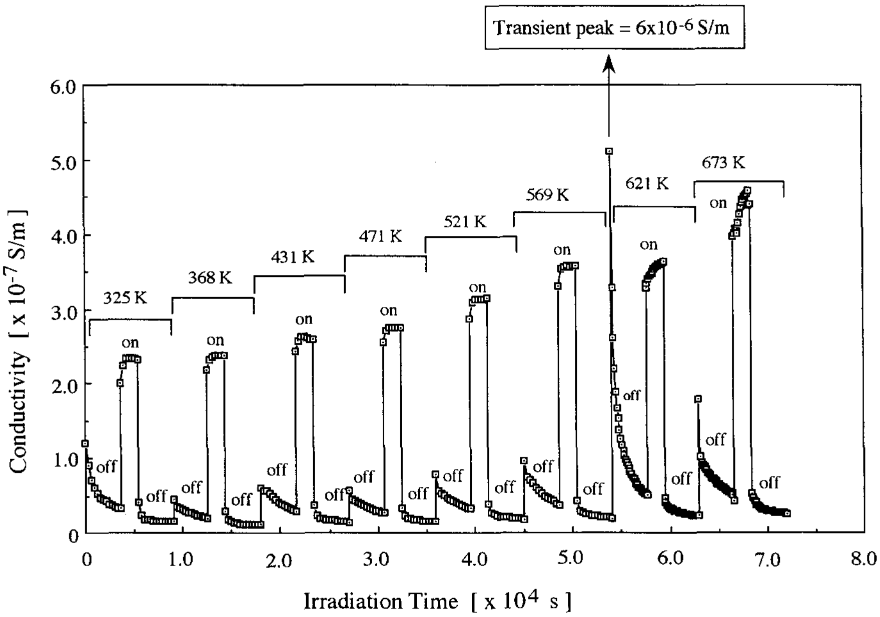


Fig. 7. Temperature dependence of electrical conductivity for a 288 μm thick α-Al_2O_3 specimen under irradiation with a 1 MeV electron dose rate of 1.5×10^{18} e/m$^2 \cdot$ s with beam-on and -off conditions.

irradiation for 1800 s followed by annealing at the temperature for 3600 s and then annealing at 50°C at a higher temperature for 3600 s as shown in Fig. 7. The conductivity measurement was done during isothermal periods and five minutes after the increase in annealing temperature to allow the specimen to reach thermal equilibrium. Like Fig. 3, at each step after turning on the electron beam the specimen current promptly raises to a certain value higher than the pre-irradiation value and then exponentially increases with time, representing RIC through excited electrons and holes associated with trapping centers such as vacancies and interstitials. When turning off the beam, the most of RIC at the annealing temperature is immediately reduced and the small amount of remaining RIC decreases exponentially with time. If the annealing temperature is raised, the specimen current increases and then exponentially slows down with time. Under non-irradiation conditions, the highest peak conductivity (5.1×10^{-7} S/m) is observed at the change of annealing temperature from 569 K to 623 K and it is greater than RIC (3.6×10^{-7} S/m). The calculated value of the transient peak conductivity before the thermal stability of the specimen at 623 K i.e. the conductivity at the first second just after raising the temperature to 623 K, is 6.0×10^{-6} S/m. This transient peak is six times greater than the limiting conductivity required for insulators of magnetic coils in ITER for which this insulator may no longer be regarded as a good insulator for magnetic coils.

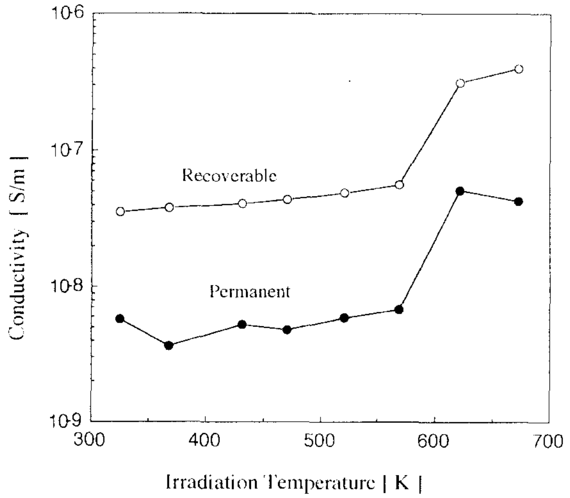


Fig. 8. Temperature dependence of recoverable and permanent conductivity from the graph shown in Fig. 7.

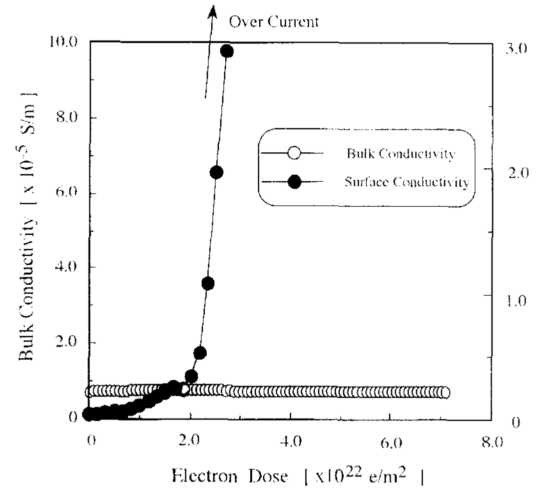


Fig. 9. Bulk and surface electrical conductivity of a 270 μm thick α-Al_2O_3 specimen under irradiation with a 1 MeV electron dose rate of 1.41×10^{18} $e/m^2 \cdot s$ with a dc applied electric field of 93 kV/m at 723 K.

Thermoluminescent (TL) and TSC measurement [8–10] have shown that the TL and TSC peaks are due to the thermal release of charge carriers mainly electrons from traps. Pickard et al. [8] have extensively investigated the thermal stimulation of electrons trapped by point defects in alumina. They showed that electrons from shallow traps in the forbidden band were accelerated during initial annealing temperature and were responsible for TSC peaks. Consequently, however, in our TSC measurement at each annealing temperature, the TSC peak arises because of the release of electrons from traps. The most peak conductivity at 623 K indicates significant charge storage in the α-Al_2O_3 specimen during irradiation, in spite of RIC which may assist to induce leakage currents to mitigate charge buildup. In addition, if the beam-off conductivity is compared with the beam-on conductivity, the preceding is resulted intrinsically [12] and gives a good agreement with the present results as shown in Fig. 7. More specifically, if irradiation does not affect the microstructure of the specimen, the beam-off conductivity should be constant. Fig. 8 shows the distinction between the contribution to the conductivity through electron beam-on and -off conditions at different temperatures. The conductivity at beam-off conditions is considered to be permanent and increases with increasing temperature. The difference in conductivity between the beam-on and -off situations represents a recoverable value. The results considered in Fig. 8 reveal that the beam-off conductivity (permanent conductivity) is several times lower than the recoverable conductivity. The difference between these two conductivity values becomes larger with temperature and does not show any severe permanent degradation, contradicting the results of Hodgson [4] and Patuwathavithane et al. [7].

Following Hodgson's demonstration of RIED, several studies [15,17,19,20] have been done at almost the same conditions as Hodgson's experiments and have found RIED in some insulators, thereby the RIED phenomenon has become more important from that time. Due to the RIED effect, ceramic insulators in fusion reactors may not perform well and their grade will be lowered. The RIED phenomenon has become controversial when several research groups [7,13,17,18] have failed to observe RIED in some grades of alumina where RIED should occur. However, in order to maintain the standardization of the RIED experiment of ceramic insulators, the International Atomic Energy Agency (IAEA) has given a few recommendations. The recommendations are directly surface temperature measurement, making guard ring geometry, verifying ohmic behavior and post irradiation measurement of the specimen. We followed those recommendations and irradiated α-Al_2O_3 for a long time to observe RIED. The bulk and surface conductivity as a function of dose are shown in Fig. 9 where a specimen was irradiated under a 1 MeV electron dose rate of 1.4×10^{18} e/m^2 s in a dc electric field of 93 kV at 723 K. No substantial bulk degradation is seen up to 7.1×10^{22} e/m^2 (7.7×10^{-5} dpa) but the RIED like abrupt increasing of surface conductivity is confirmed from a dose 3×10^{22} e/m^2 (3.3×10^{-5} dpa) possibly due to the deposition of carbon contamination layer on the surface. The observed abrupt increase in the surface conductivity is more likely to be compatible to the results of Kesternich et al. [13] who found severe leakage conductance. Moslang et al. [17] have irradiated the central part of the electrode both of Vitox and Wesgo alumina with alpha particles and found RIED only in Vitox grade. Further, Morono et al. [16] observed a different result of surface degradation in Wesgo grade under electron irradiation. Recently, Pells and Hodgson [20] carried out an electron irradiation on single crystal α-Al_2O_3 (sapphire) at 723 K and found bulk RIED. It follows that the RIED related studies do not concur one another but may be due to the discrepancy of measurement technique, variety of impurity in materials and variety of irradiation conditions. It also strongly suggests that the proper selection of grades of α-Al_2O_3 may solve the RIED problem. In fact, Hodgson [22] showed that in the case of lower dose rates, RIED requires lower total doses and for higher dose rates, the situation is vice-versa. The comparison of Hodgson's [22] dose versus dose rate graph with the present results of RIED shows that there is a possibility of occurring RIED near 10^{-3} dpa in our present RIED experiment. However, if one accumulates the RIC and RIED studies, one can see that albeit RIED is present in some studies [7,13-20,23], this RIED can represent as a bulk effect [15,19,20,23], surface degradation [13,16,18], micro cracking [14] or as an artifact [13]. It does not a matter whether the degradation occurs in bulk or in surface but it must be examined microstructurally. More extensive microstructural observations of irradiated specimens are necessary to resolve this fact. However, in conclusion, we like to emphasize that RIC and RIED are not so critical for insulators in ITER but TSC could be most critical. Further experimental works are designed to investigate this fact including environmental effects on specimens.

4. Conclusions

Electronic excitation associated with radiation induced defects controls RIC of α-Al_2O_3. Experimental results show that the effect of the thermally stimulated current, especially the transient peak resulting from excess stored charge in defects, may affect the performance of α-Al_2O_3 in fusion reactors. In fact, permanent conductivity indicates RIED and it is not so critical for α-Al_2O_3. Furthermore, a significant surface conductivity from a dose 3×10^{22} e/m^2 (3.3×10^{-5} dpa) is possibly due to the deposition of the carbon contamination layer and/or diffusion of the electrode metal into the specimen. However, no substantial bulk degradation is found even with an irradiation dose of 7.1×10^{22} e/m^2 (7.7×10^{-5} dpa) at 723 K. Thus RIED is likely to be a beyond bulk effect. In conclusion, it is emphasized that RIC and RIED of α-Al_2O_3 are not severe for insulators in ITER but TSC could limit their application in ITER.

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