STUDY OF OPTICAL PROPERTIES OF IRRADIATED AlN CERAMIC

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In the course of research, dependencies of defect formation processes, radiation resistance, changes in structural and optical characteristics of AlN ceramics under irradiation with 40 keV He^{2+} ions were established. Evaluation of changes in ceramics properties was carried out using the methods of thermally stimulated luminescence, optical absorption spectrophotometry and X-ray structural analysis. It was found that for irradiated samples, a decrease in optical absorption density spectra is observed with a sharp increase in dichroism, which is caused by an increase in concentration of implanted He ions in the structure, a change in the interplanar distances, and the formation of regions of disorder as a result of irradiation, leading to a strong distortion of the crystal structure. The change in structural characteristics due to the occurrence of defects and the introduction of helium ions into the lattice leads to a change in ceramics density, with the subsequent formation of porous inclusions in the structure, which significantly affects the optical properties of ceramics.

Keywords: radiation defects; swelling; blisters; ceramics; structural materials.

Introduction

Today, one of the important problems of nuclear energy and radiation materials science is the development of new structural materials with high radiation resistance to ionizing radiation [1]. As a rule, in the reactor industry, STM steels alloyed with various additives of nickel, chromium, which have good operational and heat-conducting characteristics are used. However, the high probability of swelling of STM steels as a result of prolonged use is a limiting factor for increasing the burnout of nuclear fuel and increasing the capacity of nuclear facilities [2-4]. The main difficulties in increasing the duration of nuclear installation campaigns are related to the swelling and embrittlement of structural materials. One way to solve this problem is to use new classes of various ceramics based on nitrides (AlN, Si_3N_4, GaN), oxides (Al_2O_3, ZrO_2, SiO_2), carbides (SiC, WC) with high electrical resistivity, thermal conductivity, high melting point, excellent insulating properties. Due to these properties, ceramic construction materials have found their application in nuclear power, optics, microelectronics as heat absorbers or the basis for microelectronic boards. In turn, nitride materials, in particular AlN, are one of the most promising candidate materials for shells and covers of the first wall of fission and fusion reactors. The potential of their use is due to the low vacancy swelling and creep, a small amount of induced activity, a high rate of radiation resistance to various types of external influences [5]. Also, this class of materials is able to withstand temperatures above 1200-1400°C, which makes them promising materials for GenIV reactors.

During irradiation, complex processes of defect formation occur in the crystal structure of the material, as a result of which various radiation irregularities, vacancies, and displaced atoms appear. In addition, a certain amount of products of nuclear reactions, such as helium and hydrogen, can accumulate in the structure, which can later cause a sharp reduction in the service life. When irradiated at low and room temperatures, as is known, helium can accumulate in the structure as a solid interstitial solution, followed by the formation of helium bubbles. The accumulation of helium in the structure is due to its low solubility and its ability to agglomerate into larger spherical clusters. In addition to the accumulation of helium in the structure during irradiation, another mechanism for the introduction of helium is its use as a coolant or aggregate of gas gaps in fuel rods.

In connection with this, it is particularly interesting to study the effect of helium on defect formation processes, as well as to reveal the evolution mechanisms of micro- and macroscopic properties of structural materials based on nitride ceramics.

Experimental part

AlN samples were irradiated at the DC-60 heavy ion accelerator of the Astana branch of the Nuclear Physics Institute with 40 keV ions of He^{2+} with fluence from 10^{15} to $5x10^{17}$ ions/cm^2 at the irradiation temperature of 300 K. The samples are polycrystalline structures with a hexagonal-type lattice, similar to the structure of wurtzite, with a concentration of impurity Al_2O_3 inclusions of not more than 4%. The thickness of the samples was 15 µm. Before irradiation, the samples were polished to obtain a surface roughness of not more than 3–4 nm.

TSL measurements were performed when samples were excited at room temperature for ten minutes with an ultraviolet deuterium lamp (50 W). After excitation, the removal of TSL spectra was carried out with linear heating of samples in the range of 300–673 K using an optical multichannel analyzer in the range of 300–800 nm.

Results and discussion

Thermally stimulated luminescence and optical absorption spectrophotometry were used to assess the effect of temperature and fluence of irradiation on the optical properties of ceramics. Figure 1 shows the evolution dynamics of the TSL emission spectra of the main peak at 400 K, depending from irradiation conditions. The position of the spectral lines was determined using the Gaussian decomposition.

According to the data obtained, the observed spectral line in the region of 3.2 eV belongs to the main recombination centers in AlN. The presence of small maxima at 1.8–2.0 eV is due to the presence of an insignificant amount of Al_2O_3 impurities in the structure. With small fluences of irradiation, the change in intensity of the spectral line is insignificant, which may be due to the occurrence of single defects in the structure and minor distortions in the structure. An increase in the irradiation fluence leads to a sharp decrease in

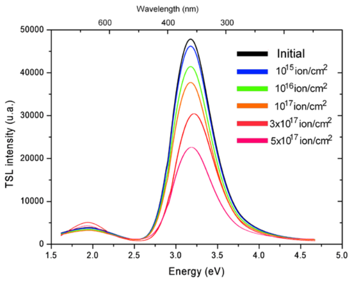

Fig. 1. Spectral distribution of the main TSL peak (400 K) depending on irradiation fluence

intensity of the spectral line and a slight shift of the maximum. An increase in concentration of embedded helium in the structure can lead to the replacement of Al and N atoms in the crystal lattice and a subsequent change in the concentration of absorbing centers and an increase in defects in the structure. Figure 2 shows the dependence of the change in the polarized absorption spectrum on the irradiation conditions. The spectrum was filmed in the range of 6–8 eV. According to the data obtained, dichroism is observed in the absorption spectra of samples under study in the range of 7–8 eV, which is characteristic of anisotropic absorption bands in the structure of ceramics. For irradiated samples, a decrease in the optical absorption density spectra with a sharp increase in dichroism is observed. A decrease in the absorption spectra may be due to an increase in the concentration of implanted He ions in the structure, a change in the interplanar distances, and the formation of regions of disorder as a result of irradiation, leading to a strong distortion of the crystal structure.

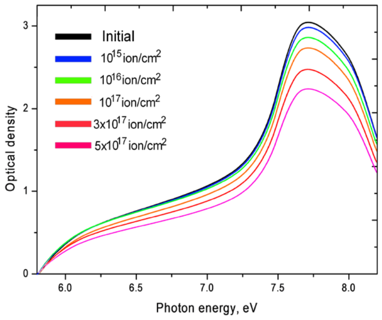

Fig. 2. Polarized absorption spectra of AlN ceramics depending on irradiation conditions

According to the data obtained, an increase in the irradiation fluence and, therefore, an increase in the concentration of implanted helium in the structure leads to an increase in the absorption value. An increase in the magnitude of absorption at large fluences indicates the predominance of defect formation processes in the introduction of helium ions with their subsequent migration and the creation of complexes of defects over the processes of recombination and annihilation of defects in the structure. Figure 3 shows a plot of the change in the helium concentration in the structure as a function of the radiation fluence. As can be seen from the presented data, with small irradiation fluences, the helium content in the structure is insignificant, however, an increase in fluence to 10^{17} and higher results in a sharp increase in the helium concentration. Increasing the concentration leads to the formation of large agglomerations of helium in the structure, and the subsequent formation of helium bubbles, leading to a change in the structural characteristics.

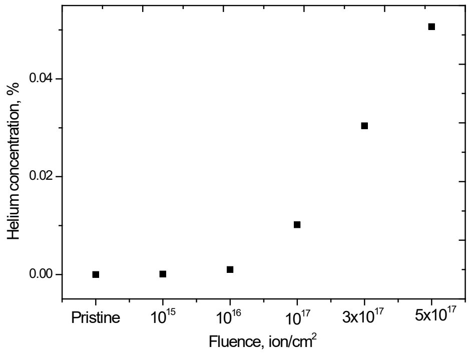

Fig. 3. Dynamics of changes in the concentration of embedded helium in the structure during irradiation

According to the X-ray diffraction data, the initial samples are AlN polycrystalline structure with a hexagonal lattice type, similar to the wurtzite structure. In this structure, there are three selected texture directions (100), (002) and (101), along which the crystallites are oriented. The presence of a low concentration of impurity Al2O3 inclusions (no more than 4%) in the structure of ceramics is due to the technological processes of obtaining ceramics from Al2O3 powder during sintering. According to earlier studies of the structural characteristics, the distribution of impurity inclusions in the structure is equally probable by volume [5]. As can be seen from the presented data, an increase in the irradiation fluence leads to a decrease in intensities of diffraction peaks and a change in lines shape, which indicates the appearance of additional distortions and stresses in the crystal structure, as well as a change in the crystallite size during irradiation. With an increase in the irradiation fluence (above 10^{17} ions/cm^2), on the diffraction patterns, the formation of a halo is observed, which is characteristic of X-ray amorphous or strongly disordered impurity inclusions. The determination of these inclusions was carried out using the Rietveld method. The formation of these inclusions may be due to the introduction of He ions with the formation of complex compounds with vacancies and impurity atoms, which subsequently leads to the rupture of chemical and crystalline bonds, as well as the degradation of the structure as a result of a large concentration of interplanar spacing.

Conclusions

The paper presents the results of changes in structural and optical properties of nitride ceramics under irradiation with low-energy helium ions. It was found that for the irradiated samples, a decrease in the optical absorption density spectra is observed with a sharp increase in dichroism. That is caused by an increase in the concentration of implanted He ions in the structure, a change in interplanar distances, and the formation of regions of disorder as a result of irradiation, leading to a strong distortion of the crystal structure. Using the method of X-ray structural analysis, it was established that an increase in the irradiation fluence leads to a decrease in intensities of diffraction peaks and a change in the lines shape, which indicated an increase in concentration of distortions and stresses in the crystal structure. It has been established that with an increase in the concentration of implanted ions and distortions of the structure, the density of ceramics drops sharply, which leads to an increase in the integral porosity.

References

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