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Electrical and optical characteristics of dielectrics for fusion use under irradiation

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Abstract

High values of electrical conductivity reported in several papers and ascribed to radiation-induced conductivity (RIC, with the magnitude of ionizing efficiency K exceeding 10^{-10} s/Gy Ω m) and radiation-induced electrical degradation in applied electric field (RIED) can not be explained within the framework of a reasonable physical model of bulk conductivity. Owing to charge carrier recombination on traps the RIC is not so high as to restrict the application of dielectrics in fusion reactors. The phenomenon of 'bulk RIED' is probably caused by surface contamination of dielectric materials during irradiation testing. More important problems concerning electric charging of dielectrics under irradiation and subsequent dielectric break-down are discussed. The radiation-induced dielectric break-down has been shown to occur in alumina without applying the electric field. The electric charging as well as features of RIC and RIL in dielectrics are attributed to the common mechanism of non-equilibrium charge carrier relaxation, namely, recombination on electron and hole traps. The presented results on RIL in Al_2O_3 based materials indicate local charge accumulation in the bulk of the materials caused by charge partitioning under irradiation. Electric charging and break-down are probably caused by charge partitioning in dielectrics under irradiation rather than primary particle charge accumulation. © 1998 Elsevier Science B.V.

1. Introduction

The stability of the properties (electrical, optical, mechanical, etc.) of dielectrics under long-term irradiation is an important problem in designing the diagnostic systems for fusion reactors. The materials based on BeO, MgO, Al_2O_3, $MgAl_2O_4$, BN, Si_3N_4 and AlN are either used or considered applicable in nuclear engineering. In fusion reactor relevant conditions these materials have to withstand a fluence up to 10^{26} n/m^2 and 10^{11} Gy, temperature up to 700°C and a voltage gradient up to 6×10^4 V/m.

The following phenomena have been reported to occur in dielectrics under irradiation: (a) radiation-induced electrical conductivity (RIC), luminescence (RIL) and optical absorption and (b) radiation-induced degradation (RID) concerning all of the properties (electrical, optical, mechanical) of dielectrics.

In recent studies the radiation-induced bulk electric degradation of dielectrics in applied electric field (RIED) was observed with conflicting values [1–17]. However, in this case a very complicated problem has arisen to extract bulk material properties from the 'material-facility' system. In this paper several well-known studies are reviewed to show that the high values of conductivity in dielectrics under irradiation ascribed to RIC (corresponding to the magnitude of ionizing efficiency exceeding 10^{-10} s/Gy Ω m) and RIED phenomena cannot be explained with the help of a reasonable physical model of bulk conductivity and the most probable cause of high conductivity values reported in several papers was leakage currents affecting the results of conductivity measurement under irradiation.

Other important problems are concerned with electric charging of dielectrics under irradiation and subsequent

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dielectric break-down. In our opinion the electric charging and break-down can be caused by electron–hole pair partitioning and subsequent local charge accumulation in dielectrics under irradiation rather than charge accumulation from the deposited primary particles. To monitor the local electric charging in Al_2O_3-based materials, RIL spectroscopy is applied.

2. Electric measurements under irradiation

The increase of conductivity of materials due to RIC is described by the equation [1–3]

$$\sigma = \sigma_0 + KR^{\delta}, \tag{1}$$

where σ_0 is the conductivity in the absence of irradiation, R is the ionizing dose rate and K (ionizing efficiency) and δ are constants depending on the material and irradiation conditions. For undoped Al_2O_3, the magnitude of δ is generally equal to 0.5–1, the magnitude of K varying in the range from 10^{-8} to 10^{-14} s/Gy Ω m [1–8]. It should be noted that the values of K much exceeding 10^{-10} s/Gy Ω m can not be explained with the help of a reasonable physical model, because these values contradict the fact that the electron lifetime in the conduction band of the tested dielectrics can not exceed 10^{-9} s.

In recent studies much attention was paid to the radiation-induced electric degradation of dielectrics in applied electric field (RIED) [1,8]. The most significant RIED (considerable increase of σ_0-component in Eq. (1)) has been observed at the temperature of 400–700 K, applied electric voltage exceeding 4×10^4 V/m, high ratio of ionizing to displacive constituents of irradiation [1,2]. Concerning the values of RIED presented in papers, a considerable discrepancy takes place. In Al_2O_3, the electric conductivity increased up to 10^{-5}–10^{-6} S/m under reactor irradiation [1,9–11] or 2 MeV electron irradiation [12]. No RIED has been found in other papers [1,3,5–8,13,14]. Similar values of the conductivity ascribed to RIED were observed at different irradiation conditions [9–12].

Both RIC and RIED magnitudes were reported to increase with the dose [15], i.e. the assumed bulk degradation (a possible cause may be electrical contact degradation [15]) results in an increase of the electron lifetime in the conduction band. However, taking into account a possible increase of trap concentration due to degradation, this lifetime should decrease.

The conductivity increase which has often been attributed to RIED was also observed at the temperatures exceeding the upper limit for RIED reported in the papers [1,2]. In-core measurements of conductivity of Al_2O_3 single crystals and ceramics were carried out under neutron flux of 7×10^{17} n/m^{2}s, the ionizing dose rate (γ-irradiation) being equal to 70 Gy/s ([8], reactor BR-10). The temperature of the samples at the maximum power of the reactor was 850–900 K and the applied voltage was 1.1×10^5 V/m. The RIC in Al_2O_3 samples was found to be 4×10^{-11} S/m. At the initial stage of the irradiation a substantial increase of the conductivity up to 10^{-5} S/m was temporarily observed (Fig. 1). Similar values registered in Refs. [9–12] following irradiation at lower temperatures were ascribed to RIED.

The measurements of RIC in pyrolitic BN (foils of 40 μm thickness) were carried out under 7 MeV proton irradiation with the flux of 1.3–2×10^{16} p/m^{2}s [8]. The experimental set-up provided measurements at the ionizing dose rate of 5×10^3 Gy/s and ionizing dose up to 10^8 Gy under normal conditions in an applied electric field up to 2.5×10^6 V/m, the temperature of a sample not exceeding 370 K. The pyrolitic BN electrical conductivity does not exceed 10^{-10} S/m, the initial value being equal to 10^{-12} S/m. Also, the irradiation with the dose of about 2×10^{20} and 5.8×10^{19} p/m^2 in the applied electrical field of 2.5×10^6 V/m does not change the electrical conductivity and its value remains at the initial level of 4×10^{-12} S/m [8].

According to these measurements the coefficient K in Eq. (1) was equal to 2×10^{-14} and 6×10^{-13} s/Gy Ω m for BN and Al_2O_3, respectively. This value can be explained [8] assuming the full particle energy transmitted into electron–hole pair formation and taking the band gap equal to 8 eV for BN and 9 eV for Al_2O_3, the electron lifetime in the conduction band equal to 10^{-13} and 10^{-11} s (caused by trapping at the temperature up to 1000 K) and the electron mobility equal to 10^{-4} m^2/(V s).

It was pointed out in Refs. [1,4,8,13,16] that the cause of high conductivity values reported in several papers was leakage currents affecting the results of conductivity measurement under irradiation. In the RIC measurements the leakage currents due to gas ionization, γ-photoeffect and surface contamination may arise. The formation of amorphous carbon and hydrocarbon layers on the surface of dielectrics can increase the surface conductivity and produce electrical behaviour that is similar to 'bulk' RIED [4,8,13,17]. In Ref. [15] the cause of apparent RIED in ceramics was attributed to radiation-enhanced diffusion of atoms from electrodes. The formation of conducting layers on the surface of Al_2O_3 single crystal and ceramic specimens has been studied by optical spectroscopy [8]. The bands 1650 and 1610 cm^{-1} in IR-reflection spectra were ascribed to the phase containing C=C (sp^2-hybridization) bonds in their structure [8].

The conducting layer deposition rate depends on experimental conditions. It was pointed out [4] that under irradiation in an applied electric field, the deposition of ionized molecules and radicals on the surface accelerates. The hydrocarbon dissociation on the oxide material surface is noticeable at the temperature higher than 500 K. At higher temperature the oxygen mobility in oxides increases, accelerating the oxidation of hydrocarbons and amorphous carbon. These reactions determine the upper temperature limit

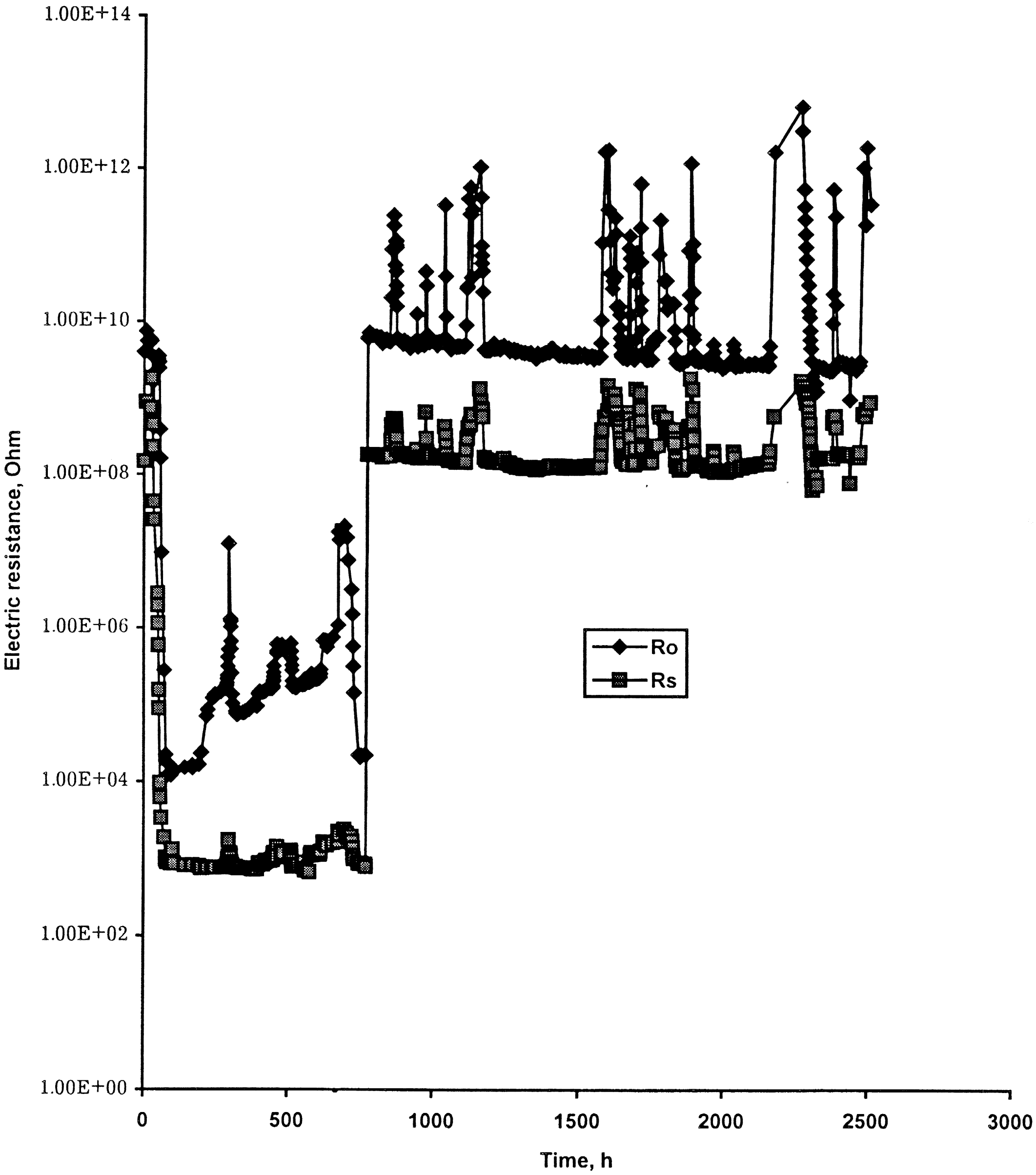

Fig. 1. Electrical resistance of Al_2O_3 single crystal specimen under reactor irradiation at 850–900 K versus irradiation time [8]. R_0 and R_s are the volume and surface resistance, respectively.

of conducting layer existence on the surface. The conditions for RIED observation correspond to conditions necessary for the formation of a conducting surface layer due to hydrocarbon deposition.

3. Radiation-induced charging and electric break-down in dielectrics

Radiation-induced dielectric break-down is necessary to consider using ceramics under prolonged irradiation. The radiation-induced dielectric break-down has been shown to occur in alumina. The alumina plates were exposed to electron irradiation from a Sr–Y-source in air at room temperature without applying the electric field. The dose of electrons with the energy exceeding 1 MeV was 3×10^{15} e/m^2, flux was about 10^9 e/m^2s. During prolonged irradiation (34 days $\sim 3 \times 10^6$ s) dielectric break-down occurred. Visible craters had appeared on the surface of the samples. The depth of the break-down region is equal to 0.8 mm.

According to the study on radiation-induced dielectric break-down in several dielectric materials [18], the observed break-down traces are evidence that the motion of electron avalanche occurred from the bulk to the surface of the materials under both electron and proton irradiation. Hence, accumulation of charged primary particles in the bulk of dielectrics was not responsible for charging, probably, due to RIC. However, irradiation causes charge partitioning by means of electron–hole pair formation with the efficiency of about 10^5 pairs per 1 MeV of primary particle energy (the precise value depends on the dielectric and primary particle). Therefore, it can be proposed that the electric charging and break-down are caused by charge partitioning and subsequent local charge accumulation in dielectrics under irradiation rather than primary particle accumulation.

The electric charging is responsible for certain features of RIC and RIL in dielectrics during irradiation and it is caused by the specific mechanism of non-equilibrium charge carrier relaxation predominating up to a dose rate of 10^{12} Gy/s, namely, recombination on electron and hole traps [19]. According to this mechanism, RIL intensity is proportional to the rate of electron trapping.

RIL related to F- and F^+-centres in Al_2O_3 is caused by charge carrier trapping and subsequent re-charging of intrinsic oxygen vacancies [19]. The concentration of oxygen vacancies arising under irradiation is much smaller than the initial vacancy concentration before irradiation. One can conclude (Fig. 2, dose dependence of F^+-band) that the rate of electron trapping on oxygen vacancies in Al_2O_3 single crystals under 8 MeV proton irradiation (flux 1.5×10^{16} p/m^2s) is constant, which is in agreement with the re-charging kinetics in steady state under irradiation. However, the rate of electron trapping on F^+-centres (Fig. 2, dose dependence of F-band) slowly increases with the dose, which is caused by increase of F^+-centre concentration. The increase of F^+-centre concentration is a response of negative charging in the microregions of the bulk of material due to charge partitioning in proton tracks. The density of negative charge excess has been estimated to be about of 10^4–10^6 Q/m^3. Taking into account this charge excess and the projected range of protons (0.2 mm), the size of track vicinity where this charge excess arises, is about 10–100 nm. This evaluation is in agreement with the size of track vicinity (> 10 nm), where trap re-charging occurs during 8 MeV proton irradiation of Al_2O_3 single crystals [19].

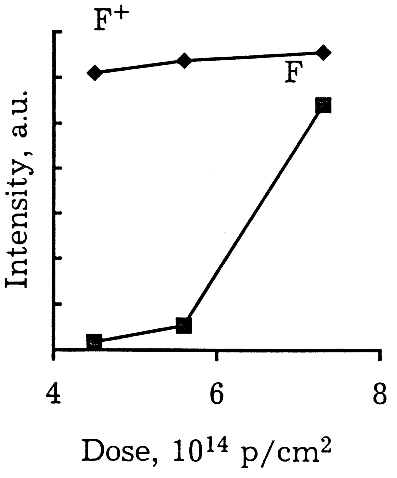

Fig. 2. Dose-dependent intensities of F^+- and F-centre bands (330 and 415 nm, respectively) of RIL of Al_2O_3 single crystal irradiated with 8 MeV proton at a flux of 10^{16} p/m^2 s.

Charge partitioning in tracks and trap re-charging cause kinetic charging of the regions with structural irregularities (grain boundaries, clusters, stacking faults, inclusions) owing to increase of the local trap concentration $N(x)$ in these regions and the difference in charge carrier mobility. The charge density can be written as

$$Q(x) \sim \int_{-\infty}^{\infty} \frac{\partial^2 N(x-u)}{\partial x^2} e^{-c|u|} \, du, \tag{2}$$

where the magnitude of c depends on material and temperature. Radiation-induced kinetic charging on irregularities is an efficient mechanism of the initial charge accumulation causing electric break-down in dielectrics under irradiation.

Also, the increase in the intensity of the RIL band of Cr^{3+} in ceramic Al_2O_3 with the dose is caused by an excess of positive charge in the grain bulk and negative charging at the grain boundaries. On increasing the dose of 8 MeV proton irradiation (flux 3×10^{16} p/m^2s) the RIL intensity of Cr^{3+}-band (690 nm) in ceramic Al_2O_3:Cr^{3+} has been observed to increase. The increase is equal to 11% at the end of irradiation, the intensity of the similar band in single crystal Al_2O_3:Cr^{3+} being constant.

The difference in the dose dependence of the Cr^{3+}-band in these materials is due to different contributions of luminescence of structural constituents, subsurface regions and the bulk of the materials. In optically transparent single crystals the main contribution is the luminescence in the bulk. In ceramics the main contribution is the luminescence of subsurface regions (layer thickness similar to grain size of 1–10 μm). Moreover, chromium ions on grain boundaries are inactive in the corresponding spectral range (690 nm) owing to changing the position symmetry. The elementary process of luminescence occurs after trapping an electron by a Cr^{4+} ion

$$e + Cr^{4+} = (Cr^{3+})_{\text{excited}} = Cr^{3+} + h\nu.$$

Cr^{4+} ion formation is controlled by hole trapping on Cr^{3+} ions, which is recompensed by Cr^{2+} ion formation during electron trapping on Cr^{3+} ions. The positive charged regions (especially, bulk of grains) giving contributions in the RIL of ceramics result in the increase of a Cr^{3+}-band owing to an enhancement of Cr^{4+} ion concentration. The magnitude of charge density does not exceed 10^2 Q/m^3, because the associated electric field is less than the electric break-down strength (10^8 V/m). The constant intensity of the Cr^{3+}-band of the RIL in single crystal Al_2O_3:Cr^{3+} indicates general electric neutrality of the bulk of the material.

4. Conclusion

High values of conductivity reported in several papers and ascribed to RIC (corresponding to the magnitude of ionizing efficiency exceeding 10^{-10} s/Gy Ω m) or RIED cannot be explained within the framework of a reasonable physical model. Owing to the re-charging of the initial charge carrier traps the RIC is not so high to restrict the application of dielectrics in fusion reactors. The phenomenon of ‘bulk RIED’ is probably caused by surface contamination of dielectric materials during irradiation testing.

More important problems are connected with electric charging of dielectrics under irradiation and subsequent dielectric break-down. The radiation-induced dielectric break-down has been shown to occur in alumina without applying the electric field. The electric charging as well as features of RIC and RIL in dielectrics are caused by the common mechanism of non-equilibrium charge carrier relaxation, namely, recombination on electron and hole traps.

The presented results on RIL in Al_2O_3 based materials indicate local charge accumulation in the bulk of the materials caused by charge partitioning under irradiation. Electric charging and break-down may be caused by charge partitioning in dielectrics under irradiation rather than charge deposition by primary particle accumulation.

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