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Study of change in beryllium oxide strength properties as a result of irradiation with heavy ions

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The paper presents data on changes in strength properties, including data on microhardness, crack resistance, bending strength and wear of BeO ceramics as a result of irradiation with heavy accelerated ions. The following types of ions were selected as heavy ions: O^{2+} (28 MeV), Ar^{8+} (70 MeV), Kr^{15+} (147 MeV), Xe^{22+} (230 MeV). Radiation doses were 10^{13}-10^{15} ion/cm^2, which make it possible to assess the effect of both single defects arising from radiation, and cluster overlapping defective areas occurring at large radiation doses. During the studies carried out, it was found that an increase in the ion energy and, consequently, in the damaging ability and depth of the damaged area, leads to a sharp decrease in the strength mechanical characteristics of ceramics, which is due to an increase in defective areas in the material of the near-surface damaged layer. However, an increase in irradiation dose for all types of exposure results in an almost equilibrium decrease in strength characteristics and the same trend of change in strength characteristics. The obtained dependencies indicate that the proposed mechanisms responsible for changing the strength properties can, under certain assumptions, be extrapolated to various types of exposure to heavy ions in the energy range (25-250 MeV).

Keywords: strength, heavy ions, hardness, ceramics, radiation damage.

Introduction

Today, one of the promising materials for practical application in the field of structural materials for nuclear power, in particular high-temperature reactors, is ceramic based on beryllium oxide (BeO) [1-3]. Interest in it is due to the huge potential in the field of application of refractory materials operating at high temperatures, high neutron fluxes, exposure to aggressive media, etc. The choice of beryllium oxide and ceramics based on it for structural materials will significantly increase the temperature of the working core, as well as high parameters of the neutron capture cross section allow using these ceramics as neutron moderators in the core [4-7]. However, despite such a wide range of application prospects, a number of questions still remain regarding the preservation of the stability of the strength properties of ceramics under the influence of irradiation with heavy ions comparable to fission fragments of uranium nuclei. According to the literature data of radiation resistance of beryllium oxide, today there are only scattered data on resistance to exposure to heavy ions [8-14]. Most research works are devoted to the study of the effect of neutron radiation and date back to 1950-2000 [15-20]. Current data on the resistance of beryllium oxide to heavy ion irradiation are insufficient, making such research in this field the most promising in view of their large potential for use as structural materials for nuclear power [21-24]. Based on the above, the main goal of this work is to study the effect of irradiation with heavy ions such as O^{2+} (28 MeV), Ar^{8+} (70 MeV), Kr^{15+} (147 MeV), Xe^{22+} (230 MeV) on the strength mechanical properties of ceramics exposed to irradiation. Interest in this study is due to the possibility of obtaining new experimental data on the resistance of ceramics to radiation, as well as determining their resistance to radiation and maintaining strength properties.

Experimental part

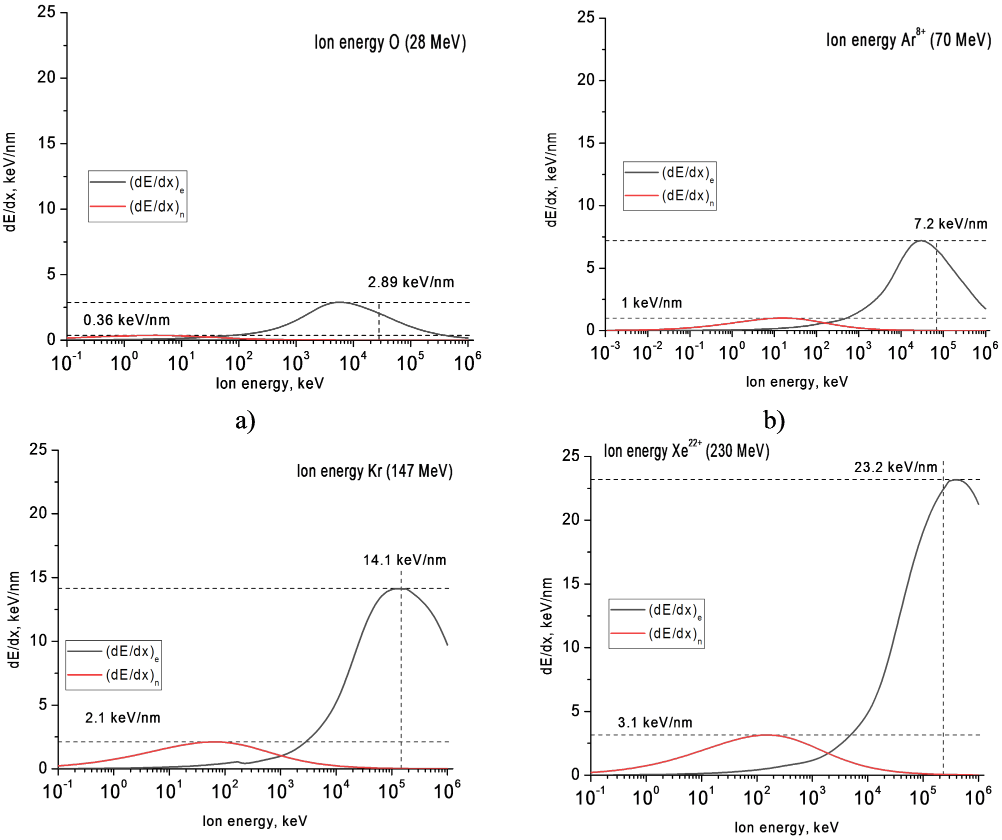
Polycrystalline ceramics of the hexagonal type BeO, spatial system P63mc (186) were chosen as objects of study. According to the PDF2-2016 database, the crystal lattice parameters for the studied samples after refinement with the DiffracEVA v4.2 program code were a=2.671 A, c=4.332 A (PDF-01-077-9751).

The study of the effect of irradiation with heavy O^{2+} (28 MeV), Ar^{8+} (70 MeV), Kr^{15+} (147 MeV), Xe^{22+} (230 MeV) ions was carried out at the DC-60 heavy ion accelerator located in Nur-Sultan, Kazakhstan. Irradiation was carried out on water-cooled targets in order to avoid overheating of the samples under the action of irradiation. The irradiation doses were 10^{13}-10^{15} ion/cm^2. The calculation of the energy losses of incident ions, as well as the maximum depth of ion penetration into the ceramics, was carried out on the basis of model data on the ceramics density using the SRIM Pro 2013 program code [25].

The mechanical properties of ceramics and their changes as a result of irradiation were measured according to standard methods for determining microhardness using indentation, determining the bending strength and wear of ceramics as a result of friction, and determining crack resistance. All methods comply with GOSTs for determining the mechanical strength properties of materials.

Results and discussion

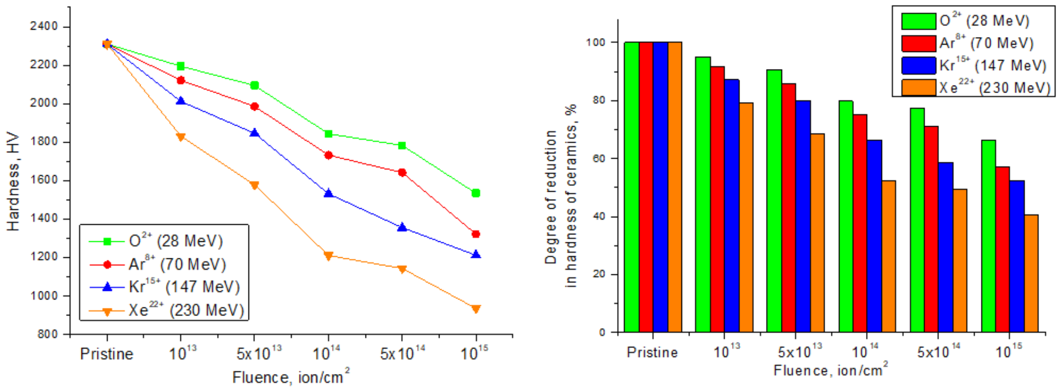
Figure 1 shows the results of modeling the radiation losses of incident ions in BeO ceramic depending on the type of ions and the energy of incident ions. Modeling of energy losses was carried out in the SRIM Pro 2013 program code based on the radiation damage model of Kinchin-Pisa. The following conclusions can be drawn from the presented energy loss diagrams. An increase in the energy of ions and a change in their type leads to an increase in energy losses both in interaction with nuclei and with electronic shells. At the same time, the difference between electronic and nuclear losses is 5-7 times for all types of ions. Based on the calculated data, ion path lengths in ceramic were calculated, which ranged from 7 to 20 μm depending on the type of ions.



a) b) c) d)

Figure 1. Calculated data of energy losses of incident ions calculated using the SRIM Pro 2013 program code a) O^{2+} (28 MeV); b) Ar^{8+} (70 MeV); c) Kr^{15+} (147 MeV); d) Xe^{22+} (230 MeV).

Figure 2 shows the results of changes in the microhardness value depending on the radiation dose, as well as the degree of decrease in the hardness value depending on the type of radiation. The general view of the obtained dependences of the change in the value of hardness can be divided into two characteristic stages. The first stage is typical for irradiation doses of 10^{13}-5×10^{13} ion/cm^2 and is characterized by a slight decrease in the hardness of ceramics for samples irradiated with O^{2+}, Ar^{8+}, and Kr^{15+} ions. This behavior indicates a small radiation damage influence degree on the strength properties of ceramics. The second stage for these types of ions is characterized by a decrease in the strength by 25-40% of the initial value, which indicates a partial degradation of the structure and its disorientation, leading to a decrease in the strength of ceramic. A distinctive feature are the cases of irradiation with Xe^{22+} ions for which a sharp decrease in hardness is observed even at low fluences, which indicates the low stability of the near-surface layer to irradiation with Xe^{22+} ions. With large fluences of irradiation with Xe^{22+} ions (10^{14}-10^{15} ion/cm^2), the drop in microhardness values of the near-surface layer of ceramic was more than 50% of the nominal value before irradiation. This behavior of changes in microhardness may be due to the large contribution of energy losses to disordering and defect formation, which leads to the formation of microcracks and amorphous inclusions in the near-surface layer. At the same time, one should take into account the fact that the damage area along the trajectory for Xe^{22+} ions, in contrast to all other types of ions, is much larger and is approximately 30-50 nm (approximate estimates according to the SRIM Pro 2013 program code), which leads to the fact that at irradiation doses of 10^{13}-5×10^{13} ion/cm^2, a partial overlap of the defective regions is observed. This overlap causes a large number of point defects to remain in the structure of the near-surface layer, resulting in additional distortions and deformations of the ceramic structure.



a) b)

Figure 2. a) Graph of microhardness change versus type of external effects; b) Diagram of the degree of reduction in hardness of ceramic versus irradiation fluence and ion type.

Figure 3 shows the results of the change in the crack resistance of BeO ceramic depending on the ion type and radiation dose. The general view of the change in the crack resistance has the same nature of the change as the microhardness of ceramic under the influence of radiation. A decrease in crack resistance as a result of an increase in fluence indicates deformation mechanisms of damage to the ceramic structure as a result of radiation damage, as well as distortion of the crystal lattice, which lead to swelling. An increase in the distortions contribution and deformations in the structure leads to a deterioration in strength and, consequently, a decrease in crack resistance.

Table 1 shows the results of measurements of such values as bending strength and wear of ceramic as a result of friction obtained during the measurement of

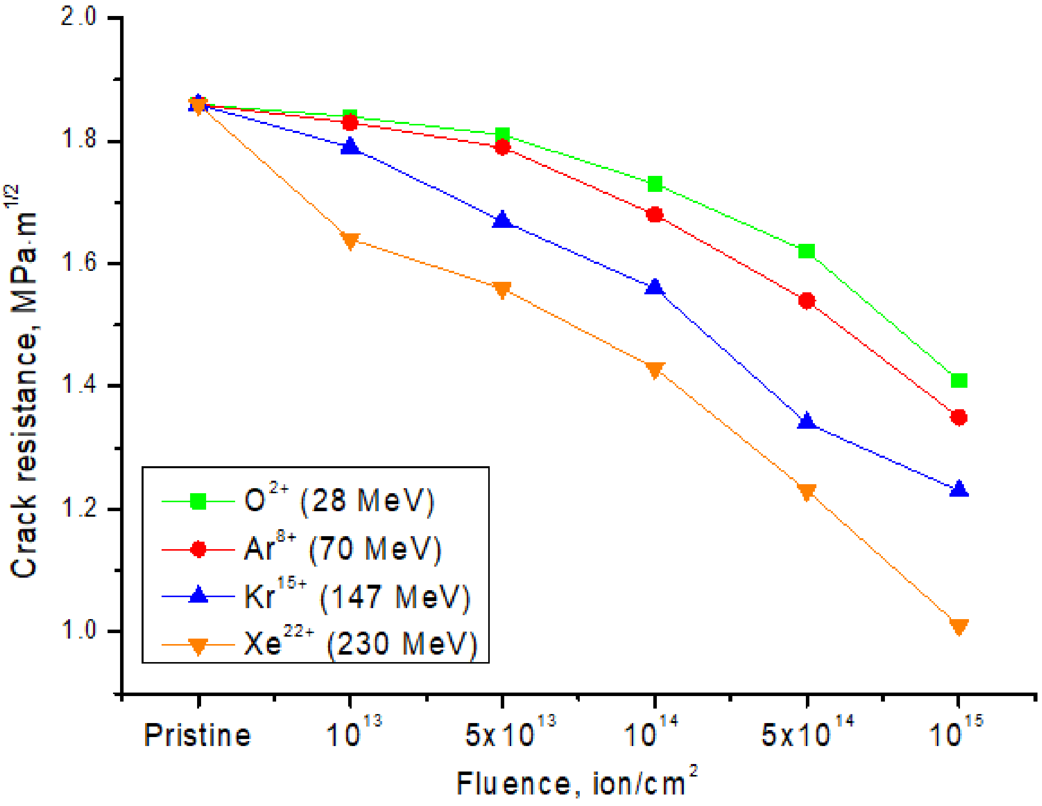


Figure 3. Graph of crack resistance variation of BeO ceramic to irradiation.

strength characteristics.

As can be seen from the data presented, the greatest changes in strength properties are observed for samples irradiated with Kr^{15+} and Xe^{22+} ions, for which the greatest changes in hardness and a decrease in crack resistance were observed. At the same time, the greatest wear during friction tests was more than 40 percent for samples irradiated with Xe^{22+} ions, which indicates deterioration in ceramic strength. The general view of the obtained data on changes in strength characteristics indicates the resistance of ceramics to irradiation with O^{2+} and Ar^{8+} ions, and weak resistance to irradiation with Xe^{22+} ions, which has a direct dependence of strength characteristics on the energy of incident ions and their damaging ability due to energy losses. In fact, it was shown in [22, 23] that irradiation with heavy ions with energies above 70 MeV at fluences above 10^{13}-10^{14} ion/cm^2, the main structural distortions are associated with partial disordering and the formation of a large number of point and vacancy defects leading to destruction and partial swelling of the irradiated layer of ceramic. At the same time, according to the general theory of radiation damage based on the model of thermal bursts arising along the trajectory of ions in the material for very short periods of time (10^{-13}-10^{-15} s) and leading to a sharp increase in temperature in local regions of the structure, formed point defects are capable of generating primary knocked-out atoms and cascades of knocked-out electrons. Most of these defects, formed as a result of the collision of heavy ions with the crystal lattice, annihilate (more than 90% of all defects), but the surviving part of point defects is capable of introducing significant deformations and distortions

Table 1.
Data on bending strength and wear of ceramic as a result of friction.

Parameter	Pristine	10^{13} ion/cm^2	$5\cdot10^{13}$ ion/cm^2	10^{14} ion/cm^2	$5\cdot10^{14}$ ion/cm^2	10^{15} ion/cm^2
O^{2+} ion irradiated						
Three-point bending strength (MPa)	154±3	153±2	151±4	146±3	141±2	134±2
Impact toughness (kJ/mm2)	1.42±0.11	1.42±0.13	1.40±0.11	1.38±0.12	1.32±0.11	1.21±0.21
Wear value of ceramic after 10000 test cycles under pressure of 100 N, %	3-5%	3-5%	3-5%	7-9%	11-13%	14-16%
Ar^{8+} ion irradiated						
Three-point bending strength (MPa)	154±3	152±2	149±3	139±3	129±2	121±4
Impact toughness (kJ/mm2)	1.42±0.11	1.41±0.13	1.34±0.11	1.22±0.12	1.11±0.11	1.04±0.15
Wear value of ceramic after 10000 test cycles under pressure of 100 N, %	3-5%	3-5%	5-7%	11-14%	19-21%	25-28%
Kr^{15+} ion irradiated						
Three-point bending strength (MPa)	154±3	151±1	141±2	129±3	111±4	104±3
Impact toughness (kJ/mm2)	1.42±0.11	1.38±0.12	1.21±0.12	1.06±0.22	0.92±0.12	0.76±0.15
Wear value of ceramic after 10000 test cycles under pressure of 100 N, %	3-5%	5-7%	11-13%	19-22%	25-27%	31-33%
Xe^{22+} ion irradiated						
Three-point bending strength (MPa)	154±3	144±2	121±3	103±2	84±2	64±5
Impact toughness (kJ/mm2)	1.42±0.11	1.22±0.12	1.01±0.13	0.85±0.16	0.65±0.13	0.49±0.13
Wear value of ceramic after 10000 test cycles under pressure of 100 N, %	3-5%	11-13%	21-24%	29-31%	35-37%	43-45%

into the ceramic structure. Also, in turn, an increase in the energy losses of incident ions due to an increase in their energy leads to the fact that when heavy ions collide with energy losses, much more bonding energy occurs (20-30 eV for beryllium oxide ceramics according to various data), chemical and crystalline bonds break, which leads to the formation of a large number of primarily knocked out atoms. At the same time, an increase in radiation fluence above 5×10^{13} ions/cm^2 leads to an increase in the effect of overlapping such regions, the radius of which, according to calculated data, varies from 10 to 50 nm depending on the type of ions. Such overlap leads to the cumulative effect of radiation damage and the occurrence of disordered areas in the structure, as well as structurally distorted inclusions. According to the data of morphological studies presented in [22, 23], the authors found that irradiation with heavy ions leads to a sharp change in the surface morphology, as well as partial swelling of ceramic. Such behavior of the near-surface layer under irradiation can be caused by deformation processes that lead to the extrusion of the deformed volume along the grain boundaries to the surface, thereby destroying the crystalline and chemical bonds. Such extrusion can lead to a sharp deterioration in the strength of ceramic, which is clearly demonstrated in this work, and has good confirmation for other types of ceramics exposed to irradiation.

Conclusion

The work is devoted to the study of the effect of irradiation with heavy O^{2+} (28 MeV), Ar^{8+} (70 MeV), Kr^{15+} (147 MeV), Xe^{22+} (230 MeV) ions on the strength and mechanical properties of BeO ceramics. This study is aimed at obtaining new experimental data on the resistance to radiation embrittlement and the decrease in the strength of ceramics with potential use as a basis for structural materials. During analysis of the obtained experimental dependences, it was found that this type of ceramic has the least radiation damage resistance to irradiation with Xe^{22+} ions. At the same time, the greatest decrease in strength properties is due to an increase in the radiation damage contribution due to large energy losses of incident ions in the material.

The obtained data can significantly expand the theory of radiation damage in the future, as well as make a significant contribution to the development of the theory of the radiation damage mechanisms in ceramics.

Further research will be aimed at studying the efficiency of reducing radiation damage as a result of irradiation through thermal annealing or pretreatment of ceramic.

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