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Significance of sample thickness and surface segregation on the electrical conductivity of Wesgo AL995 alumina under ITER environments

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Abstract

The electrical conductivity of 158, 295 and 610 μm thick Wesgo AL995 alumina was measured under 1 MeV electron irradiation with an electric field of 300 kV/m at temperatures up to 723 K. A significant increase in the conductivity with increasing the sample thickness is confirmed, but no substantial bulk degradation is found under irradiation up to a dose of 7.0×10^{22} e/m^2 (7.97×10^{-5} dpa) at 723 K. However, surface breakdown is found only in 295 and 610 μm thick specimens. The non-existence of the surface breakdown in 158 μm thick specimen is thought to be due to the sinks effect of point defects at the surface. The X-ray analysis of the virgin and degraded specimens through scanning electron microscopy (SEM) reveals the segregation of impurities along the grain boundaries on the degraded surface. The segregation of impurities assists leaking of surface current along the grain boundaries.

1. Introduction

Distinct sizes of insulating ceramics such as alumina, aluminum nitride are expected to be used in fusion reactors like international thermonuclear experimental reactor (ITER) for electrical insulation of heating and control devices. Alumina has been proved to be a potential candidate insulating material in fusion reactors [1] because of its high thermal conductivity [2], high resistance against radiation [3] and low dielectric loss [4]. The electrical conductivity induced in insulating materials during irradiation due to the electrons excited from valence to conduction band is called radiation-induced conductivity (RIC) [5]. Usually, the electrical conductivity under irradiation returns back to the unirradiation value after ceasing the irradiation source, if not the specimen is called to be a degraded one. This phenomenon is called radiation-induced electrical degradation (RIED) [6]. In other words, RIC and RIED are temporary and permanent degradation, respectively.

Although distinct sizes of alumina are to be used in fusion reactors, no studies have ever been done on the thickness dependence of RIC under fusion environments. In addition, the fact that different works have found discrepant absolute values of RIC, may be due to discrepant type and thickness of the samples including radiation spectrum and sample impurities. On the other hand, the interaction of plasma produced particles deserves to check the irradiation effects with high-energy electrons on the sample surface. In addition, recently, we have found the strong thickness dependence of RIC [7] and a RIED-like surface degradation [8] in single crystalline alumina. However, the works on the role of thickness including the impact of sample surface of Wesgo AL995 alumina on the electrical conductivity are not available. The purpose of this paper is to report the significance of sample thickness together with the effect of surface on the electrical conductivity through the measurements of electrical conductivity of Wesgo AL995 alumina under 1 MeV electron irradiation.

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Table 1
Chemical composition of impurities in Wesgo AL995 alumina used in the present work

Element	Concentration (wppm)
B	<30
Ba	22 ± 6
Ca	480 ± 50
Cu	<15
Fe	500 ± 70
Li	20 ± 10
Mg	1200 ± 150
Mn	<10
Na	350 ± 100
Ni	<30
Si	1400 ± 170
Ti	80 ± 20
Zn	10 ± 3
Zr	40 ± 20

2. Experimental

Polycrystalline Wesgo AL995 alumina specimens having diameter of 5.5 mm and thickness of 158, 295, 610 μm were used in the present study. Table 1 shows the impurity contents in polycrystalline Wesgo AL995 alumina. Titanium was deposited in vacuum to make the three-electrode system in this study. The temperature dependence of electrical conductivity before irradiation was measured in a bell jar at 10^{-4} Pa in the course of heating from room temperature (RT) to 723 K at the rate of ~3 K/min. The irradiation experiments were carried out in a high-voltage electron microscope (HVEM) under 1 MeV beam-on and -off conditions at temperatures ranging from RT to 723 K. The pressure of the HVEM was $\sim 10^{-5}$ Pa. Potential was applied to the back electrode and low side measurement technique was used. 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. The surface of the virgin and degraded specimens were analyzed using scanning electron microscopy (SEM). Readers who like to be acquainted with the specimen holder are referred to see our previous works [8,9].

3. Results and discussion

3.1. Dose dependence

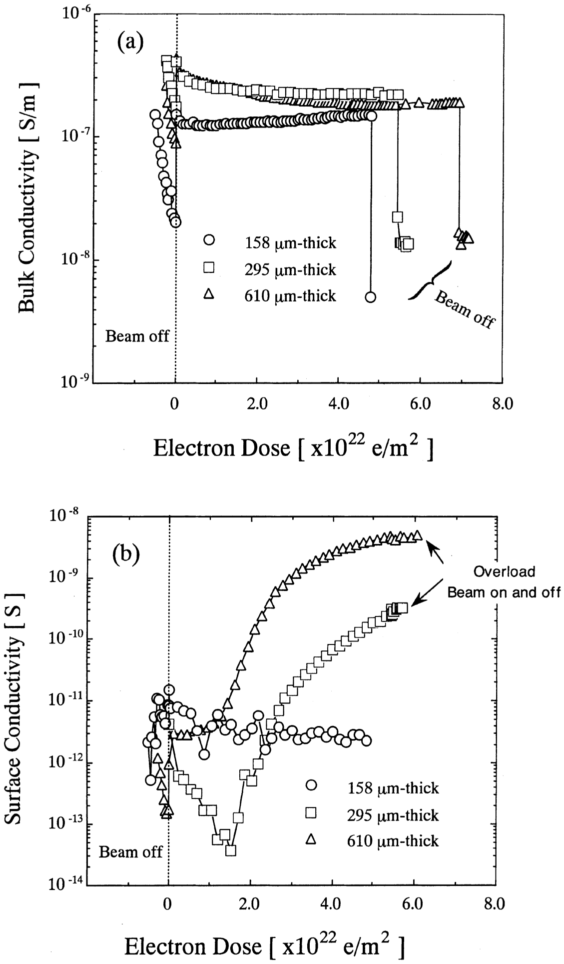

Fig. 1. (a) Bulk and (b) surface conductivity of 158 (○), 295 (□) and 610 (△) μm thick Wesgo AL995 alumina specimens having Ti electrodes under irradiation with a 1 MeV electron flux of 1.4×10^{18} e/m^2 s in a dc electric field of 300 kV/m at 723 K.

Fig. 1 shows a comparison of electron dose dependence of the bulk and surface RIC for 158, 295 and 610 μm thick Wesgo AL995 alumina specimens with a 1 MeV electron dose rate of 1.4×10^{18} e/m^2 s (1.6×10^{-9} dpa/s and 8.7×10^4 Gy/s) in a dc field of 300 kV/m at 723 K. The potentials applied to 158, 295 and 610 μm thick specimens were 47.4, 88.5 and 183 V, respectively. The bulk and surface conductivity measurements were performed simultaneously. No bulk RIED has been observed up to 9.4×10^{-5} dpa. The surface conductivity of 295 and 610 μm thick samples, first decreases until $\sim 1.5 \times 10^{22}$ e/m^2 and then starts to increase until the total dose of 6×10^{22} e/m^2. After the total accumulation dose, even though the bulk conductivity of the specimens returned back to the unirradiation value, the surface conductivity of 295 and 610 μm thick specimens did not return back. In addition, after turning off the beam, the specimen was heated for 30 min but no change in the surface degradation was observed. So the surface degradation in 295 and 610 μm thick samples is apparent. The decreasing and increasing of the surface conductivity in both specimens may indicate polarization effect (displacement of charges with respect to each other) during irradiation with electric field. On the other hand, no surface degradation was detected in 158 μm thick specimen. Surfaces would act as the dominant sinks for the point defects produced in thin specimen. The possibility of surviving point defects in the thick specimen would be higher than that of the thin specimen. The diffusivity of the 158, 295 and 610 μm thick specimens is estimated to be a order of 10^{-8} cm^2/s [10]. Therefore, the existence of the surface breakdown of 295 and 610 μm thick Wesgo AL995 alumina and the non-existence of surface breakdown in 158 μm thick specimen can be correlated to whether the point defects survive or not. The coloration change on both of the center and back electrodes of the surface degraded samples has been found in this work. Moroño and Hodgson [11] have found the similar surface degradation in Wesgo AL995 alumina which may be happened due to the large conduction along grain boundaries but no bulk RIED is reported up to 3.0×10^{-5} dpa under 1.8 MeV electrons with the electric field of 100 and 500 kV/m in high vacuum. The coloration at and near the negative electrodes is found in their work as well. The results are interpreted in terms of the mobility of cation impurity with electric field, which produces electrolysis.

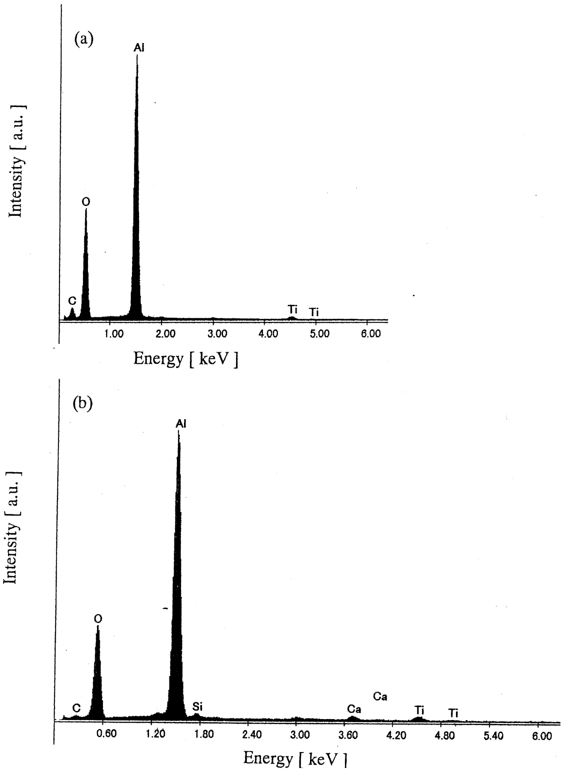

Fig. 2. Post-irradiation SEM X-ray analysis of the degraded surface of a 295 μm thick Wesgo AL995 alumina specimen: (a) in the grain; (b) along the grain boundary. More intense peaks due to impurities along the grain boundary than those of the grain are observed.

Impurity segregation at the degraded surface was found by using SEM [11]. In the quest for searching the reason of surface degradation, we have examined the surface between the center and guard electrodes of the degraded and the virgin Wesgo AL995 alumina specimens using SEM under the same conditions. Wesgo AL995 alumina is found to be a very porous material and the size of the grains is estimated to be in the range of ~10 to ~20 μm which is different from that of the other works [11,12]. In the case of degraded surface specimen, the distribution of impurities along and in the grain boundaries on the surface region between the center and guard electrodes is investigated. The X-ray intensity spectrum for degraded specimen reveals that the impurities of Mg, Si, Ca, Ti etc. segregate along the grain boundaries, but not in the grains (Fig. 2). Similar measurements of longitudinal distribution of elements in the grains and along the grain boundaries for the virgin specimen are performed. No segregation of impurities could be detected for the virgin specimen along the grain boundaries and in the grains. It is possible that the Mg, Si, Ca and Ti impurity atoms in Wesgo AL995 alumina can be electrically biased to make an electric current passing along grain boundaries through making special paths. Kesternich [13] has argued that due to the temperature gradient between the sample and sample holder, there are inherent thermal mechanical strain and stresses in the specimen which produces cracks and which may be the origin of specimen degradation. Along the cracks in the ceramics, there could be leakage current contributing to the apparent conductivity. Zinkle and his co-workers [14] have found the apparent increasing in the conductivity in amorphous alumina films which is due to the radiation-induced micro-cracks in ceramics but such kind of behavior is not very much known yet. Furthermore, although the round robin groups have done in situ measurements of RIC on Wesgo AL995 alumina over a wide range of ionizing dose rate, no bulk RIED is detected to 3 dpa regardless of radiation sources. Readers are advised to read Ref. [12] for more details of RIED results of Wesgo AL995 as well as other polycrystalline alumina. However, the SEM analysis for degraded and virgin specimens in this work indicates that the segregation of impurities along the grain boundaries is responsible for the apparent surface breakdown of polycrystalline Wesgo AL995 alumina at the field-assisted conduction.

3.2. Thickness dependence

The thickness dependence of the RIC of Wesgo AL995 alumina specimens under 1 MeV electrons dose rate of 1.4×10^{18} e/m^2 s (1.6×10^{-9} dpa/s and 8.7×10^4

Gy/s) in a dc field of 300 kV/m at various temperatures is shown in Fig. 3. Like single crystalline Kyocera alumina [7], the RIC increases nearly linearly with increasing sample thickness except at 673 K. The reasons of this non-linearity for 295 µm thick sample at 673 K is unknown. In addition, these results of thickness dependence of RIC indicate that a part of the electron beam current deposits in the samples. The deposited current increases with increasing specimen thickness and affects the value of the specimen current. The highest RIC in 610 µm thick specimen seems to be due to the highest amount of electrons charge deposition in the sample (compare with the graph of k at Fig. 4).

The ionizing efficiency, k, which is defined by $\sigma = \sigma_0 + kR^{\delta}$ where σ is the RIC, σ_0 the conductivity in the absence of radiation, R the dose rate and δ is the dose rate exponent), varies with temperature by two orders of magnitude from 300 to 730 K and it is higher for thicker specimen and vice versa. Quantitatively the value of k at RT for 158 µm thick sample is 9×10^{-15} s/(Gy Ω m) and for 610 µm thick sample is 1.2×10^{-13} s/(Gy Ω m). The overall ionizing efficiency for polycrystalline Wesgo AL995 alumina is found to be in the range of $\sim 10^{-14}$ to $\sim 10^{-12}$ s/(Gy Ω m) which is lower by two orders of magnitude to that of single crystalline Kyocera alumina [7]. This behavior represents Wesgo AL995 alumina to be a less ionizing material (because of highly porosity) comparing to the sapphire for which the quantity of energy deposited in sapphire is higher than that of Wesgo AL995 alumina. Similar behavior of thickness dependence of RIC of Wesgo AL995 alumina to that of single crystalline Kyocera alumina allows us to proceed in the same track of explanation for this results. We have found that ~5 to ~22% of the 1 MeV electron beam current may deposit in the 158–610 µm thick specimens. The estimated value of beam deposition current for 158, 295 and 610 µm thick specimens will be 3.5×10^{-8}, 8.4×10^{-8} and 1.5×10^{-7} A, respectively, at an incident dose rate of 1.4×10^{18} e/m^2 s. If we assume that all the beam current passes through the 158 µm thick Wesgo AL995 alumina, then the estimated beam deposition current ratios for 158, 295 and 610 µm thick alumina specimens will be 1, 2.4, and 4.2 (calculated from beam current), respectively, which are larger than the ionizing efficiency ratio of 1, 1.6, and 2.4 for the respective specimens. This fact indicates that the beam deposition or ionizing efficiency is not only responsible for the thickness dependence of RIC, however, is due to induced field and is discussed latter.

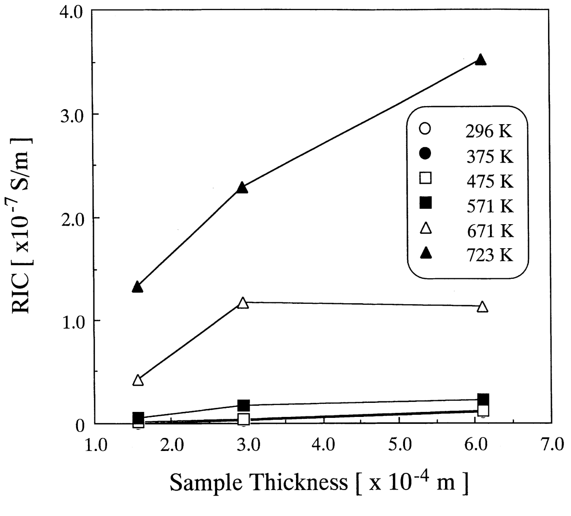

Fig. 3. Thickness dependence of RIC of Wesgo AL995 alumina with a 1 MeV electron dose rate of 1.4×10^{18} e/m^2 s in an electric field of 300 kV/m at different temperatures. The data at 296 and 375 K are overlapped with those at 475 K.

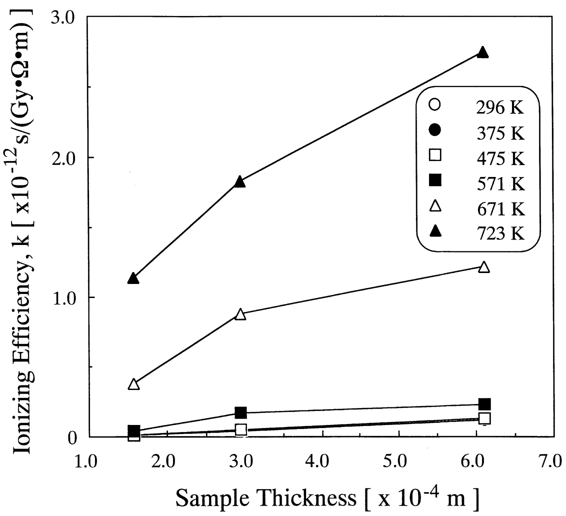

Fig. 4. Thickness dependence of ionizing efficiency (k) of Wesgo AL995 alumina specimens at various temperatures. The data at 296 and 375 K are overlapped with those at 475 K.

Further, the electric fields induced in 158, 295 and 610 µm thick Wesgo AL995 alumina specimens during 1 MeV electrons irradiation are estimated to be 84, 117, and 140 kV/m, respectively. This amount of applied field could have substantial impact on the penetration depth of the injecting beam, causing higher deposition in the thicker specimens. Frederickson and his co-workers [15] have also shown that as time goes on, the retarding behavior of the induced field on the injection beam causes greater charge build up in the thicker specimens, because the induced electric field during irradiation reduces the projected range of electrons. The resulting increase in the number of electrons deposited in the sample adds to the conduction current, which may give the answer to the divergence of RIC from linearity. However, the estimated induced fields which are 28 (potential of 13 V), 39 (potential of 34 V) and 46% (potential of 85 V) of the total applied field and are sufficiently good enough to fit the amount of specimen current varies due to thickness. Therefore, the beam deposition current or the ionizing efficiency and the induced field during irradiation with the diffusion of electrode materials through grain boundary are the plausible mechanism for RIED. In conclusion, we believe that the electric field induced by charge deposition during irradiation is comparable to the threshold electric field for RIED, may influence RIC and may be the apparent cause of RIED.

4. Conclusions

Transmission and distribution behavior of electron charge in the insulators has great influences on the RIC and these influences correlate with the thickness of the samples during irradiation. Concurrent analysis of the virgin and degraded specimens in the SEM reveals that there is no segregation of impurities in the grains and along the grain boundaries of the virgin specimen. On the other hand, the segregation of impurities is confirmed along the grain boundaries on the degraded surface. The segregation of impurities along the grain boundaries would assist leaking of specimen current along the grain boundaries.

The RIC increases with increasing specimen thickness, which is believed to be resulted from the higher amount of electron charge deposition in the thicker specimen associated with the induced field. The RIC associated with these charge depositions and induced field may cause the RIED-type degradation of alumina. Ionizing efficiency is found to be temperature and thickness dependent. Therefore, the specimen thickness is a new parameter for RIC, which was not considered in the past but reasonably could affect the RIC of alumina. However, in conclusion, we would like to emphasize that the RIC could be exceeded above the design value of electrical conductivity for insulators in ITER and the surface degradation is likely to be the dominant effect rather than that of the bulk.

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