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STUDY OF THE INFLUENCE OF RADIATION-INDUCED DAMAGE ACCUMULATION DURING THE INTERACTION OF HEAVY Xe^{22+} IONS ON CHANGES IN THE THERMOPHYSICAL PARAMETERS OF ZIRCONIA CERAMICS

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The aim of this paper is to evaluate the influence of the processes of radiation-induced damage formation in the form of point defects, dislocations and vacancies, as well as their accumulation and the formation of locally disordered regions in the near-surface layer of zirconia ceramics under irradiation with heavy Xe^{22+} ions with an energy of 230 MeV, on the change in the thermophysical properties of ceramics. The choice of the ion type for irradiation is due to the possibilities of modeling radiation damage processes comparable to the impact of uranium nuclei fission fragments during nuclear reactions in nuclear fuel. The choice of materials for irradiation in the form of ZrO_2 ceramics is due to the prospects for their use as the main material for inert matrices of dispersed nuclear fuel for new generation reactors. This choice is due to the physicochemical, thermophysical and strength properties of ZrO_2 ceramics, which are more resistant than other types of oxide ceramics. During research, it was found that the formation of isolated locally heterogeneous regions at low irradiation fluences does not lead to significant changes in the thermophysical properties of the damaged ceramic layer. However, polymorphic transformations of the $t\text{-}ZrO_2 \rightarrow c\text{-}ZrO_2$ type, which occur at irradiation fluences above 10^{12} ion/cm^2, lead to a decrease in thermal conductivity and the appearance of heat losses associated with the disruption of the phonon heat transfer mechanisms in the damaged layer.

Keywords: zirconia ceramics, radiation defects, thermophysical properties, radiation damage, heavy ions, dispersed nuclear fuel

Introduction

One of the important areas of research in modern materials science is the study of the resistance of ceramic materials to various types of radiation exposure [1-3]. Interest in this area of research consists in the need to expand the database, as well as gain new knowledge about the processes of formation, further interaction and accumulation of radiation damage in ceramics - one of the promising materials for nuclear energy. It should be noted that in ceramic materials, in contrast to metals and alloys, the processes of accumulation and further evolution of radiation damage have significant differences, as well as a pronounced relationship with the dielectric nature of ceramics [4,5]. These differences primarily consist in the processes of accumulation of radiation-induced damage and defects, which, due to the dielectric nature of ceramics, are limited in the damaged volume of the material and have low mobility. Such a difference in comparison with metals and alloys leads to the likelihood of the formation of so-called latent tracks or amorphous regions in some types of ceramics, and can also lead to the initialization of polymorphic or phase transformations [6-8]. These types of structural changes in the case of prolonged irradiation, and as a result, the accumulation of defects, can have a significant negative effect on the strength and thermal parameters. At the same time, if in the case of strength properties, small changes in the damaged volume can be ignored, since they have an insignificant effect on the stability of ceramics, then in the case of thermophysical parameters, everything is more serious. Any loss of thermal conductivity of materials must be taken into account, since a decrease in heat removal from the system will not allow all the generated heat to be removed, which can lead to the appearance of areas of local overheating of the reactor core. In the case of dispersed nuclear fuel, which is based on the technology of placing fissile material in an inert matrix based on ceramics, these effects are the most dangerous. The appearance of local areas of overheating in a dispersed fuel can lead to an acceleration of its destruction processes, which will lead to negative consequences, and for some types of oxide ceramics, the appearance of such areas can lead to an acceleration of structural disorder processes [9-11].

One of the promising materials in this direction is zirconium dioxide, the combination of properties of which makes it one of the promising materials for use as a basis for inert matrices of dispersed nuclear fuel. However, despite all the prospects, these ceramics have a number of disadvantages, primarily associated with the processes of polymorphic transformations, which can be initiated either as a result of mechanical impacts or during the accumulation of radiation damage [7,12-14]. In this regard, during evaluation of the applicability of these ceramics as materials for inert matrices of dispersed nuclear fuel, it is necessary to take into account the probability of occurrence of polymorphic transformation processes that can lead to a change in the properties of ceramics. Based on this, the main goal of this work is to study the effect of irradiation with heavy ions, and the processes of polymorphic transformations caused by them, on the change in the heat-conducting properties of ceramics, as well as to determine the dynamics of changes in the value of heat losses.

1. Experimental part

Polycrystalline two-phase ceramics based on zirconium dioxide (ZrO_2) with the dominant tetragonal phase of the P42/nmc(137) spatial syngony were chosen as the objects of study. The content of this phase is more than 80 %. The samples were irradiated at a DC – 60 heavy ion accelerator (Institute of Nuclear Physics, Astana, Kazakhstan). Heavy Xe^{22+} ions with a total ion energy of 230 MeV (1.75 MeV/nucleon) were chosen as projectile ions. Irradiation was carried out at room temperature, which was maintained by placing the target on a special water-cooled target holder. Irradiation fluences were chosen from 10^{10} to 10^{15} ion/cm^2, while the range of 10^{11} – 10^{13} ion/cm^2 was performed with a small step to determine most accurately the stage of formation of overlapping regions of locally disordered regions in the structure of the damaged layer, the appearance of which has a significant effect on the processes of structural disorder and polymorphic transformations of the t-ZrO_2 → c-ZrO_2 type. The polymorphic transformation processes of the t-ZrO_2 → c- ZrO_2 type, accompanied by rearrangement of the crystal structure and displacement of the tetragonal phase with subsequent dominance of the cubic phase, according to the data of [7,13,14], occur in the fluence range of 10^{12}–10^{13} ion/cm^2 and are accompanied by structural changes.

To determine the thermophysical parameters, a standard method for measuring the longitudinal heat flux was used, which makes it possible to determine the change in the thermal conductivity coefficient and the amount of heat loss that occurs when the properties of the material subjected to irradiation change. For the measurement, a KIT-800 thermal conductivity device (Teplofon, Moscow, Russia) was used, which makes it possible to determine changes in the thermal conductivity coefficient in the range from 100 to 800ºC. For measurements, samples were used before and after irradiation, with geometric dimensions of 8 x 8 mm and a thickness of 50 μm. Based on the measured values of the thermal conductivity coefficient, the value of heat losses was calculated, reflecting changes in thermophysical parameters in samples subjected to irradiation.

2. Results and discussion

As shown in several works [7,12-14], irradiation of ceramics with irradiation fluences above 10^{12} – 10^{13} ion/cm^2 with heavy ZrO_2 ions leads to the initialization of polymorphic transformations of the t-ZrO_2 → c-ZrO_2 type, which consist in a change in the phase composition and subsequent deformation of the crystal lattice, as well as the accumulation of radiation-induced damage. In this case, as is known, the polymorphic transformation processes are associated with a destructive change in volume as a result of the transformation of a tetragonal structure into a cubic one, accompanied by a change in volume and density. A change in density during the action of deformation distortions leads to the appearance of porous inclusions, which have a negative impact not only on the structural properties, but also on the thermophysical parameters. So, for example, as was shown in [15], the change in the thermophysical properties of ceramics is greatly influenced by the grain size and porosity, which can change both under varying conditions and the choice of the method for obtaining ceramics, and in the case of external influences. At the same time, the occurrence of heterogeneities in the composition of ceramics also plays a very important role in the change in thermal conductivity, as well as the occurrence of heat losses in ceramics [15].

According to the assessment of the phase composition of ZrO_2 ceramics in the initial state, these ceramics are a mixture of two phases: the dominant tetragonal ZrO_2 phase with the P42/nmc(137) spatial system and the cubic ZrO_2 phase with the Fm-3m(225) spatial system. The ratio of tetragonal (t-ZrO_2) and cubic (c-ZrO_2) phases is close to 4:1 (80%:20%). At the same time, the structural ordering degree in the initial state of ceramics is more than 97 %, which indicates a low content of amorphous or disordered inclusions, the presence of which is due to the ceramics manufacturing technology.

According to the data on changes in the phase composition depending on the irradiation fluence, the following was established. In the range of irradiation fluences of 10^{10} – 10^{12} ion/cm^2, no changes in the ratio of the t-ZrO_2 and c-ZrO_2 phases were observed, which indicates that the polymorphic transformation processes do not occur at these fluences. This absence of polymorphic transformations in this range of irradiation fluences can be explained as follows. At irradiation fluences below 10^{12} ion/cm^2, the interaction of incident ions with the ceramic structure occurs in local isolated regions along the ion motion trajectory in the material. Moreover, the diameter of these areas, according to the calculated estimates, is no more than 5 – 10 nm, which, at low irradiation fluences, indicates a low probability of the formation of overlapping of these damaged areas from two incident ions or the hit of two ions in one place sequentially. In this case, structural changes occur in locally isolated regions, and the accumulation of defective inclusions occurs very slowly in view of the fact that most of the defects formed in these regions annihilate among themselves in very short time intervals. During analysis of the structural changes and swelling of the crystal lattice volume depending on the irradiation fluence presented in Figure 1, it can be seen that at given irradiation fluences (10^{10} – 10^{12} ion/cm^2), the main changes occur when the fluence increases above 5 × 10^{11} ion/cm^2, for which the probability of overlapping local defective regions approaches 1, and the average distance between isolated regions becomes less than 50 – 100 nm.

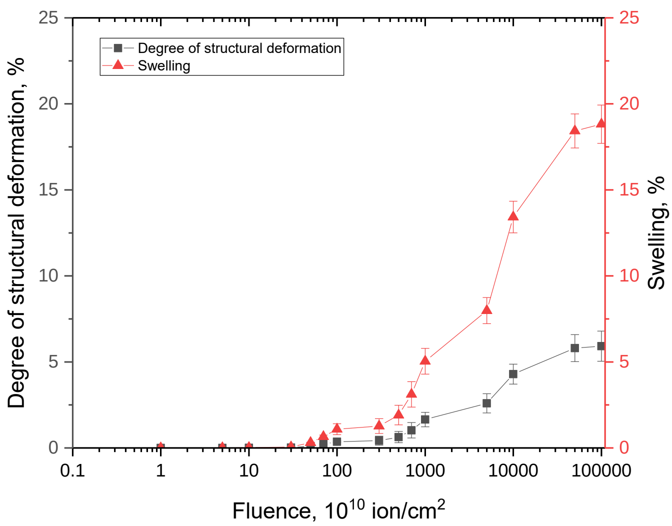

Fig.1. Results of deformation contributions in the damaged layer in ZrO_2 ceramics under heavy ion irradiation

As can be seen from the data presented, the main changes in the crystal structure deformation occur at fluences above 10^{12} ion/cm^2, for which, according to the phase analysis, the initialization of the polymorphic transformation processes of the t-ZrO_2 → c-ZrO_2 type is observed, which ends with the complete displacement of the tetragonal phase and the dominance of the cubic phase. At the same time, according to the assessment of deformation contributions, polymorphic transformations of t-ZrO_2 → c-ZrO_2 lead to a sharp increase in volumetric swelling, which is due to destructive changes associated with phase transformations. It should also be noted that at fluences above 10^{14} ion/cm^2, there is a noticeable decrease in the rate of swelling and destructive deformation of the crystal structure in the damaged layer, which may be due to the effects of saturation of the damaged layer with defects at high irradiation fluences.

As is known, deformation distortions and changes in the crystal lattice volume led to a change in the density and an increase in the porosity of ceramics. At the same time, according to the data presented in Figure 1, the processes of polymorphic transformations lead to a sharp increase in volumetric swelling associated with destructive distortion of the crystal lattice during the transformation of the tetragonal phase into a cubic one. Figure 2 shows the dependences of changes in the density and porosity of ceramics, calculated on the basis of volumetric changes in the crystal lattice with increasing fluence. The general view of the presented results in Figure 2 characterizes the change in the damaged layer associated with its deformation, change in density and the formation of porous inclusions. At the same time, according to the data presented, the main changes in density and, as a consequence, the formation of porous inclusions, occur during the initialization of the polymorphic transformation processes, accompanied by the transformation of the tetragonal phase into a cubic one, followed by a destructive increase in the crystal lattice volume. At the same time, it should be noted that at high irradiation fluences of 10^{13} – 10^{14} ions/cm^2, a decrease in changes in density destruction is observed, which indicates the effect of accumulation of radiation distortions and their influence on structural changes, which was also noted when interpreting the data on changes in deformation contributions.

A change in the density and porosity of ceramics, as well as its deformation, should affect the change in thermophysical parameters, the change of which is very important in assessing the applicability of these ceramics as structural materials for the nuclear industry. At the same time, volumetric changes in the damaged layer can have a different effect on the mechanism of heat transfer, since, unlike metals, for dielectric ceramics, the main mechanisms of heat transfer are phonon mechanisms. Figure 3 shows the dependence of the change in the thermal conductivity coefficient (K_{eff}) on the irradiation fluence, which characterizes the change in the thermophysical properties of ceramics exposed to irradiation with heavy ions.

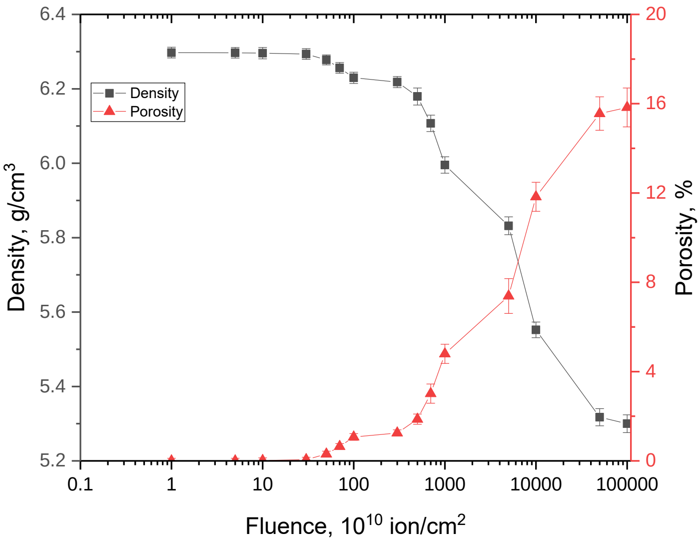

Fig.2. Results of changes in the density and porosity of ZrO_2 ceramics depending on the irradiation fluence

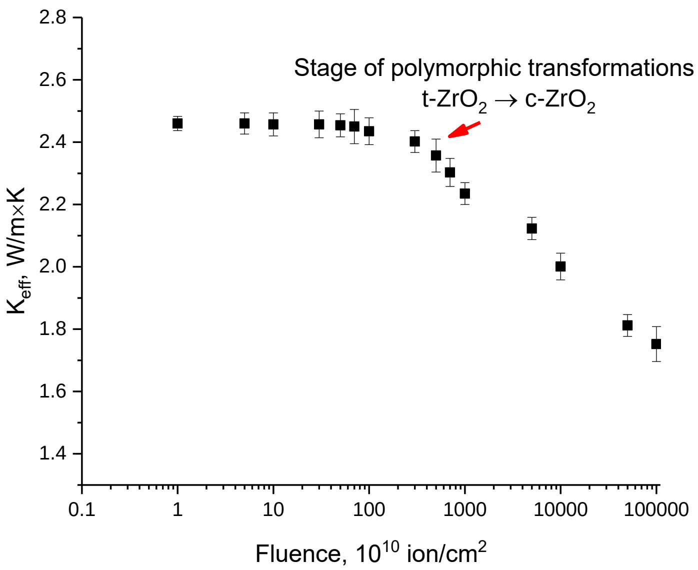

Fig.3. Results of the change in the thermal conductivity coefficient from the irradiation fluence

The general view of the presented changes in the thermal conductivity coefficient has several distinct areas that depend on the irradiation fluence and structural changes associated with the accumulation of radiation damage. At low irradiation fluences (below 10^{12} ion/cm^2), practically no changes in K_{eff} are observed, which indicates that structural distortions resulting from the formation of locally isolated regions of structural damage along the ion trajectory in the surface layer do not affect the heat transfer mechanisms associated with phonon heat transfer. At the same time, an increase in deformation distortions at fluences above 5×10^{11} ion/cm^2 leads to a slight decrease in the thermal conductivity coefficient, fitting into the measurement error, which indicates that deformation distortions resulting from the occurrence of areas of overlap of local defective fractions create obstacles to heat transfer in the form of highly disordered inclusions.

The polymorphic transformation processes of the t-ZrO_2 → c-ZrO_2 type, which occur at fluences above 10^{12} ion/cm^2, lead to a sharp drop in thermal conductivity, which indicates a change in the heat transfer mechanisms, as well as a decrease in thermophysical parameters. A further increase in the irradiation fluence, which leads to a destructive change in the volume of the crystal lattice and the density of ceramics, leads to a decrease in thermal conductivity, as well as an increase in heat losses. Figure 4 shows dependences of the change in the value of heat losses on the porosity of ceramics, which increases as a result of polymorphic transformations and destructive changes in structural parameters.

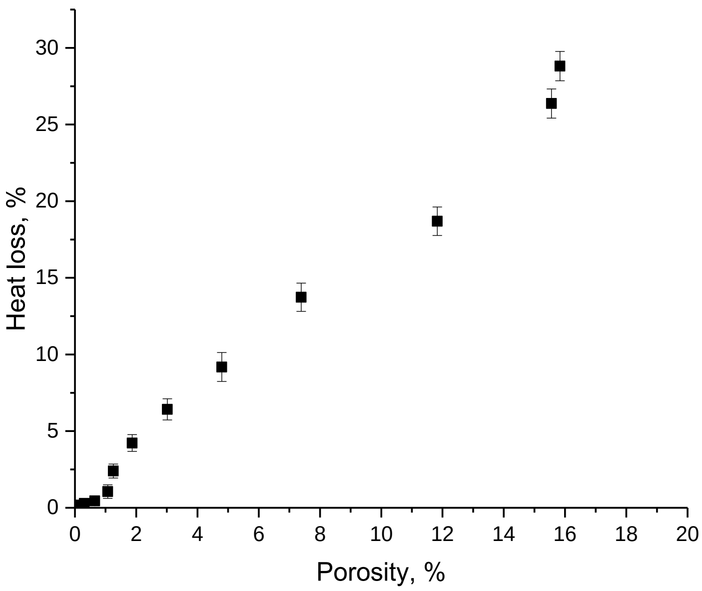

Fig.4. Results of a comparative analysis of changes in the value of heat losses from the porosity of ceramics subjected to irradiation

As can be seen from the data presented, an increase in the porosity of ceramics above 2% leads to an increase in heat losses, while the change in the value of heat losses due to porosity is not linear and has a pronounced increase at maximum irradiation fluences. Such a change in the value of heat losses at maximum irradiation fluences (5×10^{13} – 10^{14} ion/cm^2) can be due to the fact that, at these fluences, highly defective regions with a highly disordered structure and amorphous-like inclusions are formed, the appearance of which has a negative effect on the heat transfer mechanisms. Figure 5 shows the dependences of the change in the thermal conductivity and thermal diffusivity on the density of ceramics, which changes as a result of the damaged layer destruction.

As can be seen from the data presented, a decrease in the density of ceramics associated with processes of destructive volume change, as well as processes of polymorphic transformations, leads to a loss of thermal conductivity and a decrease in thermal diffusivity, which affects the thermophysical properties of ceramics. At the same time, the maximum decrease in values is more than 25 % of the initial value, which indicates the destructive effect radiation exposure on the mechanisms of heat transfer and heat transfer.

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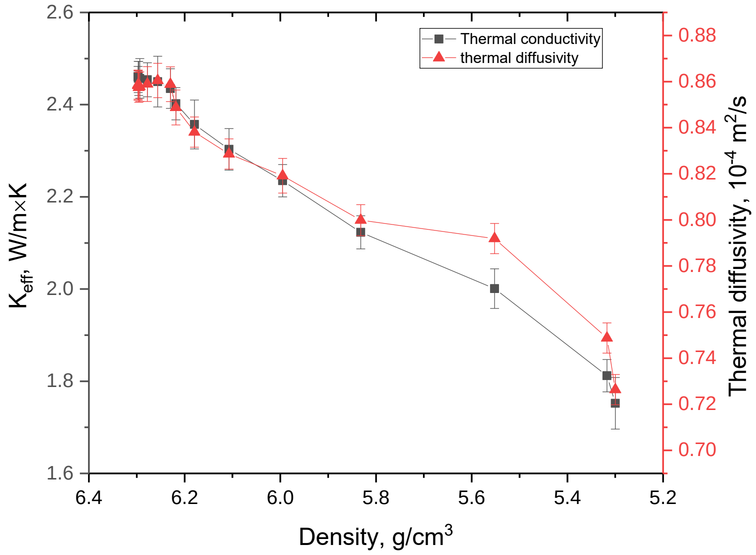

Fig.5. Dependence of changes in thermophysical parameters on the density of ceramics

Conclusion

The paper presents the results of estimating the change in thermophysical parameters in ZrO_2 ceramics subjected to irradiation with heavy Xe^{22+} ions in the range of irradiation fluences corresponding to the formation of single local defect regions and the overlapping of locally heterogeneous regions causing polymorphic transformation processes of the t-ZrO_2 → c-ZrO_2 type. During the studies, it was found that the main contribution to the structural disorder of the damaged layer of ceramics at fluences above 10^{12} ion/cm^2 is made by volumetric swelling associated with a destructive change in the structure during polymorphic transformations. According to the assessment of changes in thermophysical parameters, it was found that the main changes associated with an increase in heat losses occur during the initialization of polymorphic transformation processes. At the same time, deformation processes at low irradiation fluences do not have a significant effect on the decrease in thermal conductivity. An analysis of the change in thermophysical parameters depending on the change in structural properties and porosity showed that the main contribution to the decrease in thermal conductivity and thermal diffusivity is made by a decrease in the density of ceramics, and the associated change in porosity.

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