RADIATION INDUCED CONDUCTIVITY IN CERAMICS INSULATORS FOR THERMIONIC SYSTEMS

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Abstract

Insulators in thermionic systems must be able to sustain high temperature, radiation damage and electric fields while maintaining their mechanical stability, high electrical resistivity and thermal conductivity for long periods of time. The most common insulator used in these nuclear systems is alumina, either in the single crystal form (sapphire) or fine grain polycrystals. It has been demonstrated that the simultaneous application of electric field and radiation results in a dramatic increase in conductivity. This paper describes the investigation at Auburn University which examines the radiation-induced conductivity process in alumina using a variety of controlled radiations. These include high energy protons and alpha particles from a 2 MeV light ion accelerator, gamma radiation from a Co^{60} source and x-radiation from an x-ray tube. The experiments were conducted at temperatures up to 800 K with different radiation intensities. These various types of radiation provide different ratios of electronic energy loss versus nuclear energy loss. The former is expected to yield instantaneous conductivity change whereas the latter is responsible for permanent damage. Both instantaneous radiation-induced conductivity and permanent effects were determined under controlled environment. It was found that the two process were not completely separable. The structure damage produced by radiation also affects the recoverable radiation effects. Efficiencies, normalized to ionization, for induced conductivity were also measured and compared. Controlling parameters for the electrical degradation process were elucidated based on available data.

INTRODUCTION

One major degradation process in alumina insulators is radiation-induced conductivity (RIC) which can be recoverable or permanent. This radiation-induced conductivity in alumina was investigated experimentally by Hodgson and Pells and Shikama. Hodgson (1988, 1989, 1991) irradiated alumina with 1.8 MeV electrons from ambient to 1000 K with a simultaneous application of an electric field of 130 kV/m and found that the conductivity of the materials dramatically increased after an incubation period. Pells (1988, 1991) at Harwell also observed a significant increase in the electrical conductivity of alumina between 373 and 773 K with 18 MeV protons with an applied field of 500kV/m. More recently, Shikama (1992) irradiated alumina with reactor neutrons under similar conditions and his results were in accord with ion and electron simulation studies. In all these studies, it was observed that RIC increased with radiation time and radiation flux (or rate) and saturated at high doses. The saturation was reached more rapidly as the dose rate increased. Experimental work by Pells (1991) showed that the activation energy for the electrical conductivity of Al_2O_3 vanished when the temperature was increased from 283 K to 773 K under a combined action of radiation, temperature and electrical field. The activation energy for conduction was about 2 eV prior to radiation. It was also found that the change of conduction mechanism occurred only after radiation dose reached a certain threshold level. This threshold dose is a function of radiation temperature and material. It is obvious from the investigations mentioned above that the combined effects of field and radiation are fundamentally different that the two individual consequences. This paper describes the study at Auburn University which addresses the radiation-induced electrical degradation in alumina under protons, alpha ions, x-ray and gamma radiation.

EXPERIMENTAL SETUP

A 2 MeV light ion accelerator at the Nuclear Science Center at Auburn University was used for proton and helium ion irradiations. A sample holder was designed and fabricated for the study and to refine the experimental technique. A thin layer (10 microns thick) of Al_2O_3 was produced on a pure aluminum (99.99%) substrate using an anodization process. The thickness of this layer was controlled electrolytically. The aluminum substrate formed one side of the contact. A thin layer of gold was then vapor deposited on the alumina surface to form the other contact. The resistance of this layer was on the order of giga-ohms at room temperature over an area of 0.4 cm^2 in agreement with the expected values for alumina. The sample assembly was heated using radiation heating from a tungsten filament. The sample was mounted on a ceramic rod and the entire assembly was loaded into the sample chamber on the 45° arm of the accelerator system. The sample chamber was evacuated using a diffusion pump to a pressure of 10^{-6} torr. The electrical connections were carefully designed and experiments were conducted to ensure the reliability of the resistance measurements during irradiation and that these measurements were not influenced by the ion beam.

The effects of gamma radiation were examined using the high intensity Co^{60} source (with 1.17 MeV and 1.33 MeV gammas) at Auburn University's Nuclear Science Center. A specimen holder was designed and constructed which was capable of heating the specimens to 700 K in a vacuum of 10^{-6} torr during in-situ irradiations. The dose rate of the source at the sample was 0.53 Gy/s. For x-radiation, a 40 keV x-ray generator (from a Rigaku X-Ray Diffractometer unit) was used and the samples were maintained at room temperature in air.

RESULTS AND DISCUSSION

Figures 1 and 2 show the results obtained from the two experiments using energetic protons. Both experiments were conducted at 675 K. The beam current density was approximately 800 nA/cm^2. The difference between the two runs was the duration of the experiments. Both samples exhibited an instantaneous increase in conductance when the beam was turned on. This abrupt change is less apparent in figure 2 due to the difference in the y-axis scale. The digital data values from the early times of figure 2 agree with those from Figure 1. This abrupt increase is due to radiation-induced spontaneous conductivity where the high energy ions produce electron hole pairs. A gradual increase in conductance with irradiation dose followed the initial discrete change. This is a consequence of permanent microstructure change due to irradiation. Upon removal of the ion beam, the insulators recovered portion of the radiation-induced conductance. At low dose (Figure 1), this recovery corresponds closely to the initial rise indicating that short irradiation does not alter the ion-induced electron hole production process. However, the recovery after high dose irradiations (Figure 2) is much larger than the initial rise. This suggests that the radiation excitation process has been altered due to a change in the permanent microstructure induced by irradiation.

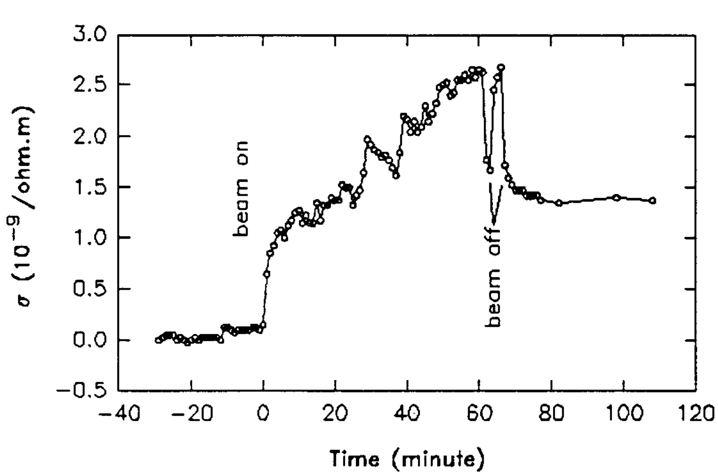

FIGURE 1. Radiation Induced Conductance in Alumina Irradiated With 2 MeV Protons at 673 K.

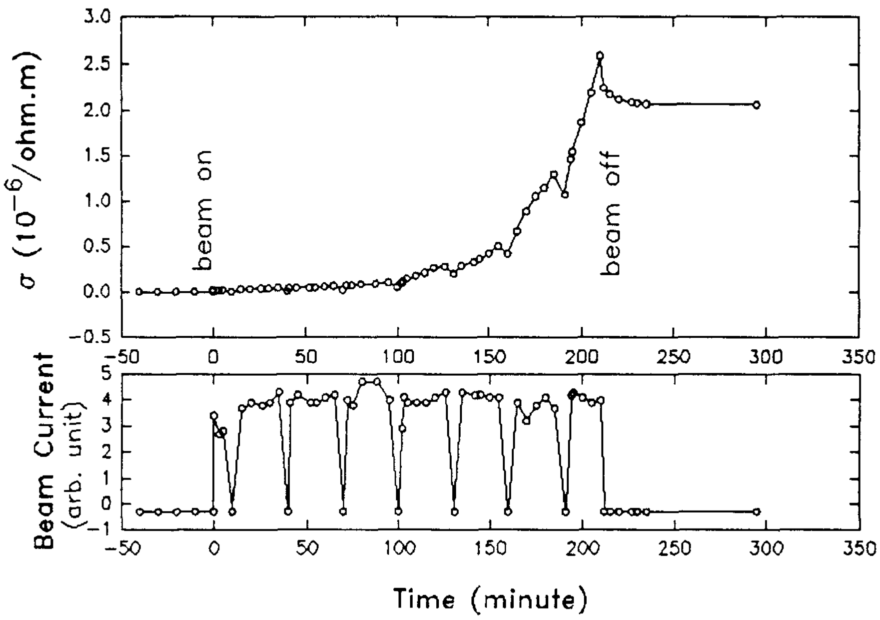

FIGURE 2. Same as Figure 1 but for Longer Duration of Irradiation; Notice the Difference in Scale and the Rapid Increase in Conductance After 200 minutes.

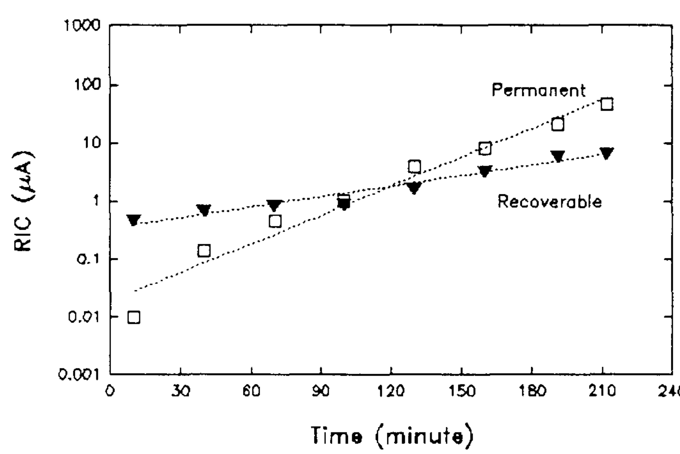

FIGURE 3. Effects of Proton Irradiation on Recoverable and Permanent Conductivity.

Analysis of the results obtained to-date indicates that there are two types of radiation-induced conductivity (RIC): recoverable and permanent. In order to separate the dose dependence of the two types of conductivity, the proton beam in the second experiment (Figure 2) was periodically interrupted (by inserting the in-line Faraday cup) and the dose-dependent recoverable conductivity was measured as a function of irradiation time. The beam current history is illustrated in the bottom of Figure 2. Results from this study are shown in figure 3 for proton irradiated alumina. Both types of induced conductivity (recoverable and permanent) increase with irradiation fluence. The recoverable conductivity is related to the excitation of electron-hole pairs induced by the ionization radiation whereas the permanent conductivity is governed by the microstructure of the material. The observed dose dependence in both phenomena suggests that the band structure as well as the microstructure of alumina is changing with irradiation. A similar experiment was conducted using 2 MeV helium ions which generated higher density cascades damage in the structure. Results are shown in Figure 4. A comparison between Figures 2 and 4 indicates that helium ions result in significantly higher instantaneous radiation-induced conductivity (evident from the large instantaneous rise in conductivity when the beam is turned on). Similar to the proton irradiation, a breakaway phenomenon was observed after an incubation period of approximately 200 minutes.

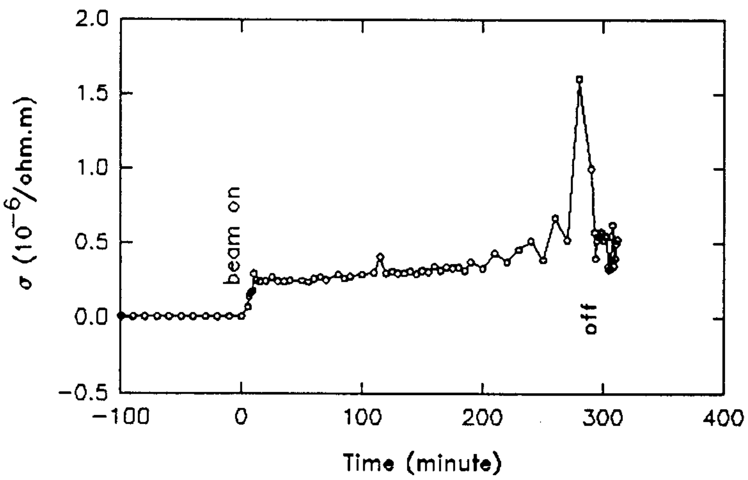

FIGURE 4. Radiation-Induced Conductance in Alumina Irradiated With 2 MeV Alpha Ions; Notice the Large Initial Radiation-Induced Conduction.

Results from alumina irradiated with gamma radiation from a Co^{60} source at temperatures between ambient and 753 K are given in Figure 5. In this experiment, an electric field of 160 kV/m was used. The specimen was exposed to the gamma source for a duration of 5 minutes at each temperature and the radiation-induced conductivity was recorded. The presence of gamma radiation induced an instantaneous increase in the conductivity. The removal of the beam at the end of the irradiation period resulted in an instantaneous and complete recovery indicating that no permanent electrical degradation has occurred due to irradiation. It is evident that electrical conductivity induced by gamma is recoverable and is independent of temperature as expected for photon-induced band to band transition.

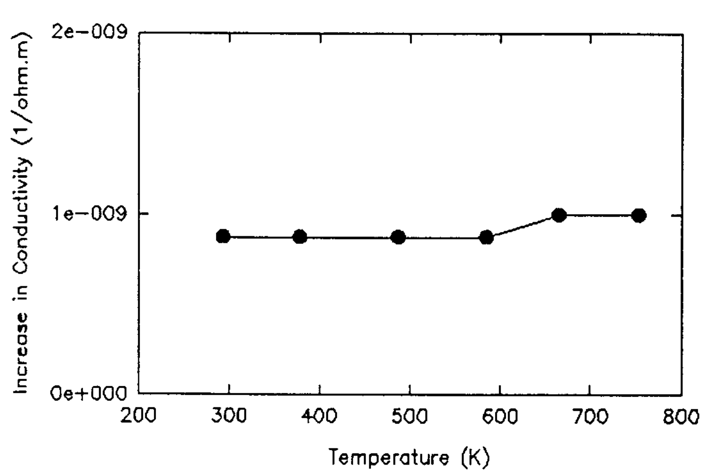

FIGURE 5. Effects of Gamma Radiation on Conductance in Alumina.

Unfortunately, the rate dependence could not be investigated since the intensity of the gamma source is not adjustable. Instead an x-ray beam from an x-ray diffractometer was used for this purpose. Results are shown in Figure 6 for an x-ray with a peak energy of 40 keV. The radiation induced conductivity was found to increase with x-ray intensity in a non-linear manner (power of about 0.7). This induced conductivity was completely recoverable (the resistance returned to the original value upon removal of the beam). Fowler has developed a model for radiation-induced conductivity in materials with a band gap. According to this model, the excitation process in which electrons are excited to the conduction band by radiation and holes are left in the valance band without recombination contributes to the conductivity of the materials. The nonequilibrium electron density in the conduction band depends on the number of electrons being excited by radiation and the average time an electron resides in the conduction band. The change in conductivity due to radiation is frequently described by a power law of the form $\Delta\sigma = k\ \Phi^n$, where k is a constant and Φ is the radiation intensity. The exponent n is usually below one. This is in agreement with the observation of n=0.7 obtained in this study.

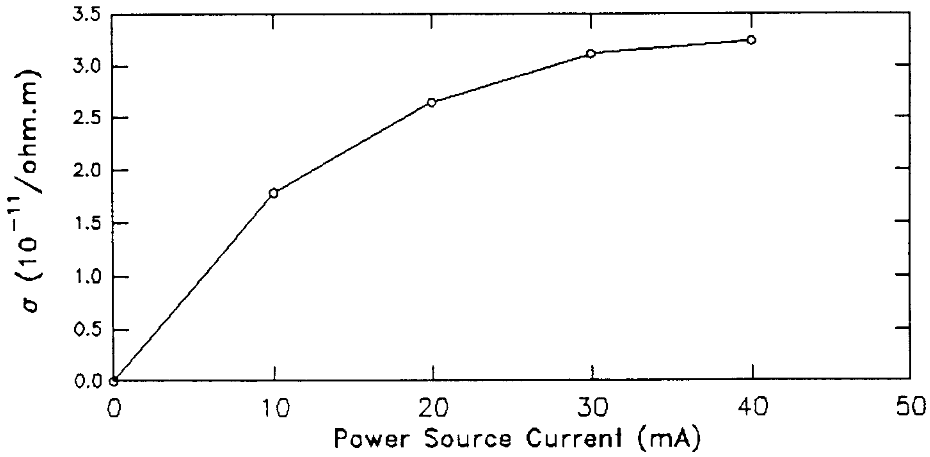

FIGURE 6. Effects of X-Radiation on Conductance in Alumina.

A comparison of the relevant parameters from different irradiation studies (including neutrons, ions, electrons gamma and x-ray) is given in Table 1.

TABLE 1. Comparison of Various Radiation Effects.

	Temp (K)	E (kV/m)	Damage Rate (dpa/s)	Ionization Rate (Gy/s)	$\Delta\sigma$ $10^{-10}(\Omega m)^{-1}$	Ionization Efficiency	Incubation Dose (10^{-4} dpa)
Neutron[a]							
On	773	25-500	$3.4x10^{-6}$	$1.4x10^{4}$	600	0.04	150
Off	773	25-500	0	150	3	0.02	---
Proton[b] (18MeV)	773	500	$4x10^{-7}$	$5x10^{5}$	10,000	0.02	10
Electron[c] (1.8MeV)	773	130	10^{-10}	278	200	0.7	0.1
Proton (2 MeV)	673	500	$3x10^{-8}$	$2x10^{5}$	200	0.001	2
Helium (2 MeV)	703	500	$2x10^{-7}$	$2x10^{6}$	2000	0.001	20
Co^{60} (γ)	300-753	160	0	0.5	0.2	0.4	---
X-Ray	300	160	0	0.28	0.002	0.006	---

[a]From Shikama 1992.
[b]From Pells 1991.
[c]From Hodgson 1991.

The damage rates (given in displacement per atom per second, dpa/s) and the ionization rates were calculated according to the type of irradiation and the intensity. The former is related to the structure damage introduced by the irradiation whereas the latter controls conductivity via band-to-band transition. The parameter $\Delta\sigma$ is the instantaneous increase in conductivity when the irradiation is turned on. Ionization efficiency is defined as the ratio of radiation-induced conductivity ($\Delta\sigma$) to ionization rate and it is a measurement of the effectiveness of the particular ionization mechanism to generate conductivity. It is evident that electron and gamma irradiations are most effective in promoting conductivity due to their high probability of interacting with electrons in the material. It appears that neutrons also possess relatively high ionization efficiency which is unexpected due to the neutral state of the particle. However it is important to note that neutron irradiations were conducted in a nuclear reactor where high intensity gamma was also present. The ionization process in this case is due to such electromagnetic radiation. The last column in table 1 gives the incubation dose (in dpa) when breakaway conductivity was observed under different irradiation conditions. It appears that alumina irradiated with electrons requires the least incubation prior to onset of breakaway (10^{-5} dpa as opposed to 0.015 dpa for neutron). This indicates that electrostatic separation of isolated defects produced by low energy transfer process in electron irradiation is more effective in generating conductivity than high energy cascades produced by ions and neutrons. This suggests that the damage structure responsible for radiation-induced electrical degradation is due to radiation-enhanced separation of the ionic species.

SUMMARY

Both recoverable and permanent radiation-induced conductivity in alumina are observed in proton and helium ion irradiations. The recoverable conductivity is related to the excitation of electron-hole pairs induced by ionization whereas the permanent conductivity is governed by the microstructure of the materials. Comparison of various radiation effects indicates that electrons and gamma rays have higher ionization efficiencies and the damage structure responsible for radiation-induced permanent electrical degradation is due to radiation-enhanced separation of the ionic species.

Acknowledgment

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