

Study of structural changes in ZrO_2 ceramics irradiated with heavy ions of Kr^{15+} with an energy of 147 MeV.

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This paper presents the results of a study of changes in structural characteristics, such as the degree of crystallinity, dislocation density, and deformation of the crystal lattice in ZrO_2 ceramics as a result of irradiation with heavy Kr^{15+} ions with an energy of 147 MeV. The choice of the type of irradiation ions is due to the possibility of simulating the processes of defect formation comparable to the irradiation of radiation-resistant materials by fragments of uranium fission. The radiation doses ranged from 5×10^{13} to 1×10^{16} ion/cm². The dependences of the change in the crystallographic parameters on the radiation dose were established. At the same time, the maximum decrease for an irradiation dose of 1×10^{16} ion/cm² was no more than 10 %, which in turn indicates a high radiation resistance to the accumulation of defects during large-dose irradiation. It was determined that the greatest change in the degree of crystallinity is observed above a dose of 1×10^{15} ion/cm². The data obtained indicate a high degree of resistance of these ceramics to radiation damage.

Key words: heavy ions, ZrO_2 ceramics, radiation-resistant ceramics, new structural materials.

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1 Introduction

To date, the most promising materials for nuclear power as structural materials are oxide or nitride ceramics, which are highly resistant to degradation, refractory and wear resistance, and good insulating properties [1-5]. Among the variety of oxide types of ceramics, one can distinguish ceramics based on zirconium dioxide (ZrO_2), which are among the most refractory oxides ($T_{melt} = 2715^\circ C$). In nature, zirconium dioxide exists in three crystalline forms: in the form of a monoclinic structure characteristic of the mineral baddeleyite, a metastable tetragonal phase, and an unstable high-temperature cubic phase [5-15]. Resistance to most types of acids and alkalis, as well as high wear resistance and refractoriness make ceramics based on ZrO_2 the basis for the manufacture of refractory, abrasive materials, superhard glasses and fuel cells [16,17]. All of the above properties make ZrO_2 -based ceramics one of the candidate materials as the basis for the materials of the first wall of high-temperature reactors. However, in the case of potential application of ZrO_2 ceramics in this direction, it is necessary to know the degree of resistance of these ceramics to the action of ionizing radiation, as well as the effect

of the accumulation of structural defects in the material [18-20].

The aim of this work is to study the structural changes in ZrO_2 ceramics as a result of irradiation with heavy Kr^{15+} ions with an energy of 147 MeV.

2 Experimental technique

Commercial samples of ceramics based on zirconium dioxide with a tetragonal type of crystal lattice were selected as objects of research. The choice of these ceramics as an object of research is due to their high resistance to chemical and temperature degradation, as well as their increased resistance to radiation damage [21-25].

The irradiation of ceramics was carried out on a DC-60 heavy ion accelerator. The ions for irradiation were Kr^{15+} ions with an energy of 147 MeV. The radiation doses ranged from 5×10^{13} to 1×10^{16} ion/cm². The choice of the type of irradiation ions is due to the possibility of simulating defect formation processes in the range of particle energies comparable to irradiation with uranium fission fragments in a reactor. The choice of the range of irradiation doses is due to the possibility of simulating defect formation processes both in the case of the begin-

ning of the overlapping of ion trajectories in the material, and in the case when the probability of overlap exceeds 1000.

The study of the effect of irradiation on structural distortions and amorphization as a result of the accumulation of defects in the irradiated material was carried out using the standard method of X-ray diffraction, which belongs to the methods of non-destructive testing of defects. The studies were carried out using X-ray diffraction of the samples in the angular range $2\theta=20-100^\circ$, with a step of 0.03° . X-ray diffraction patterns were recorded on a D8 Advance ECO powder X-ray diffractometer, Bruker, Germany. The assessment of changes in structural characteristics was carried out by determining changes in the shape and intensity of diffraction lines, as well as their distortion.

3 Discussion

Figure 1 shows the dynamics of changes in the X-ray diffraction patterns of the studied ceramics before and after irradiation. The general view of the diffraction patterns indicates a polycrystalline structure of ceramics with a high degree of structural ordering, which is evidenced by the symmetric shape of the diffraction lines in the diffraction pattern of the initial sample. The general view and angular position of the diffraction peaks are characteristic of the tetragonal

phase of zirconium oxide with the spatial symmetry P42/nmc (137), which is most common for this type of ceramics.

For irradiated samples, according to the given X-ray diffraction data, the appearance of new reflections was not detected, which indicates the absence of phase transformation processes or the formation of new phase inclusions as a result of the accumulation of defects in the structure.

The main changes in the diffraction patterns are reflected in the form of changes in the shape of the diffraction peaks, as well as their intensity. These changes indicate that the main structural changes in this case are associated with deformation and disordering of the crystal structure, changes in dislocation density, and the formation of highly disordered regions or amorphous-like inclusions in the structure. These changes are caused by the processes of defect formation, which arise as a result of elastic and inelastic collisions of incident ions with atoms of the crystal lattice, as well as the formation of displacement atoms and knocked out electrons. In this case, in the case of irradiation with high-energy ions with an energy of more than 100 MeV, the main contribution to the energy losses of incident ions is made by elastic collisions of ions with electron shells, as a result of which cascades of knocked-out electrons arise and, therefore, the electron density in the irradiated material changes.

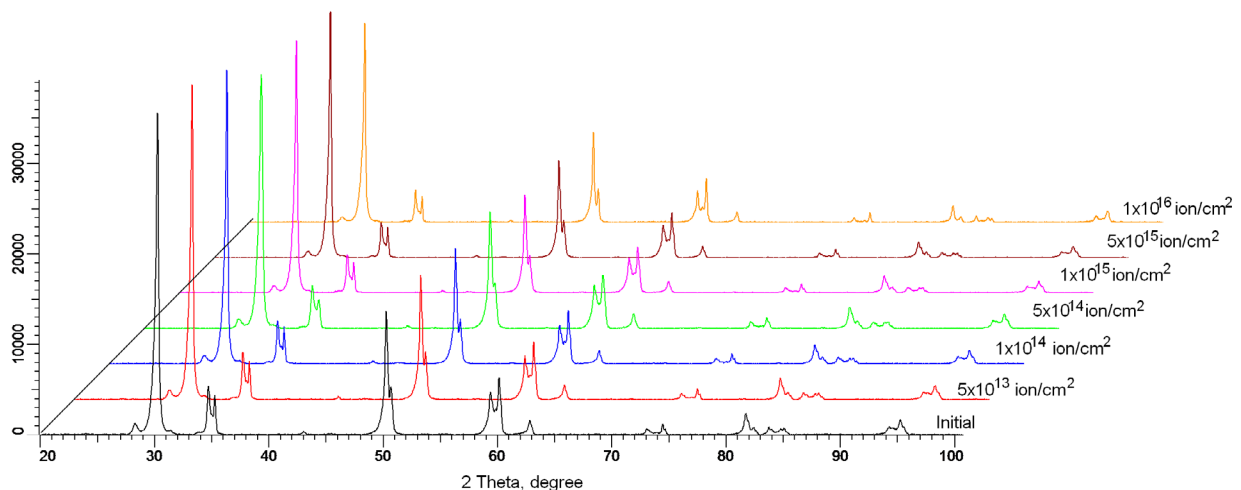


Figure 1 – X-ray diffraction patterns of ceramics before and after irradiation

Analyzing the obtained data of X-ray diffraction patterns, it was found that the greatest changes in the shape of diffraction peaks are observed at irradiation doses above 1×10^{15} ions/cm², which are characterized

by an increase in the overlapping areas of defects by more than 100 times. As is known, in the case of single ion trajectories isolated from each other in the material, most of the formed point defects are

capable of annihilating with each other, thereby not making a significant contribution to the change in the concentration of defects in the ceramic material. However, in the case when the trajectories of incident ions begin to overlap, which is observed with an increase in the radiation dose, the concentration of defects becomes much higher and can lead to the formation of regions of disorder and deformation

of the crystal lattice in the structure. In the case of high-temperature refractory ceramics, the formation of defect regions can lead to a decrease in the degree of perfection of the crystal structure due to partial disordering and deformation. Figure 2 shows the dynamics of the dependence of the change in the degree of crystallinity of the studied ceramics as a result of irradiation.

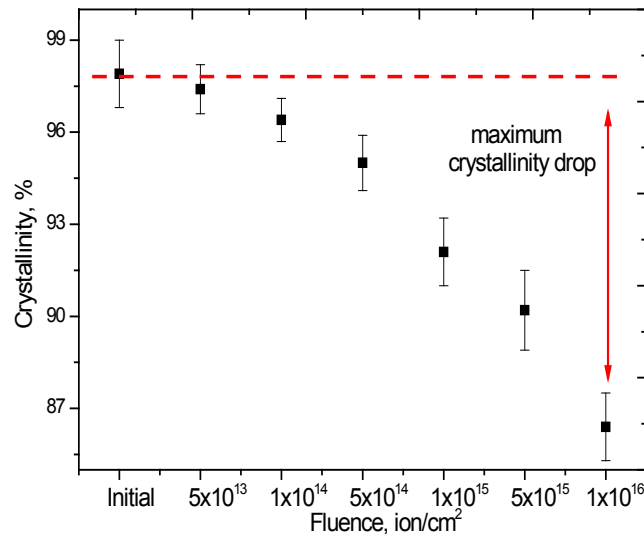


Figure 2 – Dynamics of changes in the degree of crystallinity of the studied ceramics as a result of irradiation

According to the data obtained, it can be seen that the greatest decrease in the degree of crystallinity is observed for samples irradiated with a dose of 1×10^{15} ion/cm² and above. At the same time, the maximum decrease for an irradiation dose of 1×10^{16} ion/cm² was no more than 10 %, which in turn indicates a high radiation resistance to the accumulation of defects during large-dose irradiation.

Based on the change in the shape and intensity of the diffraction peaks, the contributions from amorphous-like inclusions and highly disordered regions in the structure of ceramics, which appeared as a result of an increase in the radiation dose, were calculated. The calculation method was based on the approximation of diffraction peaks by the required number of pseudo-Voigt functions, which make it possible to separate the contribution from amorphous-like inclusions and an ordered

structure. Figure 3 shows the results of these calculations.

As can be seen from the data presented, the change in the concentration of amorphous-like inclusions in the structure of ceramics as a result of an increase in the irradiation dose is nonlinear and has a pronounced increase in the samples irradiated with a dose above 1×10^{15} ion/cm². This nonlinearity of changes in the concentration of amorphous-like inclusions is due to an increase in the regions of overlapping ion trajectories in ceramics, as a result of which the defects do not have time to annihilate, but form cluster two-dimensional and three-dimensional defects. In this case, the formation of strongly disordered regions can lead to a strong deformation of the crystal lattice, which is also clearly seen from the asymmetric shape of the diffraction maxima for samples irradiated with high doses.

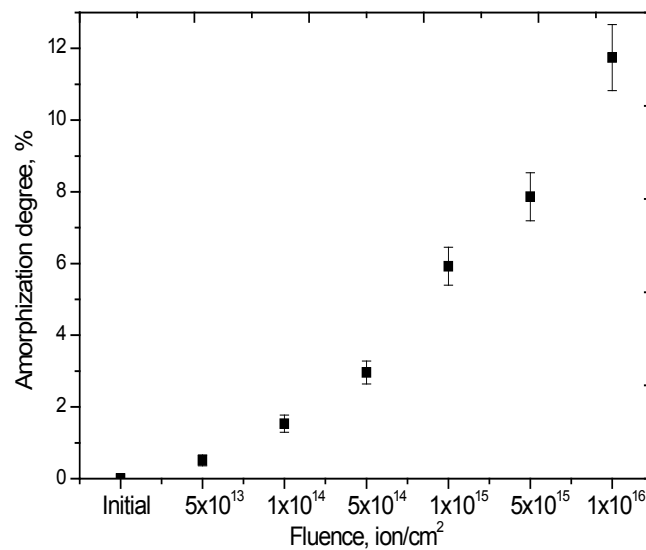


Figure 3 – Graph of the dependence of the change in the concentration of amorphous-like inclusions in the structure of ceramics as a result of an increase in the radiation dose

4 Conclusions

In conclusion, the studied ZrO₂ ceramics have a high degree of resistance to radiation damage under high dose irradiation, and the maximum degree of disordering and amorphization of the structure at high irradiation doses of 1x10¹⁵ – 1x10¹⁶ ion/

cm² is no more than 7-12 %. At the same time, no new phase inclusions or impurity formations were observed for all irradiated samples, which also indicates the high resistance of these ceramics not only to radiation damage, but also to phase transformations that can occur during large-dose irradiation.

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