Eurasian Journal of Physics and Functional Materials

Volume 9 | Number 1

Article 2

Pages: 16 - 20

3-29-2025

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A. Ibrayeva
Institute of Nuclear Physics, 050032 Almaty, Kazakhstan, a.d.ibrayeva@gmail.com

J. O'Connell
Center for High Resolution Transmission Electron Microscopy, 6001 Port Elizabeth, South Africa

A Janse van Vuuren
Center for High Resolution Transmission Electron Microscopy, 6001 Port Elizabeth, South Africa

V. Skuratov
Flerov Laboratory of Nuclear Reactions, 141980 JINR, Dubna, Russia

Recommended Citation

Ibrayeva, A.; O'Connell, J.; Vuuren, A Janse van; and Skuratov, V. (2025) "Annealing Of Irradiation Defects In Si3n4: Tem Examination," *Eurasian Journal of Physics and Functional Materials*: Vol. 9: No. 1, Article 2.
DOI: https://doi.org/10.69912/2616-8537.1237

ORIGINAL STUDY

Annealing of Irradiation Defects in Si_3N_4: TEM Examination

Anel Ibrayeva [a,b,*], Jacques O'Connell [c], Arno Janse van Vuuren [c], Vladimir Skuratov [d,e,f]

[a] Institute of Nuclear Physics, 050032, Almaty, Kazakhstan
[b] KIMEP University, 050010, Almaty, Kazakhstan
[c] Center for High Resolution Transmission Electron Microscopy, 6001, Port Elizabeth, South Africa
[d] Flerov Laboratory of Nuclear Reactions, 141980 JINR, Dubna, Russia
[e] National Research Nuclear University MEPhI, 115409, Moscow, Russia
[f] Dubna State University, 140090, Dubna, Russia

Abstract

The article presents the first results of the study of structural and morphological features of swift (220 MeV) xenon ion induced defects in polycrystalline Si_3N_4 during post-irradiation heat treatment. The approximate temperature range of ion track region recrystallization is defined as 300–800 °C, at 800 °C complete structural relaxation was observed in the *in-situ* and *ex-situ* modes. It was found that the track recovery process, as expected, depends on the grain size and thickness of the sample. For complete recrystallization in both studied modes, a short annealing time was required due to the nature of the initial defects (before heating).

Keywords: Swift heavy ions, Radiation defects, Ion tracks, High-resolution transmission electron microscopy, Annealing, Recrystallization

1. Introduction

In some solids, swift heavy ions (SHIs) are known to produce structural damages called latent tracks. These defects represent disordered amorphous areas sized of several nanometres in diameter around the ion trajectory. Post-irradiation thermal annealing may induce shrinking of amorphous regions and even complete relaxation of crystalline lattice. In other words, annealing can be considered as the reverse process of track formation [1,2]. Understanding the structural stability of these tracks over extended periods and at varying temperatures is crucial for evaluating the long-term stability of radiation-resistant materials, such as silicon nitride.

Si_3N_4 is one of the candidate materials for use as an inert matrix fuel host (IMF) for the burn-up of plutonium and other minor actinides i.e., nuclear waste [3,4]. An important property of IMF candidates is radiation stability under irradiation with fission fragments (FFs) [5,6]. SHIs are well suited to the simulation of FF radiation, since they possess masses and energies similar to those of FFs. Besides the simulation of FF damage, the effects of ion irradiation on the microstructure of materials are of interest in both science and technology. Radiation effects are one of the main limiting factors on the lifetime of reactor materials in nuclear applications. Therefore, the modelling/prediction of microstructural modification which results from ion irradiation holds substantial benefit [7].

Silicon nitride is the only nitride ceramic where amorphous ion tracks have been observed [8–10]. To the best of our knowledge, thermal heating-induced effects on track structures have been predominantly studied in oxides (e.g., Refs. [11–15]). In silicon carbide, dense electronic excitation has been shown to stimulate a process known as thermal recrystallization [16,17]. However, to date, no reports have been published on recovery processes induced by thermal annealing in nitride ceramics. This study focuses on the structural and morphological features of latent tracks in polycrystalline Si_3N_4 irradiated with Xe ions, using

Received 24 October 2024; revised 26 November 2024; accepted 6 December 2024.
Available online 29 March 2025

* Corresponding author.
E-mail address: a.d.ibrayeva@gmail.com (A. Ibrayeva).

https://doi.org/10.69912/2616-8537.1237

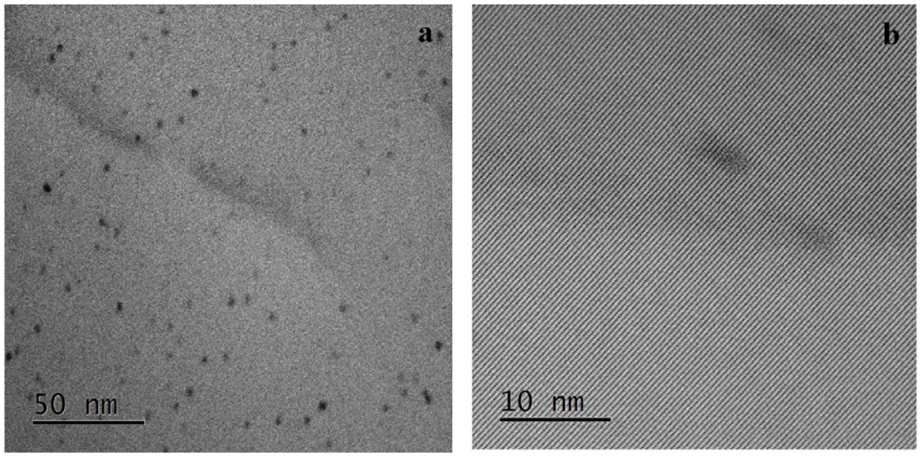

Fig. 1. BF TEM images of p-Si_3N_4 irradiated with 220 MeV Xe before annealing at: a) low magnification; b) high magnification.

high-resolution transmission electron microscopy (HR-TEM) in both *in-situ* and *ex-situ* modes. HR TEM allows to observe latent tracks directly and consequently provides insights into their characteristics and information about track annealing mechanism [15].

2. Materials and methods

Polycrystalline β-Si_3N_4 (ρ = 3.21 g/cm^3) doped with aluminium (3 at.%) were purchased from MTI Corporation (http://www.mtixtl.com). Samples were

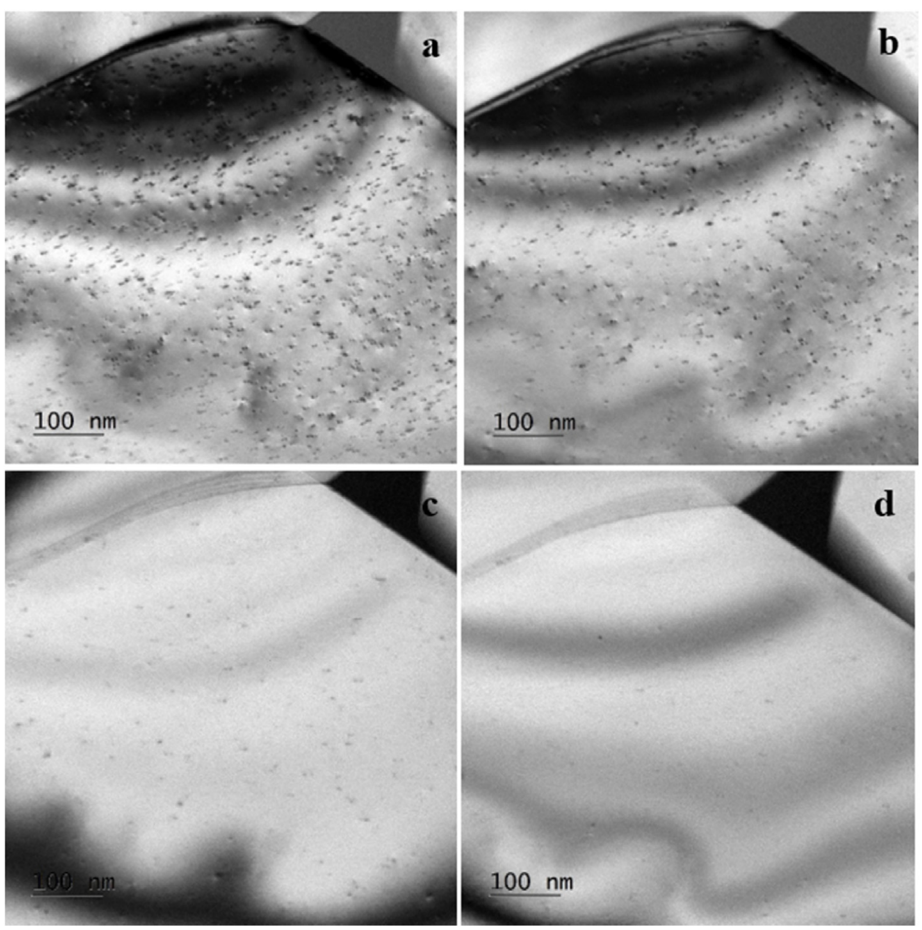

Fig. 2. BF TEM images of p-Si_3N_4 irradiated with 220 MeV Xe at different temperatures due to in-situ annealing at (a) a room temperature; (b) 300 °C; (c) 600 °C; (d) 800 °C.

irradiated with 220 MeV Xe at the DC-60 cyclotron at the INP in Astana, Kazakhstan. A fluence was 5×10^{11} cm^{-2} corresponding to a single-track mode. Irradiation was carried out at a room temperature.

TEM lamellae were prepared by means of FIB lift-out using an FEI Helios Nanolab 650. To minimise ion beam induced damage (by FIB), the samples were polished with Ga ion energies down to 500 eV. The process of a structural transformation was observed by imaging suitable specimens in a JEOL ARM 200F TEM operated at 200 kV at the Centre for HRTEM, Nelson Mandela University (Port Elizabeth, South Africa). Specimen were annealed in the *in-situ* regime using a Dens heating holder in the TEM and in the *ex-situ* using a tube furnace under an argon flow. Temperature range was 100—1000 °C. Annealing was carried out for 10 s (*in-situ*) and 20 min (*ex-situ*) at each temperature.

3. Results and discussion

In Si_3N_4 irradiation with 220 MeV Xe produces crystalline and amorphous defects as was reported in our previous works [8,10,18]. A typical bright-field (BF) images at low and high magnification are presented in Fig. 1.

Figs. 2 and 3 demonstrate typical bright-field (BF) TEM images of Si_3N_4 during *in-situ* and *ex-situ* annealing, correspondingly.

Dark diffraction contrast in bright-field (BF) TEM images indicates the presence of ion defects, even in cases of minimal amorphization. In *in-situ* mode the track density significantly decreases at 600 ° C (Fig. 2c), and at 800 ° C, the diffraction contrast becomes barely visible (Fig. 2d), so we assume that the lattice strain has been largely recovered. These images were obtained under the same diffraction conditions allowing for a comparable assessment of defect concentrations. Clearly, that the higher temperature the lower number of tracks is detected (Fig. 2). Despite it was impossible to examine the same grain in *ex-situ* specimens a similar trend of structural dynamics is assumed (Fig. 3). It is undoubtfully proved with a shrinking of amorphous areas shown in Fig. 4.

The defect formation process in a crystalline material is known to be dependent on a size of grains and a thickness of a sample. It is explained due to more

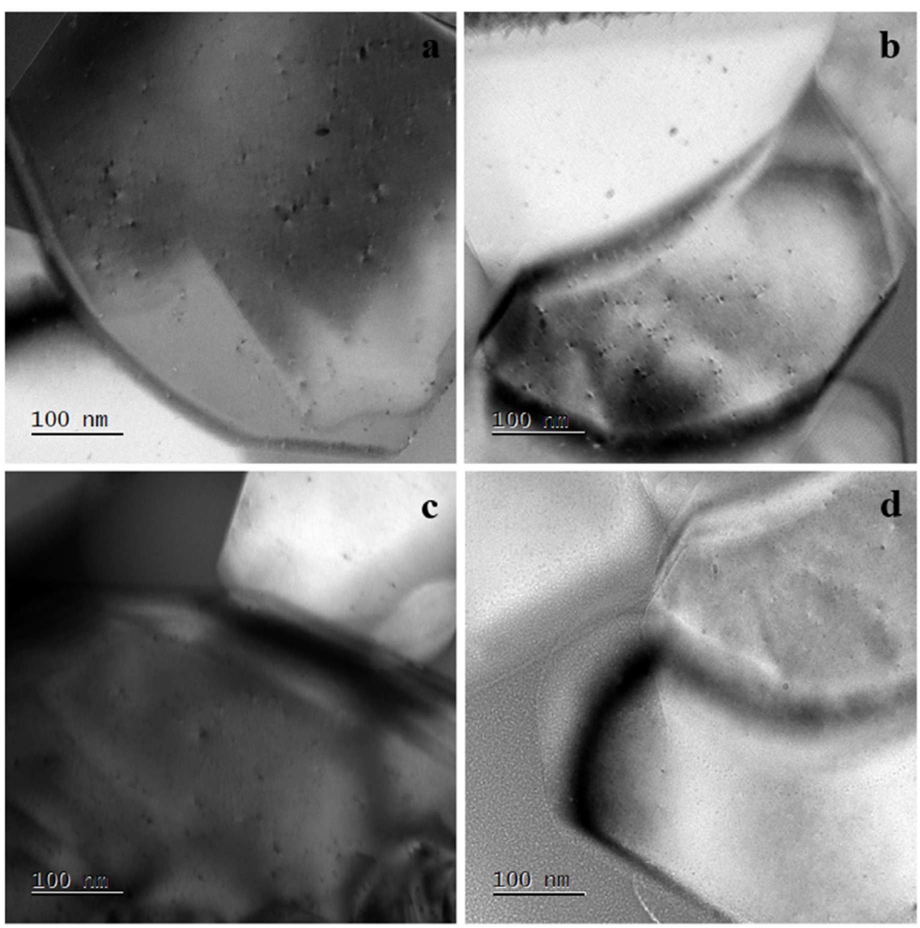

Fig. 3. BF TEM images of p-Si_3N_4 irradiated with 220 MeV Xe at different temperatures due to ex-situ annealing at (a) a room temperature; (b) 300 °C; (c) 600 °C; (d) 800 °C.

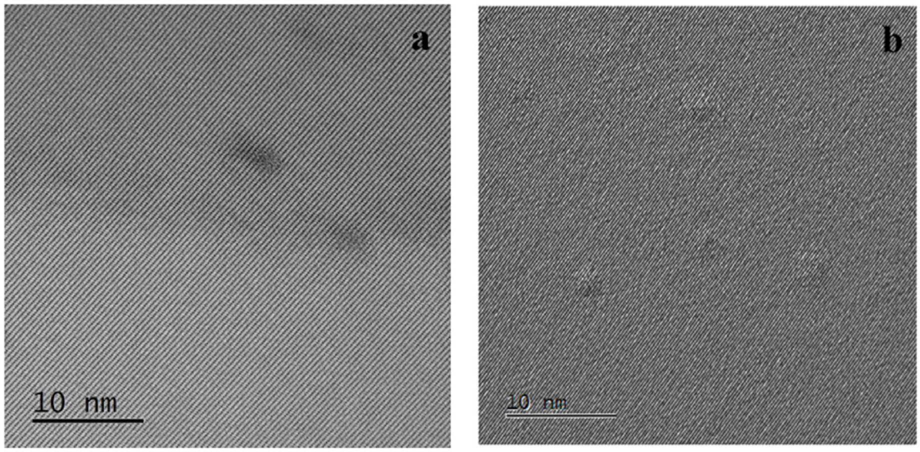

Fig. 4. BF TEM images of p-Si_3N_4 irradiated with 220 MeV Xe at different temperatures due to ex-situ annealing: at (a) a room temperature; (b) 800 °C.

localized thermal transport a structure gets damaged easier as was discussed in Refs. [19,20]. The recovery process, mentioned as the reverse of the defect formation is expected to follow a reverse evolution of defects within grains. The lower magnification TEM image of samples (Fig. 5) supports this statement complete relaxation of defects is observed in the smaller grain (compared to others), as well as in the boundary regions of larger grains.

It should be noted that annealing duration required for complete recrystallization in both modes was shorter than that typically observed in crystalline compounds, as described in, for example, [15,21,22]. We guess it is directly related to the crystalline nature of the most defects formed by the swift heavy ion impact before annealing. Therefore, the structural recrystallization of Si_3N_4 which has more amorphized regions (as a result of irradiation with ions of higher energy and/or at higher fluence) will require a higher temperature and/or longer heating duration.

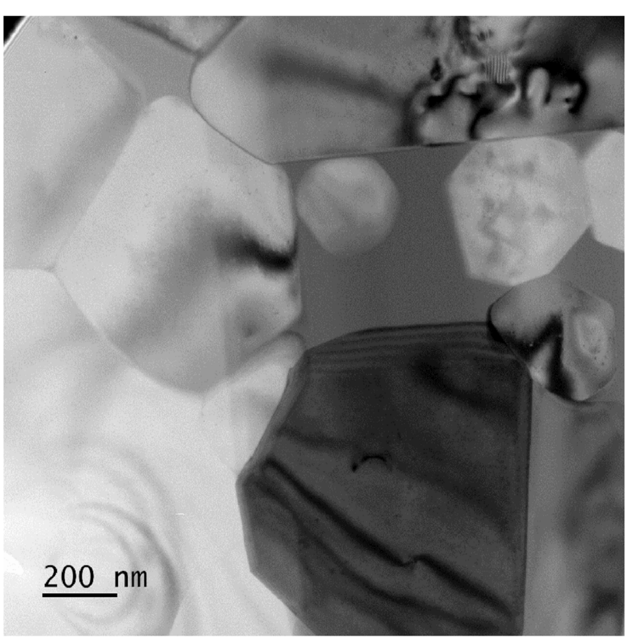

Fig. 5. BF TEM images of p-Si_3N_4 irradiated with 220 MeV Xe at 600 °C.

4. Conclusion

The approximate temperature range for recrystallization in polycrystalline silicon nitride is 300–800 °C for tracks formed by 220 MeV Xe irradiation. Complete recovery of the amorphous damaged regions in Xe-irradiated samples is observed at 800 °C in both *in-situ* and *ex-situ* heating modes. The data obtained in this study represent an initial step in understanding the dynamics of track recovery in polycrystalline silicon nitride. They are useful for further investigations of annealing-induced effects and subsequent evaluation of silicon nitride lifetime for nuclear applications.

Funding sources

This work was supported by the Ministry of Education and Science of the Republic of Kazakhstan [grant number AP19678955].

Conflict of interest

The authors declare no conflict of interest.

Acknowledgments

The authors thank DC-60 accelerators staff of the Institute of Nuclear Physics.

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