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EFFECTS OF ENERGY FLUXES ON MATERIALS

Study of the Microstructure and Phase Composition of Ceramics Based on Silicon Carbide Irradiated with Low-Energy Helium Ions

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Abstract—The effect of irradiation with helium ions with energy of 40 keV and doses from 1×10^{14} to 2×10^{17} cm^{-2} on the microstructure and phase composition of ceramics based on silicon carbide is studied. Radiation growth of the 6H–SiC crystal lattice is revealed. At a dose of 1×10^{16} cm^{-2}, a decrease in deformation is observed related to the formation of gas-vacancy clusters, which are sinks for radiation defects. Amorphization of the near-surface layer is established.

Keywords: silicon carbide, 6H–SiC, helium, amorphization
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INTRODUCTION

Silicon carbide (SiC) is a wide-bandgap semiconductor with high thermal conductivity and low thermal expansion, chemical stability, and radiation resistance, which has more than 200 polytypes with variations of the band gap. It can be used to manufacture electronic devices and structural components in fusion energy systems.

A fundamental understanding of the accumulation and recovery of radiation damage in SiC is necessary for the implementation of new technologies into production [1]. Owing to the fact that a large number of neutrons are released during the operation of a nuclear reactor, which cause nuclear reactions in the material with the formation of inert gases, helium is most often chosen for irradiation. It is known that implantation of helium into SiC results in the formation of bubbles at elevated temperatures and high radiation doses. The investigation in [2] provides evidence that helium implantation changes the mechanical properties, chemical reactivity, and electrical properties of silicon carbide. It is also known that irradiation with helium ions with energies from 15 to 50 keV and doses from 10^{15} to 10^{16} cm^{-2} leads to amorphization of the near-surface layer of SiC [1–3]. Most of the studies are carried out on 6H–SiC single crystals.

The aim of the study is to investigate the behavior of He in silicon carbide and its effect on the structure of the material.

EXPERIMENTAL

Samples of reaction-bonded Si/SiC ceramic were prepared at the Lykov Institute of Heat and Mass Transfer (Belarus), the main stages of preparation of which are shown schematically in Fig. 1.

Two fractions of silicon carbide powder were used as raw materials: SiC of M50 grade with a characteristic grain size of 50 μm and of grade M5 with a grain size of 5 μm (Volzhskii Abrasive Plant, Russia). The ratio of coarse and fine fractions was 5 : 3.

Silicon carbide powder (88 wt %) was mixed with a thermoplastic binder based on P-2 paraffin (12 wt %) and cast in a mold. Thermal removal of the binder component was carried out in air at 600°C. The resulting porous SiC matrix, after removing paraffin, was impregnated with bakelite varnish based on LBS-1 resol resins (Sverdlov Plant, Russia) at a temperature of 40°C and a pressure of 0.5 MPa. Then the preform was dried in air at 160°C for 4 h. Next, it was subjected to pyrolysis in a vacuum furnace (VacETO, Russia) at

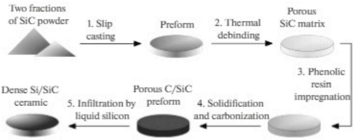

Fig 1. The main stages of the preparation of SiC ceramics [4].

a temperature of 1200°C and a pressure of 0.13 Pa for 2 h.

The same vacuum furnace was used for the final stage of siliconization, which was carried out at a temperature of 1800°C and a pressure of 0.13 Pa for 4 h. To do this, the sample was placed in a closed graphite crucible and covered with a homogeneous layer of dispersed polycrystalline silicon with a purity of at least 99.99%, which melted when heated and penetrated into the porous structure of the C/SiC preform. A chemical reaction occurred between liquid silicon and carbon in the pores to form secondary silicon carbide, which bound the original particles of primary silicon carbide powders. The remaining space remained filled with silicon. After siliconization, the surface of the sample became rough and inhomogeneous, so mechanical grinding and polishing was used to remove residual silicon and create the necessary conditions for further research of the ceramic [4].

Irradiation of samples with He^{2+} ions with an energy of 40 keV and fluences of 1×10^{14}, 1×10^{15}, 1×10^{16}, 5×10^{16}, and 2×10^{17} cm^{-2} was carried out at room temperature on a DC-60 linear accelerator of heavy ions (Institute of Nuclear Physics, Kazakhstan).

The study of the structural and phase state of the initial and irradiated silicon carbide samples was carried out using X-ray diffraction (XRD) analysis and Raman spectroscopy (RS). XRD analysis was carried out on an Ultima IV diffractometer (Rigaku, Japan) using a parallel beam geometry in copper radiation (CuK_α) with a wavelength of 0.154179 nm. Raman spectra were recorded at room temperature using a spectral-analytical complex based on a Nanofinder HighEnd scanning confocal microscope (LOTIS-TII, Belarus–Japan). The excitation wavelength was 532 nm.

RESULTS AND DISCUSSION

The maximum concentrations, damaging doses, and energy losses in the projective path of helium ions were calculated using the SRIM 2013 program (Table 1, Fig. 2).

The maximum concentration of helium ions was achieved at a depth of 348 nm. As can be seen in Fig. 2, the greatest energy losses of helium ions occur in the near-surface layer. About 80% of the energy is lost in the layer with the thickness of 300 nm.

The results of studies of the phase composition (Fig. 3) showed that the initial samples are a multiphase system: 6H–SiC with hexagonal (*P*63*mc*) phase, Si with cubic (*Fd*-3*m*) phase, and 15R–SiC with trigonal (*R*3*m*) phase. The main phase is 6H–SiC (~80%), the content of the 15R–SiC phase is ~15%, and the content of Si is ~5%. The phase content was assessed by means of the Rigaku PDXL software package using the Rietveld method.

Figure 4 shows the dependence of the relative change in lattice parameters *a* and *c* on the irradiation dose. Three areas can be distinguished on the graphs. Region I (1×10^{14}–1×10^{15} cm^{-2}) is characterized by the formation of radiation vacancies, as well as vacancy complexes. The latter are the nuclei of the for-

Table 1. Results of SRIM calculations

	Implantation dose, cm^{-2}				
	1×10^{14}	1×10^{15}	1×10^{16}	5×10^{16}	2×10^{17}
He concentration, at %	0.009	0.09	0.9	5	18
Damaging dose, displacements per atom, dpa	0.006	0.06	0.6	3	12

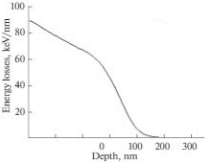

Fig. 2. Energy losses of He ions in SiC samples.

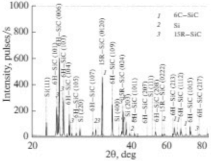

Fig. 3. X-ray diffraction pattern of the original silicon carbide sample.

mation of vacancy and gas pores (bubbles) [3, 5]. The relative change in the parameters a and c of the crystal lattice increases modulo; the difference lies in the stresses: compressive stresses arise in the direction of parameter a, and tensile stresses arise in the direction of parameter c. In region II (1×10^{15}–1×10^{16} cm^{-2}), there is a sharp drop in stresses related to the formation of helium bubbles, which are sinks for defects [6]. Therefore, for both lattice parameters, their relative change decreases. Region III (1×10^{16}–2×10^{17} cm^{-2}) is characterized by an increase in deformation, which is apparently related to both a decrease in the concentration of gas-vacancy clusters and their growth due to coalescence processes occurring in induced stress fields. An increase in the lattice parameter a and a decrease in the lattice parameter c called radiation growth should be noted. This effect is characteristic mainly of hcp lattices of metals, especially zirconium [7].

Silicon carbide 6H–SiC has hexagonal symmetry with a wurtzite structure and has C_{6v} symmetry. The selection rule of theory of groups predicts that modes A_1, E_1, and E_2 are active in the Raman spectrum. They are further divided into longitudinal (LO) and transverse (TO) optical modes [8].

To describe Raman spectra, the range of dose values can be divided into the same areas as when describing deformation (Fig. 4). The first region is characterized by a shift of the A_1 (LO) mode, which is responsible for the crystal lattice parameter c, toward longer wavelengths (Fig. 5), which is related to tensile stresses in this direction. The remaining three modes, which are responsible for parameter a of the crystal lattice, shift toward shorter wavelengths, which is related to compressive stresses in this direction. As mentioned above, in region II, there is a drop in stresses, which is evidenced in the spectra by a shift of peaks in the opposite direction relative to region I. There is also a decrease in intensity and broadening of the peaks, which is related to the accumulation of radiation defects.

With a further increase in the radiation dose, no peaks are detected in the Raman spectra. This is due to

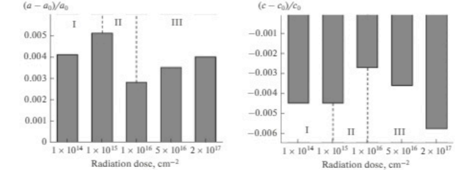

Fig. 4. Dependence of the relative change in parameters a and c of the 6H–SiC crystal lattice on the dose of irradiation with He ions with an energy of 40 keV.

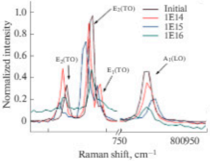

Fig. 5. Raman spectra of the initial and irradiated SiC samples.

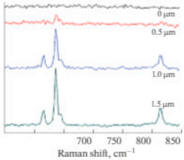

Fig. 6. Raman spectra for a SiC sample irradiated with a dose of 5×10^{16} cm^{-2} taken at various distances z from the sample surface.

an increase in the absorption coefficient because of the accumulation of optically absorbing centers induced by helium, as well as to the amorphization of the near-surface layer [9, 10].

Figure 6 shows Raman spectra for an irradiation dose of 5×10^{16} cm^{-2}, which were obtained at various distances from the irradiated sample surface (z). In a layer with a thickness of about 0.5 μm, no peaks are observed, but with a subsequent increase in depth z, first-order SiC peaks are observed.

CONCLUSIONS

As a result of irradiation of silicon carbide samples with helium ions with an energy of 40 keV and doses from 1×10^{14} to 2×10^{17} cm^{-2}, changes in the crystal lattice related to the formation of helium bubbles in the SiC structure were revealed, which can be described in three stages. At the first stage, an increase in stresses occurs (compressive ones in the direction a of the crystal lattice and tensile ones in the direction c), which is related to the nucleation of helium bubbles. Then a sharp decrease in stresses (deformations) is observed as a result of the formation of bubbles. At the final stage, an increase in stresses is observed because of an increase in the size of He bubbles. Irradiation also leads to amorphization of the near-surface layer with a thickness of about 0.5 μm.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

REFERENCES

1. Jiang, W., Thevuthasan, S., Weber, W.J., and Gröetzschel, R., Deuterium channeling analysis for He^+-implanted 6H–SiC, *Nucl. Instrum. Methods Phys. Res., Sect. B*, 2000, vols. 161–163, pp. 501–504. https://doi.org/10.1016/S0168-583X(99)00692-8
2. Ali, A.A., Kumar, J., Ramakrishnan, V., and Asokan, K., Raman spectroscopic study of He ion implanted 4H and 6H–SiC, *Mater. Lett.*, 2018, vol. 213, pp. 208–210. https://doi.org/10.1016/j.matlet.2017.11.040
3. Li, B.S., Du, Y.Y., and Wang, Z.G., Recrystallization of He-ion implanted 6H–SiC upon annealing, *Nucl. Instrum. Methods Phys. Res., Sect. B*, 2015, vol. 345, pp. 53–57. https://doi.org/10.1016/j.nimb.2014.12.049
4. Grinchuk, P.S., Kiyashko, M.V., Abuhimd, H.M., Alshahrani, M.S., Solovei, D.V., Stepkin, M.O., Akulich, A.V., Shashkov, M.D., Kuznetsova, T.A., Danilova-Tretiak, S.M., Evseeva, L.E., and Nikolaeva, K.V., Advanced technology for fabrication of reactionbonded SiC with controlled composition and properties, *J. Eur. Ceram. Soc.*, 2021, vol. 41, no. 12, pp. 5813–5824. https://doi.org/10.1016/j.jeurceramsoc.2021.05.017
5. Uglov, V.V., Surface erosion of multilayer films based on zirconium and silicon nitrides irradiated with helium ions, *Vests. Nats. Akad. Navuk Belarusi. Ser. Fiz.-Mat. Navuk*, 2018, vol. 54, no. 1, pp. 110–118. https://doi.org/10.29235/1561-2430-2018-54-1-110-118
6. Debelle, A., Crocombette, J.-P., Boulle, A., Chartier, A., Jourdan, Th., Pellegrino, S., Bachiller-Perea, D., Carpentier, D., Channagiri, J., Nguyen, T.-H., Garrido, F., and Thomé, L., Lattice strain in irradiated materials unveils a prevalent defect evolution mechanism, *Phys. Rev. Mater.*, 2018, vol. 2, no. 1, p. 013604. https://doi.org/10.1103/PhysRevMaterials.2.013604

7. Li, Y. and Ghoniem, N., Cluster dynamics modeling of irradiation growth in single crystal Zr, *J. Nucl. Mater.*, 2020, vol. 540, p. 152312. https://doi.org/10.1016/j.jnucmat.2020.152312

8. Daghbouj, N., Li, B.S., Callisti, M., Sen, H.S., Karlik, M., and Polcar, T., Microstructural evolution of helium-irradiated 6H–SiC subjected to different irradiation conditions and annealing temperatures: A multiple characterization study, *Acta Mater.*, 2019, vol. 181, pp. 160–172. https://doi.org/10.1016/j.actamat.2019.09.027

9. Li, B.S., Zhang, C.H., Zhang, H.H., Shibayama, T., and Yang, Y.T., Study of the damage produced in 6H–SiC by He irradiation, *Vacuum*, 2011, vol. 86, no. 4, pp. 452–456. https://doi.org/10.1016/j.vacuum.2011.09.011

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