INEST

Institute for Nuclear Energy Science and Technology

Fuels & Materials

Todd Allen
CORE Director

KL Murty
CORE University Lead

September 27-28, 2011

NATIONAL CHALLENGE

Degradation of materials due to harsh reactor environments limits the operation of nuclear reactors

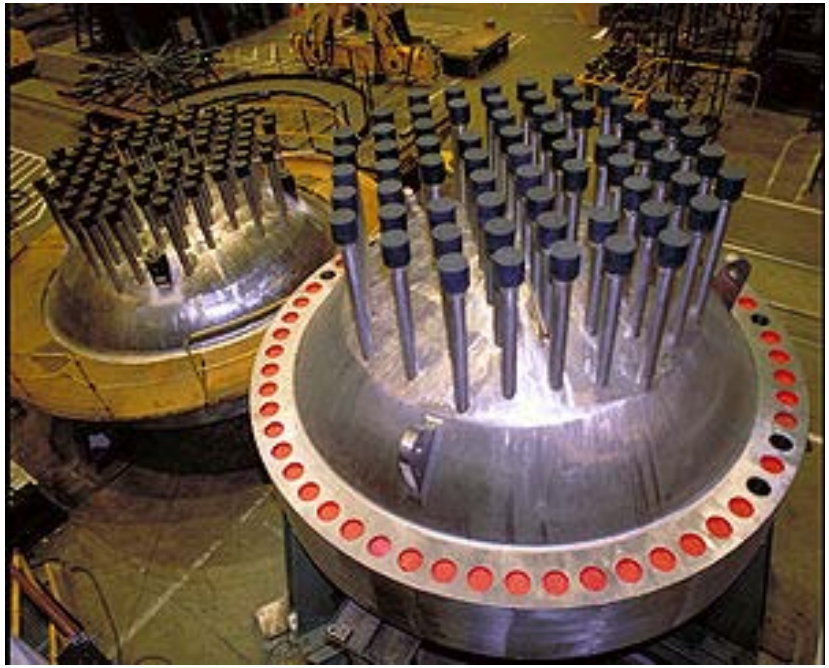
CORE MISSION

Grow technological leadership and reputation while building long-term partnerships, capabilities, tools, and techniques to advance nuclear fuels and materials. Three year plan focus:

- Apply advanced computational and experimental techniques to better understand degradation modes in LWR fuels and materials
- Research that bridges basic science and applied technology
- Develop education programs that focus on LWR technology

TECHNICAL FOCUS

- Stress corrosion cracking in LWR environments
- Mitigation of degradation mechanisms in Zr-alloy fuel cladding
- Property changes in uranium dioxide-based fuels including thermal transport and associated microstructural changes



CORE VISION

- Education programs established to improve knowledge and understanding of LWR fuel and materials research and stimulate collaborations between academia and INL researchers
- INL and the university collaborators will be recognized for their expertise in nuclear fuels and materials

CURRENT STATE

- Understanding of fundamental mechanisms degradation phenomena related to materials performance unsatisfactory
- Advancements needed to thoroughly understand issues related to life extension and safe operations
- Need to revitalize researchers with expertise in fuels and materials
- Challenges exist in the selection of materials for advanced next generation nuclear reactors

OPPORTUNITIES

Near-Term

- Materials & fuel technology needed to mitigate degradation factors in reactor core, piping materials & cladding
- Prepare research teams from INL and academia to apply for research grants

Renewed and growing federal interest in developing and funding new capability

Long-Term

- National
  - high-risk, multiple discipline research
  - pushing scientific boundaries
  - workforce education and development
  - technology leadership
- Industry
  - centers of excellence
  - knowledge repository
  - Combined intellect of lab and university
  - Cross-disciplinary, cross-institution research teams

COMPETITORS

- France, Japan, Korea and China researching LWR degradation and funded by various governmental agencies
- ORNL, ANL, PNNL and BNL pursuing research on LWRS and will target federal funding
  - PNNL also has significant LDRD aimed at LWR R&D
  - PNNL and ANL have ongoing programs sponsored by NRC

CAPABILITIES

INL

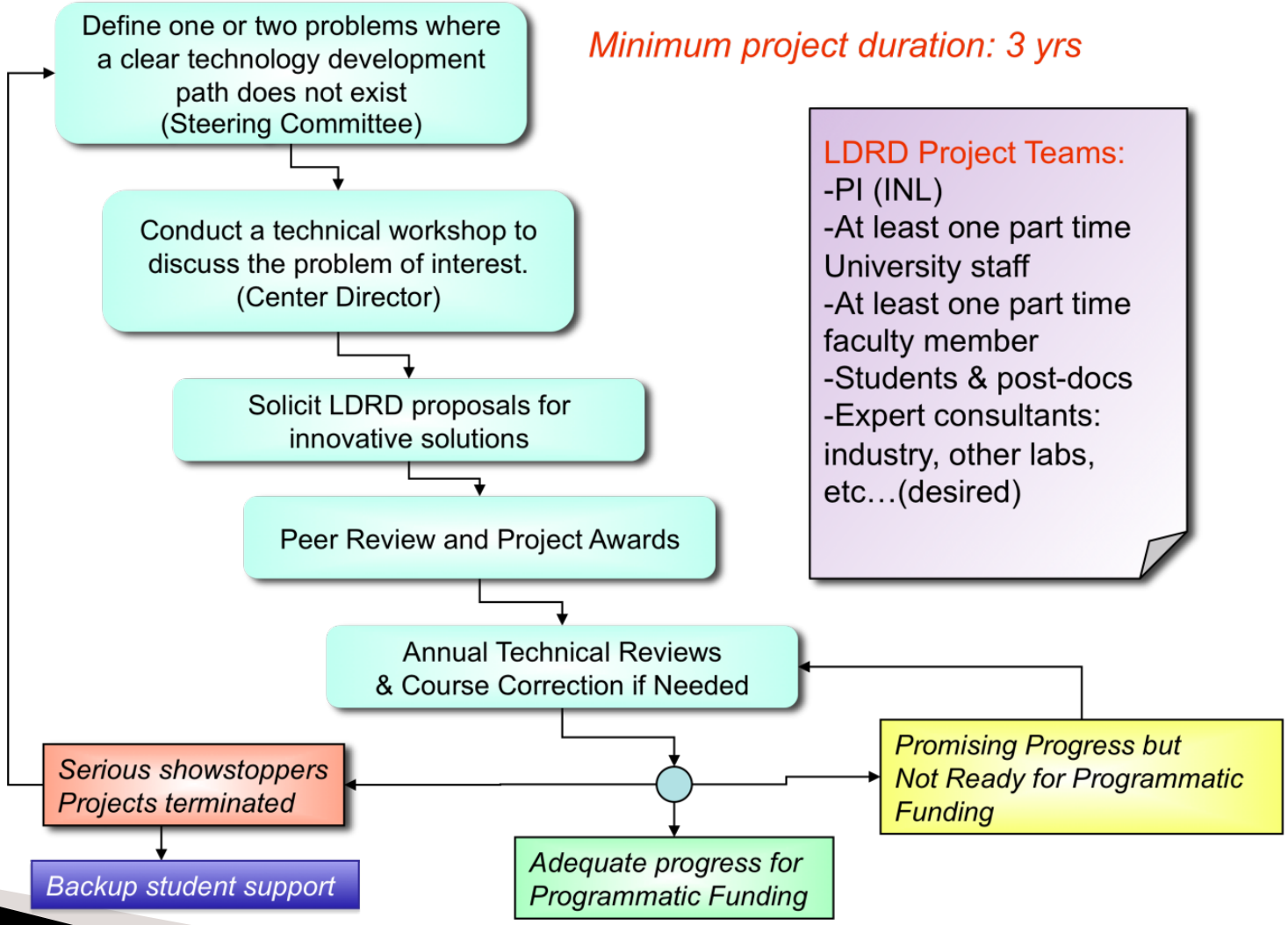
- ATR
- Hot cell facility
- PIE
- Imaging suite at CAES
- World class electron microscopes and analytical instrumentation
- Extensive personnel expertise in testing irradiated materials

NUC

- Research reactor at MIT
- Burst and Biaxial creep characterization at NCSU

COLLABORATIONS

Partnerships key to ensuring INL staff remain connected



CURRENT PROGRAM ELEMENTS

- **Fuels & Materials CORE white paper**
- **Workshop Series**
  - — June 2010 Stress Corrosion Cracking Science
  - — April 2011 Zirconium Alloy Science
  - — April 2012 Uranium Fuel Science
- **Outreach Events**
  - — Jan 2011 Regional Workshop (NCSU Raleigh)
- **LDRD**
  - — 2010 Stress Corrosion Cracking
  - — 2011 Zirconium Alloy Science
  - — 2012 Uranium Fuel Science

FY 2011 CORE FUNDING

	Allocation	How Used
CORE Management	$125,000	Management, administrative support, supplies, etc.
LDRD	$1,315,100	Five LDRD projects in SCC, UO_2, Zr Alloy science, fundamental modeling
Program Development	$36,000	Not used this year
University Contract	$71,200	NCSU Support for CORE
Workshop	$30,000	Zirconium Workshop held in May 2011

F&M CORE LDRD PARTICIPATION

Year	No. Proposals Submitted to F&M CORE	Number of Proposals Developed by INL/University Teams Formed at Workshop	Number of F&M CORE Proposals Awarded	Number of F&M CORE Proposals Awarded to INL/University Teams Formed at Workshop
2010	4	4	1	1
2011	8	4	2	1
2012	12	10	2	2

FY 2011 ACCOMPLISHMENTS

- **Workshops**
  - Regional Workshop: NCSU Raleigh Feb 2011
  - Zirconium Alloy Science INL May 2011
- **Research**
  - Irradiation-Induced Evolution of Defect and Microstructure in Nanocrystalline BCC Mo
  - Development of Positron Techniques to Understand Microstructural Evolution of U and UO_2 Under Irradiation
  - In situ characterization of an oxide film for prediction of stress corrosion cracking susceptibility
  - Characterization of the Nanomechanical Response of Material to Determine Key Variables in Stress Corrosion Cracking
  - In-situ micro-Raman spectroscopy and modeling of breakaway oxidation of zircaloy cladding

WORKSHOP QUOTE

May Workshop:

ANT International – Zirconium Alloy Science

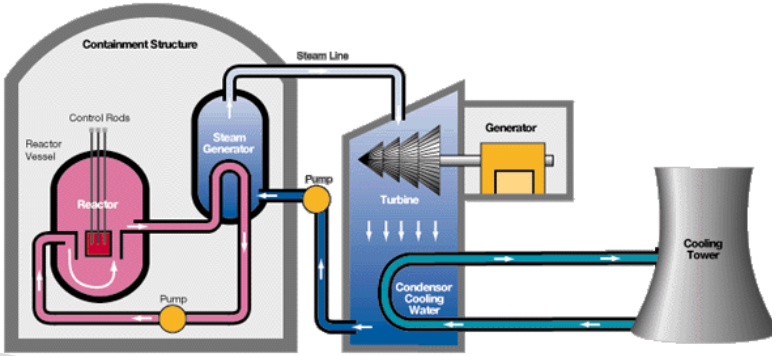
Attendees:

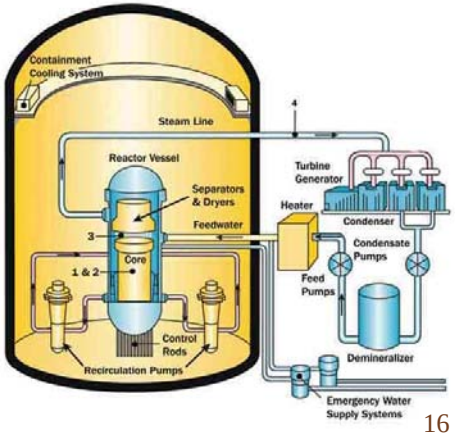
- INL: 23
- University:12
- Other Labs: 3

"A rare and exceptional workshop. These two days brought together true deep expertise, representing over 100 years of industry experience, and an enthusiastic group of young scientists eager to bring state of the art tools to bear on cutting edge problems. Such a workshop is enormously more efficient than simply letting university researchers try to ferret out the real issues and problems on their own. These two days may have saved months of work on the part of the participants. Workshops of this type and caliber should be the standard for how new research areas are launched and promoted."

INEST-Event Regional (East Coast) Meeting -F&M CORE in Feb 2011

- Held February 21, 2011 at North Carolina State University in Raleigh, NC
- The morning session was on the description of the COREs followed by a tour of the PULSTAR reactor in the nuclear engineering department at North Carolina State
- The afternoon session was devoted to presentations on nuclear fuels and materials for LWRs from experts in the field describing the current status of understanding and future challenging issues





INEST-Event Regional (East Coast) Meeting / Fuels & Materials CORE
North Carolina State University, Raleigh, NC
Monday, Feb 21st, 2011
Progress Energy Lecture Hall, 1202 Burlington Engineering Labs

AGENDA - morning

8:00am	Continental Breakfast	
8:30am	Welcome	NCSU Officials
8:45am	INEST Overview	Dave Hill/INL (Marsha substituted)*
9:00am	Fuel Cycle CORE	Patricia Paviet-Hartmann/INL
9:20am	Safety & Licensing CORE	Ray Cao/OhioState
9:40am	Fuels & Materials CORE	Todd Allen/INL (KL substituted)*
10:00am	Break	
10:15am	Overview of CASL	Paul Turinsky/NCSU
10:35am	Overview of CASL/MPO	Don Brenner/Jacob Eapen/NCSU
10:55am	Overview of NEUP	John Gilligan/NCSU Marsha Lambregt/INL
11:15am	Lunch (NSUF)	Todd Allen/INL (Marsha & KL substituted)*
	How to propose to the Fuels & Materials CORE	Greg Bala/Marsha Lambregts/INL
12:30pm	Facility Tour (PULSTAR, CASL, etc)	

*flights cancelled due to weather

INEST-Event Regional (East Coast) Meeting / Fuels & Materials CORE
North Carolina State University, Raleigh, NC
Monday, Feb 21st, 201

Afternoon session in Duke Energy Conference Room, 1121 Burlington

1:30pm	Stress Corrosion Cracking Overview	Peter Ford/Consultant Robin Dyle, EPRI
3:00pm	Break	
3:15pm	Fuel Overview	Chuck Patterson/Consultant
4:00pm	Modern Materials Science Applications to Zr	Richard Holt/Queens University

Attendance: 48 registered (~42 in attendance) - 6 from INL

Massachusetts Institute of Technology (2)
Pennsylvania State University (1)
University of Cincinnati (1)
University of South Carolina (1)
Global Nuclear Fuels (3)
F&M Advisory group (3+1)

Rensselear Polytechnic Institute (1)
University of Maryland (3)
Ohio State University (1)
University of Tennessee (2)
EPRI (1) – presenter
Remaining from NCSU

Characterization of the Nanomechanical Response of Material to Determine Key Variables in Stress Corrosion Cracking

LDRD 11-043 – (Start date: October, 2010)

- Bill Gerberich and K. Andre Mkhoyan
  University of Minnesota

- John Jackson
  Idaho National Laboratory

GOALS AND OBJECTIVES

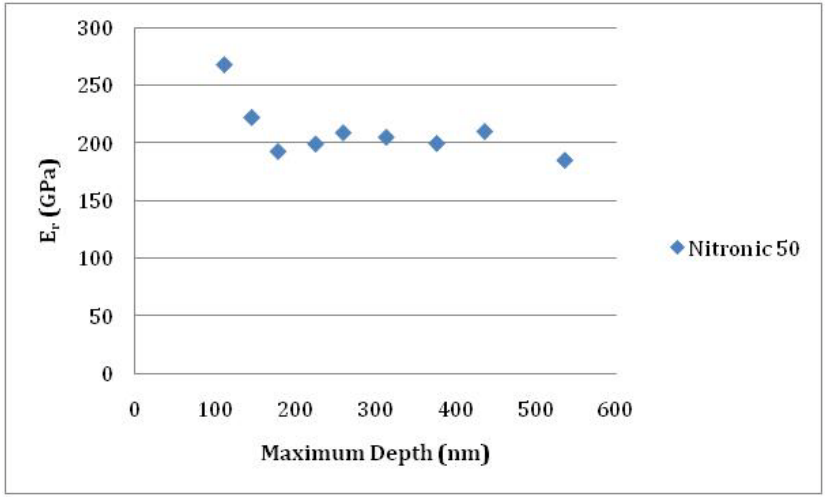
- **Proof of principle studies to determine the effect of relevant variables on crack initiation**
  - Screen variable using well controlled nano scale experiments; chemo-mechanical interactions
- **Nanopillar deformation as a function of parameters that contribute to, or govern SCC**
  - Effect of surface oxidation, alloy impurity content, surface dislocations, or hydrogen on deformation behavior and crack initiation

TECHNICAL ACCOMPLISHMENTS TO DATE

- **Acquired and implemented two MEMS transducers for use in a PI 95 TEM Picoindenter**
  - Required for force-displacement measurement
- **Refined metallography and etching techniques for preparation of electron transparent films**
  - Examination of impurity interactions at grain boundaries using TEM
- **Refined a Focused Ion Beam (FIB) milling technique for manufacture of micro-pillars**
  - Initially silicon, then 316 SS, finally CW Nitronic 50.
  - Aspect ratio and geometry are important for compression via pico-indenter.

TECHNICAL ACCOMPLISHMENTS TO DATE

Performed initial nano-hardness and modulus measurements on Nitronic 50 sample



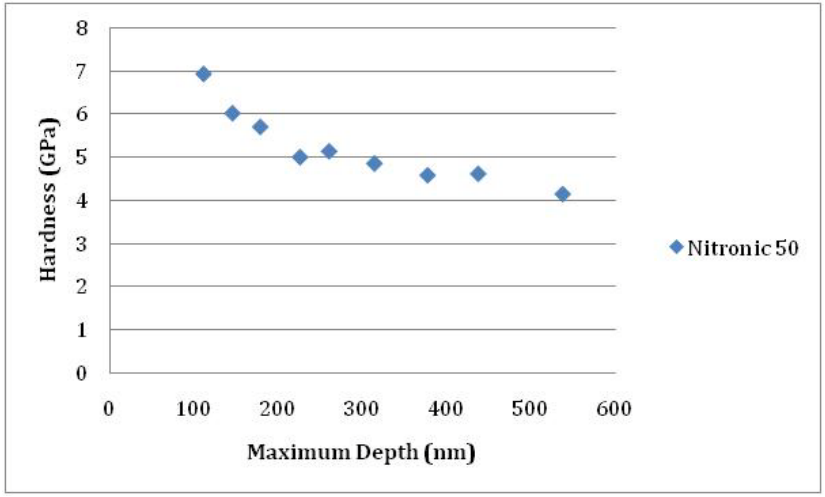


Figure 1: Elastic moduli and nanohardness from indentation experiments using a sharp Berkovich tip. Moduli approximately constant after initial readings but hardness drops due to the indentation size effect.

PUBLICATIONS AND PRESENTATIONS

- Project is in its initial stages. None to date.

FY 2012 PLANNED ACTIVITIES

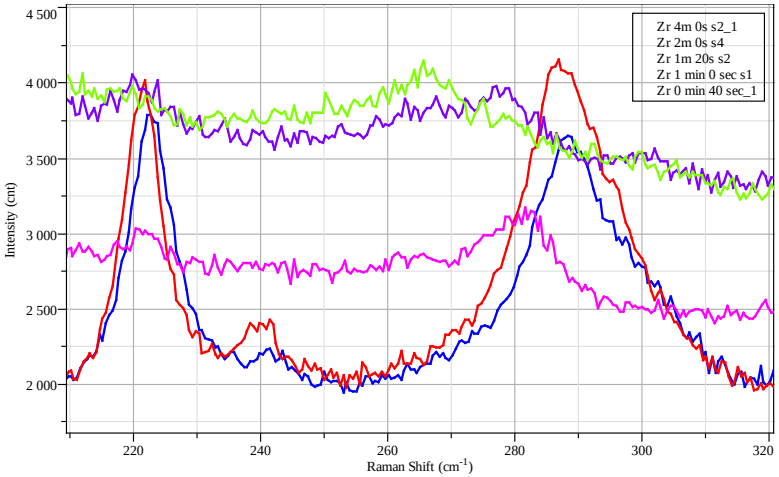
- Initial microstructural characterization of CW Nitronic 50 via indentation TEM.
- Successful in-situ TEM studies of Nitronic 50.
- Initial hydrogen charging of 316 SS and Nitronic 50 to study detailed interactions with grain boundaries and dislocations as well as the size effect on mechanical properties of material.

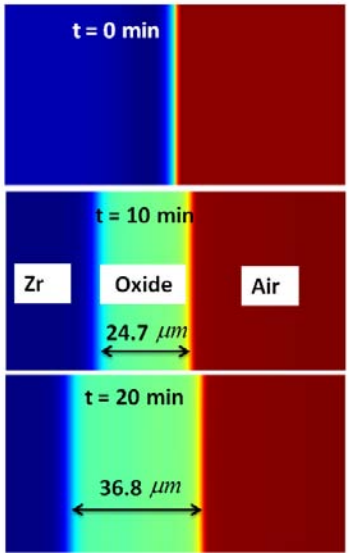
IN-SITU MICRO-RAMAN SPECTROSCOPY AND MODELING OF BREAKAWAY OXIDATION OF ZIRCALOY CLADDING

- Start: October 2010
- Participants: Zhandos Utegulov (INL), Haitham El Kadiri (Mississippi SU), Marat Khafizov (INL)
- The Goal of the project is to reliably predict the breakaway oxidation of Zry-4 using Raman Spectroscopy validated multiscale model

TECHNICAL ACCOMPLISHMENTS

- An experimental methodology was developed to observe monoclinic and tetragonal phases through micro-Raman and Atomic Force Microscopies
- Two Phase Field models were developed and numerically investigated.
  - The first models morphological evolution and coarsening of coherent precipitates that account for inhomogeneous elasticity.
  - Second simulates oxidation of a pure metal with effect of growth stresses.





PUBLICATIONS

- “A Phase-field model for selective oxidation of metals and alloys:Application to Zirconium for Nuclear”, M. Asle Zaeem, H. El Kadiri, M.F. Horstemeyer, M. Khafizov, and Z. Utegulov, accepted to Current Applied Physics
- “Morphological evolution and coarsening of coherent precipitates: effects of inhomogeneous elasticity and intermediate phase growth”, M. Asle Zaeem, H. El Kadiri, M. Khafizov, Z. Utegulov, and M.F. Horstemeyer, submitted to Acta Materialia

PLANS FOR 2012

- Perform in-situ hydrostatic pressure-dependent micro-Raman spectroscopy on tetragonal ZrO_2 polycrystalline powders to measure transformation stresses associated with the phase transition of tetragonal zirconia to monoclinic one at fixed temperatures.
- Formulate and investigate a stress-dependent phase field (PF) model for tetragonal-monoclinic transformation in thin film Zirconia. The stress dependency includes the effect of stress on transformation as well as the generation of stress due to the volume expansion by the transformation. The modeling will be done with the goal to validate the Raman stress mapping results.

Development of Positron Techniques to Understand Microstructural Evolution of U and UO_2 Under Irradiation

INL PI: Maria Okuniewski
University PIs: Prof. Kelvin Lynn and Marc Weber (Washington State University)
INL Collaborators: Doug Akers, Craig Shull, Mark Drigert, Lyle Roybal, Randy Fielding, Jason Harp, and Manny Perez

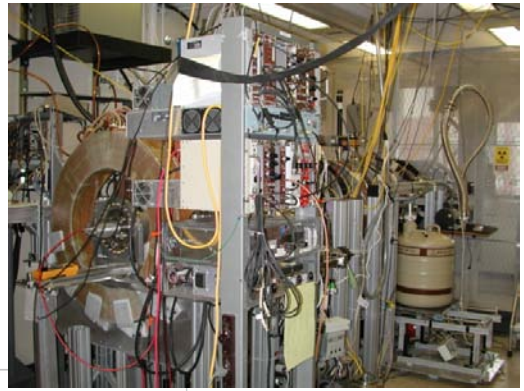
Project Initiated: Oct. 2009

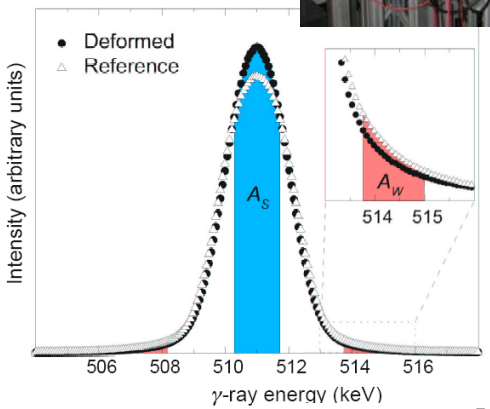
Project Goals:

(1) Understand the microstructural evolution in irradiated U alloys with positron annihilation spectroscopy (PAS) and transmission electron microscopy. This information will provide unique data, lead to a better understanding of the impact of microstructure on the thermal and mechanical properties of fuels, and provide crucial input and validation data for multi-scale, multi-physics models.

(2) Develop a PAS system to analyze irradiated materials.

Sample Types: U alloys that have been both ion and neutron irradiated.



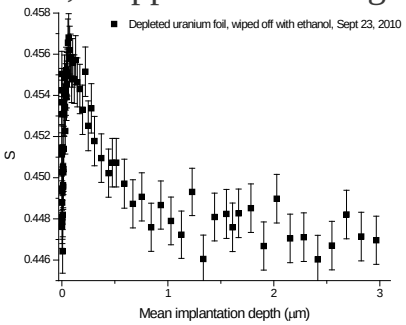


$$S = \frac{A_s}{A_0}, \quad A_s = \int_{E_0 - E_s}^{E_0 + E_s} N_D \, dE \qquad W = \frac{A_w}{A_0}, \quad A_W = \int_{E_1}^{E_2} N_D \, dE$$

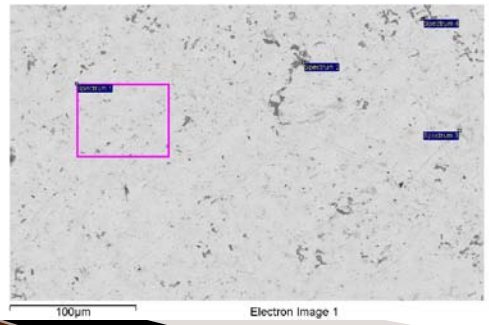
ACCOMPLISHMENTS

- Specimens
  - Fabrication of five DU-based alloys (DU, DU-10Mo, DU-20Mo, DU-10Zr, DU-15Zr – polycrystalline)
    - *Heat treatments, chemistry, and microscopy completed*
- Specimen irradiations
  - Upgrades of WSU accelerator completed (for ion irradiations)
  - Neutron irradiations for DU-10Zr pending
    - *Capsules are currently ready for insertion at reactor (waiting reactor approvals)*
- PAS Systems
  - Designed, fabricated, and tested PAS system to analyze irradiated materials (which are radioactive)
    - *Includes hardware, software, and special collimator*
  - Design of a re-configurable PAS system (Doppler broadening, lifetime, and angular correlation techniques) for analyzing low activity samples. System will have user friendly interface for multiple users (graduate students and scientists)
- PAS Studies
  - Initiated PAS studies of defects in pure DU

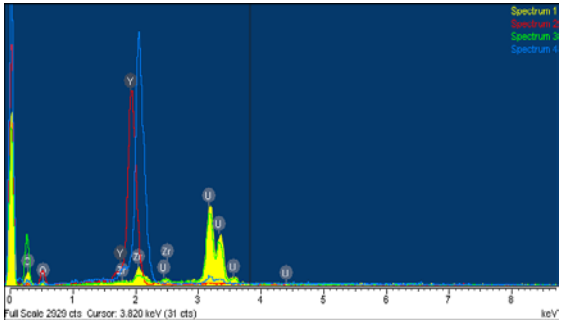
DU, Doppler broadening



DU-10Zr, as cast



DU-10Zr, as cast



PUBLICATIONS AND PRESENTATIONS

- M. A. Okuniewski, M. H. Weber, K. G. Lynn, B. W. Beeler, B. J. Good, C. S. Deo, M. I. Baskes, S. A. Maloy, and J. F. Stubbins, *Positron Annihilation Spectroscopy in Nuclear Structural Materials and Fuels*, The Twelfth International Workshop on Slow Positron Beam Techniques (Australia, August 2010).
- M.A. Okuniewski, Microstructural Evolution in Nuclear Fuels: Connecting Experiments and Simulations, Idaho National Laboratory for Secretary of Energy, Dr. Steven Chu (Idaho Falls, ID, September 2010).
- Related publications (ongoing research that will benefit from PAS studies):
  - B. Beeler, B. Good, S. Rashkeev, C. Deo, M. Baskes, and M. Okuniewski, First Principles Calculations for Defects in U, *Journal of Physics Condensed Matter*, vol. 22, n. 50 (2010), 505703.
  - B. Beeler, B. Good, S. Rashkeev, C. Deo, M. Baskes, and M. Okuniewski, First Principles Calculations of the Stability and Incorporation of He, Xe, and Kr in U, *Journal of Nuclear Materials*, currently in press.
  - R. R. Mohanty, J. Bush, M. A. Okuniewski, Y. H. Sohn, Thermotransport in γ(bcc) U-Zr alloys: A phase-field model study, *Journal of Nuclear Materials*, vol. 414 (2011), p. 211.

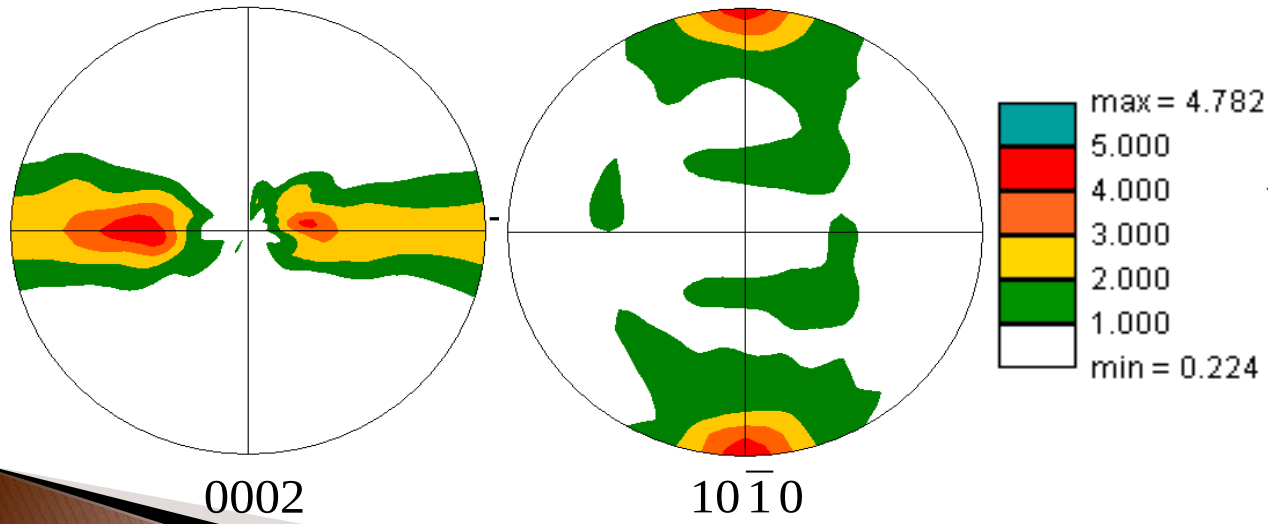
OBJECTIVES FOR 2012

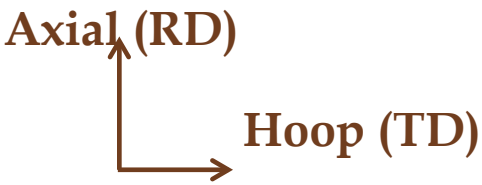
- Specimen irradiations
  - Complete ion and neutron irradiations at respective facilities
- PAS Systems
  - Continue use of PAS system to analyze irradiated materials (which are radioactive) .
  - Complete the assembly and testing of the re-configurable PAS system. System will be in constant operational mode.
- PAS Studies
  - Complete PAS studies of defects in U alloys
- Modeling
  - Relate, validate, and/or verify results obtained to ongoing modeling efforts supported via other sources
- Publish PAS results

Biaxial Creep of Zry Cladding

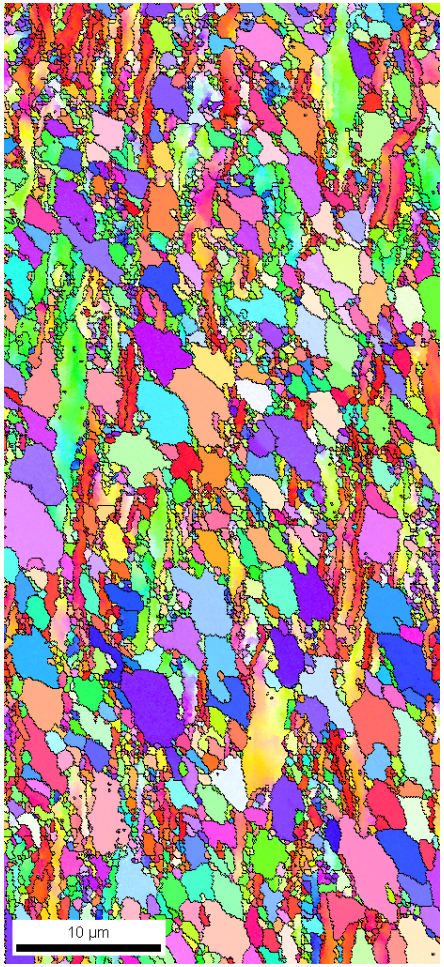
KL Murty, NC State University
(2010–11 effort)

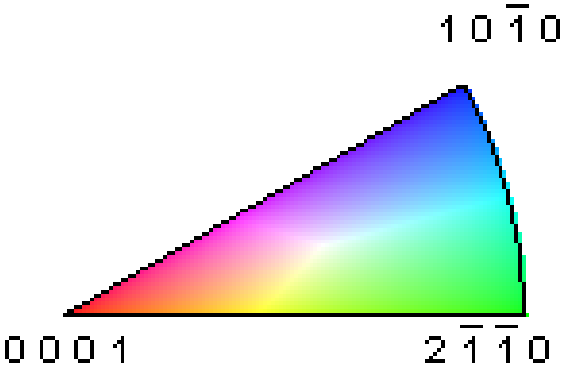
- High Temperature furnace was procured along with suitable LVDT extensometer for creep measurements at higher temperatures
- Creep testing will be performed on HANA alloy (Zr-1.5Nb-0.4Sn-0.2Fe-0.1Cr)
- Texture of as-received material was evaluated using





HANA alloy
As-received
(OIM)





IPF

IQ

INEST MEASURES OF SUCCESS

- Publications including conference, journal, and special workshop reports: ***1 published, 4 under review, 4 in preparation***
- Reports: 1 in preparation
- Number of students/post-docs supported: ***5 postdocs, 5 students***
- Number of stimulated invited talks: ***1 at a major corrosion conference in France***
- External grants stimulated at partner institutions (BES, NSF, etc.): ***Two university proposals to NSF***
- Staff exchanges, Patents, Community interface: ***None yet***

FY 2012 GOALS AND OBJECTIVES

- **New Fuels & Materials CORE white paper (set objectives for next three years)**
- **Workshop Series**
  - May 2012 Uranium Fuel Science
- **Outreach Events**
  - Feb 2011 Regional Workshop (TBD)
- **LDRD**
  - 2012 Uranium Fuel Science

FY 2013/2014 GOALS & OBJECTIVES

- **Execute vision of next Fuels & Materials CORE white paper**
- **Workshop Series**
  - Potential focus on application of advanced experimental and modeling techniques
  - Potential focus on spent fuel storage
- **Outreach Events**
  - Feb 2013/14 Regional Workshops
- **LDRD**
  - Potential focus on application of advanced experimental and modeling techniques or spent fuel storage

RISKS AND RISK MANAGEMENT

- **Lack of support for LWR-related research and development**
  - Engage with LWRs program to ensure continued interest in LWR issues. Post Fukushima and post BRC, the LWR interest areas may change and change the CORE focus. Avoid overlap with NS&T
- **Inability to engage industry in eventual support for follow-on R&D activities**
  - Work with F&M Steering Committee to engage industry in supported research and development
- **Decrease in support for university R&D programs where further growth could occur**
  - Encourage university partners to perform follow-on work through NEUP

NEEDS

- **The CORE needs to work with newly formed teams to move beyond the limits of the LDRD projects:**
  - Use NSUF capability
  - Compete for national program tasks (a venue that allowed for lab-university teams to truly partner in competitions would be beneficial)
- **The CORE, recognizing the NS&T programs are showing a greater interest in LWR issues, needs to plan for the next set of tasks that grow the intellectual range of the lab**

SUMMARY

- Fuels and Materials CORE has focused on LWR issues
- Through a series of workshops with targeted topic and attendees, we are forming teams composed of lab and university staff engaged in LWR issues
- Five projects are ongoing looking at SCC in steels and corrosion/hydriding of Zr-base alloys
- FY12 is a key year to devise the next three year plan