



U.S. DEPARTMENT OF  
**ENERGY**

**Nuclear Energy**

# Status of SiC Research for Accident Tolerant Fuels

*Shannon Bragg-Sitton*  
*Idaho National Laboratory*

*Advanced LWR Fuels Technical Lead*  
*Advanced Fuels Campaign*

*Advanced LWR Fuels Pathway Lead*  
*Light Water Reactor Sustainability Program*

*August 2013*

## Nuclear Energy

---

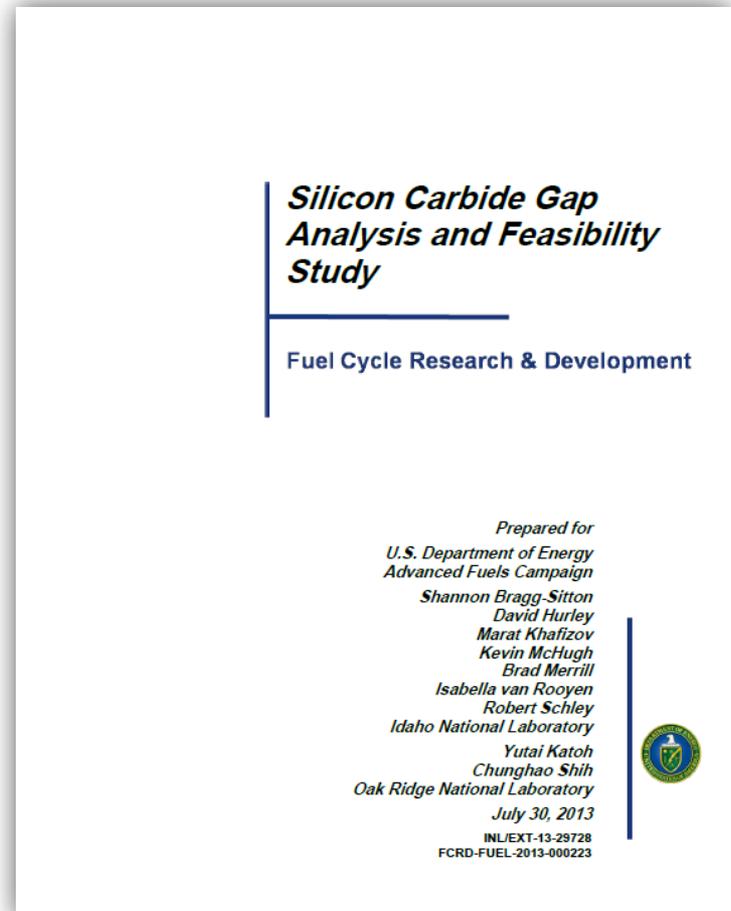
- **Overview of DOE SiC research**
- **Severe accident modeling: MELCOR analysis w/SiC**
- **Recent characterization test results**
  - Oxidation kinetics
  - Irradiation studies
  - Fuel-clad interactions
  - Elastic property measurement
  - Thermal properties
  - Failure model analysis
  - Quench testing
- **Technology development**
  - ASTM standards development
  - SiC/SiC joining technology



# SiC Gap Analysis and Feasibility Study

## ■ SiC Gap Analysis / Feasibility

- Milestone report issued July 30, 2013
- Incorporates results of work funded under multiple programs:
  - *Fuel Cycle R&D Advanced Fuels Campaign*
  - *Light Water Reactor Sustainability*
  - *Fusion Materials Research (Office of Science)*





## Key Outcomes of SiC Gap Analysis and Feasibility Study

- **Preliminary computational analysis results (SiC-modified MELCOR) suggest significant improvement in the reactor coping time with SiC materials versus Zr-alloys under a selected loss of coolant accident (LOCA) scenario**
  - Applies available monolithic SiC oxidation data
  - Includes sensitivity analysis on oxidation rate (10x) and thermal conductivity (0.1x) – minimal peak T difference
- **Many data and technology gaps remain, particularly for SiC composite materials**
- ***Insufficient data currently available to draw a clear conclusion regarding the potential licensing and adoption of SiC components in LWR applications***
- ***Specific recommendations for additional testing and characterization discussed at the end of this presentation***

# PRELIMINARY MODELING RESULTS – SEVERE ACCIDENTS



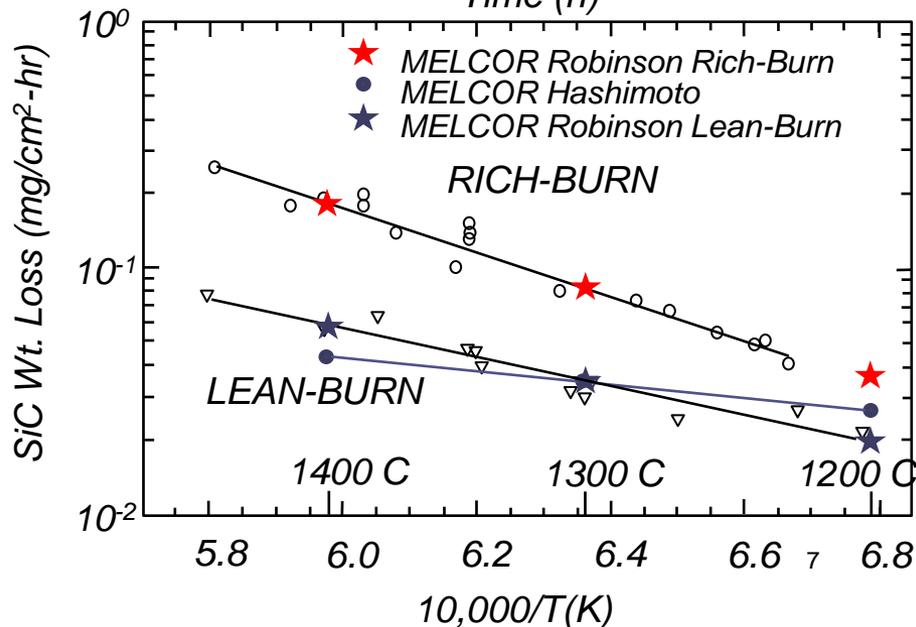
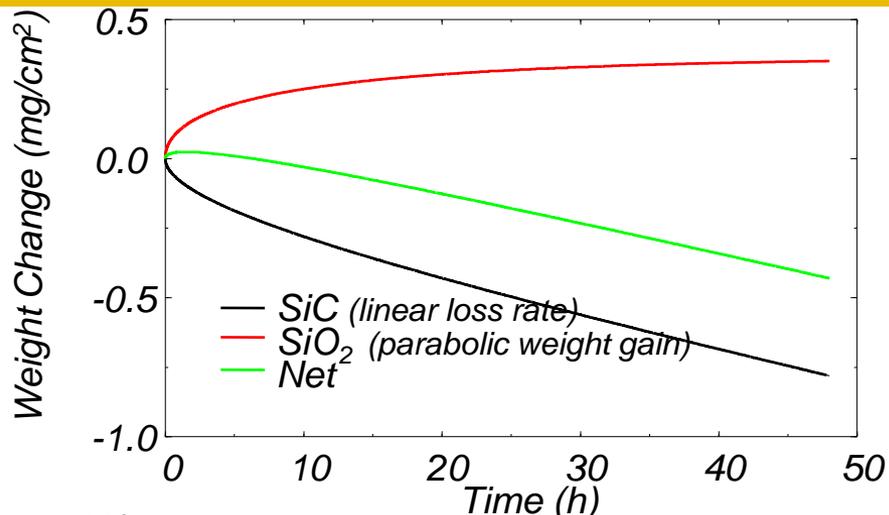
# Modeling and Analysis of Advanced Cladding and Components

- **Scoping analyses to estimate potential performance improvements for ATF concepts under normal operating conditions, off-normal conditions and severe accidents.**
  - Normal operation (BISON) –
    - *Addition of SiC material properties*
    - *Capability to model multi-layered cladding*
  - Severe Accidents (MELCOR)
    - *Addition of SiC properties (alternate for Zirconium alloy)*
    - *Currently modifying to allow additional material, multi-layered materials*
  - Scoping Probabilistic Risk Assessment
    - *Within LWRS Risk Informed Safety Margin Characterization*
    - *Incorporation of MELCOR and BISON results to perform PRA*
    - *Development of analysis framework - in progress*
  - Baseline accident scenarios (identified in International Metrics meeting):
    - *TMI-2 (INL)*
    - *Fukushima 3 (ORNL)*



# MELCOR Severe Accident Analysis for SiC Cladding and Components

- A modified version of MELCOR with Zr replaced by SiC, including the nonlinear oxidation process for monolithic SiC in steam and oxygen atmospheres
- This version of MELCOR was benchmarked against data from R. P. Arnold<sup>1</sup>, R. C. Robinson<sup>2</sup>, and E. J. Opila<sup>3</sup> to ensure that the oxidation model was working properly
- SiC-modified MELCOR was applied to a BWR (Peach Bottom) mitigated long-term station blackout and a PWR (Surry) unmitigated short-term station blackout accident scenarios<sup>4</sup>



<sup>1</sup>Master of Science Thesis, MIT, September 2011

<sup>2</sup>American Ceramic Society, 82 [7], 1999, 1817-25

<sup>3</sup>American Ceramic Society, 80 [1], 1997, 197-205

<sup>4</sup>INL/LTD-12-27861, December, 2012



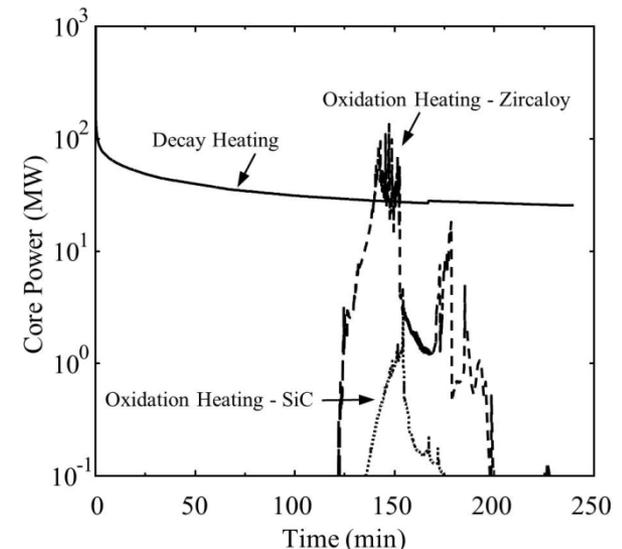
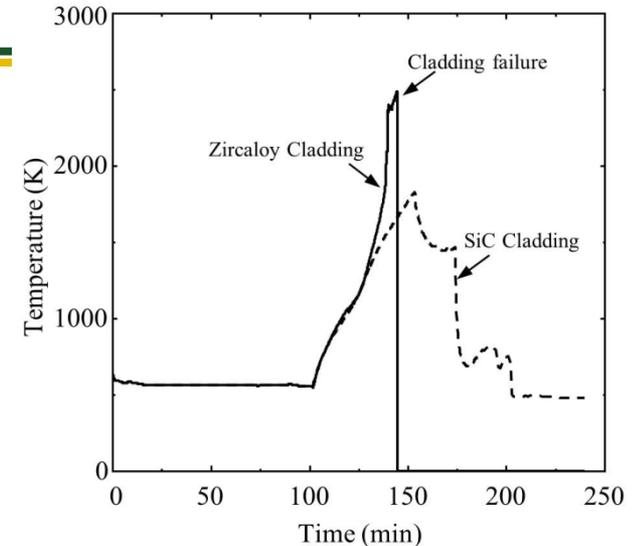
# MELCOR Severe Accident Analysis for SiC

## ■ Comparison of MELCOR predicted maximum cladding temperature during a TMI-2 accident

- Water level drops below active fuel height 100 m into accident
- At 125 m oxidation heating in Zr clad system accelerates clad T rise
- Zr clad failure at ~145 m – loss of cladding and core geometry (failure criterion is a T set point)
- SiC clad reaches peak  $T_{\text{clad}}$  of 1830K, below  $T_{\text{melt}}$  of the silica scale (1873K) – MELCOR calculations with existing SiC data show that it would not have failed

## ■ MELCOR predicted clad oxidation heating

- Two orders of magnitude less for SiC than for Zircaloy
- @150 m Zr heating exceeds core decay heat by factor of 5
- SiC produced more than 10 times less explosive gas during the accident analysis than Zircaloy

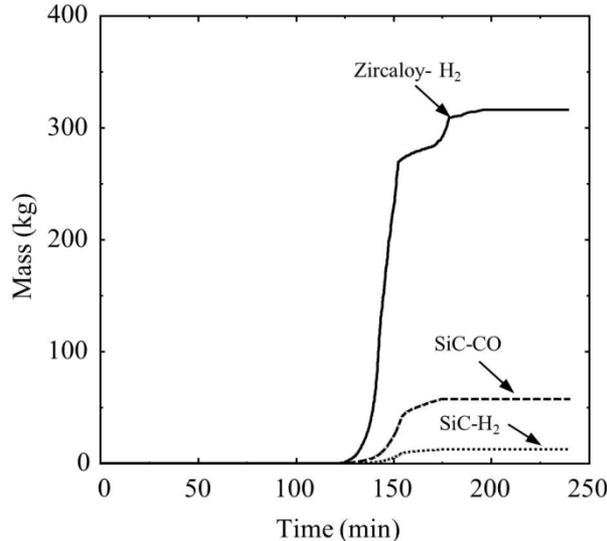




# MELCOR Severe Accident Analysis for SiC

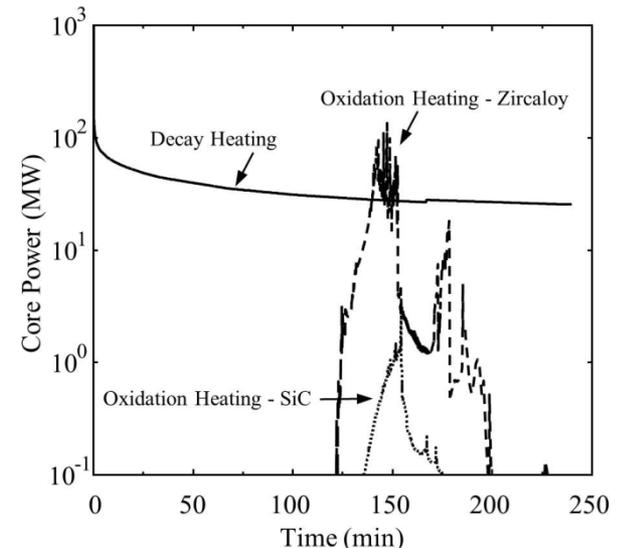
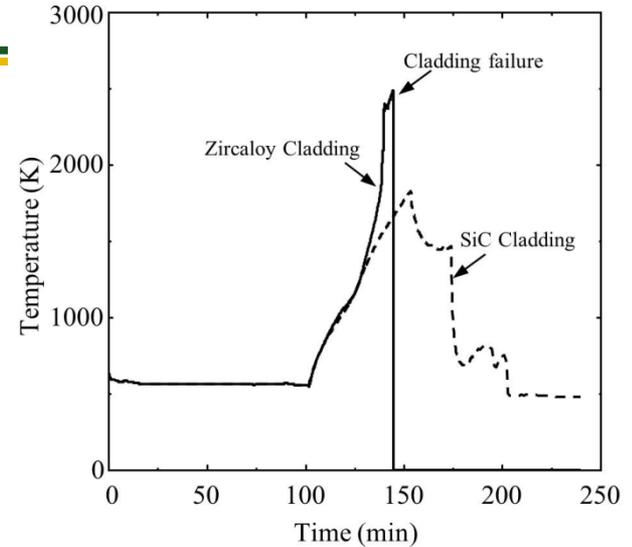
## ■ Comparison of MELCOR predicted maximum cladding temperature during a TMI-2 accident

- Water level drops below active fuel height 100 m into accident
- At 125 m oxidation heat clad T rise
- Zr clad failure at ~145 geometry (failure criterion)
- SiC clad reaches peak silica scale (1873K) – IAEA SiC data show that it w



## ■ MELCOR predicted cladding temperature

- Two orders of magnitude less for SiC than for Zircaloy
- @150 m Zr heating exceeds core decay heat by factor of 5
- SiC produced more than 10 times less explosive gas during the accident analysis than Zircaloy



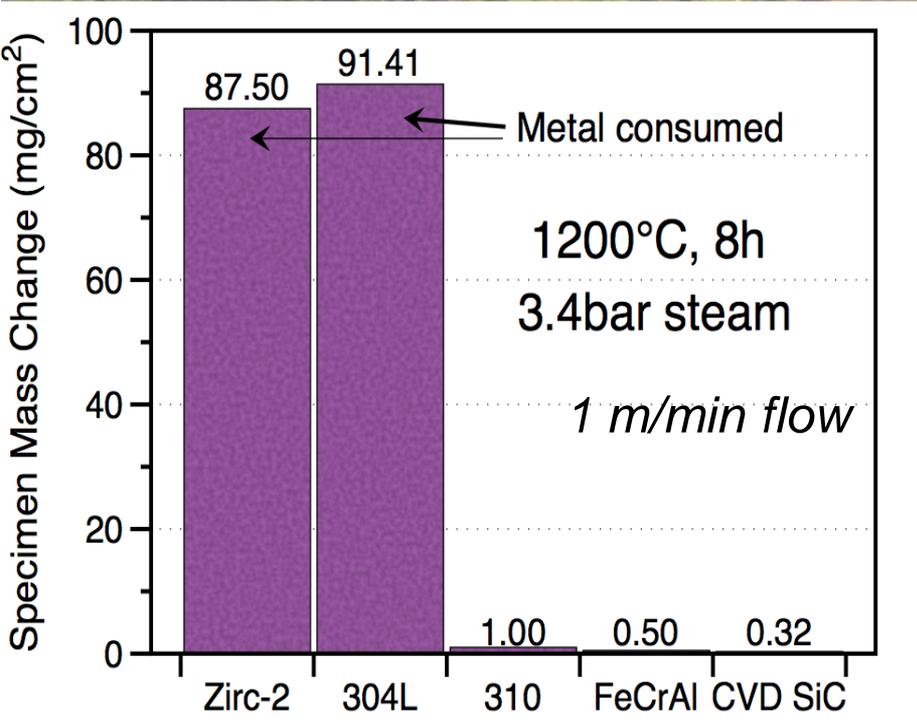
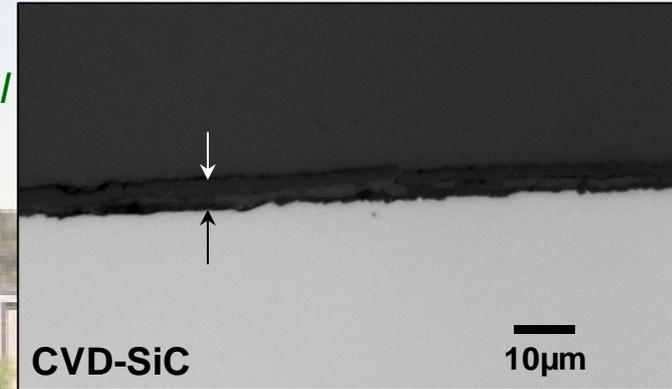
# ADVANCED CHARACTERIZATION OF SILICON CARBIDE COMPOSITES

# FCRD Advanced ATF Clad Development

initial screening – better than Zircaloy?



SiC: Minimal Reaction



Coated

As Fabricated Cross section

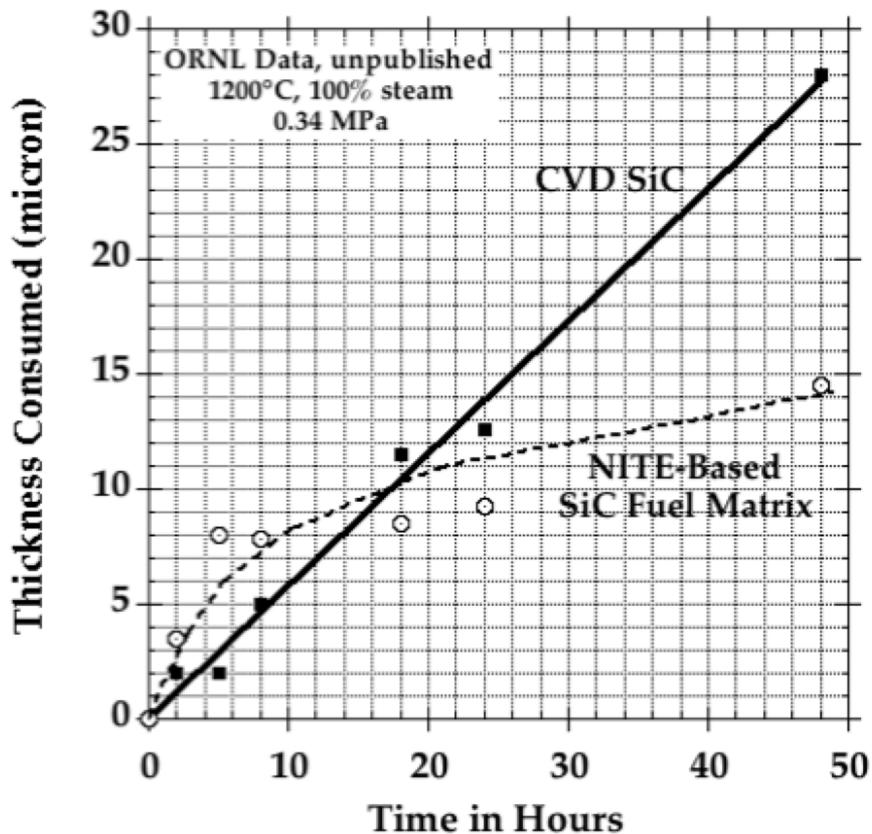


Uncoated

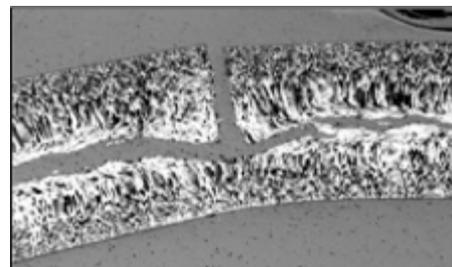
After 1300 °C and 250 psig of 100% steam for 48 hours



# Baseline Material Performance : SiC Based ATF Concepts 1200°C Screening

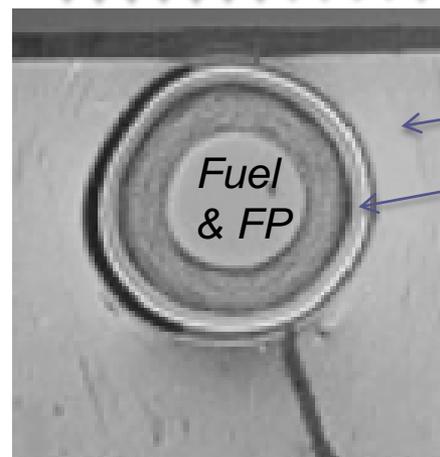


Zircaloy-4



↑ 1000 μm ↓

*Attack and Recession*



SiO<sub>2</sub>

SiC Matrix

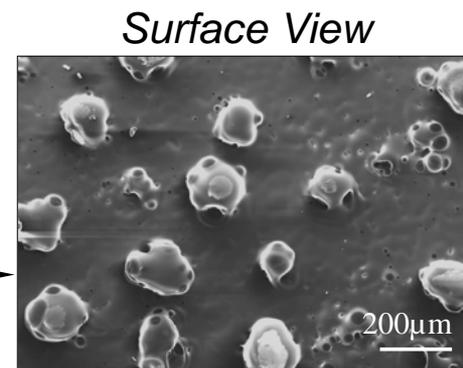
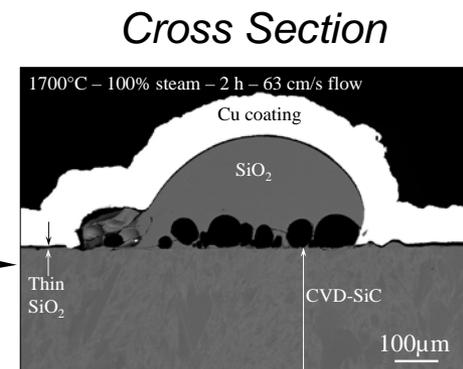
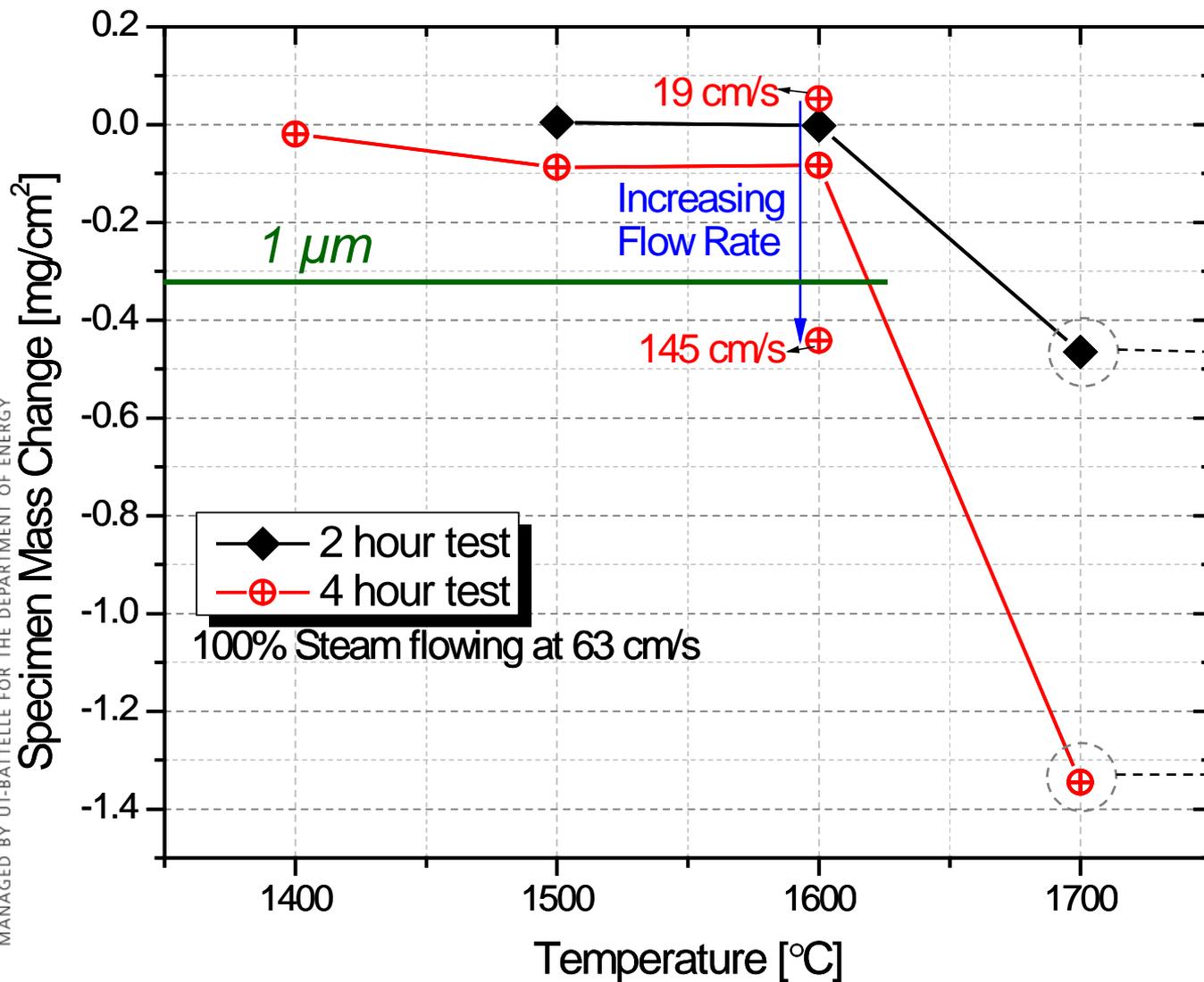
SiC Shell

Fuel & FP

*Under severe accident conditions breach of cladding leads to FP release only after SiC matrix recession and breach of SiC TRISO shell. At 1200°C recession rate ~ 10 μm/day.*



# Atmospheric Pressure Steam Tests on CVD-SiC to 1700°C

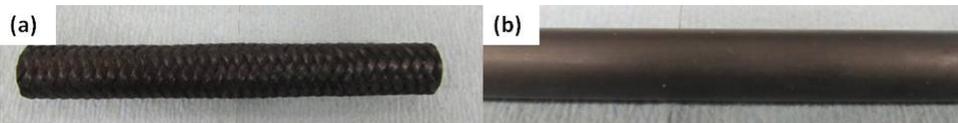




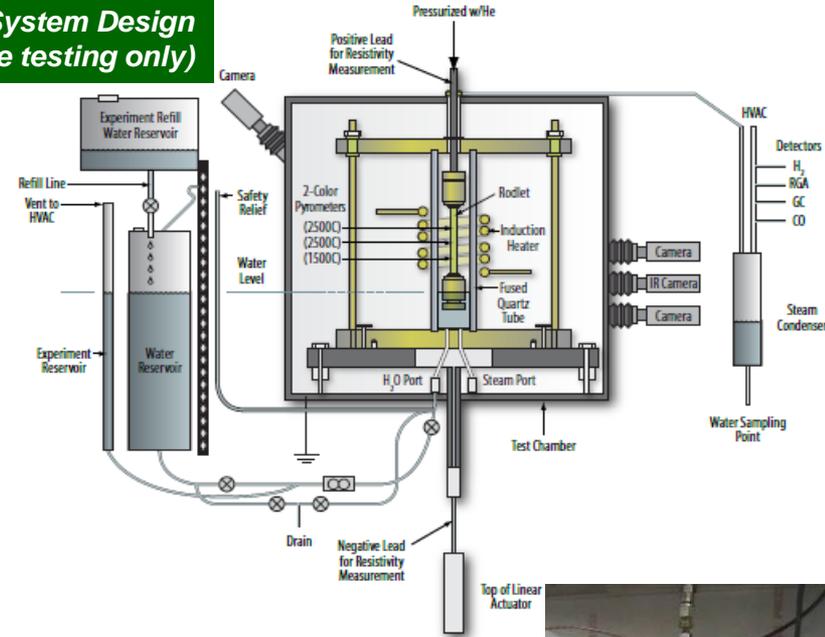
# Engineering Design and Safety Analysis: Oxidation Kinetics System (OKS @ INL) (LWRS)

## INL System Design (out of pile testing only)

- Out of pile / out of cell evaluation of advanced cladding materials under simulated LOCA conditions



SiC tube samples used in this study: (a) CMC sleeve formed from woven  $\beta$ -SiC fibers that was used to reinforce Zr-702 tubes during deformation testing; (b) monolithic  $\alpha$ -SiC tube.



Induction heating of a 2-ply SiC-CMC, Zr-702 hybrid sample.

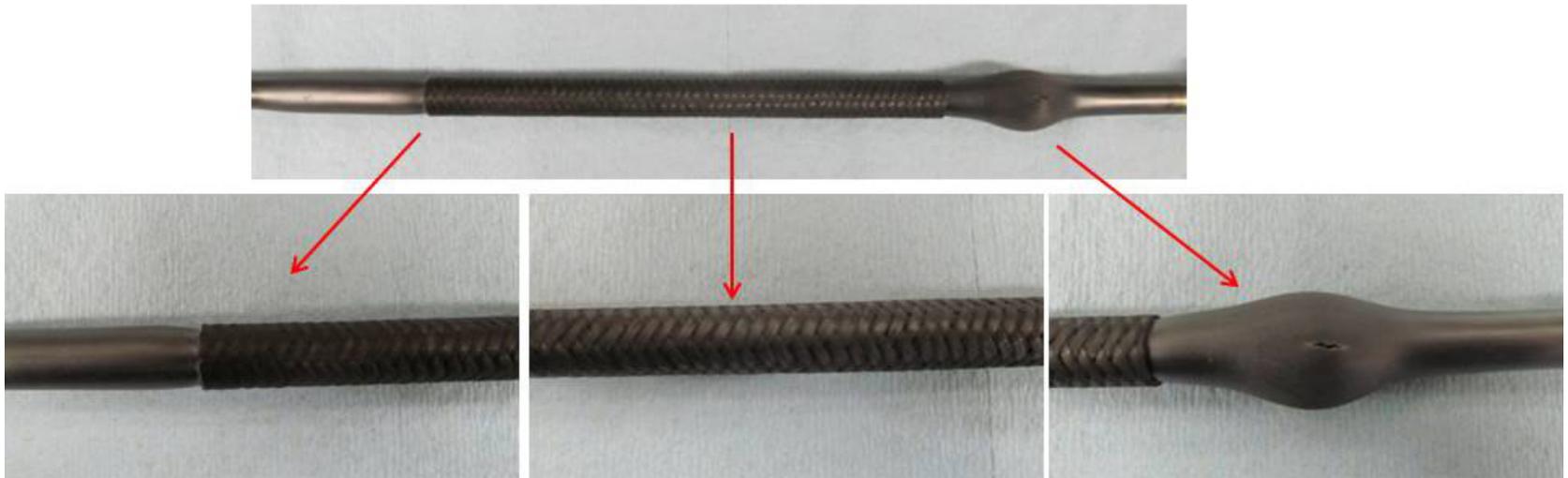


Partial immersion/refill quenching 09/25/2012



## Zr-702 tube w/SiC CMC (2-ply, 150mm) 500 psig He, 915 °C, steam (LWRS)

- SiC CMC sleeve shown to stabilize the Zr-702 tube along the length of the reinforcement



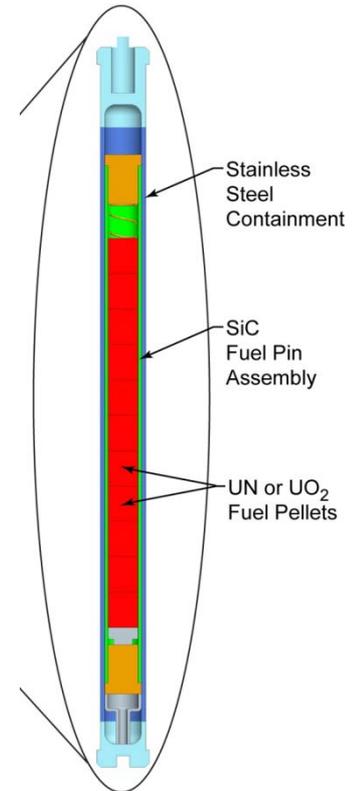
*Deformation profile of a Zr-702 tube reinforced with a 150 mm long × 11.3 mm OD SiC-CMC sleeve. Ballooning and rupture of the Zr-702 tube occurred outside the reinforced area near the end of the heated zone.*

***Results qualitatively indicate strength enhancement through the addition of a SiC CMC sleeve to an inner Zr-alloy tube. Method of “sealing” the SiC CMC to the Zr-tube requires refinement, and full-length SiC CMC sleeve has not yet been tested.***



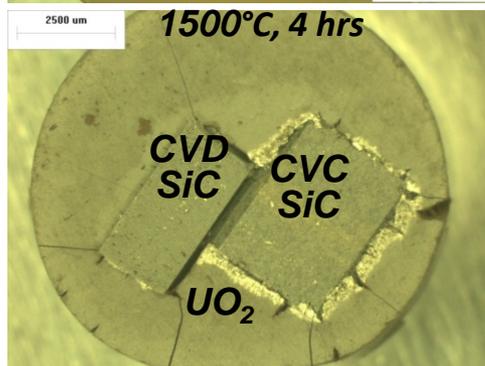
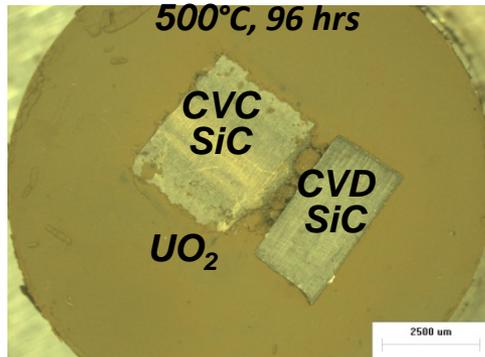
# SiC Clad Technology Development: HFIR Irradiation, PIE Status (LWRS)

- **TRIPLEX SiC / UO<sub>2</sub> capsules irradiated in HFIR**
  - 2 pins pulled: 10 GWd/MTU and 20 GWd/MTU
  - Shipped to hot cell for examination; both capsules visually examined
- **SiC/UN capsules were also irradiated, but there currently are no plans to perform PIE on these pins**
- **The PIE effort includes several key steps:**
  - Sectioning of capsules to remove outer encapsulation
  - Removal of SiC rods
  - Visual inspection of SiC rods
  - Gamma scanning
  - Puncturing and gas sampling
  - Sectioning and opening of SiC capsules
  - Metallography
  - Radiochemistry (based on time and funding available)
- **PIE is expected be completed by end of August (report December 2013)**



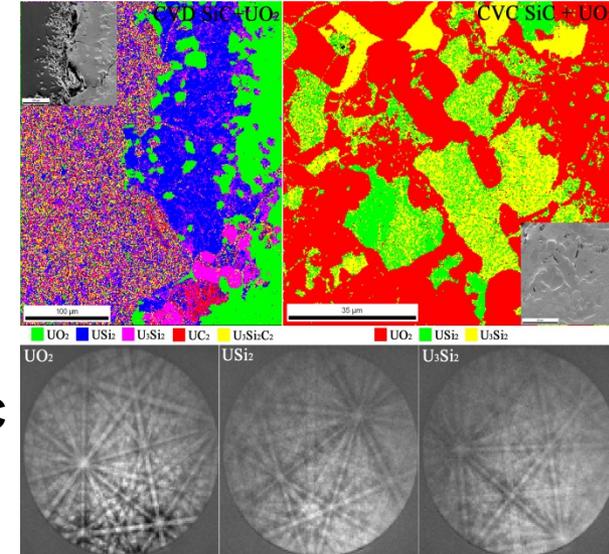


# SiC Clad Technology Development: Diffusion Couple Study (LWRS)



Optical micrographs indicating no SiC – UO<sub>2</sub> reaction at 500°C (top) and substantial reaction of UO<sub>2</sub> in particular with CVC SiC at 1500°C (bottom).

- Reactivity of high purity CVD SiC and “CVC” SiC (TREX proprietary) with UO<sub>2</sub> was experimentally examined.
- Substantial reactions were found after heat treatment at 1500°C for 4 hrs, whereas no reaction was confirmed at 500°C for 96 hrs.
- CVC SiC (inner layer of TRIPLEX clad) was found to be significantly more reactive with UO<sub>2</sub> at 1500°C.
- **Potential future work will include development of temperature-dependent reaction map and determination of reaction kinetics for high purity SiC – UO<sub>2</sub> system.**



EBSD Phase maps of CVD SiC – UO<sub>2</sub> and CVC SiC – UO<sub>2</sub> boundaries after heat treatment at 1500°C for 4 hrs. Checker flag region on left represents SiC.

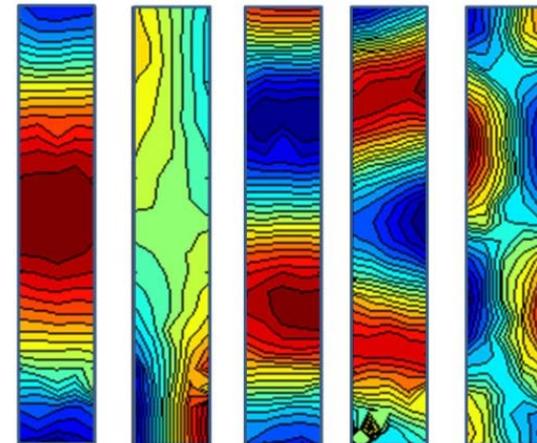
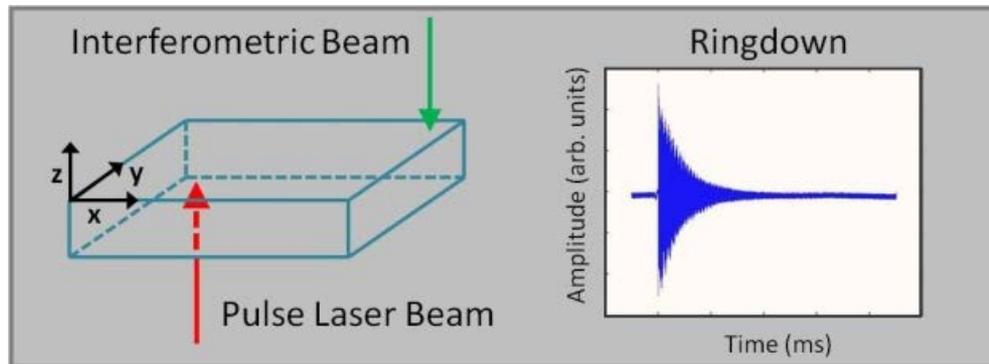
LWRS Level 3 Milestone (M3LW-13IN0502045): “Document out-of-pile testing to assess chemical interaction of fresh UO<sub>2</sub> fuel with candidate SiC clad materials”, July 1, 2013, work conducted at ORNL.



# Advanced SiC CMC Characterization: Elastic Constants (LWRS)

## ■ Measurement of elastic constants

- Accurate knowledge of elastic properties is critical to designing new CMC cladding materials with the appropriate dimensional tolerances
- Elastic properties of SiC-CMC are defined in large part by the specific fiber weave and the interaction of fibers with the matrix
- Fibers introduce directionality to the CMC causing the elastic properties to become direction dependent (elastic anisotropy)
- Dynamic elastic constants can be measured using ultrasonic techniques
- Testing to date has begun to measure the elastic properties of a candidate SiC-CMC material
- **Measurements must be performed on the specific CMC material (same fiber, weave, matrix, etc.) for the intended in-reactor application to ensure proper component design to meet dimensional tolerances**



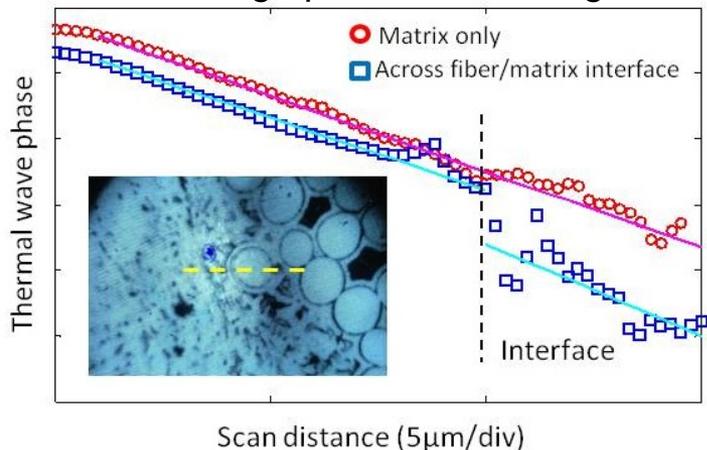


# Advanced SiC CMC Characterization: Thermal Properties (LWRS)

- **The composite matrix transports the majority of the thermal energy**
  - SiC fibers have a lower thermal conductivity than monolithic SiC
  - Possibility for considerable thermal contact resistance between the fiber and matrix further limits thermal transport
- **Understanding degradation of thermal conductivity in the matrix and the fiber is essential to understanding thermal transport in the composite material**
- **Initial evaluation: SiC-CMC composite composed of Nicalon Type S fibers**
  - Can extract thermal diffusivity and thermal contact resistance from data shown

*Thermal wave phase scan in matrix and across fiber/matrix interface.*

*Inset: micrograph of scanned region.*



## **Key Results / Conclusions:**

→ *Thermal contact resistance between the fiber and matrix may have a substantial contribution to a reduction in composite thermal conductivity*

→ *Understanding the change in contact resistance with thermal cycling and irradiation dose will help resolve the mechanisms that limit thermal transport in SiC-CMC materials*



# Failure Analysis for Wound Composite Ceramic Cladding Assembly (LWRS)

- **Purpose:** Testing and evaluation of microcracking and failure behavior of SiC CMCs to develop probability of failure estimates

- **Alpha-SiC Testing**

- Weibull statistics generated for 27 samples tested under internal pressurization conditions; associated fractography was performed to identify failure
- Internal pressurization results compared to those generated by diametral compression, axial tension, and flexure

Test Method	Characteristic Strength (MPa)	Weibull Modulus	Associated Flaw Population Sampled
Axial Tensile	320	10.0	Volume
Internal Pressurization	Original Estimate	5.3	Both Volume and Surface
	Revised Estimates	137	Both Volume and Surface
		108	Surface
		146	Volume
Diametral Compression	253	31.7	Surface
C-Ring Flexure	299	4.6	Surface Perpendicular to Circumferential Direction
Sectored Flexure	192	5.5	Surface Parallel to Circumferential Direction

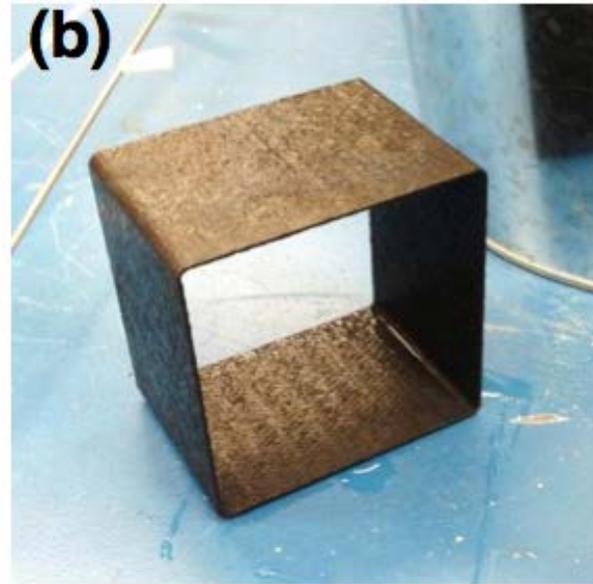
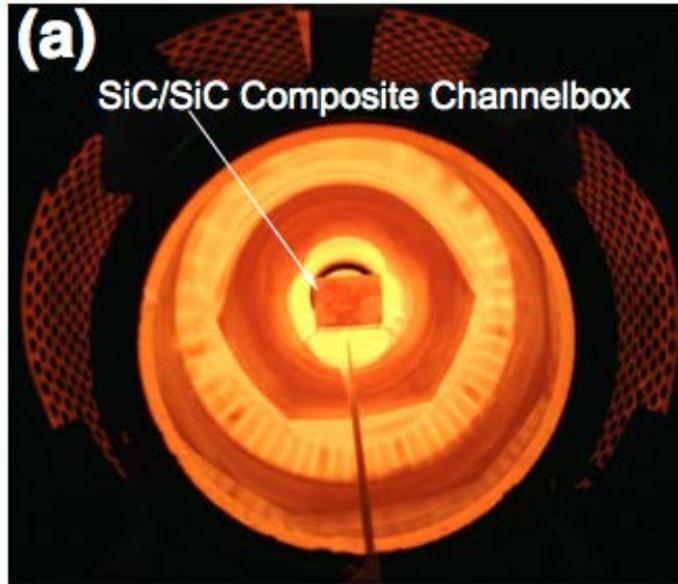
- **Beta-SiC Testing**

- Weibull statistics generated 10 ten samples tested under internal pressurization conditions; associated fractography was performed to identify failure
- Strength was much lower than that of the  $\alpha$ -SiC studied (26 MPa,  $m = 11.7$ ); failure was found to occur due to volume flaws

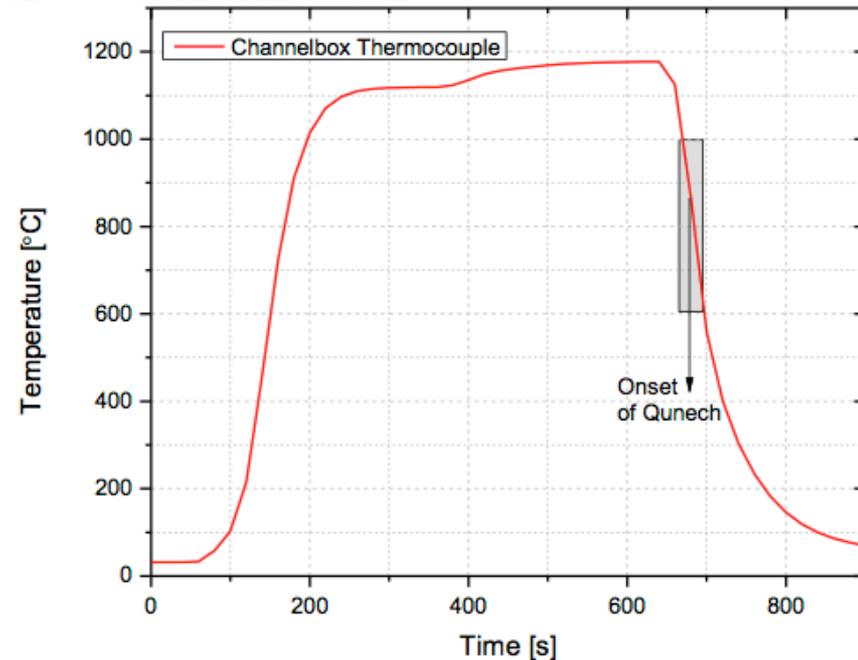
- **Composite Testing**

- Internal pressurization testing, single layer and double layer composite tubes
- Single layer tubes had an average strength of 138 MPa (stdev 3.3 MPa)
- Double layer tubes had an average strength of 172 MPa (stdev 52 MPa)

# EPRI Channel Box Quench Test



- Continuous fiber CVI SiC/SiC
- Tube furnace in air – water bath quench
- No evidence of chipping, gross micro-cracking or weight loss following quench
- Steam oxidation in 1200-1700°C range, 4 hr showed negligible material loss



Work of Ken Yueh-EPRI, Kurt Terrani-ORNL

# SILICON CARBIDE TECHNOLOGY DEVELOPMENT

# Development of ASTM Standards for Ceramic Matrix Composites in Nuclear Service

## ■ Goal

- Develop plan to codify critical standards for ceramic matrix composite (CMC) based LWR fuel cladding
- Establish full-consensus standards that are essential for development and qualification of CMC nuclear components

## ■ LWRS role:

- Supports test standards development in ASTM C28 on Advanced Ceramics in collaboration with other programs
- LWRS lead role in standards for hoop tensile properties and joint shear strength, while taking support roles in development of other standards
- Interacting with ASME design code development for nuclear CMC components

## ■ Status

- Draft standards to be submitted in Fall 2013 for first subcommittee balloting
- Receive comments; discuss at Jan 2014 subcommittee meeting



# Initial Preparations for SiC CMC Interlaboratory Round Robin Testing

## ■ SiC/SiC Composite Tubes for Tensile Inter-Laboratory Round Robin Study

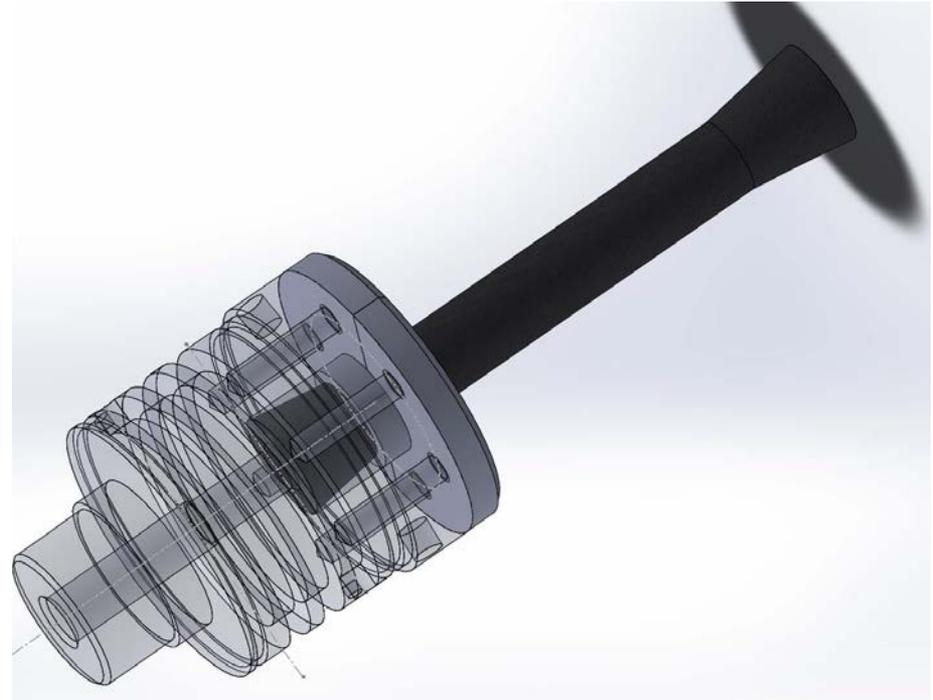


- Small diameter SiC/SiC composite tube test articles previously designed and fabricated were made available for the LWRS round robin study
- Tube specimens with tapered ends were fabricated with braided Hi-Nicalon and Hi-Nicalon Type S preforms and chemically vapor-infiltrated SiC matrices by Hyper-Therm High Temperature Composites (now Rolls-Royce High Temperature Composites)



# Preparation for Round Robin Study for ASTM C1773-13 Tube Axial Tensile Test Standard is in Progress

- Design of a self-aligning grip fixture is being finalized
- Dummy specimens (currently being prepared) will be used before starting tests with SiC/SiC test articles
- Lead test study at ORNL will establish the detailed standard test procedure to be adopted in an inter-laboratory test campaign
- Work beginning under LWRS; future funding undetermined





# Preliminary Plan for Round Robin Study on Tube Axial Tensile Test

Program Phase	Period	Task Items
Current	March 2014	<ul style="list-style-type: none"><li>• Complete inter-laboratory study plan including a preliminary list of participating teams.</li><li>• Establish detailed test procedure and practices through a lead test study at ORNL.</li></ul>
Follow-up Phase I	March 2015	<ul style="list-style-type: none"><li>• Prepare kits of test materials and instructions for distribution to participating teams.</li><li>• Organize the round robin testing process.</li><li>• Compile and analyze results.</li></ul>
Follow-up Phase II	March 2016	<ul style="list-style-type: none"><li>• Review and discuss Phase I results.</li><li>• Update C1773 with a section of Precision and Bias added.</li></ul>

# **SIC JOINING TECHNOLOGY DEVELOPMENT**

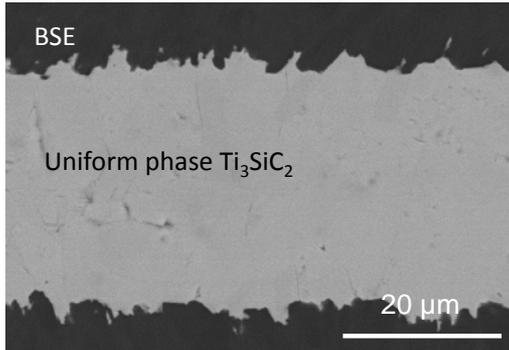
**Oak Ridge National Laboratory**

**General Atomics**

**Rolls-Royce High Temperature Composites**



# SiC Joining and Irradiation Studies at ORNL

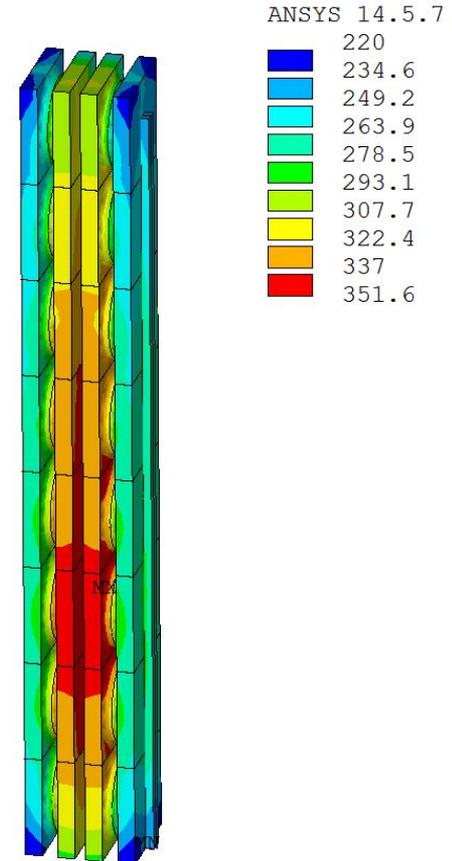


*Ti diffusion bonding showing uniform and intact bonding of two SiC substrates.*



*Miniature torsion specimens of Ti diffusion-bonded SiC joints*

- Miniature torsion specimens of SiC joints have been prepared for irradiation study following successful establishment of diffusion bonding technology with Ti or Mo active insert.
- Design and thermal analysis of rabbit vehicle are complete for neutron irradiation of these torsion specimens in a target region of High Flux Isotope Reactor.
- Progress on track toward proposed irradiation in FY-2014 (funding TBD)



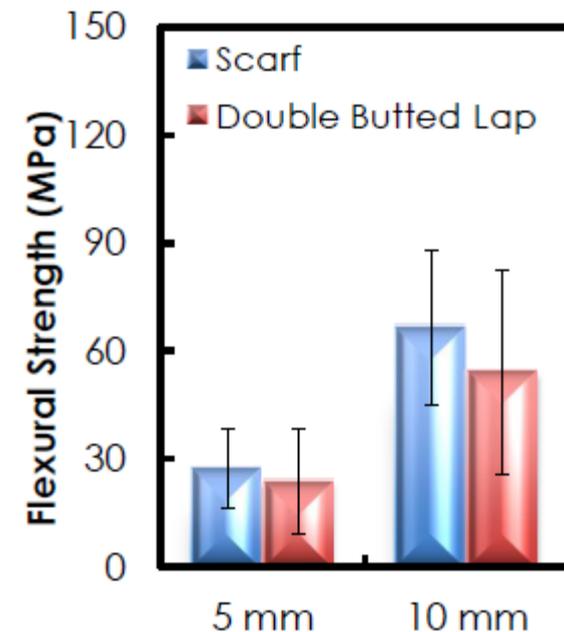
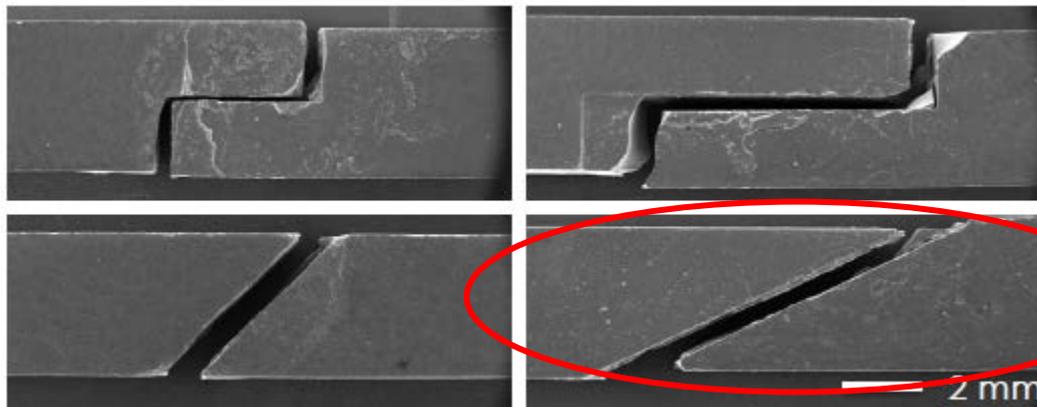
*Thermal analysis shows that temperature of the joint sections are maintained in a 290 – 320°C range.*

LWRS Level 3 Milestone (M3LW-13IN0504073): "Issue Report Summarizing SiC Joining & Irradiation Studies", July 31, 2013.



# SiC/SiC Joining General Atomics

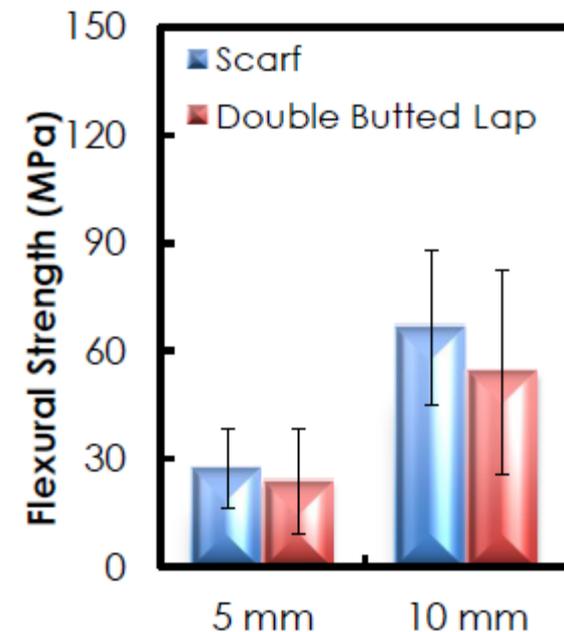
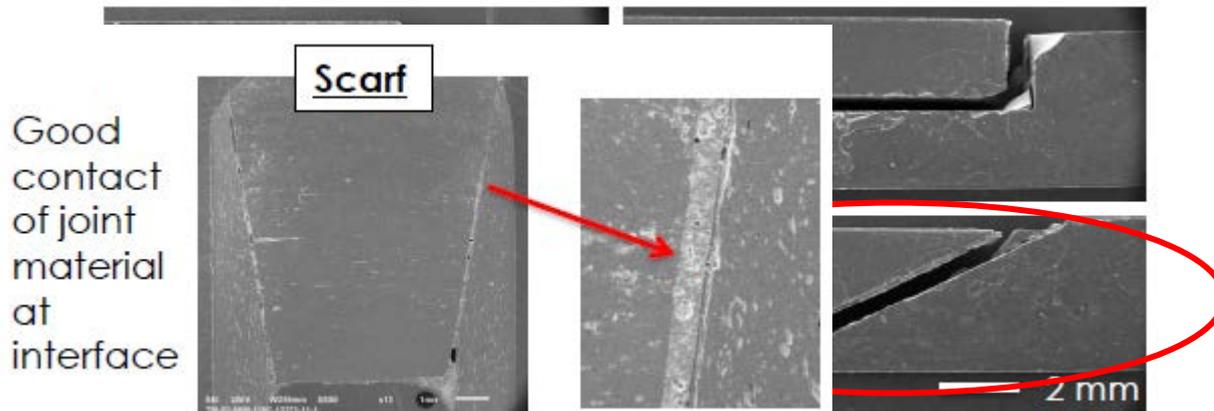
- Research to produce a mechanically robust and impermeable joint between an end plug and a SiC-SiC tube representative of an LWR SiC- SiC clad fuel rod for subsequent irradiation testing under conditions representative of an LWR core environment.
- Identification of the most successful and promising joint
  - Specifics of bonding materials and procedure proprietary
  - Mechanical strength and impermeability benefit from increased bond length
  - Quantified improvement in bond strength with bond length
    - 10 mm long scarf joint shown to be the most robust in four-point bend strength
  - Note that planar testing is insufficient for characterization of more complex cylindrical geometries





# SiC/SiC Joining General Atomics

- Research to produce a mechanically robust and impermeable joint between an end plug and a SiC-SiC tube representative of an LWR SiC- SiC clad fuel rod for subsequent irradiation testing under conditions representative of an LWR core environment.
- Identification of the most successful and promising joint
  - Specifics of bonding materials and procedure proprietary
  - Mechanical strength and impermeability benefit from increased bond length
  - Quantified improvement in bond strength with bond length
    - 10 mm long scarf joint shown to be the most robust in four-point bend strength
  - Note that planar testing is insufficient for characterization of more complex cylindrical geometries



### ■ Currently working on butted scarf joints

- Fabricated improved butted scarf endplug joint assemblies with monolithic SiC tubes
- Assemblies with varied joint surfaces were fabricated for endplug pushout testing to better understand critical parameters for good results
- Performed machining of composite tubes; required development of method for grinding to achieve desired scarf angles
- Measured permeability on butted scarf joints at 300 deg C
  - *The leak rate measured of  $\sim 5 \times 10^{-10}$  atm\*cc/sec was significantly better than scarf geometry ( $9 \times 10^{-7}$  atm\*cc/sec)*
  - *Improvement due to joint geometry AND improved technique for mating endplug and cladding tube*

### Expectations for August:

- Mechanical testing performed
- Fabrication of first composite tube joint assembly
  - High temperature thermal cycling on joined assemblies
- Deliver of samples to INL in September





# SiC/SiC Joining

## Rolls Royce High Temperature Composites

### ■ Scope:

- Demonstration of an impermeable joint using materials either known to be stable under the irradiation environment (SiC) or are currently planned for investigation due to their promise (MAX phase)
- Joining will be conducted at temperatures compatible with Nuclear Grade SiC/SiC composites reinforced with Tyranno SA or Hi-Nicalon Type S fibers

### ■ Status: CVI SiC densification of SiC/SiC fuel cladding segments for bonding is nearly complete

- Monolithic SiC tube with SiC fiber overbraid (Hi-Nicalon Type S fiber), then forming a nuclear grade SiC/SiC composite containing the nuclear grade SiC fiber
- Design is believed to combine the impermeability of a monolithic material with the toughness and non-catastrophic failure of a composite

*SiC/SiC fuel clad segments produced for bonding studies and pressure testing*





# SiC/SiC Joining Rolls Royce High Temperature Composites

- Two different braid architectures have been produced
- Design of fiber architecture for the pinned joint configuration has been completed
- Small diameter nuclear grade SiC/SiC pins will be produced from a triaxially braided architecture that has been shown to have failure strengths in excess of 700 MPa

## Near term plans:

- MAX phase and LPS SiC bonding of end caps will accelerate, including more mechanical testing
- Verification of the manufacturability and required tolerances for the pinned joint approach is planned



*Above: Lower end count braid architecture with greater bias angle on braid*

*Below: Higher end count braid architecture with lower bias angle on braid; has greater total axial reinforcement due to the higher end count*



# PATH FORWARD



## SiC Gaps – Recommended Measurements and Testing

- Further characterize **composite mechanical properties**, particularly following exposure to high temperature conditions, quench, etc.
  - Failure mode analysis: larger number of samples, composite samples to generate accurate statistics
- Additional characterization of **composite thermal properties**
- Additional characterization of **irradiation behavior of composite materials** (post-irradiation mechanical and thermal properties; irradiation creep behavior)
- Perform / complete **oxidation kinetics** testing of SiC composites
- Resolve **fabrication issues** associated with composite materials (reproducibility of thin-walled tubes; demonstrate strength and hermeticity of joint technologies; irradiation testing of complete assemblies)
- Complete development of **ASTM standards** and associated round robin testing for ceramic composite materials for nuclear service

**QUESTIONS?**