

The effect of residual mechanical stresses and vacancy defects on the diffusion expansion of the damaged layer during irradiation of BeO ceramics

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ABSTRACT

The paper presents the results of a study on the application of Raman and UV spectroscopy methods to determine the structural damage kinetics in the near-surface layer of BeO ceramics caused by high-dose irradiation with He²⁺ ions. Interest in this type of ceramics is due to the combination of its structural and thermophysical parameters, making these ceramics one of the promising classes of materials for microelectronics and structural materials for nuclear reactors, with the possibility of operation in conditions of heightened radiation background. According to the conducted studies, it was established that with the irradiation fluence growth, changes in the nature of deformation structural distortions associated with the accumulation of residual mechanical stresses of tensile and compressive types are observed. At irradiation fluences of 10^{16} – 5×10^{16} He²⁺/cm², tensile stresses play a dominant role in structural distortions, while the value of compressive stresses at fluence growth makes up a small share in the overall nature of the deformations. Moreover, an elevation in the irradiation fluence above 5×10^{16} He²⁺/cm² leads to a rise in the concentration of defects caused by the formation of oxygen vacancies, as well as He-V_O type complexes, the presence of which is indicated by the halo intensity growth in the Raman spectra, as well as a change in the intensity of the absorption bands. Analysis of changes in thermophysical parameters revealed that a rise in structural distortions associated with the accumulation of complex defects results in thermal conductivity reduction and a deterioration in heat transfer processes associated with partial amorphization of the damaged layer. Moreover, the established direct relationship between the value of residual mechanical stresses and the degradation of thermal conductivity indicates the cumulative effect of destructive changes caused by irradiation, as well as the influence of diffusion mechanisms on the damaged layer thickness growth.

1. Introduction

The use of ceramic materials based on beryllium oxide as one of the most promising materials with good thermal conductivity combined with high insulation properties in nuclear and thermonuclear energy, as well as microelectronics and optical materials, is one of the currently discussed research topics [1–4]. At the same time, in most cases, the use of ceramic materials occurs under extreme conditions, in particular, external mechanical impacts, increased radiation background, as well as thermal effects, which together imposes certain restrictions not only on

the expansion of the potential for the use of these ceramics, but also the need to take into account possible processes of degradation of the properties of ceramics due to these changes [5–7]. Unlike alloys and steels, the use of which has certain limitations in the case of high temperatures, ceramic materials can be considered as candidate materials for operation in extreme conditions, especially in the case of high radiation doses, which lead to the accumulation of large concentrations of structural distortions, as well as the formation of amorphous formations, the presence of which can negatively affect the strength and thermal parameters of ceramics [8–11].

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The main objective of this study is to comprehensively investigate the relationship between changes in structural, optical and thermal parameters caused by the accumulation of radiation damage in the near-surface layer of BeO ceramics, which are one of the promising materials in nuclear and thermonuclear energy, as well as protective coatings for protecting telescope glass and insulating materials [12–16]. The choice of ceramics based on beryllium oxide as objects of study is due to its combination of structural, optical and thermal-physical parameters, which allow it to be considered as one of the candidate materials capable of withstanding high doses of radiation [17,18], as well as being used in modes with extreme operating conditions. Moreover, in several works [19–21], it was shown that irradiation of ceramics with heavy BeO ions leads to the initiation of structural disordering processes associated with ionization processes, as well as athermal effects resulting in destabilization of the near-surface layer. In the case of high-dose irradiation with He²⁺ ions, a number of studies [22–25] established critical doses at which embrittlement of the surface layer was observed due to the formation of gas-filled inclusions; however, the mechanisms of formation, as well as the diffusion of implanted ions deep into the material and the expansion of the damaged layer, were not studied.

In this study, the main emphasis on the study of structural features and their changes caused by irradiation is made on the use of Raman and UV spectroscopy methods, the use of which in combination allows us to determine the main types of structural defects that arise during irradiation, as well as their concentration, and in the case of using Raman piezospectroscopy methods, to determine the type of deformation distortions caused by the accumulation of residual stresses in the near-surface damaged layer. It should be noted that the use of a combination of Raman and UV spectroscopy methods allows us to determine with high accuracy not only the kinetics of structural changes with increasing radiation dose, but also to establish their type associated with the formation of vacancy defects, as well as complex defects in the structure of the damaged layer [26,27]. However, in the case of low-energy ions, the use of X-ray diffraction methods is not always applicable due to the small thickness of the damaged layer, the measurement of which requires special conditions during radiography or thinning of samples to thicknesses characteristic of the damaged layer. In this case, the Raman spectroscopy method allows us to determine structural changes associated with the distortion of chemical and crystalline bonds at a shallow depth, characteristic of the damaged layer. The use of the UV spectroscopy method in assessing optical transmission spectra and induced absorption spectra built on their comparison allows us to establish structural distortions and their type, caused by the structural damage accumulation caused by irradiation. In this case, the shifts of the fundamental absorption edge make it possible to determine indirectly the kinetics of changes in the electron density distribution, a change in which in the case of dielectric ceramics can lead to the occurrence of breakdowns, which is one of the critical parameters for microelectronic devices.

2. Materials and methods of research

The samples chosen for study were BeO ceramics with a wurtzite-type structure (hexagonal type of crystal lattice), possessing a polycrystalline structure and a structural ordering degree of about 97.5 %. These samples were commercial ceramics. The degree of structural ordering was determined on the basis of X-ray structural analysis data by determining changes in the crystal lattice parameters and their deviations from reference values.

Simulation of radiation damage associated with the accumulation of implanted ions, as well as the processes of interaction of incident ions with the crystalline structure of the near-surface layer, was carried out at the DC-60 heavy ion accelerator (Institute of Nuclear Physics of the Ministry of Energy of the Republic of Kazakhstan, Kazakhstan). For the simulation, He²⁺ ions with an energy of 40 keV, the value of which allows the creation of defects that initiate structural distortions at a depth

of about 400–450 nm in the near-surface layer, were used. The choice of irradiation fluence from 10^{16} to 3×10^{17} He²⁺/cm² [22–24], the choice of which is determined by the possibilities of initializing structural distortions with deep overlap of defective areas formed along the trajectory of ion movement in the target material during irradiation. The maximum ion range in the near-surface layer of ceramics was estimated using the SRIM Pro 2013 software code. The calculations used a model that takes into account the effects of cascade mixing and multiple scattering, which made it possible to determine not only the maximum ion range, but also their deviation from the initial trajectory. Irradiation was carried out at room temperature, under vacuum conditions, the target temperature was controlled using a special target holder with water cooling, the use of which made it possible to maintain the target temperature within 25 ± 3 °C (control was carried out using thermocouples placed both on the target holder and on the samples).

The structural distortions caused by irradiation were determined by analyzing the Raman spectra of the studied samples taken using an EnSpectr M532 Raman spectrometer (Spectr-M LLC, Chernogolovka, Russia). Raman spectra were taken in two modes. The first shooting mode was performed from the surface of the samples in order to determine the general trend and kinetics of structural distortions associated with the accumulation of residual mechanical stresses and the formation of amorphous inclusions, which are indicated by the broadening of spectral lines and a decrease in intensity. The second shooting mode was carried out from the side cleavages of the samples with a step of 50 nm along the trajectory of ion penetration into the material in order to determine the diffusion mechanisms of migration of implanted helium.

The study of the damaged layer degradation mechanisms, and the assessment of the diffusion effects associated with the migration of implanted helium in the near-surface layer, causing destruction in the crystal lattice along the movement trajectory, was carried out using Raman spectra recording in depth on lateral cleavages. An assessment of the residual stress values, as well as the FWHM values by depth, made it possible to determine which type of deformation predominates in the structure of the damaged layer. In this case, the magnitude of residual stresses, expressed in the shift of spectral lines, is caused by deformation distortions in the structure, while an increase in the FWHM value indicates amorphization of the crystal structure as a result of its disordering.

The study of optical characteristics, the change of which reflects the accumulation of structural damage in the damaged layer, as well as the change in electron density associated with the accumulation of structural distortions, was carried out by evaluating the transmission spectra of the studied ceramics. These spectra were obtained using the UV spectroscopy method implemented on a SPECORD 200/210/250 PLUS spectrophotometer (Analytik Jena, Jena, Germany). The spectra were recorded in the range from 300 to 800 nm, with a step of 1 nm.

The determination of thermophysical parameters was carried out using the method of measuring the thermal conductivity coefficient by depth in order to determine the effect of the damaged layer crystalline structure degradation on the thermophysical parameters.

3. Results and discussion

Fig. 1 demonstrates the results of Raman spectroscopy of the studied BeO ceramic samples irradiated with He²⁺ ions. According to the data presented in Fig. 1a, it is evident that the spectra of samples irradiated with ions with a fluence of 10^{16} He²⁺/cm² contain a strong background characteristic of destructive changes in the damaged layer, while for other irradiation fluence values, broad peaks appear in the regions from 1000 to 1700 cm⁻¹, as well as from 2400 to 3300 cm⁻¹. Moreover, 4 peaks are clearly distinguishable in the spectra: at 337, 682, 723 and 1090 cm⁻¹, related to the modes E₂, A₁(TO), E₁(TO) and E₁(LO) of beryllium oxide of symmetry group C_{6v}, respectively.

Analysis of the change in the intensity of spectral lines that appear at

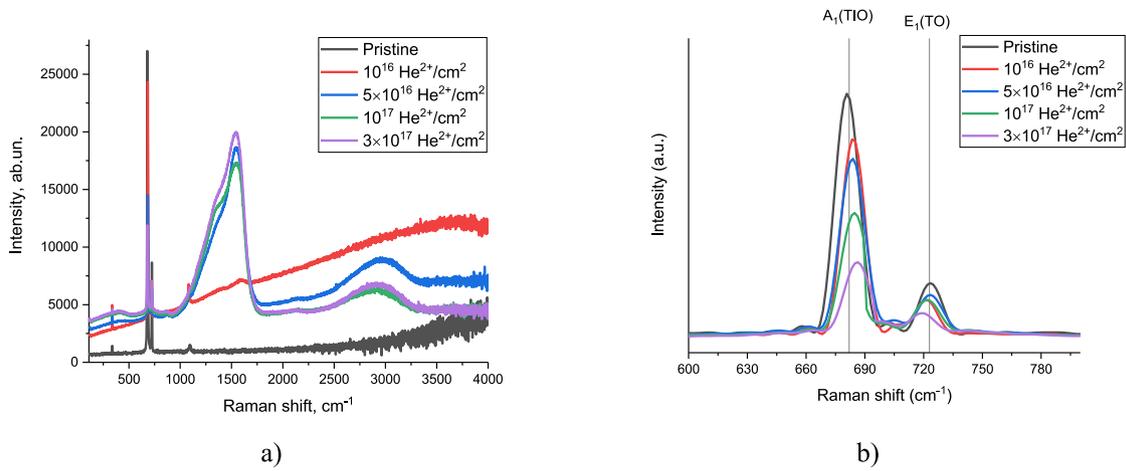


Fig. 1. a) General view of the presented Raman spectra depending on the irradiation fluence, reflecting general changes associated with deformation distortions, partial amorphization, and the accumulation of complex defects in the structure; b) Detailed representation of the change in position and intensity of the main spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ depending on the irradiation fluence, reflecting the destructive effect of irradiation.

an irradiation fluence of $5 \times 10^{16} \text{ He}^{2+}/\text{cm}^2$ and higher in the range of $1000\text{--}1700 \text{ cm}^{-1}$ and $2500\text{--}3200 \text{ cm}^{-1}$ indicates the formation of complex defects of the He-V_O type in the structure, the concentration of which increases in the damaged layer as irradiation fluence grows. It is important to highlight that at a low irradiation fluence of $10^{16} \text{ He}^{2+}/\text{cm}^2$, the presence of this band was not detected, which indicates that at this irradiation fluence, structural changes are caused by structural distortions caused by the formation of tensile and compressive residual mechanical stresses in the damaged layer structure.

A detailed analysis of the position and intensity of the spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ contingent upon the value of atomic displacements, presented in Fig. 1b, reflects the characteristic changes in the spectral lines associated with the accumulation of structural distortions, alongside partial amorphization, which is evidenced by a decrease in the intensity of the spectral lines, as well as their broadening (changes in the FWHM value). The observed alterations in the value of the shift of the Raman lines for the two main peaks relative to the initial position of the maximum for unirradiated samples depending on the value of the atomic displacements indicate a different nature of the deformation distortions of the crystal structure of the damaged layer. Moreover, in addition to differences in the type of deformation distortions observed

for chemical bonds, the change in the magnitude of the Raman peak shifts at one irradiation fluence indicates a difference in the value of tensile and compressive stresses in the structure of ceramics. As is evident from the data presented, the most pronounced changes associated with a reduction in line intensities are observed at a fluence of $10^{17} \text{ He}^{2+}/\text{cm}^2$ and higher. This indicates that at these fluences in the structure, in addition to structural deformation distortions, the formation of amorphous inclusions or disordered regions, the presence of which is associated with the formation of complex defects and their agglomeration, is also observed.

The assessment results of the shift in the positions of the maxima of the main spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ depending on the value of the atomic shifts (irradiation fluence) are presented in Fig. 2a. The presented data reflect the nature of deformation distortions arising in the structure of the damaged ceramic layer when the irradiation conditions change, while, as can be seen from the results of the presented analysis, the nature of the displacement has not only a difference in the type of displacement, but also in its trend, which indicates differences in the accumulation mechanisms of structural distortions of chemical bonds during the damage accumulation.

The analysis of the change in the FWHM value of the spectral lines

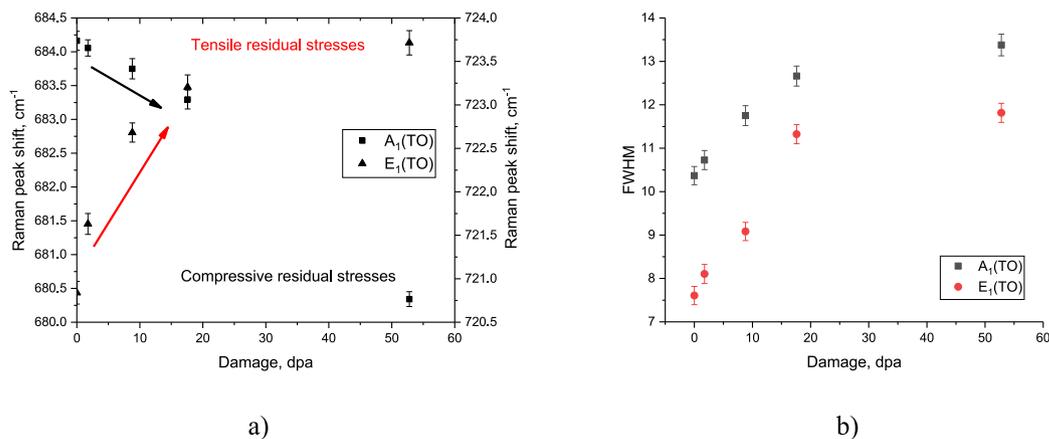


Fig. 2. a) Results of the dependence of the displacement of the position of the spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ in dependence, reflecting the accumulation of deformation distortions of the tensile and compressive type in the damaged layer structure; b) Results of changes in the FWHM values of the main spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ depending on the atomic displacement value, reflecting partial amorphization of the crystal structure.

$A_1(\text{TIO})$ and $E_1(\text{TO})$ contingent upon the atomic displacement value is presented in Fig. 2b. These changes reflect the accumulation degree of amorphous inclusions associated with the formation of complex He- V_O defects in the damaged layer structure. As can be seen from the data presented, the change in the trends of the FWHM values of the spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ has a similar character, which indicates deformation distortion of the crystal structure and the rupture of chemical and crystalline bonds caused by the accumulation of radiation damage. Moreover, the broadening of the reflexes, with a decrease in the intensity of the spectral lines (see data in Fig. 1b) indicates the destruction of the damaged layer, which is most pronounced at high irradiation fluence values. Also, from the analysis of the comparison of changes in the shifts of Raman lines and the FWHM values of the spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$, it can be concluded that the processes of deformation distortion caused by the formation of residual mechanical stresses of a tensile and compressive nature, as well as the effects of disordering caused by partial amorphization and loss of short-range order of the crystal structure make an equally probable contribution to the degradation of the damaged layer caused by irradiation. The change in the type of deformation distortions with increasing irradiation fluence in this case is due to several factors. Firstly, in the case of high-dose irradiation, the accumulation and subsequent agglomeration of implanted ions leads to the filling of voids, which has an additional deformation distortion on the structure. Secondly, the accumulation of structural distortions in the case of high-dose irradiation results in disordering of the crystalline structure, which in turn leads to more pronounced structural changes and subsequent embrittlement of the damaged layer.

The evaluation results of the value of residual mechanical stresses determined for two spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ depending on the atomic displacement value are presented in Fig. 3a. According to the assessment of the shifts of the Raman spectra, it was found that the shift of the $A_1(\text{TIO})$ maximum is characteristic of the accumulation of residual mechanical stresses of the compressive type, while the changes in the $E_1(\text{TO})$ spectral line are associated with deformation distortions caused by the accumulation of residual mechanical stresses of the tensile type. In this case, at low irradiation fluences of 10^{16} – 5×10^{16} $\text{He}^{2+}/\text{cm}^2$, the change in the value of residual mechanical stresses is of exponential nature, while in the case of high irradiation fluences (10^{17} – 3×10^{17} $\text{He}^{2+}/\text{cm}^2$), a slowdown in the trend of mechanical residual tensile stress growth was observed. In the case of compressive residual stresses at fluences of 10^{16} – 10^{17} $\text{He}^{2+}/\text{cm}^2$, the change in value is minimal, while for the maximum irradiation fluence of 3×10^{17} $\text{He}^{2+}/\text{cm}^2$, the value of

residual mechanical stresses is close to the value of residual mechanical stresses of the tensile type.

Fig. 4 reveals the assessment results of the change in the values of residual mechanical stresses and the FWHM values of the spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ in the damaged layer along the ion trajectory. The data were obtained by recording Raman spectra on the side cleavages of the samples in order to determine the mechanisms of destruction of the damaged layer caused by the accumulation of deformation stresses, as well as the concentration dependences of defective inclusions of the He- V_O type. These dependencies were also obtained in order to establish the relationship between the observed changes in the structural features of the damaged layer, associated with deformation stresses and the formation of disordered regions, and the thickness of the damaged layer, the changes of which in this case can be caused by diffusion processes that occur during high-dose irradiation [28,29]. For example, in the work [30] it was shown that diffusion processes have a clearly expressed dependence on the energy and flux density of the radiation, and are most pronounced with high-dose irradiation (about 50–100 dpa), which must be taken into account when designing the potential for using structural materials, and also not to exclude the effect of diffusion expansion of the thickness of the damaged layer in experiments on modeling radiation damage using ion irradiation. In this case, the use of Raman piezo-spectroscopy methods in the analysis of lateral cleavages not only makes it possible to evaluate the structural changes along the ion trajectory, but also to determine the thickness of the damaged layer at which deviations from the original values are observed.

The comparative analysis results of changes in the values of residual mechanical stresses and FWHM values in samples along the trajectory of ion movement in the damaged layer showed that an elevation in the irradiation fluence not only leads to a rise in the concentration of residual stresses, expressed in a growth in the values that determine them, but also, in the case of large fluences, to the damaged layer thickness growth, caused by diffusion processes. According to the data presented, an elevation in the irradiation fluence leads to a shift in the depth of deformation distortions of the crystal structure, as evidenced by changes in the values of residual mechanical stresses and FWHM observed at a depth exceeding the ion range in the material (on the graphs, this boundary is marked with a dotted red line). Moreover, alterations in the values of residual mechanical stresses in the depth of the sample at low irradiation fluences are most pronounced for $A_1(\text{TIO})$, while changes for $E_1(\text{TO})$ in depth are observed only in the case of high-dose irradiation (10^{17} – 3×10^{17} $\text{He}^{2+}/\text{cm}^2$). Such changes indicate that deformation distortions in the structure of the damaged layer can not only

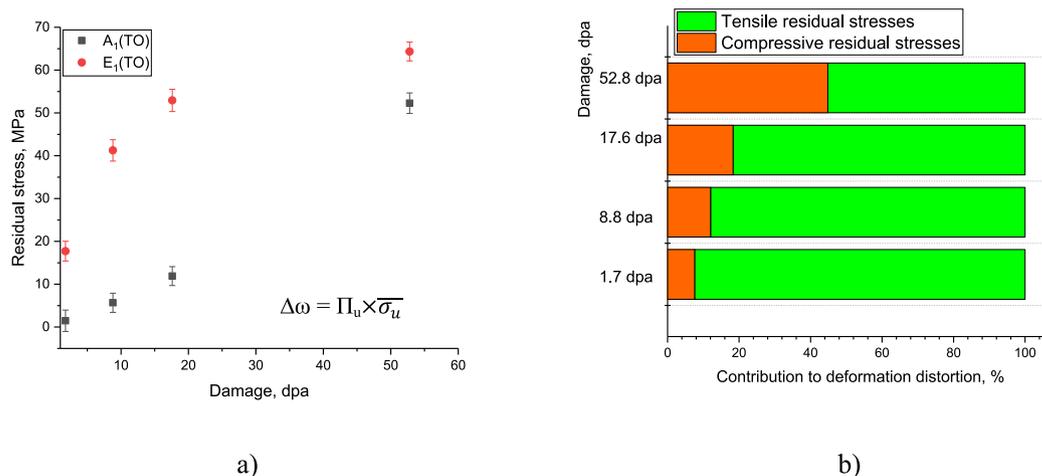


Fig. 3. a) Assessment results of the value of residual mechanical stresses in the damaged layer for the main spectral lines $A_1(\text{TIO})$ and $E_1(\text{TO})$ depending on the atomic displacement value; b) Comparison results of contributions of tensile and compressive residual stresses in the structure of the damaged layer, reflecting the kinetics of deformation distortions of the crystal lattice at radiation dose growth.

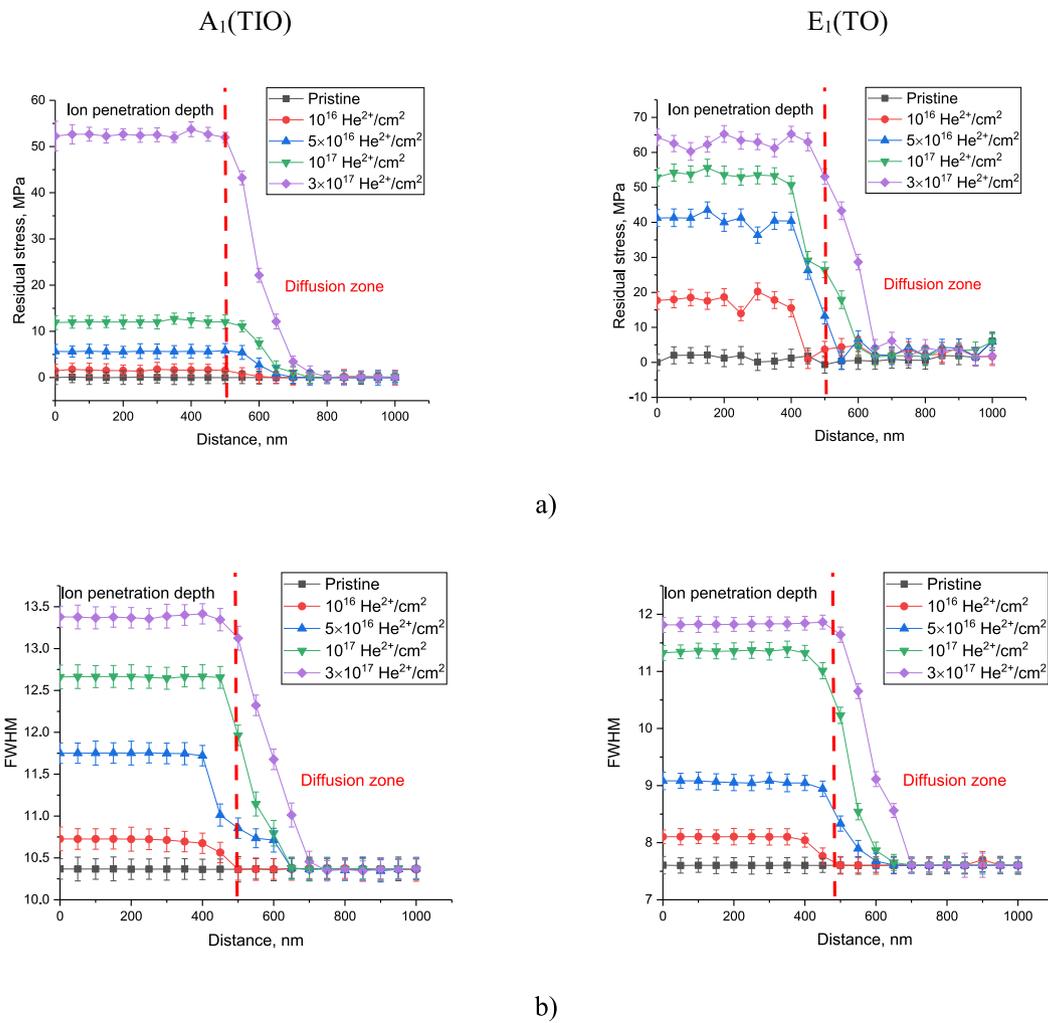


Fig. 4. Assessment results of changes in the main values of Raman spectra, reflecting changes in the depth of the damaged layer (The dotted line in the figure marks the boundary of the maximum path length of ions in the surface layer, according to the simulation results.): a) Results of changes in the values of residual mechanical stresses; b) Results of changes in the FWHM value characterizing partial amorphization of the damaged layer.

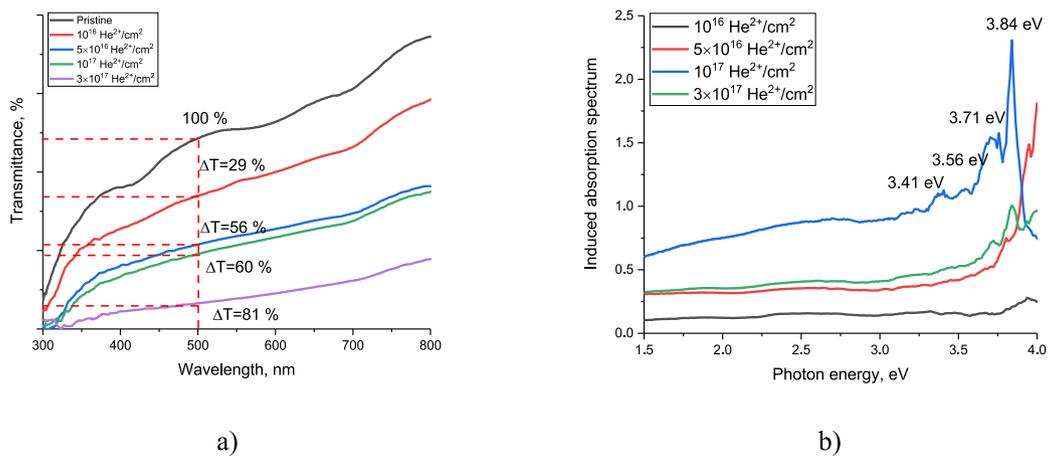


Fig. 5. a) Evaluation results of the optical transmission spectra of the studied BeO ceramics depending on the irradiation fluence (the dotted lines indicate changes in the transmission value, reflecting the formation of absorption centers in the damaged layer, leading to a decrease in the transmittance of the ceramics); b) Evaluation results of induced absorption spectra reflecting changes in the concentration of defects created by irradiation and the accumulation of structural distortions.

accumulate, but also create the effect of diffusion expansion of the damaged layer, caused by both the diffusion of implanted ions deep into the material and the migration of complex defects of the He-V_O type, resulting in crystal structure deformation. At the same time, the trend of decreasing changes in the values of residual mechanical stresses and FWHM depending on the depth indicates that the cumulative effect has a pronounced concentration dependence, the accumulation of which is associated with irradiation fluence, and as a result, with the concentration of defective formations arising as a result of the interaction of incident ions with the crystal structure. In this case, the observed structural changes can be explained by the effect of ballistic impact [31, 32] arising from irradiation, which, in the case of high irradiation doses, is expressed in the displacement of part of the structural damage into the material, thereby increasing the thickness of the damaged layer, as evidenced by the observed changes in the trends of the values of residual mechanical stresses and FWHM, the value of which characterizes in this case the contribution of the concentration of defective inclusions in the damaged layer.

The influence of the accumulation of structural distortions on the optical properties of ceramics, characterizing the change in transmission capacity, as well as the reasons causing its reduction, was determined using the evaluation method of the optical transmission spectra of the

studied samples depending on the irradiation fluence. The evaluation results of changes in the transmission spectra are presented in Fig. 5a. The analysis of the observed changes in the decrease in the transmission value indicates the formation of absorbing centers in the structure, the presence of which in this case is due to the structural disorder effects, as well as the formation of amorphous inclusions in the case of high irradiation fluences (10^{17} He²⁺/cm² and higher). In this case, the observed fundamental absorption edge shift, together with changes in the transmittance of ceramics, indicates an alteration in the electron density in the damaged layer, associated with the formation of an anisotropic charge distribution in the damaged layer, the change of which is caused by ionization processes of interaction of incident ions with the material.

The results of the induced absorption spectra calculated using the method of determining the ratio of the transmission intensities of the irradiated samples to the initial samples are presented in Fig. 5b. Analysis of the obtained data on the dependences of the absorption spectra indicates an elevation in absorption caused by irradiation, and it should be noted that the nature of the changes in the absorption spectra has a clearly expressed dependence on the type of structural changes caused by irradiation. In the case where the mechanisms of formation of residual mechanical stresses play the main role in structural changes in the structure of ceramics, the change in optical absorption spectra does

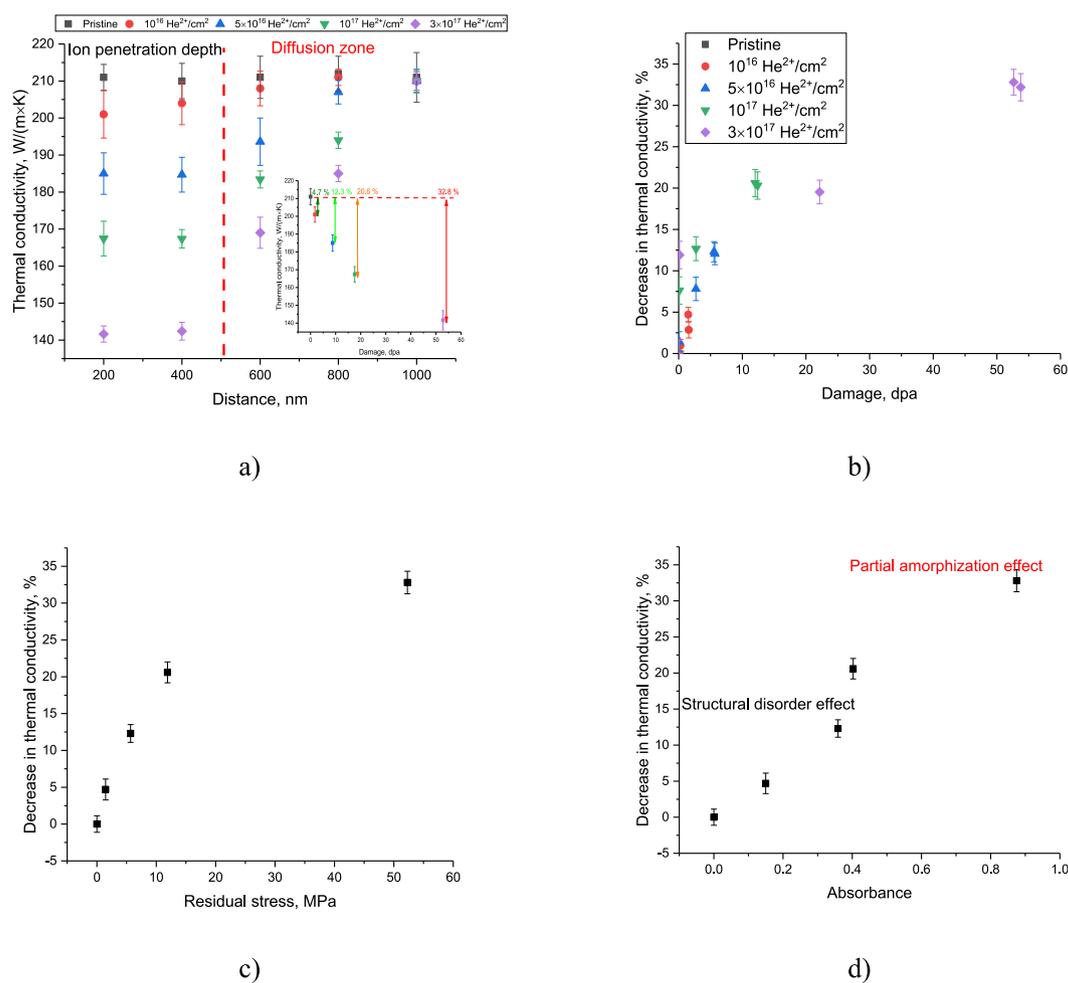


Fig. 6. a) Results of measurements of the thermal conductivity coefficient of BeO ceramics at different depths depending on the irradiation fluence (the inset shows the results of changes in the thermal conductivity coefficient at a depth of 200 nm, in comparison with the initial value); b) Results of comparison of the thermal conductivity degradation value and the value of structural damage, determined on the basis of changes in the value of residual stresses along the trajectory of ion movement; c) Assessment results of the connection between alterations in the degradation of the thermal conductivity coefficient and the value of residual mechanical stresses in the damaged layer structure; d) Results of comparative analysis of changes in the optical density value characterizing the accumulation of structural damage and absorption centers in the damaged layer and changes in the thermal conductivity coefficient expressed as a percentage of degradation of the thermal conductivity properties of irradiated samples.

not have clearly expressed maxima, from which it follows that the concentration of absorption centers associated with oxygen vacancies or complex defects of the He – V_O type is minimal. At fluences of 10¹⁷ He²⁺/cm² and higher, the formation of local maxima in the induced absorption spectra, the presence of which indicates an elevation in the structure of complex defects, and oxygen vacancies, the concentration of which grows with a change in the irradiation fluence, is observed. It should also be noted that these local maxima are in good agreement with the observed changes in the Raman spectra associated with the formation of halo peaks in the region of 1000–1700 cm⁻¹ and 2500–3200 cm⁻¹, the presence of which was explained by the formation of disordered regions associated with the accumulation of the concentration of implanted helium ions, alongside the formation of complex defects of the He – V_O type [18].

Fig. 6a shows the results of changes in the thermal conductivity coefficient of the studied ceramics in the damaged layer by depth, reflecting alterations in thermophysical parameters associated with structural changes and their accumulation with irradiation fluence growth. The general trend of change in the thermal conductivity coefficient by depth contingent upon the irradiation fluence indicates a direct relationship between the thermal conductivity degradation and the concentration of accumulated structural damage, as well as the broadening of the thickness of the damaged layer under high-dose irradiation.

As can be seen from the data presented, the change in the thermal conductivity coefficient depending on the irradiation fluence indicates the influence of the degradation of the damaged layer and the increase in its thickness on the thermal conductivity degradation. In this case, the observed reduction trend of thermal conductivity coefficient by depth depending on the irradiation fluence indicates that the structural damage accumulation leads to a decrease in the heat exchange rate, and a decrease in the concentration of structural distortions and defective inclusions deep into the sample directly affects the change in thermophysical parameters (data of the comparative analysis of the degradation of thermophysical properties by depth in comparison with the structural damage value are presented in Fig. 6b). The selection of the region of partial amorphization was performed taking into account the observed structural changes associated with a sharp increase in the contribution of deformation distortions in the damaged layer, which in this case is due to the effects of overlapping defective areas and subsequent amorphization. A reduction in the degree of structural damage associated with the ballistic nature of the transfer of defects into the damaged layer at a depth exceeding the ion range leads to a reduction in the degradation of the thermal conductivity coefficient, and at a depth of about 800–1000 nm, to a complete restoration of the value at the level of the original samples. Moreover, a comparative analysis of changes in the value of residual mechanical stresses and the degree of degradation of thermophysical parameters shown in Fig. 6c indicates a direct connection between disordering and partial amorphization of the damaged layer and deterioration of thermal conductivity. It should also be noted that the cumulative effect associated with an increase in the concentration of structural distortions leads to an increase in heat losses in the damaged layer, the change of which has a direct relationship with the concentration of absorbing centers arising as a result of disordering of the damaged layer (see data in Fig. 6d). Analysis of the change in the optical density value, reflecting the concentration of absorbing centers in the damaged layer structure, calculated on the basis of data on changes in transmission spectra, in comparison with changes in thermophysical parameters makes it possible to identify the contribution of the accumulation of the damaged layer crystalline structure deformation distortions to the degradation of thermophysical properties, and amorphous inclusions that arise during high-dose irradiation.

4. Conclusion

This article presents the results of a study of the radiation damage

accumulation effect on the change in optical and thermal parameters of BeO ceramics as a result of high-dose irradiation with helium ions, the accumulation of which occurs in the surface layer and can have a negative impact on the properties of ceramics, alongside their operating modes in the case of work in extreme conditions.

By assessing the nature of changes in deformation distortions, it was established that at low irradiation fluences, residual mechanical tensile stresses play a dominant role in distorting the crystalline structure of the damaged layer. While at the maximum irradiation fluence (3 × 10¹⁷ He²⁺/cm²) an equally probable nature of the influence of residual mechanical stresses of tensile and compressive types, due to an increase in the contribution of compressive stresses to deformation distortion, is observed. The increase in compressive residual stresses at maximum irradiation fluence is due to a rise in the concentration of complex defects of the He - V_O type in the damaged layer, as evidenced by an elevation in the intensity of the spectral line characteristic of this type of defect. The irradiation temperature growth in this case can have a negative effect on elevation of the damaged layer thickness, since under high-temperature exposure the effects of thermal expansion, as well as the diffusion of defects and implanted ions, are more pronounced.

The comparative analysis results of changes in the thermal conductivity coefficient of the studied samples by depth and structural disorder degree, determined on the basis of changes in the FWHM values of Raman spectral lines, showed a good correlation, reflecting the connection between the damaged layer degradation and the deterioration of the thermophysical parameters.

Further studies in this direction will consist in the search for enhancement of the stability of the near-surface layer of BeO ceramics to swelling by changing the grain sizes, as well as using methods of creating dispersion hardening in the near-surface layers due to external influences or size effects. In the future, much attention will also be paid to the study of degradation mechanisms of ceramics in the case of irradiation temperature variation.

CRedit authorship contribution statement

Azamat E. Ryskulov: Investigation, Formal analysis, Data curation. **Igor A. Ivanov:** Investigation, Formal analysis, Data curation. **Artem L. Kozlovskiy:** Writing – review & editing, Writing – original draft, Software, Resources, Methodology. **Marina Konuhova:** Writing – review & editing, Writing – original draft, Validation, Project administration.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

the work reported in this paper.

Data availability

Data will be made available on request.

References

- [1] F.F. Komarov, A.F. Komarov, V.V. Pil'ko, V.V. Pil'ko, Radiation resistance of structural materials of nuclear reactors on irradiation with high-energy hydrogen and helium ions, *J. Eng. Phys. Thermophys.* 86 (2013) 1481–1484.
- [2] A. Shaimerdenov, D. Sairanbayev, T. Kulsartov, S. Gizatulina, A. Akhanov, Z. Zaurbekova, M. Podoinikov, Irradiation experiments of titanium beryllide samples in the WWR-K reactor, *Ann. Nucl. Energy* 194 (2023) 110120.
- [3] Y. Liu, Y. Zhu, T. Shen, J. Chai, L. Niu, S. Li, Z. Wang, Irradiation response of Al₂O₃-ZrO₂ ceramic composite under He ion irradiation, *J. Eur. Ceram. Soc.* 41 (4) (2021) 2883–2891.
- [4] I.A. Sokolov, M.K. Skakov, V.A. Zuev, D.A. Ganovichev, T.R. Tulenbergenov, A. Z. Miniyazov, Study of the interaction of plasma with beryllium that is a candidate material for the first wall of a fusion reactor, *Tech. Phys.* 63 (2018) 506–510.
- [5] A.L. Kozlovskiy, M. Konuhova, D.B. Borgekov, Popov Anatoli, Study of irradiation temperature effect on radiation-induced polymorphic transformation mechanisms in ZrO₂ ceramics, *Opt. Mater.* 156 (2024) 115994.
- [6] L. Thomé, S. Moll, A. Debelle, F. Garrido, G. Sattonnay, J. Jagielski, Radiation effects in nuclear ceramics, *Adv. Mater. Sci. Eng.* 2012 (1) (2012) 905474.
- [7] A.I. Popov, M.A. Monge, R. González, Y. Chen, E.A. Kotomin, Dynamics of F-center annihilation in thermochemically reduced MgO single crystals, *Solid State Commun.* 118 (3) (2001) 163–167.
- [8] A.L. Kozlovskiy, M. Konuhova, D.I. Shlimas, D.B. Borgekov, M.V. Zdorovets, R. I. Shakirzyanov, A.I. Popov, Study of the effect of nanostructured grains on the radiation resistance of zirconium dioxide ceramics during gas swelling under high-dose irradiation with helium ions, *ES Materials & Manufacturing* 24 (2024) 1165.
- [9] M.A. Monge, R. Gonzalez, J.M. Santiuste, R. Pareja, Y. Chen, E.A. Kotomin, A. I. Popov, Photoconversion and dynamic hole recycling process in anion vacancies in neutron-irradiated MgO crystals, *Phys. Rev. B* 60 (6) (1999) 3787.
- [10] A. Lushchik, E. Feldbach, E.A. Kotomin, I. Kudryavtseva, V.N. Kuzovkov, A. I. Popov, E. Shablonin, Distinctive features of diffusion-controlled radiation defect recombination in stoichiometric magnesium aluminate spinel single crystals and transparent polycrystalline ceramics, *Sci. Rep.* 10 (1) (2020) 7810.
- [11] T. Inerbaev, A. Akilbekov, D. Kenbayev, A. Dauletbekova, A. Shalaev, E. Polissadova, M. Konuhova, S. Piskunov, A.I. Popov, Color centers in BaFBr crystals: experimental study and theoretical modeling, *Materials* 17 (2024) 3340.
- [12] S. Nikiforov, D. Ananchenko, A. Borbolin, A. Vokhmintsev, I. Weinstein, S. Zvonarev, Spectrally resolved thermoluminescence of anion-deficient Al₂O₃-BeO ceramics for high-dose dosimetry, *Phys. Status Solidi* 218 (1) (2021) 2000341.
- [13] V. Chakin, R. Rolli, A. Moeslang, P. Kurinskiy, P. Vladimirov, C. Ferrero, W. Van Renterghem, Study of helium bubble evolution in highly neutron-irradiated beryllium by using x-ray micro-tomography and metallography methods, *Phys. Scripta* 2011 (T145) (2011) 014012.
- [14] V. Altunal, V. Guckan, A. Ozdemir, Z. Yegingil, A calcination study on BeO ceramics for radiation dosimetry, *Mater. Res. Bull.* 130 (2020) 110921.
- [15] V.O. Altunal, V. Guckan, A. Ozdemir, K. Kurt, A.H.M.E.T. Ekcibil, Z. Yegingil, Investigation of luminescence properties of BeO ceramics doped with metals for medical dosimetry, *Opt. Mater.* 108 (2020) 110436.
- [16] V.S. Kiiko, V.Y. Vaispapi, Thermal conductivity and prospects for application of BeO ceramic in electronics, *Glass Ceram.* 71 (2015) 387–391.
- [17] S.V. Nikiforov, I.G. Avdyushin, D.V. Ananchenko, A.N. Kiryakov, A.F. Nikiforov, Thermoluminescence of new Al₂O₃-BeO ceramics after exposure to high radiation doses, *Appl. Radiat. Isot.* 141 (2018) 15–20.
- [18] V. Altunal, V. Guckan, Z. Yegingil, Effects of oxygen vacancies on luminescence characteristics of BeO ceramics, *J. Alloys Compd.* 938 (2023) 168670.
- [19] M.V. Zdorovets, A.L. Kozlovskiy, D.B. Borgekov, D.I. Shlimas, Influence of irradiation with heavy Kr¹⁵⁺ ions on the structural, optical and strength properties of BeO ceramic, *J. Mater. Sci. Mater. Electron.* 32 (11) (2021) 15375–15385.
- [20] A.V. Trukhanov, A.L. Kozlovskiy, A.E. Ryskulov, V.V. Uglov, S.B. Kislitsin, M. V. Zdorovets, D.I. Tishkevich, Control of structural parameters and thermal conductivity of BeO ceramics using heavy ion irradiation and post-radiation annealing, *Ceram. Int.* 45 (12) (2019) 15412–15416.
- [21] A.E. Ryskulov, M.V. Zdorovets, A.L. Kozlovskiy, D.I. Shlimas, S.B. Kislitsin, V. V. Uglov, Study of irradiation temperature effect on change of structural, optical, and strength properties of BeO ceramics when irradiated with Ar⁸⁺ and Xe²² heavy ions, *J. Mater. Sci. Mater. Electron.* 32 (2021) 10906–10918.
- [22] M.V. Zdorovets, D.I. Shlimas, A.L. Kozlovskiy, D.B. Borgekov, Effect of irradiation with low-energy He²⁺ ions on degradation of structural, strength and heat-conducting properties of BeO ceramics, *Crystals* 12 (1) (2022) 69.
- [23] M.V. Zdorovets, D.I. Shlimas, A.L. Kozlovskiy, D.B. Borgekov, Study of the application efficiency of irradiation with heavy ions to increase the helium swelling resistance of BeO ceramics, *Metals* 12 (2) (2022) 307.
- [24] S.B. Kislitsin, A.E. Ryskulov, A.L. Kozlovskiy, I.A. Ivanov, V.V. Uglov, M. V. Zdorovets, Degradation processes and helium swelling in beryllium oxide, *Surf. Coating. Technol.* 386 (2020) 125498.
- [25] N.J. Dutta, S.R. Mohanty, K.P. Sooraj, M. Ranjan, Surface and structural analyses of helium ion irradiated beryllium, *Vacuum* 170 (2019) 108962.
- [26] Ewen Smith, Geoffrey Dent, *Modern Raman Spectroscopy: a Practical Approach*, John Wiley & Sons, 2019.
- [27] A. Zhumazhanova, A. Mutali, A. Ibrayeva, V. Skuratov, A. Dauletbekova, E. Korneeva, M. Zdorovets, Raman study of polycrystalline Si₃N₄ irradiated with swift heavy ions, *Crystals* 11 (11) (2021) 1313.
- [28] X.Y. Chen, A.L. Wen, C.L. Ren, C.B. Wang, W. Zhang, H.F. Huang, P. Huai, Theoretical prediction of radiation-enhanced diffusion behavior in nickel under self-ion irradiation, *Nucl. Sci. Tech.* 31 (8) (2020) 79.
- [29] Todd R. Allen, Gary S. Was, *Radiation-enhanced diffusion and radiation-induced segregation. Radiation Effects in Solids*, Springer, Netherlands, 2007.
- [30] P.J. Doyle, K.M. Benensky, S.J. Zinkle, Modeling the impact of radiation-enhanced diffusion on implanted ion profiles, *J. Nucl. Mater.* 509 (2018) 168–180.
- [31] K. Nordlund, J. Keinonen, M. Ghaly, R.S. Averback, Coherent displacement of atoms during ion irradiation, *Nature* 398 (6722) (1999) 49–51.
- [32] A.V. Krashenninnikov, K. Nordlund, Ion and electron irradiation-induced effects in nanostructured materials, *J. Appl. Phys.* 107 (7) (2010).