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Electrical surface degradation of electron irradiated sapphire and silica

A. Moroño *, E.R. Hodgson, S.M. González de Vicente

Euratom/Ciemat Fusion Association, Avenida Complutense 22, 28040 Madrid, Spain

A B S T R A C T

Two types of oxides, silica and sapphire have been irradiated with 1.8 MeV electron and 54 keV He^+ in order to study surface electrical and optical degradation processes. It has been found that both materials suffer severe surface degradation as a consequence of radiation induced oxygen removal for either 1.8 MeV electron irradiation or 54 keV He^+ implantation. Degradation is higher in the case of alpha particle bombardment, but the results strongly suggest that the fundamental processes taking place are basically the same. Ionizing radiation rather than knock-on displacements appears to be the origin of the severe oxygen removal from the irradiated surface. The possibility of such surface degradation in insulators for ITER and future fusion reactors needs to be fully assessed.

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1. Introduction

Different types of electrical degradation of insulators are known to occur when these materials are subjected to a radiation field [1,2]. Electrical degradation may occur within the bulk and also at the surface of the material. Susceptibility and rate of degradation is believed to depend strongly on the ratio between displacement and ionization processes which occur during material irradiation. The aim of the work to be presented here is to address the role played by ionizing radiation on the surface electrical degradation of oxides. In order to do this, two different types of oxides have been studied: silica and sapphire. The behaviour of these two materials when subjected to a radiation field is known to be very different. Radiolytic processes giving rise to oxygen vacancy production occur within the bulk of silica [3] while in the case of sapphire such processes do not occur, knock-on collisions being necessary to produce oxygen displacements [4].

Sapphire and silica samples have been irradiated with 1.8 MeV electrons (high electronic excitation but low displacement), and with 54 keV He^+ (high electronic excitation and high displacement) at 450 °C. For both types of irradiation the experimental set-up permitted one to measure the surface electrical conductivity at different doses and as a function of temperature.

Severe surface electrical degradation occurs when either sapphire or silica are subjected to electron irradiation. Such degradation is a consequence of the loss of oxygen from the irradiated surface. A comparison with the 54 keV He^+ irradiations indicates that the electrical surface degradation is mainly due to the ionizing radiation component rather than to the knock-on displacements. This could have serious consequences for the use of alumina ceramics in fusion reactors where the gamma radiation alone may induce the loss of oxygen from the vacuum surface of the ceramic insulators eventually producing degradation of the electrical insulation.

2. Experimental procedure

Two types of materials were studied: KS-4V quartz glass kindly provided by the Russian Federation within the ITER programme and Union Carbide UV grade sapphire. The samples 1 mm in thickness were optically polished, and then either irradiated with 1.8 MeV electrons or implanted with 54 keV He^+ ions at 450 °C in high vacuum (10^{-6} mbar). Implantations were performed using the CIEMAT Danfysik 60 kV ion implanter with a beam current of 0.5 μA/cm^2 and up to a dose of 1.5×10^{17} ions/cm^2, and electron irradiations were performed in a Van de Graaff accelerator with 1.8 MeV electrons at 700 Gy/s up to 200 MGy. In both cases the samples were mounted on an oven allowing one to perform implantations and irradiations at controlled temperatures, and the irradiating beam was defined by a 6 mm diameter stainless steel collimator. To measure the surface conductivity two platinum electrodes separated by 1.5 mm were sputtered onto the sample face to be irradiated. In this way a surface area of approximately 6×1.5 mm^2 was defined. Following irradiation DC electrical surface conductivity was measured as a function of temperature from 20 to 450 °C for different doses, by applying 100 V between the electrodes, the current sensitivity was 10^{-11} A.

Optical absorption spectra from 195 to 3000 nm were measured with a Varian Cary 5 spectrophotometer. SEM X-ray analysis was performed before and after irradiation with a Hitachi S-2500 microscope, and the voltage acceleration used to perform the X-ray analysis was 25 kV.

* Corresponding author.
E-mail address: morono@ciemat.es (A. Moroño).

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3. Results

The evolution of the surface current with dose for both sapphire and KS-4V quartz glass irradiated with 1.8 MeV electrons at 450 °C is shown in Fig. 1 where one can see that surface electrical degradation occurs for both materials, the degradation being higher for sapphire. Surface electrical currents as a function of dose for both materials implanted with 54 keV He^+ at 450 °C is shown in Fig. 2. Again both materials exhibit severe surface electrical degradation. Sapphire tends to saturate more rapidly than KS-4V quartz glass.

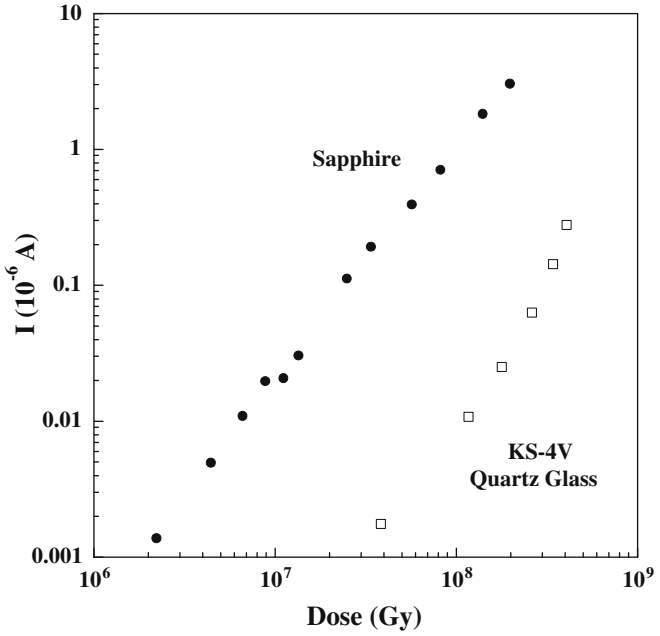

Fig. 1. Surface electrical current as a function of dose for sapphire and KS-4V quartz glass electron irradiated at 450 °C.

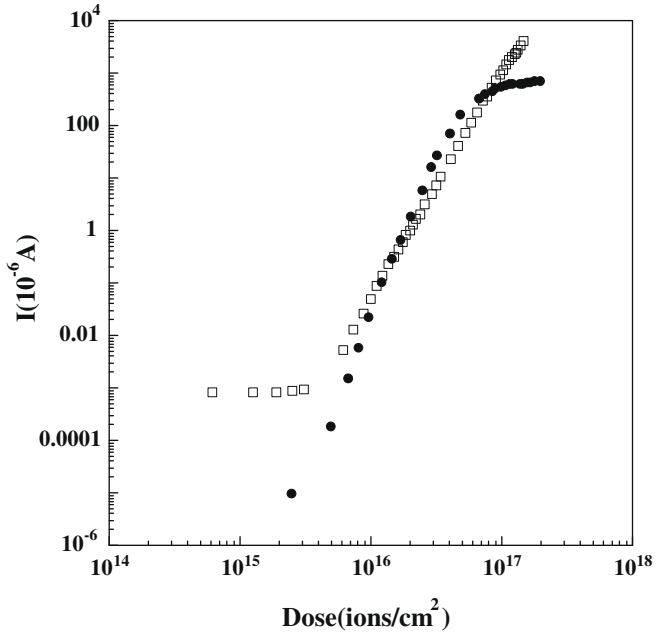

Fig. 2. Surface electrical current as a function of dose for sapphire (dots) and KS-4V quartz glass (squares) implanted with 54 keV He^+ ions at 450 °C.

Fig. 3 shows the Arrhenius plots at different doses for electron irradiated and He^+ implanted sapphire. One can see that the activation energy (slope) decreases after electron irradiation and He^+ implantation, decreasing more for the implantation case. Similar results were obtained for KS-4V quartz glass, as one can see in Fig. 4 where the Arrhenius plots for He^+ implanted and electron irradiated samples are shown. Also for this material the activation energy is reduced for both types of bombarding particles, again decreasing more for the implantation case. The results show that He^+ implantation induces higher surface electrical degradation than electron irradiation.

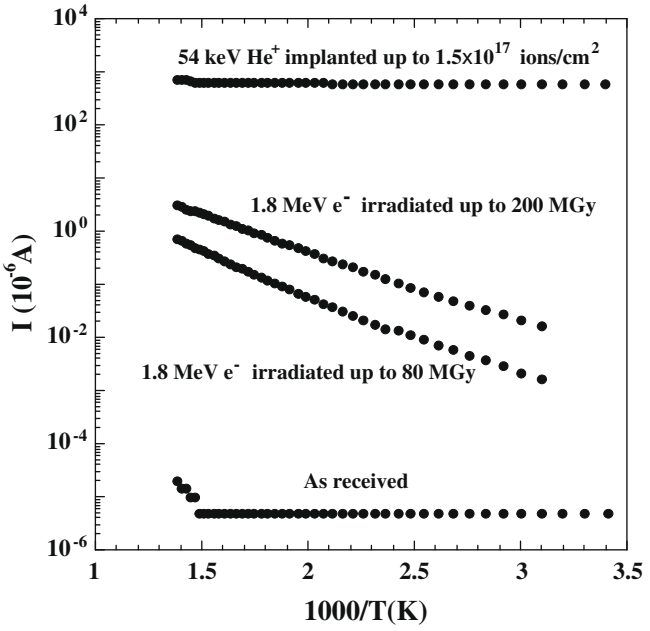

Fig. 3. Surface electrical current as a function of the inverse of temperature for both electron irradiated and He^+ implanted sapphire.

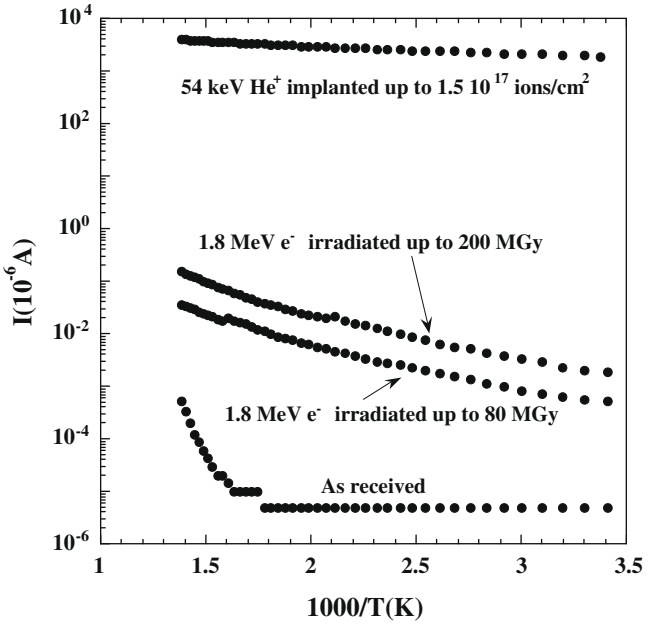

Fig. 4. Surface electrical current as a function of the inverse of temperature for both electron irradiated and He^+ implanted KS-4V quartz glass.

Optical absorption spectra induced by 1.8 MeV electron irradiation and 54 He^+ implantation for sapphire and silica are shown in Figs. 5 and 6, respectively. In both figures the optical absorption before either implantation or irradiation has been subtracted in order to show the induced darkening. The induced optical absorption is much higher in the case of He^+ implantation for both materials.

In Fig. 7 the SEM X-ray analysis is given for electron irradiated and unirradiated sapphire. One can clearly see that the oxygen proportion is much lower in the case of the irradiated sapphire. A similar comparison for silica (KS-4V) is shown in Fig. 8 where SEM

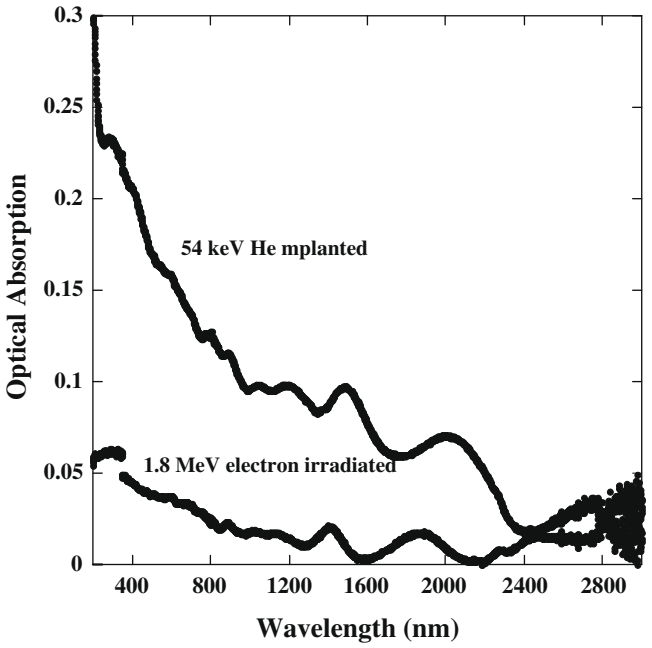

Fig. 5. Optical absorption for electron irradiated and He^+ implanted sapphire.

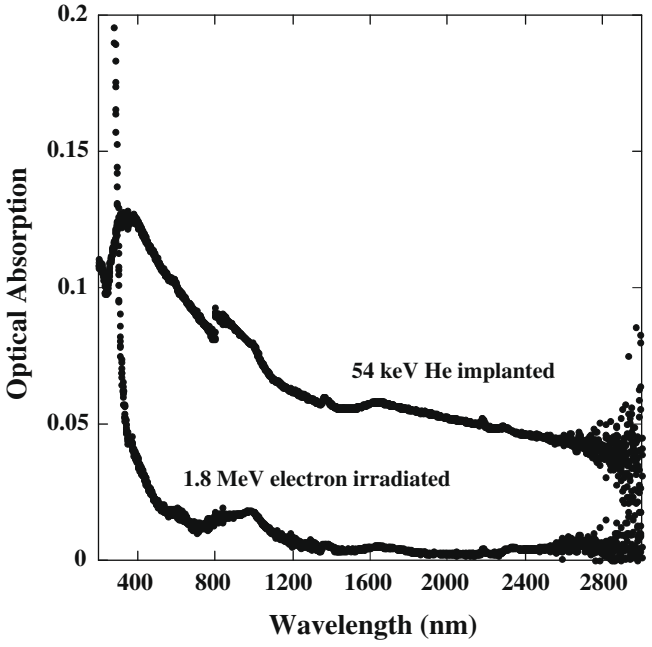

Fig. 6. Optical absorption for electron irradiated and He^+ implanted KS-4Vquartz glass.

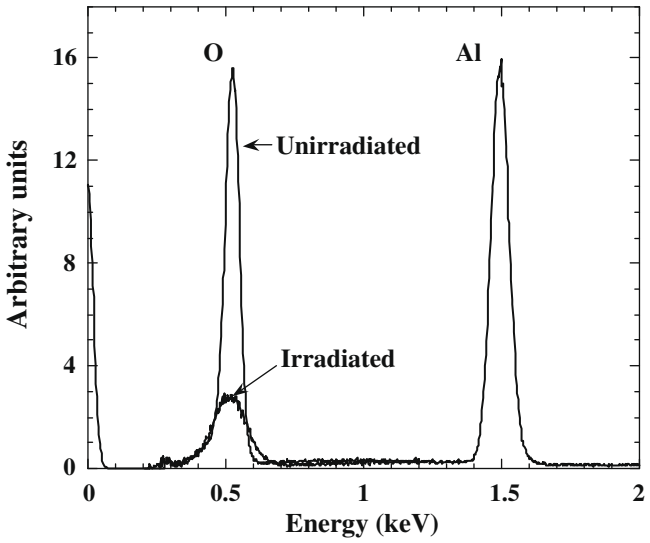

Fig. 7. SEM X-ray analysis for unirradiated and electron irradiated sapphire.

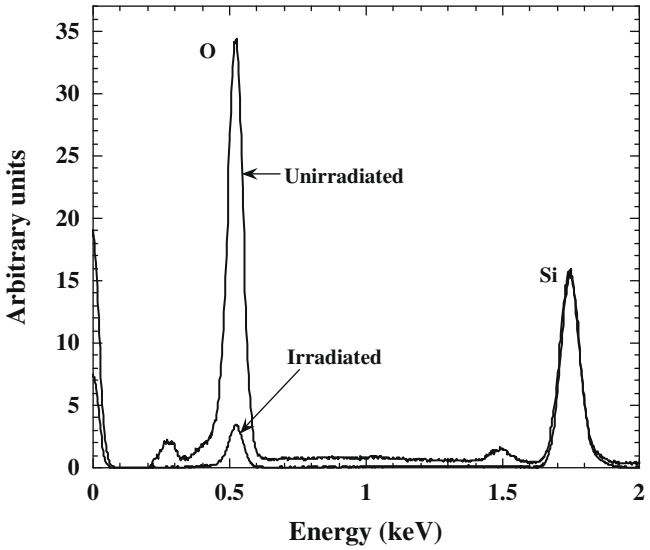

Fig. 8. SEM X-ray analysis for unirradiated and electron irradiated KS-4V quartz glass.

X-ray analysis for irradiated and unirradiated materials are shown. Also in this case a clear reduction in the oxygen content occurs after electron irradiation.

4. Discussion

Previous work clearly demonstrated that severe surface degradation of oxides after either proton [5] or alpha particle [6] bombardment occurs, characterized by oxygen loss, electrical conductivity degradation, and optical absorption increase. The experimental results shown here indicate that both 1.8 MeV electron irradiation and 54 keV He^+ implantation produce basically the same surface effects: drastic reduction of oxygen content and as a consequence an enormous increase in electrical conductivity. However, the magnitude of the fundamental damage processes for each type of irradiation is very different in principle. In the case of 54 keV He^+ implantation there is a high number of atomic

displacements within a very thin layer (of the order of 0.8 μm), high electronic excitation and also physical sputtering, while in the case of 1.8 MeV electron irradiation the number of atomic displacements is extremely low and physical sputtering is negligible. Furthermore, the same behaviour has been observed for the two types of oxides studied here which are known to behave in a very different way when exposed to a radiation field, the main difference being that silica suffers radiolysis when exposed to purely ionizing radiation while sapphire does not (knock-on collisions are necessary in order to produce atomic displacements). The observed preferential loss of oxygen suggests that the fundamental process taking place is extremely selective, and that a type of preferential radiolytic sputtering induced by the ionizing component is the main mechanism giving rise to drastic oxygen loss. The mechanism must be very similar to the radiolytic damage observed to occur for the alkali halides [7,8]. Oxygen loss induces a band gap reduction as seen in Figs. 5 and 6. Although the processes seem basically the same, the magnitude of the surface degradation in terms of surface conductivity and induced optical absorption is much less for electron irradiation. This must be a consequence of the higher density of energy deposition in the case of He implantation. The average density of energy deposited for sapphire implanted up to a dose of 1.5×10^{17} ions/cm^2 assuming a penetration of about 0.8 μm is of the order of 8×10^9 J/Kg (8000 MGy) while in the case of the 1.8 MeV electron irradiation the penetration is of the order of 3 mm and the total dose was 200 MGy, i.e. the dose in the case of He bombardment is about 12 times higher. In addition the much higher rate of displacements due to He bombardment in comparison with 1.8 MeV electron irradiations must also play an important role in the surface degradation process. In particular interstitial oxygen is generated as a consequence of displacements enhancing the flux of oxygen from the interior to the surface.

Refractory oxides such as sapphire are known to be extremely resistant to gamma radiation damage in the bulk, however these experimental results demonstrate that severe vacuum surface degradation may occur in an ionizing radiation environment. In the case of a fusion reactor the situation will be worse because the insulators not only will be subjected to gamma and neutron irradiation, but also to energetic particle bombardment due to the presence of electromagnetic fields and ionized residual gas [9].

5. Conclusions

Similar surface optical and electrical degradation for SiO_2 and Al_2O_3 occur after either 1.8 MeV electron irradiation or 54 keV He^+ implantation, due to oxygen removal from the irradiated surface. The results indicate that the cause is oxygen preferential sputtering due ionizing radiation rather than ion displacements due to the knock-on collisions. The possibility of such surface degradation in insulators for ITER and future fusion reactors needs to be fully assessed.

Acknowledgements

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