STUDY OF PROPERTIES OF IRRADIATED BeO CERAMIC

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The paper presents study results of structural and optical changes in heavy ion irradiated BeO ceramics. Irradiation was carried out on DC-60 heavy ion accelerator using Ni^{12+} ions with an energy of 100 MeV with irradiation fluence of 10^{13}-10^{14} ions/cm^2. It has been determined that change in magnitude of atom displacements from lattice sites is exponential, which is conditioned by defect overlap regions occurrence at fluence of 10^{14} ions/cm^2, followed by formation of a large number of migrating defects in structure, leading to crystal structure distortion and deformation due to chemical bonds rupture. In case of defect overlap areas generation, characteristic for irradiation fluences of 5×10^{13} - 10^{14} ions/cm^2, amorphous-like inclusions formation of more than 5% was observed, that leads to thermal conductivity decrease by 15-20%.

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

Introduction

Therefore, studies related to processes of defect formation and radiation resistance are of not only scientific, but also practical interest in predicting behavior of ceramics under irradiation, which will allow us to estimate applicability domain and service life of structural materials for the new IV-th generation of nuclear reactors [1-3]. The main sources, that form defects in construction materials of nuclear reactors during operation, are neutron radiation and uranium fission fragments with an energy of 100-150 MeV. Studies on radiation resistance of beryllium and its oxide to neutron radiation originate in the middle sixties of the XX century [4-6]. At the same time, most data on radiation resistance cannot be compared with each other due to variety of sample types (single crystals, polycrystals, samples with different impurities), which is related to obtaining methods as well as to difference in operating parameters of nuclear reactors.

As a result of the studies performed, it was established that at high radiation doses (10^{13}-10^{21} neutrons/cm^2), the main defects in the structure are changes in grain sizes as well as crystallites crushing and formation of amorphous-like inclusions. At the same time, changes in structural properties are usually conditioned by energy transfer from incident particles to crystal lattice atoms, followed by formation of primary knocked-on atoms and cascades of secondary defects (in case of uranium fission fragments interaction) and defects accumulation due to nuclear reactions with following impurity inclusion formation as a result of interaction with neutrons [5-7]. One of the ways to estimate neutron irradiation effect on structural properties was proposed by Wigner, which consist in estimation of atom displacements from lattice sites. According to the theoretical model, which was later confirmed experimentally, atoms displaced from lattice sites can lead to formation of cascades of primary knocked-on atoms if their energy exceeds the binding energy, that has a significant effect on change in physical dimensions and conductivity of nuclear structural materials.

The paper presents study results for heavy ion irradiation of ceramic materials based on beryllium oxide. Choice of Ni^{12+} ions with an energy of 100 MeV allows you to simulate the impact of radiation on near-surface layer depth of more than 10-12 μm and radiation defects creation, that is comparable to the neutron effect on material.

Experimental part

BeO based ceramics with the thickness of 15 μm and area of 5×5 mm was selected as original samples. Berlox® brand samples were purchased from American Beryllia Inc., USA. Chemical purity is 99.999%.

Irradiation was carried out on DC-60 heavy ion accelerator (Institute of Nuclear Physics, Ministry of Energy of the Republic of Kazakhstan) by Ni^{12+} ions with an energy of 100 MeV, irradiation fluence was 10^{17}-10^{18} ions/m^2, which corresponds to formation of defects overlap regions during ions interaction with the crystal structure. The beam current when irradiated with 100 MeV Ni^{12+} ions is 100 μA, and water cooling of the target substrate is used. Vacuum in chamber during irradiation was 5×10^{-5} Torr. Samples in vacuum chamber are attached to electrically insulated cooled target holder. Measurement of target charge allows us to control irradiation fluence gained by the target. Magnetic system for suppressing secondary electron emission has been installed in the chamber in front of the target to improve measurement accuracy. The system is an iron demountable frame, measuring 10×10 cm, that serves as an external magnetic conductor with a counter-oriented set of permanent magnets installed in it. A scanning system is used for uniform distribution of the ion flux. This system is based on magnetic principle of the ion beam deflection horizontally and vertically with a frequency of (85 - 300) Hz. The frequencies for vertical and horizontal deviations are different, which allows you to form a grid on the target (Lissajous figures) for the purpose of uniform irradiation of the target. Current measurement on the target is performed using an ORTEC 439 digital current integrator, working in combination with an ORTEC 994 counter. Study of the dynamics of changes in structural properties and basic crystallographic characteristics before and after irradiation was conducted by X-ray diffraction analysis on D8 ADVANCE ECO diffractometer (Bruker, Germany) using CuK_α radiation. Bruker AXS DIFFRAC.EVA v.4.2 software and the ICDD PDF-2 international database were used to identify phases and study the crystal structure.

Results and discussion

As can be seen from the presented data, the initial sample surface is characterized by a low degree roughness (no more than 3-10 nm), which is caused by technological processes of samples manufacturing. For irradiated samples a change in surface relief is observed with formation of porous inclusions, the average size of which is from 50 to 100 nm, and increase in roughness degree up to 50-100 nm. At the same time, at large irradiation fluences cleavages and height differences are observed on the studied samples surface, presence of which may be conditioned by near-surface layer peeling as a result of amorphization and stress concentration increase in the structure under irradiation.

The X-ray analysis method was used to estimate irradiation influence on structural properties and concentration of distortions and deformations in the crystal lattice. Dynamics of changes in X-ray diffraction patterns before and after irradiation is shown in Figure 1.

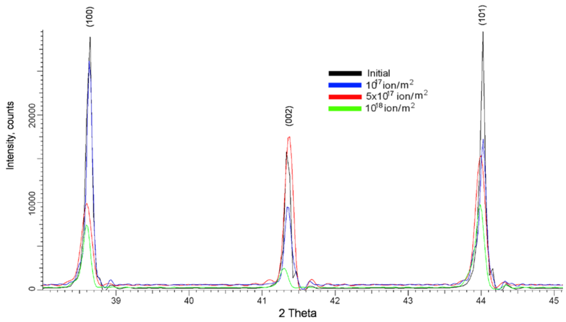

Fig. 1. Dynamics of changes in main diffraction maxima as a result of irradiation

The irradiated samples show a sharp change in shape and intensity of diffraction maxima, which indicates appearance of additional microstresses and defects in the structure as a result of irradiation. At the same time, for samples irradiated with fluence of 10^{18} ions/m^2 a sharp decrease in diffraction maxima intensity is observed as well as amorphous inclusions formation in the structure, which confirms the data of optical measurements of degradation of the structure. At that, the largest amorphous halo formation is observed along the textural direction (101), which may be caused by more active defects migration along this textural direction and low radiation resistance of ceramics in this texture plane. According to the presented diffraction patterns, as irradiation fluence increases a change in shape and width of the diffraction lines is observed, which is conditioned by a change in concentration of distortions and deformations of the structure. A sharp increase in the distortions and deformations of the crystal lattice leads to a change in the interplanar distances and a shift of the diffraction maxima to the region of small angles. The change in interplanar distances and lattice parameters is due to migration and penetration of initially knocked-on atoms from lattice sites into the interstitial sites, as well as an increase in concentration of defects and local regions of disorder as a result of arising of overlapping regions of cascade defects. In case of heavy ion irradiation with an energy about 100 MeV an enormous amount of energy is transferred into the crystal structure, and "injection" intensity of the energy into the electronic subsystem is 100–1000 times greater than into the nuclear one. It should be noted that only a part of kinetic energy of incident particles and primary knocked-on atoms is dissipated in the form of displaced atoms potential energy. Most of the energy is stored as excitation energy and is transformed into thermal energy. As a result, a shock wave appears in the structure, which carries out a part of interstitial atoms from cascade region, forming reduced atomic density areas with generation of zones of disorder or amorphous inclusions.

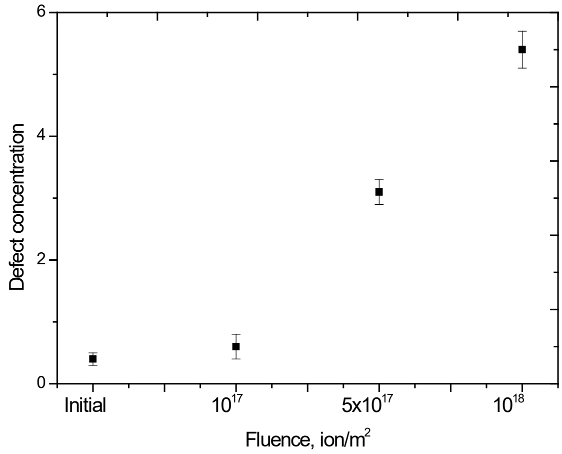

Fig. 2. Dependence graph of defect concentration in the structure on irradiation fluence

As it is seen from the presented data, with small irradiation fluences, in which single point defects formation is typical, most of which recombine, defect concentration in the structure is insignificant. For large fluences, which are characterized by defect cascades formation, leading to formation of thermal bursts overlapping regions, an increase in defect concentration is observed in the structure. Concentration increase is conditioned by amorphization processes, displacement of atoms from crystal lattice sites, distortions and deformations increase in the crystal structure. The method of measuring magnitude of root-mean-square atom displacements from lattice sites was applied for estimation distortions and deformations of the crystal structure resulting from external influences.

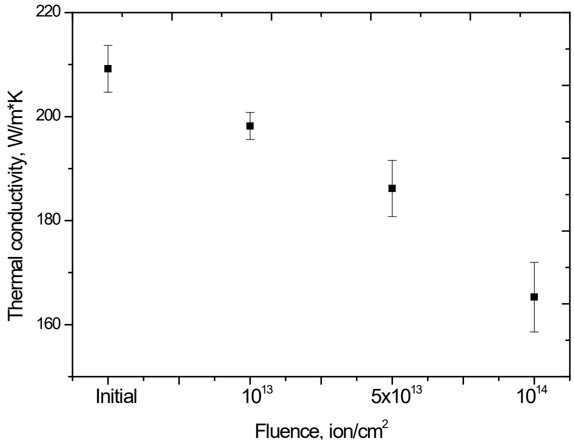

Fig. 3. Dependence graph of change in the thermal conductivity coefficient on irradiation fluence

Drop in thermal conductivity may be caused by changes in the ceramics density and perfection degree of the structure, change dynamics of which is presented in Figure 3. According to the data obtained, an increase in atom displacements from lattice sites as well as porous inclusions formation in the structure, leads to a sharp decrease in density and crystallinity degree of the ceramics, that indicates amorphous-like inclusions appearance in the structure. Amorphous-like inclusions formation leads to crystal structure deformation and distortion, which has a significant impact on heat-conducting and insulating characteristics of the ceramics. In turn, decrease in the value of thermal conductivity can lead to performance degradation of nuclear reactor because of reduction in heat exchange and conducting characteristics.

Conclusions

In the course of the conducted research, the dependences of defect formation processes and radiation resistance of BeO ceramics on the fluence of heavy Ni12+ ions irradiation with an energy of 100 MeV were established.

According to X-ray structural analysis, atomic force microscopy and scanning electron microscopy it has been established that an increase in concentration of distortions and displacements of atoms from lattice sites is typical in case of defect overlap regions formation at irradiation fluences of 5x1017 ions/m2 and above. At these irradiation fluences the processes of formation and accumulation of defects and vacancies prevail over the processes of spontaneous recombination. As a result, it was established that decrease in optical properties and deterioration of heat-conducting characteristics are caused by defect concentration increase under irradiation in the structure, with subsequent occurence of regions of disorder in near-surface layer and formation of hillocks.

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