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# Radiation-induced electrical conductivity in aluminium nitride ceramic

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## Abstract

The electrical conductivity of aluminium nitride ceramics, doped with 8 wt% $Y_3O_3$ and obtained from fine raw powders synthesized in plasma, was measured with and without gamma-neutron irradiation (the maximum fast neutron flux was $3 \times 10^{15}$ n/m$^2$ s and in the gamma-dose rate range of $10^3$–$10^6$ Gy/h) in the temperature range of 40–400°C. The electrical conductivity at room temperature was $10^{-12}$ S/m and temperature-induced increase of conductivity has an activation energy of 1.06 eV at a temperature range of above 160°C. Radiation-induced conductivity obeys the usual power law dependence on gamma-ray dose rate. The electrical conductivity at high dose rate is less than $10^{-6}$ S/m and is little dependent on temperature. © 1998 Elsevier Science B.V.

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## 1. Introduction

The growing importance of ceramic insulating materials for the successful operation of fusion reactors is now well established. Initially oxide ceramics, particularly alumina, were mainly proposed as insulating materials. But attention is also turned to aluminium nitride (AlN) as one of the feasible materials with high electrical resistance and thermal conductivity [1–4]. It is known that the intrinsic electrical conductivity of AlN ceramics depends on purity, amount of defects, technology of production, temperature and surrounding gases [2–4]. The sintering additives, for example $Y_2O_3$, which are used for ensuring high thermal conductivity, also improve the dielectric properties. Such sintering additives bind oxygen, which is dissolved in the AlN lattice and thereby diminish the concentration of aluminium vacancies in the lattice and electrical conductivity of AlN materials [3].

The radiation-induced conductivity (RIC) of alumina, magnesia and spinel ceramic during irradiation was investigated [5–9]. The electrical conductivity during irradiation may be expressed as an empirical relation

$$\sigma = \sigma_0 + \sigma_r = \sigma_0 + KP^d, \qquad (1)$$

where $\sigma_0$ is the conductivity in the absence of the irradiation, and $P$ is the ionizing dose rate. It is known that $K$ and $d$ are not constants, but are dependent on irradiation temperature, dose rate, material defects and impurities. For example, in the case of alumina the value $d$, which is directly obtainable from the experimental curve, is a complex function vs. temperature [5]. The value of the radiation dose rate exponent $d$ is generally between 0.5 and 1.0 according to a quasicontinuum of discrete states and is determined by the balance between production of free charge carriers by ionization and trapping and recombination of electron–hole pairs. The value of the proportionality constant $K$ has a strong dependence on material with typical values at room temperature ranging from $10^{-12}$ to $10^{-9}$ (S/m)/(Gy/s) [10].

In some cases the radiation-induced electrical degradation (RIED) of electrical properties in insulating oxides

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irradiated at high temperatures in electrical field was observed [6,11–13]. It can be expected that in AlN having predominantly covalent bonding in comparison with alumina the degradation of insulator properties would be decreased. Some results about AlN behavior in radiation field are given in Refs. [10,14].

In the present paper we show the results of resistivity measurements of aluminium nitride ceramics under gamma-neutron irradiation in situ in the Latvian 5 MW light water–water research reactor IRT.

## 2. Experimental procedure

Fine AlN and $Y_2O_3$ powders with specific surface area of 40 and 30 $m^2/g$, respectively, produced by the plasma synthesis in the Institute of Inorganic Chemistry were used for preparation of ceramic samples [15]. The samples were disc-shaped, 10 mm in diameter and 0.8 mm thick. Sintering was carried out in nitrogen (0.12 MPa) in a tungsten resistance furnace at 1810 ± 5°C. The total oxygen content of sample was 3.4 ± 0.2 wt%, including 1.7 wt% bound in $Y_2O_3$ and 1.7 wt% bound in $Al_2O_3$ (3.0 wt%) in as prepared AlN powder. Approximately equal mole-content of $Y_2O_3$ and $Al_2O_3$ was also present. The density of samples was $(3.34 \pm 0.01) \times 10^3$ $kg/m^3$.

The $Y_2O_3$ additive allows one to obtain non-porous AlN ceramics with practically theoretical density and thermal conductivity above 150 W/m K. Such thermal conductivity corresponds to the oxygen content of 0.1–0.2 wt% in AlN lattice and the AlN lattice also becomes less defective [16].

The horizontal experimental channel (diameter of 100 mm and length of 2.8 m) of the research reactor IRT was used as a radiation source [17]. Maximum gamma dose rate measured with RFT gamma-dosimeter was $10^6$ Gy/h and maximum fast neutron ($> 0.1$ MeV) flux was of $3 \times 10^{15}$ $n/m^2$ s. It may be assumed [18–20] that in these experimental conditions the generation rate of charge carriers by gamma irradiation is of one to two orders higher in comparison with the generation rate of the charge carriers which are determined by the charged knock-on atoms produced by the elastic scattering of fast neutrons. Irradiation facility (Fig. 1) allows one to transport the sample holder to the active core of reactor and to change the gamma dose rate of three orders (1 kGy/h–1 MGy/h) placing the sample 3.0 to 0.2 m from the active core [17].

The samples with two thermally deposited silver electrodes with a diameter of 3 mm are held sandwich-like between two flat, spring loaded nickel electrodes which enable the application of the electric field to the sample. The samples with measuring cables are located in the resistance furnace which allows to keep the temperature during irradiation up to 500°C. The temperature of the samples was measured in situ with chromel–alumel thermocouple inserted in the heater 2–3 mm behind the sample. In this case there is some difference between the data of thermocouple and the temperature of sample depending on heating rate.

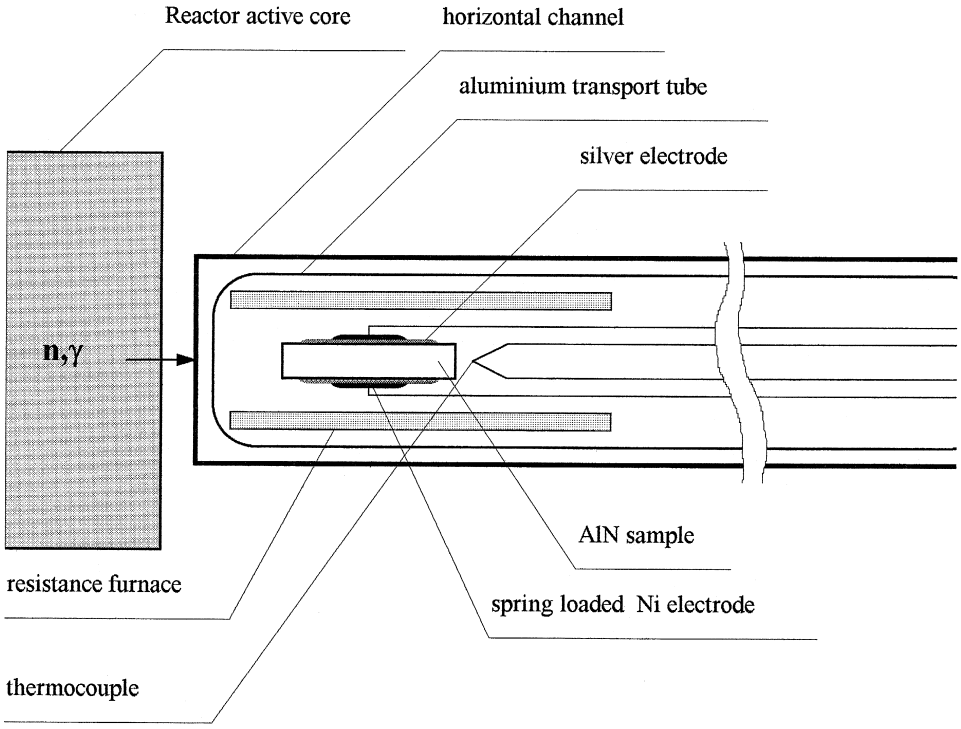

Fig. 1. Schematic illustration of irradiation system for in situ measurements of electrical conductivity.

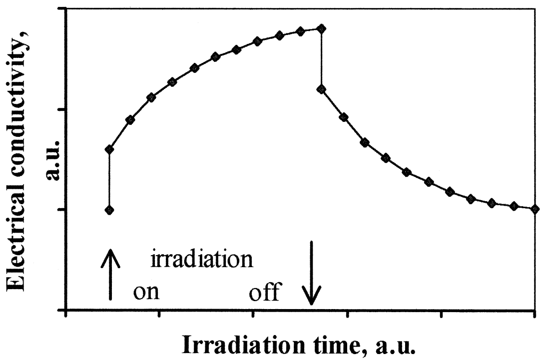

Fig. 2. Radiation-induced electrical conductivity of AlN ceramics versus irradiation time.

The electrical resistance of samples was measured by the conventional dc two terminal method in air. There are some peculiarities in measuring the electrophysical properties of insulating materials during irradiation — the radiation induced change of neighboring medium, construction materials and measuring detectors. The reactor in situ measurements have a lot of technical problems. The intense radiation field in the reactor is ionizing the air around the sample. As a result a parasitic electric current is generated, which can disturb the measurements. The analysis of in situ electrical conductivity measurements in radiation fields is given in Refs. [21–23] and the guard ring technique and vacuum system are recommended to avoid the influence of the surface conductivity. Due to the experimental constraints this technique was not applicable in the present case. Since a guard ring was not used and the conductivity was measured in air, some stray leakage current may have been included in the conductivity values and therefore pre- and post-irradiation conductivity values at low temperature can be used only for approximate comparison. A similar situation is discussed in Ref. [12] where using two-terminal dc electrodes in air and assuming that surface current will be very small to compare the radiation-induced conductivity. The electrical conductivity measurements were made at constant measuring voltage in the range of 2.5–100 V (usually 10 V) to decrease the influence of the polarization effects. We have performed some experiments using air and different materials (MgO, alumina, etc.) in the place of AlN sample to estimate the role of additional currents due the surface conductivity and surrounding air ionization.

During investigations it was observed that the RIC depends on irradiation time. Fig. 2 shows typical behavior of RIC of our samples during irradiation time at one step. The RIC value consisted of an instantaneous conductivity and of an irradiation time dependent component: conductivity increases from the initial value until the finite value immediately after the irradiation is switched on and increases asymptotically with the time to a constant value for prolonged irradiation (for AlN ceramic up to 10 min at 100°C). After termination of irradiation the RIC of the sample did not disappear instantaneously, but rather the RIC initially diminished rapidly followed by a slow and gradual decay. This behavior of RIC repeated after each step change of the dose rate and was taken into account to obtain conductivity vs. dose rate dependence.

## 3. Experimental results and discussion

The measured dc conductivity in the reactor gamma-neutron field has a contribution from both temperature and ionization dose rates. The effect of temperature on the electrical conductivity before and during irradiation in AlN (8% $Y_2O_3$) ceramic samples is presented in the usual manner where the ordinate representing the electrical conductivity is plotted on a logarithmic scale as a function of the reciprocal of the absolute temperature (Fig. 3). The curves 1 and 2 show the temperature dependence of the conductivity of preirradiated samples and it is obvious that during the first thermal cycling (curve 1) when the temperature increases up to 160°C, the decrease of conductivity takes place. During repeated thermal cycling only increase of conductivity versus temperature is observed and a negligible difference between results of second and third thermal cycling exists (curve 2). Similar experimental curves were obtained in Ref. [24] for single crystal $Gd_3Ga_5O_{12}$ when electrical conductivity was measured by two and three electrode technique. This fact may be explained by the surface contamination, moisture, etc., which contribution is more obvious at lower temperatures. Also, the

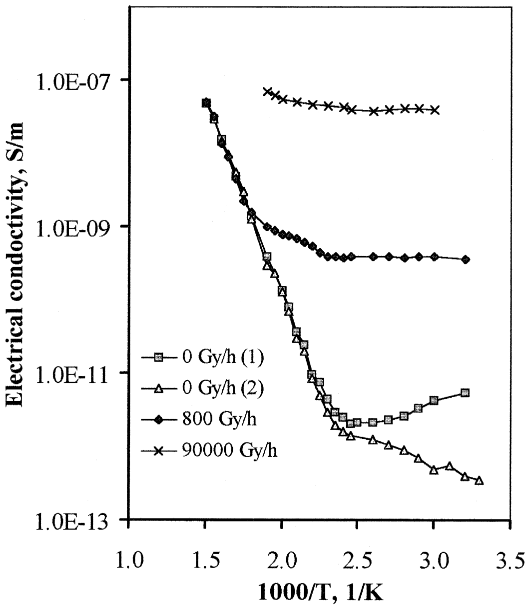

Fig. 3. Electrical conductivity of AlN ceramics versus reciprocal temperature with and without irradiation. (1) The sample as prepared and (2) after thermal cycling.

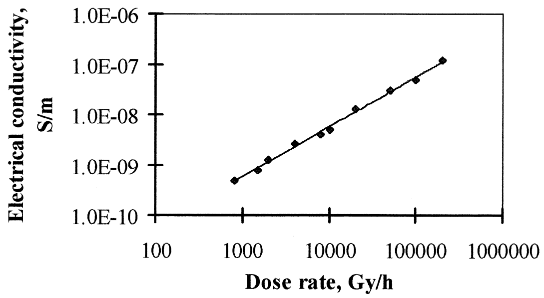

Fig. 4. Electrical conductivity of AlN ceramics versus gamma dose rate at 80°C.

guard ring has an influence on the experimental results in particular at temperatures below 100°C.

The measured electrical conductivity at room temperature in the absence of radiation was $10^{-12}$ S/m, coinciding with the literature data [1]. We assume that the surface conditions increase the measured conductivity and the intrinsic value could be somewhat lower.

The electrical conductivity of AlN before irradiation has the activation energy of 1.06 eV above 160°C and changes insignificantly below this temperature (the activation energy is approximately 0.16 eV). The conductivity dependence on temperature is similar to that for another oxide ceramic, for example for alumina [7,9]. For such dielectric materials the production of charge carriers in temperature range from room temperature to 500°C is connected with the electrons and holes, released from different trapping levels in forbidden band [5].

Radiation influences the electrical conductivity more in the region of low temperatures. During irradiation the activation energy of electrical conductivity decreases and for the dose rate of 800 Gy/h the value of 0.2 eV in a temperature range of 160–280°C was calculated. For high dose rates ($> 4 \times 10^4$ Gy/h) the conductivity is practically independent of temperature. The magnitudes of the observed conductivity and activation energy are about the same as the reported values of RIC and activation energies in alumina and other ceramics [7–9].

The important result of this investigation is that the electrical conductivity of AlN sample is less than $10^{-6}$ S/m at dose rates up to $10^5$ Gy/h. According to Ref. [10] such conductivity is low enough for application of AlN ceramics as insulator materials under gamma-neutron irradiation.

The conductivity dependence on the dose rate was measured at 80°C (Fig. 4). It is shown that the radiation-induced changes of conductivity versus gamma-dose rate is practically linear in the dose rate interval up to $10^6$ Gy/h. Our experimental data also reveal that the dose rate exponent $d$ and constant of proportionality $K$ are independent on the dose rate and their values ($d = 1.0$ and $K = 6 \times 10^{-13}$ (S/m)/(Gy/h)) are near to the corresponding values for other ceramic materials [6–9].

To increase the reality of these results we have measured the resistance of air and some dielectric materials (MgO, $Al_2O_3$, $Gd_3Ga_5O_{12}$) vs. temperature and dose rate in the same experimental conditions. The resistance of the air at room temperature before irradiation is of the same order as the AlN sample. With increasing the temperature we observe a weak change of air resistance and at $T >$ 160°C the resistance of the sample is one to two orders less than for air. We must consider that the air shunting path during sample measurement is 8–10 mm and thus the electric field applied to the sample is less than $10^4$ V/m. The dose rate dependence for air is similar to that for AlN, but the exponent constant is 0.6 compared with $\sim 1.0$ for AlN. The conductivity of AlN at room temperature without irradiation is strongly dependent on surface conductivity, but at higher temperature the role of surface conductivity must be diminished. We assume that during irradiation the influence of surface conductivity is also small; we observe no irreversible changes of conductivity before and after irradiation and practically no hysteresis effect in conductivity vs. dose rate, increasing and decreasing dose rate. The dose and temperature dependencies for other materials (MgO, alumina and gadolinium gallium garnet single crystals) measured in analog manner [25] are similar in character but different in activation energies and dose rate exponents. We observe no change of optical absorption in these crystals. Our experimental results in first approximation coincide with results of Lindau and Möslang [14] for AlN ceramic irradiated with 104 MeV α-particle beam and conductivity measured with three electrode system in vacuum (before irradiation $\sigma_0$ (80°C) $= 2 \times 10^{-12}$ S/m and $E_a = 0.9$ eV, in dose rate range $10^2$–$10^6$ Gy/s d ~ 1.0 eV).

Thermally stimulated conductivity (TSC) measurements of irradiated samples have confirmed the role of the

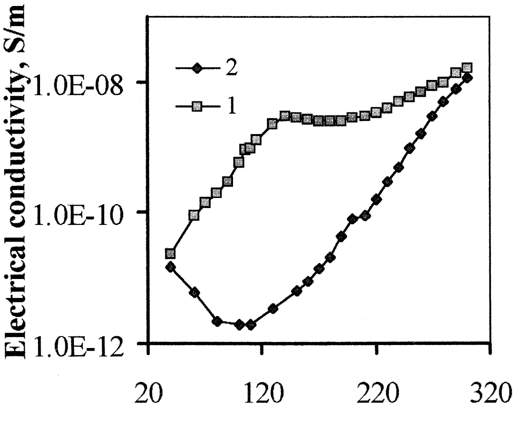

Fig. 5. Electrical conductivity of irradiated AlN ceramics versus temperature. 1 – directly after irradiation, 2 – after thermal cycling.

charge carriers trapping–detrapping processes during irradiation. Fig. 5 shows TSC measurements with a heating rate of 10°C/min carried out with sample previously irradiated with a dose of $10^5$ Gy (curve 1). We observed a broad TSC maximum at 150°C associated with the trapping centers with the activation energy of 0.5–1.0 eV. A repeated measurement, also after thermal cycling of the same sample (curve 2), shows practically the same electrical conductivity change versus temperature as before irradiation. It gives evidence that only the distribution of the donor and acceptor levels in the forbidden zone was changed as a result of the irradiation.

## 4. Conclusions

The electrical conductivity of AlN ceramic doped with 8 wt% of $Y_2O_3$ at the room temperature is $10^{-12}$ S/m. Temperature-induced increase of conductivity have activation energy of 1.06 eV at the temperature range above 160°C.

Radiation-induced conductivity in such ceramics possess the usual power law dependence on gamma-ray dose rate. The dose rate exponent is practically independent on the dose rate in the region of $10^3$–$10^6$ Gy/h. The electrical conductivity at high dose rate is less than $10^{-6}$ S/m and little dependent on temperature.

Thus the AlN ceramics produced from fine powders may possibly be used as an insulator material in fission and fusion devices.

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