ITS MODEL-BASED ANALYSIS OF TRACK FORMATION IN CRYSTALLINE AND AMORPHOUS SILICON NITRIDE

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Silicon nitride is a promising candidate material for inert matrix fuel host to be used for transmutation of minor actinides via nuclear reactions. Radiation-induced changes in this material are subjects of extensive studies. Most corresponding literature data focus on radiation damage produced by neutron and charged particle radiation in elastic collisions and much less is known about defects formed via relaxation of dense electronic excitations associated with high-energy heavy ion irradiation. The purpose of this report is the analysis of latent track parameters in polycrystalline and amorphous Si3N4 irradiated with swift (700 MeV) Bi ions deduced from transmission electron microscopy (TEM) examination. We discuss the applicability of the «core-shell» approach based on the inelastic thermal spike model to describe the latent formation in this ceramic comparing experimental and calculated track sizes. The threshold vaporization energy level corresponding to the core radii found from TEM data is equal Eb=1.6 eV/atom for polycrystalline silicon nitride. Additionally, the threshold electron loss energy necessary for track formation was estimated as ~14 keV/nm for amorphous Si3N4 and ~18 keV/nm for polycrystalline Si3N4.

Keywords: swift heavy ions; ceramics; silicon nitride; radiation defects; latent tracks.

Introduction

Nowadays disposal of spent nuclear fuel and radioactive waste resulting from the nuclear fuel cycle stay the actual problem of the modern material science. One of the approaches to reduce the radiotoxicity of waste consists of the transmutation of minor actinides using so-called inert matrix fuel hosts [1]. Silicon nitride is considered as candidate material for such matrices because of its physical properties, which are well suited to reactor conditions [2, 3]. Structural response of Si3N4 against swift heavy ion (SHI) irradiation, simulating fission fragments (FFs) still remains less studied in comparison with conventional types of radiation. In particular, there is an evident lack of experimental data on latent track parameters in this material as well as atomistic models of track formation. It should be noted that Si3N4 is the only nitride ceramic where latent tracks have been registered [3].

This study presents the comparative analysis of the latent track parameters in crystalline and amorphous silicon nitride in the framework of the inelastic thermal spike (iTS) model.

Experimental

Commercially available Al doped polycrystalline (p-SiN, average grain size about 1 micrometer) and amorphous silicon nitride (a-SiN) films (thickness 300 nm) were used as target materials in this work. Amorphous film samples were pre-thinned to TEM transparency using Technoorg ion milling technique. The samples were irradiated with 700 MeV Bi ions to fluences ranging from 5×10^{11} cm^{-2} to 1.23×10^{13} cm^{-2} at room temperature using U-400 cyclotron facility at FLNR JINR, Dubna. Selected p-SiN samples were exposed by 1030 MeV Bi ions to fluence 10^{10} cm^{-2} at U-400M FLNR JINR cyclotron and 220 MeV Xe ions at DC60 cyclotron (INP, Nur-Sultan, Kazakhstan).

TEM lamellae from polycrystalline Si_3N_4 were produced by means of a FEI Helios Nanolab FIB-SEM. The lamellae were investigated with either a JEOL 2100 LaB6 or a JEOL ARM 200F TEM both operated at 200 kV. Microscopic researches were conducted in the Centre for HRTEM in Port Elizabeth, Nelson Mandela University, South Africa.

Results

An example of high-resolution TEM image of amorphous latent tracks in p-SiN is given in Fig. 1.

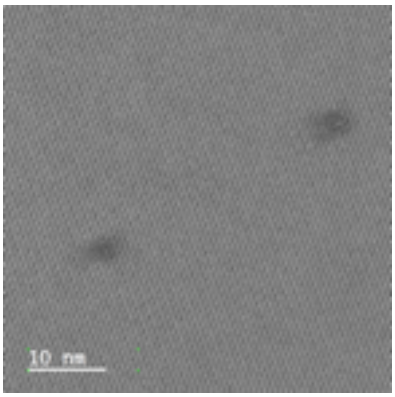

Fig. 1. TEM image of isolated latent tracks in polycrystalline Si_3N_4 irradiated with 700 MeV Bi ions

Fig. 2 shows BF TEM micrograph of p-SiN irradiated in the ion track overlapping regime (ion fluence, demonstrating amorphous latent tracks (white spots) in almost completely SHI amorphized material containing some crystalline remains).

Typical HAADF-STEM image of latent track induced by swift Bi ions in a-SiN is given in Fig. 3.

The distribution of latent track sizes is shown in Fig. 4. As can be seen, 700 MeV bismuth ions produce tracks with a broad diameter distribution ranging from 1.5 to 5 nm. The mean track diameters in amorphous and polycrystalline Si3N4 are equal 2.3±0.2 nm and 3.3±0.3 nm, respectively.

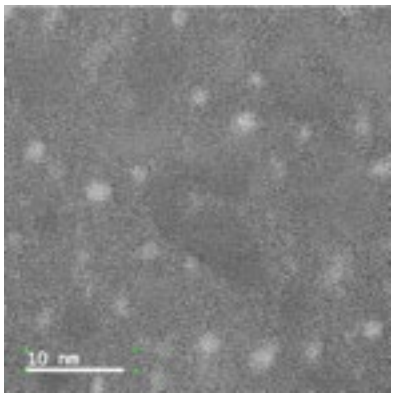

Fig. 2. 700 MeV Bi ion induced tracks in amorphized Si_3N_4

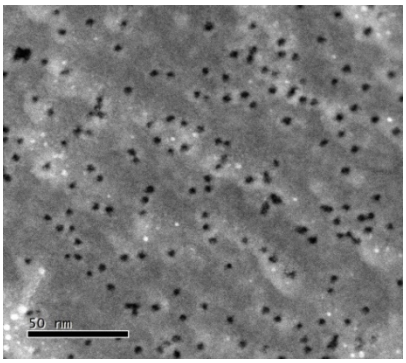

Fig. 3. Latent tracks of 700 MeV Bi ions in amorphous Si_3N_4

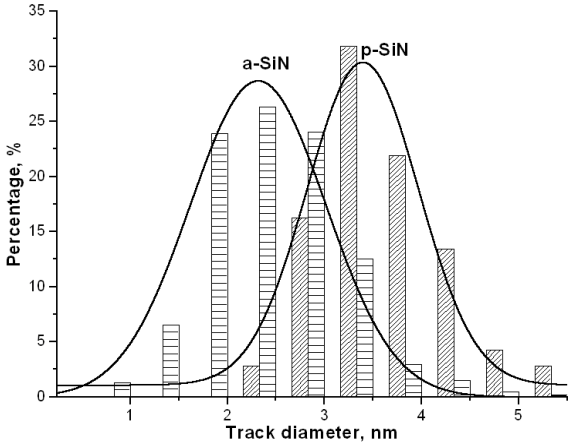

Fig. 4. Latent tracks diameter distribution for polycrystalline and amorphous silicon nitride irradiated with 700 MeV Bi ions

Discussion

As known, several models have been developed to understand latent track formation. One of them, most often used in analysis of experimental data, is inelastic thermal spike (iTS) based on two main equations describing the energy exchange between electronic and ionic subsystems in SHI irradiating solids [4]:

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(rK_e(T_e)\frac{\partial T_e}{\partial r}\right) - \lambda(T_e - T_i) + A(r,t)$$

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where C_e, C_i are specific heat capacities, T_e, T_i are temperatures, K_e, K_i are thermal conductivities (indices *i* and *e* refer to the ionic and electronic subsystems of the material, respectively), λ is the constant (velocity) of the electron-phonon interaction, A (*r*, *t*) is a space-time source describing the heating of the material electronic subsystem by an incident ion.

The iTS model has been successfully applied to determine track parameters in variety of materials. However, standard iTS model, where track formation is explained by quenching of a molten region around ion trajectory, predicts incorrect track size in Si_3N_4 [5]. To fit experimental results, Kitayama et al. had used the approach based on the idea of the «core-shell» construction of defects [5]. According to it, the track core was suggested to be formed via the vaporization of material whereas the shell was a result of its melting. Following this approach there were evaluated Bi ion induced track parameters in polycrystalline, amorphized and amorphous silicon nitride.

Table 1 presents experimental and calculated data on track parameters in a-SiN and amorphized Si_3N_4 (az-Si). As was found, experimental track radii may be successfully reproduced using the electron–phonon coupling constant λ=3 nm and the energy corresponding to the melting and boiling processes E_m= 0.62 eV/atom and E_b= 2.5 eV/atom, respectively. This consequence is in a good agreement with results presented in [5]. The results of calculation are also given in Fig. 5 together with experimental data from papers [5-7] as a function of electronic stopping power.

Table 1. S_e- electronic stopping power

Ion, energy, MeV	S_e, keV/nm	R, nm a-SiN	R, nm az-SiN	R, nm iTS
Bi, 700	31.46	1.2±0.2	1.5±0.3	shell 6 core 1.7

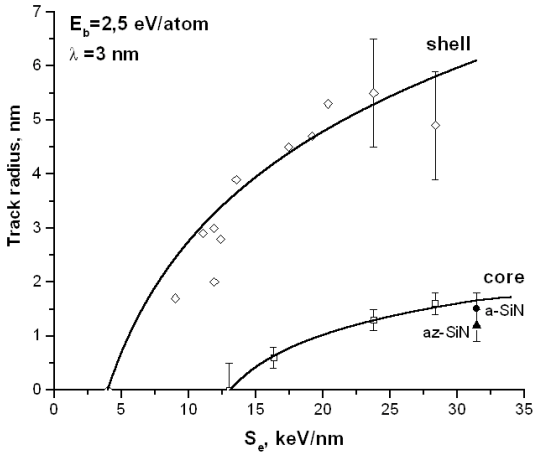

Fig. 5. The core and shell radii in amorphous Si_3N_4 as a function of electronic energy loss

The comparison of experimental and calculated mean track radii in polycrystalline Si_3N_4 is shown in Table 2 and Fig. 6.

Except of the data on latent fusion heat L_{fus}=1400 J/g [5] and the decomposition temperature T_{melt}=2775 K [2], we failed to find correct parameters

Table 2. The comparison of experimental and calculated mean track radii in polycrystalline Si_3N_4

Ion, energy, MeV	S_e, keV/nm	R, nm	iTS R, nm Eb=1.6 eV/atom λ=4.3 nm
Bi, 700	33.5	1.8±0,3 (5e11)	shell 4.5 core 2.1
Bi, 1030	33	1.6±0,3	shell 4.2 core 2.1
Xe, 220	21.9	1±0.1	shell 3.1 core 0.9

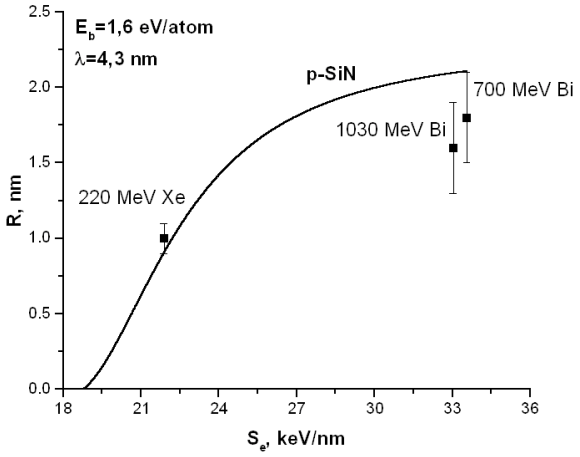

Fig. 6. The core radius in polycrystalline Si_3N_4 as a function of electronic energy loss

for the vaporization of this material. So, based on the experimental data the best fit result of the iTS calculations was received with Eb = 1.6 eV/atom for the polycrystalline silicon nitride. The electron-phonon interaction constant was taken as λ=4.3 nm. These values were chosen using the empirical relation between λ and bandgap of crystalline nitride solids (standard values usually vary within 4.3 and 4.8 nm).

Conclusions

It was found that experimentally determined track core radii can be obtained in the framework of the iTS model using vaporization energy value Eb=1.6 eV/atom for polycrystalline silicon nitride. Additionally, the threshold electronic stopping powers ~14 keV/nm and ~18 keV/nm were deduced for amorphous and polycrystalline Si3N4.

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