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

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# Presentation Outline

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- *Proposed Study*
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# Nuclear Reactor Core Structural Materials Research

- *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 <u>estimating</u> 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

- *<u>Definition of preconditioning</u> – 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, <u>preconditioning is not proposed to replace self-ions.</u>*
- *Instead, it is proposed as a next-level irradiation response characterization tool that allows a second asssessment 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 - → self-ions → 300+ dpa (days)
LWR Internals - → self-ions → 100+ dpa (days)

alloy revision

***<u>Preconditioning</u> of First Downselect***

**Phase 2**

FR Clad/Duct - → neutrons (1-2 yrs) → ~50 dpa → self-ions (days) → 300+ dpa
LWR Internals - → neutrons (1-2 yrs) → ~20 dpa → self-ions (days) → 100+ dpa

**Further Downselect and Max Neutron Dose**

**Phase 3**

FR Clad/Duct - → neutrons (5-10 yrs) → 150+ dpa
LWR Internals - → neutrons (5-10 yrs) → 100+ dpa

# Project Overview

- *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

# Prior Preconditioning Studies

- *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.*

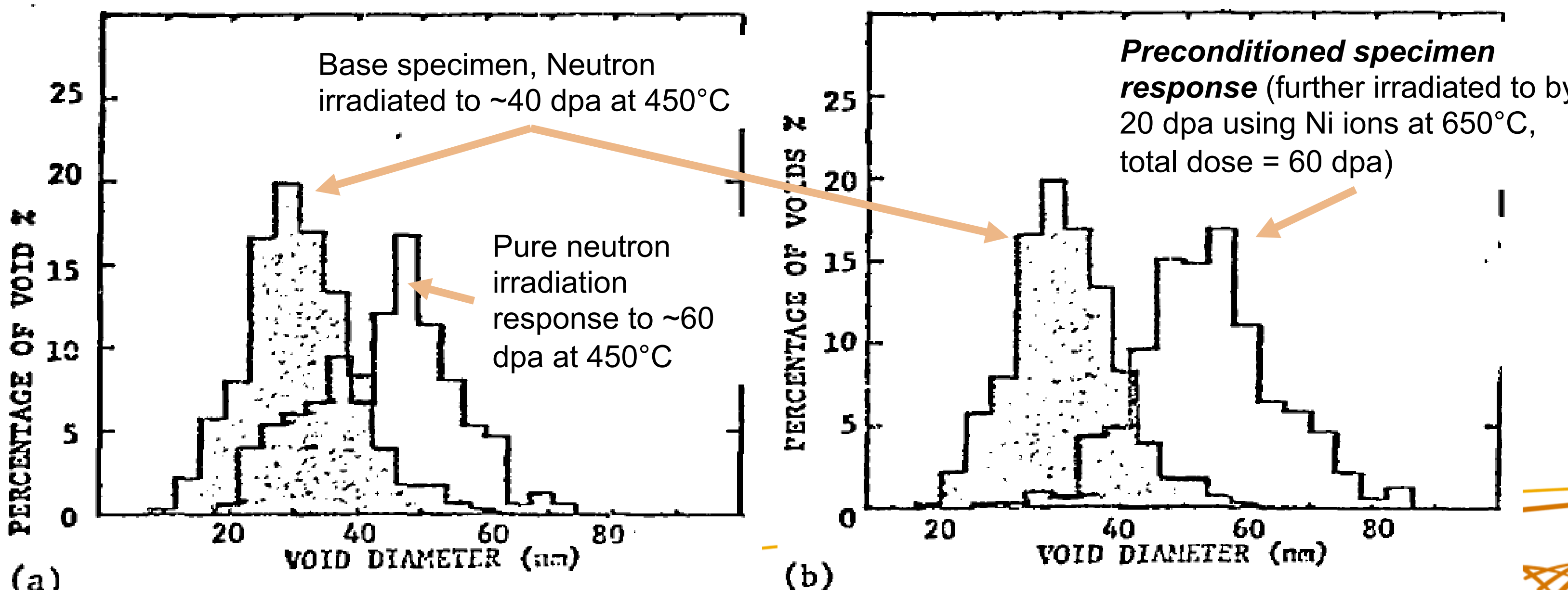
Some results from historic neutron preconditioning studies

| Materials | Neutron irradiation | Ion irradiation | Conclusion | Ref |
|---|---|---|---|---|
| 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<br>• Observed different thickness of void denuded zone between preconditioned and pure electron irradiated thin foil specimens<br>• In central region of thin foil, the neutron produced voids densities were preserved or increased as expected. | [1] |
| 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. | [2] |
| 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.<br>• When preconditioned at higher temperature, precipitates dissolution were observed in ion irradiation. | [3] |
| 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.<br>• Swelling response of solution-treated PE16 was altered by preconditioning due to promoting of γ' precipitates. | [4] |

[1] Garner, et al., ASTM STP 570, 1975, 433-448
[2] Lee, et al., J. Nucl. Mater., 85&86 (1979) 577-581
[3] Rowcliffe et al., US/USSR Breeder Reactor Info. Exch., 1979
[4] Gelles, et al., J. Nucl. Mater., 108&109 (1982) 504-514

# 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.*





[3] Rowcliffe et al., US/USSR Breeder Reactor Information Exchange, 1979

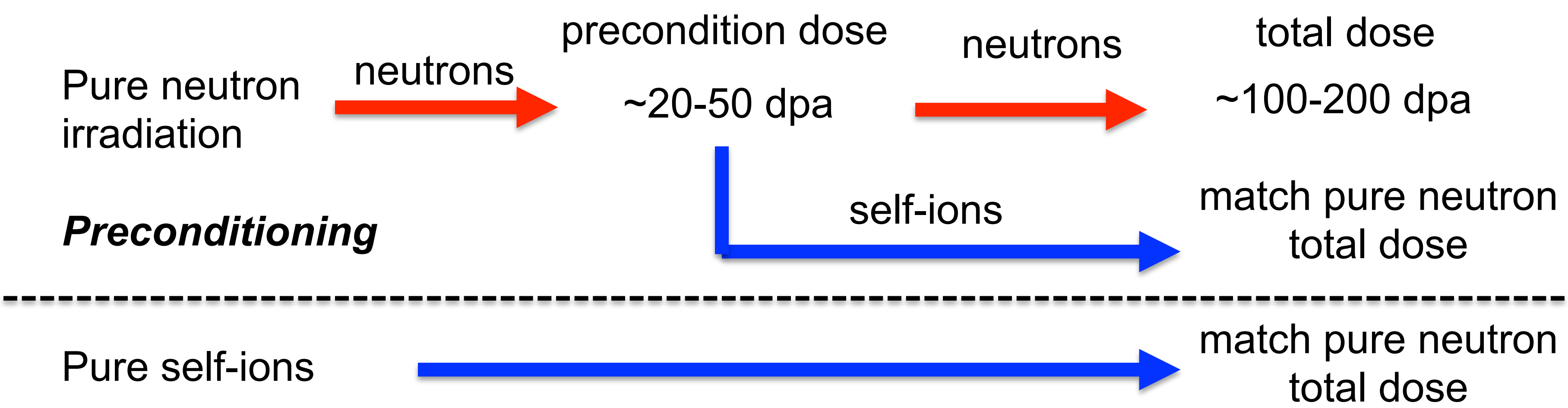
# Shortcomings of Prior Preconditioning Studies

- *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.*

# Proposed Study

- *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




Materials available for neutron preconditioning study

| Material/Heat | Neutron Irrad. Temperature (°C) | Low Neutron Dose (dpa) | High Neutron Dose (dpa) |
|---|---|---|---|
| HT-9/91353 | 370 | 7 | 44 |
| HT-9/91354 | 412 | 37 | 112 |
| HT-9/91353 | 490 | 19 | 77 |
| HT-9/92235 | 495 | 38 | 65 |
| HT-9/91353 | 550 | 20 | 100 |
| HT-9/84425 | 550 | 20 | 100 |
| T-91/30176 | 414 | 73 | 150 |
| 10Cr-1Mo/04479 | 605 | 37 | 120 |
| MA957/DBB0122 | 495 | 34 | 48 |

# 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

| Material/Heat | Neutron Irradiation Temperature (°C) | Neutron Dose (dpa) |
|---|---|---|
| **HT-9/84425/ACO-3** | 443 | 155 |
| **HT-9/91353** | 392 | 164 |
| **HT-9/91354** | 412 | 112 |
| **HT-9/91354** | 546 | 109 |
| **T-91/30176** | 413 | 180 |
| **MA957/DBB0111** | 412 | 112 |
| **MA957/DBB0111** | 546 | 113 |
| **Mod. HT-9/XA3607** | 414 | 139 |
| **316 SS** | 412 | 175 |

# Timeline

| Project Tasks | Q1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| i | | | | | | | | | | | | |
| ii | | | | | | | | | | | | |
| iii | | | | | | | | | | | | |
| iv | | | | | | | | | | | | |
| v | | | | | | | | | | | | |

*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, mechanial testing*
*iv. Data interpretation, comparison, ongoing reporting*
*v. Preparation of final report*

# Partners

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

# Capability - Ion Irradiation

- *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 ~100 dpa/day using Fe self-ions.*

1.7 MeV Tandetron accelerator at Texas A&M University

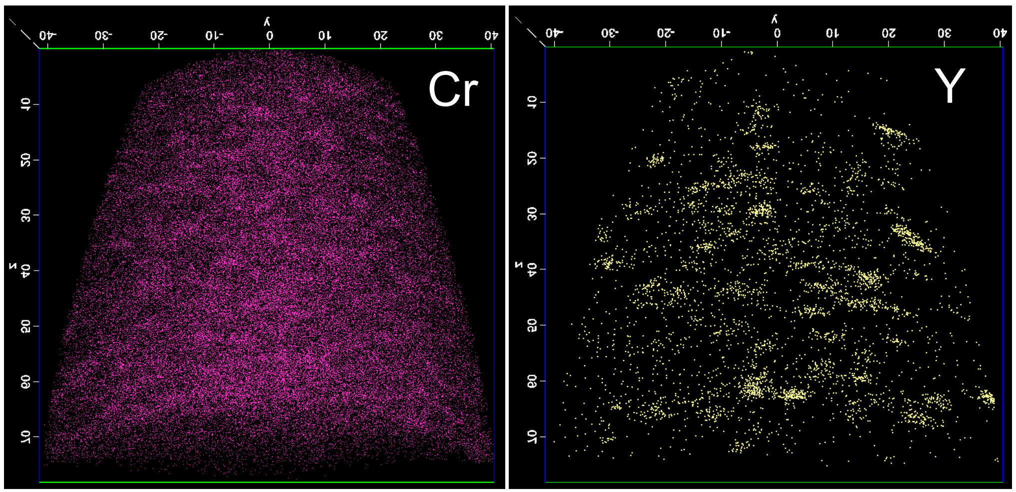
Pacific Northwest
NATIONAL LABORATORY

# Capability - Advanced Characterization

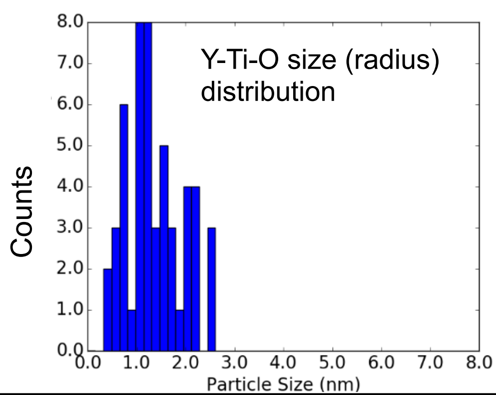
- *Transmission electron microscopy (TEM) and atom probe tomography (APT) will be routinely used to characterize microstructure and microchemistry.*
- *TEM examination will provide structural information, such as dislocation structures, second phases evolution, void swelling and grain sizes, etc.*
- *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.*

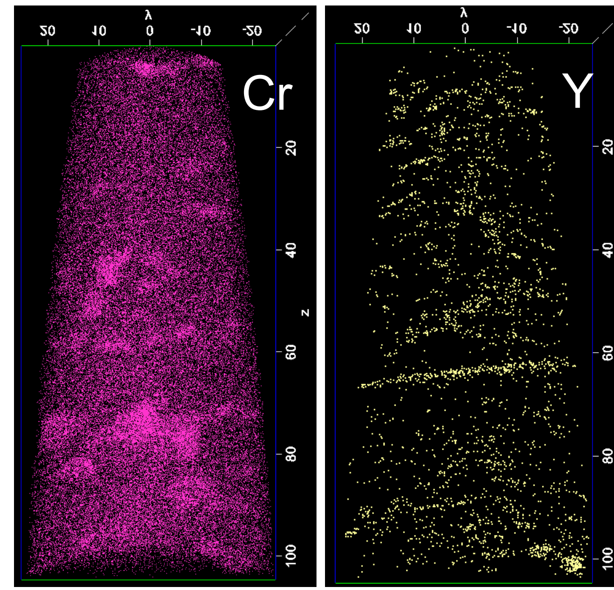
# Capability-Advanced Characterization

- *The PNNL group has extensive experience in characterization 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.*

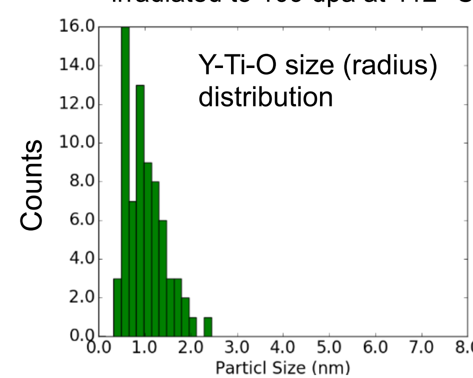



Ion map of a MA957, Cr ion irradiated to 100 dpa at 400 °C




Ion map of a MA957, neutron irradiated to 109 dpa at 412 °C



# 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.*