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The effect of irradiation with swift heavy ions on the structural and morphological properties of beryllium oxide ceramics

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In this study the results of structural and morphological changes in Ni^{12+} heavy ion irradiated BeO ceramics are presented. 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 inclusions formation of more than 5% was observed, that leads to thermal conductivity decrease by (15-20)%.

Keywords: heavy ions, radiation defects, nuclear power engineering, construction materials, radiation resistance.

Introduction

In the modern world, great attention is paid to new construction materials for nuclear power engineering, which are operated in harsh environments, such as high temperatures, intense fluxes of ionizing radiation and corrosive environments [1, 2]. In turn, BeO ceramics stands out among all types of ceramics due to unique combination of high thermal conductivity with high electrical resistivity. Also due to low value of thermal neutron absorption cross section, high chemical and corrosion resistance and thermal conductivity, BeO ceramics have great potential for use as structural materials for nuclear and atomic industry [3-5]. The main requirement for structural materials is to ensure stability of working characteristics, such as thermal conductivity, electrical resistance, high radiation and corrosion resistance to external influences. When ceramics are irradiated with ionizing radiation, in particular by heavy ions, an enormous number of defects appear in their structure, most of which annihilate in a very short time (10^{-14}-10^{-12} s) as a result of defects radiation annealing [6, 7]. However, the remaining part of defects can lead to significant change in physicochemical, structural, and optical properties of the ceramics by creating cascades of secondary defects and displaced atoms. At the same time, defects accumulation in structure can lead to a change in structural characteristics, accumulation of amorphous inclusions and areas of disorder in near-surface layer, which, subsequently, can lead to deterioration of thermal conductivity and working characteristics of structural materials [8-11]. 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 [12-15]. 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 [16, 17]. 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 [18-20]. In the course of previously conducted studies, a procedure was developed for comparing different types of radiation, which is based on measuring changes in structural parameters, defect concentration in the structure and magnitude of atom displacements from lattice sites [21-23]. 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 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 [24]. 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 [25]. Under neutron irradiation with large fluences, nuclear reactions can occur in materials, resulting in impurity and amorphous inclusions formation, that leads to swelling and destruction of near-surface layer from 1 to 15 microns thick [26, 27]. However, It is worth noting that reactor tests of studied samples during neutron irradiation are carried out for quite a long time, which can be more than 10 years, while the samples after irradiation need to be hold for a certain time in order to reduce background radiation. It is also worth noting that reactor tests of studied samples during neutron irradiation are carried out for quite a long time, which can be more than 10 years, while the samples after irradiation need to be hold for a certain time in order to reduce background radiation. In turn, the use of heavy ion accelerators makes it possible to simulate almost all standard effects on structural, conducting, and insulating features of materials that are observed upon neutron flux irradiation in nuclear reactors [28]. At that, heavy ion irradiation is very valuable and useful tool in studies of neutron radiation effect on radiation resistance and defect formation processes in structural materials. High-accuracy control of irradiation conditions, such as fluence, incident particles energy, irradiation temperature, makes it possible to simulate exposure conditions of ionizing radiation close to the real ones [29]. So in the works of W.J. Weber et al., the prospectivity of ion beams application to simulate defect formation processes and ceramics amorphization mechanisms, comparable to the neutron effect, is shown [30, 31].

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.

Materials and Methods

BeO based ceramics with the thickness of 15 μm and area of 5×5 mm were selected as original samples. The studied samples were hexagonal-type lattice polycrystalline structures with lattice parameters a = 2.671 Å, c = 4.332 Å(*Bromellite* , PDF – 01-077-9751).

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^{13}-10^{14} ions/cm^2, which corresponds to formation of defects overlap regions during ions interaction with the crystal structure. Maximum free path of Ni^{12+} ions is 12.7 ± 0.5 μm, radial deviation - 500 ± 50 nm, number of vacancies resulting from the interaction - 11200 ± 100 vacancies/ion.

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 CuKα 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.

The morphological properties were studied by using Scanning Electron Microscope (SEM) JEOL JSM-7600F at an accelerating voltage of 15.0 kV, LEI regime.

Results and Discussion

The dynamics of changes in surface morphology and lateral cleavages of the studied samples before and after irradiation is shown in Figures 1-4. 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.

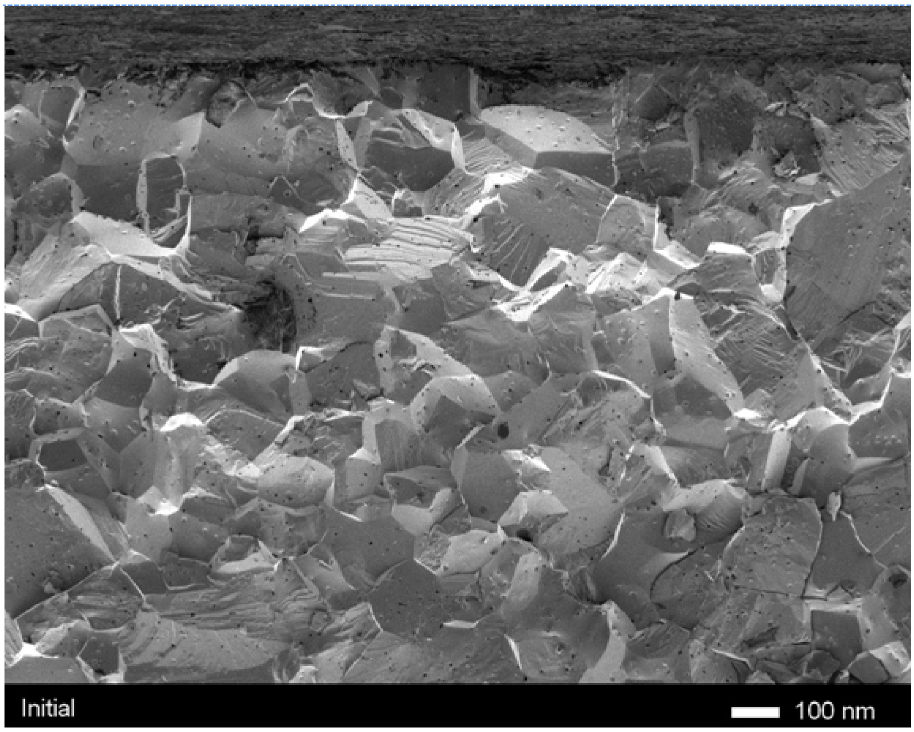

Figure 1. Lateral cleavages of the studied samples after irradiation: the arrow indicates the maximum path length of nickel ions in ceramics: initial sample.

As can be seen from the presented data, when ceramics are irradiated with heavy ions, there is a change in crystallite size and formation of porous inclusions, average size of which does not exceed (10-15) nm. Irradiation fluence increase leads to amorphous inclusions formation in ceramics structure, which confirms the results of changes in structural characteristics. At large irradiation fluences, a

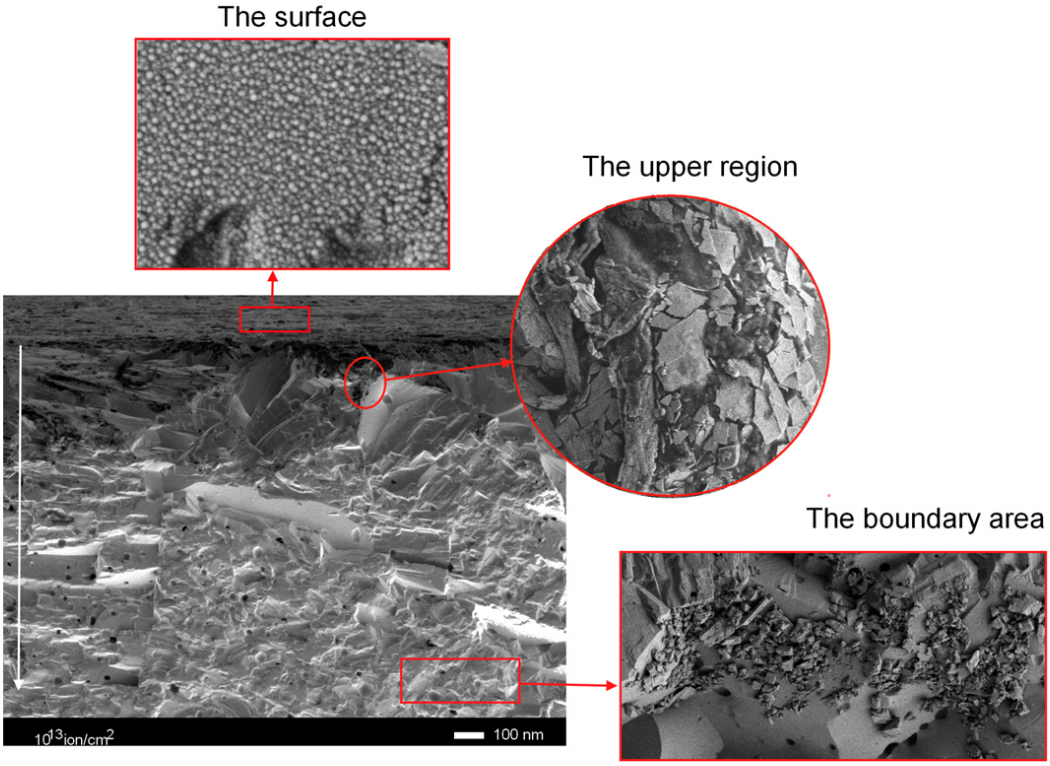

Figure 2. Lateral cleavages of the studied samples after irradiation: the arrow indicates the maximum path length of nickel ions in ceramics: 10^{13} ion/cm^2.

coarsening of amorphous inclusions in the ceramics near-surface layer is observed, as well as grains fragmentation as a result of incident ions interaction with crystal structure of ceramics. According to the data obtained, spherical inclusions formation is observed on the irradiated samples surface, density and dimensions of which increase as the irradiation fluence raises with the subsequent formation of cracks on the surface. Larger defect inclusions are observed in near-surface layer close to grain boundaries and defect sinks, as well as a high content of amorphous inclusions is observed at the depth of the maximum ion range. Formation of regions of disorder as a result of elastic and inelastic collisions of the incident ions with crystal lattice atoms leads to structural characteristics deterioration and a significant change in crystallographic characteristics of the studied samples.

The X-ray analysis method was used to estimate irradiation influence on structural properties and concentration of distortions and deformations in the crystal lattice. Figure 5 shows dynamics of changes in main diffraction peaks with Miller indices (100), (002) and (101) 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^{14} ion/cm^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. Shift of maximum

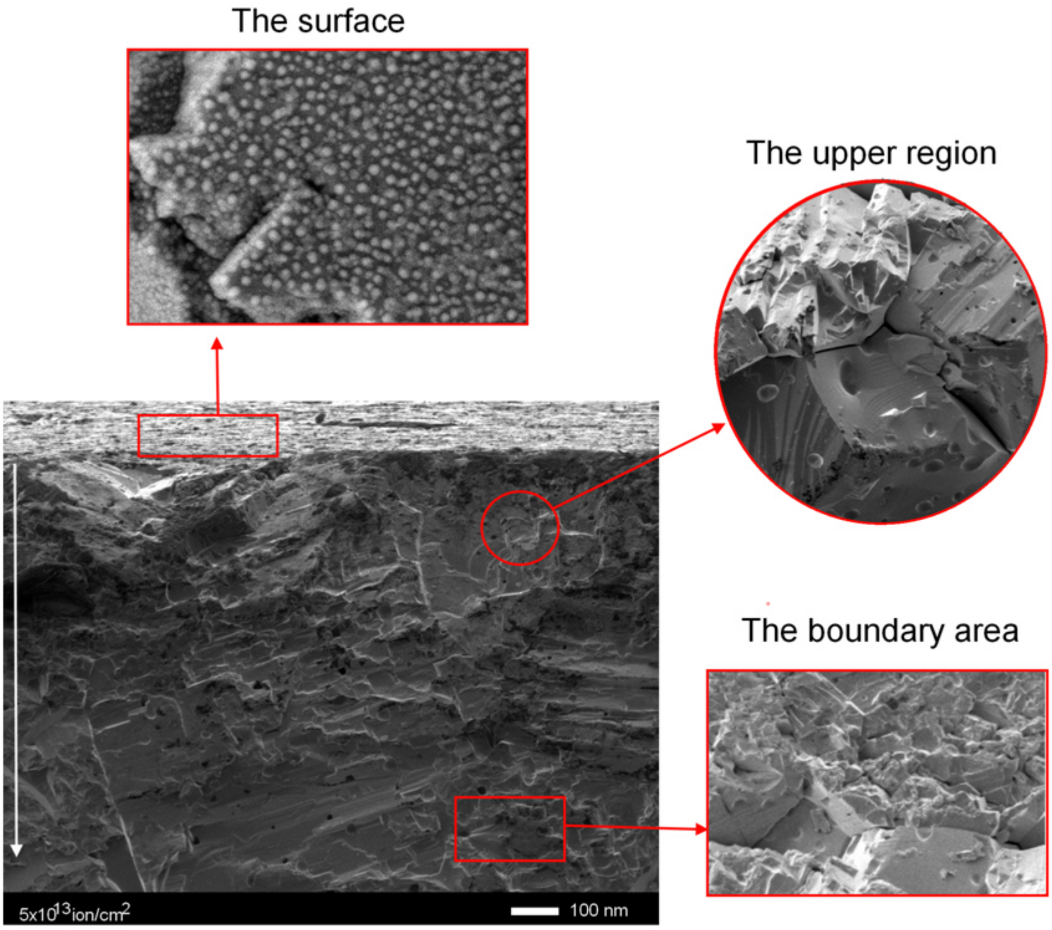

Figure 3. Lateral cleavages of the studied samples after irradiation: the arrow indicates the maximum path length of nickel ions in ceramics: 5×10^{13} ion/cm^2.

of diffraction line to small angles indicates interplanar distances increase, that is induced by migration and penetration of knocked-on atoms from lattice sites into interstitial sites as well as by increase in concentration of defects and local regions of disorder in the structure. Change in diffraction line shape can be conditioned by two factors: size effect and crystal structure distortion or deformation. According to shape and width analysis of the diffraction lines using the Williamson-Hall method, both factors have equally probable effect pattern on structural properties change for irradiated samples. Results of changes in defect concentration in the structure are presented in Figure 6a.

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. The displacements quantity estimation is performed by measuring ratios of the two

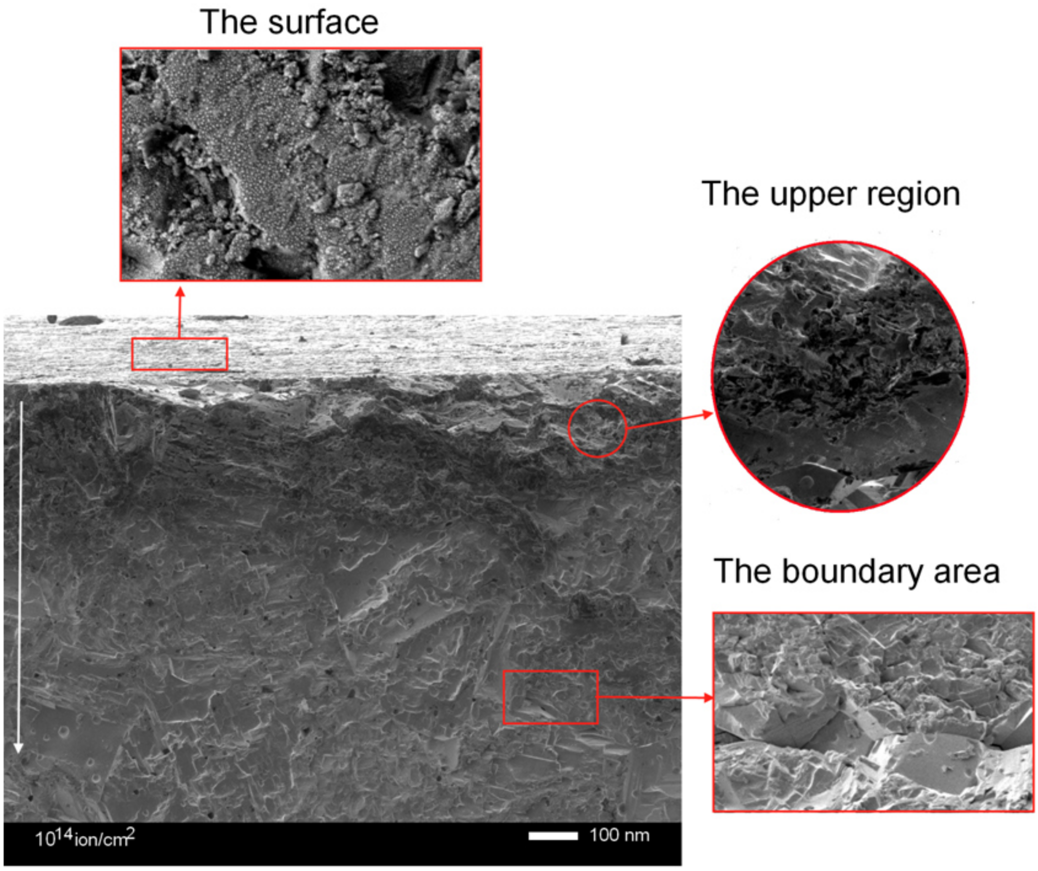

Figure 4. Lateral cleavages of the studied samples after irradiation: the arrow indicates the maximum path length of nickel ions in ceramics: 10^{14} ion/cm ^{2} .

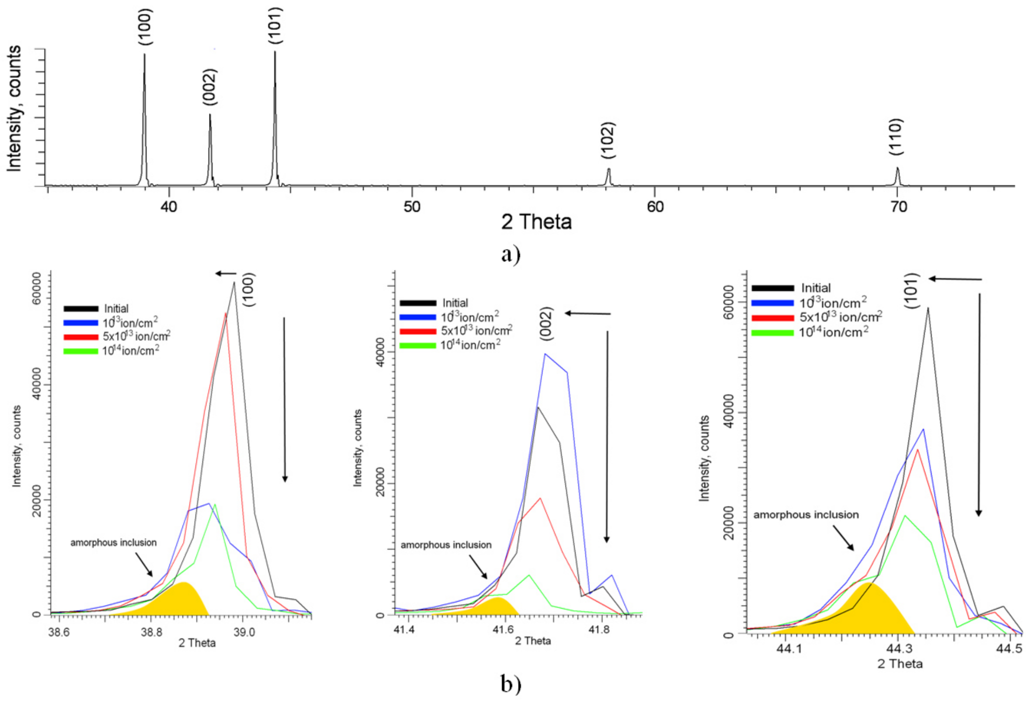

Figure 5. a) X-ray diffraction pattern of the original sample; b) Dynamics of changes in main diffraction maxima as a result of irradiation.

most intense lines of the same sample before and after irradiation, and is calculated by the formula (1):

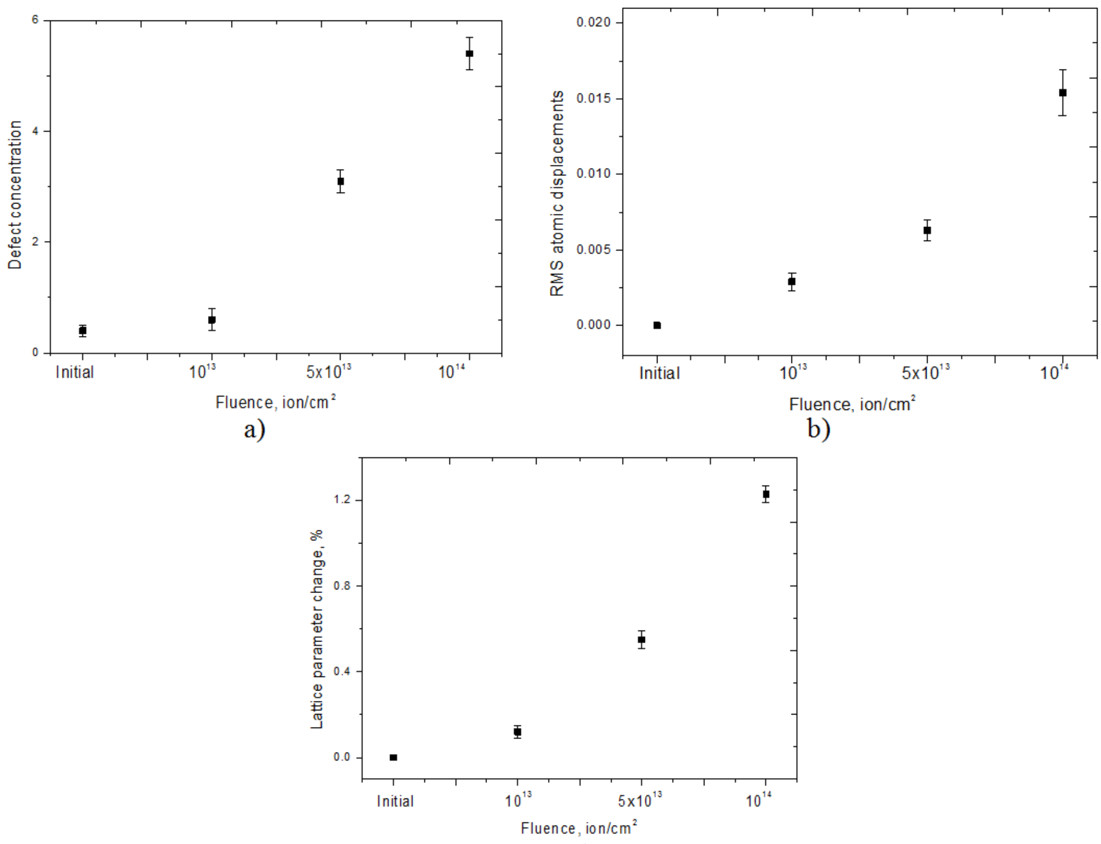

Figure 6. a) Dependence graph of defect concentration in the structure on irradiation fluence; b) Dynamics of changes in root-mean-square atomic displacements caused by irradiation; c) Dynamics of changes in crystal lattice parameters during irradiation.

$$U^2 = 3a^2 \ln\left[(I_1/I_2)_{irridiated} \,/\, (I_1/I_2)_{initial}\right] /$$

$$/4\pi^2 \left[(h_2^2 + k_2^2 + l_2^2) - (h_1^2 + k_1^2 + l_1^2)\right], \quad (1)$$

where a – lattice parameter, $(I_1/I_2)_{initial}$, $(I_1/I_2)_{irradiated}$ – diffraction lines intensity ratios before and after irradiation, respectively. Evaluation of displacements magnitude was carried out by studying the changes in shape and intensity of main diffraction lines on x-ray patterns, which is caused by changes in interplanar distances and crystal structure deformation as a result of external influences. Figure 6c shows dynamics of changes in crystal lattice parameters after irradiation. According to the data obtained, at small irradiation fluences, at which single defects formation is observed, the change in crystal lattice parameter value is insignificant. This is conditioned by the fact that most of the defects have time to annihilate in a very short time. (10^{-13} s). As irradiation fluence rises, a sharp increase in the parameter and root-mean-square displacement of atoms from lattice sites is observed, which is caused by distortions concentration increase in the structure. Formation of cascades of point defects and vacancies, as well as primary knocked-on atoms is characteristic at large irradiation fluences. After 10^{-13} s, atoms displacement in the structure stops, and athermal rearrangement or spontaneous recombination begins, which is not associated with thermal excitations of atoms. Spontaneous recombination lifetime is 10^{-11} s, as a result of which neighboring defects annihilation or same type defects recombination occurs, followed by formation of vacancy or interstitial type complexes. However, in case of overlapping defects regions formation in the structure, specific for irradiation fluences of 5×10^{13} - 10^{14} ions/cm^2, defects formation and accumulation processes prevail over the processes of spontaneous recombination and annihilation, that leads to density change and formation of amorphous inclusions in the structure.

Conclusion

The paper presents study results of structural and morphological changes in Ni^{12+} heavy ion irradiated BeO ceramics. The choice of nickel ions with an energy of 100 MeV is conditioned by possibility of simulating the effect of radiation on near-surface layer depth of more than (10-12) μm and creating radiation defects, which is comparable to neutron influence on the material. In accordance with X-ray analysis data it has been determined that change in magnitude of atom displacements from lattice sites is exponential, which has great influence on crystal structure distortion and deformation in case of defect overlap regions formation at large irradiation fluences.

References

[1] Lu Cihang et al., Annals of Nuclear Energy **114** (2018) 277-287.
[2] Bonal, Jean-Pierre et al., MRS bulletin **34**(1) (2009) 28-34.
[3] W. Zhou et al., Annals of Nuclear Energy **81** (2015) 240-248.
[4] Isokawa Yuya et al., Optical Materials **76** (2018) 28-33.
[5] A. Kozlovskiy et al., Vacuum **163** (2019) 45-51.
[6] A.J. Leide et al., Journal of Nuclear Materials **514** (2019) 299-310.
[7] J.D. Fowler et al., Journal of the American Ceramic Society **60**(3-4) (1977) 155-161.
[8] H.J. De Bruin et al., Philosophical Magazine **16**(140) (1967) 427-430.
[9] Gong Yihao et al., Philosophical Magazine **98**(2) (2018) 95-106.
[10] P.J. Horodek et.al., Vacuum **138** (2017) 15-21.
[11] A. Bhattacharya et al., Acta Materialia **165** (2019) 26-39.
[12] D.G. Walker, Journal of Nuclear Materials **14** (1964) 195-202.
[13] T. Teichmann et al., Journal of Instrumentation **13**(10) (2018) 10015.
[14] D.G. Walker, Journal of Nuclear Materials **14** (1964) 187-194.
[15] C.B. Garcia et al., Metallurgical and Materials Transactions E **4**(2-4) (2017) 70-76.
[16] V.V. Uglov et al., Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **435** (2018) 228-235.
[17] V.V. Uglov et al., Surface and Coatings Technology **344** (2018) 170-176.
[18] Kong, Xirui et al., Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **422** (2018) 12-17.
[19] E.G. Njoroge, Eric G. et al., Vacuum **144** (2017) 63-71.
[20] W. Dienst Journal of nuclear materials **191** (1992) 555-559.

[21] S.B. Austerman, Journal of Materials Science **1**(3) (1966) 249-260.
[22] F.P. Korshunov et al., Vacuum **83** (2009) 131-133.
[23] W.J. Weber, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms **166** (2000) 98-106.
[24] He Shixiong et al., Corrosion Science **122** (2017) 108-117.
[25] Lu Chengjie et al., Journal of the European Ceramic Society **36**(14) (2016) 3319-3327.
[26] K. Dukenbayev et al., Materials Research Express **5** (2018) 065502.
[27] A. Kozlovskiy et al., Vacuum **155** (2018) 412-422.
[28] Yang, Tengfei et al., Journal of Nuclear Materials **513** (2019) 120-128.
[29] A. Kozlovskiy et al., Optical Materials **91** (2019) 130-137.
[30] W.J. Weber et al., Journal of Materials Research **13**(6) (1998) 1434-1484.
[31] W.J. Weber et al., Journal of Nuclear Materials **250**(2-3) (1997) 147-155.