HIGH ENERGY HEAVY ION TRACKS IN NANOCRYSTALLINE SILICON NITRIDE

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Silicon nitride is the only nitride ceramic where latent tracks have been registered. There is almost no data devoted to nanocrystalline Si_3N_4 despite the actuality of studying properties of nanomaterials, in particular a radiation stability. This work was dedicated to study effect of irradiation with high energy heavy ions in nanocrystalline silicon nitride by TEM means being the unique direct research method available for nanomaterials. The threshold electronic loss power was estimated at ~17 keV/nm level based on the determined track radii. The comparative analysis of present results and previous data for polycrystalline Si_3N_4 irradiated at the same conditions gives a reason to expect the radiation stability of silicon nitride should not depend on the crystal grain size sufficiently.

Keywords: nanomaterials; silicon nitride; swift heavy ion irradiation; latent track; radiation stability.

Introduction

Nanocrystalline materials are an actual task for researchers over the world recently, as a decrease of the grain size can induce changes in their properties, in particular in a radiation stability [1-4]. On the one hand, a high density of grain boundaries can act as a highly efficient sink for annihilation of interstitial point defects and vacancies created during irradiation [1-5] and improves radiation characteristics. On the other hand, a difficult energy transfer through the material promotes a growth of number of radiation damages due to the localized energy distribution within the grain diameter sized in a nanometer scale [5-7]. The mechanism of radiation damages in the nanostructured material is determined depending on which of processes above is dominant in a specific case.

Among isolators, SiO_2 and Si_3N_4 play the unique role as materials of silicon micro- and nanoelectronics [8]. Silicon nitride being the only nitride ceramic where latent tracks have been registered is of interest for study its radiation behavior. By now, most previous works have been aimed to study radiation-induced changes in properties of amorphous films [7-13] and polycrystals [8, 14-18] of Si_3N_4, whereas there is almost no data about its nanocrystalline state (nc-Si_3N_4).

A special attention should be paid for methods to study properties and effect of latent tracks. In major cases means as RBS, SAXS, AFM and FTIR, chemical etching [for instance, 1, 8-11, 18, 19] were used, and fewer works [1-2, 7, 14-17] presented results obtained by transmission electron microscopy (TEM) being by the way one and only direct method to study SHI defects. High resolution TEM allows to determine track characteristics as a continuity/discontinuity and a size with a minimal error, what gives a base for a verification of different models and as a consequence for widening a data about the mechanism of forming defects under different irradiation conditions. Additionally, TEM should be underlined to be the most suitable method to study defects in individual crystallites, which is principally actual for nanomaterials. Thus, based on all points above the present work is decided to aim to study a high energy heavy ions impact on radiation stability of nanocrystalline silicon nitride.

Experimental

Commercially available nanocrystalline powder Si_3N_4 sedimented on TEM grids were used as targets for irradiation. Specimen were irradiated with 710 MeV Bi to fluence 5·11 cm^{-2} at U-400 cyclotron in the FLNR of JINR (Dubna, Russia) and 220 MeV Xe to fluence 5·11 cm^{-2} at DC-60 cyclotron in INP (Nur-Sultan, Kazakhstan). Different thick aluminum foils were used to variate electronic stopping powers. Microstructural analysis was done with a JEOL JEM 2100 LaB_6 or ARM200F TEM operated at 200 kV in the Centre for HRTEM in Nelson Mandela University (Port Elizabeth, South Africa), and with Talos™ F200i S/TEM operated at 200 kV in the FLNR of JINR.

Results

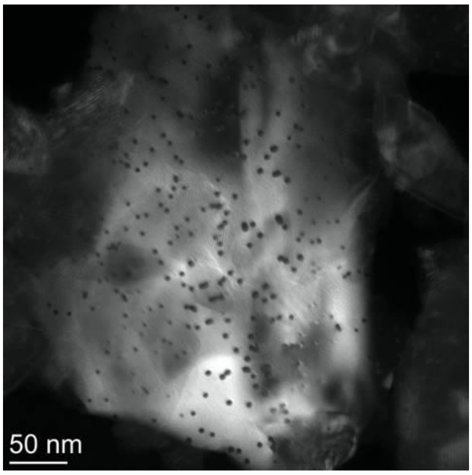
Typical TEM images of nanocrystalline silicon nitride (nc-Si_3N_4) irradiated with high energy heavy ions are presented in Fig.1.

Results of electronic stopping powers of incident ions calculated with SRIM-2016 and average radii of latent tracks in nc-Si_3N_4 are given in Table 1 and Fig.2. No tracks were found in samples irradiated with 710 MeV Bi and thicker than 27 μm Al foil, that means threshold electronic stopping power S_{et} should be more than 16.5 keV/nm. Analysis of points in Fig.2 confirms this statement and allows to estimate a S_{et} value of nanocrystalline silicon nitride ~17 keV/nm.

Table 1. Electronic stopping powers S_e and mean track radii in nc-Si_3N_4

Ion, energy, MeV	Foil thick, μm	S_e, keV/nm	R, nm
^{209}Bi, 710	no	33.58	1.7±0.2
	14	30.4	1.5±0.3
	17.4	28.83	1.7±0.2
	19	27.33	1.6±0.3
	23	23.62	1.2±0.4
	27	19.04	0.9±0.4
	28.7	16.53	no
	34.7	11.34	no
^{131}Xe, 220	no	22.12	1.12±0.2

In [14] of Zinkle et al and our previous works [15-17] track sizes were determined in polycrystalline silicon nitride p-Si_3N_4 irradiated under the same conditions in the individual track regime (710 MeV Bi and 220 MeV Xe to fluence of 5e11 cm^{-2}). The radii for p-Si_3N_4 were equal 1.7±0.2 nm and 1.0±0.2 nm



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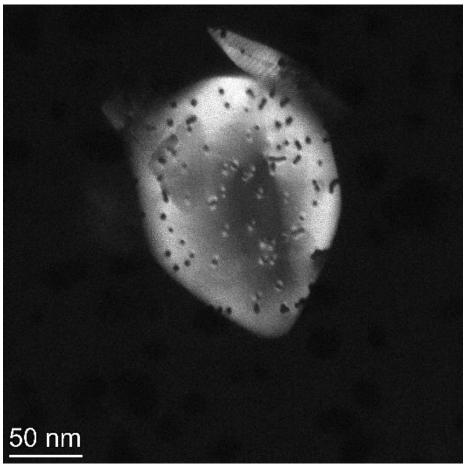

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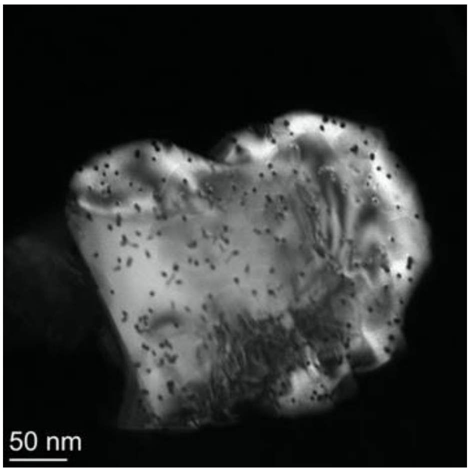

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Fig. 1. TEM images of nc-Si_3N_4 irradiated with 710 MeV Bi a) without foil; b) with 14 μm Al foil; c) with 23 μm Al foil

for bismuth and xenon ions respectively. Matching the previous results and the data in this work shows almost same values for poly- and nano- silicon nitride within the margin of error that allows to suppose their comparable radiation stabilities too. Additionally, it confirms with the estimated threshold conditions

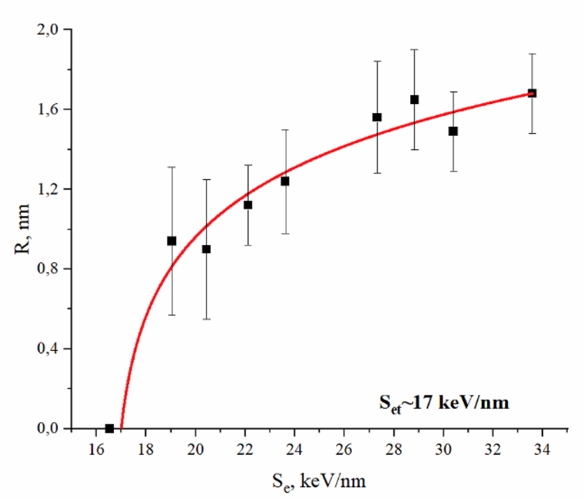

Fig 2. Latent tracks radii in nc- Si_3N_4 as a function of electronic energy loss

~18 keV for p-Si_3N_4 in [16, 17] and ~17 keV for nc-Si_3N_4 obtained in the present work.

Conclusion

Based on experimentally determined track radii for 710 MeV Bi and 220 MeV Xe irradiation of nc-Si_3N_4 threshold electronic stopping power was found to be equal ~17 keV/nm. The comparative analysis of data for this material in poly- and nanostate determines almost the same values of latent track size and threshold conditions and, consequently, it is expected that a radiation stability of Si_3N_4 should not depend on the grain size sufficiently.

Acknowledgements

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