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Application of pulsed multi-ion irradiations in radiation damage research: A stochastic cluster dynamics simulation study

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ABSTRACT

Under the multi-ion irradiation conditions present in accelerated material-testing facilities or fission/fusion nuclear reactors, the combined effects of atomic displacements with radiation products may induce complex synergies in the structural materials. However, limited access to multi-ion irradiation facilities and the lack of computational models capable of simulating the evolution of complex defects and their synergies make it difficult to understand the actual physical processes taking place in the materials under these extreme conditions. In this paper, we propose the application of pulsed single/dual-beam irradiation as replacements for the expensive steady triple-beam irradiation to study radiation damages in materials under multi-ion irradiation.

1. Introduction

Charged particle irradiation has been widely used to accelerate the testing process of materials used in nuclear applications. This approach has many advantages over neutron irradiation since it allows to achieve higher damage rates and shorten irradiation time. Besides the exclusion of radioactivity concerns, experimental conditions can be easily controlled in these ion-beam accelerators. However, charged particle irradiation in accelerated material testing facilities also comes with several disadvantages. The biggest drawback is that materials may not be subjected to simultaneous implantation of different element types as in actual nuclear reactors thus synergistic effects due to interactions between implanted species and radiation displacements may be overlooked as a consequence. Experiments carried out at the TIARA triple-ion facility in Japan show that complex synergies arise when Fe-Cr ferritic model alloys are irradiated with Fe^{3+},He^{+}, and H^{+} ions simultaneously [1]. The volume-swelling ratios measured in these alloys are significantly higher and noncumulative compared to those obtained from dual-ion irradiation experiments of Fe^{3+} with either He^{+} or H^{+} ion-implantation.

Recently, we introduce the Stochastic Cluster Dynamics (SCD) model that enables the simulation of complex defect evolution in materials under simultaneous multi-ion irradiations [2]. SCD obviates the need of solving the system of ordinary differential equations (ODEs) in traditional rate theory models, which are widely used to simulate radiation damage evolution in materials, by applying the Gillespie's SSA method to generate trajectories that are statistically correct samples of the probabilities of the system governed by the chemical master equation (CME) [3–5]:

$$\frac{\partial P(x,t|x_0,t_0)}{\partial t} = \mathscr{R}P(x,t|x_0,t_0) = \sum_{j=1}^{M} [a_j(x-\nu_j)P(x-\nu_j,t|x_0,t_0) - a_j(x)P(x,t|x_0,t_0)], \tag{1}$$

where $\mathscr{R}$ is the generating matrix for the Markov chain describing the chemical reactions, and $P(x,t|x_0,t_0)$ denotes the possibility that the population of defect cluster S_i at time t will be x given that at time t_0 is x_0.

Monte Carlo method is used to generate an ensemble of trajectories to form a basis for the statistical analysis. The time to the next reaction $\triangle t$ and the next reaction event R_k are determined as $\Delta t = -log(r_1)/\sum_j R_j$ and $\sum_{j=1}^{k} R_i > r_2 \sum_{j=1}^{M} R_j = r_2 R_{tot}$ where r_1,r_2 are two random numbers uniformly distributed in (0,1), and R_{tot} is the sum of all reaction rates R_j. This simulation step is repeated until the desired irradiation dose or irradiation time is reached. This process, however, can be computationally-intensive for large systems or large ensembles, and recent improvements have been made to SCD to accelerate the simulations and allow it to handle reactor relevant irradiation doses [6].

Using SCD, we carry out computer simulations of synergistic effects developed in pure bcc-Fe under Fe^{3+},He^{+}, and H^{+} multi-ion irradiations under conditions similar to those in Tanaka's experiments [7]. Our study suggests that only if we place a lower bound, or a 2 nm in diameter specifically, on the size of defect-clusters that may contribute to

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the volume-swelling level in the irradiated bcc-Fe samples then can we produce the synergistic effects observed in Tanaka's experiments. Otherwise, there is no significant difference in volume-swelling measurements in the simulated Fe^{3+},He^{+}, and H^{+} triple-ion irradiation and the simulated Fe^{3+}, He^{+} dual-ion irradiation case. Since many small defects are still irresolvable even in the most advanced microscopes nowadays, the disagreement between our simulations and Tanaka's experiments at small scales may directly arise from the difference in the way we account for the contribution to volume-swelling levels by these small defect-clusters. Since we can track every defect in our simulations, all of them contribute to the volume-swelling in our calculation. Therefore, we suggest more experimental quantification of volume-swelling ratio due to small defect-clusters formed under dual and triple-ion irradiations experiments should be performed to validate the presence of synergistic effects observed in Ref. [1] if they were not considered in Tanaka's previous report. Here, volume-swelling ratio is calculated as $\sum_i N_i/(\Omega V_0)$ with N_i is the number of vacancies in cluster S_i,Ω is the atomic density, and V_0 is the reaction volume.

Although triple-ion irradiation facilities may produce conditions comparable to those encountered in actual nuclear reactors, they usually come at high construction costs thus only a few of such facilities are available around the world at present. As a result, experimental investigations of materials for nuclear applications are carried out in less expensive dual-ion or single-ion irradiation facilities where the energetic elements are implanted sequentially instead of simultaneously. These approaches may not generally allow observing possible synergistic effects of He and H gas ions observed in Tanaka's experiments. An interesting question arises is whether it is possible to modify the configurations of existing single/dual-beam facilities to enable them to produce experimental conditions similar to those obtained in the scarce triple-beam irradiation facilities. A possible and inexpensive solution is implanting heavy ions and gas species into the samples in alternating pulses from one or two beamlines. Ion-irradiation experiments in facilities equipped with fast beam-switching systems were carried out earlier [8,9]. In dual-source mass analyzed low-energy ion beam systems, the length of irradiation pulses can be narrowed down to about 20 to 200 μs, the pulse frequency is adjustable in the range of 10–50 Hz, and the ion source switching can be completed within 100 ms using electromagnetic mass separation systems [9].

So far, most theoretical investigations of radiation damage evolution have been focused on steady irradiation with little attention paid to pulsed irradiation conditions. However, provided the inherent pulsing capabilities of existing and proposed irradiation facilities such the SNS at ORNL, MTS at LANL, IFMIF and ITER, both theoretical and experimental study of radiation damage under pulsed irradiations is of great interest. Simulation studies have been reported on the impact of pulsed irradiation on defect evolution in comparison to steady irradiation, it has been shown that significant differences between these two types of irradiation method [10–14]. However, these calculations were mostly performed only to very small doses, typically less than 0.1 dpa. Besides, these earlier studies did not go beyond single species implantation, and complex-structured defects were not even considered therefore simplified the problem significantly. With SCD, we can efficiently achieve reactor-relevant irradiation doses, handle complex-structured defects and vary simulation parameters to simulate a wide range of experimental conditions.

In this study, we apply SCD models to examine the feasibility of using pulsed single- or dual-beam irradiation to produce damage distributions that are similar to those formed under expensive steady triple-beam irradiation in pure bcc-Fe, thus suggesting cost-effective methods to verify the actual presence of synergistic effects observed in Ref. [1] that can be performed at existing accelerator facilities with fewer than three beamlines. To the best of our knowledge, these types of simulations have never been attempted before.

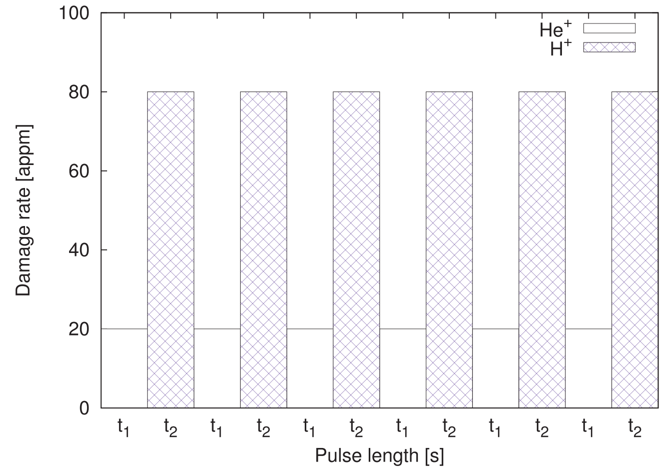

Fig. 1. Pulsed irradiation schematic of the second beamline in the dual-beam irradiation configuration. In the first beamline, Fe^{3+} emits at a steady dose rate of 1.6×10^{-3} dpa/s same as in the steady triple-ion irradiation case.

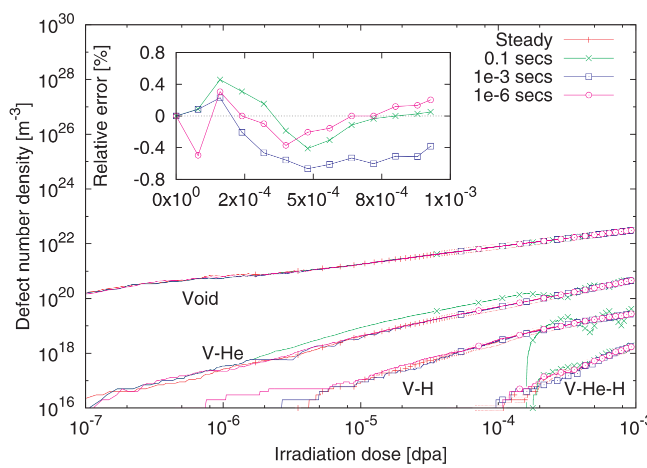

Fig. 2. Number density of various types of defects formed under pulsed dual-beam irradiations with short pulse-length compared to steady triple-beam irradiations. The inset shows relative differences in volume-swelling ratios obtained from these two irradiation methods.

2. Methods

We carry out simulations with similar conditions to those applied in Tanaka's experiments. The energies of Fe^{3+}, He^{+}, and H^{+} ions are 10.5 MeV, 1.05 MeV, and 0.38 MeV, respectively. The dose rate of Fe^{3+} is 1.6×10^{-3} s^{-1} and implantation rates of He^{+} and H^{+} are 10 appm/dpa and 40 appm/dpa. The reaction volume is 10^{-13} cm^3, and all simulations are carried out at 783 K at which largest cavities are observed in Tanaka's experiments [1].

Here, the assumed dual-beam experiment configuration consists of two beamlines: first beamline consists of a steady Fe^{3+} source just like that in the triple-beam experiment configuration, the second beamline is connected to both He^{+} and H^{+} ion sources and can implant these gas ions in alternate pulses with pulse-lengths t_1 and t_2 as shown in Fig. 1. To compensate for the loss of He^{+} and H^{+} fluxes during their beam-off periods in the second beamline, we increase their implantation rates by a factor of $(t_1 + t_2)/t_1$ and $(t_1 + t_2)/t_2$, respectively. Since defect formation in the material is sensitive to the He^{+}: H^{+} implantation ratio, it is important to keep this ratio consistent with that in the triple-beam irradiation case to produce the same results [7].

Besides pulsed dual-beam irradiation, we also investigate the possibility of further simplifying the accelerator configurations by reducing the number of beamlines to only one. We perform SCD simulations of

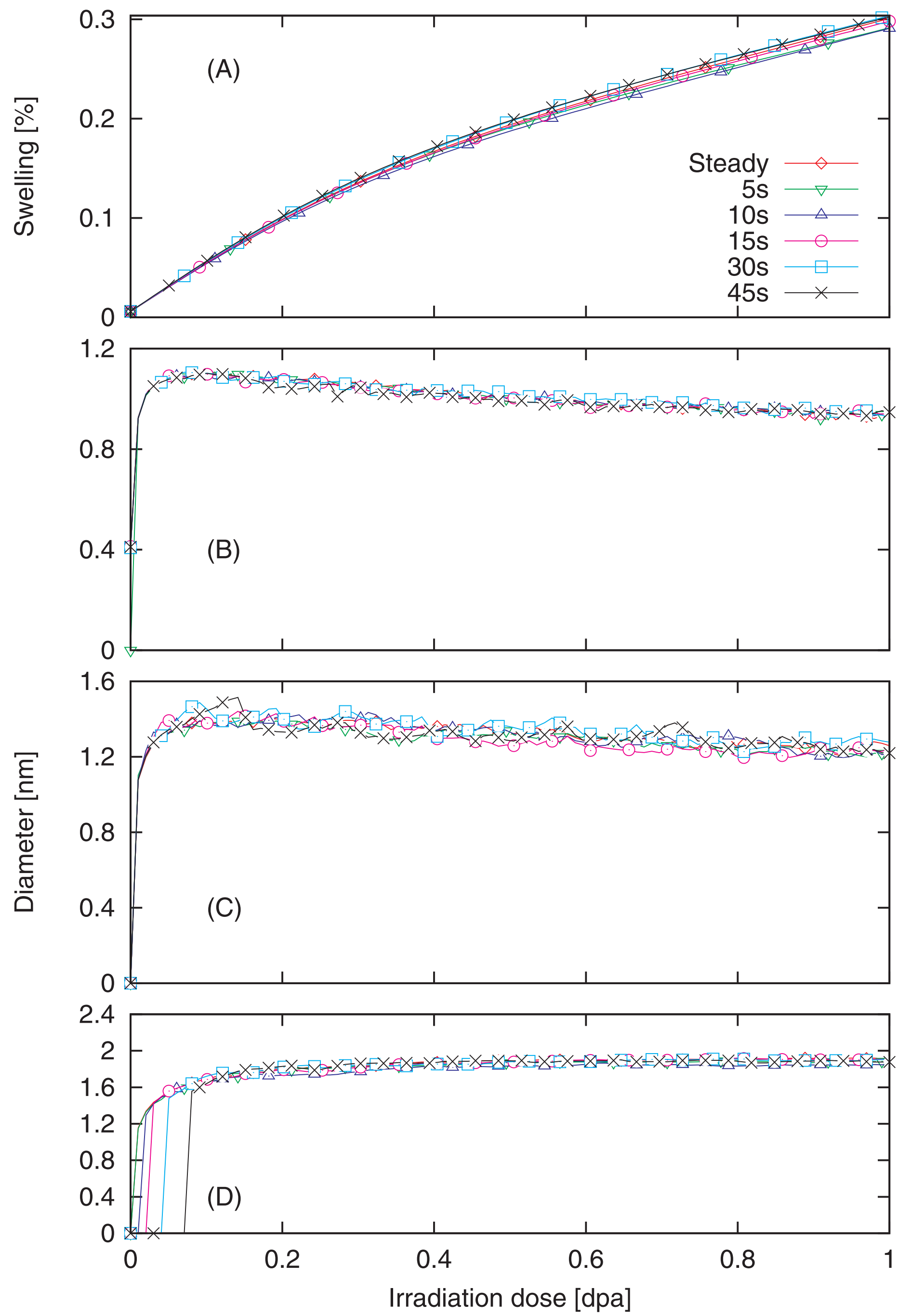

Fig. 3. Volume-swelling (A) and average sizes of various types of defects, such as voids (B), V-He clusters (C), and V-He-H clusters (D), formed in bcc-Fe under pulsed dual-beam irradiations with various pulse-length compared to steady triple-beam irradiations at 783 K.

pulsed single-beam irradiations in place of triple-beam irradiation in which all Fe^{3+},He^{+} and H^{+} ions are implanted in separate pulses housed in one single beamline. Similar to the pulsed dual-beam irradiation case, the dose rate of incident ion in each pulse is adjusted accordingly to compensate for the loss of flux during the beam-off periods. The new irradiation dose rate of Fe^{3+} is $\Gamma' = (t_1 + t_2 + t_3)/t_1\Gamma$, and the new implantation rates of He^{+} and H^{+} gas ions are also increased by factors of $(t_1 + t_2 + t_3)/t_2$ and $(t_1 + t_2 + t_3)/t_3$, respectively with t_1,t_2, and t_3 are the lengths of the Fe^{3+},He^{+}, and H^{+} pulses, respectively, and Γ is the irradiation dose rate in the reference steady triple-beam case. The simulation conditions are the same as those applied in the previous pulsed dual-beam simulations with an irradiation temperature of 783 K and total damage dose of 0.15 dpa. Here, for the sake of simplicity, we set the Fe^{3+}, He^{+} and H^{+} pulse-lengths be the same thus the corresponding irradiation fluxes are tripled in this pulsed single-beam irradiation case. Similar to the pulsed dual-beam irradiation case, we start with short pulse simulations to examine if pulsed single-beam irradiation can reproduce results obtained from the reference steady triple-beam irradiation case before pursuing the more expensive simulations with longer and more practical pulses, from 0.1 s to 5 s. These simulations with very short pulse-lengths allow us to verify the fidelity of our model.

3. Results and discussion

Fig. 2 shows the population densities of various types of defect clusters formed in bcc-Fe under pulsed dual-beam irradiation as functions of dose and pulse-length, and the relative differences in the

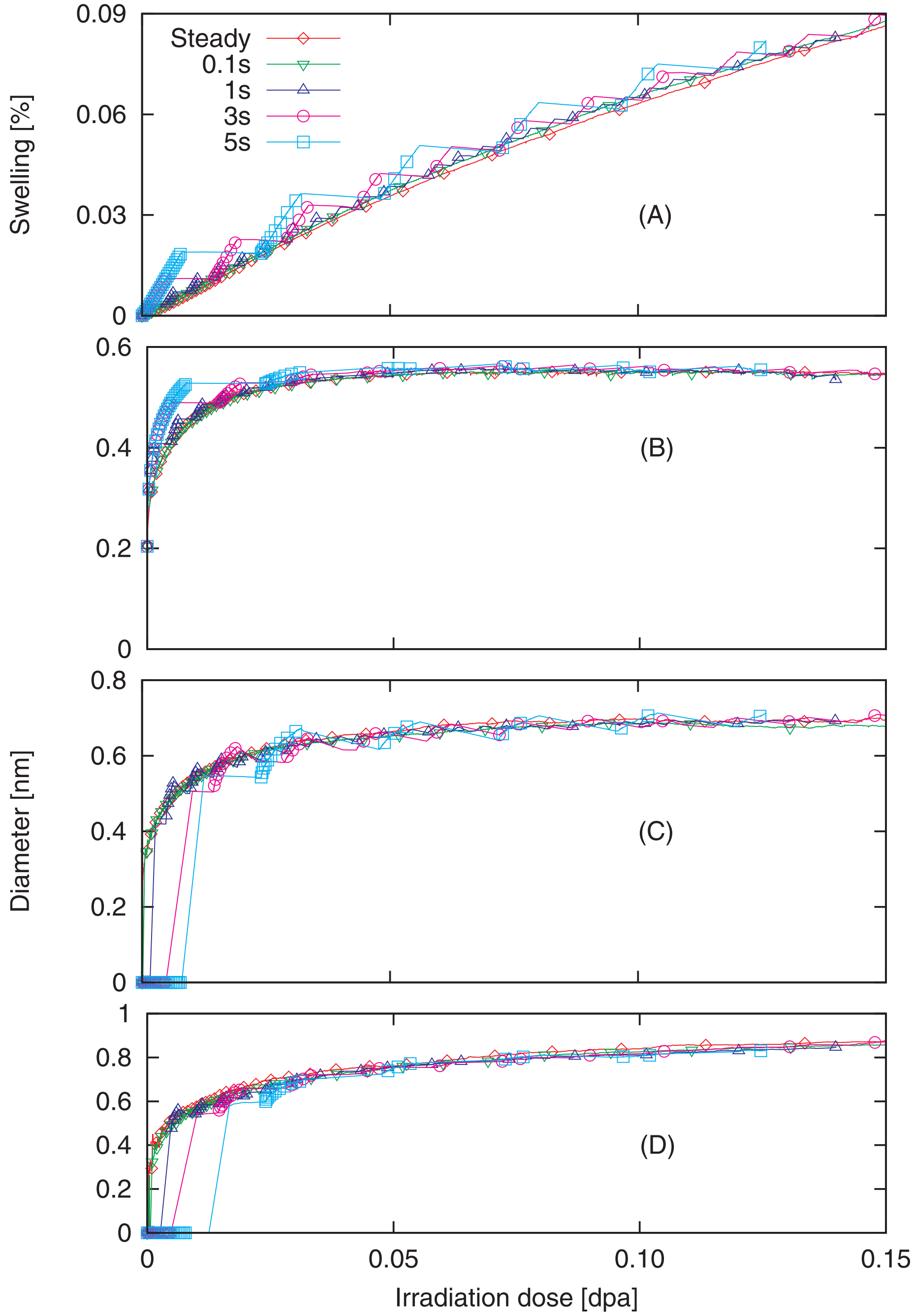

Fig. 4. Volume-swelling (A) and average sizes of various types of defects, such as voids (B), V-He clusters (C), and V-He-H clusters (D), formed in bcc-Fe under pulsed single-beam with various pulse-length compared to steady triple-beam irradiations at 783 K.

predicted volume-swelling ratios compared to the one obtained under steady triple-beam irradiation is shown in the inset. We start with short pulse simulations to examine if pulsed dual-beam irradiation can reproduce results obtained from the reference steady triple-beam irradiation case before pursuing the more expensive simulations with longer and more practical pulse-lengths. Another advantage of performing short pulsed simulations is that they allow us to verify the fidelity of our model, i.e. with very short pulses, results obtained from pulsed simulations must converge to the steady irradiation case. In these simulations, we set t_1 and t_2 both equal to 1 µs, 1 ms or 0.1 s, respectively. As a consequence, the implantation rates of He^+ and H^+ gas ions in each of these pulses are doubled compared to the corresponding values in the steady triple-ion irradiation case, to 20 appm/dpa and 80 appm/dpa, respectively.

As can be seen in Fig. 3, pulsed dual-beam irradiations produce similar results to those obtained from steady triple-beam irradiation. As we shorten the pulse-lengths t_1 and t_2, the results converge to the reference triple-ion irradiation case. It can be seen from the inset of Fig. 2 that the differences in the predicted volume-swelling ratios obtained from pulsed dual-beam irradiations are within 0.5% relative to that of the triple-beam case. The average sizes of defect clusters are also observed to be nearly identical between these two irradiation methods.

Even though dual-beam irradiations with short pulses produce results closest to those obtained from steady triple-beam irradiation, they are not very practical since designing and implementing rapidly switching ion sources can pose significant technical challenges. Therefore, we also examine whether it is possible to extend these pulses thus reducing the switching times of the He^+,H^+ ion sources. Fig. 3

shows the overall volume-swelling ratios and average sizes of various defect types obtained in bcc-Fe under pulsed dual-beam irradiations with longer pulses, up to 45 s, and higher doses, up to 1 dpa, and compared these values to the reference steady triple-beam case. It can be seen that, even with long pulses, results obtained from these two different irradiation methods are still convergent. Therefore, on the basis of our SCD simulation results, we suggest that pulsed dual-beam irradiations, with pulse-length up to 45 s, can be used in place of triple-beam irradiation for investigation of radiation damage evolution in bcc-Fe under Fe^{3+},He^{+}, and H^{+} irradiation. To justify our suggestion, experimental validation will be of great value.

As can be seen in Fig. 4, pulsed single-beam irradiation can also reproduce results obtained from steady triple-beam irradiation. It is apparent that defect populations formed under pulsed single-beam irradiations approach those of the steady triple-beam irradiation reference case. Better agreement between these two irradiation methods is achieved at short pulse-lengths, which is reasonable since pulsed single-beam irradiation effectively converges to steady-state triple-beam irradiation when the pulse-lengths are shorten. Similar to the pulsed dual-beam irradiation case, we examine if this agreement still holds at longer pulse-lengths. Fig. 4 also shows the volume-swelling and average sizes of various defect types obtained in bcc-Fe under pulsed single-beam irradiation with longer pulses, up to 5 s. Compared to the pulsed dual-beam irradiation results shown in Fig. 3, the curves in Fig. 4 exhibit some oscillations, larger amplitudes with longer pulses, but they do not deviate from the corresponding steady triple-beam curves. Therefore, it is reasonable to conclude that, from the simulation point of view, pulsed single-beam irradiation can also be used in place of steady triple-beam irradiation or at least can provide us reliable estimates of the results that would be obtained from the steady triple-beam irradiation experiments.

4. Conclusion

We apply the SCD models to examine if pulsed single or dual-beam irradiation can potentially be used in place of steady triple-beam irradiation to test materials used for nuclear applications. These findings provide us alternative options to investigate the complex kinetics taking place in actual nuclear materials under different irradiation conditions, whenever the access to triple-beam testing facilities is limited. For the pulsed dual-beam configuration, He^{+} and H^{+} gas ions are implanted into the material in alternating pulses whereas the Fe^{3+} ion source is kept steady as in the steady triple-ion irradiation case. With pulsed single-beam configuration, Fe^{+}, He^{+}, and H^{+} ions are all implanted into the materials in alternating pulses. The most important requirement is that Fe^{+},He^{+}, and H^{+} implantation rates must be adjusted accordingly to compensate for the loss of irradiation fluxes due to the pulse nature of these beamlines. The pulse-length is also important, short pulses provide results closer to those obtained from the steady triple-beam reference case. Our current objective is to explore if less expensive ion-beam configurations can substitute the expensive and limited access triple-beam accelerators for studying of complex microstructure evolution in nuclear materials, they are especially suitable for validating experimental results report by Tanaka et al. in Ref. [1]. Exactly how other parameters such as irradiation dose rate, temperature or ion pulse order, length and frequency of pulses will eventually affect the defect populations or the synergies between gas ions, and under which conditions pulsed irradiations will fail to replicate steady irradiations are the subjects of ongoing research.

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