Influence of High Dose Reactor Irradiation on Some Parameters of Al_2O_3 Crystals

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Abstract – The characteristic features of the process of radiation-defect formation and change in the structural, optical and electrical properties of Al_2O_3 crystals exposed to gamma and reactor radiation have been investigated by some spectroscopic methods. The dose dependence's of the generation of color and luminescence centers, change of the electrical and structural parameters and this energies activation of a crystals was determined. The possible mechanism of damage structure of a samples irradiated in a reactor is discussed.

1. Introduction

Al_2O_3 oxide is one of the prospective high-k electroinsulators and construction oxides materials, in particular, for the ceramical fuel material and for a first wall thermonuclear arrangement. Besides, this oxide is used widely as an active element or substrate at creation of laser and MOS systems, as a film coating and receiver of IR-radiation [1–7]. From here this work is the aim at investigation of radiation stability of the physical properties and a structure of Al_2O_3 crystalline oxide after irradiated in the reactor of a high fluence and cource Co^{60}.

2. Results and discussion

In this connection the peculiarities of radiation effect on lattice parameters (a,c), form and position of several reflections, reflection coefficient and frequency of valence and deformation oscillations of Al-O bonds, electric characteristics of the oxide were studied with techniques of X-diffraction, dielectric, optic and IR-reflection spectroscopy.

The fracture our maximum on the dose curves Y(F) of intensity photoluminescence (FL) at 330, 390 and 510 nm, colour centers 205–460 nm and 570 nm in the range of identical doses was determined at the analysis of the obtained results. Fig. 1 presents the temperature-dose dependence of the intensity of the 330 nm band in FL spectrum of crystals. As the T=600–700^{0}C of intensity this band increasing, this can be used in creating tunable lasers [4].

Table 1. Temperarure dependence (I/I_o, arb. un.) at t (s) of band 510 nm

	T_i, ^oC (at $F=5\cdot10^{18}$ cm^{-2})			
t, s	150	400	800	900
120	0.98	0.84	0.98	0.99
360	0.93	0.70	0.96	0.94
600	0.93	0.60	0.95	0.94

A similar kinetics of a process of decoloration of 510 nm FL band was established when temperature was varied. The basic obtained results here are listed in Table 1. It appeared that also the temperature dependence of photodecoloration of this band has an extremum character, because the maximum rate of the process is attained at 600^{0}C. A subsequent in the temperature leads to its retardation, which can be seen up to T>900^{0}C on a change in the time UV illumination of plates from 0 to 600 sec. The $I/I_o(\Phi, T, t)$ graphs obtained can be approximated by summary two exponential dependence's with the constant of decay i = 0.01–0.10, which is a function of temperature and dose.

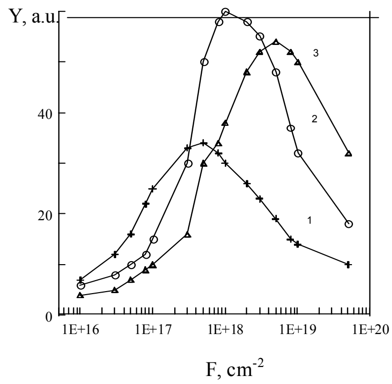

Fig. 1. Dose dependence Y(F) for bans 330 nm at T=300–1, 600–2, 700^{0}C-3

For example, in present communication it is submitted the results of X-ray structure investigation of after irradiation by high neutron doses (F). X-diffraction patterns were obtained using a roentgen diffractometer. Particular attention was given to dynamics of form and positions some representative reflexes with hkl = 014, 110, 03.12 and ets. The diffraction patterns were found essential changes of peak intensity and position depending upon F, the Bragg angle 2Θ decreases with an increase in dose, the regularity is shown if $K\alpha_1$ and $K\alpha_2$ doublet is splintered. For example, in Figure 2 was given to radiation dynamics of the function 2Θ (F) for this doublet ($K\alpha_1$ and $K\alpha_2$) in the reflexes 03.12.

From Fig. 2 is shown that the displacement of this peak $\Delta(2\Theta)$ = 40–50^0 at dose $F=10^{20}$–10^{21} cm^{-2} (for the reflexes 02.10 – in Table 1). At high doses the reflexes at $2\Theta > 70^0$ are eroded and weakened and doublets are not splitting, the lattice parameters of crystals (a, c, d/n) was undergoes anisotropy expansion (Table 2), at very high dose of change this parameters is composed not so many as Δc= 0.0038 nm and Δa = 0.0014 nm.

Table 2. Influence of neutrons on some parameters of crystals

F, cm^{-2}	$1\cdot10^{17}$	$1\cdot10^{18}$	$5\cdot10^{18}$	$8\cdot10^{18}$
D_1, a.u.	0.27	0.62	1.29	1.29
D_2, a.u.	0.12	0.35	0.51	0.64
F, cm^{-2}	1.10^{19}	8.10^{19}	1.10^{20}	5.10^{20}
D_1, a.u.	1.27	1.70	1.95	2.15
D_2, a.u.	0.93	1.00	1.21	1.99
c, nm	1.298	1.299	1.300	1.301
d/n, nm	0.2379	0.2381	0.2383	0.2386
$(2\Theta)^o$	89.03	88.98	88.95	88.86

Besides, it is shown that at this doses some halo appears at the diffraction patterns. In Table 2 was given dose dependence of generation in the absorption spectra a bands 257 and 358 nm (D_1, D_2, F- and F-aggregate defects). The mechanism of atom displacement out of lattice knots plays the main role in radiation damage of irradiated of high doses crystals.

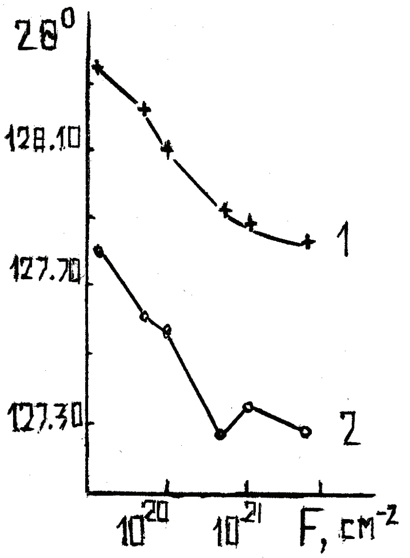

Fig. 2. Dose dependence of 2Θ (F) for doublet$K\alpha_1$ (1) and $K\alpha_2$(2) of reflexes (03.12)

The optical characteristics – reflection coefficient and frequency of valence (736, 614 cm^{-1})and deformation (464 cm^{-1}) oscillations of Al-O bonds of the oxide were studied with techniques of IR-reflection spectroscopy in region 400–1200 cm^{-1}. The partify decrease of intensity R and frequency ν of this modes at fluence $8\cdot10^{19}$ cm^{-2} was observed. The change of these parameters were found near a neutron fluence $(6–8)\cdot10^{19}$ cm^{-2}. ome change of these optical (a valence and deformation oscillations of Al-O bonds) and structural parameters (the lattice parameters of crystals (a, c, d/n) and positions some representative reflexes, of peak intensity and the Bragg angle), a density and a linear size of irradiated samples were found near a neutron fluence 10^{20} cm^{-2} and 10^{21} cm^{-2}, but the structure state remained stable at the following increase of the irradiation dose (at F= $1\cdot10^{21}$ cm^{-2} and $7\cdot10^{21}$ cm^{-2} and higher).

Features of dose and temperature dependencies of electric properties (σ) were determined after influence of different doses of ionizing radiation. Increase of the dielectric permittivity was found near temperatures (25–400 °C) and high doses. Besides, at a gamma-dose of 10^3–10^5 Gy the electric conductivity decreased. The opportunity of describing dependence σ(T,D) has been showed in irradiated Al_2O_3 oxide through the analytic equation, consisting of the exponential and power functions with corresponding coefficients. The significance of those coefficients in the temperature range where the σ (irradiation stimulated) is prevailing, was calculated. Their satisfied consenting with the significance of coefficients in aluminum ceramics UF-46 were determined. It is clear that the discovering result is very interesting, because of its certifications that the σ in reactor irradiated ceramics was determined mostly by the gamma-irradiation, influencing on the crystalophase of ceramics.

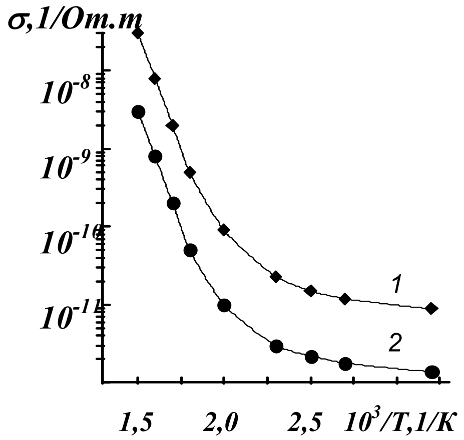

Fig. 3. The dependence σ(T) of samples before (1) and after(2) irradiation

Table 3. Activation energy for the electrical conductivity of corundum

D, Gy	T,°C	E_1, eV	T,°C	E_2, eV
O	< 150	0.29	>150	0.68
10^4	<150	0.20	>150	0.78

Irradiation to dose D=10^4 Cy leads to σ decrease, however the shapes of the σ_1(T) dependencies for samples N 1 and N 2 type (cut parallel and normal on the main optical axis) after gamma-irradiated are alike (Fig. 3). Activation energies (E) of conductivity for the low- and high-temperature regions were calculated for a samples – E_1 and E_2 accordingly (Table 3).

Experimental dependence of σ(T, D) for single crystals of Al_2O_3 oxide at T<150°C and T>150°C gamma-irradiation practical not influence on the activation energy. This indicates on retain at the irradiation of character conductivity of the crystals. What is concern of some increasing E_2 in irradiated samples (in the high temperature region) we can say, that in case of $T>T_b$ on low-temperature conductivity is laying the σ which connected with motion, for example, oxygen ions by the lattice vacancies, what is good according to Dince about the conductivity mechanism at more than room temperature. The proximity of gained results for activation energy of conductivity gamma-irradiated crystals E_2 to such kind of activation energy of the reactor irradiated UF-46 ceramics are indicated the additional fact, which certifying the most contribution to conductivity of crystalophase.

The dose dependencies of the conductivity in this plates was found to be nonmonotonic. For example, on the curve σ(D) in a samples № 1 and № 2 at the $D=10^5$ Gy of the minimum was marked. These temperature-dose kinetics of the dependence σ(T, D) for the temperatures 25–100°C were established. So, in the first time were established, that the σ(D) curve has the extremums, because of near the 10^{5} Gy dose it passing though the minimum. It is interesting, that near the doses $D\geq10^6$ Gy initial conductivity behaviour of samples is restoring. The problems connected with the possible of causes were established of the peculiarities of kinetics in the existence models are discussed on the base of these electrical characteristics analysis.

3. Conclusions

Thus, such thermal-radiation treatment may serve as a method leading to the permittivity growth at elevated irradiation doses and temperatures, decrease and improvement of several electric, mechanic and optic characteristics. This testifies to prospect of application of this high-k oxide (Al_2O_3) in a composition at other oxides metals in the quality construction, insulation and fuel materials in the reactor, cosmic and semiconductor technique, under the condition of irradiation influence. A conclusion about the radiation-induced modification of the physical properties of Al_2O_3 crystals and about the action of large fluences neutron on the order of a structure, a parameters of lattice and the formation of region of disordering at the places of accumulation of the radiation defects as a result of implementation of the mechanism of displacement of atoms in the crystal lattice of a dielectric has been made.

References

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