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U.S. DEPARTMENT OF  
**ENERGY**

**Nuclear Energy**

## Fuel Cycle Research and Development: **Core Materials Technologies**

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**Core Materials Technical Lead for Fuels**  
**Los Alamos National Laboratory**

**DOE-NE Materials Cross-Coordination  
Meeting**

**8/14/12**

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## Grand Challenge for Core Materials for High Burnup and Next Generation LWR Fuels

**Develop and test advanced alloys for clad and duct and other high dose core components to >400 dpa over the clad /duct operating conditions**

- Irradiation tolerant
  - *Resists swelling and creep*
  - *Does not accumulate damage (resists hardening and embrittlement)*
  - *Stable microstructure (resists radiation induced segregation)*
  - *Manages helium or other gas buildup*
  - *Stable with Transmutation impurity buildup*
- Resist chemical interaction with fuel (for the cladding)
  - *Not reactive with fuel*
  - *Prevent diffusion into cladding*
- Corrosion resistance with coolant
  - *Protective oxide layer*
  - *Non reactive with coolant*
- Weldable and Processed into tube form

**Develop and test advanced alloys for Next Generation LWR Fuels with Enhanced Performance and Safety and Reduced Waste Generation**

- Low Thermal Neutron Crossection
  - *Element selection (e.g. Zr, Mg)*
  - *Reduce cladding wall thickness*
- Irradiation tolerant to 20-40 dpa
  - *Resists swelling and irradiation creep*
  - *Does not accumulate damage*
  - *Stable microstructure (resists RIS)*
- Mechanically robust under loading and transportation conditions
- Compatibility with Fuel and Coolant
  - *Resists stress corrosion cracking*
  - *Resists accident conditions (e.g. high temperature steam)*
  - *Resists abnormal coolant changes (e.g. salt water)*
- Weldable and Processed into tube form
  - *Maintain hermetic seal under normal/off-normal conditions*



# Outline-Approach

## h Develop the knowledge base up to 200 dpa- High Dose Core Materials Irradiation Data

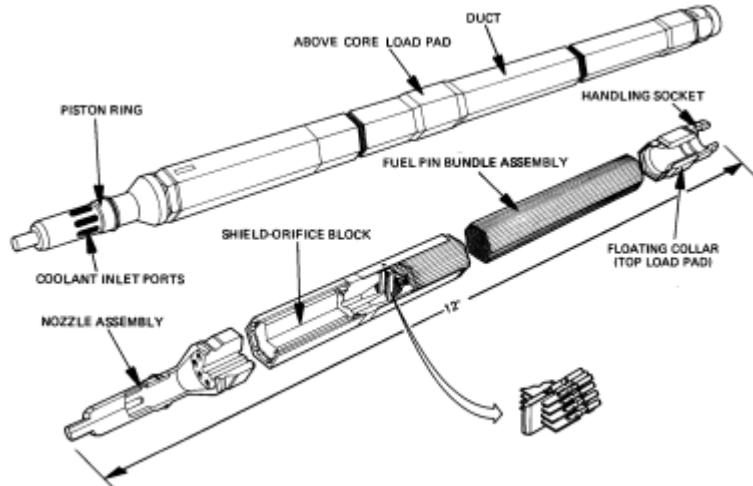
- ACO-3 Duct Testing
  - *Rate Jump testing*
- FFTF/MOTA testing

## h Advanced Material Development

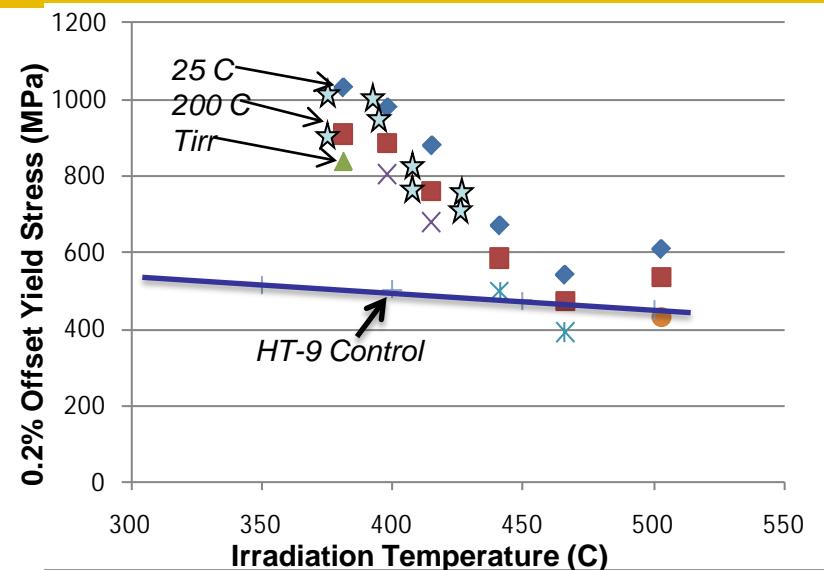
- Develop coatings/liners to Mitigate FCCI
- Develop and test Advanced Cladding materials
  - *Improved Processing of Advanced ODS Alloys*
  - *Tube forming processes for ODS alloys*
  - *Steam oxidation tests*

## h International and University Collaborations

## h Outlook

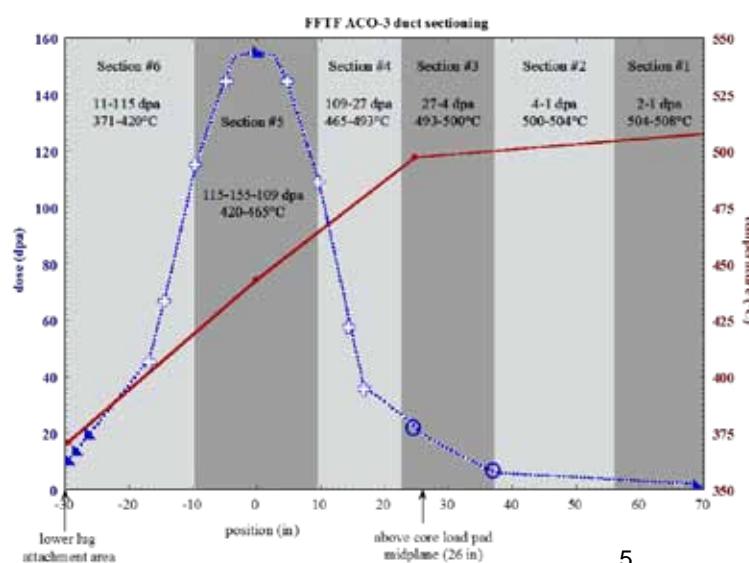


# Analysis of Specimens from ACO-3 Duct



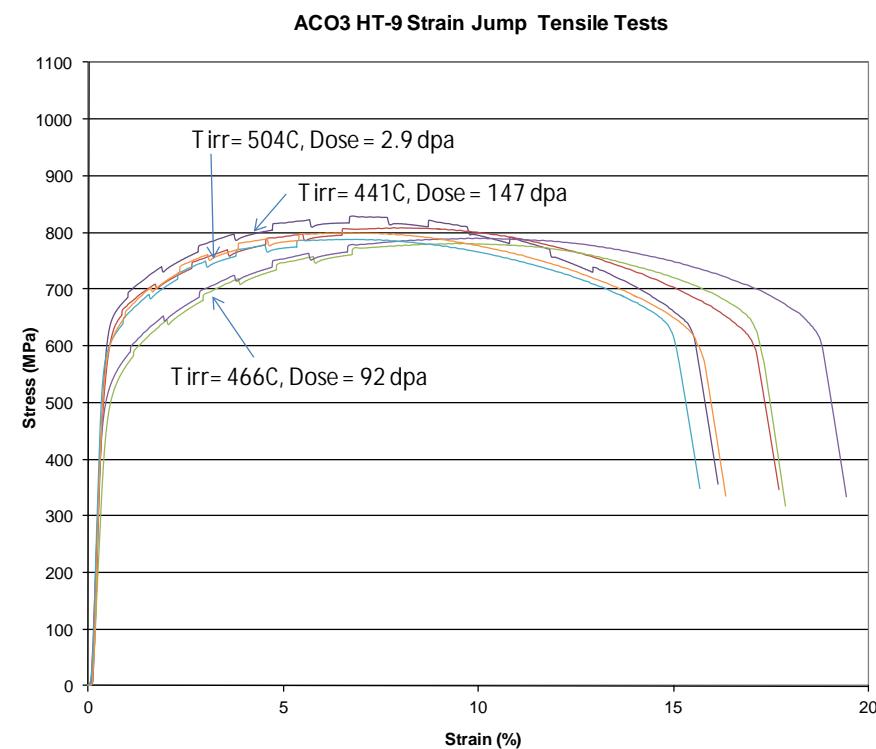
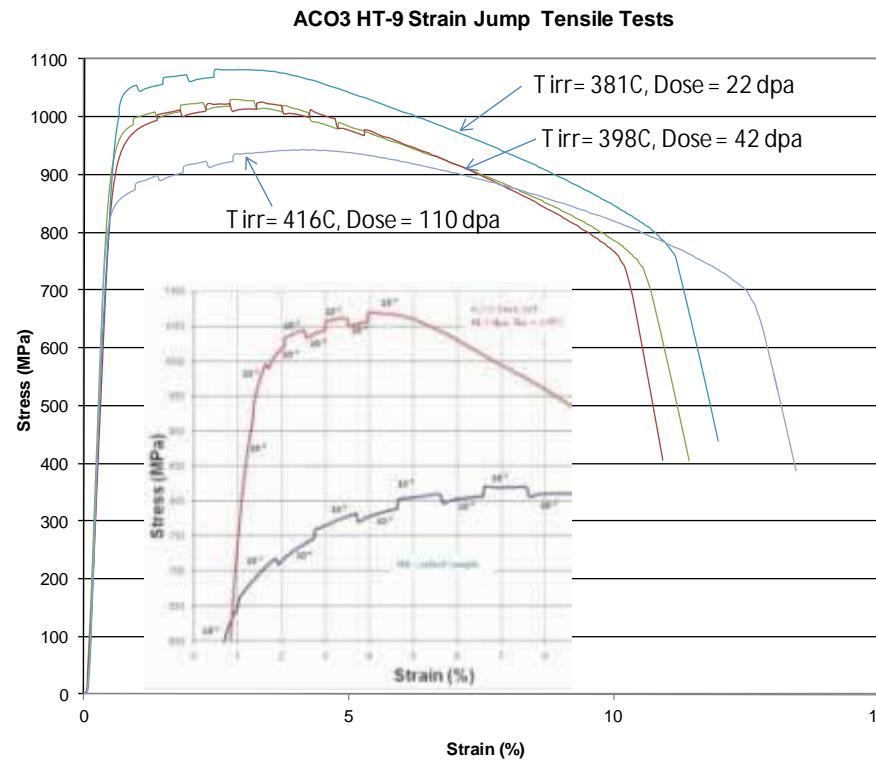
## Significance of ACO-3 duct Analysis

- Provides actual data on a component irradiated in a sodium fast reactor to very high dose (155 dpa)
- Opportunity to perform more detailed analysis
  - TEM from FIB'ed specimens
  - Atom probe analysis
  - SANS analysis
- Aids in physics-based model development
  - Provide new data required for model development
  - Provides a complete data set for model verification
- Provides direction in future alloy development for radiation tolerant materials





# Rate Jump testing results completed at RT on ACO-3 Duct

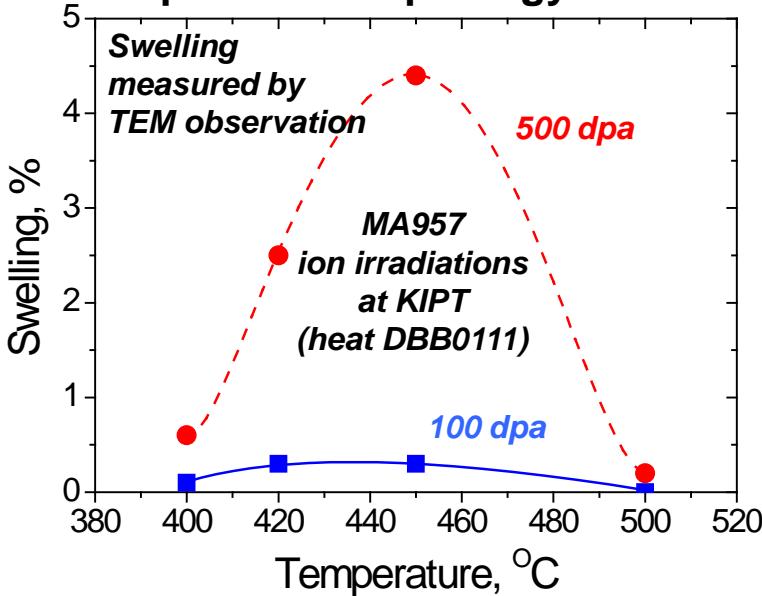


- Two rate jump tests performed at each of 6 irradiation conditions for the ACO-3 duct
- Strain rate varied between  $10^{-3}$  and  $10^{-4}$ /s.
- No change in strain rate sensitivity observed for materials after irradiation.
- Data is being shared with NEAMS modelers to aid in clad model development.

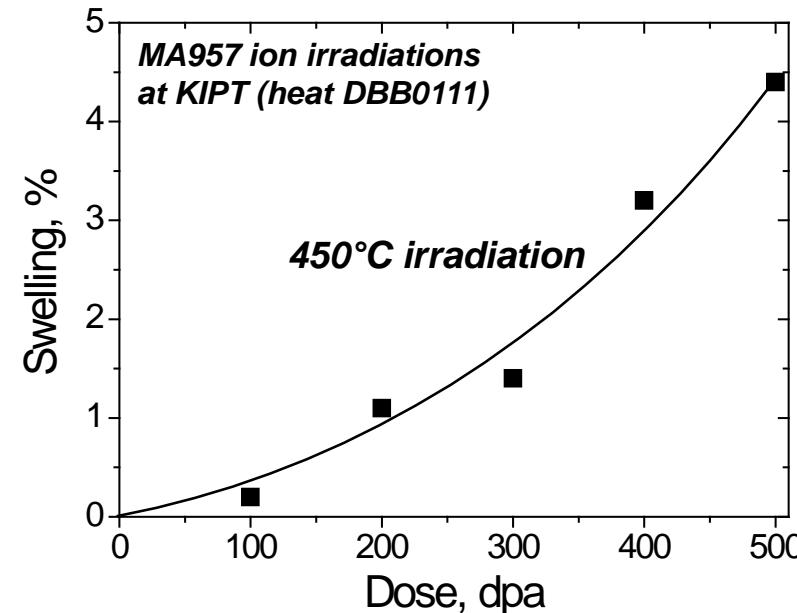


# PNNL High Dose MA957 ODS Steel Examination

1. Continued TEM observations on 110 dpa neutron irradiated MA957 with focus on identifying relationship between oxide particles and dislocations in creep microstructures.
2. Continued APT examination of oxide particles in neutron irradiated and ion irradiated MA957 to study the effect of irradiation dose and temperature on oxide particle morphology.



3. Ion irradiations at KIPT in the Ukraine.
  - ▀ Performed ion irradiations on MA957 tubing to match doses obtained on neutron irradiated MA957 tubing (~110 dpa). Then ion irradiate to 500 dpa.
  - ▀ Almost no swelling after 100 dpa at 400 ° C and 500 ° C, consistent with neutron irradiated results.
  - ▀ Continuous swelling from 100 dpa





# High Dose MA957 ODS Steel – APT Examinations at PNNL/UC Berkeley

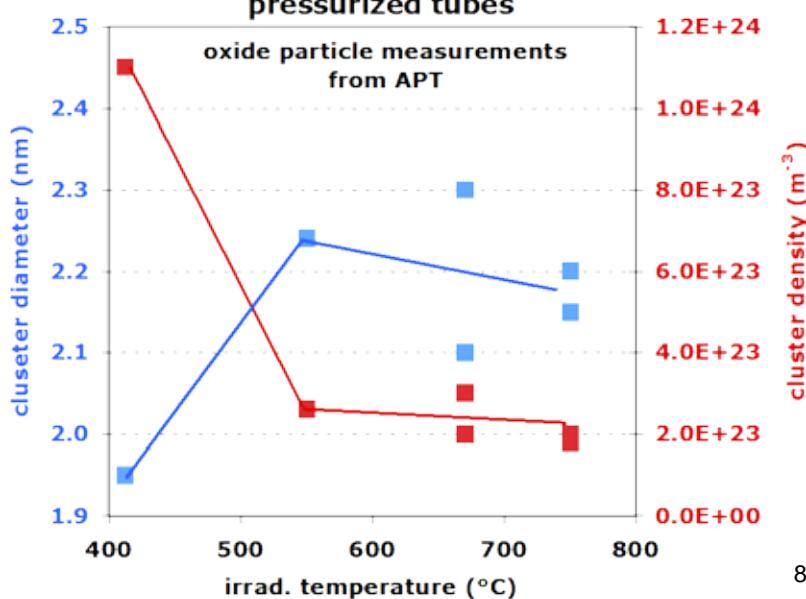
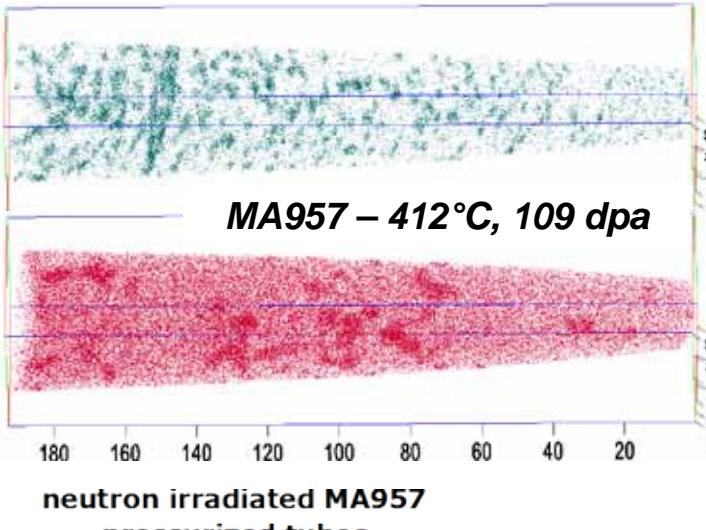
- Objective - study effect of irradiation dose and temperature on oxide particle morphology.
- Using 110-120 dpa neutron irradiated MA957 from in-reactor pressurized tube creep specimens.
- Preliminary APT examinations completed on specimens irradiated at 412, 550, 670, and 750°C to 109-121 dpa.

## Initial Results

- MA957 pressurized tubes have a small oxide particle size of ~2 nm similar to newer ODS steels such as 14YWT.
- No ballistic dissolution at these irradiation temperatures, but small difference in oxide particle population at 412°C vs higher temperatures.
- Cr-rich alpha-prime clusters observed at 412°C irradiation temperature.

*TiO signal  
from oxide  
particles*

*Cr-  
rich  
alpha  
prime*





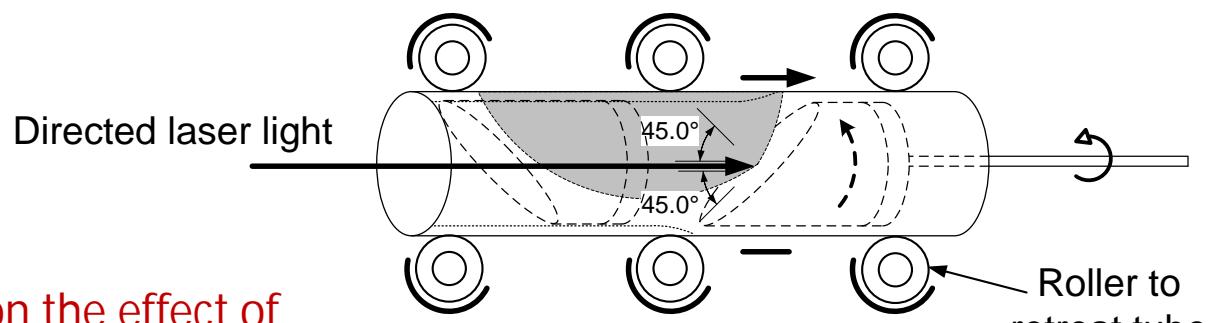
# Advanced Material Development

- **Developing Coatings and Liners to prevent FCCI**
- **Obtaining Irradiation data on Advanced Alloys (international collaborations)**
  - MATRIX irradiations- Samples to be shipped in early 2013?
  - STIP irradiations – Samples from STIP IV being analyzed. Tensile testing completed at LANL and ORNL.
- **Investigating Possible Future irradiations**
  - Domestic Facilities (MTS (18 dpa/yr) or HFIR) – Collaborating in ATR irradiations
  - International collaborations
    - *Collaborating with Terrapower for irradiations in BOR-60 in Russia*
    - *Initial discussions under way for future irradiation in the CEFR in China.*
- **Advanced Material Development**
  - Friction stir ODS material processing
  - Mechanical alloying ODS material processing
  - Development of high temperature steam resistant LWR cladding mats.

# Coating and Diffusion Couple Study for FCCI Mitigation

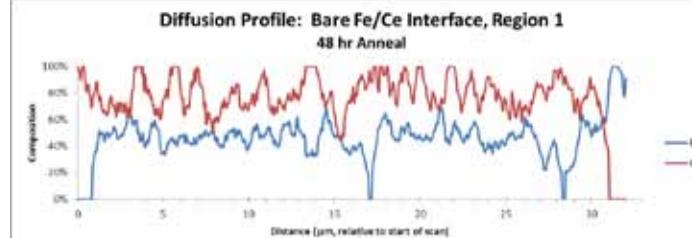
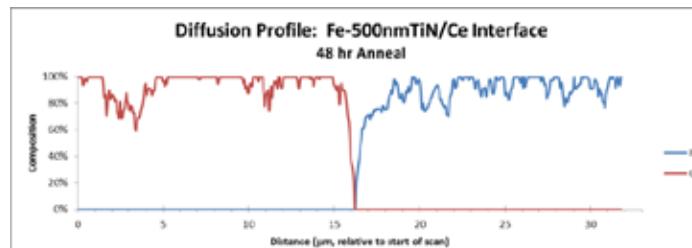
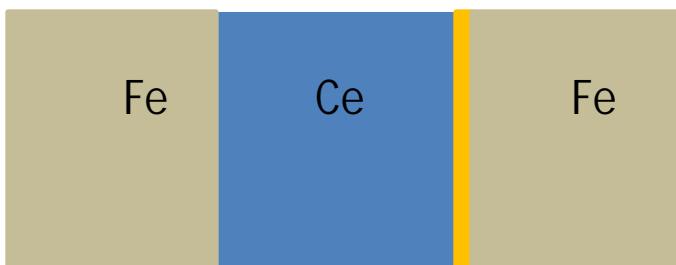
A customer-designed laser deposition system for inner wall coating of long tube will be available at Texas A&M University for the FY2012 coating work

Retreating tube with rotating target pellet holder



Diffusion couple studies on the effect of ceramic coating on suppressing fuel-cladding-chemical-interaction (550 – 600 °C for 12-24 hours)

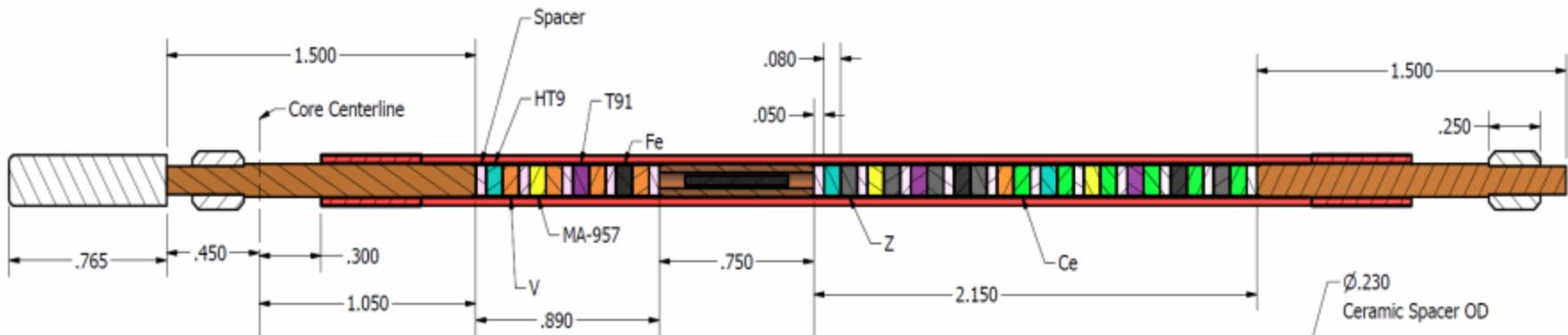
Ceramic coating



The effect of a 500 nm thin TiN coating on Fe-Ce interaction (550°C/48 hrs)

# Diffusion Couple Studies on FCCI Mitigation

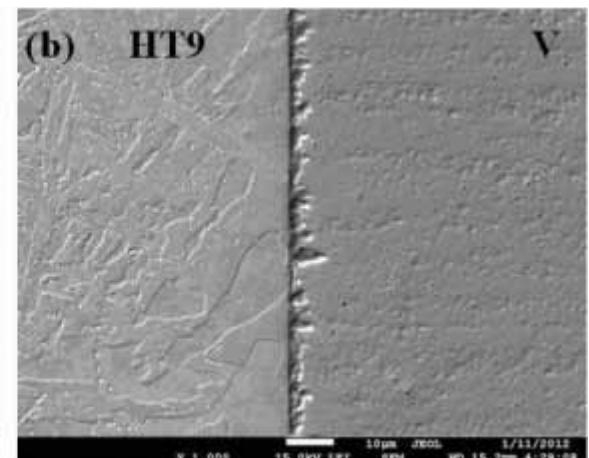
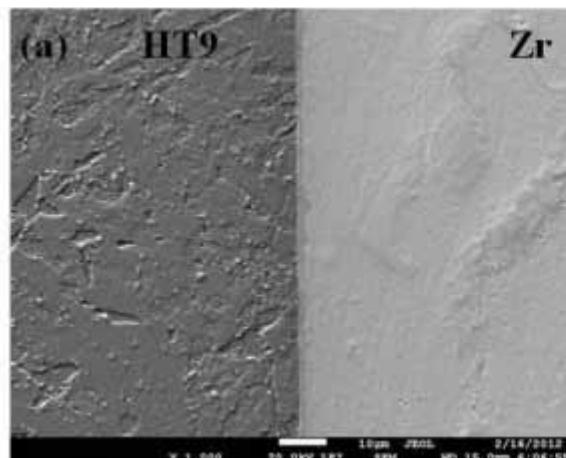
Re-design for diffusion couple irradiation experiment in ATR (550 °C for 50 days) to meet the temperature and post-irradiation-examination requirements.



Interaction zone thickness: 0.5  $\mu\text{m}$   
Interdiffusion zone thickness: 18  $\mu\text{m}$

Interaction zone thickness: 8  $\mu\text{m}$   
Interdiffusion zone thickness: 28  $\mu\text{m}$

Diffusion couple thermal annealing studies on chemical compatibility at the cladding – liner interface (HT-9 vs. V or Zr). (704 – 815 °C for 50-200 hours)



704 °C for 200 hrs

# High Toughness ODS Alloy Development in FC R&D (I-NERI)

- *Development and Characterization of Nanoparticle Strengthened Dual Phase Alloys for High Temperature Nuclear Reactor Applications*
- To develop high toughness NFAs for high temperature (700°C) high dose (>300 dpa) applications:  $100 \text{ MPa} \sqrt{\text{m}}$  over the range of RT - 700°C.
- Use grain boundary strengthening/modification techniques.
- *ORNL (TS Byun & D.T. Hoelzer) – KAERI (JH Yoon)*
- *Dec. 1, 2010 – Nov. 30, 2013*

\* *Nanostructured Ferritic Alloys (NFAs) vs. Oxide Dispersion Strengthened (ODS) Alloys*



# Production of Base Materials (9YWTV)



Two alloy power heats (8 kg each) have been produced by gas atomization process at ATI Powder Metals:

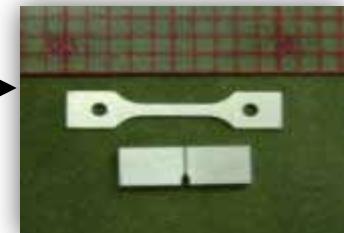
**Fe-9Cr-2W-0.4Ti-0.2V-0.12C+0.3Y<sub>2</sub>O<sub>3</sub>** &  
**Fe-9Cr-2W-0.4Ti-0.2V-0.05C+0.3Y<sub>2</sub>O<sub>3</sub>**



Ball milling for 40 hours in Zoz CM08 machine (6 loads)/ Canned & degassed (6 cans, 920g each)



Extruded below 850°C  
Cut into 4 inch long blocks



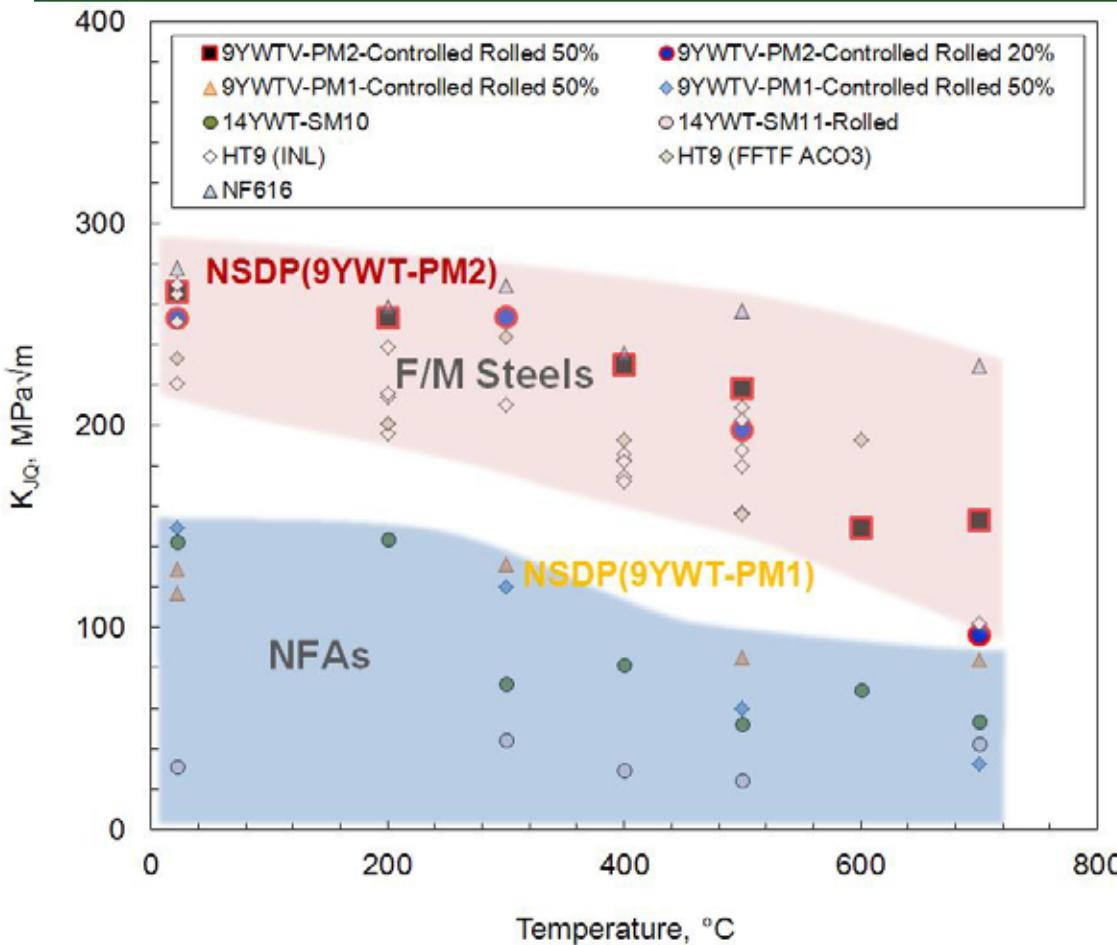
Characterization

## Goals of Yrs 2 & 3:

- Post-Extrusion TMT Optimization
- Micro & High Temp. Characterization
- Feedbacks for new processing



# Preliminary Results for Fracture Toughness



- 9YWT-PM2-850C-200m: Annealed at 850°C for 200 minutes.
- 9YWT-PM2-850C-20H: Annealed at 850°C for 20 hours.
- 9YWT-PM2-900C-50%R: Hot-rolled for multi-step 50% reduction after annealing at 900°C.

- ∅ Fracture toughness can be significantly improved by some controlled TMTs, and the  $K_{JQ}$  values are between those of other NFAs and FM steels.
- ∅ Further development/optimization of processing is underway.

# Scale Up Processing Study of 14YWT

- The first ball milling experiment of the large L2314 powder heat was completed at the Zoz pilot plant
- ~55 kg of powder was separated into 3 size ranges
  - *Coarse: 150-500 mm*
  - *Middle: 45-150 mm*
  - *Fines: <45 mm*
- Prior ball milling experiment using CM08 at ORNL indicated that the middle and fine size particles could be mixed and ball milled together
- Ball milling of three 15 kg batches of coarse, fine and medium powder was completed by Zoz
  - 40 h in Ar using parameters supplied by FCRD processing team
  - Powder samples were taken at 20 h and 40 and chemically analyzed
  - EPMA analysis completed on 20 h and 40 h samples



# Coarse powder ball milled using CM100 (100 liter capacity)

- CM100 and extracted samples for analysis



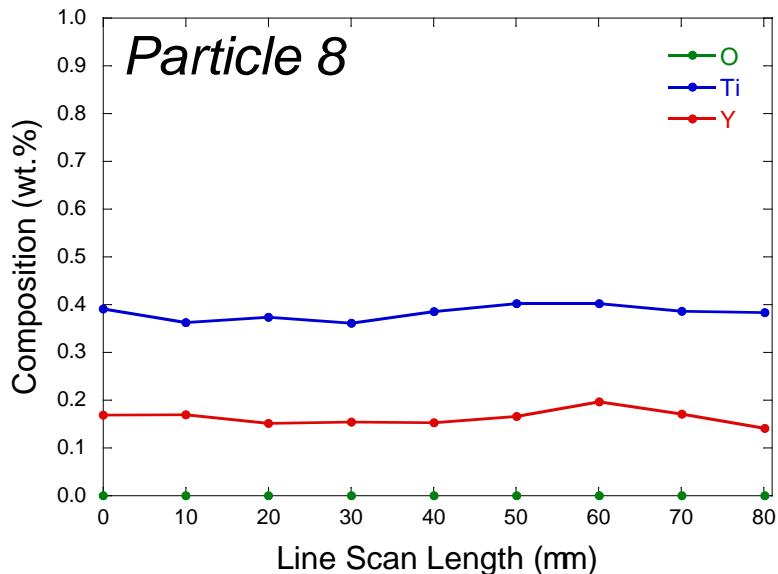
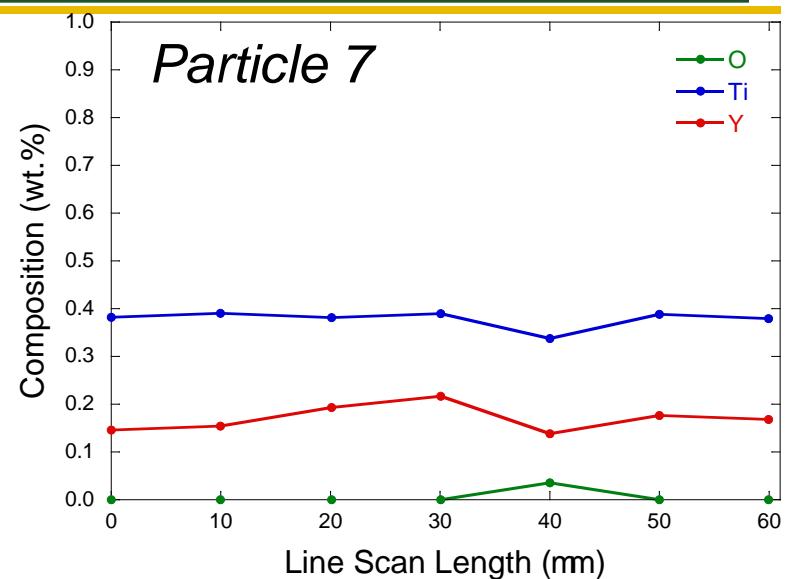
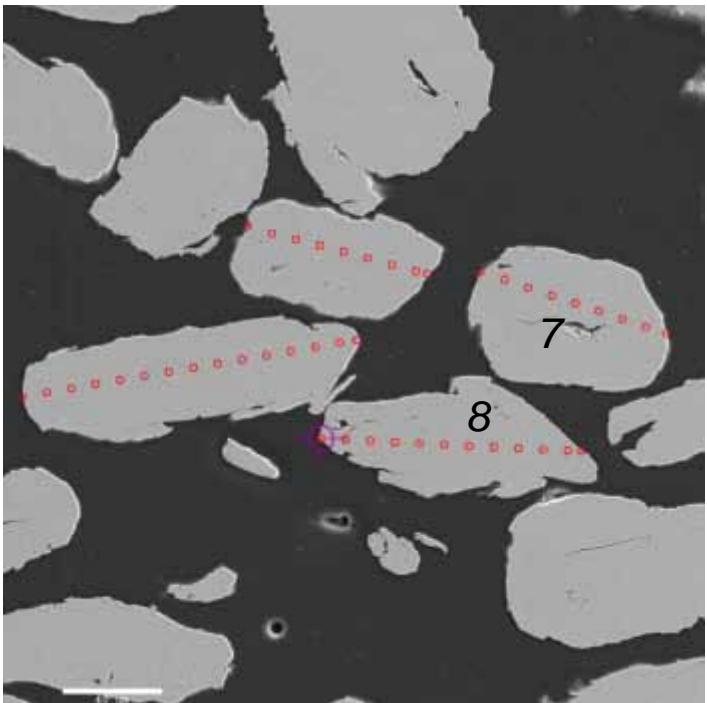
- Chemical analysis showed the ball milling condition achieved the desired goal of elevated O level and low N and C levels

Sample Identification	<u>20hr Milling</u>	<u>40hr Milling</u>
V540-01	%	%
Oxygen	.091	.111
Nitrogen	.006	.008
Carbon	.012	.019
Chromium	13.7	13.7
Tungsten	2.90	2.88
Titanium	.38	.38
Yttrium	.18	.19



# Electron Probe Microanalysis of Ball Milled Coarse Powder

- For Medium and fine powders, uniform mixing observed at 40 h.
- For Coarse powders >40 h ball milling is required for uniform mixing



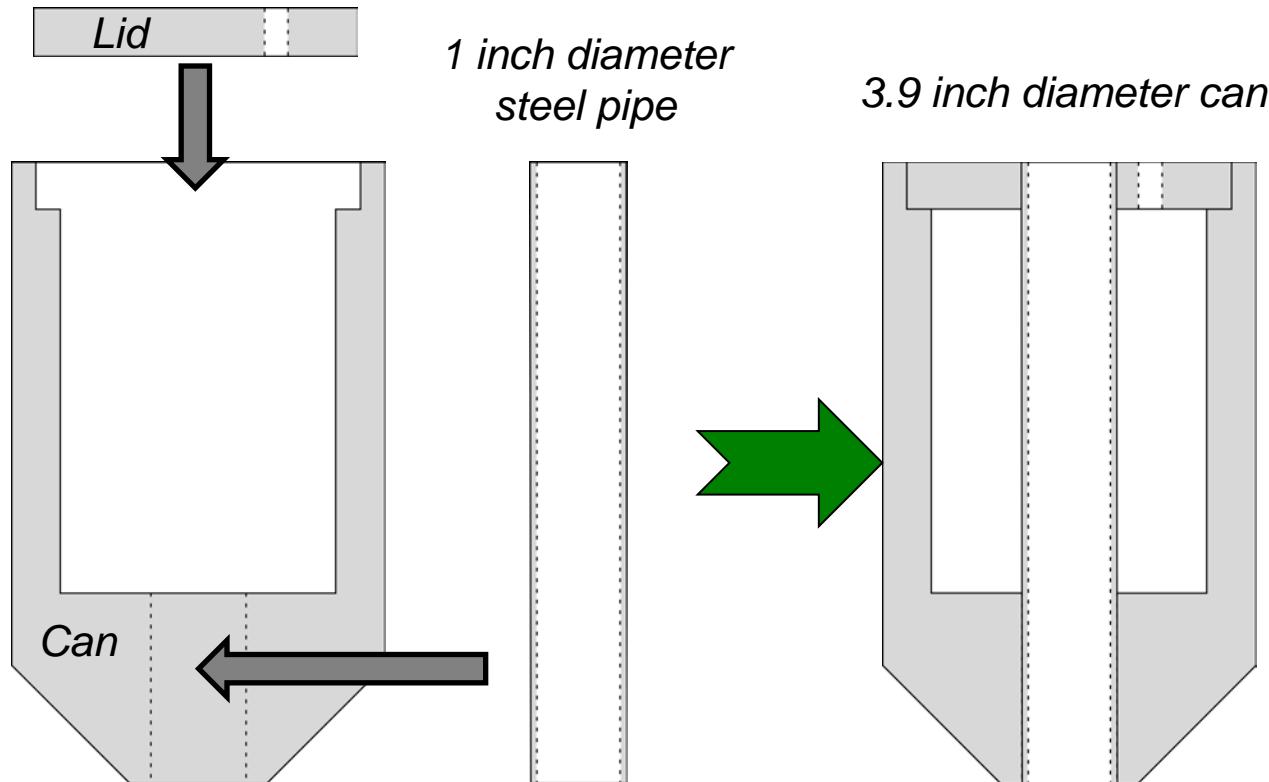


# Production of Thick Walled Tubing from 14YWT Heats

- Can was designed for producing thick wall tubing of 14YWT
- Design will use a 0.82 inch diameter mandrel
- 3 cans are currently being fabricated in local shop
- Powder is currently being ball milled with CM08

## Procedure

- Steel pipe is welded in the bored hole in the can
- Can is filled with ball milled powder
- Lid is welded to can and steel pipe
- Powder is degassed
- Can is extruded with mandrel through circular die
- Mandrel removed to form thick walled tube



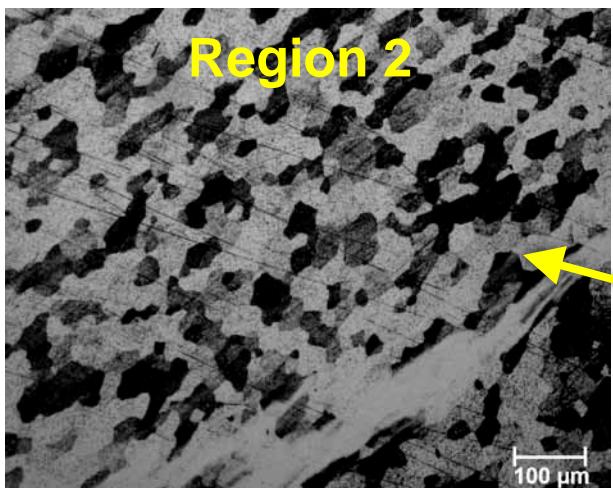


# PNNL Innovative Fabrication of ODS Steel Tubing

- Objective: Create economical method to fabricate ODS steel tubing directly from powders using a friction stir consolidation and extrusion method.

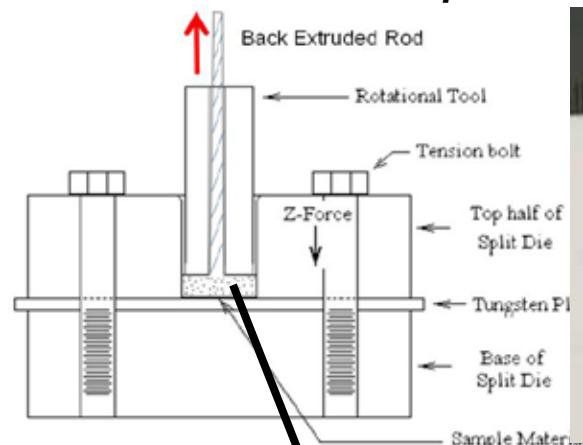
## FY12 Activities

- First attempt at consolidating and extruding 14YWT gas-atomized powders into rod.
- Powders successfully compacted but did not extrude through the throat.
- Several regions of different microstructure



20-50  $\mu\text{m}$  size grains observed in regions 1-4

*schematic of FCE concept*

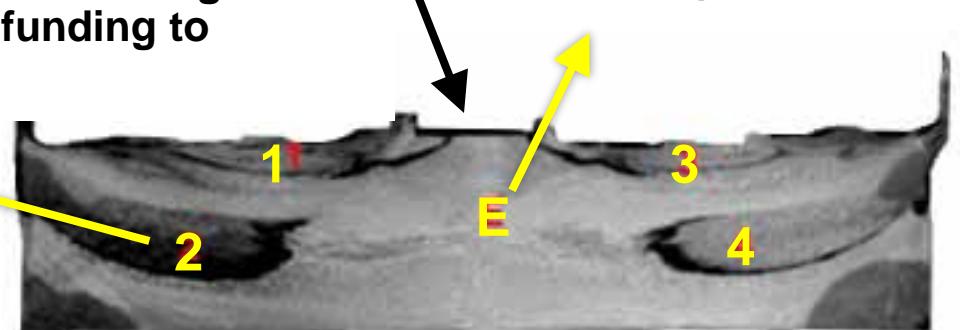


*friction stir tool face*



- Regions of nearly equiaxed grains of desirable size.
- Good start... awaiting additional funding to continue.

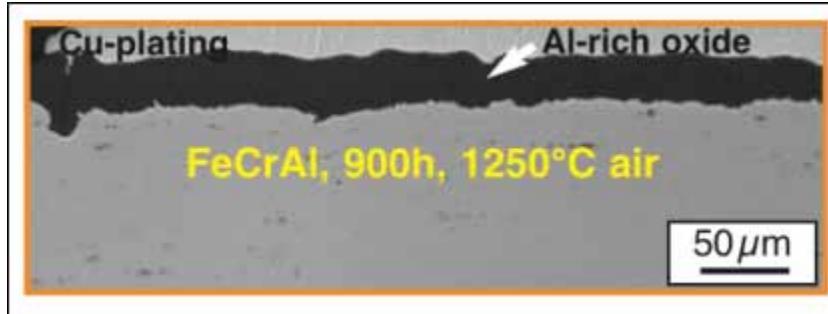
Region "E" had ultrasmall ~2  $\mu\text{m}$  grains



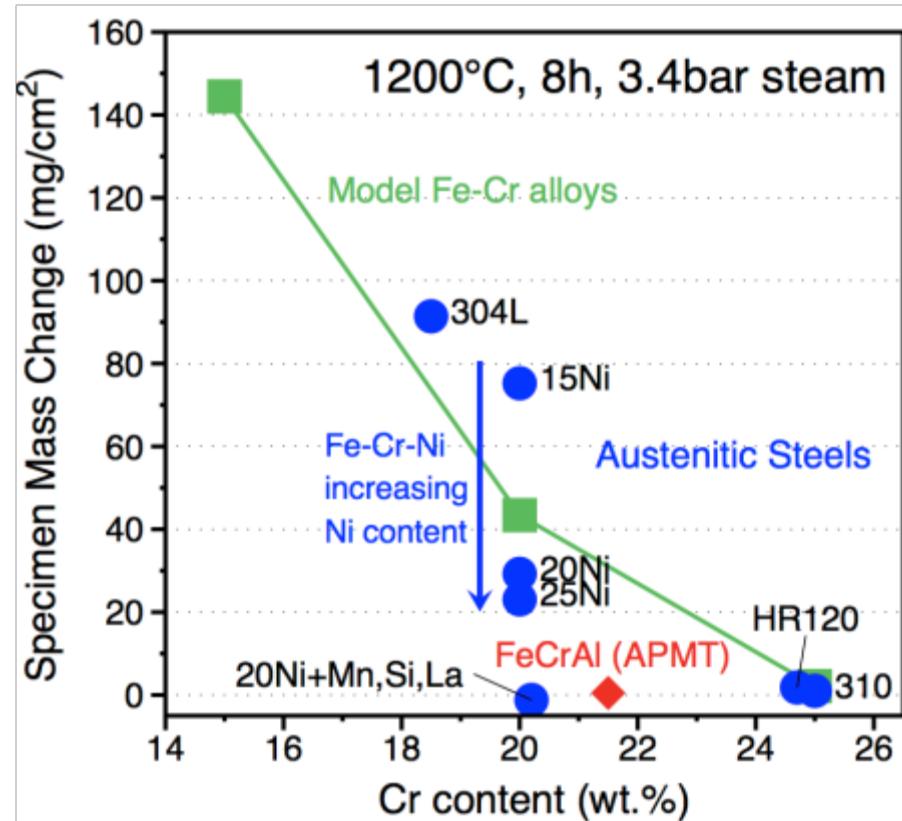
Friction consolidated gas atomized 14YWT powders

# Properly alloyed metals as protective as Si-based ceramics at 1200 °C

- Example from ORNL experiments at 1200 °C in steam at 3.4 bar (50 psia) for 8 h
- All low mass gain: 310SS ( $\text{Cr}_2\text{O}_3$ ), FeCrAl, Kanthal APMT ( $\text{Al}_2\text{O}_3$ ), CVD SiC ( $\text{SiO}_2$ )



- In the ~1200°C range alumina forming alloys (AFA's) compete well with silica formers (SiC .)
- Above 1200°C it is anticipated that AFA's will outperform silica formers, though to what extend is not known.



# Materials Integration and University and International Collaborations

## Integrate FCRD Core Materials Activities

- Fuels Core Materials Work- (INL, PNNL, LANL, ORNL, LLNL)
  - *Materials teleconferences monthly*
- University Materials Research (attend university review, review quarterly progress reports)
  - UCSB- *Optimized Compositional Design and Processing-Fabrication Paths for Larger Heats of Nanostructured Ferritic Alloys*
  - TAMU-*Bulk nanostructured austenitic stainless steels with enhanced radiation tolerance*
  - U. Ill Urb/Champaign-*Development of Austenitic ODS Strengthened Alloys for Very High Temperature Applications*
- ATR Reactor Irradiations (provide materials and preparing to collaborate in testing)

## Working group meetings and Workshops

- LANL will host next NE Materials Cross-cut Meeting through a webinar in August 2012

## International Collaborations

- INERI-GETMAT- 14Cr ODS material development
- INERI-KAERI- 9Cr ODS material development
- Participant in IAEA Coordinated Research Project on “Benchmarking of Structural Materials Pre-selected for Advanced Nuclear Reactors” – met in Vienna, May 2-6, 2011.
- DOE-CIAE Collaboration – Proposed irradiation in CEFR
- DOE-Russia – Proposed irradiation in BOR-60
- LANL-Terrapower CRADA – proposed irradiation of ACO-3 specimens in BOR-60



# Core Materials Research and Development – 5 Year Plan

## Qualify HT-9 for high dose clad/duct applications (determine design limitations)

FFTF (ACO-3 and MOTA) Specimen Analysis

Rev. 6 of AFCI (FCRD) Materials Handbook

Data to 250-300 dpa on F/M and 100-150 on Inn. Material

Re-irradiation of FFTF specimens in BOR-60

## Advanced Material Development (improved radiation resistance to >400 dpa)

STIP- IV (PSI) Specimen PIE

Materials Test Station Irradiations

MATRIX-SMI and 2 (Phenix) Specimen PIE

Data on Advanced Materials to 80-100 dpa

ODS Ferritic Steel Material Development

Develop ODS Tubing and Weld specifications for innovative Weld material

Produce ODS Tubing

Advanced Materials Irradiation in BOR-60 and CEFR

## Advanced Material Development (improved FCCI resistance to >40 % burnup)

Development of Coated and Lined Tubes

PIE on Lined Irradiated Tube

## Advanced Material Development (improved accident tolerance for LWR's)

Oxidation testing in Steam on Advanced Steel Alloys

Continued development of Advanced LWR alloys in collaboration with FOA

Thin walled tube development for advanced steel alloys

Develop Steel thin-walled tubing

FY'11

FY'12

FY'13

FY'14

FY'15

FY'16

Provides data for NEAMS model development of Cladding



# Core Materials Research and Development – ATF- Clad Development- 5 Year Plan

## Advanced Material Development (improved accident tolerance for LWR's)

Oxidation testing  
in Steam on  
Advanced Steel  
Alloys

Oxidation testing in Steam  
on Advanced Steel Alloys

Oxidation testing in Steam  
on Advanced Steel Alloys  
after irradiation

Final report of LOCA  
testing on ATF clad  
materials

Thin walled tube development  
for advanced steel alloys

Develop Steel thin-walled tubing

Weld development for thin walled tubing

Qualified weld procedure for thin  
walled tubing

Produce thin walled tubing for ATF  
irradiation

ATR irradiation of ATF cladding with  
advanced fuels

Report on  
irradiation  
testing of  
clad  
materials to  
at least 15  
dpa

Irradiation testing and PIE on ATF clad materials

Corrosion testing on ATF clad materials

IASCC testing on ATF clad materials

Summary  
report on  
corrosion  
resistance  
of ATF clad  
materials

Coordinate and collaborate with ATF fuel development with industry through FOA

FY'11

FY'12

FY'13

FY'14

FY'15

FY'16



## Future work in FY12

- Prepare mechanical test specimens from ACO-3 duct for re-irradiation in BOR-60
- Perform mechanical testing on STIP-IV irradiated specimens (total dose up to 24 dpa)
- Complete summary report on FFTF/MOTA specimens tested up to 200 dpa
- Fabricate coated tube for performing fuels irradiation
- Development and testing of 9Cr NFA and large scale heat of 14YWT