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MD and TEM evaluation of swift Xe ion induced latent tracks in Si_3N_4

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As known silicon nitride ceramics is considered as a candidate material for inert matrix fuel hosts used for transmutation of minor actinides. Unfortunately, by now very limited data is devoted to investigate of its stability under high energy heavy ion irradiation simulating fission fragment impact. Aim of our present study is a comparative analysis of parameters of latent tracks induced with swift Xe ion irradiation in polycrystalline Si_3N_4 using molecular dynamic (MD) simulation and high resolution transmission electron microscopy (TEM). Silicon nitride samples were irradiated with 220 MeV xenon ions at room temperature to fluence 5×10^{11} cm^{-2} that correspond to ion track non overlapping regime and allows to analyze single ion track regions. The calculated MD and experimental TEM values of track diameter for 220 MeV Xe irradiation are found to be in a good agreement and equal about 2 nm, whereas the threshold ionizing energy loss for track formation predicted by MD is lower than threshold value from TEM measurements (11.5 keV/nm vs 15 keV/nm). The averaging of all presented data gives the threshold energy loss level $\sim 13 \pm 2$ keV/nm.

Keywords: swift heavy ions, latent tracks, threshold energy loss, silicon nitride, MD, TEM.

Introduction

Structural sensitivity of dielectrics to swift heavy ions (SHI) irradiation is determined, first of all, by magnitude of specific ionizing energy losses required to form specific extended radiation defects, so called latent tracks. That is why experimental and theoretical studies aimed at finding of this threshold value is of considerable interest, especially for nuclear materials irradiating with fission fragments, i.e. atoms with a mass ranging from 80 to 155 and an energy of about 100 MeV. Among materials demonstrating very high resistance to such SHI impact are nitride-based ceramics. To date, latent track associated with radiation damages have been revealed in silicon nitride only [1-4]. At the same time, the data on latent track parameters are still very limited for both crystalline and amorphous Si_3N_4 and further work is needed to find out the threshold for track formation as well as the track size versus electronic stopping power, the irradiation temperature, structural state of material etc.

Nowadays, many experimental methods are available to study radiation damage in solids, among which transmission electron microscopy (TEM) is the only direct technique for observation and analysis of radiation-induced structural changes such as latent tracks. Another comprehensive approach to investigate a latent track morphology is molecular dynamic simulation (MD). This method has been successfully applied for describing processes of defect formation via relaxation of dense electronic excitations in some insulating materials, like Al_2O_3, MgO, YAG [5-8]. By our knowledge, this powerful technique has not been used for simulation latent tracks in silicon nitride yet.

In this work we compare: 1) track radii found from MD simulation and TEM examination; 2) threshold ionizing energy losses required for latent track formation predicted by MD and deduced from analysis of corresponding microscopic data.

TEM examination

Commercially available polycrystalline β-Si_3N_4 obtained from MTI Corporation was used in this investigation (Figure 1). The specimens were irradiated at room temperature with 220 MeV Xe ions to a fluence of 5×10^{11} cm^{-2} using facility at the DC-60 cyclotron, INF, Nur-Sultan. Ion beam homogeneity over the irradiated specimen surface was controlled using beam scanning in the horizontal and vertical directions and was better than 10%. TEM lamellas oriented parallel to the implanted surface were prepared by means of an FEI Helios Nanolab 650. To minimize ion beam induced damage the samples were prepared with ion energies down to 500 V. Structural analysis was done with a C_s corrected JEOL ARM200F transmission electron microscope operated at 200 kV.

Diameters of tracks formed in silicon nitride specimens as a result of irradiation with Xe ions were measured from high angle annular dark field (HAADF) STEM images (Figure 2). This type of micrographs is considered well suited for defect size determination

due to tracks being lower density areas are visible as darker spots than

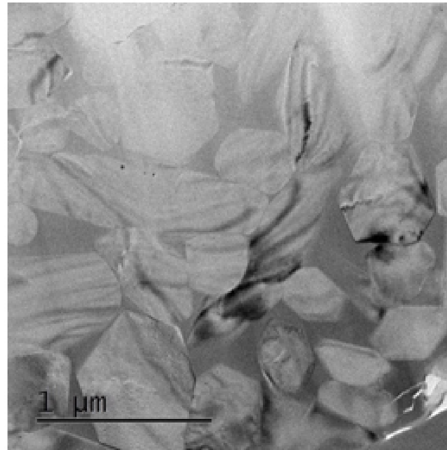

Figure 1. Bright field TEM image of polycrystalline Si_3N_4.

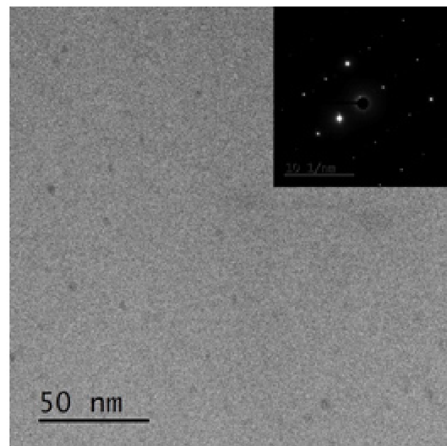

Figure 2. HAADF image of Si_3N_4 irradiated with 220 MeV Xe.

surrounding matrix. In the insert of Figure 2 selected area diffraction pattern is presented. It demonstrates that irradiated target keeps its crystalline structure under this Xe ion fluence. In addition, in our case tracks are obviously not completely amorphous, this is why not so sharp contract was got in images, that consequently leads increasing an inaccuracy of measurements. Nevertheless, to find the average track diameter a distribution of track sizes was fitted by Gaussian function, in this case the statistical reliability reached 95% (Figure 3). So, the average track size was equal $\sim 1 \pm 0.1$ nm in radius.

Besides the experimental point found in present work, results from [1] and [12, 13] were used to evaluate the threshold energy loss level (Figure 4). Fitting line was chosen from [12, 13] because of it describes the present work data quite well. Obviously, that three points are not enough for accurate fitting, however, they can give as at least some evaluating based on empirical results. So this parameter was determined at level of ~ 15 keV/nm.

MD simulation

The approach used in this study to simulate Xe ion induced latent tracks in silicon nitride is based on a hybrid scheme described the coupled kinetics

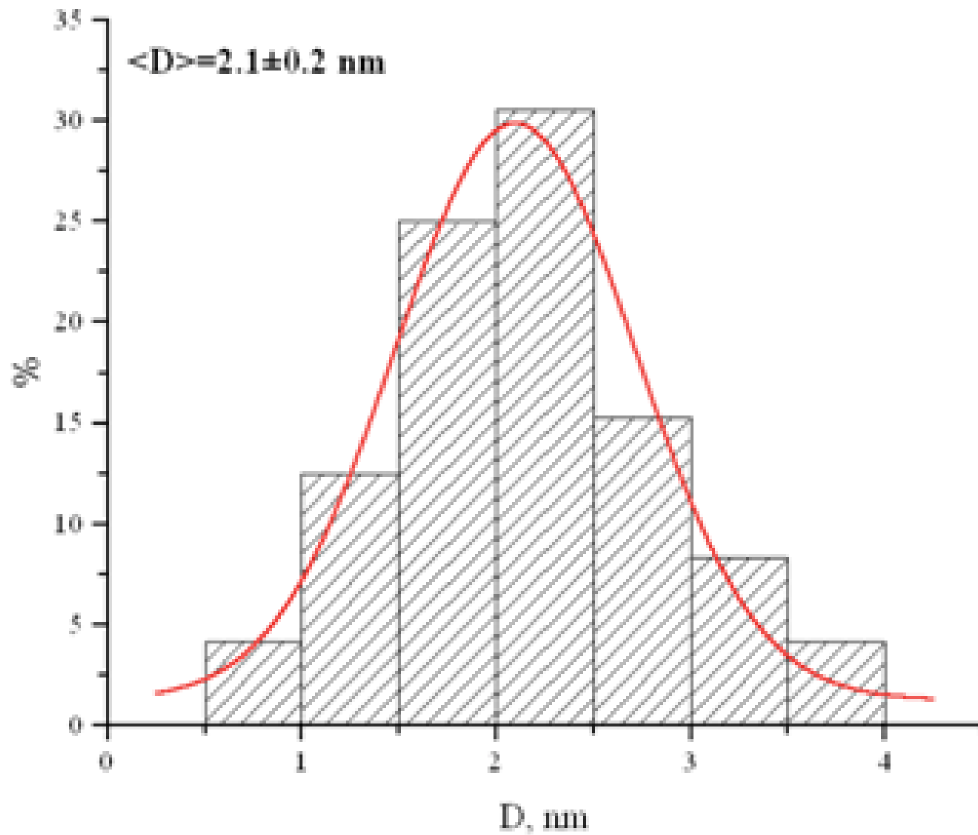

Figure 3. Distribution of track sizes of Si_3N_4 irradiated with 220 MeV Xe.

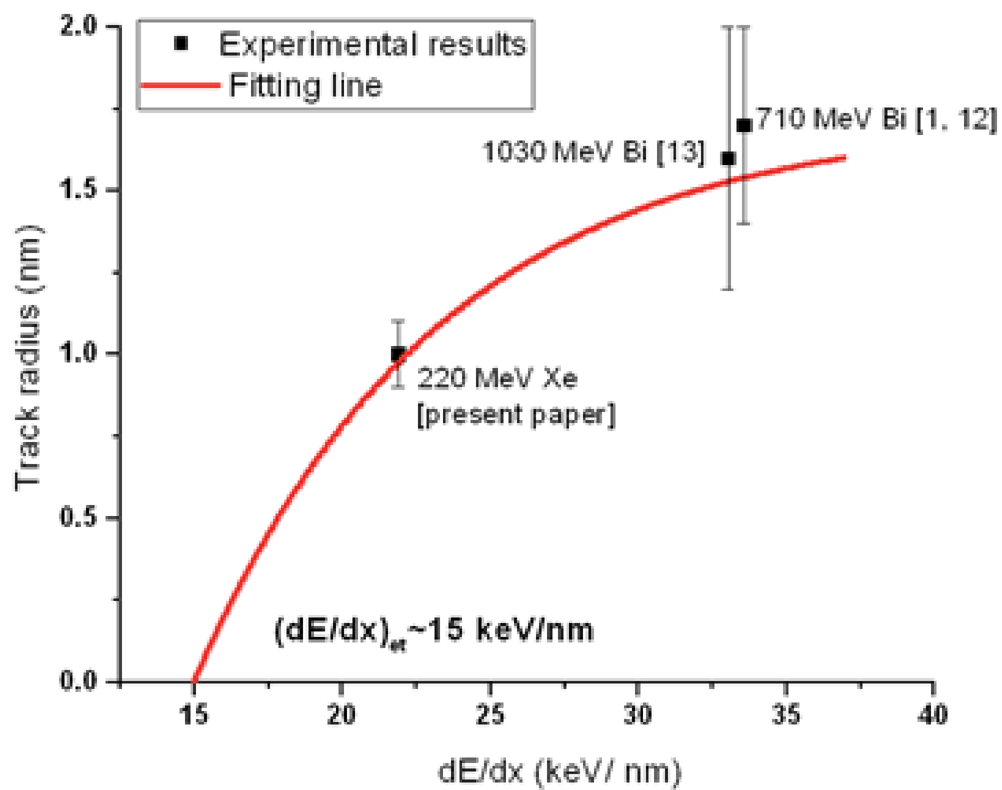

Figure 4. Radiation induced track radius in polycrystalline Si_3N_4 vs electronic stopping power.

of the excitation and relaxation of both electronic and the atomic sub-systems of irradiating material [7, 8]. To determine the initial parameters which are characteristic of an excited state of the ensemble of electrons as well as energy transferred to lattice atoms via electron-lattice coupling in vicinity of ion trajectory we used the asymptotic trajectory Monte Carlo code TREKIS [9, 10]. The calculated radial distribution of the energy transferred into the lattice, shown in Figure 5 for several Xe ion energies, was then used as input data for classical molecular dynamics code LAMMPS [11] to simulate subsequent lattice relaxation and structure transformations.

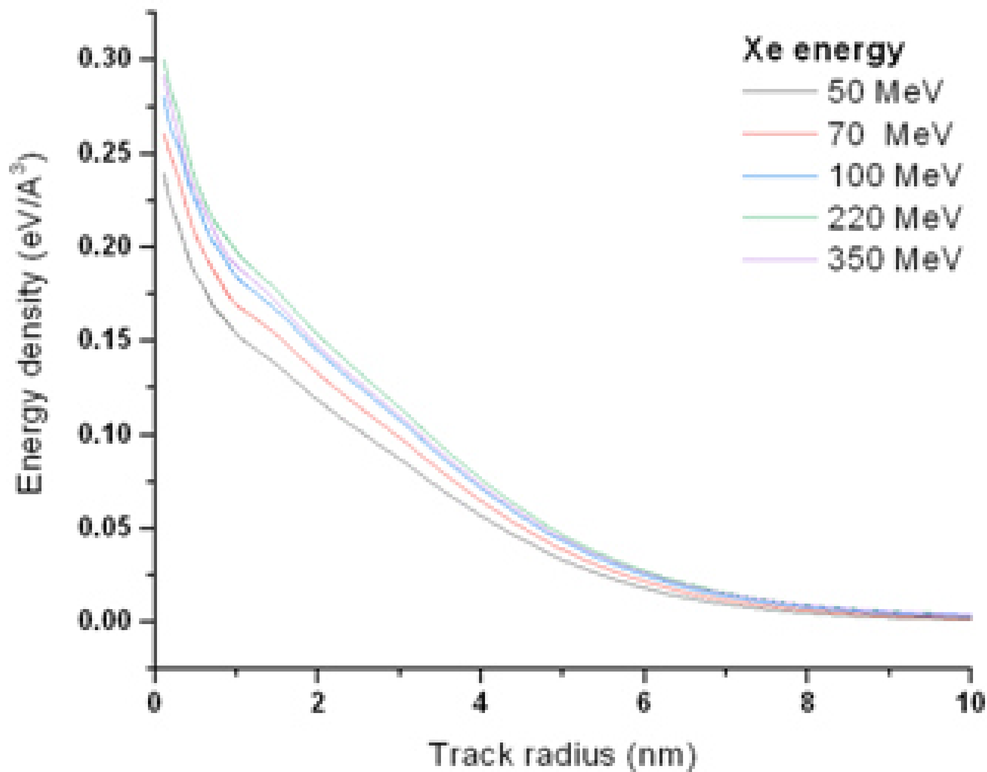

Figure 5. Radial energy distribution in electron sybsystem around of Xe ion trajectory in silicon nitride.

Figure 6 shows calculated dependence of specific ionizing energy loss using TREKIS code in comparison with SRIM data. As seen, TREKIS gives lower magnitude of energy losses. For example, the difference for 220 MeV is 2.5 keV/nm. In our opinion, such discrepancy, about 10%, is in the range of accuracy of SRIM code. As known, the accuracy can be even 20% for some combinations ion + target atoms [www.srim.org].

All MD simulations were done for β-phase structure of Si_3N_4. Interactions between atoms in Si_3N_4 are calculated using a Vashishta type potential with parametrization taken from [12]. Xe ion trajectories were parallel to Z axis of the cell ([001] direction of Si_3N_4 lattice). The supercell sizes used in the MD simulations were $24.2 \times 23.5 \times 19.9\ nm^3$ (1096000 atoms) with the periodic boundary conditions in all directions. All details of the applied methods can be found in [5-8]. Track evolution was traced up to 150 ps, after which the cell temperature dropped below 350 K, and no structural changes are expected after this time. Simulated atomic

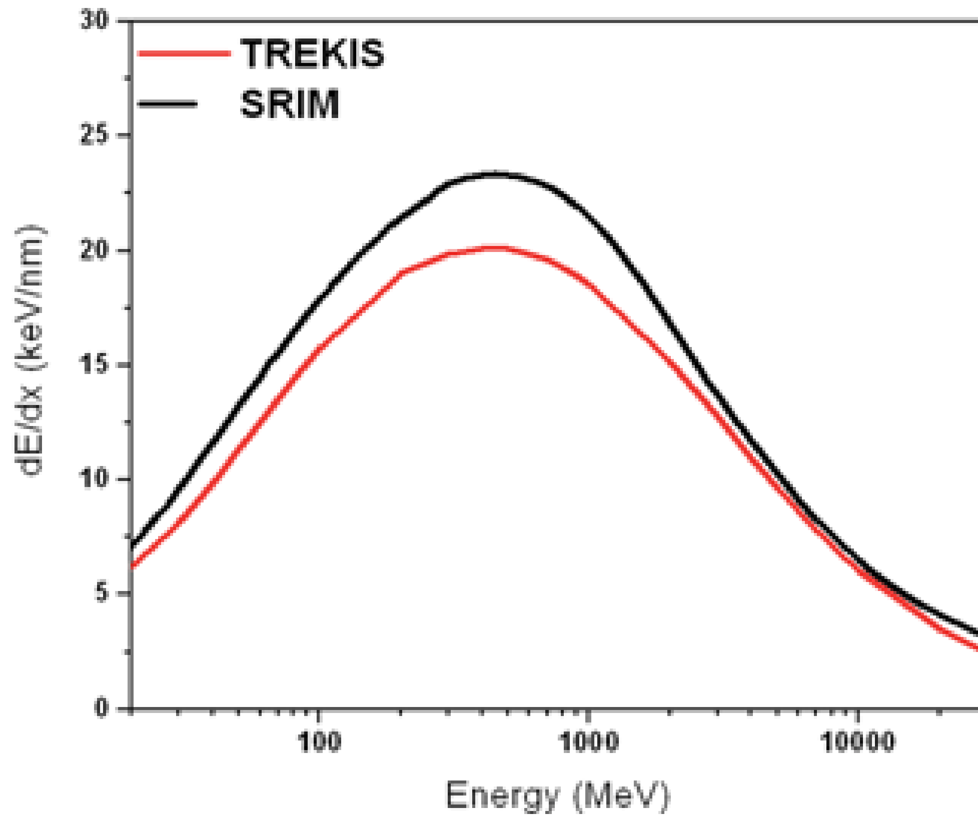

Figure 6. Electronic stopping power of Xe ions in silicon nitride calculated using TREKIS and SRIM codes.

snapshot of Si_3N_4 lattice in vicinity of Xe ion trajectories at 150 ps after ion impact are shown in Figure 7a-d.

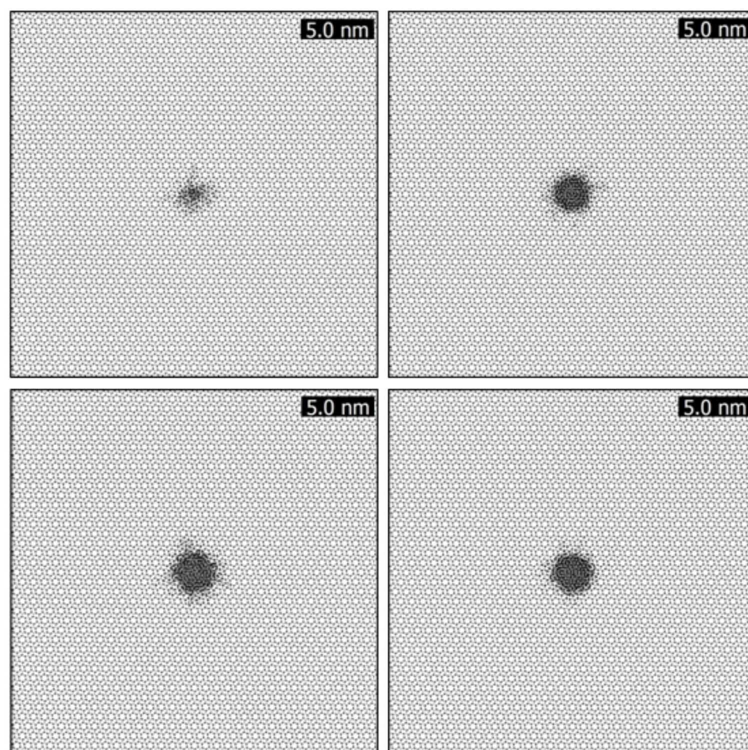

Figure 7. Snapshots of simulated latent tracks for different energies at 150 100 fs after Xe ion passage.

Simulated atomic snapshot of Si_3N_4 lattice in vicinity of Xe ion trajectories at 150 ps after ion impact are shown in Figure 7a-d. As seen, MD simulation predicts the formation of latent tracks for Xe ion energies till 70 MeV, when they become similar to point defect clusters. Actually it is not possible to state if such disordered discontinuous regions surrounding ion trajectories, which remain crystalline at this ion energy, may be recognized or considered as latent tracks. At energy 100 MeV and above, that corresponds to dE/dx $\leq$ 15.9 keV, simulated tracks are completely amorphous and continuous. The size of tracks was determined from the density profile passing through the centre of a track region. Then profile has been fitted by a Gaussian hat function and the FWHM was used as the track diameter.

Track radii found from MD together with experimental data for high energy Xe and Bi ions received in current study and previous works [1, 12, 13] are presented in Figure 8.

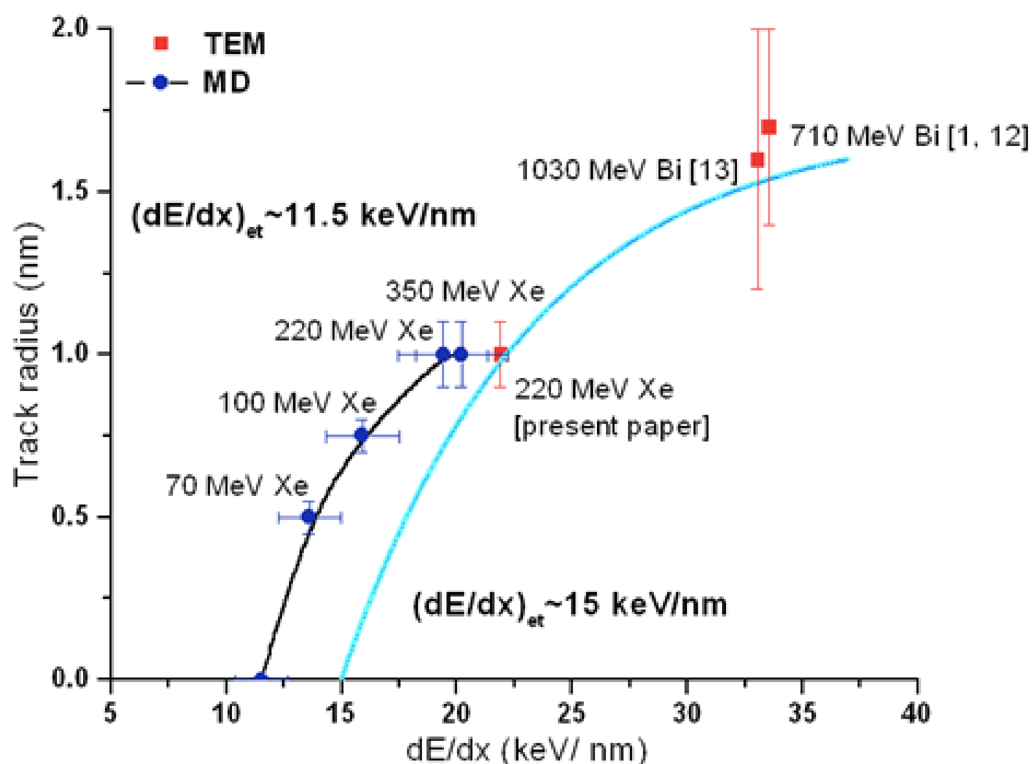

Figure 8. Calculated and experimental track radii as a function of electronic stopping power in silicon nitride.

One should note that no other data on tracks in crystalline Si_3N_4, besides shown in the Figure 8, are known from the literature. Taking into account the shift in electronic stopping power, one can state that calculated radius is in good agreement with experimentally determined track size for 220 MeV Xe ions. The results of simulation, as seen from Figure 7, imply that threshold value of ionizing energy loss necessary to form latent track in silicon nitride is about 11.5 keV/nm. This is lower than threshold value evaluated from approximation of experimental dependence. This dependence is based on limited number of experimental points and, evidently, exact determination of the threshold $(dE/dx)_{et}$ magnitude requires additional structural examinations using high resolution TEM, especially in the range of dE/dx $<$ 20 keV/nm. Further work is also needed to study the latent track

morphology in order to find threshold conditions when latent tracks in silicon nitride become discontinuous. Preliminary TEM results received on samples prepared in "cross-section" geometry show that we detect discontinuous tracks after irradiation with 167 MeV Xe ions. This will give additional criteria to evaluate the ability of molecular dynamic simulation to describe radiation damage formation in this material via relaxation of dense electronic excitations.

Conclusion

In framework of present study latent tracks induced by 220 MeV Xe irradiation have been investigated using transmission electron microscopy and molecular dynamics simulation. The average track radius values found by both approaches are in a good agreement and equal $\sim 1 \pm 0.1$ nm. However, the threshold energy loss levels are different for experimental data and calculation. Corresponding stopping powers have been determined as ~ 15 keV/nm and ~ 11.5 keV/nm, respectively. This difference can relate, first of all, to lack of available experimental data points, leading to some inaccuracy in determining the fitting line. Secondly, used parameters of Vashishta type potential may be not well-suited to a material with such complicated response of radiation induced properties as Si_3N_4. Thus, further investigation of silicon nitride under swift heavy ions impact is needed to specify a fitting line and consequently a level of threshold energy loss for track formation in this ceramic. By this moment, we could conclude that value $(dE/dx)_{et}$ could be between our determined points and evaluated as $\sim 13 \pm 2$ keV/nm.

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