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LAWRENCE LIVERMORE NATIONAL LABORATORY

Measuring radiation damage dynamics by pulsed ion beam irradiation

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1 Cover page data

1.1 Federal Agency and Organization Element to Which Report is Submitted

U.S. Department of Energy, Office of Nuclear Energy, NEET-Reactor Materials.

1.2 Federal Grant or Other Identifying Number Assigned by Agency

Work package # CT-14LL040514. Work authorization number NT/0114/14/AL/50.

1.3 Project Title

Measuring radiation damage dynamics by pulsed ion beam irradiation.

1.4 PI Name, Title, and Contact Information

Sergei Kucheyev, physicist, kucheyevllnl.gov, phone: (925) 422-5866.

1.5 Name of Submitting Official, if other than PD/PI

See Sec. 1.4.

1.6 Submission Date

December 30, 2014.

1.7 DUNS Number

Unknown.

1.8 Recipient Organization (Name and Address)

Lawrence Livermore National Laboratory, 7000 East Ave, Livermore, California 94550.

1.9 Project/Grant Period (Start Date, End Date)

Start date: January 1, 2014. End date: December 31, 2016.

1.10 Reporting Period End Date

December 30, 2014.

1.11 Report Term or Frequency (annual, semi-annual, quarterly, other)

Annual.

1.12 Signature of Submitting Official

Sergei Kucheyev

2 Accomplishments

2.1 Major goals of the project

The major goal of this project is to develop and demonstrate a novel experimental approach to access the dynamic regime of radiation damage formation processes in nuclear materials. In particular, the project exploits a pulsed-ion-beam method in order to gain insight into defect interaction dynamics by measuring effective defect interaction time constants and defect diffusion lengths. For Year 1, this project had the following major milestone: demonstration of the concept of the pulsed ion beam method for a prototypical nuclear ceramic material, SiC. The target date for this milestone is January 1, 2015. As we describe below, this milestone has been met.

2.2 Specific accomplishments for the reporting period

Major activities. This was the first year of this project. Major activities consisted of (i) the acquisition of high purity SiC single crystals; (ii) development and testing of a variable-temperature sample holder with accurate dosimetry, (iii) ion bombardment of Si and SiC crystals under well controlled pulsed-ion-irradiation conditions; and (iv) advanced characterization of irradiated targets by ion channeling. We performed pulsed ion beam experiments with both SiC (a prototypical nuclear ceramic material) and Si (the best studied and arguably the simplest material) targets. By doing it, we investigated radiation damage dynamics in SiC and tested the robustness of the pulsed ion beam method (with both Si and SiC targets).

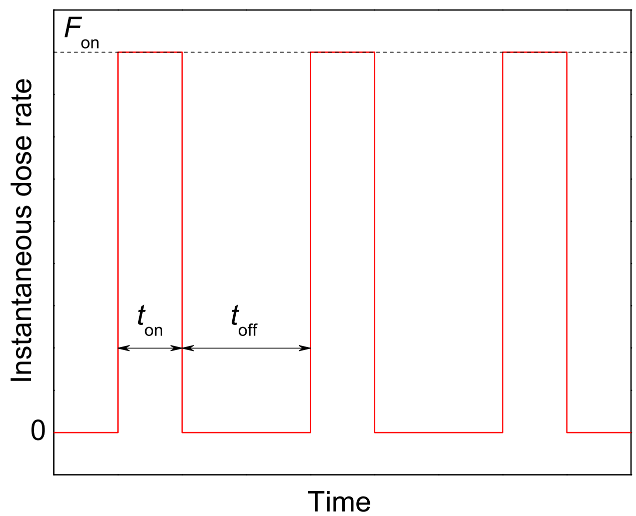

Figure 1: Schematic of the time dependence of the dose rate, defining t_{on}, t_{off}, and F_{on} (the maximum instantaneous dose rate).

Specific objectives. The specific objective was to demonstrate the robustness of the concept of the pulsed ion beam method and apply it for the best studied and readily commercially available nuclear ceramic material, SiC.

Significant results. We executed a successful series of pulsed ion beam irradiation experiments that demonstrated the robustness of the pulsed ion beam method for studying radiation defect interaction dynamics. Such pulsed ion beam experiments were aimed at direct measurements of effective time and length constants of defect interaction processes. The defect interaction time constant τ was measured directly by studying the dependence of lattice disorder on the passive part of the beam duty cycle, t_{off}, while the effective defect diffusion length L_{d} was estimated from the dependence of damage on the active part of beam cycle, t_{on}. The additional pulsed-beam-related irradiation parameters ($t_{\rm on}$, $t_{\rm off}$, and $F_{\rm on}$) are defined in Fig. 1, which shows a schematic of the time dependence of the dose rate during pulsed ion beam irradiation. In such pulsed-beam experiments, the total ion dose is split into a number of equal square pulses. These experiments involved ion irradiation of a series of specimens to the same total dose with all except one of the irradiation parameters fixed. Irradiation is followed by measurements of the level of stable disorder in the crystal bulk and at the surface with ion channeling spectrometry.

We performed systematic studies of radiation damage in Si and 3C-SiC bombarded with 500 kev Ar ions in the temperature range of $25-250$ °C in the pulsed ion beam mode. Figure 2 shows a summary of the buildup of bulk damage in 3C-SiC for different temperatures. Figure 3 shows the dependence of bulk damage in 3C-SiC on the duration of the passive part of the cycle, t_{off}, and all the other irradiation parameters fixed. It reveals a second order decay process for all the temperatures studied:

$$n(t_{off}) = n_\infty + \frac{n_0 - n_\infty}{1 + \frac{t_{off}}{\tau}},$$

where n is the maximum defect concentration measured by ion channeling in the crystal bulk where the nuclear energy loss is maximum. Figure 4 summarizes temperature dependences of τ and the dynamic annealing efficiency (DAE) in 3C-SiC bombarded with 500 keV Ar ions. Such a DAE is defined as $\frac{n_0-n_\infty}{n_0}$. It is seen from Fig. 4 that, while the DAE monotonically increases with increasing irradiation temperature, the $\tau(T)$ dependence is more complex. It suggests the participation of an efficient thermally-activated process that dominates defect interaction dynamics in the temperature range of $60-200$ °C.

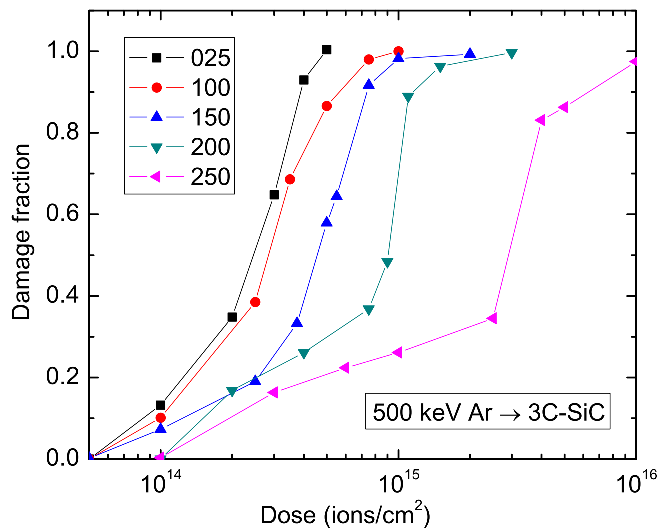

Figure 2: Dose dependences of relative disorder at the maximum of the bulk defect peak for 3C-SiC bombarded with 500 keV Ar ions at different temperatures (indicated in the legend in °C).

As a comparison, Fig.5 summarizes temperature dependences of τ and DAE in Si bombarded with 500 keV Ar ions. In contrast to the case of 3C-SiC (Fig. 4), Fig. 5 shows a monotonic increase in the DAE and a monotonic decrease in τ with increasing irradiation temperature for Si. These results demonstrate that the pulsed ion beam method provides a wealth of information about defect interaction dynamics.

We also studied defect interaction dynamics in Si crystals at room temperature bombarded with (pulsed beams of) 500 keV Ne, Ar, or Xe ions. A defect lifetime (τ) in the range of $3-8$ ms and a characteristic defect diffusion length of about $20-30$ nm are measured for Si irradiated at room temperature under different conditions. Both the τ and diffusion length of defects (L_d) were found to be essentially independent of the maximum instantaneous beam flux, total ion fluence, and dopant concentration within the ranges studied. However, both τ and L_d increase with increasing ion mass. This suggests that the type of radiation defects in Si depends on the average density of collision cascades. We have also found that τ depends on the level of pre-damage in the crystal lattice. We will investigate this intriguing effect in more detail next year.

Manuscripts detailing these findings are currently in preparation.

2.3 Opportunities for training and professional development

Training. One graduate student (Joseph Wallace) and one postdoc (Bimo Aji) are working full time on this project. The project has provided opportunities of one-on-one work (training) of the PI (Kucheyev) with both the graduate student and the postdoc.

Professional development. The PI (Kuchyev) has participated in two conferences, giving oral presentations based on results of this project. These two conferences were

- 2014 Spring MRS Meeting (Symposium EEE: Materials Behavior under Extreme Irradiation, Stress or Temperature, held in San Francisco, April 21 – 25, 2014);
- The 26th International Conference on Atomic Collisions in Solids Conference (ICACS-26), held in Debrecen, Hungary, July 13 – 18, 2014.

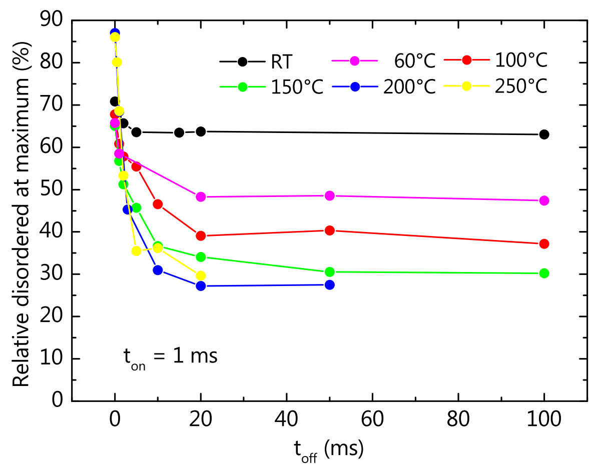

Figure 3: Dependence of relative disorder at the maximum of the bulk defect peak in 3C-SiC bombarded at different temperatures with a pulsed beam of 500 keV Ar ions on the duration of the passive part of the cycle, t_{off}, and all the other irradiation parameters fixed.

2.4 Dissemination of results

The major focus of the first year of this project was on obtaining results rather than dissemination of results. Until about October 2014, there was not much to disseminate except for the concept of the pulsed beam method and some preliminary data for single crystalline Si, demonstrating a tremendous promise of the pulsed beam method but also calling for more experimental data to validate the robustness of, hence, usefulness of the method. Nevertheless, results were disseminated at the two international conferences listed in the preceding paragraph.

2.5 Plan to accomplish the goals of the next reporting period

For the next year, we plan the following activities:

- Complete measurements of the temperature dependence of the defect interaction time constant (τ) in 3C-SiC and Si and document these results in journal articles.
- Measure the dependence of τ (for Si and SiC) on the density of collision cascades (determined by ion mass and energy), the instananeous defect generation rage, and the level and type of pre-existing disorder.

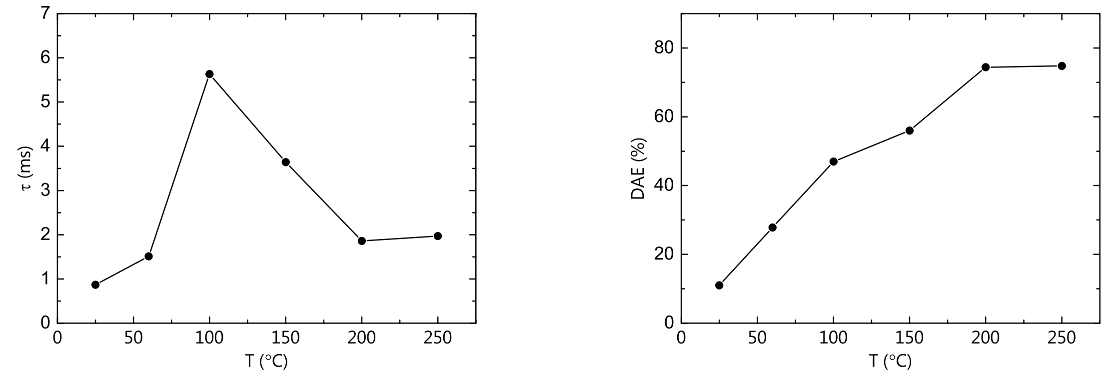

Figure 4: Temperature dependences of (left) the effective time constant of defect interaction processes, τ, and (right) the efficiency of dynamic annealing in 3C-SiC bombarded with 500 keV Ar ions.

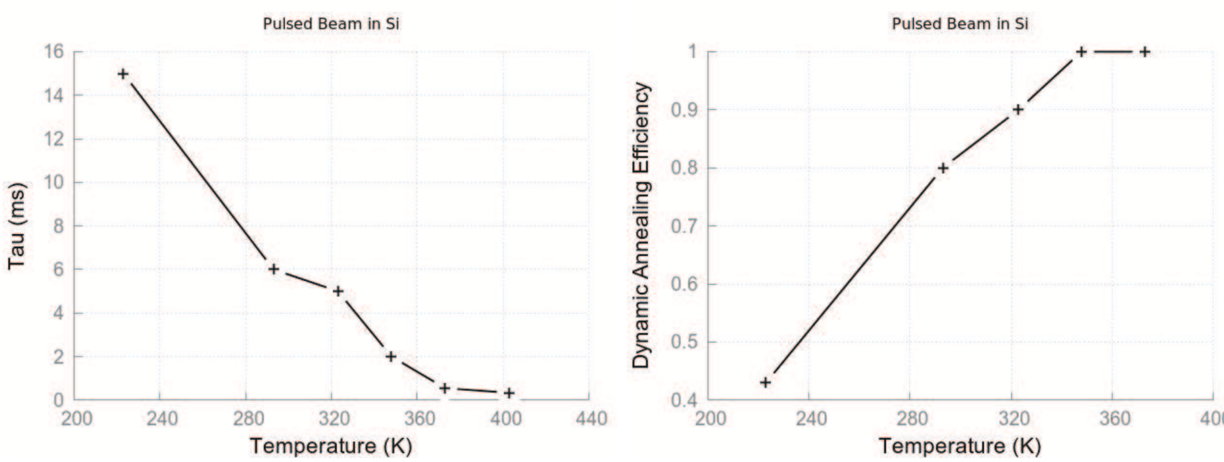

Figure 5: Temperature dependences of (left) the effective time constant of defect interaction processes, τ, and (right) the efficiency of dynamic annealing in Si bombarded with 500 keV Ar ions.

- Compare defect interaction dynamics in different major polymorphs of SiC (3C, 4H, and 6H).
- Explore kinetic-Monte Carlo and chemical rate equation approaches in order to describe defect interaction dynamics in Si and SiC. Our results (described in Sec. 2.2) have revealed the second order kinetics for both Si and 3C-SiC and strong (and non-trivial) temperature dependences of τ for both materials. We will attempt to develop quantitative models to describe our results.

3 Participants and other collaborating organizations

3.1 Individuals who have worked on the project

The following three individuals have worked on the project:

Sergei Kucheyev

- Name: Sergei Kucheyev
- Project Role: PI
- Nearest person month worked: 2
- Contribution to Project: Sergei has led the project and participated in all activities of this project.
- Funding Support: Funding only from this award.
- Collaborated with individual in foreign country: No
- Country(ies) of foreign collaborator: None
- Traveled to foreign country: Yes
- If traveled to foreign country(ies), duration of stay: 1 week (to attend the ICACS conference held in Hungary)

Joseph Wallace

- Name: Joseph Wallace
- Project Role: graduate student
- Nearest person month worked: 4
- Contribution to Project: Joseph has performed pulsed ion beam irradiation and ion beam analysis experiments
- Funding Support: Joseph's salary in 2014 was paid by the Livermore Graduate Scholar Program (LGSP)
- Collaborated with individual in foreign country: No
- Country(ies) of foreign collaborator: None
- Traveled to foreign country: No
- If traveled to foreign country(ies), duration of stay: None

Bimo Aji

- Name: Bimo Aji (full name: Leonardus Bimo Bayu Aji)
- Project Role: postdoc
- Nearest person month worked: 2
- Contribution to Project: Bimo has performed pulsed ion beam irradiation and ion beam analysis experiments
- Funding Support: Funding only from this award.
- Collaborated with individual in foreign country: No

- Country(ies) of foreign collaborator: None
- Traveled to foreign country: No
- If traveled to foreign country(ies), duration of stay: None

3.2 Other organizations that have been involved as partners

Texas A&M University (TAMU)

- Organization Name: Texas A&M University (TAMU)
- Location of Organization: TAMU, College Station, Texas
- Partners contribution to the project: Work of Joseph Wallace (a graduate student from TAMU) at LLNL and advice of Prof. Lin Shao (Joseph's graduate student adviser) from TAMU.
- Financial support: None
- In-kind support: Supervisory guidance of graduate student (Joseph Wallace) by Prof. Lin Shao from TAMU
- Facilities: None in 2014
- Collaborative research: None in 2014
- Personnel exchanges: Joseph Wallace (a student from TAMU) works full time at LLNL in Livermore while being financially supported by the Livermore Graduate Scholar Program (LGSP).
- More detail on partner and contribution (foreign or domestic): LLNL offers a highly competitive fellowship (LGSP) to employ graduate students who are willing to perform full time research at LLNL toward a degree from their degree-granting institution. The salary of Joseph Wallace was fully supported by the LGSP in 2014. Sergei Kucheyev is the LLNL mentor of Joseph Wallace, who worked in 2014 on this NEET pulsed beam project. Since Prof. Lin Shao from TAMU is the graduate adviser of Joseph Wallace, Prof. Shao is also involved in the project although with zero costs to the project.
- Have other collaborators or contacts been involved? No.

4 Impact

4.1 Impact on the development of the principal discipline of the project

This project offers an excellent opportunity to pioneer a new direction of advanced reactor materials characterization techniques and tools. It could establish the pulsed ion beam method as the primary approach to study defect interaction dynamics in nuclear materials. Understanding radiation defect dynamics is vital to the development of new materials for service in advanced nuclear reactors. It is critical if we want to extend our laboratory findings to nuclear material lifetimes and to time scales of geological storage of nuclear waste.

Experimental data on defect interaction dynamics is essential for building realistic and physically sound models to describe the formation of stable radiation defects. Indeed, the level of sophistication of a physical model of damage accumulation increases dramatically if this model is benchmarked against defect dynamics dataset measured in this project (instead of against damage buildup curves). The development of truly predictive tools requires both general understanding of radiation damage processes gained from experiments and specific experimental data to guide such theoretical efforts. This project is both developing a novel experiment approach to gain such understanding of dynamic defect processes and providing benchmark experimental data needed to link theory and modeling to experiments and critically assess radiation damage models.

Moreover, for several decades, many laboratories have been using ion beams to simulate neutron-induced radiation damage in nuclear materials. Such ion beam irradiation experiments inevitably use rastered ion beams, resulting in an effectively pulsed beam operation for any given location on the sample. The shape of the pulse and the pulsed-beam parameters of typical raster irradiation experiments are, however, difficult to control since they depend on the focused beam shape (determining the instantaneous dose rate) and specific rastering conditions. The fundamental understanding of defect dynamics revealed in this project could be used to guide future ion irradiation experiments with rastered ion beams for, at least, first order estimates of the importance of instantaneous and average dose rates.

4.2 Impact on other disciplines

The method developed in this project could be used to study implantation damage effects in semiconductors where ion irradiation is exensively used for doping the material and for dry etching. It could also be used to understand radiation effects in the field of astrophysics.

4.3 Impact on the development of human resources

This project has provided a research opportunity and challenge for Joseph Wallace, a graduate student for TAMU.

4.4 Impact on physical, institutional, and information resources that form infrastructure

This project has resulted in the development and implementation of a variable temperature holder for high-dose pulsed-ion-beam irradiation experiments with accurate dosimetry.

4.5 Impact on technology transfer

Nothing to report.

4.6 Impact on society beyond science and technology

Nothing to report.

4.7 Dollar amount of the awards budget spent in foreign country(ies)

The percentage of the award's budget spent in a foreign country is 1.6% (∼$1.6k out of $103k). It was spent during the conference trip to ICACS in Hungary (in July 2014), where the PI gave an invited talk describing results of this project. ICACS is one of the top international conferences in the field of ion-solid interactions.

5 Changes/Problems

5.1 Changes in approach and reasons for change

There were no changes in the objectives, scope, or approach during the reporting period.

5.2 Actual or anticipated problems or delays and actions or plans to resolve them

During Year 1, the project encountered two (unrelated) delays.

The first was a delay with hiring a postdoc to replace the originally identified postdoc (Sup Charnvanichborikarn, who decided to leave LLNL during the US federal government shutdown of 2013). The process of hiring isn't fast at LLNL. It was further delayed by the fact that the first best candidate, whose hiring package went all the way through the system, declined the LLNL offer and accepted a prestigious postdoc fellowship at NIST. So, we had to restart the hiring process again in May 2014. An excellent candidate was finally hired on Nov. 10, 2014.

The second delay was caused by a cascade of technical issues with the 4 MV ion accelerator on which the project depends. In fact, the group spent most of September and the first week of October 2014 chasing and fixing accelerator problems. These included issues with the Einzel lens, GVM unit, the charging system, and the $E \times B$ velocity selector. Before that, the 4 MV accelerator was operating almost trouble free for over 10 years. So, such a large load of problems was very unusual. All these problems were related. They were fixed, and pulsed-ion-beam irradiation and ion-beam analysis experiments were restarted in mid October.

5.3 Changes that have a significant impact on expenditures

The delay with re-hiring a postdoc, described in Sec. 5.2, has resulted in a significantly lower level of expenditures during Year 1. Expenditures were further reduced by the full time involvement of Joseph Wallace, who is a graduate student with the salary paid by the Livermore Graduate Scholar Program (LGSP). See Sec. 3.1 for additional details. The remaining two years of the project will have an increased effort, within the budget, to ensure that the project objectives are met.

5.4 Significant changes in use or care of human subjects, vertebrate animals, and/or Biohazards

Nothing to report.

5.5 Change of primary performance site location from that originally proposed

Nothing to report.

6 Budgetary information

As of the reporting period end date (December 30, 2014), the total costs of the project are $103k.