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Flerov Laboratory of Nuclear Reactions

FINAL REPORT ON THE SUMMER STUDENT PROGRAM

Latent Track Formation in Crystalline and Amorphous Silicon Nitride

Supervisor:
Dr. Vladimir Alexeevich Skuratov

Student:
Anel Ibrayeva, Kazakhstan
L.N. Gumilyov Eurasian National University

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Introduction

Nitride ceramics are considered as candidate materials for inert matrix fuel hosts for transmutation of minor actinides [1]. So, silicon nitride attracts a great attention due to its remarkable thermal and mechanical properties [2,3]. Furthermore, for mentioned applications an irradiation stability of materials is required, in particular impact of ions with energy of fission fragments because it still stays less studied and consequently less understood than neutron and conventional (low energy) ion irradiation effects [2, 3]. As known, this impact can be simulated by swift heavy ions (SHI) E≥1 MeV/amu, which in turn can cause a formation of specific radiation defects called "latent tracks" when a certain threshold value of specific ionization energy loss S_{et} is exceeded. At the same time S_{et} and track radii R_{tr} should be noted as only experimentally determined "input parameters" used to verify theoretical models of track formation.

By now, among available experimental methods to investigate radiation-induced defects in solids TEM observation is considered as a comprehensive one for a study of latent tracks since it is a direct measurement technique. In turn, among theoretical models able to describe such complicated process inelastic thermal spike (iTS) model is widely used as it has been well recommended itself for describing track formation process under swift heavy ions impact in many insulators. Taking into account special features of Si_3N_4 the "two-threshold" variation of iTS model seems to be more suitable [4].

Because silicon nitride is the only nitride ceramic where latent tracks have been registered, to investigate its radiation-induced response becomes more motivating and actual task, especially by both experimental and calculation methods. The purpose of our work is to analyze latent track parameters in amorphous and crystalline Si_3N_4 by high resolution TEM and in framework of iTS model.

Experimental

Commercially available amorphous silicon nitride films (a-SiN) and polycrystalline Si_3N_4 (p-SiN) with grain size from several hundred nanometers to 2 microns were purchased from MTI Corporation (http://www.mtixtl.com). Pristine samples were irradiated with 710 MeV Bi ions up to fluence of 5×10^{11} 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.

TEM lamellas oriented parallel to the implanted surface were prepared by means of an FEI Helios Nanolab 650. To minimise ion beam induced damage the samples were prepared with ion energies down to 500 V. Structural analysis was done with a Cs corrected JEOL ARM200F transmission electron microscope operated at 200 kV

Results

An example of high-resolution TEM image of amorphous latent tracks in p-SiN and a-SiN is given in Fig. 1. The average track sizes were determined based on analysis of HAADF microscopic images using Gaussian fitting function with statistical reliability coefficient ~95% (fig.2).

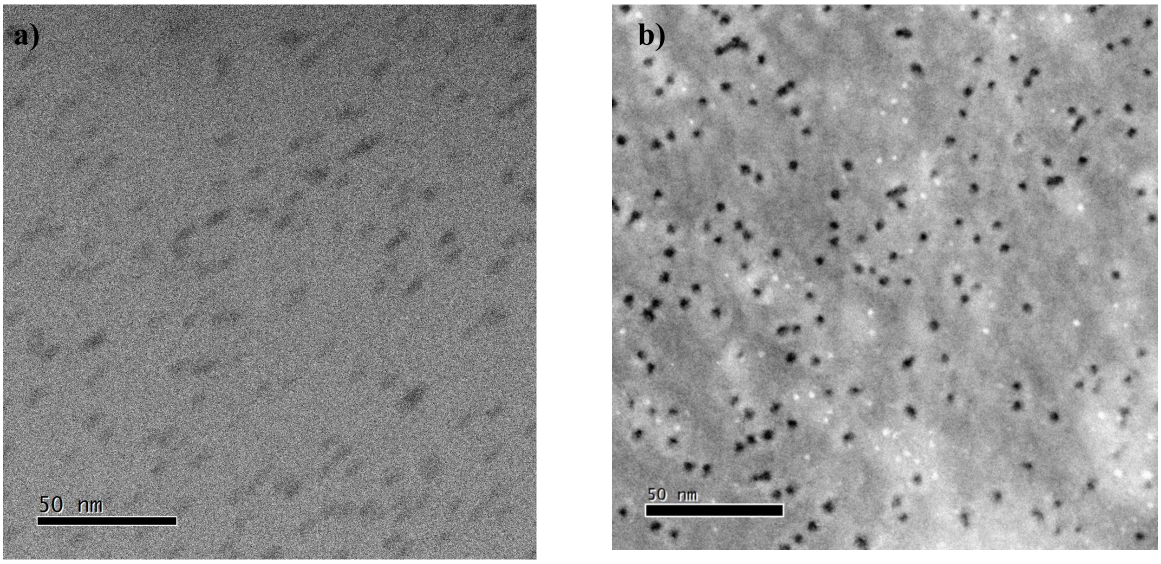

Figure 1. HAADF STEM images of polycrystalline (a) and amorphous (b) Si_3N_4 irradiated with 710 MeV Bi up to fluence of $5x10^{11}$ cm^{-2}

As seen, 710 MeV bismuth ions produce tracks with a broad diameter distribution ranging from 0.5 to 5 nm and from 0.5 to 4.5 nm for amorphous and polycrystalline specimens, respectively, the corresponding average values are equal to 2.3±0.3 nm and 3.3±0.3 nm.

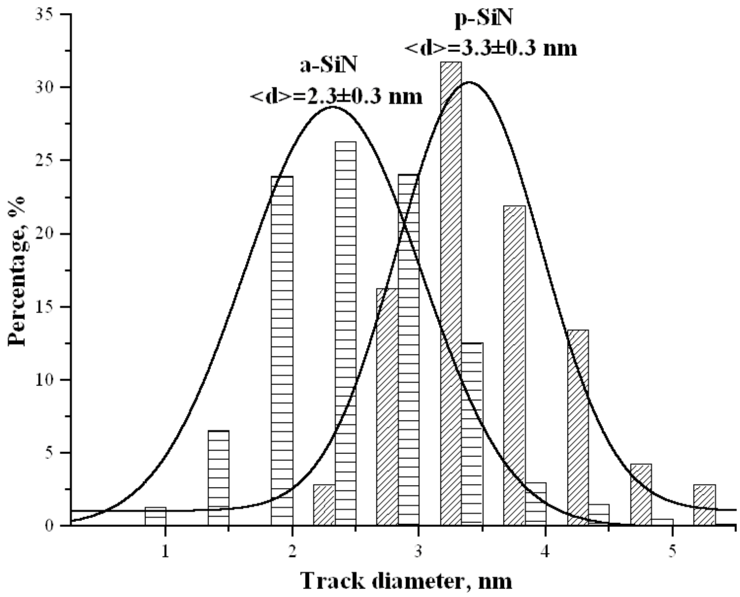

Fig. 2. Distribution of latent track sizes formed in amorphous and polycrystalline Si_3N_4 irradiated with 710 MeV Bi up to $5x10^{11}$ cm^{-2}

Discussion

As has been mentioned, the inelastic thermal spike (iTS) is the most often used in analysis of experimental data devoted to SHI impact. It is based on two main equations describing the energy exchange between electronic and ionic subsystems in solids [4, 5]:

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

$$C_a(T_a)\frac{\partial T_a}{\partial t} = \frac{1}{r}\frac{\partial}{\partial r}\left(rK_a(T_a)\frac{\partial T_a}{\partial r}\right) + g(T_e\text{-}T_a)$$

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.

Unfortunately, despite iTS model has been successfully applied to determine track parameters in variety of materials, its standard variation, where track formation is explained by quenching of a molten region around ion trajectory, predicts incorrect track size in Si_3N_4 [4, 6]. Kitayama et al. has suggested to use two threshold variation of i-TS where a track is believed to have a "core-shell" structure. According to it, a track core is formed as a result of evaporation, whereas a shell is a melted region. Figures 3 and 4 present results of calculations and our experimental results together with from [4] as a function of electronic stopping power.

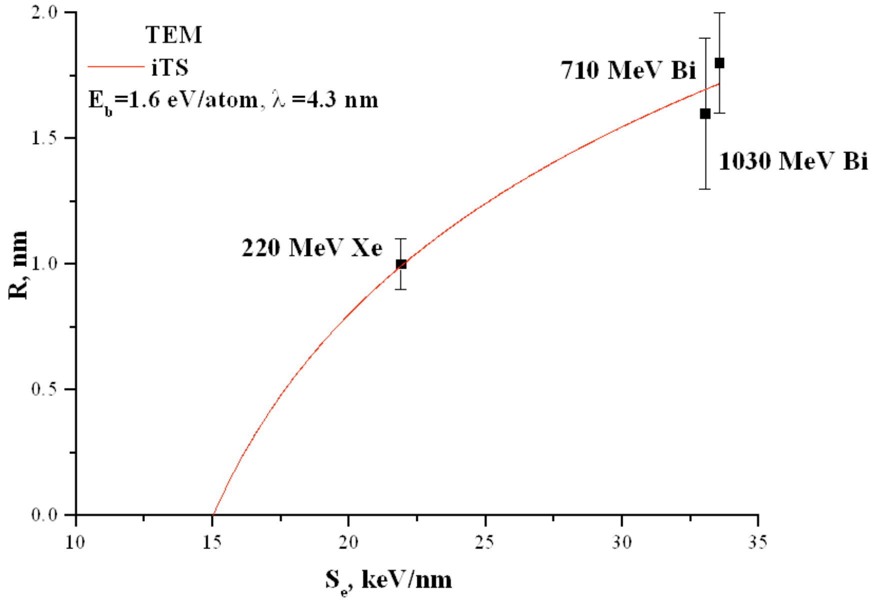

Fig. 3 The core radii in polycrystalline Si_3N_4 as a function of electronic energy loss with iTS fitting line

As was found, experimental track radii in polycrystalline silicon nitride may be successfully predicted by i-TS with the electron–phonon coupling constant λ=4.3 nm and the boiling energy E_b= 1.6 eV/atom. The energy value value corresponding to evaporation/boiling process has been considered as free because we failed to find correct data for Si_3N_4. For amorphous films free parameters λ=3 nm and E_b=2.5 eV/atom which are in a good agreement with results in []. Constants of the electron-phonon interaction were chosen as recommended on basis of the empirical relation between λ and bandgap. Thus, the threshold energy loss for a track core formation is found at ~15 keV/nm for p-SiN and ~13 keV/nm for a-SiN.

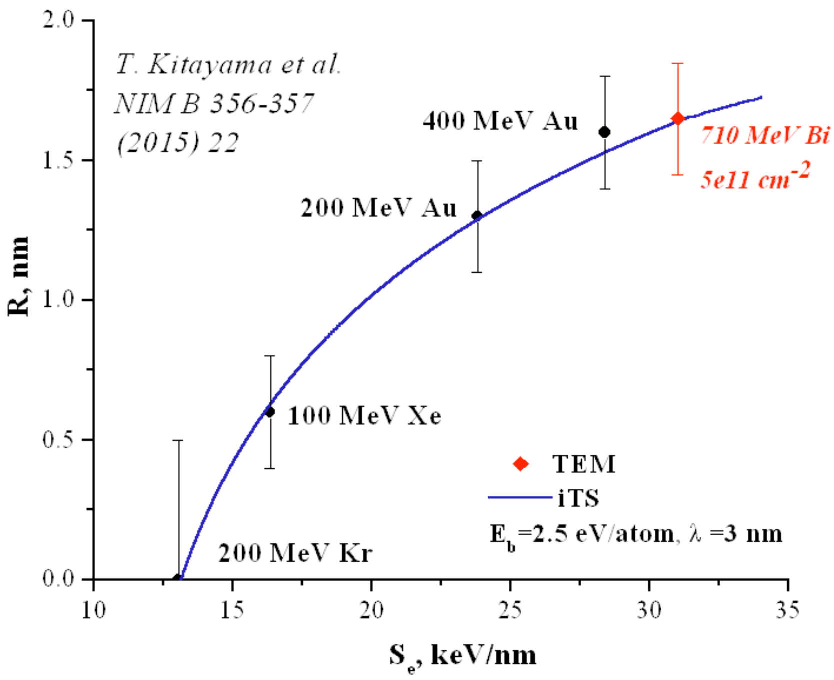

Fig.4 Track core radii in amorphous Si_3N_4 as a function of electronic energy loss with iTS fitting line

Conclusion

Consequently, latent tracks were registered by TEM in Si_3N_4 irradiated with 710 Bi ions at fluence $5x10^{11}$ cm^{-2}. Experimentally determined track core radii were shown to be correctly predicted in the framework of the two threshold version of iTS model using parameter E_b=1.6 eV/atom and 2.5 eV/atom for polycrystalline and amorphous silicon nitride. The corresponding threshold electronic stopping powers were determined as ~15 keV/nm and ~13 keV/nm.

References:

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