The Use of Neutron Irradiation Preconditioning Followed by Self-Ion Irradiation to Characterize the Irradiation Response of Nuclear Reactor Structural Materials—An Overview

Mychailo Toloczko, Principle Investigator
Scientist, Pacific Northwest National Laboratory (PNNL)

Jing Wang, Participant, Ph.D. Intern
Pacific Northwest National Laboratory and Texas A&M University (TAMU)

Frank Garner, Collaborator
Research Professor, Texas A&M University

Osman Anderoglu, Collaborator
Scientist, Los Alamos National Laboratory (LANL)

Andrew Minor, Collaborator
Professor, University of California at Berkeley (UCB)

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Presentation Outline

- Irradiation Needs for Fast Reactors and Light Water Reactors
- Introduction to Preconditioning
- Project Overview
- Background
  - Prior Preconditioning Studies
  - Shortcomings of Prior Preconditioning Studies
- Proposed Study
  - Materials and Methods
  - Project Timeline
  - Partners and Capabilities
- Summary
Interest in improved materials for nuclear reactor core internals is as strong as its ever been.

The meaning of "high dose" for fast and light water reactors has increased dramatically in recent years. Fast reactors seek 300+ dpa cores, and light water reactors want 100+ dpa internals.

The use of alternative irradiation techniques is being driven by:
- High dose (150+ dpa) neutron irradiations are perceived more and more as impractical due in part to the low availability of materials irradiation facilities.
- More rapid front line screening. A necessary first step in alloy development is now estimating neutron irradiation response.

Self-ion irradiations are currently perceived as the primary tool.
- Many advantages – quickly attain dose, non-activated specimens
- Some drawbacks – strongly different than neutron irradiation, mechanical properties is difficult
- Revived interest – recent studies presenting methods to better estimate neutron irradiation response, modern microscopy is simplifying specimen analysis
- Concerns continue to exist for the ability to predict high dose neutron irradiation response.
Introduction to Preconditioning

- **Definition of preconditioning** – Neutron irradiation (preconditioning) followed by charged particle irradiation.

- The concept is that preconditioning sets up a neutron irradiation microstructure that can be extended to higher dose with charged particles.

- This may produce a more neutron-prototypic high dose microstructure than pure charged particle irradiations.

- This project seeks to assess the value of neutron preconditioning as a tool for estimating the high dose neutron irradiation response of fast and light water reactor core structural materials.

- Since self-ions have key advantages of rapid irradiation and non-activation that are ideal for front line screening/characterization, preconditioning is not proposed to replace self-ions.

- Instead, it is proposed as a next-level irradiation response characterization tool that allows a second assessment of high dose response to further downselect for high dose neutron irradiations.
Proposed Usage

Initial Screening and Revision Using Heavy Ions

Phase 1
FR Clad/Duct - LWR Internals -

self-ions
300+ dpa
100+ dpa
(days)

alloy revision

Preconditioning of First Downselect

Phase 2
FR Clad/Duct - LWR Internals -

neutrons
~50 dpa
~20 dpa
(1-2 yrs)

self-ions
300+ dpa
100+ dpa
(days)

Further Downselect and Max Neutron Dose

Phase 3
FR Clad/Duct - LWR Internals -

neutrons
150+ dpa
100+ dpa
(5-10 yrs)
This project seeks to assess the value of neutron preconditioning as a tool for estimating the high dose neutron irradiation response of fast and light water reactor core structural materials.

Very limited number of prior studies performed 30-40 years ago where its value was considered.

PNNL has a large cache of neutron irradiated alloys up to doses of ~100-200 dpa from FFTF. LANL will also supply some high dose HT-9 specimens. Some of them are listed in below.

Will use optimized self-ion irradiation methods and modern microscopy techniques to assess preconditioning as a high dose characterization tool.

List of some high dose materials

- **HT-9** ~400°C, 100-180 dpa; ~550°C, 100-120 dpa (two heats)
- **Modified HT-9** (10Cr-1Mo alloy): 414°C, 135-140 dpa
- **T-91** 413°C, 175-185 dpa
- **MA957**: 412°C, 110 dpa; 550°C, 113 dpa
During 1970s and 1980s, a few neutron preconditioning experiments were carried out with heavy ion or electron irradiation in austenitic stainless steels. Primarily focused on void swelling responses. Provided limited microstructural information on chemical evolution, dislocation densities. Some neutron preconditioning experiments successfully reproduced similar swelling response as for pure neutron irradiations.

### Materials

<table>
<thead>
<tr>
<th>Materials</th>
<th>Neutron irradiation</th>
<th>Ion irradiation</th>
<th>Conclusion</th>
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</table>
| 304 SS, 316 SS | EBR-II ~6-72 dpa at 400-780 °C | 1 MeV electron to 9.8-19.4 dpa at 515 and 535 °C | Exploratory research  
• Observed different thickness of void denuded zone between preconditioned and pure electron irradiated thin foil specimens  
• In central region of thin foil, the neutron produced voids densities were preserved or increased as expected. |
| 316 SS | EBR-II ~43 dpa at 450 and 584°C | 4 MeV Ni ion to 60 dpa at 550-650 °C | Growth of void produced by neutron was suppressed by injected ions near damage peak. |
| 316 SS | EBR-II ~43 dpa at 450 and 584°C | 4 MeV Ni ion to 60 dpa at 550-650 °C | Dislocation densities and void size distribution were similar between neutron irradiated specimen and preconditioned specimen irradiated by Ni ion at higher temperature.  
• When preconditioned at higher temperature, precipitates dissolution were observed in ion irradiation. |
| PE16, 310, A286 | EBR-II ~20-30 dpa, 425-650°C | 1 MeV e- to 15 dpa at 500-700 °C | Swelling response of some specimens were insensitive to neutron preconditioning followed by electron irradiation.  
• Swelling response of solution-treated PE16 was altered by preconditioning due to promoting of $\gamma'$ precipitates. |

Example for Void Swelling

- Example of successful preconditioning study in sustenitic stainless steel [3].
- The following figures compare void size distributions in solution-annealed 316 stainless steel after pure neutron irradiation and after preconditioning. Shaded area in (a) and (b) represents size distribution after initial neutron irradiation to ~40 dpa.
- Pure neutron irradiation to ~60 dpa is shown in (a); and (b) shows further Ni ion irradiation on another specimen by 20 dpa at 650°C to 60 dpa total on a ~40 dpa.
- Note strong difference in ion irradiation temperature due to temperature shift.

Most of these studies focused on void swelling response, while limited microstructural information, such as chemical evolution and dislocation density were provided.

Charged particle dose levels were relatively small compared to neutron dose.

Void swelling in Ni-ion irradiated specimens was measured using a step height method, which is considered less accurate; the dose assignment in Ni-ion irradiation was calculated using EDEP-1, in which stopping power of Ni-ion was found to be overestimated in comparison to SRIM (causing undeestimation of ion range).

Only limited technical details were provided for charged particle irradiation procedures, which has been recently recognized as being capable of imposing huge influence over microstructural evolution.

HVEM experiments were relatively well controlled but were intrinsically limited by strong surface effects and lack of damage cascades, etc.
This project proposes to provide an in-depth assessment of the effectiveness of neutron preconditioning for evaluating high dose radiation response for relevant materials.

As illustrated, three sets of parallel irradiation experiments will be conducted.

Self-ion irradiations will utilize current best-practice methods.

The microstructural evolution will be examined by advanced microscopy characterizations, such as a combination of TEM and APT, and by nano-/micro-mechanical testing.
Prototypic Preconditioning Assessment Study

Pure neutron irradiation

Preconditioning

Precondition dose

~20-50 dpa

self-ions

Total dose

~100-200 dpa

Pure self-ions

match pure neutron total dose

Materials available for neutron preconditioning study

<table>
<thead>
<tr>
<th>Material/Heat</th>
<th>Neutron Irrad. Temperature (°C)</th>
<th>Low Neutron Dose (dpa)</th>
<th>High Neutron Dose (dpa)</th>
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<tr>
<td>HT-9/91353</td>
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<tr>
<td>HT-9/91353</td>
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<td>T-91/30176</td>
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<td>10Cr-1Mo/04479</td>
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<td>MA957/DBB0122</td>
<td>495</td>
<td>34</td>
<td>48</td>
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Alternative Preconditioning Assessment Study

Preconditioning

- Neutrons
- Preconditioning dose: 100-200 dpa
- Self-ions
- Total dose: 300+ dpa

Pure self-ion irradiation

- Self-ions
- Match total dose

Materials available for neutron preconditioning study

<table>
<thead>
<tr>
<th>Material/Heat</th>
<th>Neutron Irradiation Temperature (°C)</th>
<th>Neutron Dose (dpa)</th>
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### Timeline

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i. Ion irradiation of non-preconditioned materials: Temperature dependence and dpa dependence

ii. Ion irradiation neutron preconditioned materials: temperature dependence and dpa dependence

iii. Characterization: TEM, APT, mechanical testing

iv. Data interpretation, comparison, ongoing reporting

v. Preparation of final report
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<th>Partners</th>
<th>Role</th>
<th>Equipments</th>
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| • Mychailo Toloczko, Jing Wang, Dan Edwards, PNNL | • Overall lead for the project.  
• Expertise in neutron irradiation effects and microstructure  
• Substantial prior experience with heavy ion irradiations for fast reactor structural materials studies  
• Expertise in APT and TEM methods  
• Provide unirradiated and neutron irradiated materials  
• Sample preparation and microstructural characterization of irradiated specimens  
• Data comparison, interpretation, and linkage between ion and neutrons | • Cameca LEAP 4000X HR  
• JEOL ARM cold FEG (S) TEM aberration corrected TEM  
• FEI Quanta 3D SEM/FIB |
| • Frank Garner, Lin Shao, TAMU   | • Ion beam irradiations  
• Microstructural characterization of irradiated specimens  
• World expert in fast reactor history, neutron and ion irradiation effects, ion irradiation techniques, and microstructure of irradiated materials  
• Data comparison, interpretation, and linkage between ions and neutrons | • Ionex 1.7 MV Tandetron Accelerator  
• Tescan LYRA-3 SEM-FIB  
• FEI Tecnai G2 F20 ST FE-TEM |
| • Osman Anderoglu, LANL          | • Expertise in irradiation effects and microstructure of irradiated materials  
• Expertise in TEM methods  
• Supplying some neutron irradiated material | • FEI Tecnai TF30-FEG  
• FEI Helios SEM/FIB |
| • Andrew Minor, Peter Hosemann, UCB | • Expertise in nano- and micro-mechanical tests | • micromaterials nanoindenter  
• Hysitron exsitu nanoindenter |
Heavy ion irradiations will be performed at Texas A&M University following well-established procedures for ion irradiation.

This includes using a defocused, non-rastering beam; using 3.5 MeV Fe self-ions to reduce sputtering on the target surface and compositional change in the optimal examination region; maintaining vacuum level of $1 \times 10^8$ torr in target chamber; and calculate dose profiles in SRIM using Kinchin-Pease model; inclusion of temperature-shift effects.

The facility has been licensed to handle and ion irradiate neutron-activated materials and can deliver dose to $\sim 100$ dpa/day using Fe self-ions.

1.7 MeV Tandetron accelerator at Texas A&M University
Transmission electron microscopy (TEM) and atom probe tomography (APT) will be routinely used to characterize microstructure and microchemistry.

TEM examination will provide structural information, such as dislocation structures, second phases evolution, void swelling and grain sizes, etc.

APT is good at produce 3-D reconstruction of chemical distribution with nanometer level spatial resolution and excellent mass resolution in a small needle size specimen.

Nano-/micro-mechanical testing, such as nano-indentation and micropillar compression, will be performed to explore the possible mechanical property-microstructure relationship after irradiation experiments.
The PNNL group has extensive experience in characterization of irradiated microstructures. Below is an example from previous preliminary studies for comparison between ion and neutron irradiated materials.

The APT 3-D chemical reconstruction is very effective in detecting nanometer scale features in both specimens.
Summary

- Intent is to assess the value of preconditioning as a Phase 2 irradiation response characterization tool of FR and LWR core structural materials that would take place after initial screening using pure self-ion irradiations.

- Potential value is that it may provide a more accurate representation of high dose neutron irradiation response for microstructural evolution.

- The history and extent of prior research was briefly discussed.

- Will utilize up-to-date ion irradiation techniques, and modern microscopy methods.

- PNNL and partner knowledge-base, neutron-irradiated materials library, available accelerators, and materials characterization capability are an excellent foundation to perform this work.