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DEFECT FORMATION IN POLYCRYSTALLINE α-Al2O3 UNDER EXPOSURE TO NEUTRAL AND CHARGED PARTICLE BEAMS

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ABSTRACT

In this work, point and low-dimensional defects formed in the near-surface layer of single-crystal α-Al2O3 under the influence of high doses of 4 MeV electron irradiation were investigated. The experiments were carried out on the Elektronika U-003 accelerator in the dose range of 1014–1017 cm-2 with current pulses of 4.5·10-6 s duration and a beam density from 0.4 to 3 μA/cm2. Using scanning electron microscopy with local elemental analysis, the formation of stable fluorine and magnesium impurities in the irradiation region was established, which indicates radiation-stimulated transmutation processes in the crystal lattice of aluminum oxide. The obtained data are important for understanding the radiation resistance of aluminum oxide-based ceramics under extreme conditions, for example, as structural and insulating materials in nuclear power engineering and space technology.

KEYWORDS

Corundum, electron irradiation, surface defects, electron microscopy, IR spectroscopy.

Introduction

Corundum (polycrystalline) without metal impurities is used in nuclear energy, thermonuclear installations, and electronic engineering, as it has the densest hexagonal packing of oxygen anions and is the strongest, most refractory, and radiation-resistant dielectric material.

A nanostructure (200 nm scale) of the γ-Al_2O_3 phase formed on the surface of α-Al_2O_3 ceramics was discovered when exposed to an IR laser pulse of 10^{-3} s with a power density of 10^5 Vt/cm^2 [1]. The paper demonstrates the possibility of effectively filling deep centers in anion-defective aluminum oxide single crystals with a pulsed electron beam at room temperature, which is proven by observing a high-temperature TL peak at 750 K [2]. In [3], the influence of deep traps filled with a pulsed electron beam on the thermoluminescence (TL) of the dosimetric peak at 450 K in anion-defective aluminum oxide single crystals was investigated. It was suggested that in the temperature ranges of 600−750 and

900−1000 K, predominantly electron traps are emptied, while at T=780−900 and above 1000 K, hole traps are emptied.

In [4], the effect of thermal annealing on the optical absorption induced in Al_2O_3 single crystals during reactor irradiation was investigated. Irradiation with a dose of $1.5 \cdot 10^{21}$ n/cm^2 resulted in the appearance of background absorption in the wavelength range of 215 – 560 nm and optical absorption bands at 413, 256, 227, and 205 nm. During thermal annealing, changes in optical absorption occurred at temperatures from 350 to 900 K. The annealing onset temperature of the 413 nm band corresponded to the temperature at which the single crystals were maintained after reactor irradiation. Oxygen vacancies formed during neutron irradiation change their electrical charge under the influence of ionizing radiation, forming F^+ and F-centers, which are, respectively, singly positively charged and neutral oxygen vacancies. They are responsible for the aforementioned absorption bands at 205–256 nm, as well as photoluminescence at 413 nm (F-centers) and 328 nm (F^+-centers). It is known that the functional characteristics of Al_2O_3 are largely determined by the presence of impurity and intrinsic defects [5,6]. Impurity defects are represented predominantly by $3d^n$ elements, which are always present in Al_2O_3 regardless of the synthesis method [5,7]. Another type of defect, always present in Al_2O_3, are intrinsic defects in the form of anion vacancies in various charge states [6,8,9]. In this work, the VUV luminescence of unirradiated and electron-irradiated corundum single crystals with a dose of $6 \cdot 10^{17}$ el/cm^2 was studied using time-resolved spectroscopy with synchrotron radiation at low temperatures. It was shown that in irradiated corundum crystals, the excitation of a self-canceling exciton extends into the region of interband transitions. It was found that in irradiated corundum, the decay times of the 7.6 eV emission band are 7 ns and 24 ns [10].

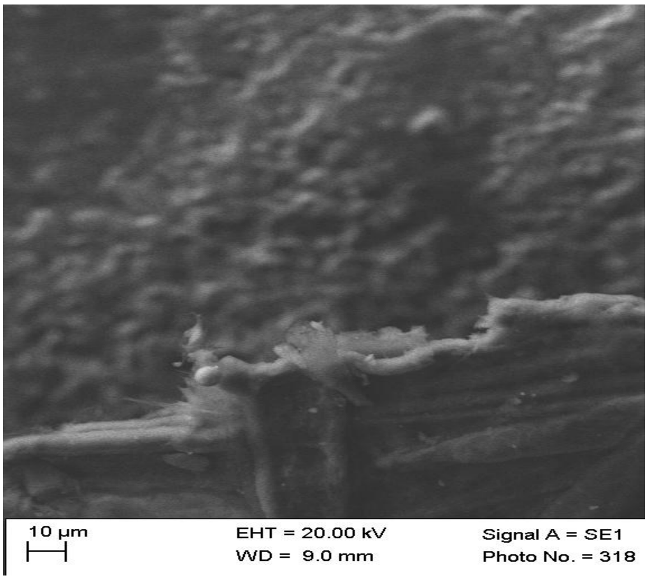
We have previously discovered a significant shortening of the duration and a decrease in the intensity of the afterglow and PL upon laser excitation (12 ns and 337 nm) of pure α-Al_2O_3 crystals after ^{60}Co γ-irradiation with doses from 10^4 to 10^7 Gr due to the predominant recombination of close electron-hole pairs [11]. Microscopic examination of the irradiated surface of α-Al_2O_3 is of scientific and practical interest.

In this work, point and low-dimensional defects formed in the surface layer of α-Al_2O_3 under the influence of high doses of irradiation with particles with energies above 1 MeV were investigated.

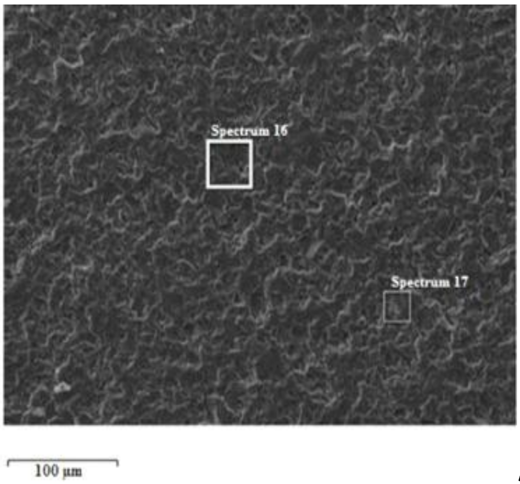
Study object and methods

Pure corundum microceramic samples 0.5 mm thick were used for the experiments. Irradiation with 4 MeV electrons was performed in air without cooling on an Elektronika U-003 accelerator, with doses of 10^{14}–10^{17} cm^{-2}, current pulses of $4.5 \cdot 10^{-6}$ seconds, and beam current densities from 0.4 to 3 μA/cm^{-2}. The surface microstructure was also irradiated with an Ekspla UV laser ($2.8 \cdot 10^{-11}$ s, 350 nm, 10 mJ). The microstructure of unirradiated and irradiated surfaces and the local elemental composition were studied using an EVO MA10 scanning electron microscope at different scales with an EDS (SDD) attachment. An electrically conductive graphene nanofilm was deposited on the dielectric surface using magnetron sputtering of a graphite cord in a vacuum. The vibration spectra of the Al-O bonds were studied on a Nicolet iS50 Fourier transform IR spectrometer in the 4000–400 cm^{-1} range in transmission and internal reflection modes, with IR spectra analyzed using databases.

Figures 1*a*, *b*, and *c* show images of the original and irradiated microcorundum surfaces at a current density of 1 μA/cm^2 with a dose of 13 MR, and Figure 2 *a*, *b*. A micrograph of the microcorundum surface after electron irradiation at a current density of 400 nA/cm^2 with a dose of 1.3 MR. The microstructure of the irradiated surface was then analyzed using a scanning electron microscope, and the local elemental composition was determined at different locations to determine deviations from the stoichiometry of the main elements of the corundum matrix, as well as the content of inclusions or even the appearance of possible impurities as a result of radiation-induced reactions on the surface.



a



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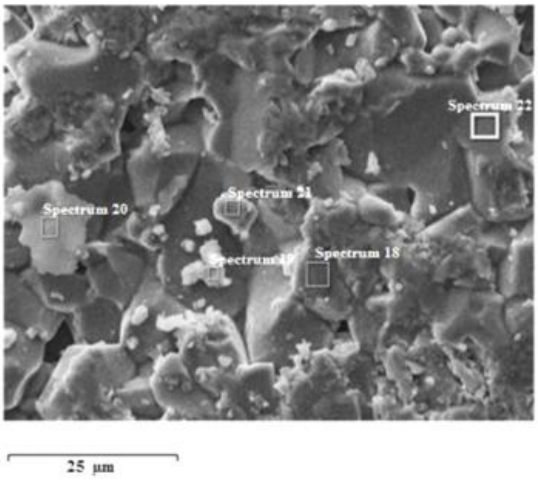

c

Fig. 1 *a*, *b*, *c*. Micrographs of the microcorundum surface after irradiation with 4 MeV electrons at a current density of 1 µA/cm^2 with a dose of 13 MR

In Figures 1 *a*, *b*, *c* and 2 *a*, *b* it is evident that in the indicated different areas of the surface where the characteristic spectra of X-ray radiation were recorded (for light elements this is mainly the K-series), excited by an electron beam with an energy of 100 keV to determine the local elemental composition: where 4 MeV electrons hit and far from where the beam hit.

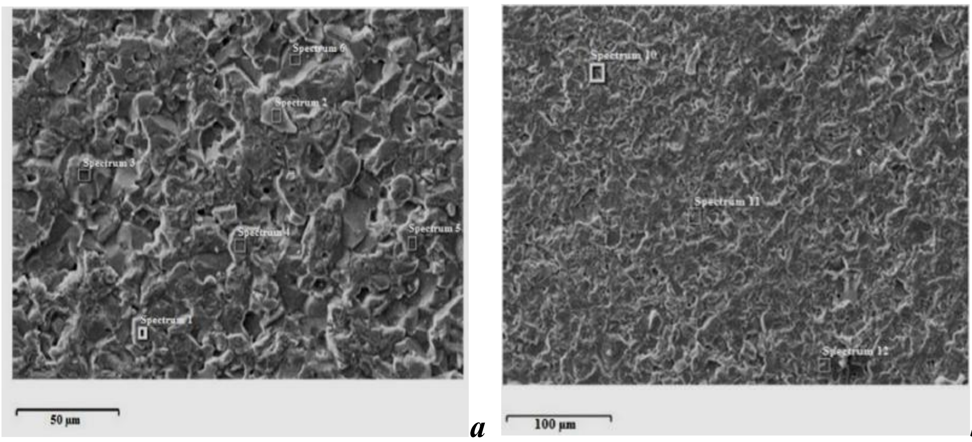

Fig. 2. *a*, *b*. Micrograph of the microcorundum surface after electron irradiation at a current density of 400 nA/cm^2 with a dose of 1.3 MR

The random detection of calcium, sodium and silicon indicates that these are surface inclusions (most likely particles of paper in which the samples were wrapped), so their contrast in Figs. 1 and 2 differs from the matrix and they are not associated with irradiation. It was assumed that impurities adsorbed as a result of radiation-induced reactions on the dielectric surface charged by electron irradiation would appear. However, this would lead to a systematic increase in nitrogen content in the beam field, forming aluminum nitride or oxygen. However, this was not observed experimentally because even with excess nitrogen in the atmosphere, it cannot replace oxygen in the oxide due to its lower chemical potential.

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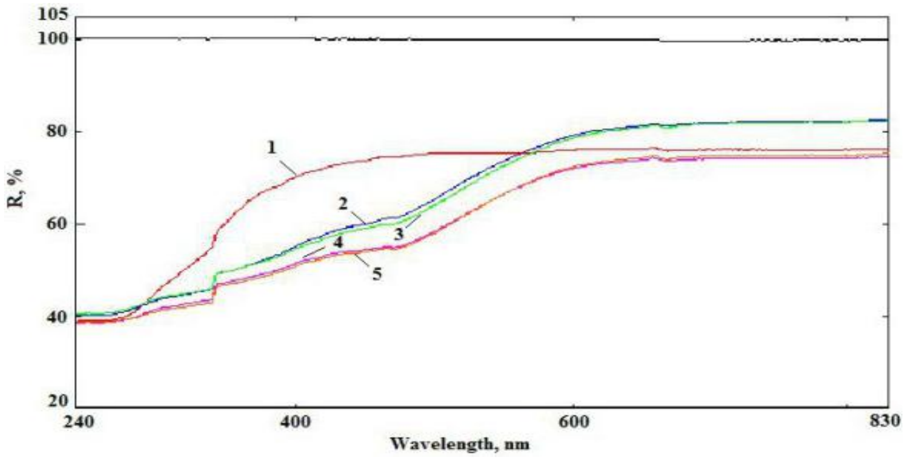
Fig. 3 shows the optical reflectance spectra of microcorundum before (curve 1) and after irradiation with 4 MeV electrons, taken from the exposed and back surfaces: (curves 2, 3) - at a current density of 3 μA/cm^2 with a dose of 10^{17} cm^{-2}, (curves 4, 5) - at a current density of 0.4 μA/cm^2 with a dose of 10^{15} cm^{-2}; a jump at 340 nm due to the change of lamps.



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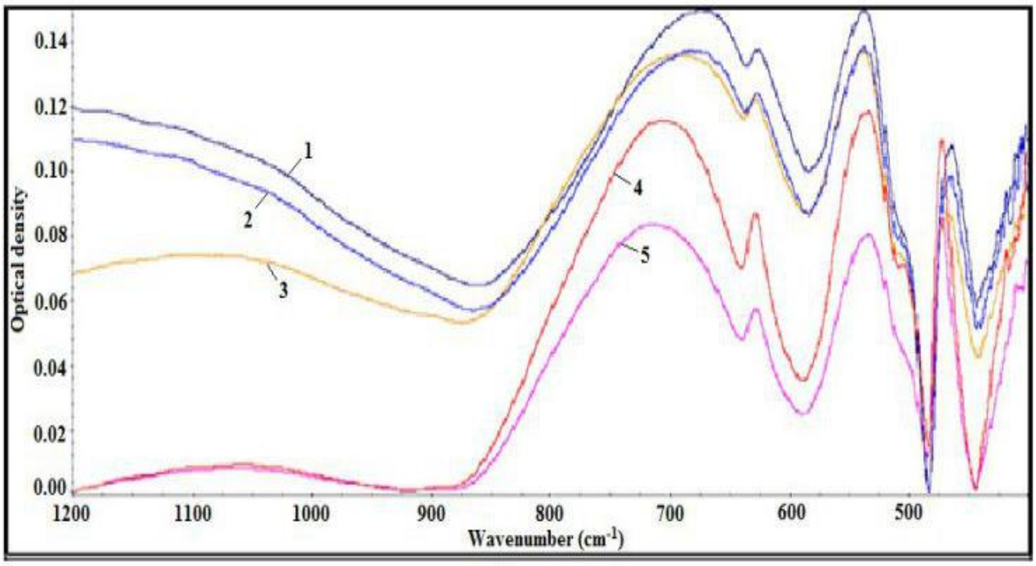
It is evident from the figure (Fig. 3, curve 1) that in unirradiated microcorundum ceramics, the reflectance in the visible region is at the level of 75%, and at the absorption edge it decreases to 38%. After irradiation at 0.4 μA/cm^2 with a dose of 10^{15} cm^{-2} (Fig. 3, curves 4, 5), reflectivity decreases in the 300–600 nm range due to radiation absorption by oxygen-deficient color centers. However, after more intense irradiation at 3 μA/cm^2 (Fig. 3, curves 2, 3), with a 100- fold higher dose, an increase in reflectivity in the 550–830 nm range is observed above that in the unirradiated sample.

In Fig. 4 *a*, *b*, *c* vibrational IR spectra of microcorundum before (curve 1) and after intense irradiation with 4 MeV electrons at a current of 0.4 μA with doses from 10^{15} cm^{-2} to 10^{16} cm^{-2} (*a*), 1 μA with a dose of 10^{17} cm^{-2} (*b*) and 3 μA with doses of 10^{16} and 10^{17} cm^{-2} (*c*), taken from the exposed (curve 4.5) and back (curve 2.3) sides in reflection mode.



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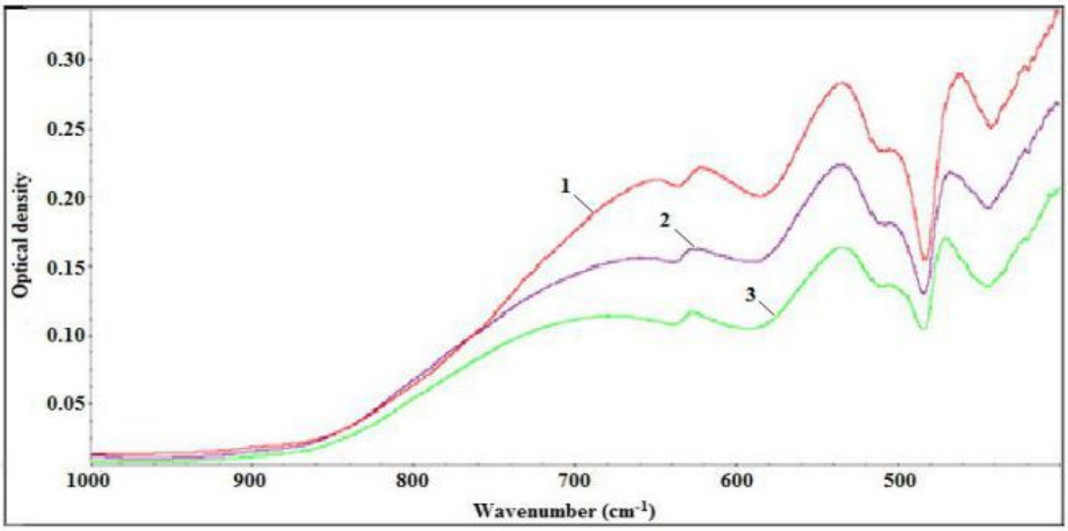

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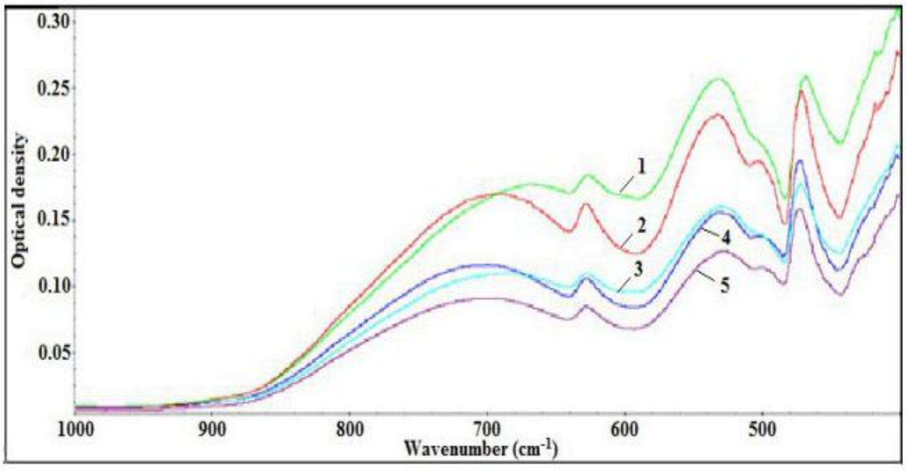

c

Fig. 4 *a*, *b*, c. Vibrational IR spectra of microcorundum before (curve 1) and after intense irradiation with 4 MeV electrons at a current of 0.4 μA with doses from 10^{15} cm^{-2} to 10^{16} cm^{-2} (*a*), 1 μA with a dose of 10^{17} cm^{-2} (*b*) and 3 μA with doses of 10^{16} and 10^{17} cm^{-2} (*c*), taken from the exposed (curve 4.5) and back (curve 2. 3) sides in reflection mode

It can be seen that the optical lamp of the exposed beam is located lower than that of the back and unexposed beams. The tensile and bending vibrations of the O-Al-O chain after electron irradiation are weakened and are maximal in the short-wavelength region, indicating the accumulation of deformations in the corundum crystal lattice. Spectrum 18 in Fig. 1 *c* and spectrum 3 in Fig. 2 *a* are similar in many ways: an increase in fluorine content correlates with an increase in the absorbed dose, while the oxygen content drops significantly and the aluminum content increases. This is consistent with the IR spectra in Fig. 3, where the O-Al-O bond vibration bands are significantly weakened due to a decrease in the oxygen content.

In [1], based on the appearance of IR absorption at 900–950 cm^{-1}, it was established that, during surface melting under the influence of laser and electron radiation, some Al^{3+} ions are converted from oxygen octahedra to tetrahedrons.

Figure 5 shows the current flowing in the near-surface region of irradiated Al_2O_3 ceramic samples with a carbon layer deposited on them. Electron irradiation with a dose of 1 – 10^{15} cm^{-2}, 2 – 10^{16} cm^{-2}, 3 – 10^{17} cm^{-2}.

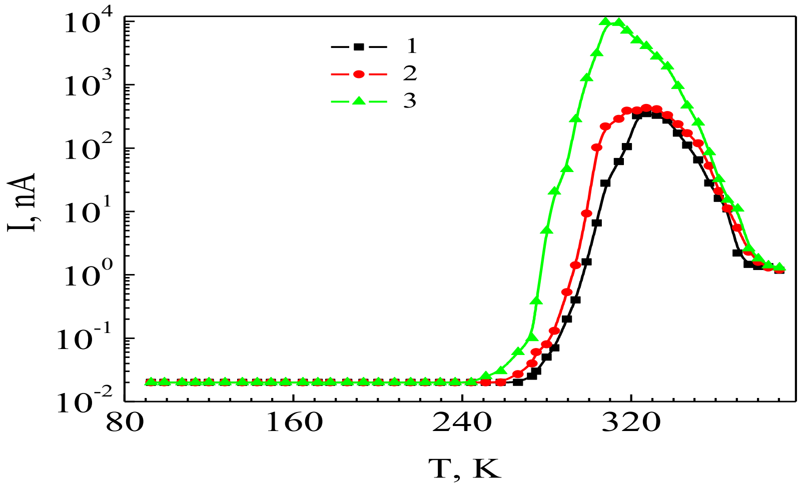

Fig. 5. Current flowing in the near-surface region of irradiated Al_2O_3 ceramic samples coated with a carbon layer. Electron irradiation with doses of 1 – 10^{15} cm^{-2}, 2 – 10^{16} cm^{-2}, and 3 – 10^{17} cm^{-2}

Electron irradiation at a current of 406 nA, 450 nA and 2880 nA, respectively. The appearance of the current peak is most likely due to the fact that, according to Surdo [12], the highest efficiency of exciton excitation, and consequently the generation of excitons bound to them, is observed at T = 295 K, i.e. close to the maximum temperature of the peaks observed in our experiment (337 K; 325 K and 313 K, respectively, fluences), and such orientations at which excited levels of F^+- and F-centers are present at the bottom of the conduction band.

Conclusions

Thus, exposure to high doses of irradiation with particles with energies exceeding 1 MeV leads to significant structural and chemical changes in the near-surface aluminum oxide layer, including microstructural modification and deviation from stoichiometry. The Al-O-Al absorption band at 1600 cm^{-1} is initially greatly weakened, then disappears after maximum fluence, replaced by another band at 1530 cm^{-1}. The absorption profile becomes characteristic of scattering by quasi-free carriers, as in p-metals. Apparently, the concentration of charged oxygen vacancies in the near-surface layer becomes so high that nanoclusters of metallic aluminum, as well as another crystalline phase of aluminum oxide, are formed. Scanning electron microscopy studies combined with local elemental analysis (EDS) demonstrated significant changes in the elemental composition of the aluminum oxide (Al_2O_3) surface layer under the influence of an intense 4 MeV electron beam. This change also reduced the oxygen content, as determined independently by two spectral methods: IR and EDS.

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