ELSEVIER
Nuclear Instruments and Methods in Physics Research B 166–167 (2000) 159–164
NIM B
Beam Interactions with Materials & Atoms
www.elsevier.nl/locate/nimb

Electrical insulating potential of aluminum nitride under irradiation with fast electrons

M.M.R. Howlader, C. Kinoshita *, K. Shiiyama, M. Kutsuwada

Department of Applied Quantum Physics and Nuclear Engineering, Kyushu University 36, Hakozaki 6-10-1, Fukuoka 812-8581, Japan

Abstract

In order to evaluate the electrical insulating potential of aluminium nitride as a radio frequency (rf) heating device, neutral beam injectors, magnetic coils, etc. in fusion reactors environments, the current–voltage (*I–V*) characteristics and electrical conductivity of poly-crystalline aluminum nitride, doped with 1 wt% Y_2O_3 (AlN–1 wt% Y_2O_3) have been measured before and during irradiation with 1 MeV electrons at temperatures up to 723 K. The *I–V* characteristics of AlN without irradiation are non-linear, while those with irradiation exhibit hysteresis. Blocking effect without irradiation is considered, and it changes with fast electron irradiation. Inconsistency in the *I–V* characteristics of AlN between positive and negative polarity during irradiation is found, and it is thought to be due to the electron charge deposition onto the single surface of the specimen. No bulk and surface RIED are found up to 1.5×10^{-5} dpa. If the alpha-particles produced by high neutrons are assumed to be negligible, then AlN can be used as an alternative candidate material for the coating on the blanket of fusion reactors, because of the absence of severe thermally stimulated conductivity (TSC) and bulk and surface degradation under fusion reactors environments. © 2000 Elsevier Science B.V. All rights reserved.

PACS: 72.20.–i; 81.40.Rs
Keywords: Radiation induced conductivity; Radiation induced electrical degradation; Thermally stimulated conductivity; Aluminum nitride; Electron irradiation

1. Introduction

Aluminum nitride (AlN) is potentially being used in microelectronic packaging applications [1] and recently is proposed for a variety of fusion reactors applications as a first wall coating, magnet insulation, invertor, diverters, radio frequency (rf) windows and instrumentation [2], as well as for high radiation environments due to its high insulating behavior, high thermal conductivity [3] and large band gap energy of 6.5 eV [4]. AlN is characterized by mixed ionic and covalent bonding, and the defects present in the crystals act as the source of conduction electrons at temperatures. Irradiation with high energy radiation can energize these defect produced electrons, which are enough

* Corresponding author. Tel.: +81-92-642-3771; fax: +81-92-642-3771.
E-mail address: c.k.tne@mbox.nc.kyushu-u.ac.jp (C. Kinoshita).

PII: S0168-583X(99)00740-5

to results in radiation induced conductivity (RIC) through the excitation of electrons from valence to conduction bands [5]. AlN may also exhibit losses under irradiation in the presence of an electric field, which causes fracturing. In fact, the electrical properties of insulating materials may degrade permanently when irradiating for a long time in the presence of electric field at moderate temperatures, so called radiation induced electrical degradation (RIED) [6], and the RIED can cause malfunctioning in instrumentation.

In order to get insights into the mechanisms of the conduction process, it is necessary to understand the processes involved in the current–voltage (*I–V*) behavior and electrical conductivity of ceramic insulators before and during irradiation. To do this, it is indispensable to consider the conditions for Ohmic, neutral, and blocking or Schottky barrier (which occur between the electrode and specimen surface) of $\phi_i > \phi_c$, $\phi_i = \phi_c$ and $\phi_i < \phi_c$, respectively [7]. Here, ϕ_i and ϕ_c represent the work function of insulator and electrode material, respectively. The purpose of this study is to understand the conduction mechanisms responsible for Ohmic and non-Ohmic behavior and to get more understanding of the conduction process of AlN with and without electron irradiation for evaluating the electrical insulating potentiality in fusion environments.

2. Experimental procedure

Brownish-colored, well-polished polycrystalline AlN with 1 wt% Y_2O_3 (AlN–1 wt% Y_2O_3 having purity greater than 99.0%) specimens of 5.5 mm diameter and 200 and 500 μm-thicknesses were used in this study. Titanium was deposited on the specimen surfaces to act as center, guard and base electrodes in the vacuum pressure of 10^{-4} Pa. The configuration of the three electrodes system in this work is same as described previously [8]. Electrical measurements were done before and during irradiation. Before irradiation, a bell jar with vacuum pressure of 10^{-4} Pa was used to perform the experiments in the temperature range of 300–723 K with an electric field of 150 kV/m. During irradiation, the experiments were carried out in the HVEM at Kyushu University under 1 MeV electrons irradiation with beam-on and -off conditions in the same temperature range and applied electric field as the unirradiation experiments.

3. Results and discussion

3.1. Unirradiated specimen

An Arrehenius plot of electrical conductivity for a 200 μm-thick AlN–1 wt% Y_2O_3 specimen with an applied electric field of 150 kV/m before and during irradiation is given in Fig. 1. The irradiation data consist of the conductivity at two ionizing doses of 1.8×10^4 and 7.4×10^4 Gy/s. Two distinct bands of activation energy in the unirradiation conductivity were observed. Between room temperature and 480 K the activation energy was 0.26 eV, and from 480 to 723 K it was 0.81 eV. Fig. 2 shows the *I–V* curves of a 200 μm-thick AlN–1 wt% Y_2O_3 specimen at 423 and 723 K before irradiation. In these measurements, the voltage was applied to the specimen for 1 and 5 min. The amplitude of the negative quadrant of *I–V*

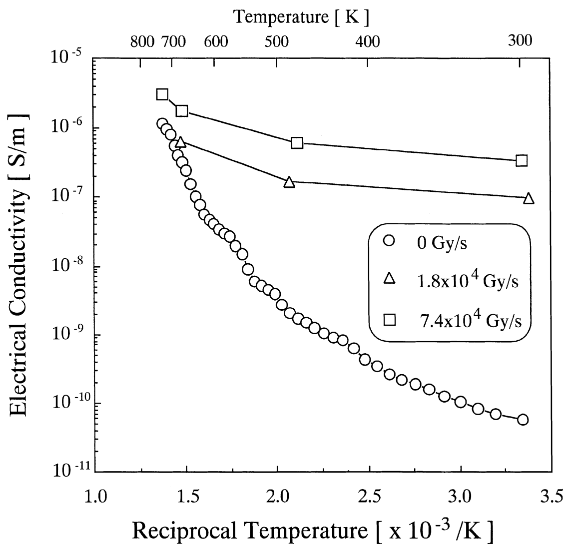

Fig. 1. Semi-log plot of electrical conductivity as a function of reciprocal temperature for a 200 μm-thick poly-crystalline AlN before and during irradiation with 1 MeV electrons in an electric field of 150 kV/m.

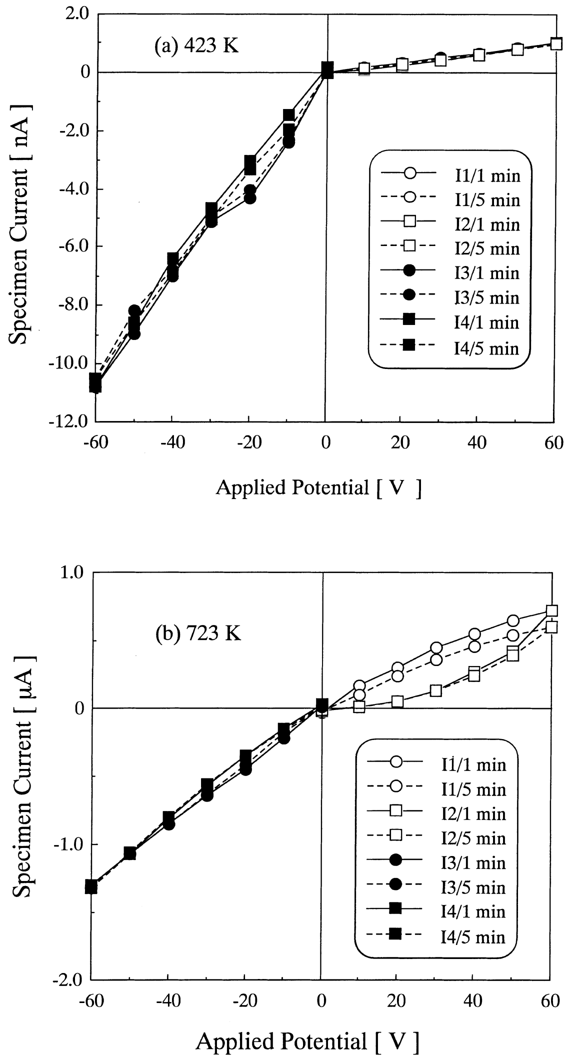

Fig. 2. *I–V* relationships of a 200 μm-thick poly-crystalline AlN with Ti electrodes before irradiation at (a) 423 K and (b) 723 K. The solid and dotted lines correspond to the specimen current for 1 and 5 min, respectively. The open and filled symbols correspond to the potential applied to the base and center electrodes, respectively. Here, I1, I2, I3 and I4 represent the first, second, third and fourth run numbers of the experiments.

curves is higher than that of the positive quadrant at various temperatures. Time dependent reduction of the specimen current at various potentials is found. According to Simmons [7], Ohmic, blocking (Schottky) and neutral contacts can be formed across the interface between the electrode metal and insulator in the consequence of $\phi_i > \phi_c$, $\phi_i < \phi_c$ and $\phi_i = \phi_c$, respectively. The Ohmic contact acts as a reservoir of charge, which is capable of supplying electrons to the insulator; thereby space charge conduction is present in the insulator. In the case of blocking contact, electrons flow from the insulator into the electrode for maintaining thermal equilibrium. For the neutral contact, zero charge transfer exists between interfaces resulting in no space charge in the insulator and no bending in the band. Consequently Ohmic and neutral contact characteristically are bulk limited whereas blocking (Schottky) contact is electrode limited. The non-Ohmic behavior is apparent in Fig. 2. The non-symmetric behavior of *I–V* curves with respect to the polarity of applied voltage was analyzed based on the electrode-limited blocking effect, including space charge conduction.

3.2. Irradiated specimen

In order to understand the effect of irradiation with fast electrons on blocking effect, *I–V* measurements of a 200 μm thick AlN–1 wt% Y_2O_3 specimen irradiated with a 1 MeV electron dose rate of 1.2×10^{18} e/m^2 s (1.4×10^{-9} dpa/s and 7.4×10^4 Gy/s) at 296 and 480 K were performed and their results are shown in Fig. 3. Unlike unirradiation *I–V* measurements, the specimen current was measured here after reaching the stable reading with the application of electric field (after about 1 min) at each step. Hysteresis and asymmetry between the positive and negative quadrant value are present in the *I–V* curves. The apparent amplitude at the positive quadrant of *I–V* curves is higher than that of the negative quadrant, contrasting to the unirradiation *I–V* curves which might be due to the continuous irradiation onto the single surface of the specimen. Hence, the *I–V* curves with and without irradiation indicates that irradiation changes the blocking effect and deposits electronic charge in the specimen [9,10]. Therefore, it is concluded from the *I–V* behavior that the specimen current is bulk dominant under these conditions. Also, the activation energy reduces to 0.08 eV during irradiation with 7.4×10^4 Gy/s, indicating the change in the conduction process due to irradiation. Fig. 4 shows

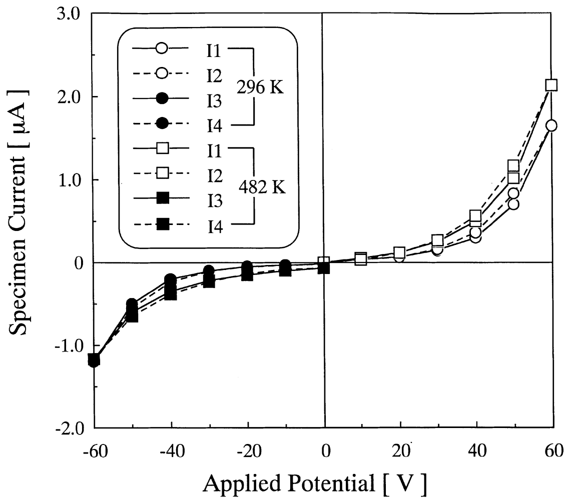

Fig. 3. *I–V* relationships of a 200 µm-thick AlN with Ti electrodes with a 1 MeV electron flux of 1.2×10^{18} e/m^2 s at 296 and 723 K. The solid and dotted lines correspond to the increasing and decreasing applied potential, respectively. Here, I1, I2, I3 and I4 represent the sequential numbers for the experiments.

the time dependence of electrical conductivity of a 200 µm-thick AlN–1 wt% Y_2O_3 specimen with a 1 MeV electron dose rate of 1.2×10^{18} e/m^2 s with beam-on and -off at 296 and 482 K. The conductivity suddenly increases at both temperatures after turning the electron beam on by almost four orders of magnitude compared to unirradiated state and then varies almost constantly with time. The result also indicates that the difference in the conductivity at 296 and 482 K under beam-off condition is higher than that under the beam-on condition. No TSC peaks are found in the AlN specimen. The abruptly increasing behavior of electrical conductivity with increasing temperatures, indicating thermal excitations of electrons from impurity levels. The decrease in electrical conductivity with increasing annealing time at each temperature is most likely due to the annealing of the trapping centers associated with point defects produced during irradiation.

The dose rate dependence of electrical conductivity of AlN–1 wt% Y_2O_3 specimen is plotted in Fig. 5 along with the results of the Lindau and Möslang [11] and Ulmanis and Palcevskis [12]. The conductivity is proportional to both the ionizing dose rate and the temperature. The results also imply that the conductivity increases with increasing beam intensity irrespective of irradiation type and energy. The results suggest that AlN can be used for the coating materials of the blanket in fusion reactors provided that the alpha-particles produced by high neutrons are not dominant.

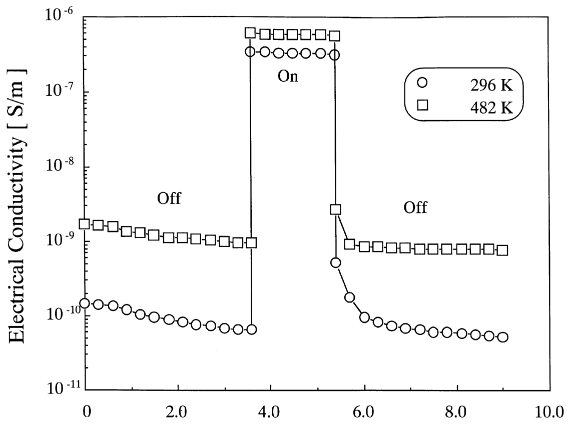

Fig. 4. Time dependence of electrical conductivity of a 200 µm-thick AlN specimen having Ti electrodes with a 1 MeV electron dose rate of 1.2×10^{18} e/m^2 s with beam-on and -off in a dc electric field of 150 kV/m at 296 and 482 K.

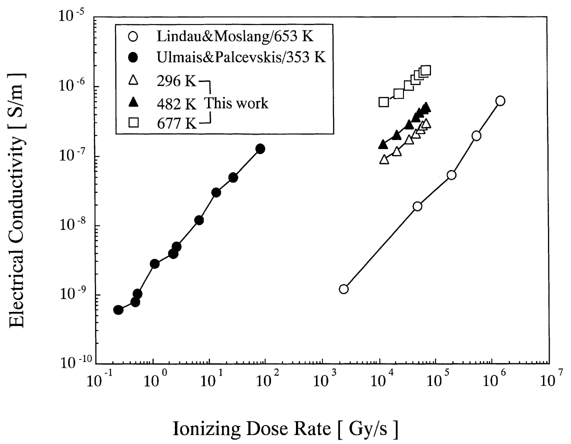

Fig. 5. Ionizing dose rate dependence of electrical conductivity for a 200 µm thick AlN specimen at 296, 482 and 677 K compared with those of Lindau and Möslang [11] and Ulmais and Palcevskis [12].

Fig. 6 shows the total dose dependence of the bulk and surface conductivity for 200 and 500 μm-thick AlN–1 wt% Y_2O_3 specimens under irradiation with a 1 MeV electron dose rate of 1.2×10^{18} e/m^2 s in a dc field of 150 kV/m at 723 K. Before irradiation, the bulk conductivity decreases with time; however, the bulk conductivity suddenly increases after turning on the beam, by more than one order of magnitude compared to the unirradiated value, and then gradually increases with time. After turning off the electron beam, the conductivity reduces to the unirradiated value and then recovers with respect to time. The analogous increase and decrease in RIC of alumina under 1 MeV electron irradiation have been found [8] and interpreted in terms of the effect of electronic excitation and the interaction between radiation-induced defects and free electrons in the insulator. The surface conductivity during irradiation increases by factors of two or three over the unirradiated surface conductivity, and then gradually decreases with time. No bulk and surface RIED have been found up to 1.5×10^{-5} dpa. Lindau and Möslang [11] have irradiated AlN using 104 MeV alpha-particles during beam-on and -off with a dc electric field of 100 kV/m up to 0.08 dpa at 573, 623 and 773 K. They have found two to three orders of magnitude difference between the beam-on and -off conductivity. They have also checked

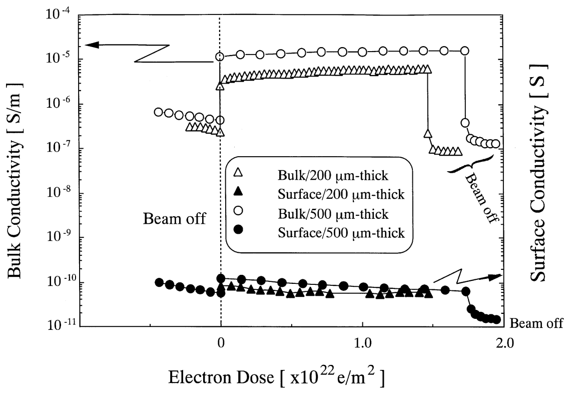

Fig. 6. RIC of 200 and 500 μm-thick AlN specimens having Ti electrodes under 1 MeV electron irradiation with increasing and decreasing beam intensity at 296, 482 and 677 K in a dc electric field of 150 kV/m. The open and filled symbols correspond to increasing and decreasing beam intensity, respectively.

the error (surface conductivity) in the bulk conductivity according to Kesternich [13] and argued that the conductivity changes are bulk limited. In addition, no significant RIED has been evident up to 0.08 dpa in their study. On the other hand, in this study, the beam-on and -off conductivity differs by a magnitude of four, two and half and one and half orders, respectively at 296, 480 and 723 K. This fact indicates that the electrical conductivity of AlN at higher temperatures is dominated by thermal rather than ionization processes, as in Wesgo AL995 alumina [14]. However, the results of bulk and surface conductivity also show the different amplitudes for 200 and 500 μm-thick specimens. The same trend of explanation of the thickness dependence of the RIC of Kyocera single crystalline alumina and polycrystalline Wesgo AL995 alumina [14] is also applicable for this result of AlN–1 wt% Y_2O_3 specimen.

4. Conclusions

The *I–V* characteristics and electrical conductivity of poly-crystalline aluminum nitride, doped with 1 wt% Y_2O_3 (AlN–1 wt% Y_2O_3) specimen have been measured before and during irradiation in a bell jar and in a high voltage electron microscope with 1 MeV electrons, respectively, at temperatures up to 723 K. The *I–V* characteristics of AlN specimen without irradiation are non-linear while those with irradiation exhibit hysteresis. Blocking effects observed without irradiation change under fast electron irradiation. Inconsistency in the *I–V* characteristics of AlN between positive and negative polarity during irradiation is thought to be due to the electron charge deposition onto the surface of the specimen. The RIC is due to the excitation of band electrons. No bulk and surface RIED are found up to 1.5×10^{-5} dpa. Electron irradiation and subsequent annealing at different temperatures demonstrated the quality of irradiation produced trapping centers. Finally, AlN should be considered as a candidate material for coating the blanket of fusion reactors unless the alpha-particles produced by high neutrons are assumed to be dominant.

References

[1] J.S. Throp, D. Evans, M. Al-Naief, M. Aktaruzzaman, J. Mater. Sci. 25 (1990) 4965.
[2] F.W. Clinard Jr., J. Nucl. Mater. 85–86 (1979) 393.
[3] G.A. Slack, R.A. Tanzilli, R.O. Pohl, J.W. Vandersande, J. Phys. Chem. Solids 48 (7) (1987) 641.
[4] R.H. French, J. Am. Ceram. Soc. 73 (3) (1990) 477.
[5] S.J. Zinkle, E.R. Hodgson, J. Nucl. Mater. 191–194 (1992) 58.
[6] E.R. Hodgson, J. Nucl. Mater. 179–181 (1991) 383.
[7] J.G. Simmons, DC Cond. Thin Films, 1971.
[8] M.M.R. Howlader, C. Kinoshita, T. Izu, K. Shiiyama, M. Kutsuwada, J. Nucl. Mater. 239 (1996) 245.
[9] B. Gross, in: G.M. Sessler (Ed.), Electrets, Topics in Appl. Phys., Springer, New York, 1980, Vol. 33, p. 217.
[10] T. Marbeut, S.K. Krawczyk, Induced Defects in Insulators, 1984, p. 77.
[11] R. Lindau, A. Möslang, J. Nucl. Mater. 233–237 (1996) 1294.
[12] U. Ulmanis, E. Palcevskis, J. Nucl. Mater. 252 (1998) 195.
[13] W. Kesternich, F. Scheuermann, S.J. Zinkle, J. Nucl. Mater. 219 (1995) 190.
[14] M.M.R. Howlader, C. Kinoshita, K. Shiiyama, M. Kutsuwada, to be submitted.