INL/MIS-21-61426-Revision-0

Understanding TRISO Coated Particle Neutron Irradiation Behavior:Evolution of Advanced Micro Analysis and Electron Microscopy Approaches

Changing the World's Energy Future

Isabella J Van Rooyen

Idaho National Laboratory

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February 2021

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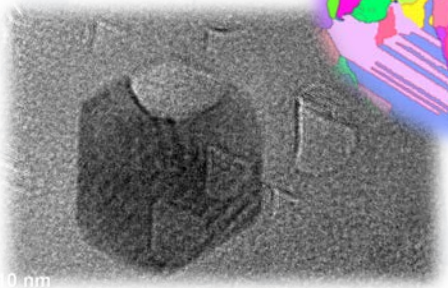
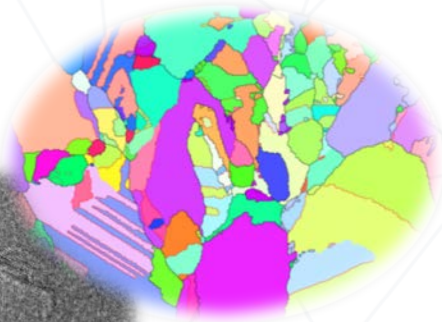
Isabella J van Rooyen
Distinguished Staff Scientist
January 25, 2021





ART
ADVANCED REACTOR TECHNOLOGIES

Advanced Microscopy for Nuclear Fuels and Materials; Virtual Workshop, January 25-27, 2021

Advanced Gas Reactor Fuel Development and Qualification Program

ADVANCED REACTOR TECHNOLOGIES

Objectives and motivation

- Provide data for fuel qualification in support of reactor licensing
- Establish a domestic commercial TRISO fuel fabrication capability

Reduce market entry risk

Submit AGR TRISO fuel performance, PIE, and safety test results in topical reports to NRC by 2025 for use in licensing TRISO-fueled advanced reactors.

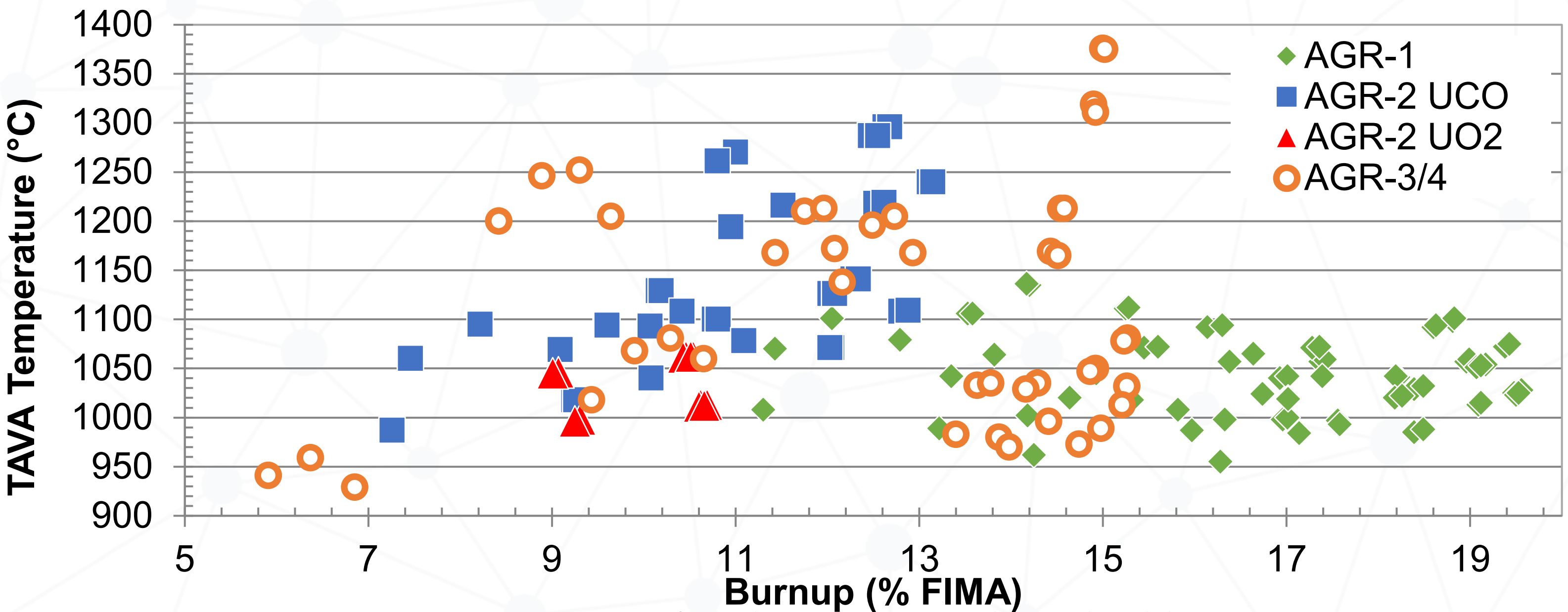
Approach

- Focus is on developing and testing **UCO** TRISO fuel
  - **Develop fuel fabrication and QC measurement methods**, first at lab scale and then at industrial scale
  - **Perform irradiation testing** over a range of conditions (burnup, temperature, fast neutron fluence)
  - **Perform post-irradiation examination and safety testing** to demonstrate and understand performance during irradiation and during accident conditions
  - **Develop fuel performance models** to better predict fuel behavior
  - **Perform fission product transport experiments** to improve understanding and refine models of fission product transport

[Paul Demkowicz]

AGR Irradiation Experiments

Experiment	Fuel Production Scale			Purpose
	Kernels	TRISO Coatings	Compacts	
AGR-1	Engineering	Laboratory	Laboratory	Shakedown of irradiation test train, baseline UCO fuel performance, enabled selection of TRISO coating Variant 3 for AGR-2.
AGR-2	Engineering	Engineering	Laboratory	Test performance of TRISO coatings produced at engineering scale. Compare UCO and UO_2 performance.
AGR-3/4	Engineering	Laboratory	Laboratory	Fission product transport measurements for UCO fuel graphitic matrix and nuclear graphite
AGR-5/6/7	Engineering	Engineering	Engineering	Fuel qualification and verification of UCO performance. Expands irradiation temperature range higher and lower.



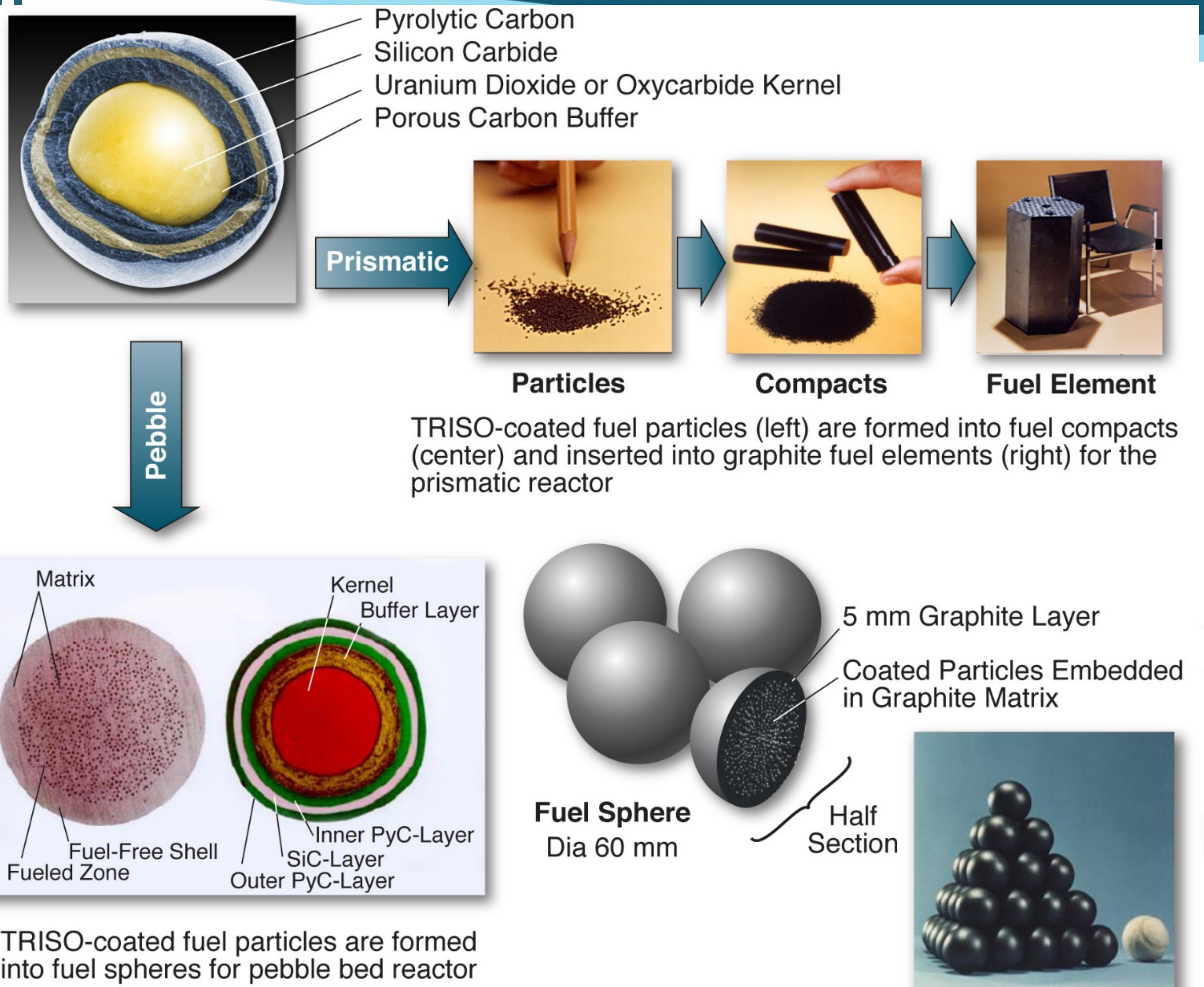
[John Stempien]

Introduction

ART
ADVANCED REACTOR TECHNOLOGIES

- Several advanced reactor designs incorporate tristructural isotropic (TRISO) fuel particles to achieve high coolant temperatures and increased thermodynamic efficiency
- TRISO coatings are deposited on the fuel kernel by chemical vapor deposition (CVD)
- Pyrocarbon and SiC layers retain fission gases. SiC layer is primary “pressure vessel” and retains condensable fission products (e.g. cesium, europium, strontium, etc.)

[TMS2019 Annual meeting, March 10-14, 2019, San Antonio, Texas, USA]



[T. Allen et al., Materials Today, 13 (2010) 14-23]

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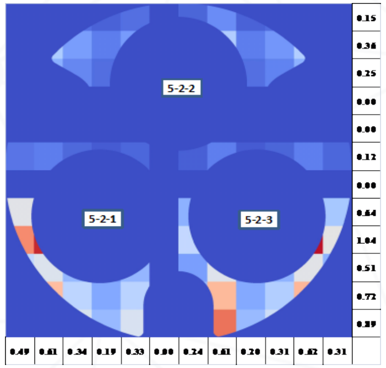
Studying Failed Particles: Understanding Fuel Performance

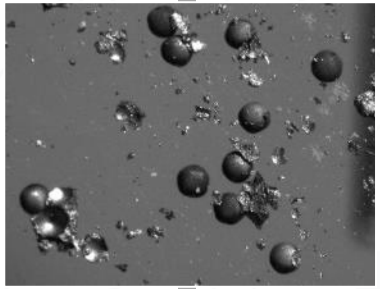
Identify compacts with leakers



Gamma scan to identify cesium hot spots and compact location

Identify particles with failed coatings

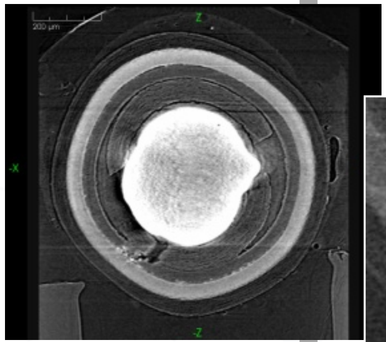
Gamma count to find particles with low cesium retention

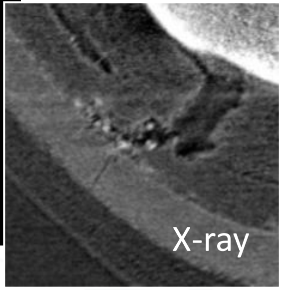


Deconsolidation to separate particles from compact

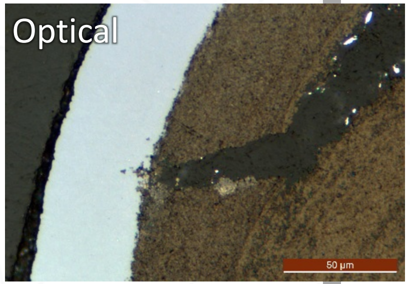
Study particles with failed coatings

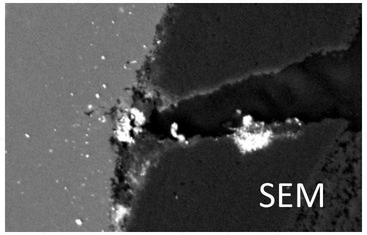
X-ray tomography to locate failures



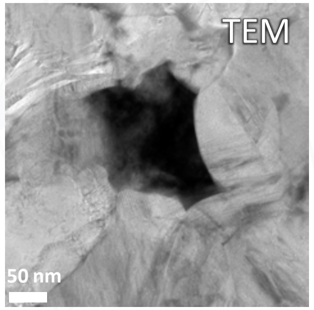


Materialography to expose defective region for analysis





Advanced microscopy to study coating layers in detail



[John Stempien]

Advanced Microscopy Objectives

OBJECTIVES

- Understanding Effects of Irradiation on TRISO layers
- Fission product chemistry and behavior in UCO kernel
- Identify and Understand Fission Product Transport Mechanisms in TRISO Coated Particles

OUTCOMES and IMPACT

- Impact on Performance
- Improve Predictive Behavior Modeling
- Kernel Behavior: Release from kernel; release from whole particle
- Known Fission Product Transport Mechanisms

Methods identified during AGR-1 Shakedown

2011-2017

- Initial work scope [PLN-2828] for AGR-1 includes basic SEM with elemental analysis EDS, WDS and EBSD
- The SEM examinations provided information on macro- and micro- level distribution of fission products
- Knowledge gaps which basic SEM could not provide:
  - Accurate quantification of elemental composition
  - Accurate distributions of elements in a specific precipitate
  - Phases of precipitates
  - Identification of Ag in these fission product precipitates
  - Fission product transport mechanisms through “intact” layers---specifically Ag and Pd
- Leads to exploration of more advanced electron microscopy and micro-analysis techniques

The deployment and adaption of advanced nano-scaled techniques set a benchmark for future studies.

[van Rooyen I. J., T. Lillo, H. Wen, K. Wright, J. Madden, J. Aguiar, 2017, Advanced Electron Microscopy and Micro analytical technique development and application for Irradiated TRISO Coated Particles from the AGR-1 Experiment, INL/EXT-15-36281, January 2017].

ART ADVANCED REACTOR TECHNOLOGIES

Advanced Microscopy and Micro-analysis Techniques

Fission Product Identification & Location

Microstructure
(Neutron Irradiation Damage (Voids, loops…))

FEI Tecnai G² F30 STEM at the Center for Advanced Energy Studies (CAES)

Decontamination in glove box

Electron probe micro-analyzer (EPMA)

Scanning Electron Microscopy (SEM)
- Main elements of precipitates
- Precipitate distribution
- Identify areas for TEM

Electron back scatter diffraction (EBSD)

Focused Ion Beam (FIB)
- Preparation for TEM, STEM, APT, HRTEM
- EBSD, TKD; ASTAR collection

Transmission Electron Microscopy (TEM)
- Micro-Nano sized imaging
- Analytical tools (EDS)
- 2 D imaging

Scanning Transmission Electron Microscopy (STEM)

Energy Dispersive Spectroscopy (EDS)
Energy Filtered TEM (EFTEM)
Electron Energy Loss Spectroscopy (EELS)

- Nano sized imaging
- Analytical tools
- 2 D imaging

Atom Probe (APT)
- 3D analytical tool

High Resolution Transmission Electron Microscopy (HRTEM)
- Atomic structure and imaging
- EELS higher resolution

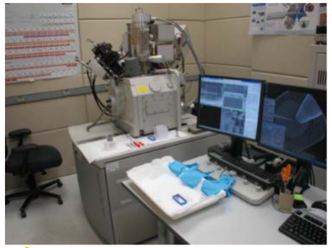
Transmission Kikuchi Diffraction (TKD)
- Crystallographic information

Precession Electron Diffraction (PED)
- Crystallographic information

Microstructure

Fission Product Transport

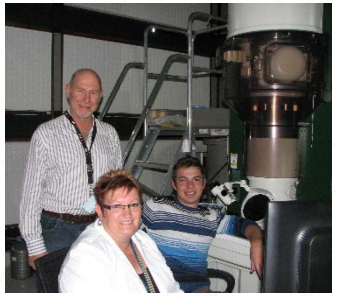
Grain boundary Characteristics



FEI Quanta 3D FEG Dualbeam FIB at EML at MFC

Preferred Methods after AGR-1 Shakedown

Specialized Applications

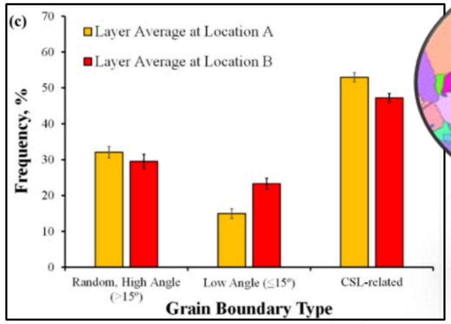


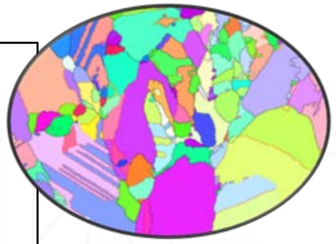
JEOL ARM 200F TEM two CEOS spherical aberration correctors

Cameca LEAP 4000XHR at CAES

Current Focus of TRISO Advanced Microscopy and Micro-Analysis

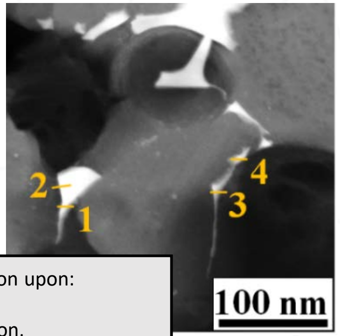
ADVANCED REACTOR TECHNOLOGIES





AGR-2 particle examination
Predominantly SiC, IPyC

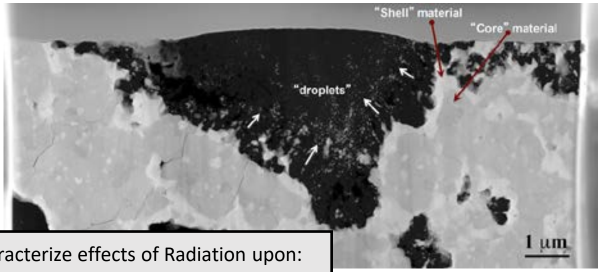
INL: Subhashish Meher
Karen Wright
Tom Lillo
Isabella van Rooyen
BSU: Yaqiao Wu



- Characterize effects of Radiation upon:
  - ❖kernel porosity
  - ❖layer degradation or corrosion,
  - ❖layer debonding,
  - ❖fission product precipitation,
  - ❖Microstructure; grain characteristics.
- Determine microstructural differences between particles exhibited high and low releases of Ag-110m

1

LANL: Terry Holesinger
UF: Yong Yang, Zhenyu Fu
INL: Boopathy Kombaiah
Mukesh Bachhav
Karen Wright
Isabella van Rooyen



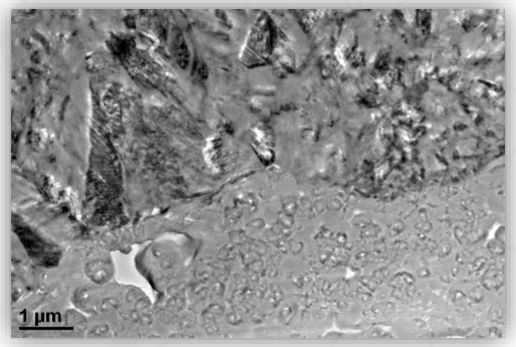
Kernel Examination

2

Characterize effects of Radiation upon:
- ❖kernel porosity
- ❖layer debonding,
- ❖fission product precipitation,
- ❖microstructure

3

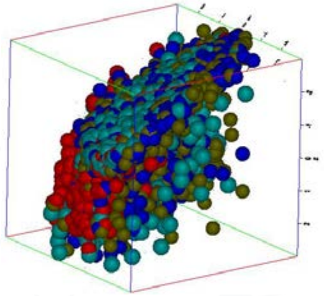
Neutron Irradiation Damage



INL:
Subhashish Meher
Tom Lillo
Jhonathan Rosales
Isabella van Rooyen

4

Fission Product Transport Mechanism White paper

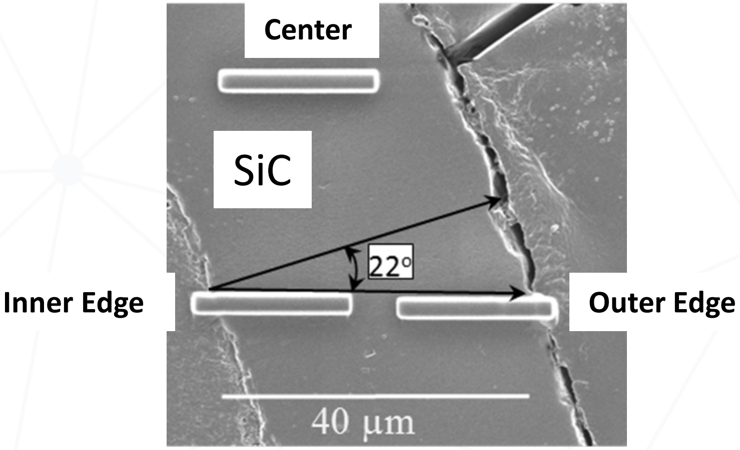


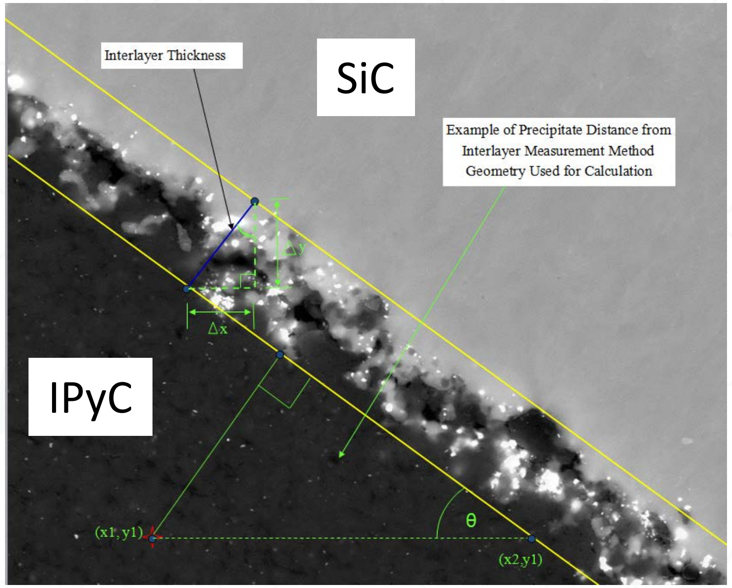
INL: Isabella van Rooyen
Paul Demkowicz
Paul Humrickhouse
William Skerjanc
Subhashish Meher
and others
Sandia: To be determined

ADVANCED REACTOR TECHNOLOGIES

1

SiC Layer Examination





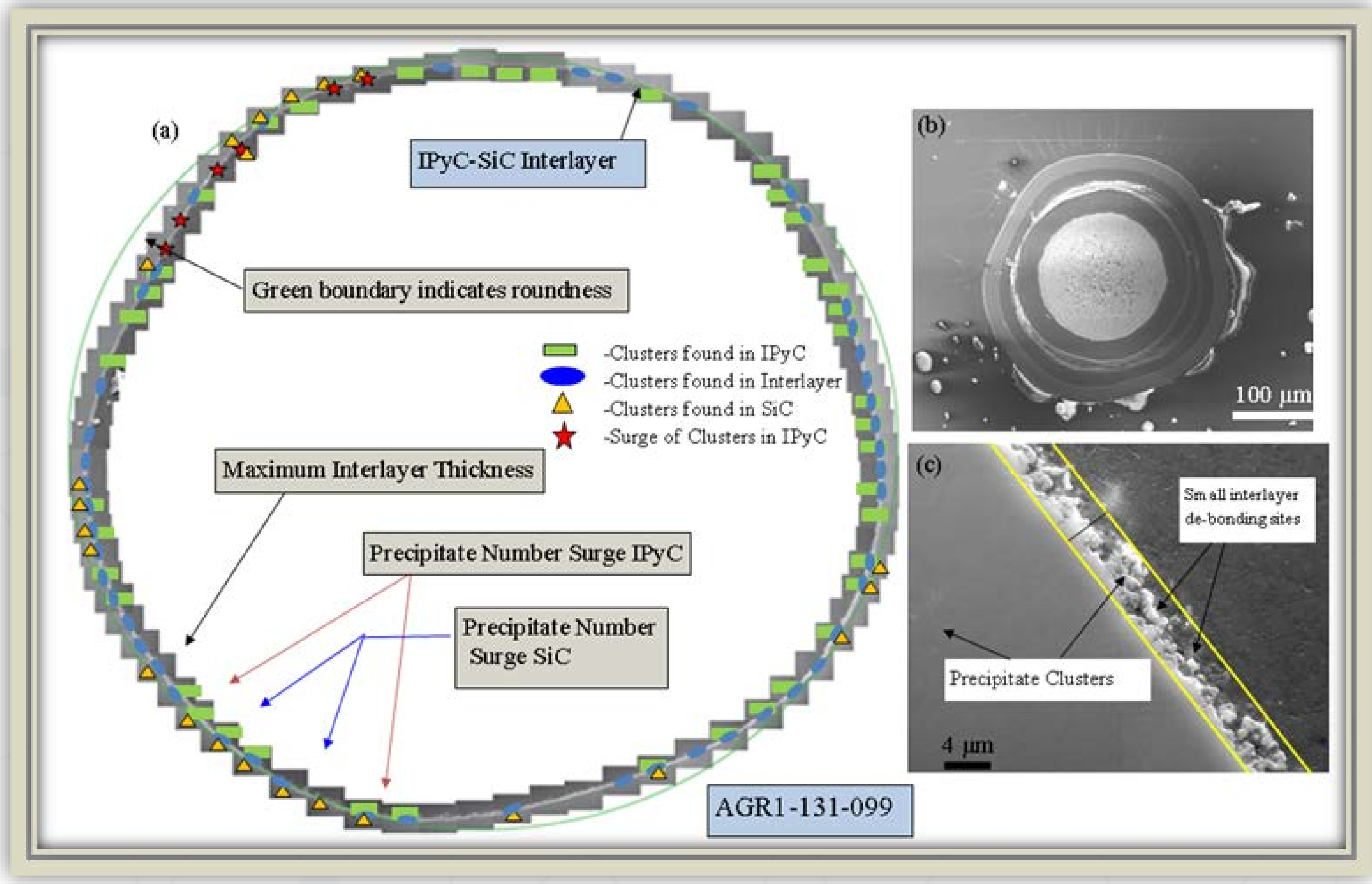
SEM SiC-IPyC Interface, Precipitate Distribution and Quantification

2011-2014

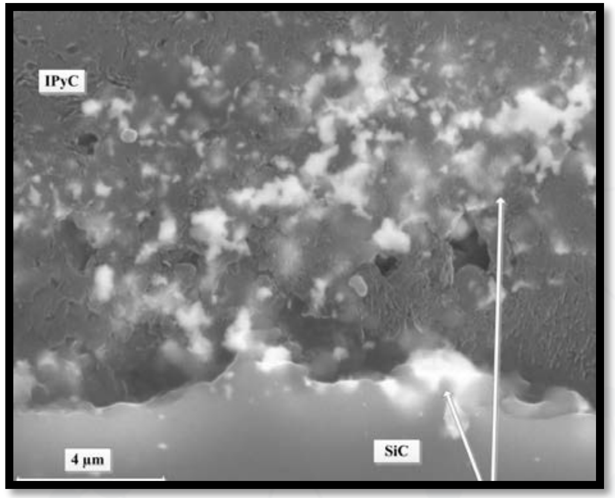
Phased Approach:

- JEOL 7000 FEG SEM: sequential secondary electron images of the inner-pyrolitic carbon (IPyC)-SiC interface at 4000x magnification
- ~81 micrographs per particle
- Identify Pd precipitates with energy-dispersive x-ray spectroscopy (EDS)
- Integrate in montage Adobe Photoshop
- Identify Pd precipitate clusters
- Measure depth of Pd precipitates in IPyC and SiC
- Measure interlayer thickness

[Ref. Van Rooyen et al., TMS 2015, 144th Annual Meeting, March 15-19, 2015; Walt Disney World Orlando, Florida, USA]



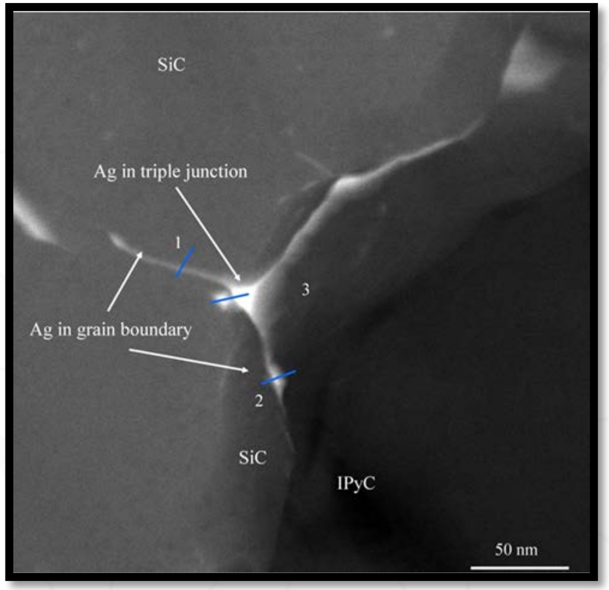
Precipitate Identification Evolution within SiC layer



[Van Rooyen, I. J., D. Janney, B. Miller, P. Demkowicz, J. Riesterer, “Electron microscopic evaluation and fission product identification in irradiated TRISO coated particles from the AGR-1 experiment: A preliminary review”, Nuclear Engineering and Design, 271 (2014) 114-122 (Also presented at the HTR2012 Tokyo 28 October-1 November 2012, paper HTR2012-3-023)]

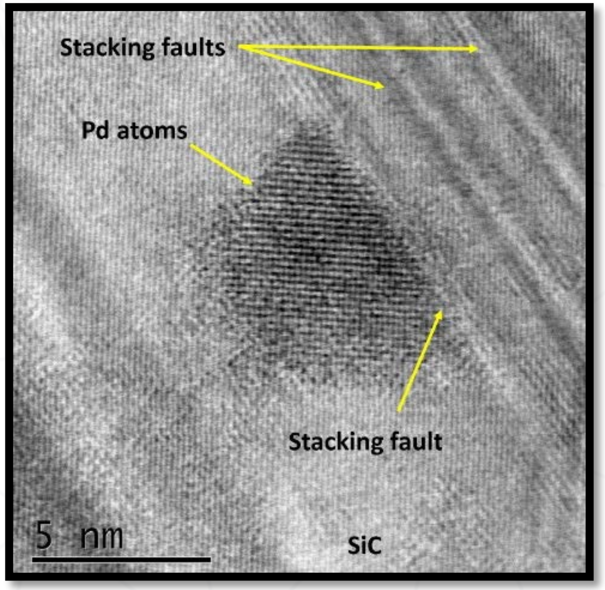
SEM 2012

First Ag identified



[Van Rooyen, I. J., Y. Q. Wu, and T. M. Lillo, “Identification of Silver and Palladium in Irradiated TRISO Coated Particles of the AGR1 Experiment,” Journal of Nuclear Materials 446 (2014) 178-186].

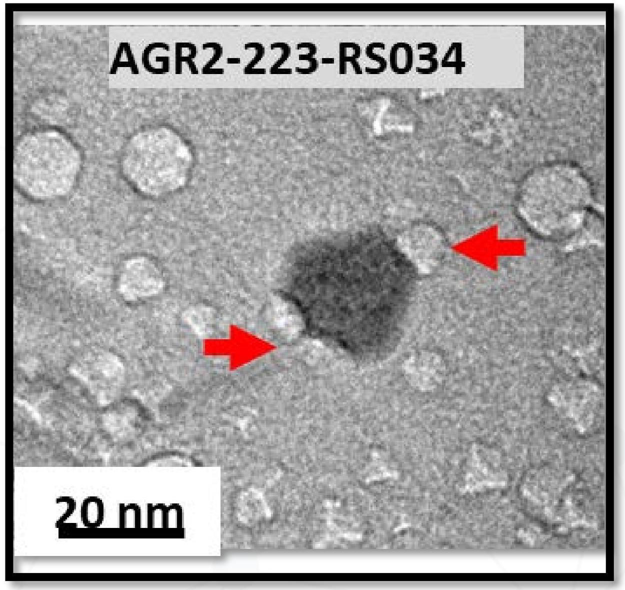
STEM 2013/2014



[Van Rooyen, I. J., E. J. Olivier and J. H Neethling, “Investigation of the Fission Products Silver, Palladium and Cadmium in Neutron Irradiated SiC using a Cs Corrected HRTEM“, Journal of Nuclear Materials, 476 (2016) 93 – 101]

[Olivier, E.J., J.H. Neethling and I.J. van Rooyen, Investigation of the structure and chemical nature of Pd fission product agglomerations in irradiated TRISO particle SiC, Journal of Nuclear Materials, https://doi.org/10.1016/j.jnucmat.2020.152043, Volume 532, 15 April 2020, 152043]

HRTEM 2014/2015/2016 & 2020



[Meher, S., I.J. van Rooyen, T.M. Lillo, A Novel Dual-Step Nucleation Pathway in Crystalline Solids under Neutron Irradiation, Scientific Reports (2018) 8:98 | DOI:10.1038/s41598-017-18548-8]

[Chao Jiang, Isabella J. van Rooyen, and Subhashish Meher, “Ab initio study and thermodynamic modeling of the Pd-Si-C system”, https://doi.org/10.1016/j.commatsci.2019.109238, Computational Materials Science, Volume 171, January 2020, 109238].

HRTEM 2018

ADVANCED REACTOR TECHNOLOGIES

AGR-1 Experiment: Precipitate Element Combination Summary

Particle	Precipitate Element Combinations in the SiC Layer		
	Inner Area	Center Area	Outer Area
AGR1-632-035 (High (79%) Ag retention)	Pd, Ag	Pd, Ag, Ce	Pd
	Pd-Ag, Pd-Pu, Pd-U, Pd-Ce	Pd-Ag, Pd-U, Pd-Ce,	Pd-Ag, Pd-Eu, Pd-Ce
	Pd-Ag-Cd, Pd-U-Pu, Pd-Ag-Cs-U		Pd-Ag-Ce, Pd-Ce-Eu
AGR1-531 - 038 (Low (< 19%) Ag retention)	Pd	Pd, Ag	Pd
	Pd-U, Pd-Pu, Pd-Ag	Pd-Ag, Pd-Pu, Pd-U, Pd-Ce	
	Pd-Cs-Eu, Pd-Ce-Eu, Pd-Cs-Ag, Pd-U-Pu, Pd-Ag-Eu	Pd-Ag-U, Pd-U-Pu, Pd-Ag-Pu	Pd-U-Pu
AGR1-531 - 031 (High (105%) Ag retention)	Pd, Cs, Pu, U, Ce,		
	Ag-U, Cs-U, Ce-U, U-Pu		
AGR1-131-066 (High (39%) Ag retention)	Pd, U	Pd	Pd
	Pd-Si, U-Si, Pd-U, Cs-U	Pd-Si, Pd-U	Pd-U, Pd-Ce
	Pd-Si-U	Pd-Si-U	Pd-Ce-U
AGR1-131-099 (Low (<6%) Ag retention)	Pd, U	Pd	Pd
	Pd-U, U-Cs, Pd-U	Pd-U	Pd-U
	Pd-U-Ce, Pd-U-Cs		
	Pd-U-Cs-Ce		
AGR1-433-001 (Low (66%) Ag retention)	Ag, Pd	Pd	Pd
	Ag-Cs, Pd-Ce, Pd-Ag, Pd-U, Ce-U, Pd-Pu	Pd-U,	Pd-Ag, Pd-Eu, Pd-Ce
	Pd-Ag-Ce, Pd-Ce-U, Pd-Ce-Pu, Pd-U-Pu, Pd-Ag-U	Pd-Ce-Eu, Pd-Cs-Pu, Pd-Ce-U	Pd-Eu-U
	Pd-Ce-U-Pu, Pd-Eu-U-Pu, Pd-Ce-Eu-U, Pd-U-Pu-Ce	Pd-Ce-Eu-Pu, Pd-Ag-Ce-Eu	Pd-Ag-Cs-Eu-U
		Pd-Ag-Cs-Eu-U	
		Pd-Ag-Ce-Eu-U-Pu	
AGR1-433-004 (High (98%) Ag retention)	Pd, Ag	Pd	Pd
	Pd-U, Pd-Ce, Pd-Ag, Pd-Eu	Pd-Ce	Pd-Ag
	Pd-Ce-Pu, Pd-U-Pu, Pd-Ag-Ce		Pd-Cs-Eu

[van Rooyen I. J., T. Lillo, H. Wen, K. Wright, J. Madden, J. Aguiar, 2017, Advanced Electron Microscopy and Micro analytical technique development and application for Irradiated TRISO Coated Particles from the AGR-1 Experiment, INL/EXT-15-36281, January 2017].

ADVANCED REACTOR TECHNOLOGIES

AGR-1 Experiment: Precipitate Element Combination Summary

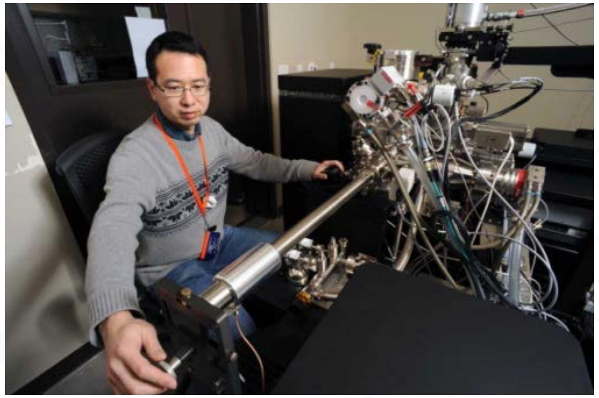
Particle	Precipitate Element Combinations in the SiC Layer		
	Inner Area	Center Area	Outer Area
AGR1-632-035 (High (79%) Ag retention)	Pd, Ag	Pd, Ag, Ce	Pd
	Pd-Ag, Pd-Pu, Pd-U, Pd-Ce	Pd-Ag, Pd-U, Pd-Ce,	Pd-Ag, Pd-Eu, Pd-Ce
	Pd-Ag-Cd, Pd-U-Pu, Pd-Ag-Cs-U		Pd-Ag-Ce, Pd-Ce-Eu
		Pd-Ag-Ce-Eu-U-Pu	
AGR1-433-004 (High (98%) Ag retention)	Pd, Ag	Pd	Pd
	Pd-U, Pd-Ce, Pd-Ag, Pd-Eu	Pd-Ce	Pd-Ag
	Pd-Ce-Pu, Pd-U-Pu, Pd-Ag-Ce		Pd-Cs-Eu

Combinations of elements in more than 700 precipitates that were examined:

- Complex and varying in nature
- More element combinations exist for precipitates from particles with relatively low Ag retention compared to particles with relatively high Ag-retention irrespective of fuel type.
- Cs present in particles from all compacts evaluated.
- Often other elements (e.g., Eu, Ce, Pu, and Cs) can be present in precipitates that predominantly contain Pd, Si, Ag, and U.
- U is predominantly found in combination with other elements and is only found alone in precipitates from Compact 1-3-1, which is a Variant 3 fuel compact.
- U and Ag are only found as a combination in the low Ag retention safety-tested particle AGR1-433-001.

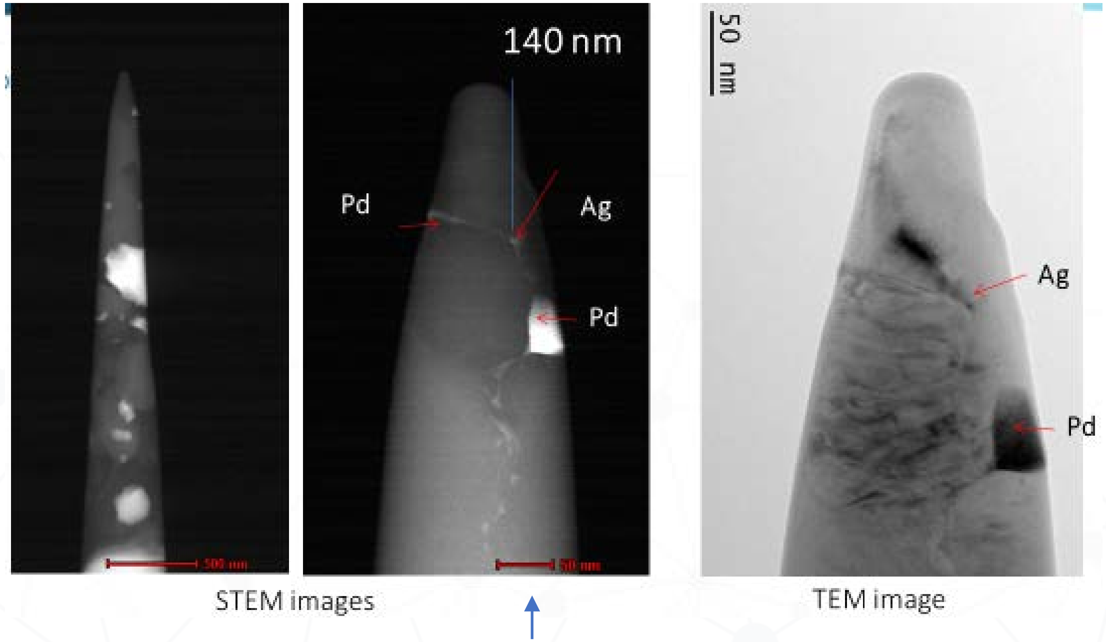
Challenges in Atom Probe Tomography

- Difficulties in preparing LEAP sample tips having features close enough to the tip tops
- Configuration of tips
- Irradiated SiC is more brittle due to neutron irradiation induced defects
- Interpretation: Element/molecule identification in mass-spectrum



(Cameca LEAP 4000XHR at the Center for Advanced Energy Studies (CAES))

FY2014/2015
Unirradiated and Irradiated SiC
NSUF-RTE Awards



The only tip contains a Ag-rich phase at a triple-junction about 140 nm below tip top. Other precipitates are found to be Pd-rich.

[NSUF User's week 2014, Idaho Falls, June 4-6, 2014]

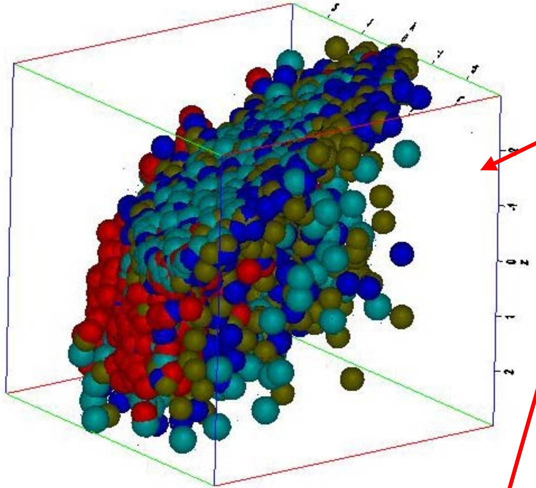
Non-homogeneous Fission Products Distribution

ADVANCED REACTOR TEC

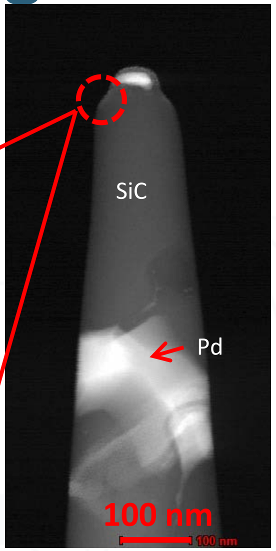
BOISE STATE UNIVERSITY

AGR1-632-034

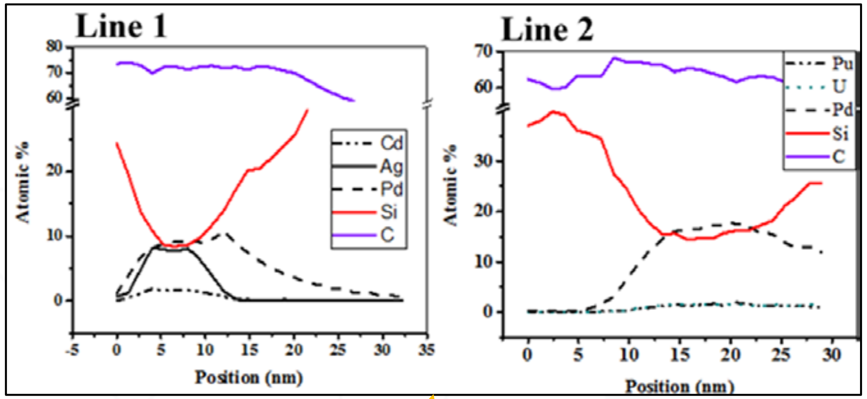
Volume size: 6 x 6 x 6 nm^3



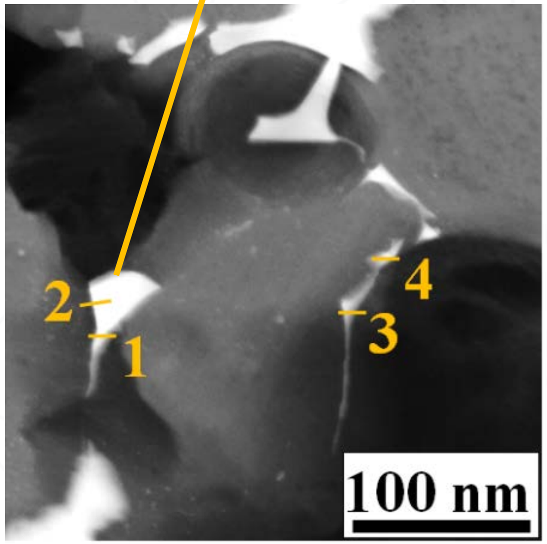
● Pd (9.0 at.%) ● U (2.5 at.%)
● Ag (2.7 at.%) ● Ru (0.4 at.%)

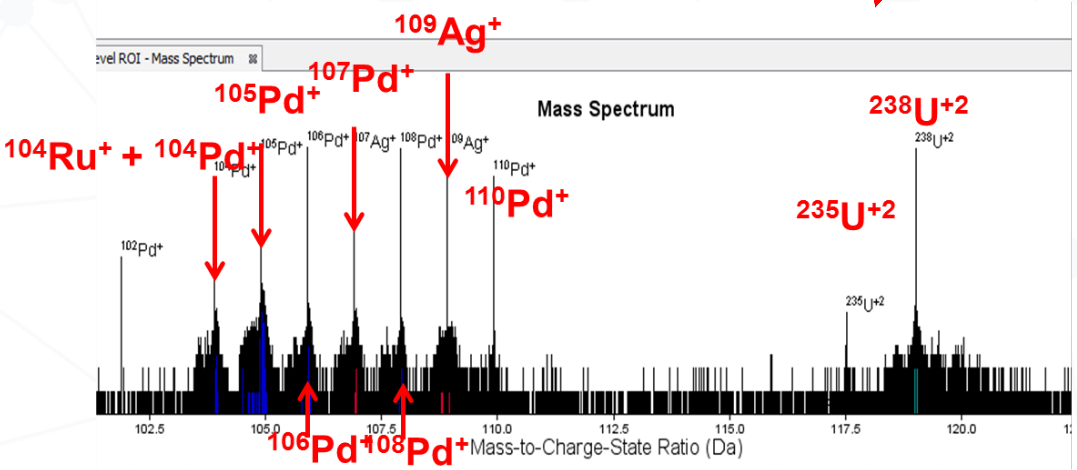


Irradiated SiC APT tip



AGR2-223-RS06





Indexed based on physics predictions of isotopic inventory

[NSUF-RTE-13-412 (partial) & NSUF-RTE-14-541]

[Advanced Gas Reactor Fuels Program Meeting, July 18-19, 2017, Idaho Falls, Idaho]

SiC Grain Boundary Characteristics (GBC)

FY2018-2021

PED: Program Funded

FY2013

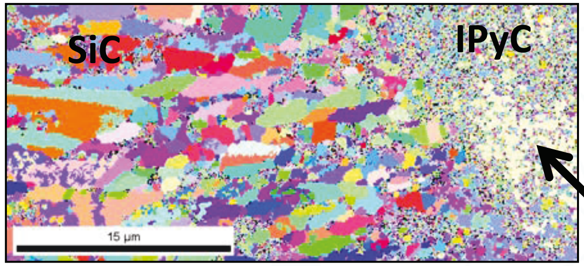
Conventional EBSD
Mounted sample
Micro examination
SEM

Transmission Kikuchi Diffraction
TEM lamella
Micro & Nano Examination
SEM

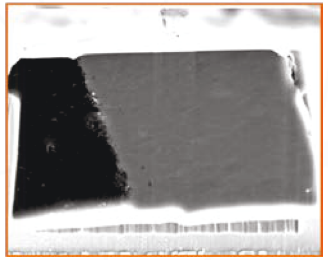
ASTAR PED
TEM lamella
Micro & Nano Examination
TEM

PED: NSUF Funded

FY2014



Irradiated



FIB preparation method developed as part of NSUF-RTE funded SiC Strength/EBSD project

Polished mounted TRISO particles
- Tested successfully on unirradiated SiC (various references)
- Not achieved yet on irradiated TRISO

FIB polished surface
- Tested successfully on unirradiated AGR SiC
- Not feasible on irradiated TRISO mounts (no cutting allowed, size limitation)

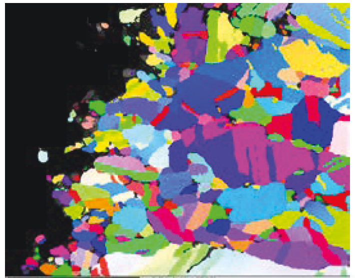
FIB prepared Thick Lamella
- Modified method tested on unirradiated SiC (NSUF project)
- Partially successful on irradiated AGR1 SiC (equipment limitations/ damage)

- Time delays between modification of surface prep
- Detectors out of operation
- Equipment Scheduling conflicts

FIB-TEM lamella
- Tested successfully on unirradiated SiC from AGR-1 TRISO(May 2013)
- Tested successfully on unirradiated & irradiated research SiC (June 2013)
- Tested successfully irradiated SiC from AGR-1 TRISO (**Sept 2013**)

- No cost collaboration (Brucker & NMMU)
- Can be used as micro- and nano measurement
- Resolution ~ 10 nm

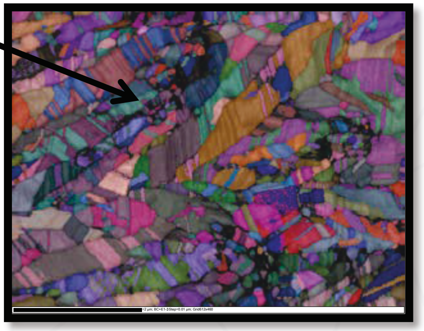
Irradiated

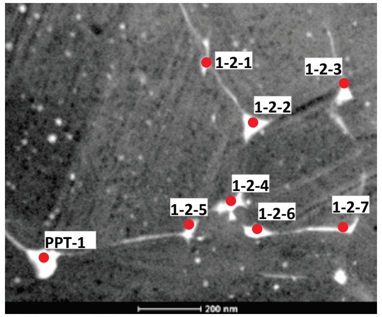


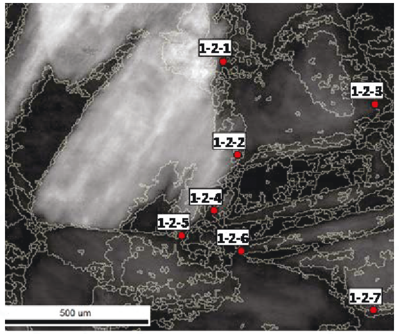
FIB-TEM lamella
- Not tested yet, no funding
- Boise State University research SiC (Sept 2013)
- INL-LDRD funding proposal
- Sept NSUF funding proposal with BSU

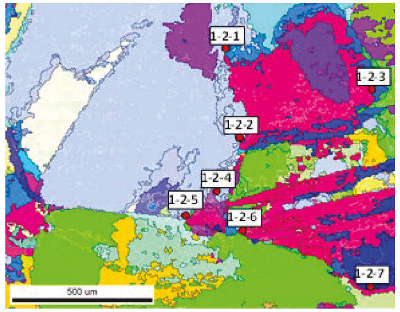
- Very advantageous
- Resolution ~ 1 to 2 nm (TEM mode)

Unirradiated

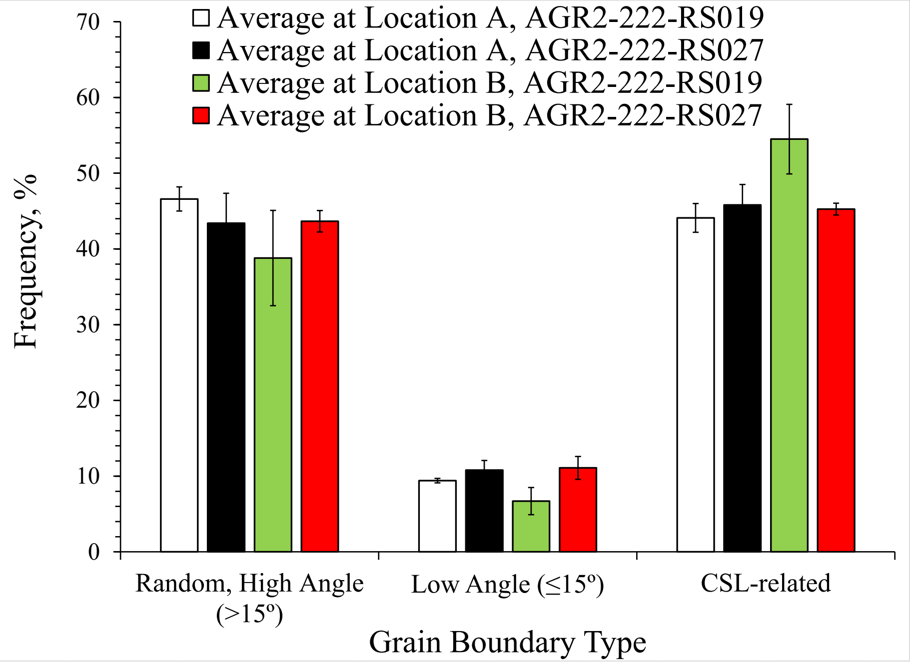


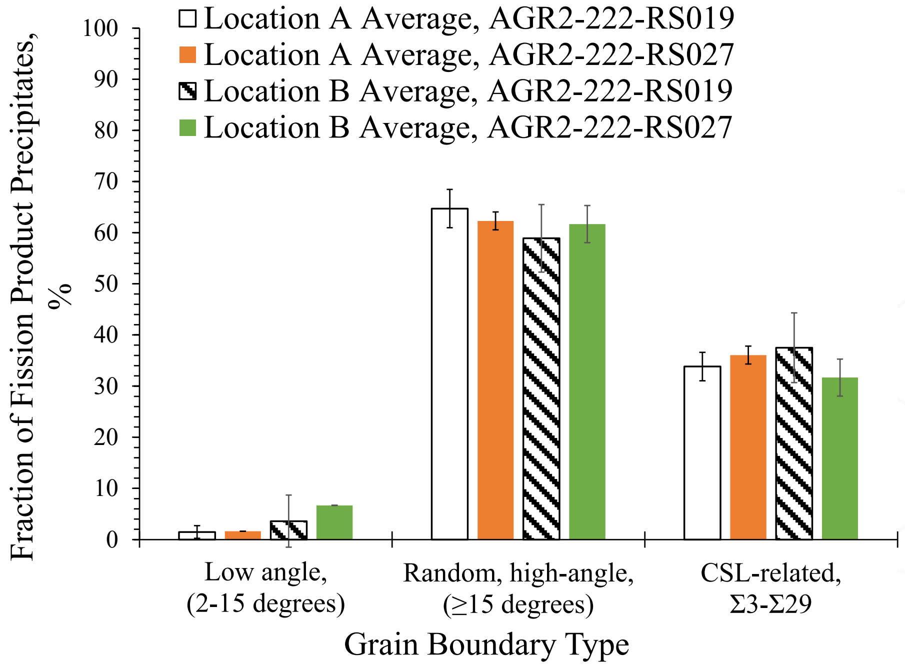






SiC layer Examination: Grain boundary nature & grain boundary precipitation





- The grain boundary distribution nature in both AGR-2 particles are very similar
- As expected, most of the fission product precipitates are observed at **high angle grain boundaries**

[Advanced Reactor Technologies Gas-Cooled Reactor Program Review, July 14-16, 2020, GoToWebinar]

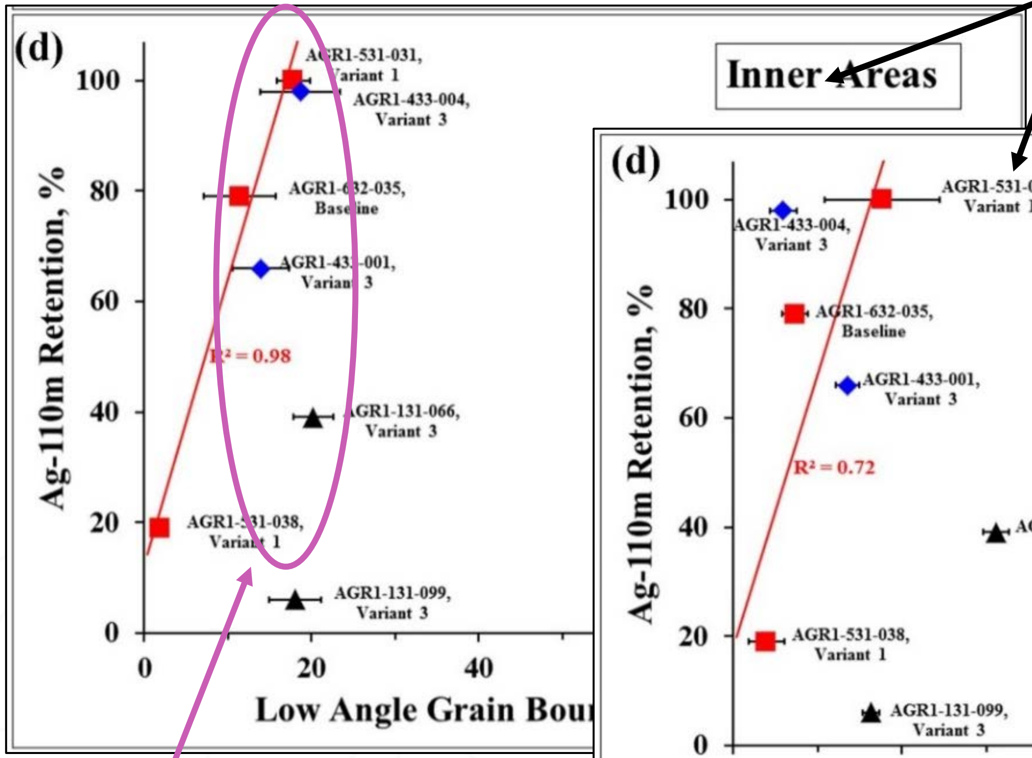
ADVANCED REACTOR TECHNOLOGIES

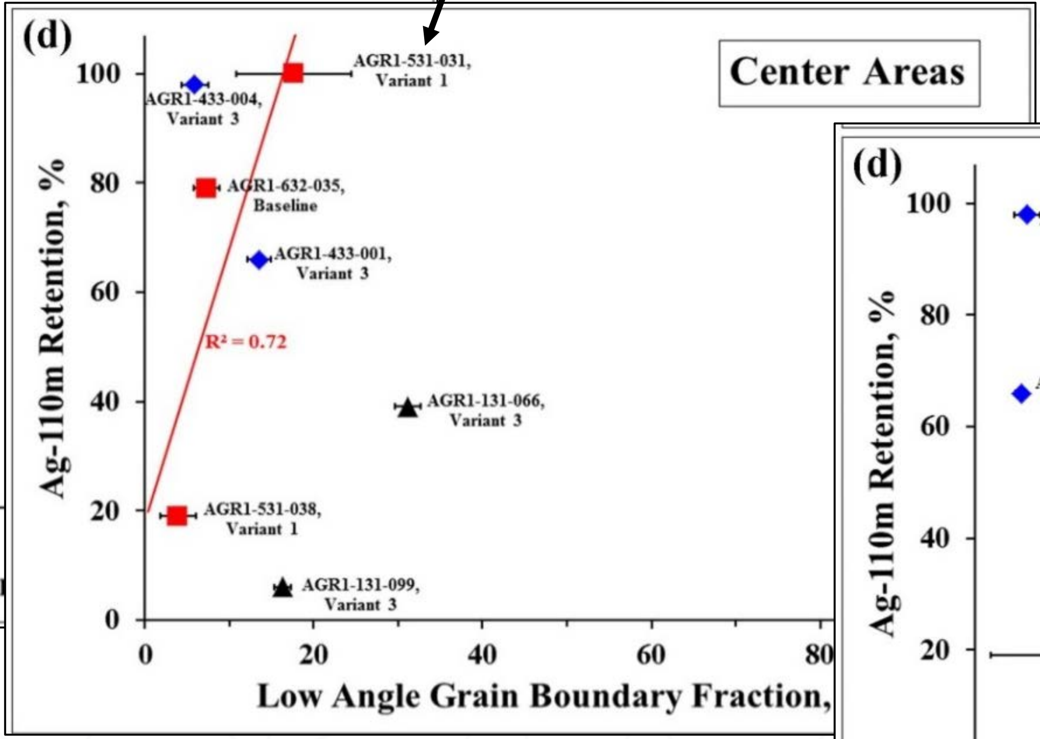
AGR-1 GBC: Low Angle Boundary

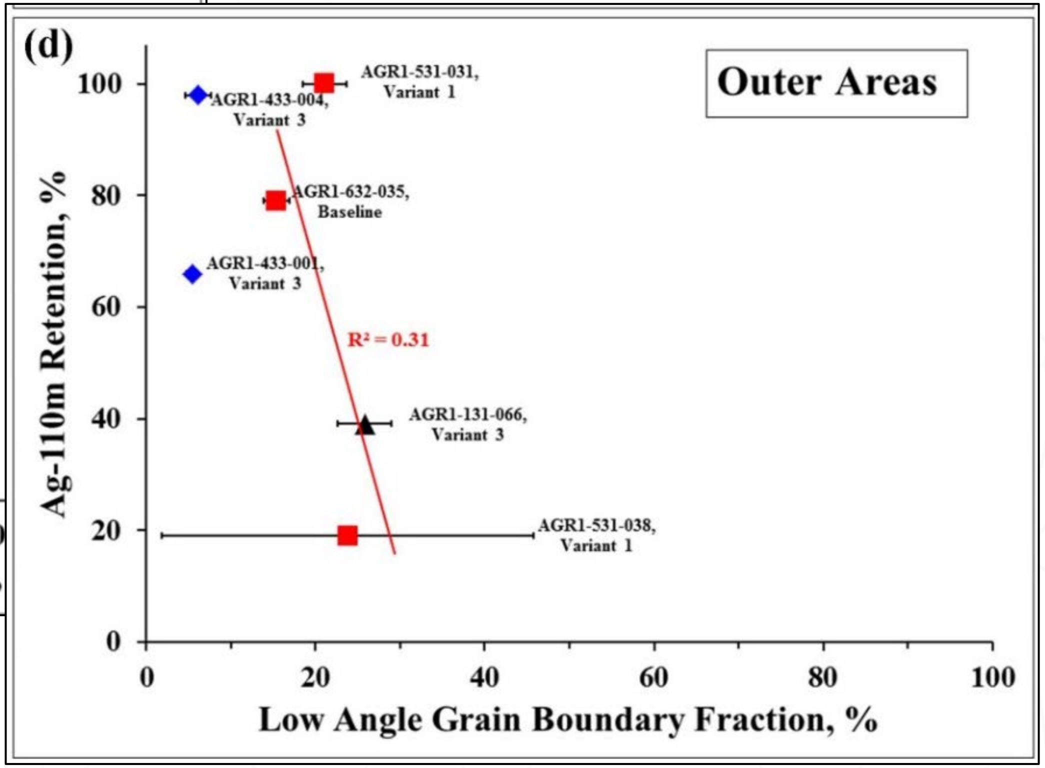
Good + correlation between Ag-110m retention and low-angle grain boundary fraction.

EXPECT
> low angle boundaries = aid Ag retention

[Advanced Gas Reactor Fuels Program Meeting, July 18-19, 2017, Idaho Falls, Idaho]







Variant 3 particles: narrow band ~ 20% low angle boundaries

Combining datasets from inner, center, and outer regions: weaken correlations between Ag-110m and various grain boundary parameters.

- Relook at increments chosen to determine if groupings consistent and relevant

ADVANCED REACTOR TECHNOLOGIES

2

UCO Kernel Examination

ART ADVANCED REACTOR TECHNOLOGIES

Kernel and Kernel-Buffer interlayer: Scope & Matrix

Microstructure

Fission Product Identification, Quantification & Location

Why?

- 1: effect of fuel type and burnup
- 2: effect of lab scale vs pilot scale (approx. same burnup)
- 3: comparison of AGR-1 vs AGR-2 safety tested kernel
- 4: Effect of Ag retention
- 5: lowest and highest radiation level particles available (approx. same Ag retention)

Other relationships can still be explored

Particle	Ag retention	Fuel Type	Burnup (%FIMA)
AGR1-632-034	65%	AGR-1 Baseline	11.4
AGR1-523-SP01	16%	AGR-1 Variant 1	17.4
AGR1-131-066	39%	AGR-1 Variant 3	15.3
AGR1-433-001 Safety tested	66%	AGR-1 Variant 3	18.6 & Safety tested
AGR2-223-R06	8%	AGR-2	10.8
AGR2-222-RS36 Safety tested	80%	AGR-2	12.55 & Safety tested
AGR2-633-RS28	<21%	AGR-2	7.5
☆ AGR2-633-RS09	88%	AGR-2	7.5
☆ AGR1-411-030	90%	AGR-1 Variant 3	19.4

Future work ☆

Completed

LANL completed

Impact release from kernel & therefore release from whole particle

UCO Examination

AGR2-222-RS036

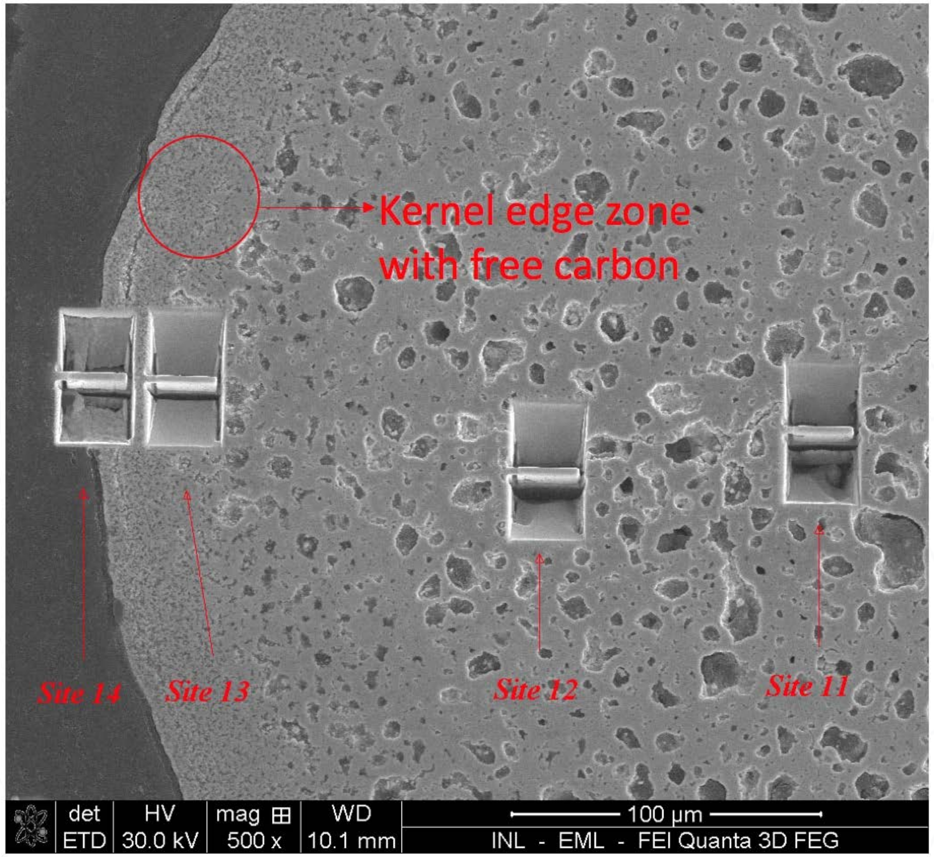


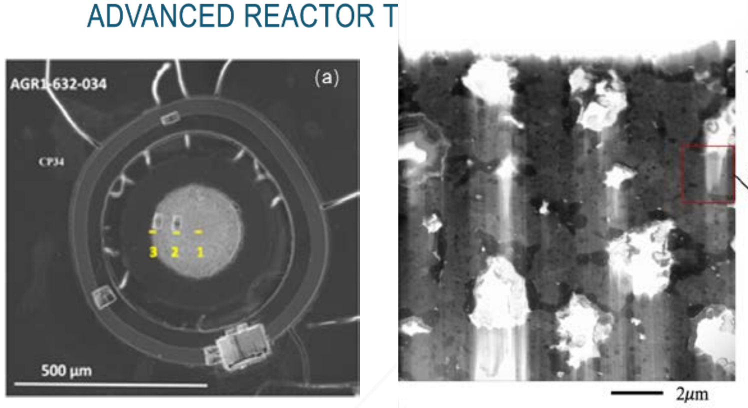
Figure 1: SEM image showing lamellae locations: fuel center, half center, fuel edge, and interface between fuel kernel and buffer layer.

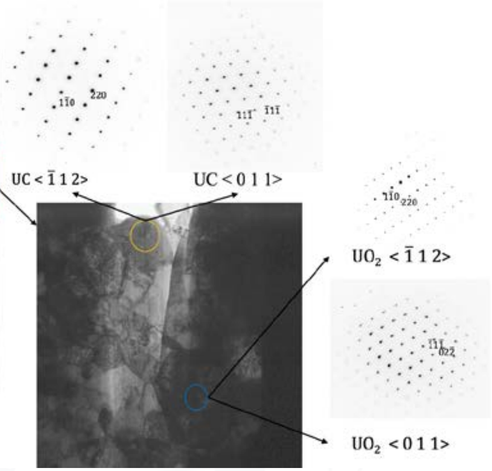
- UC, UC_2 and UO_2 ratio
- Fission product location and composition
- Determining Stoichiometry Changes in the Fuel Kernels Through Quantifying the Light Elements (C, O)

- Techniques
  - ✓ Atom probe tomography (APT)
  - ✓ Electron energy loss spectroscopy (EELS)
  - ✓ Selected Area Diffraction (SAD)
  - ✓ Scanning Transmission Electron Microscopy - Energy Dispersive Spectroscopy (STEM-EDS)

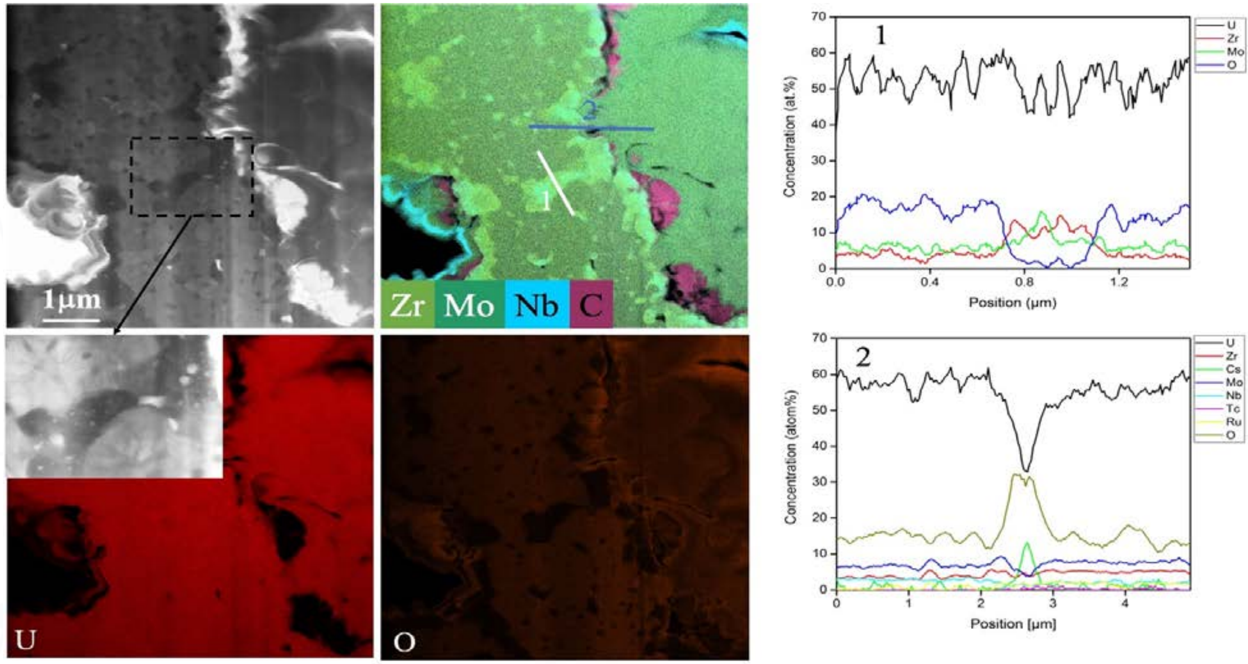
Irradiated Microstructure of AGR-1 Fuel Kernel

[Advanced Reactor Technologies Gas-Cooled Reactor Program Review, July 14-16, 2020, GoToWebinar]





Compact	AGR1-632-034
Burnup (%FIMA)	11.4
Fast neutron fluence (x 10^{25} n/m^2), E>0.18 MeV	2.55
Time-average volume-average temperature (°C)	1070
Time-average peak temperature (°C)	1144



- Fuel matrix consists of UC and UO_2, and UO_2 presents as the dominating phase.
- Zr forms carbide in the solid solution of UC
- Mo, Ru and Tc also enrich in UC phase, and Nb tends to enrich at pore surface.
- Ultra-fine Fission gas bubbles located in UC phase, while UO_2 is free of fission gas bubbles.

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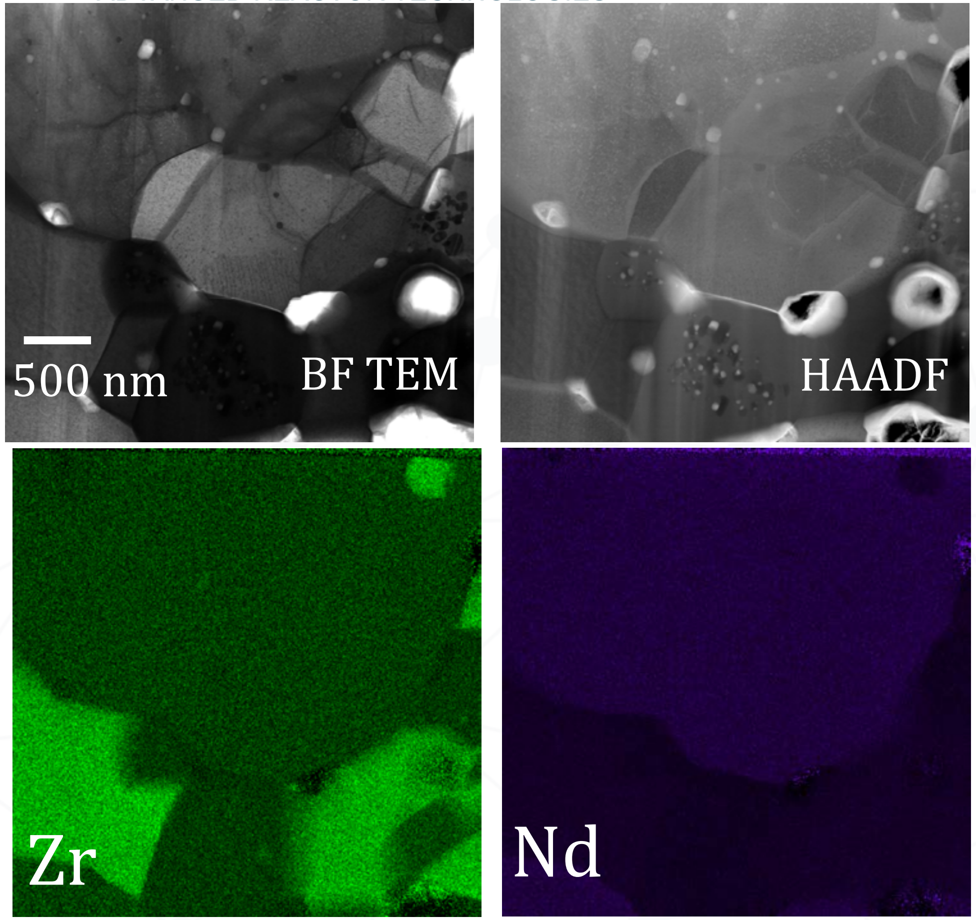
Other Fission Products: Ce, Nd, Pr, Sr and Eu

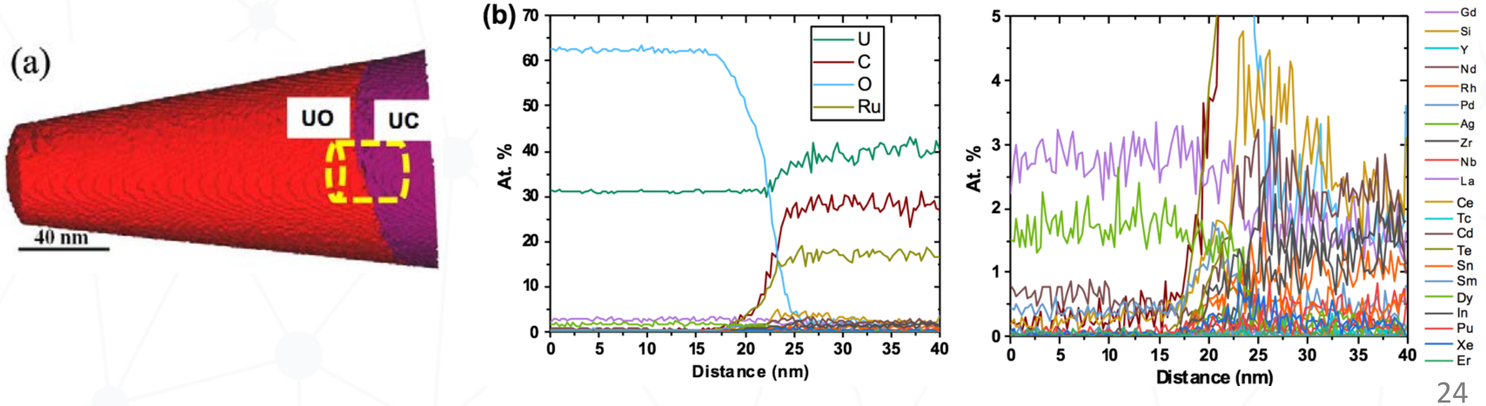


EDS maps from the fuel kernel center for AGR2-222-RS19 particle

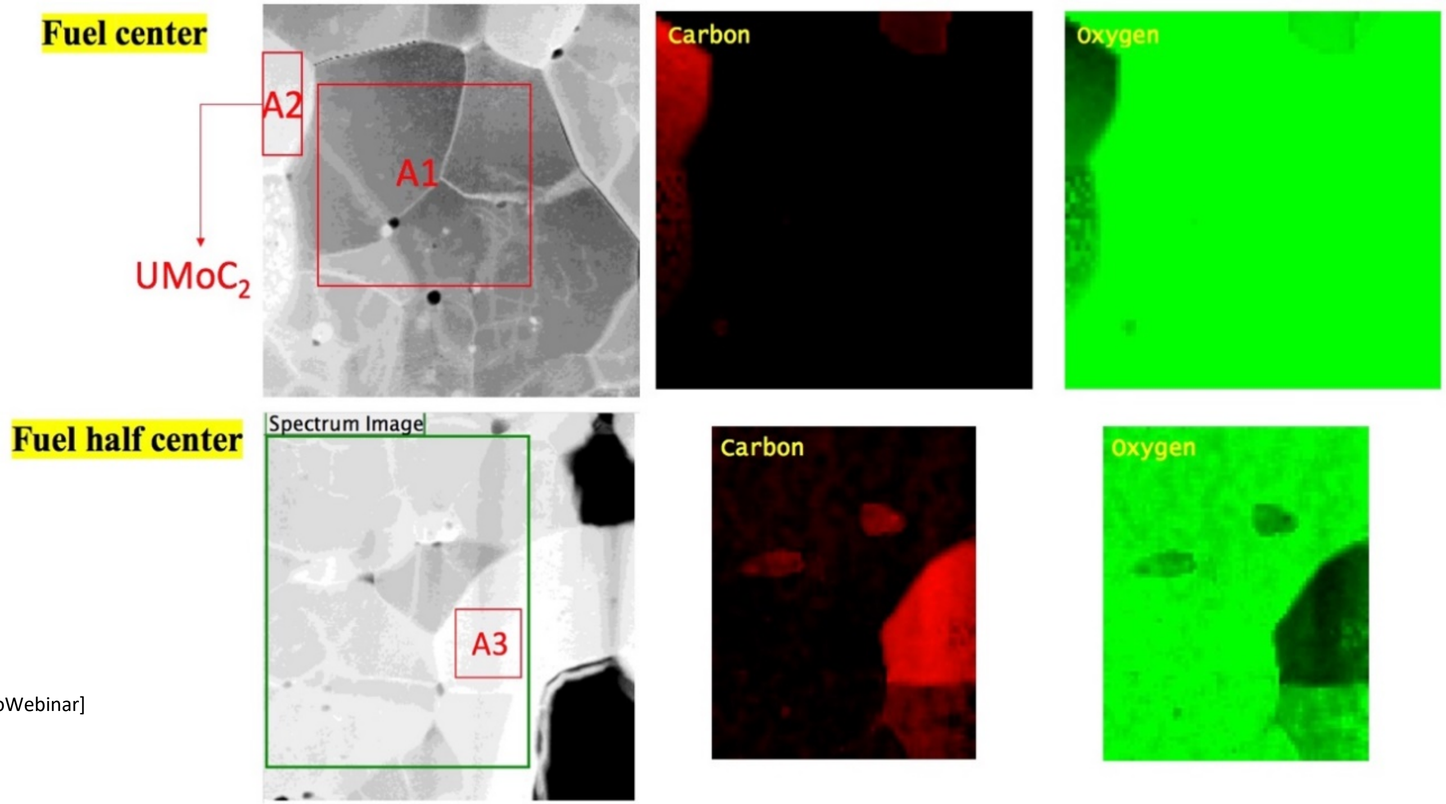
- From EDS analysis, only Nd shows enrichment in UO_2 phase, while the other elements were not positively detected (under the detection limit).
- Limited APT work shows enrichment of Nd, Pd in UO_2
- More APT and TEM work is needed to locate Sr, Eu and other interested fission products. (APT should be calibrated for laser energy as Dy below might be mis-identified).

[Advanced Reactor Technologies Gas-Cooled Reactor Program Review, July 14-16, 2020, GoToWebinar]

Preliminary analysis of initial APT was performed, specifically on interface



Preliminary EELS study: AGR2-222-RS036



Area	Element	Relative concentration (at.%)
A1	C	$0 \pm 6 \times 10^{-6}$
	O	100.0 ± 5.0
A2	C	83.0 ± 4.0
	O	16.5 ± 0.7
A3	C	83.0 ± 4.0
	O	16.5 ± 0.7

[Advanced Reactor Technologies Gas-Cooled Reactor Program Review, July 14-16, 2020, GoToWebinar]

- Additional data collections needed using optimized energy windows and exposure times.
- In both the fuel center and half-center, the UO_2 phase consistently showed almost no carbon, whereas the UC/$UMoC_2$ phase contained a small fraction of oxygen.

ADVANCED REACTOR TECHNOLOGIES

3

SiC Layer: Neutron Irradiation Damage Examination

Neutron Irradiation Damage: Scope and Matrix

What?

- Correlate neutron-induced microstructural
  - defect density and size,
  - volume fraction, and
  - morphology with neutron irradiation parameters (i.e., neutron fluence and temperature)
- Analyze the defect density and distribution in the vicinity of fission product precipitates

Microstructure (Neutron Irradiation Damage (Voids, loops…))

Fission Product Transport

Particle	Ag retention	Fuel Type	Burnup (%FIMA)
AGR1-632-034	65%	Baseline	11.4
AGR1-523-SP01	16%	Variant 1	17.4
AGR1-131-066	39%	Variant 3	15.3
AGR1-433-001 Safety tested	66%	Variant 3	18.6
AGR2-223-R06	8%	AGR-2	10.8
AGR2-222-RS036 Safety tested	84%	AGR-2	12.6
★ AGR2-633-TBD	TBD	AGR-2	7.5
★ AGR1-411-030	90%	Variant 3	19.4

To be determined: TBD | Completed | Future work ★

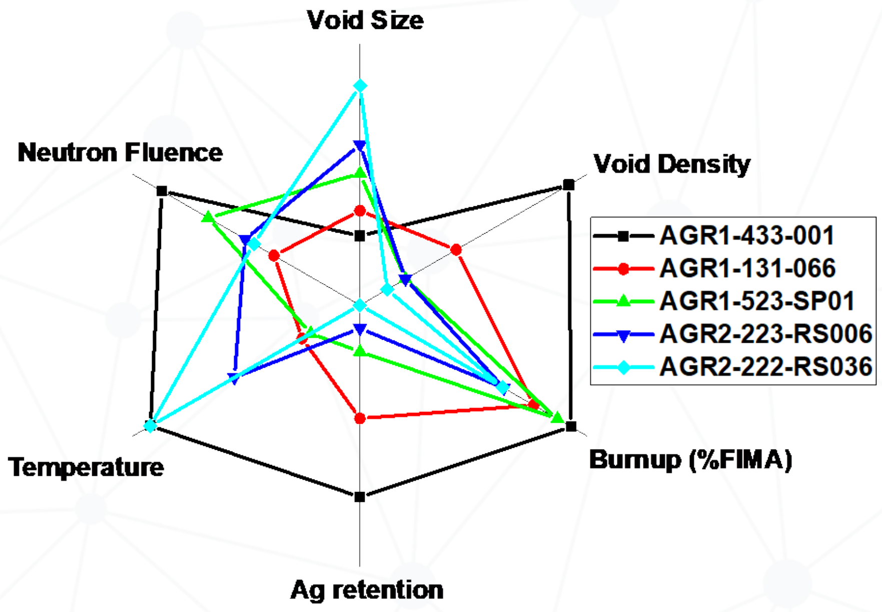
Why?

- 1: effect of fuel type and burnup
- 2: effect of lab scale vs pilot scale (approx. same burnup)
- 3: comparison of AGR-1 vs AGR-2 safety tested particles
- 4: effect of radiation level on variant 3 fuel from AGR-1 particles
- 5: lowest and highest radiation level particles available

Effect of Neutron Irradiation Damage on Ag Retention

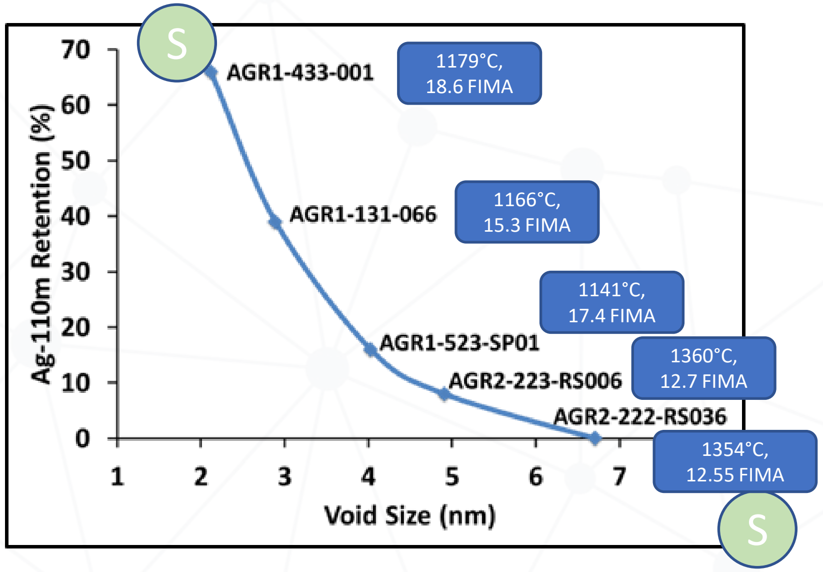
ADVANCED REACTOR TECHNOLOGIES

Irradiation parameters relationship with neutron damage



Inverse relation of void density and size due to possible defect coalescence

Effect of defect size on Ag retention



Ag-110m retention in the SiC layer appears to have an inverse relation with void sizes

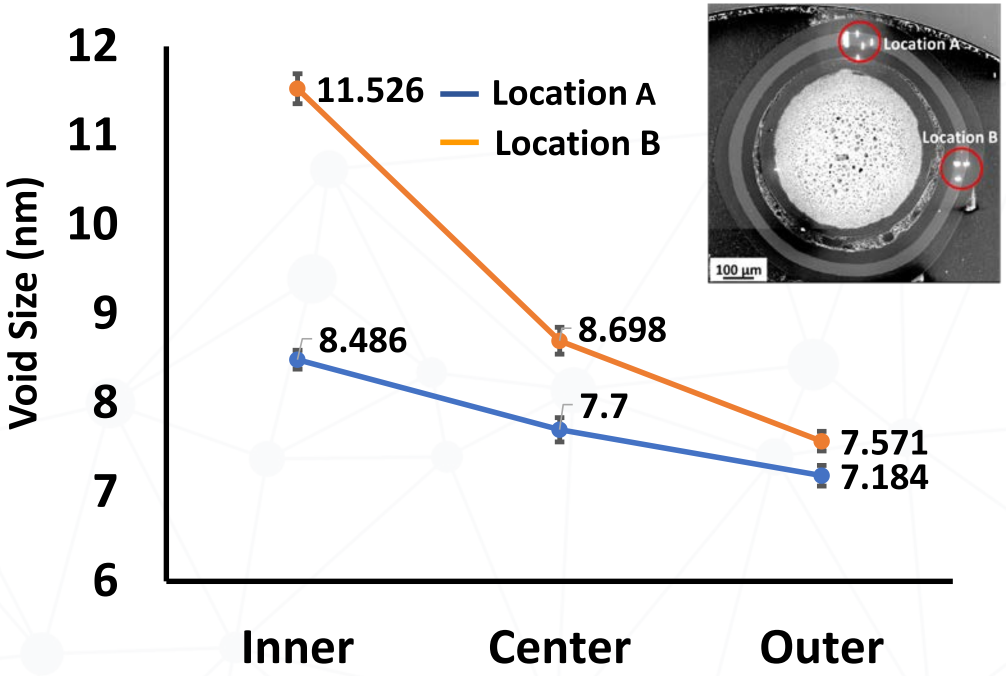
2018

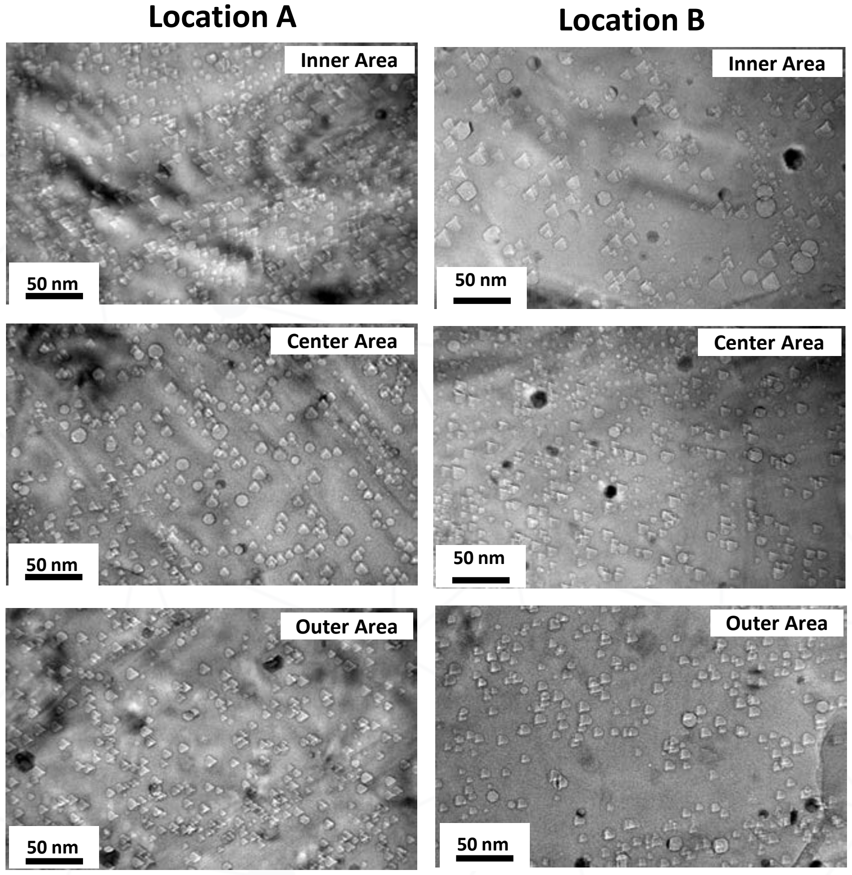
[van Rooyen et al, **Proceedings of HTR** 2018 –Paper No. 3013]

ADVANCED REACTOR TECHNOLOGIES

Effects of Irradiation on TRISO- SiC layer (AGR2-222-RS019)

2020





[Advanced Reactor Technologies Gas-Cooled Reactor Program Review, July 14-16, 2020, GoToWebinar]

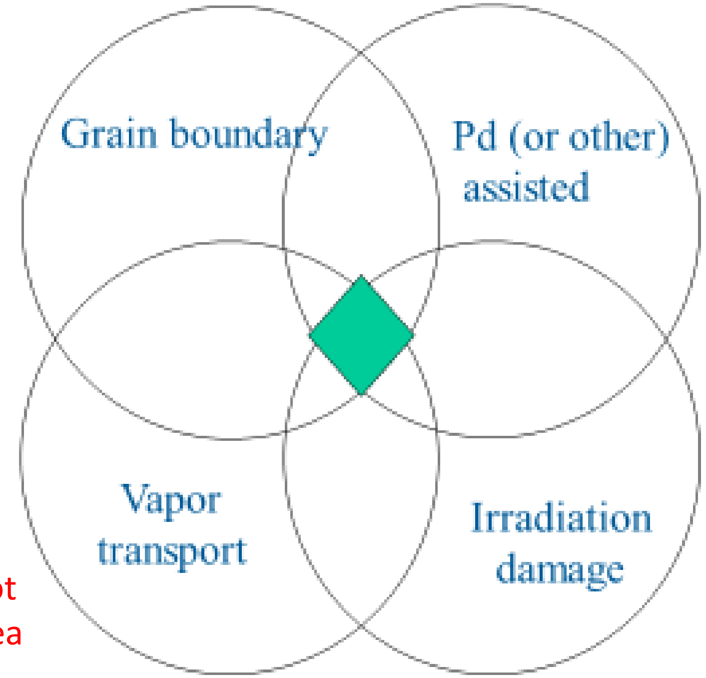
- The void sizes are larger in SiC layer adjacent to region where buffer layer is broken.
- The observed void size variation with integrity of buffer layer can potentially affect the fission product retention.
- Comparison with AGR-1 and other AGR-2 particles next to be completed

ADVANCED REACTOR TECHNOLOGIES

4 Fission Product Transport Mechanisms Complex Interactive Mechanisms?

Present at GB, no significant consistent trends observed (PED)

Needs re-evaluation and updating based on new results; Full integration of results and potential AI needs to apply for trend analysis as big data



Budget and work scope did not allow for work in this topic area

Multiple Combinations of chemical compounds and phases (STEM, APT, EELS, EPMA)

Needs re-evaluation and updating based on new results & needs to compare AGR-2 vs AGR-1

Localized SiC phase transformation (STEM, TEM, HRTEM)

In Progress:

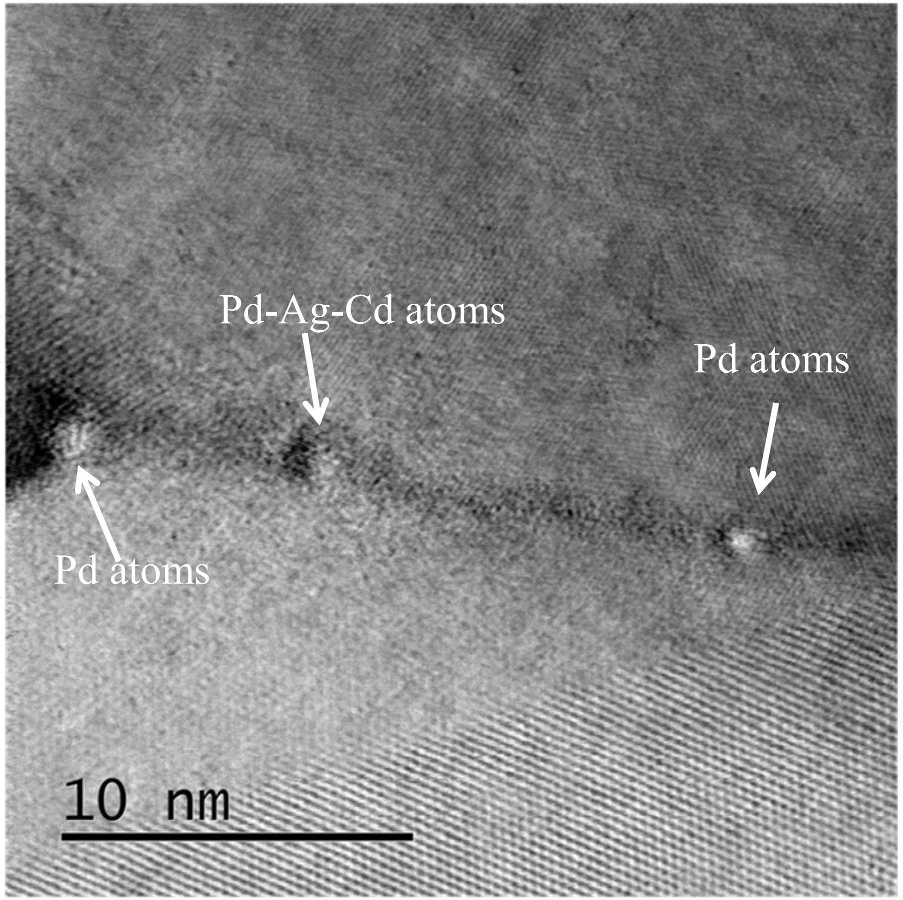
- High temperature in-situ TEM

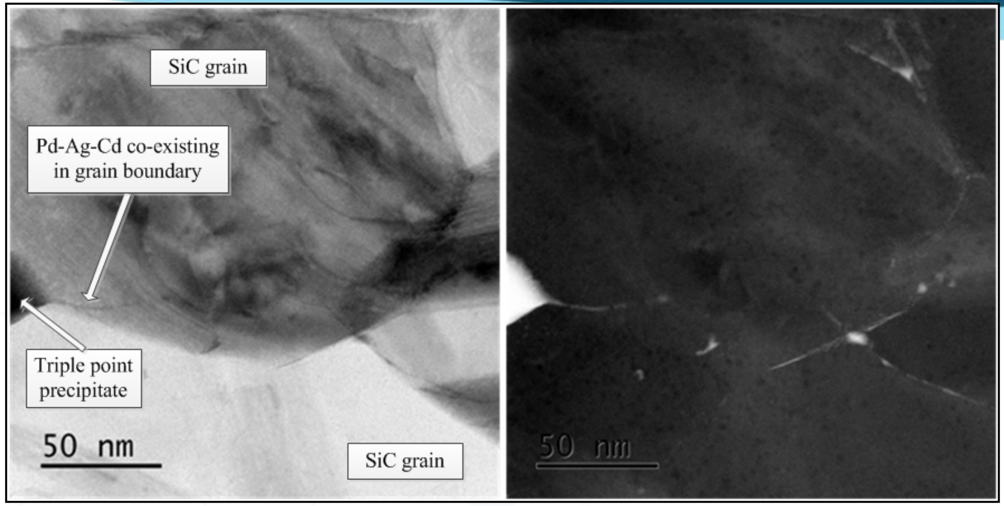
Needs re-evaluation and updating based on new results & reporting outstanding;

HRTEM

FY2013

First-of-a-kind results and has high impact potential on Ag transport mechanism





AGR1-411-030-1b

[Van Rooyen, I. J., E. J. Olivier and J. H Neethling, "Investigation of the Fission Products Silver, Palladium and Cadmium in Neutron Irradiated SiC using a Cs Corrected HRTEM", Journal of Nuclear Materials, 476 (2016) 93 – 101]

- Ag-Pd-Cd co-existing in same grain boundary
- Not possible to differentiate between different atoms
- Ag and Pd always co-exist in the same triple point

- JEOL ARM 200F TEM, operated at 200 kV
- Two CEOS spherical aberration correctors for correction in TEM and STEM modes
- Oxford Instruments XMAX 80 EDS detector
- Gatan Quantum Image filter with dual EELS capabilities
- Imaging and analysis: using sub-angstrom sized probe with probe current density of ~ 68 pA

AGR-1 Conclusions: Fission Product Transport Mechanisms

- Although this work was not predominantly focused on fission product mechanistic studies, results and observations contributed toward knowledge on transport mechanisms.
  - No single mechanism hypothesis can be reported.
  - Complexity of mechanisms is further highlighted by the multiple variations of elemental combinations found in the fission product precipitates.
  - Not necessarily true that a chemical-assisted transport mechanism is dominant.
  - Presence of Ag predominantly on grain boundaries suggests that grain boundary transport mechanism may be prominent.
  - Neutron damage and its effects on fission-product transport needs to be considered in future work

[I. J. van Rooyen, H. Nabielek, J. H Neethling, M. Kania and D.A. Petti, PROGRESS IN SOLVING THE ELUSIVE AG TRANSPORT MECHANISM IN TRISO COATED PARTICLES: "WHAT IS NEW?" Paper 31261, Proceedings of the 2014 International HTR-2014 Conference of High Temperature Reactors, Weihai, China, 2014]
[I.J. van Rooyen, T.M. Lillo, H. Wen, K.E. Wright, J. Madden, J. Aguiar, Advanced Electron Microscopy and Micro Analytical Technique Development and Application on Irradiated TRISO Coated Particles from the AGR-1 Experiment, INL/EXT-15-36281, January 2017]
[Advanced Gas Reactor Fuels Program Meeting, July 18-19, 2017, Idaho Falls, Idaho]

ADVANCED REACTOR TECHNOLOGIES

What is next?

- Kernel Level 3 Milestone Report: March 17, 2021
  - ✓ Isabella J van Rooyen, Yong Yang, Zhenyu Fu, Boopathy Kombaiah, Karen Wright, *“Advanced Microscopy Report on UCO fuel kernels from selected AGR-1 and AGR-2 experiments”*

- Techniques:
  - ✓ APT optimization for irradiated kernel and SiC layers
  - ✓ EELS - EDS integration
  - ✓ GBC (PED) - EDS integration (BIG DATA)
  - ✓ EPMA results fully integrated with APT-EDS?

Acknowledgements

- Transmission electron microscopy work was carried out at the Center for Advanced Energy Studies (CAES) Microscopy and Characterization Suite (MACS); IMCL(MFC) and University of Florida

This work was sponsored by the U.S. Department of Energy's Office of Nuclear Energy, under U.S. Department of Energy Idaho Operations Office Contract DE-AC07-05ID14517, as part of the Advanced Reactor Demonstration Program and the Nuclear Scientific Users Facility–Rapid Turnaround Experiments program.

ADVANCED REACTOR TECHNOLOGIES

Questions??

Isabella van Rooyen

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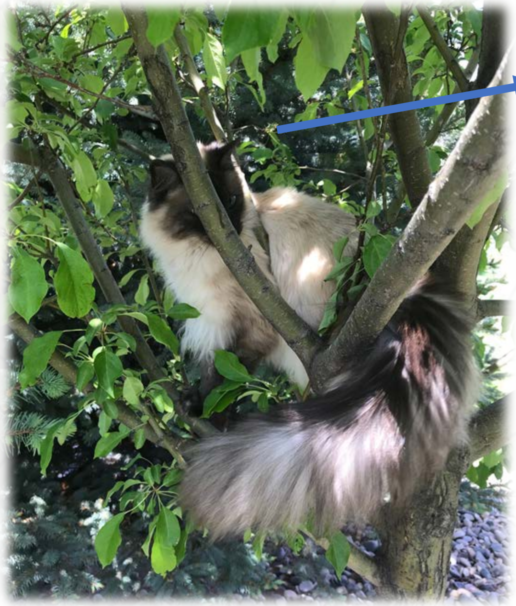
I want also to work on this TRISO fuel research!!



So much to do!

INL

Idaho National Laboratory

ADVANCED REACTOR TECHNOLOGIES

EXTRA SLIDES

AGR-1 and AGR-2 Particle Irradiation History and Characterization

Particle	Ag Retention	Fuel Type	Burnup (% FIMA)	Kernel	SiC layer: FP Distribution and Microstructure				
					EPMA	FIB	STEM	PED	Radiation Damage Measurement (STEM)
AGR1-632-034	0.65	Baseline	11.4						
AGR-523-SP01	0.16	Variant 1	17.4						
AGR1-131-066	0.39	Variant 3 ORNL Lab	15.3						
AGR1-433-001	0.66		18.6 Safety tested*						
AGR1-433-004	0.66		18.6 Safety tested*						
AGR1-433-003 AGR1-433-007	0.66		18.6 Safety tested*						
AGR2-223-R06 (Mount D07)	0.08	AGR2 BWXT	10.8						
AGR2-223-R034 (Mount D06)	0.84		10.8						
AGR2-222-RS36 (Mount D25)	Not detectable Eu = 0.8		12.55 Safety tested*						
AGR2-222-RS19 (Mount D26)	0.20 Eu = 0.54		12.55 Safety tested*						
AGR2-222-RS27 (Mount D26)	0.11 Eu = 0.51		12.55 Safety tested*						
AGR2-633-RS28 (Mount D42)	<0.21		7.46					FY2021**	FY2021**
AGR2-633-RS09 (Mount D43)	0.88		7.46				FY2021	FY2021	FY2021
AGR2-633-RS01 (Mount D43)	0.76		7.46	TBD	TBD	TBD	TBD	TBD	
Unirradiated Baseline As-Fabricated	N/A	AGR1	N/A						
Unirradiated Baseline As Fabricated and Compacted	N/A	AGR2	N/A				FY2021	FY2021	

Completed by Jan 2021

Planned FY2021

* 1600°C, 300h

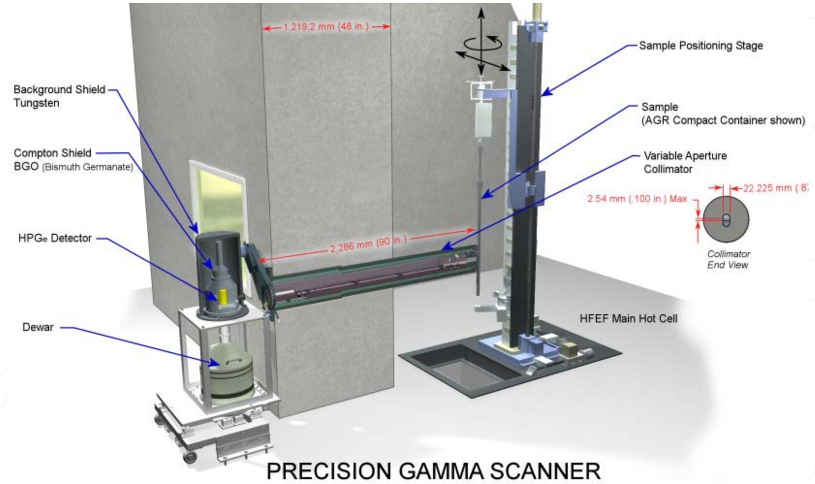
** in progress

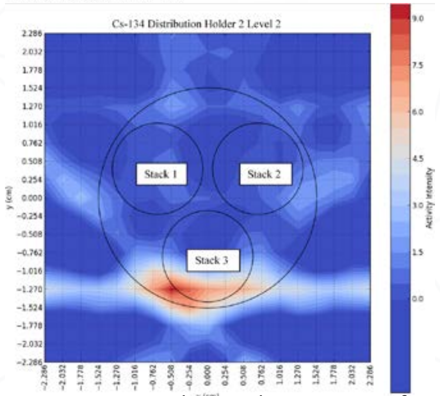
Mass Balance as an Indicator of Fuel Performance

ADVANCED REACTOR TECHNOLOGIES

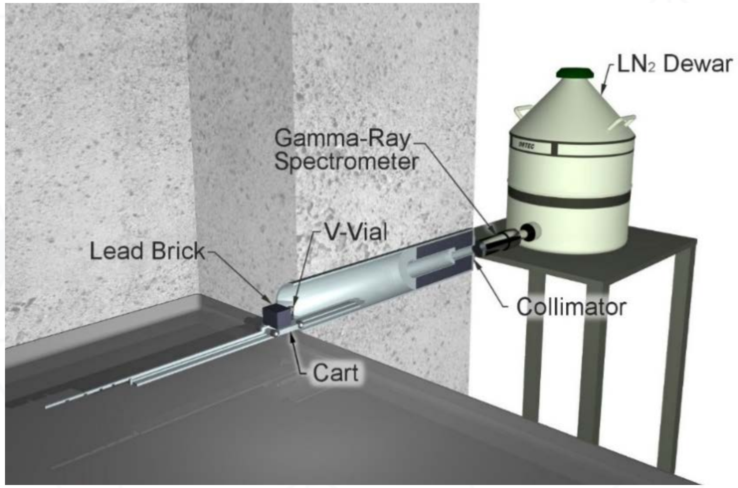
- Analyze irradiation experiment components to determine fission product release from fuel compacts

Individual Fuel Compact and Graphite Holder Gamma Scanning

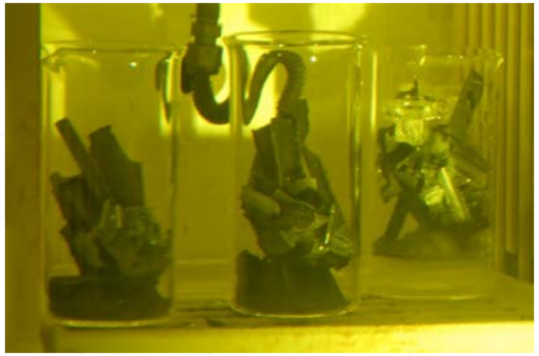




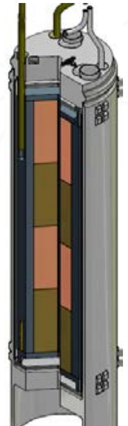
Capsule Components Counting



Burn-leach of graphite and leach of ceramic components for Sr-90



Leach of metal components for gamma-emitters and Sr-90

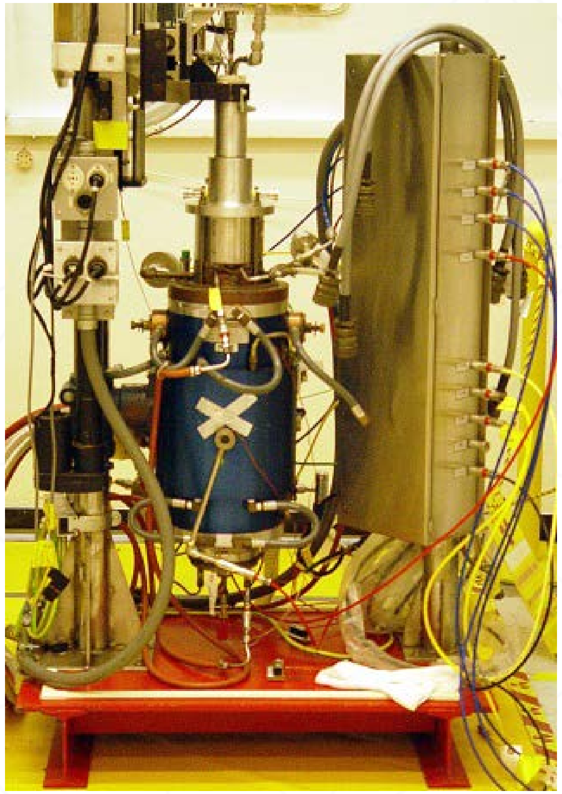


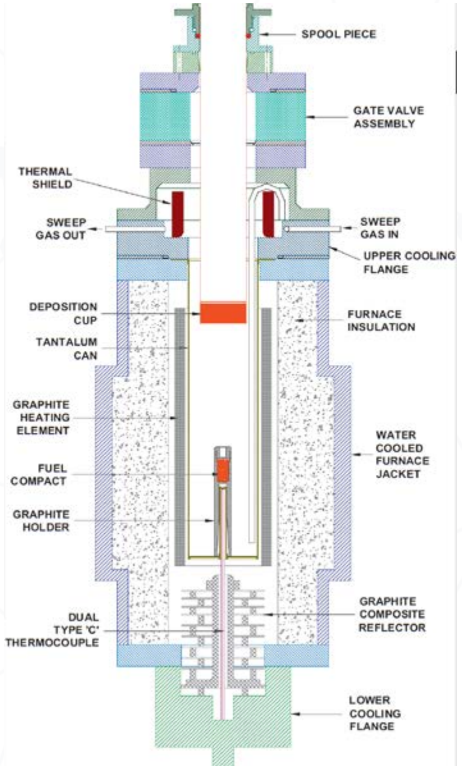
[John Stempien]

Safety Tests in Helium

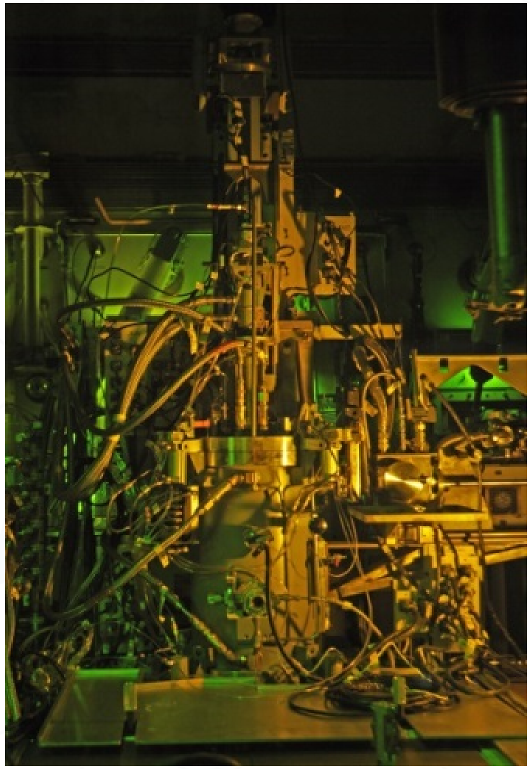
- High-temperature heating tests 1500-1800°C at INL and ORNL
- Determine SiC and TRISO failure rates

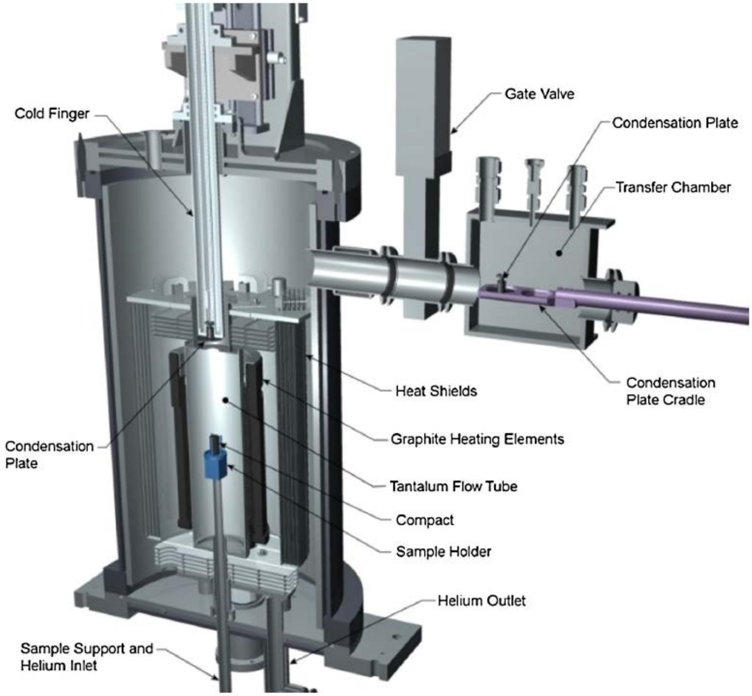
Core Conduction Cooldown Test Facility (CCCTF)





Fuel Accident Condition Simulator Furnace (FACS)

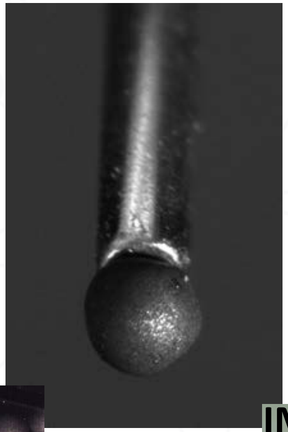


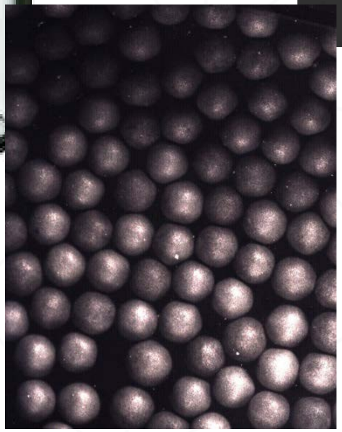


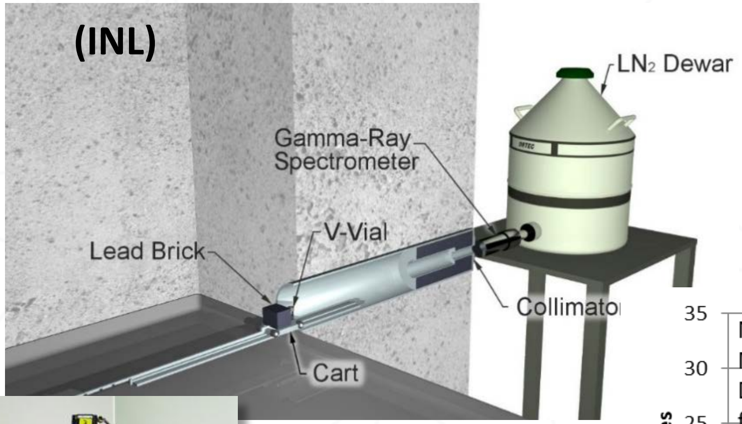
[John Stempien]

Fuel Particle Integrity and Compact Fission Product Retention

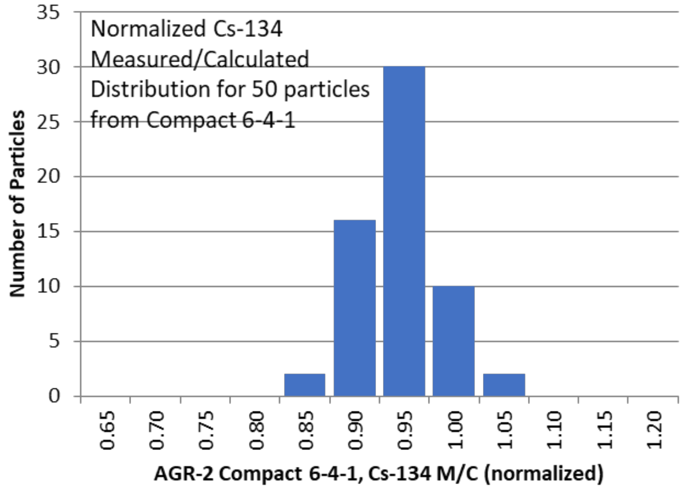
- Compact destructive analyses: deconsolidation leach-burn-leach (DLBL)
- Particle inspection and gamma counting
  - Small subset at INL
  - All particles via irradiated microsphere gamma analysis (IMGA) at ORNL







Particles equivalent inventories in AGR-2 Compact 6-4-1 outside of the SiC layers. Indicates no failed SiC layers.				
Ag-110m	Cs-134	Eu-154	Sr-90	U-238
<1.5E+0	2.38E-3	1.62	2.53E-1	1.69E-2



Electron Probe Micro-Analysis (EPMA)

ADVANCED REACTOR TECHNOLOGIES

Fission Product Identification, Quantification and Location

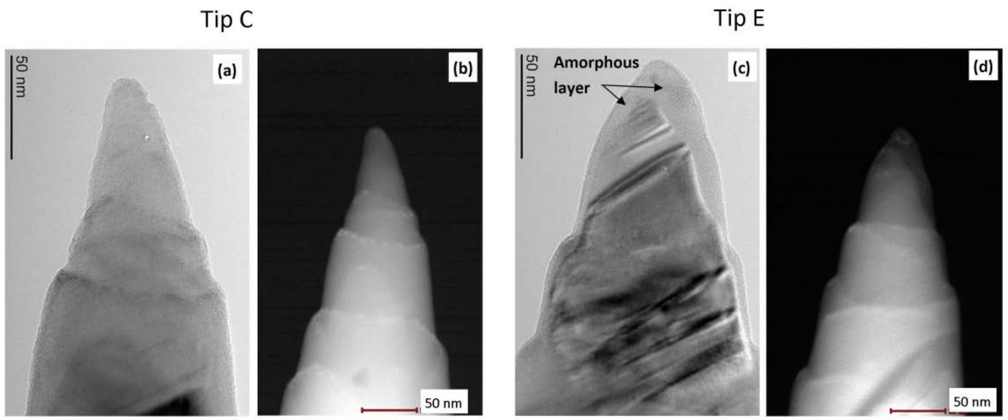
EPMA is an electron beam instrument similar to a scanning electron microscope, but unlike an SEM, an EPMA is optimized for quantitative chemical composition analysis rather than imaging.

Shows how a large number of fission products distribute within the particle.

Shielded enclosure surrounding INL's EPMA

ADVANCED REACTOR TECHNOLOGIES

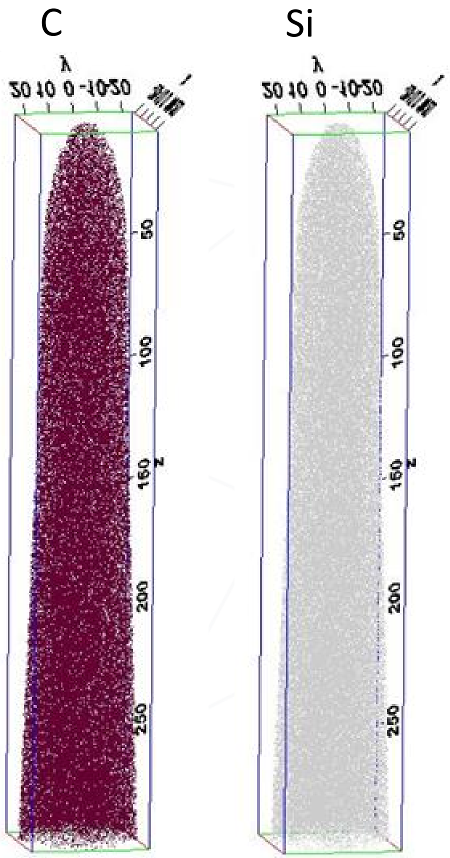
Optimization with unirradiated SiC



Run #	R33_04253
# ions	14 M
Mode	laser
Temperature (setting, K)	90
Laser energy (pJ)	90
Pulse frequency (kHz)	200
Detection Rate (%)	0.1

Run #	R33_04254
# ions	1.7 M
Mode	laser
Temperature (setting, K)	90
Laser energy (pJ)	90
Pulse frequency (kHz)	200
Detection Rate (%)	0.1

Two large datasets up to 300 nm in depth, which contained up to 15 million atoms, were acquired

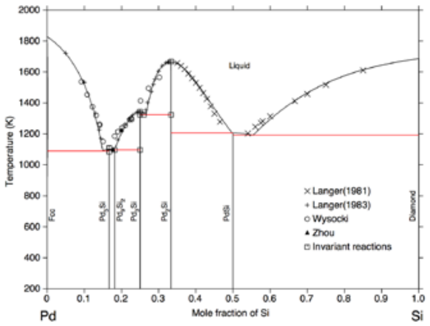


- The most successful measurements were obtained at a set of parameters of a temperature of 90 K and laser energy of 90 – 100 pJ. Laser parameters for the earlier FY2014 APT work on irradiated SiC consisted generally at lower temperature, i.e., 90 K for current vs. 40 – 80 K for previous.

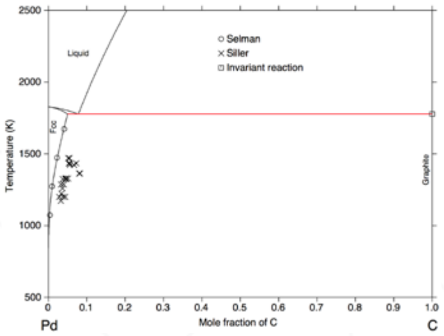
- APT on irradiated SiC , August 2015

A new thermodynamic model for Pd-Si-C system

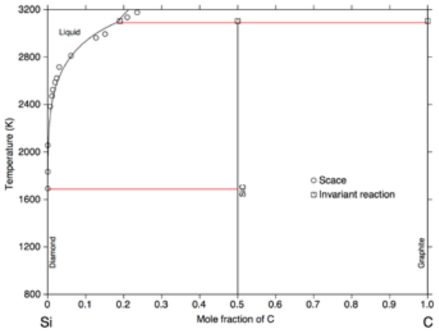
- Palladium, a major fission product of uranium, is known to form palladium silicides and other complex multi-element compounds
- A new physics-based, high fidelity thermodynamic model has been developed in FY19 for understanding the chemical reaction between fission product Pd and SiC coating
- Model predicted phase diagrams in excellent agreement with experiments



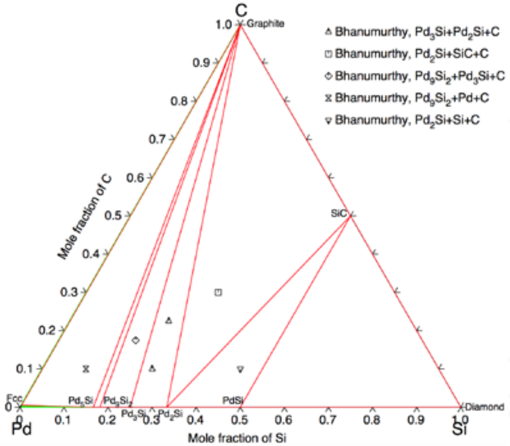
Pd-Si



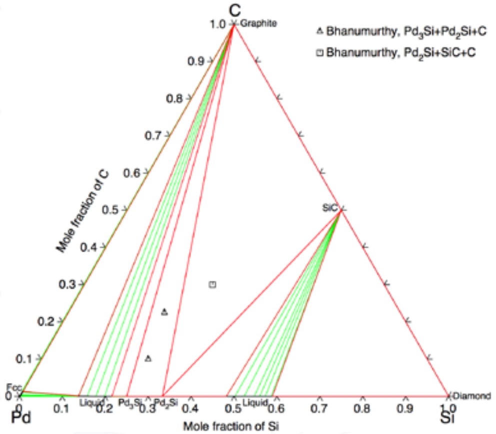
Pd-C



Si-C



Pd-Si-C at 800℃

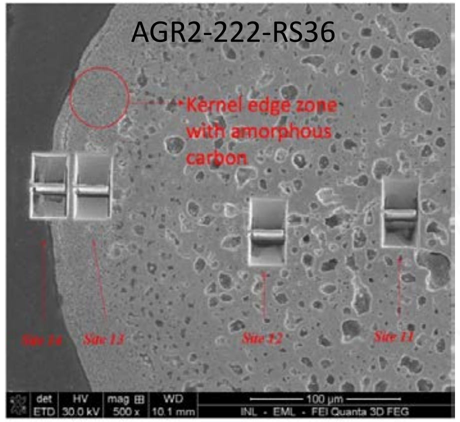


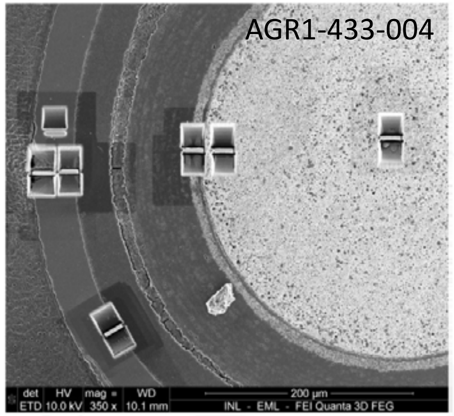
Pd-Si-C at 1000℃

[Chao Jiang, Isabella J. van Rooyen, and Subhashish Meher, “Ab initio study and thermodynamic modeling of the Pd-Si-C system”, Computational Materials Science.]

ART ADVANCED REACTOR TECHNOLOGIES

Safety test irradiated AGR-1 and AGR-2 Comparison





Compact	AGR2-222-RS36	AGR1-433-004
Burnup (%FIMA)	12.55	18.63
Fast neutron fluence (x 10^{25} n/m^2), E>0.18 MeV	3.39	4.16
Time-average volume-average temperature (°C)	1287	1094
Time-average peak temperature (°C)	1354	1179
Safety tested	300 hrs at 1600(°C)	300 hrs at 1600(°C)

- AGR-2 fuel kernel mainly consists of UC, UO_2, $UMoC_2$, $U_2Ru(Tc)C_2$. Safety testing clearly promotes the precipitation of $UMoC_2$ and $U_2Ru(Tc)C_2$ phases.
- Few UC_2 grains were identified at the fuel kernel half center.
- No U_2RuC_2 or $UMoC_2$ precipitates positively identified in AGR-1 fuel kernels. (possible correlation between presence of U_2RuC_2/$UMoC_2$ and initial fuel chemistry (significant high content of $UC_{1.86}$ in the AGR-2)).
- Various rod-shaped precipitates observed in fuel center and edge zone of the AGR-1 fuel kernel. EDS maps show that the precipitates are enriched with Rh, Ru, Tc, Pd, with/without Mo.

UCO fuel kernel		AGR-1	AGR-2
Diameter (μm)		348.4 ± 8.3	426.7 ± 8.8
Density (g/cm^3)		10.7 ± 0.026	11.0 ± 0.030
^{235}U enrichment (at.%)		19.74	14.03
Chemistry (mole%)	UO_2	67.9	71.4
	$UC_{1.86}$	0.4	12.3
	UC	31.7	16.4

[Advanced Reactor Technologies Gas-Cooled Reactor Program Review, July 14-16, 2020, GoToWebinar]