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DETERMINATION OF CRITICAL DOSES OF RADIATION DAMAGE TO ALN CERAMIC UNDER IRRADIATION OF HELIUM AND HYDROGEN IONS

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The work is devoted to the study of the radiation damage kinetics to heat-conducting, insulating and mechanical properties in polycrystalline ceramics based on aluminum nitride under irradiation of helium and hydrogen ions, as well as the determination of critical doses that cause maximum irreversible consequences. The choice of ions for irradiation is due to the ability to simulate the radiation damage processes during the accumulation of helium and hydrogen ions in the structure of the near-surface layer with the subsequent formation of gas-filled bubbles. During the studies carried out, it was found that at doses of irradiation with helium ions above 1x10^{17} *ion/cm*^{2}*, there is a sharp deterioration in thermal conductivity and a decrease in ceramic resistance, which is associated with the onset of the formation of helium bubbles and partial embrittlement of the near-surface layer. However, an increase in the radiation dose above 5x10*^{17} *ion/cm*^{2} *does not lead to significant changes in thermal conductivity and insulation characteristics, which indicates the effect of radiation damage accumulation and a decrease in the ceramic degradation rate. In contrast to irradiation with helium ions, irradiation with hydrogen ions to doses higher than 1-3x10*^{17} *ion/cm*^{2} *does not lead to significant changes in the thermal insulation characteristics, which indicates the ceramic resistance to hydrogen absorption processes.*

Keywords: aluminum nitride, ceramics, structural materials, radiation damage, helium swelling, embrittlement.

Introduction

One of the promising materials for nuclear power, in the case of creating high-temperature nuclear reactors, are nitride ceramics such as Si_3N_4, AlN, TiN, BN et.al. [1-5]. As is known, nitride ceramics have a number of unique properties such as high thermal conductivity, radiation and corrosion resistance, chemical inertia, high melting point (more than 1800-2300°C), good insulating properties [6,7]. Among all nitride ceramics, it is worth noting separately polycrystalline aluminum nitride, which, unlike silicon nitride, has high binding energy values, which leads to the absence of extended radiation damage along the trajectory of ions in the material, called latent tracks [8-10]. As is known, the formation of latent tracks in a ceramics material or dielectrics is associated with local changes in the electron density and its redistribution in the structure. At the same time, unlike metals, in which electrons torn from their places during radiation damage can return to their positions, for dielectrics such a return is very difficult [11-13].

The key factor determining the use and service life of ceramics as structural materials is radiation damage resistance, as well as the preservation of insulating and mechanical characteristics under prolonged radiation exposure [14-18]. At the same time, unlike irradiation with heavy ions and radiation damage caused by them in the form of local areas with changed electron density, as well as deformation of the crystal lattice, the most unstable are damage caused by irradiation of ceramics with helium or hydrogen ions [19-25]. In this case, not only the formation of local heterogeneities and distortions of the structure is observed, but also due to the weak solubility and high mobility of helium and hydrogen in the structure, the formation of gas-filled inclusions is possible, leading to swelling and embrittlement of ceramics. However, the exact data of critical radiation doses causing irreversible, catastrophic damage to the crystal structure and properties of ceramics has not been established for aluminum nitride to date, which is the main motivation for such studies [26-30].

Based on the foregoing, the purpose of this work is to conduct research to determine the critical radiation doses at which the maximum effect of swelling and degradation of the near-surface layer of ceramics based on aluminum nitride is observed. Determination of the radiation damage kinetics, as well as the subsequent evolution of gas-filled inclusions in the case of irradiation with helium and hydrogen ions, was carried out by irradiating ceramic with helium and hydrogen ions in a wide dose range from 10^{15} ion/cm^2 to $1x10^{18}$ ion/cm^2.

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The choice of the upper threshold of irradiation is due to the literature data, according to which the most pronounced swelling processes for carbide and nitride ceramics are observed at doses above 10^{17} ion/cm^2.

1. Experimental part

Polycrystalline ceramic of aluminum nitride (AlN) with a hexagonal type of crystal lattice and a density of 3.26 g/cm^3 were selected as objects of study. This type of ceramic is a commercial material used as a base for insulating substrates, as well as a base for structural materials.

Radiation damage was simulated by two types of ions, helium and hydrogen (protons). Irradiation with helium ions (He^{2+}) was carried out on a DC-60 heavy ion accelerator (Nur-Sultan, Kazakhstan), the ion energy was 40 keV, the irradiation dose was 10^{15} – 10^{18} ion/cm^2. Irradiation with 1.5 MeV hydrogen ions (protons) was carried out at the UKP-2 accelerator (Almaty, Kazakhstan), the irradiation doses were 10^{15} – 5x10^{17} ion/cm^2. To avoid overheating of samples during irradiation and to initiate thermal annealing of defects as a result of accumulated heat, the samples were placed on water-cooling targets during irradiation, which allow the temperature of the samples to be maintained near room temperature.

Investigation of mechanical properties of ceramics before and after irradiation was carried out by determination of value of crack resistance, bending strength and impact toughness. The study of heat-conducting properties was carried out using a stationary method for measuring the absolute longitudinal heat flux, followed by determining the value of the thermal conductivity coefficient using formula (1):

$$\lambda = \frac{q\delta}{t_{c1} - t_{c2}}, \tag{1}$$

where q is the heat flux density, W/m^2; t_{c1} and t_{c2} are the temperature constants on the hot and cold sides of the wall, K; δ is the wall thickness, m; λ is the coefficient of thermal conductivity of the wall material, W/(m·K).

2. Results and discussion

Figure 1 shows the results of changing the value of the thermal conductivity coefficient depending on the type of ions and the radiation dose. The value of thermal conductivity characterizes the properties of materials to give off and conduct heat, which for structural materials is one of the important operating parameters that contribute to the stable removal of heat from the reactor zone. Moreover, in the event of a drop in this value, a decrease in thermal conductivity can lead to irreversible consequences, accompanied by overheating of the core. The change in the value of thermal conductivity under the action of irradiation is primarily associated with radiation damage caused by irradiation, which leads to the creation of additional defects, as well as regions of disorder, which leads to partial amorphization of the structure.

As a rule, the critical value of the decrease in thermal conductivity is a decrease by more than 20 % from the nominal value, which characterizes a large degradation of the material. In this case, in contrast to mechanical damage, the change in thermal conductivity can be more pronounced, which indicates changes at the crystalline level, as well as the formation of a large number of amorphous inclusions. As can be seen from the data presented, in the case of irradiation with hydrogen ions, even at maximum irradiation doses, the decrease in thermal conductivity coefficient does not exceed 10 %, which is within the permissible limits, and indicates the resistance of ceramic to irradiation by protons.

Another variation in the thermal conductivity is observed for samples irradiated with He^{2+} ions. The general nature of the changes can be divided into three characteristic regions, which will be hereinafter referred to as the I – irradiation dose region 10^{15} – 10^{17} ion/cm^2, the II – irradiation dose region 10^{17} – 5x10^{17} ion/cm^2, and the III – irradiation dose region 5x10^{17} – 10^{18} ion/cm^2. Region I is characterized by an insignificant decrease in thermal conductivity, comparable to the changes under irradiation with protons. Region II is characterized by a sharp decrease in thermal conductivity from 91 % to 74 % of the initial value, which indicates a sharp change in the structural properties of the irradiated ceramic. Region III is characterized by a decrease in thermal conductivity coefficient by 3-5 % in comparison with the changes characteristic of region II. Such a change for region III indicates the effect of the radiation damage accumulation and a decrease in the material degradation rate. In fact, according to previous studies, it has been established that the mechanism of helium swelling at doses above 3x10^{17} ion/cm^2 slows down, since the radiation damage accumulation obeys an exponential law, and at high radiation doses it reaches the so-called saturation plateau of radiation defects [31, 32]. In turn, a sharp drop in thermal conductivity at radiation doses of 10^{17} – 5x10^{17} ions/cm^2 indicates the formation of a cumulative effect of radiation-induced defects in the structure of the near-surface layer, as well as the formation of helium inclusions and bubbles that can cause partial amorphization of the near-surface layer, which leads to a decrease in heat removal and deterioration of the insulation properties of ceramic. The obtained thermal conductivity variation dependences indicate that this type of polycrystalline ceramic has rather high indicators of irradiation resistance with both proton and helium beams up to doses of 3-5x10^{17} ion/cm^2.

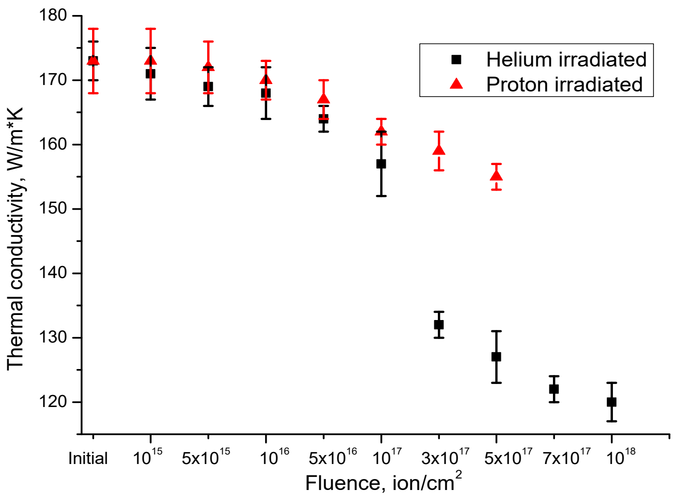

Fig.1. Graph of thermal conductivity coefficient change depending on type of ions and irradiation dose.

Important characteristics of the use of ceramics as structural materials are their mechanical and strength properties, the change in which under the influence of radiation can affect the operational characteristics, as well as the service life of these materials. One of these quantities characterizing the change in the materials strength is the crack resistance of the materials, which characterizes the pressure required to create a microcrack in the near-surface layer. Figure 2 shows a graph of the change in the crack resistance depending on the irradiation dose, which reflects the dynamics of crack resistance of ceramic as a result of the change in the concentration of defects formed by the irradiation. As can be seen, the change in crack resistance in the case of proton irradiation is two-step in nature, which is characterized by small changes at irradiation doses up to 10^{17} ion/cm^2 and a drop in crack resistance by 7-11 % for irradiation doses 3x10^{17} and 5x10^{17} ion/cm^2, respectively. In the case of irradiation with He^{2+} ions, the main changes occur at doses above 3x10^{17} ions/cm^2, and are characterized by a decrease in crack resistance by 20-30 % of the initial value. This behavior of crack resistance changes indicates a deterioration in the strength of the ceramic and a decrease in their cracking resistance as a result of external influences.

Deterioration of crack resistance of ceramic at high radiation doses is related to processes of accumulation of implanted helium, which, as shown earlier in works [33-35], due to its mobility, is able to form helium bubbles, the dimensions of which vary from 50 nm to several microns depending on the radiation dose. According to the general theory of the helium bubbles formation [35], the mobility of helium leads to the filling of voids formed as a result of deformation and crystal structure distortion as a result of radiation damage, as well as the subsequent evolution of point defects in the structure of the damaged layer. The filling of such voids with helium leads to the formation of additional stresses in the crystal lattice, which are capable of exerting pressure on the structure, thereby squeezing the deformed volume onto the surface, forming hillocks or gas-filled bubbles. A further increase in the concentration of implanted helium leads to an increase in the volume of the bubbles, and, consequently, to an increase in the stress in them, which ultimately leads to their rupture with the formation of craters or microcracks. This behavior is well described in [33–35], where the authors studied various mechanisms of the formation of gas-filled bubbles and blisters upon irradiation with helium ions, and their further evolution, leading to degradation of the near-surface layer. The destruction of the near-surface layer at irradiation doses above 3x10^{17} ions/cm^2 leads to a sharp decrease in the ceramic strength, as well as an increase in the likelihood of cracking as a result of external influences.

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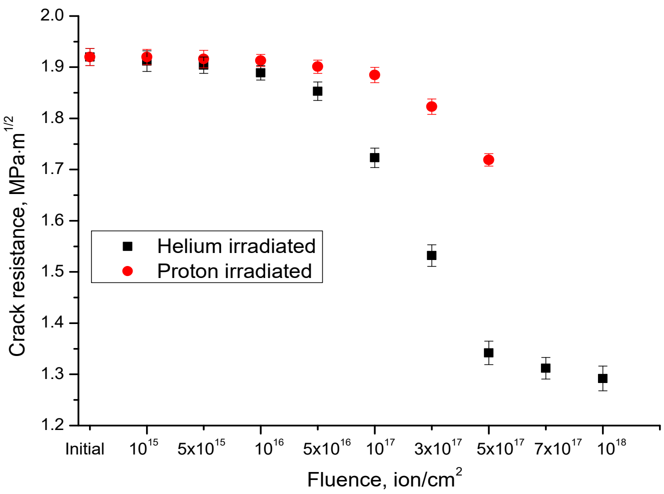
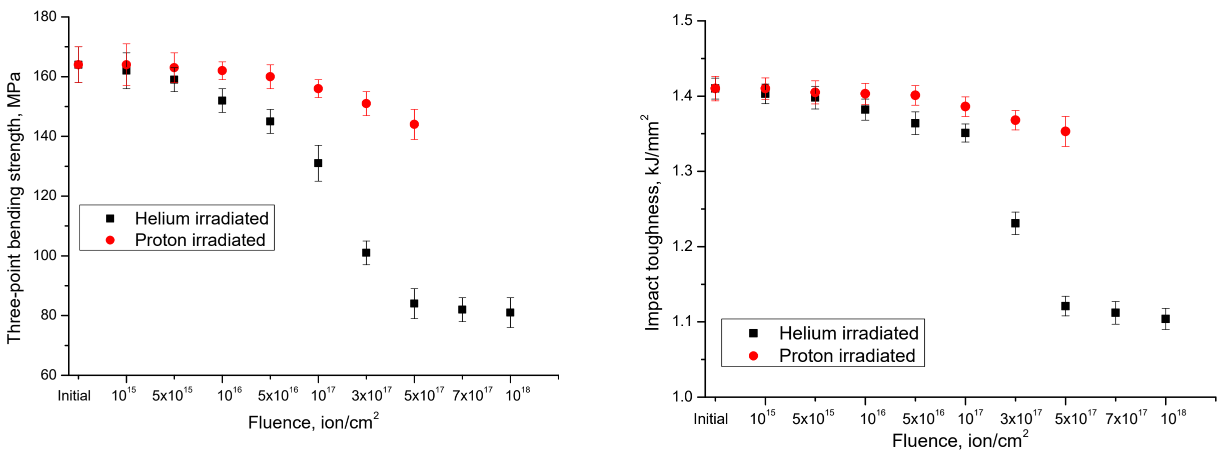


Fig.2. Graph of the change in crack resistance depending on the radiation dose

Figure 3 shows the results of changes in bending strength and impact strength of ceramic exposed to hydrogen and helium ions.



a) b)

Fig.3. a) Graph of bending strength change depending on irradiation dose and type of ions; b) Graph of impact strength change depending on irradiation dose

The general appearance of the change in these values is characteristic of such changes in the crack resistance of ceramic and indicates that, as in the case of crack resistance, irradiation with protons does not lead to a significant decrease in strength characteristics, while irradiation with He^{2+} ions at doses higher than $3x10^{17}$ ions/cm^2 leads to a sharp deterioration in strength characteristics.

Conclusion

In conclusion, we can summarize the results of the experimental work, which are as follows. First, the AlN polycrystalline ceramic under study showed good stability in maintaining mechanical and heat-conducting properties in the dose range of 10^{15}-10^{17} ion/cm^2 for both types of selected ions. This behavior indicates that this type of ceramic has a high degree of resistance to radiation damage arising from the interaction of ions and the subsequent formation of gas-filled inclusions. Secondly, in the case of irradiation with protons, the studied ceramic in the entire dose range showed a decrease in heat-conducting and strength characteristics within 5-10 %, which is good indicators of radiation resistance. At the same time, for samples irradiated with He^{2+} ions, the critical values of doses at which there is a decrease in heat-conducting and strength properties by more than 25-30% are doses of $3\text{-}5x10^{17}$ ions/cm^2, however, a further increase in irradiation doses does not lead to significant changes in the properties of ceramic, which indicates the radiation damage cumulative effect and a decrease in the material degradation rate.

The results obtained can later be used to expand the theory of radiation damage to nitride ceramic, as well as to predict and design high-temperature reactors of a new generation, based on which it is planned to use a new class of structural materials based on ceramics.

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