

# Computational Modeling and Post Irradiation Examination Capabilities at the Advanced Test Reactor National Scientific User Facility

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Metallurgical and Materials Engineering Department

ATR-NSUF Users Week

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**COLORADO SCHOOL OF MINES**  
EARTH • ENERGY • ENVIRONMENT

# Presentation Outline

- Missouri S&T/CSM FSRT background
- Research summaries
  - Microstructural investigation of high-burnup uranium oxide fuels
  - Microstructural investigation of TRISO fuel compacts
  - Atomistic modeling of vacancy and noble gas transport in uranium oxide
  - Microstructural FEA analysis of U-Mo dispersion-type fuels
- Questions?

# Faculty-Student Research Team (FSRT)

- ▶ In 2009, the ATR-NSUF funded two faculty-student research teams
- ▶ The Missouri S&T/Colorado School of Mines team consisted of 3 faculty and 4 students
  - Dr. Jeffrey King, Missouri S&T/Colorado School of Mines
  - Dr. Brian Gorman, Colorado School of Mines
  - Dr. Greg Hilmas, Missouri S&T
  - Vaibhav Khane, Missouri S&T
  - Rita Kirchhofer, Colorado School of Mines
  - Torey Semi, Colorado School of Mines
  - Melissa Teague, Missouri S&T

# FSRT Goals

- ▶ The goal of the Missouri S&T/Colorado School of Mines team was to combine post-irradiation examination and computational methods to enhance the capability to analyze and model high-burnup reactor fuel
- ▶ This evolved into four parallel lines of inquiry
  - Microstructural investigation of high-burnup uranium oxide fuels (Melissa Teague)
  - Microstructural investigation of TRISO fuel compacts (Rita Kirchhofer)
  - Atomistic modeling of vacancy and noble gas transport in uranium oxide (Torey Semi)
  - Microstructural FEA analysis of U-Mo dispersion-type fuels (Vaibhav Khane)

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# SIMFUEL Development

- Used MCNPX to calculate 145 GWd/tM Burn-up LWR fuel composition
- Selected the 10 most abundant elements
- Made substitutions based on availability and chemical compatibility
- $\text{BaCO}_3$ ,  $\text{CeO}_2$  (Np),  $\text{La}_2\text{O}_3$  (Am, Cm),  $\text{MoO}_3$ , SrO,  $\text{Y}_2\text{O}_3$ ,  $\text{ZrO}_2$ ,  $\text{Rh}_2\text{O}_3$ , PdO,  $\text{RuO}_2$  (Tc),  $\text{Nd}_2\text{O}_3$  (Pr, Pm, Sm)

SIMFUEL Composition (by weight %)		
$\text{ZrO}_2$	90.96%	
$\text{CeO}_2$	1.93%	
$\text{ZrO}_2$	1.28%	
$\text{MoO}_3$	1.48%	
$\text{Nd}_2\text{O}_3$	1.26%	
PdO	1.30%	
SrO	0.26%	
$\text{BaCO}_3$	0.76%	
$\text{La}_2\text{O}_3$	0.44%	
$\text{Pr}_2\text{O}_3$	0.33%	

# Processing of SIMFUEL

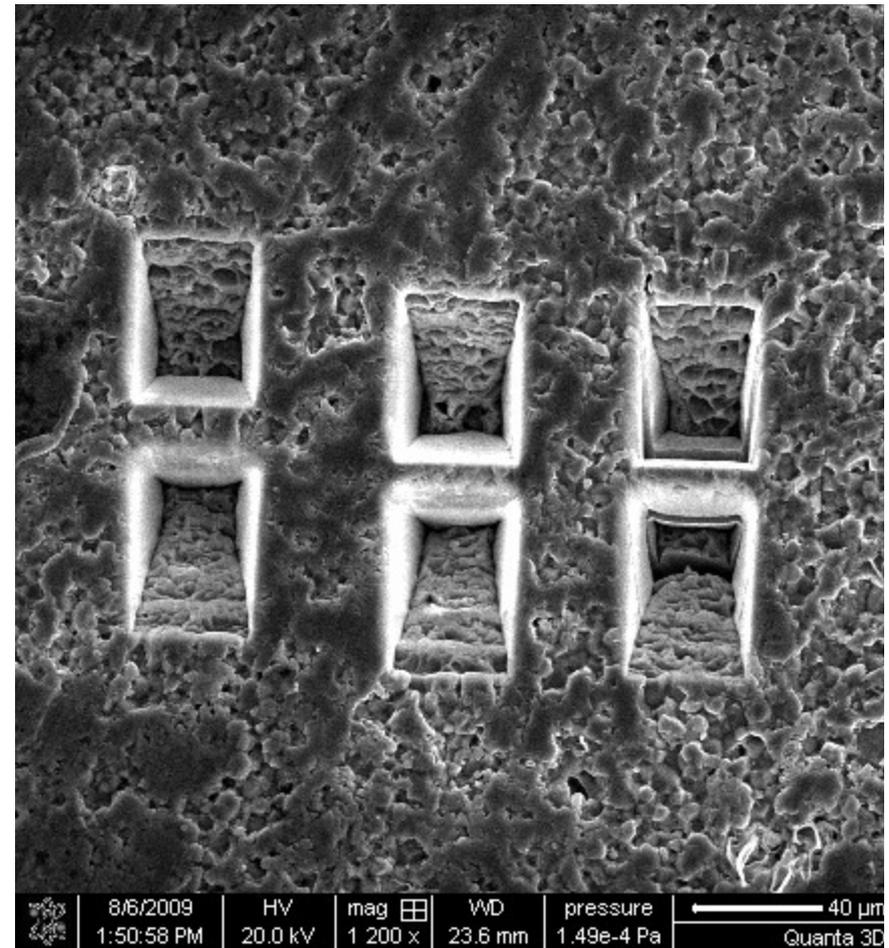
- Pellets uniaxially pressed at 4 ksi
- Cold Isostatic pressed to 35 ksi
- Sintered under 90/10 Ar/H<sub>2</sub> up to 1650°C at 10°C/min, then natural furnace cooled
- Thermally shocked upon cooling, indicated slower cooling rate needed.



SIMFUEL  
sintered at  
1650°C for 30min  
in flowing 90/10  
Ar/H<sub>2</sub>

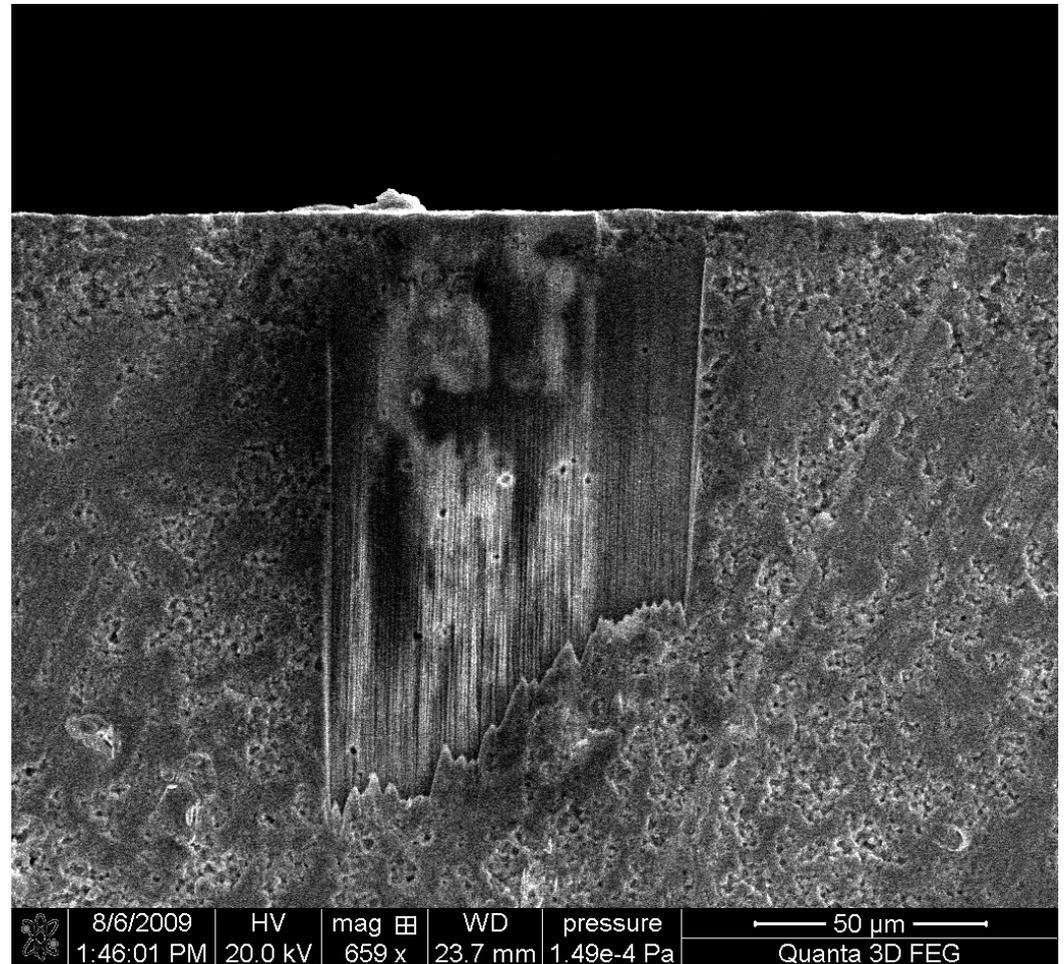
# Characterization of SIMFUEL

- Mechanically polished to 1200 grit
- Used FIB to mill 3 trenches for monopole installation



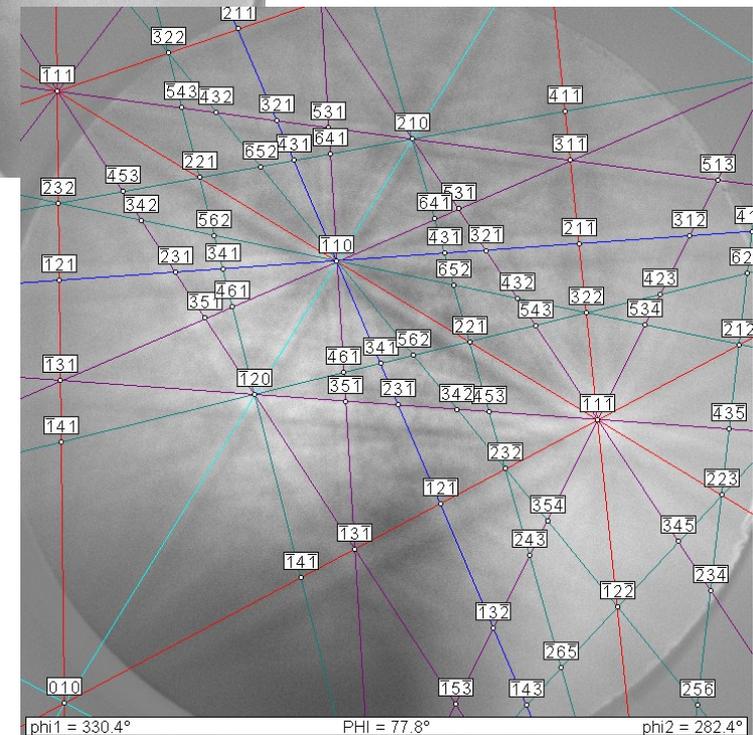
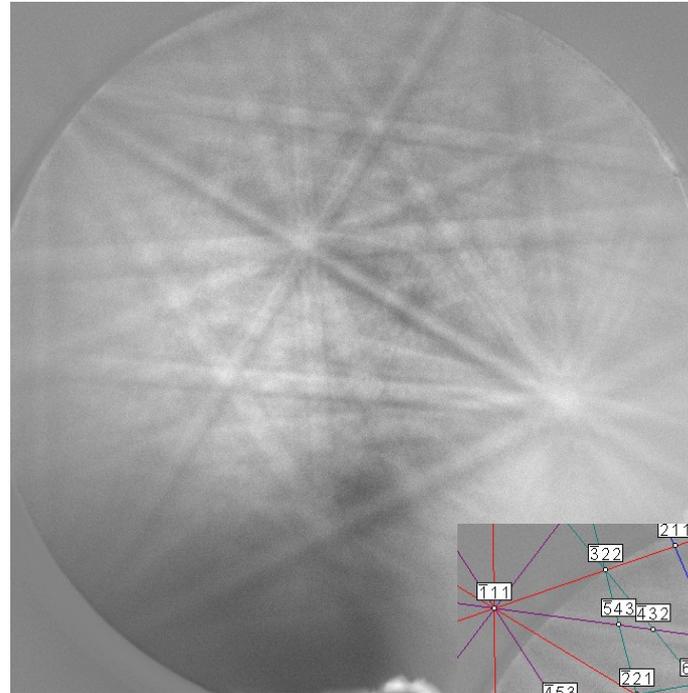
# EBSD

- Milled at grazing incident angle
- Rough Milling
  - 30 kV
  - 6° tilt
  - 15 nA
- Cleaning
  - 5 kV
  - 6° tilt
  - 8 nA



# EBSD Patterns

- Collected using OIM software
- Indexed to m3m cubic  $\text{ZrO}_2$

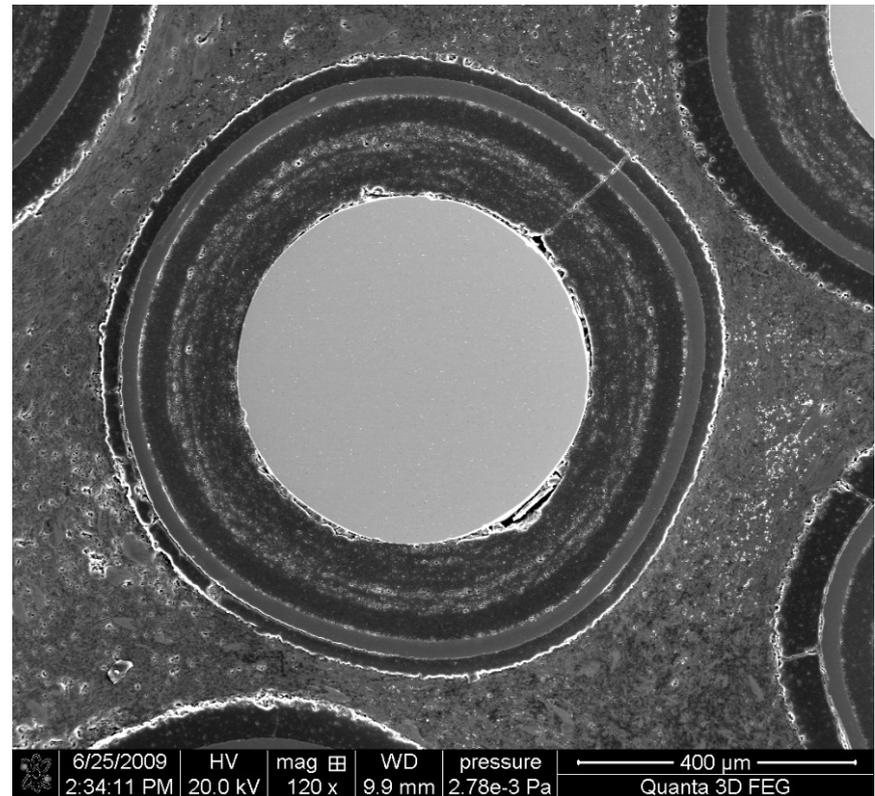


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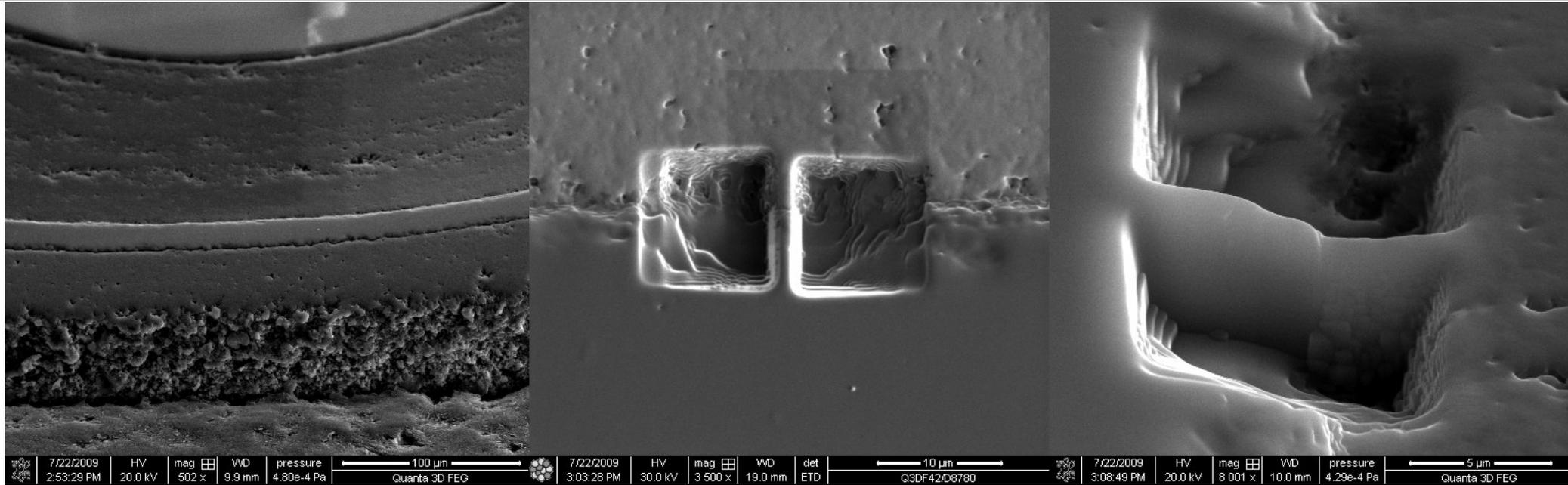
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# TRISO Specimen Preparation

- Sample preparation of a TRISO compact and individual particles
- Use focused ion beam milling to prepare specimens for further analysis
  - Special interest in milling the interfaces
  - Prepare surface for EBSD analysis

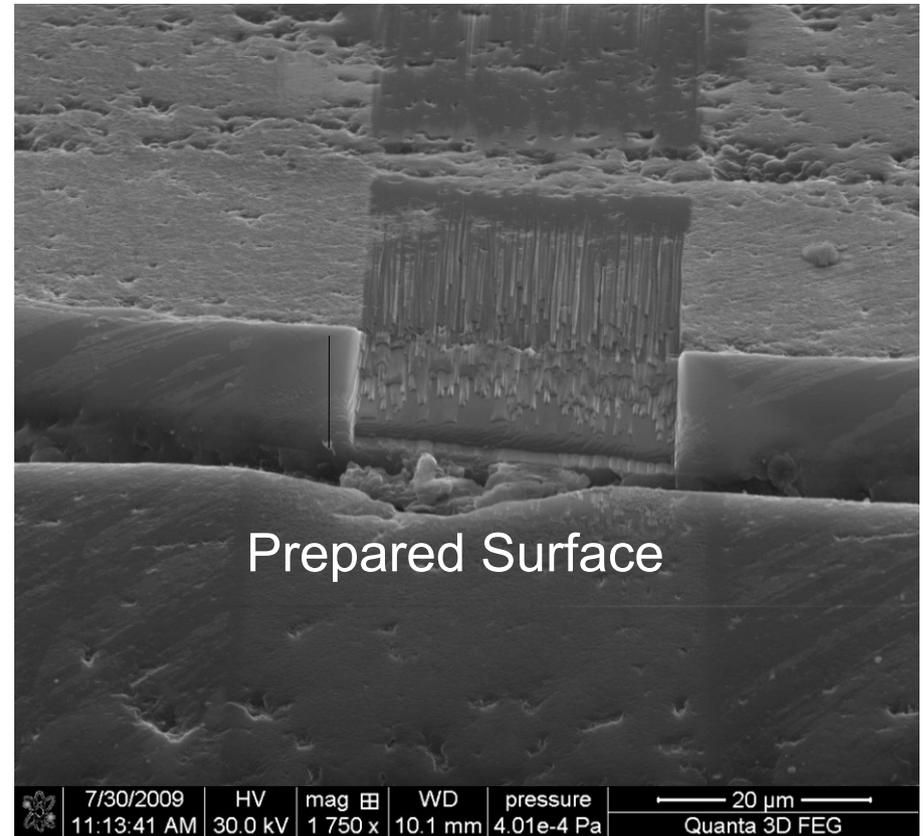
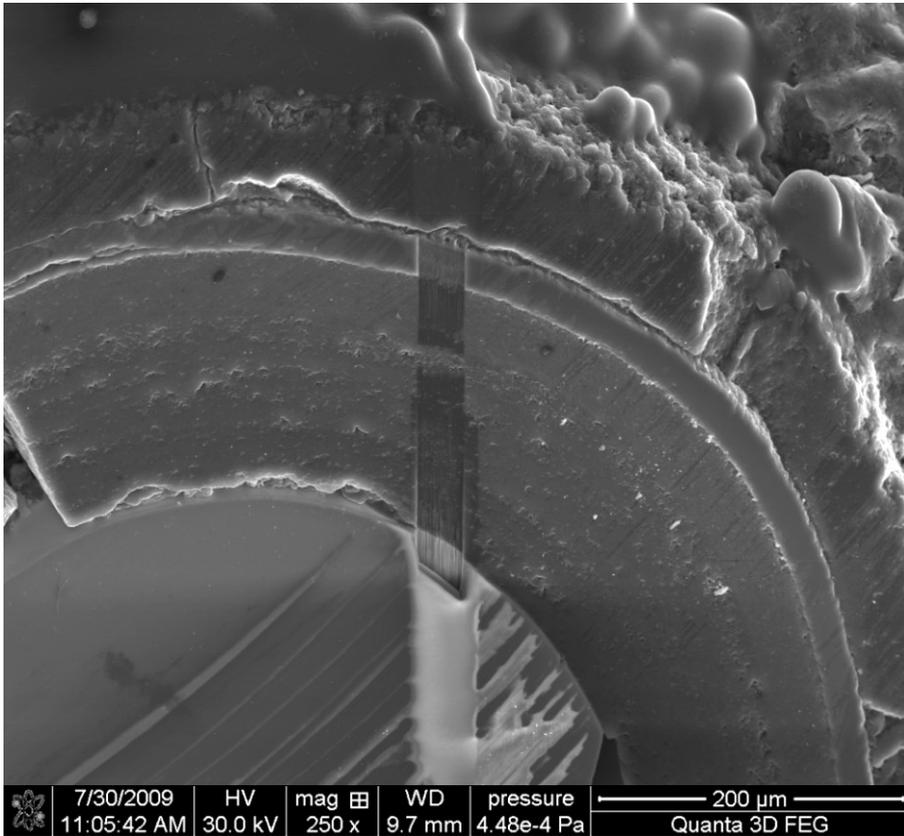


# Focused Ion Beam Milling



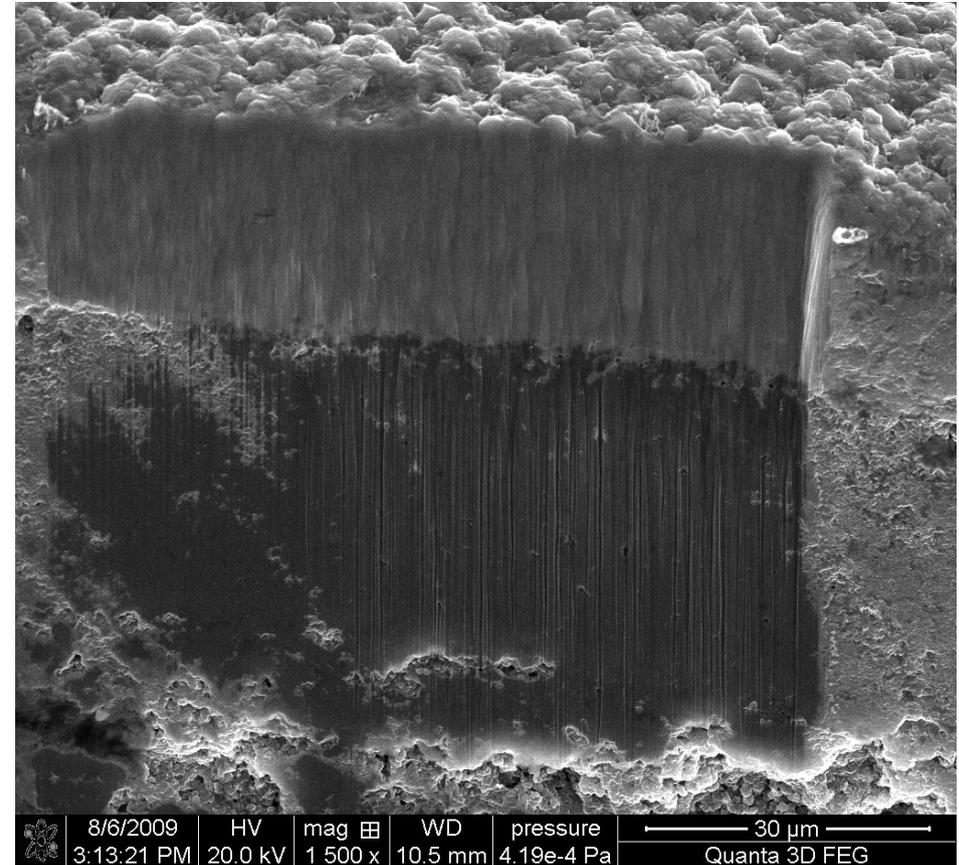
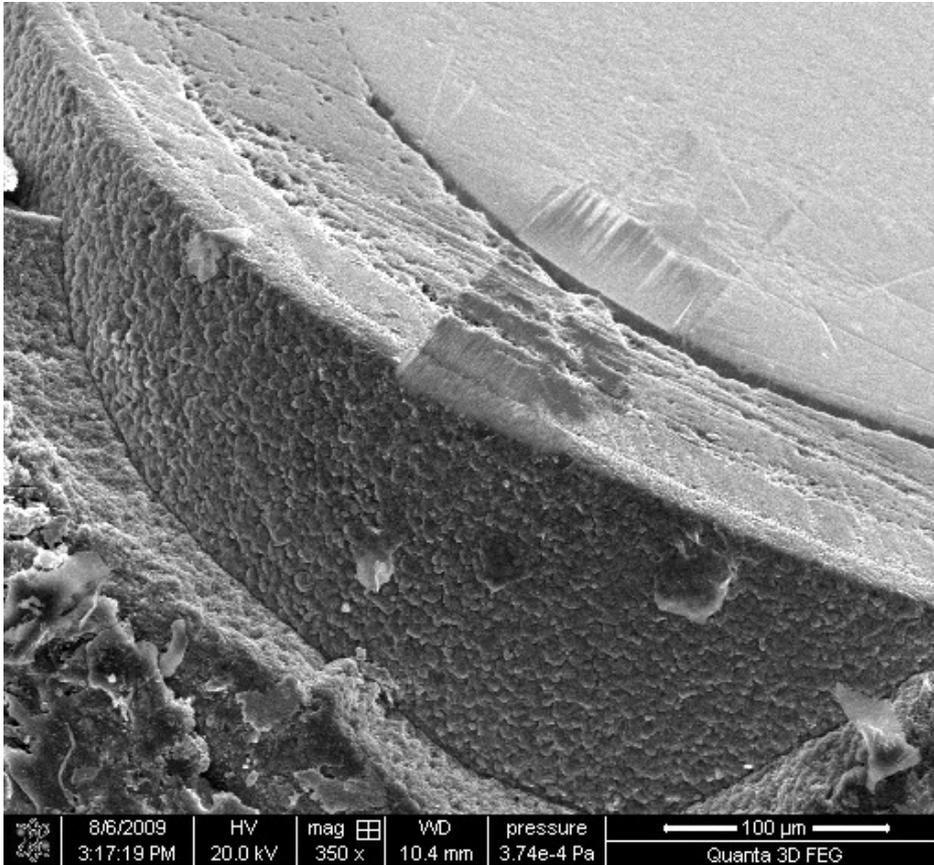
- Milling at the interface of the SiC and PyC layers
  - Milling rates are different for the different materials
  - Can obtain a great surface finish

# Sample Preparation for EBSD



- Specimen preparation for EBSD
  - Special interest in the SiC layer and PyC of the TRISO coated particle.
- EBSD analysis of the SiC
  - Get polymorph information

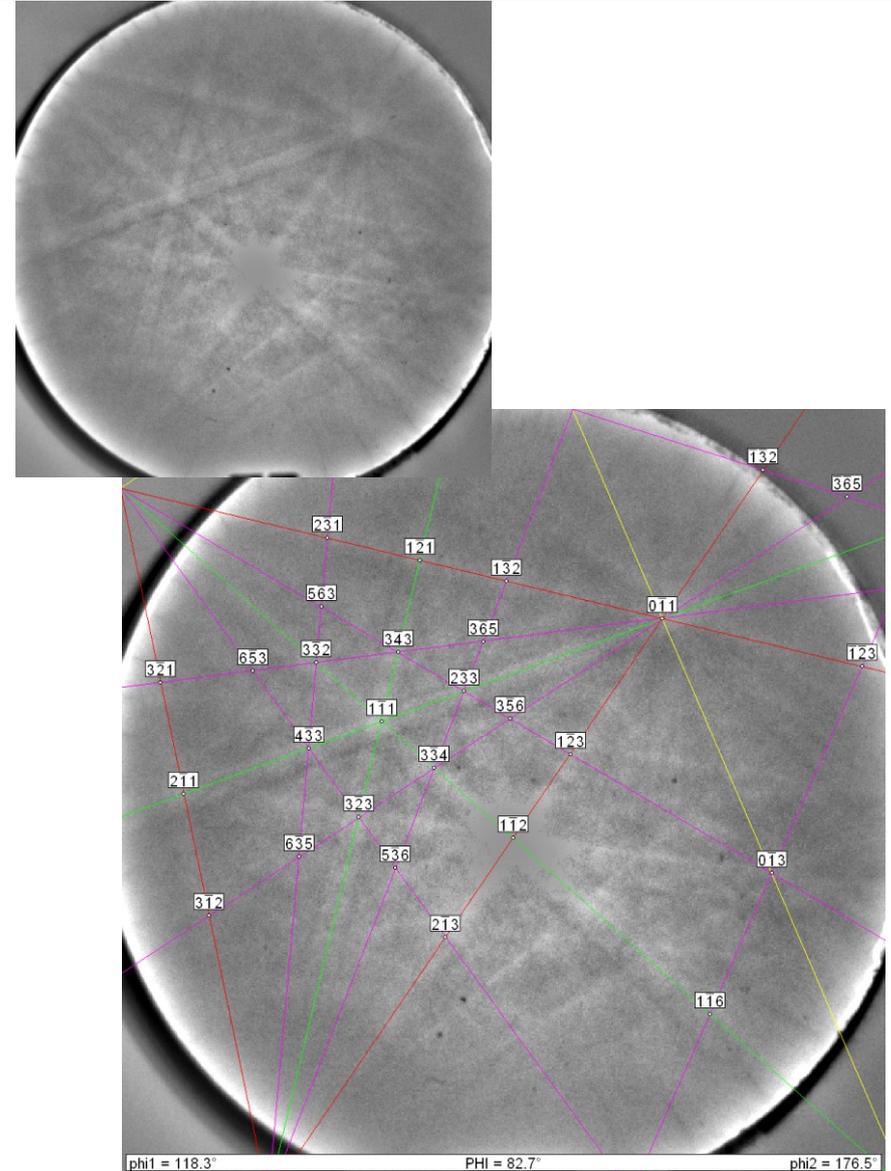
# Sample Preparation for EBSD



- Reduce curtain effect by using a low accelerating voltage (also reduces amorphization)
- Need to use Pt layer to protect against ion impingement

# EBSD Patterns for SiC

- EBSD data obtained after surface preparation with the FIB
- Determine crystallographic information:
  - Lattice parameter
  - Crystal structure
- EBSD mapping



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# Xenon and Krypton

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Noble gases cause two major problems in high-burnup nuclear fuels:

- Fission gas bubble swelling
  - Leads to degradation of fuel integrity
  - Reduces local thermoconductivity
- Gas release from reactor fuel
  - Can lead to breach of cladding
  - Release of radioactivity

# Fundamental Questions

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- Where do FP gas atoms settle in lattice?
- How do these atoms come together to form small clusters?
- Can the clusters move?
- Do bubbles (large clusters) form? Is there a limit on their size?
- Can bubbles move?
- How do FP gas atoms end up at grain boundaries?
- What role do vacancies play in all of this?

# Research Strategy

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- Focus on uranium dioxide
- Use density functional theory (DFT) for configuration, mechanical and diffusion studies.
- Elucidate motion of fission product atoms and formation/motion of small clusters of fission product atoms

# Research Strategy, cont.

- Vacancies
  - One U vacancy, no strain:
    - Goal: Determine diffusivity of vacancy
    - Method: Transition State Theory (activation energy)
  - Modify vacancy, perform similar calculations
    - Frenkel Defects
    - Schottky Defects
  - Add strain:
    - Account for Soret effect, temperature gradients, other forces
    - Model by lattice dilation
    - Repeat goals using same method

# Research Strategy, cont.

- Gas atoms (Kr)
  - Introduce into (vacancy) defects
    - Determine energetically preferred defect site
      - Frenkel, Schottky, O vacancies, U vacancy
- Clusters
  - Quantify tendency of motion of  $N > 1$  vacancies, Kr atoms
    - Agglomeration
    - Energy barriers
    - Temperature dependence
    - Critical size
  - Describe fate of O atoms

# Density Functional Theory

Primary principle:

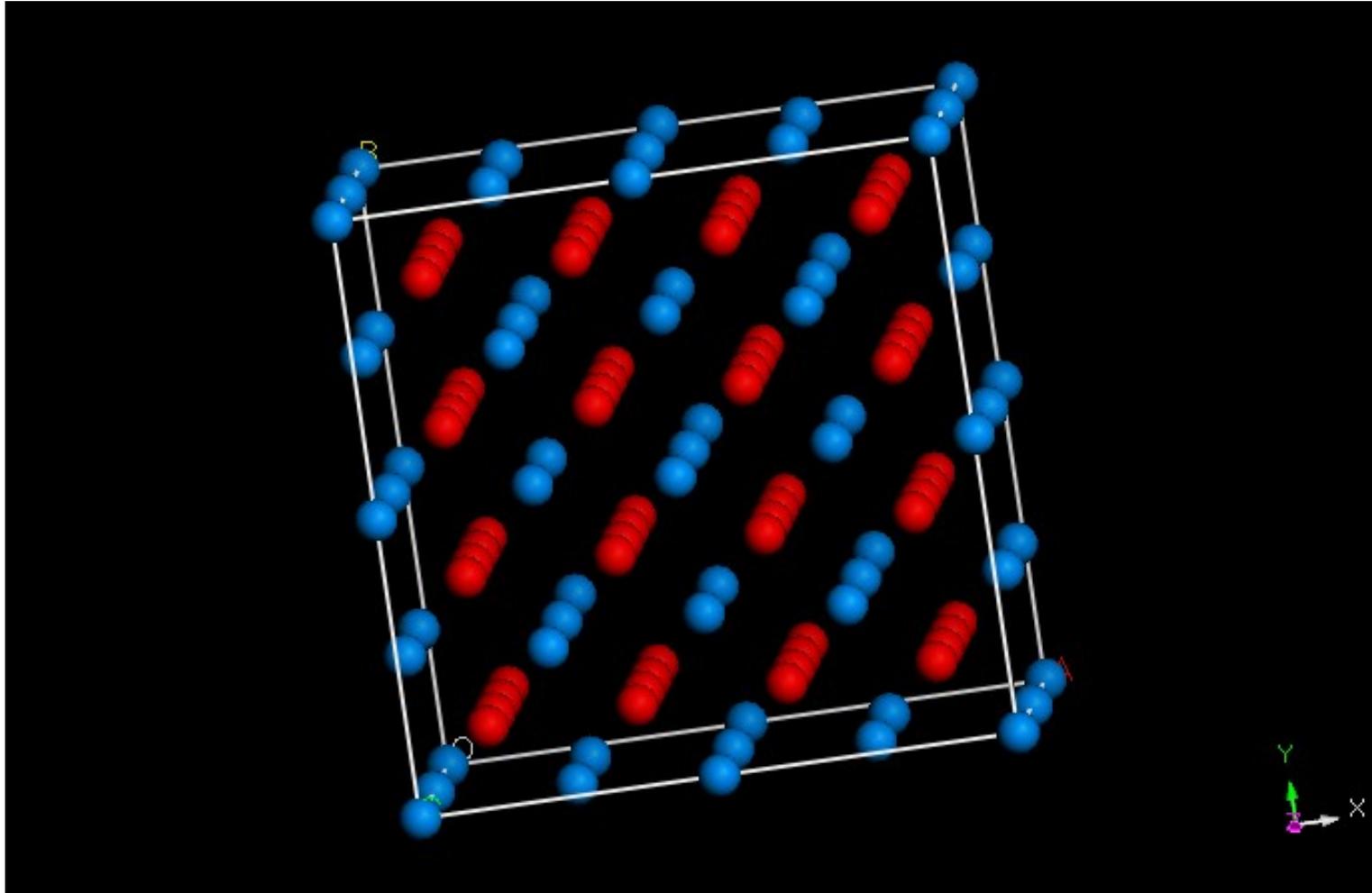
Any property of a system of many interacting particles can be represented as a functional of the ground state density.

- Scalar function of the ground state density determines all information in the many-body wavefunctions for the ground state (and in principle, all excited states)
- Can solve Schrodinger equation for 3 variables instead of for  $3N$  variables
- DFT does not specify how to formulate these functionals!!

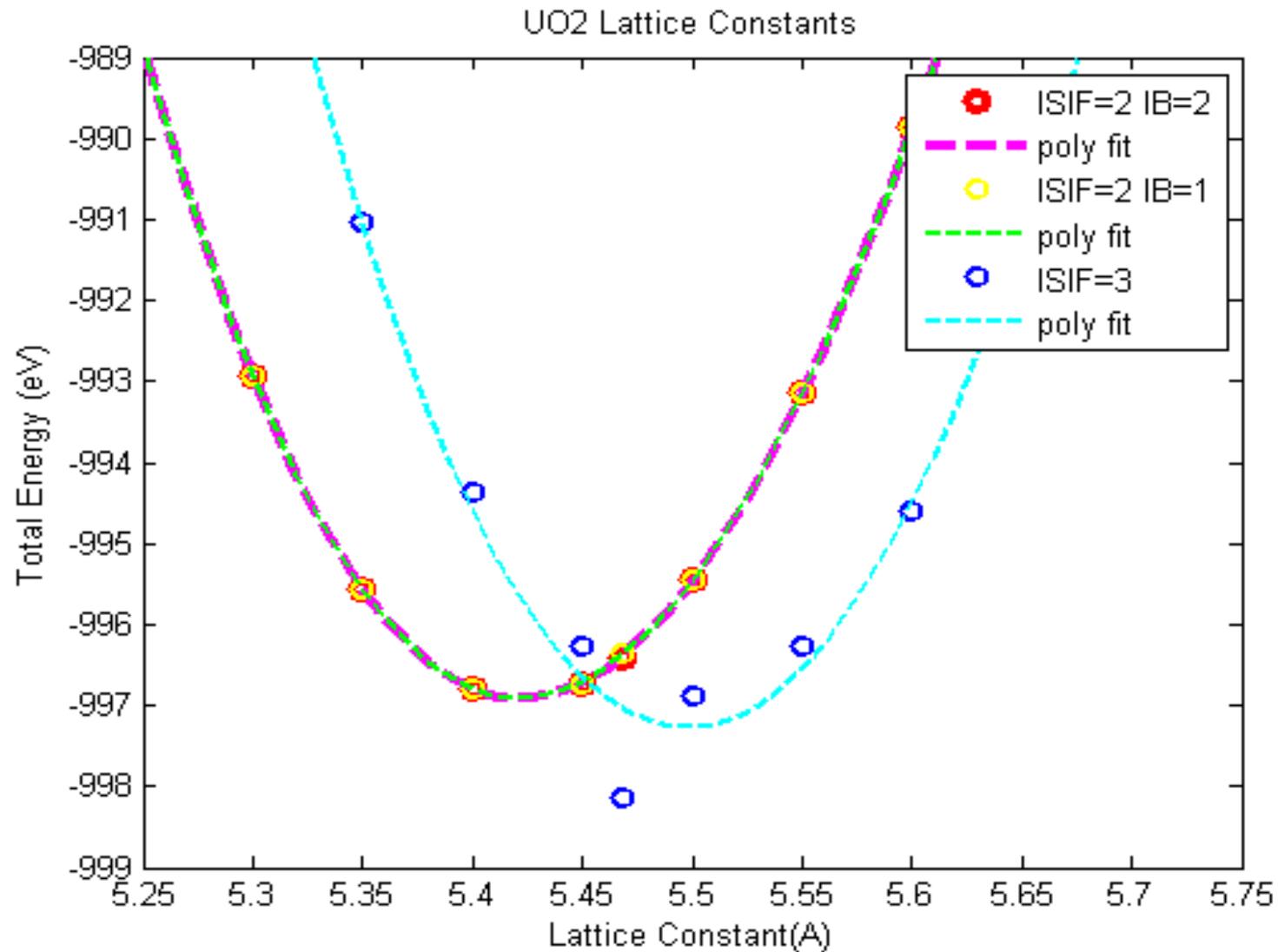
# Computational Methods

- VASP
  - plane-wave pseudo-potential code
    - treats core electrons as inert
    - calculations done on valence electrons only
  - uses PAW potentials considered best available
- RSPt
  - all-electron code (LAPW)
  - FP-LMTO
  - advantage: can massage potentials to work with f-electrons that may penetrate core
  - disadvantage: settings can be difficult to choose for convergence
- codes are complementary
- exchange-correlation functionals: LDA, GGA

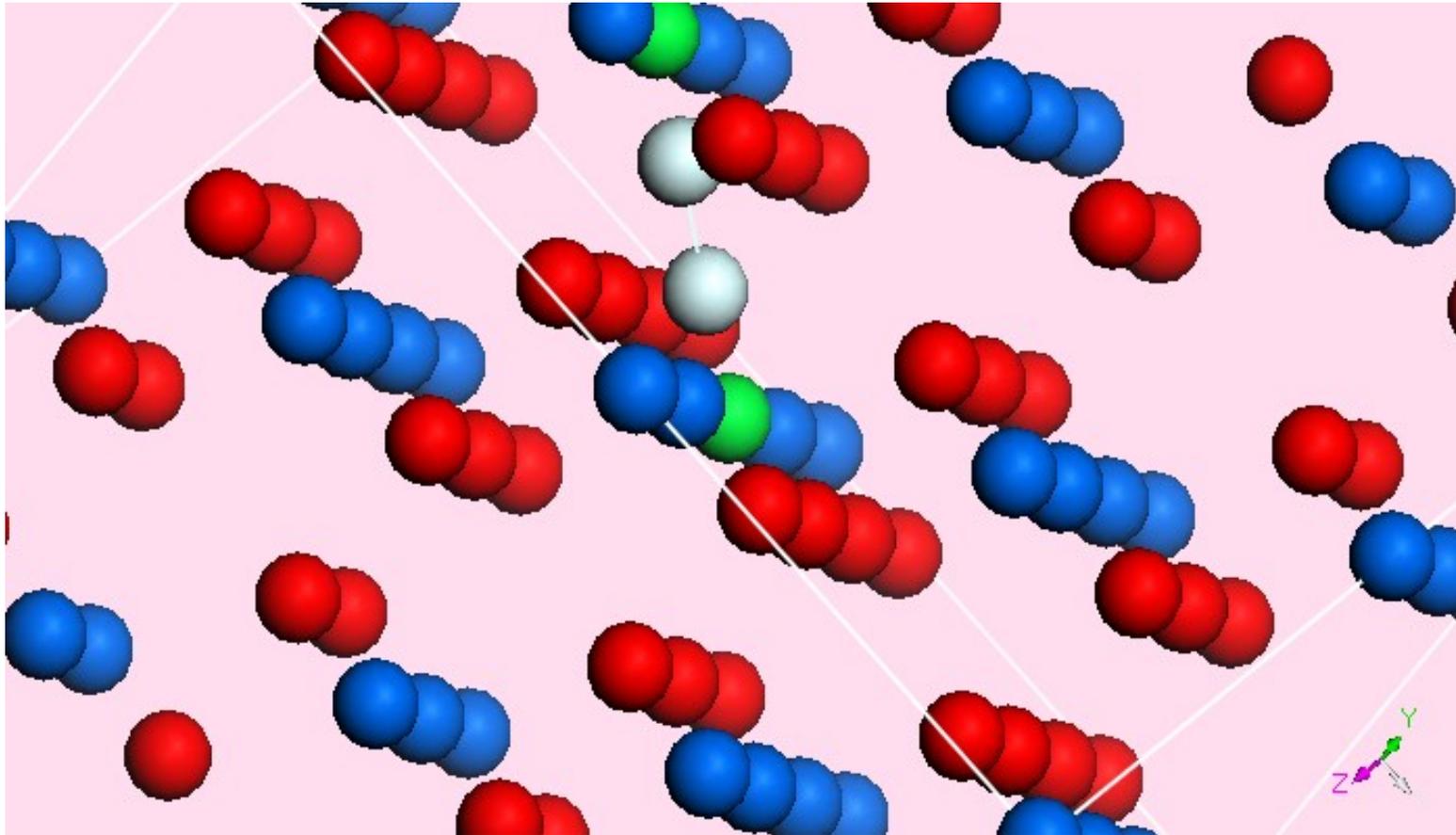
# 2x2x2 UO<sub>2</sub> Supercell



# UO<sub>2</sub>: Lattice Constant Calculations



# Possible U(Pu) Vacancy Path



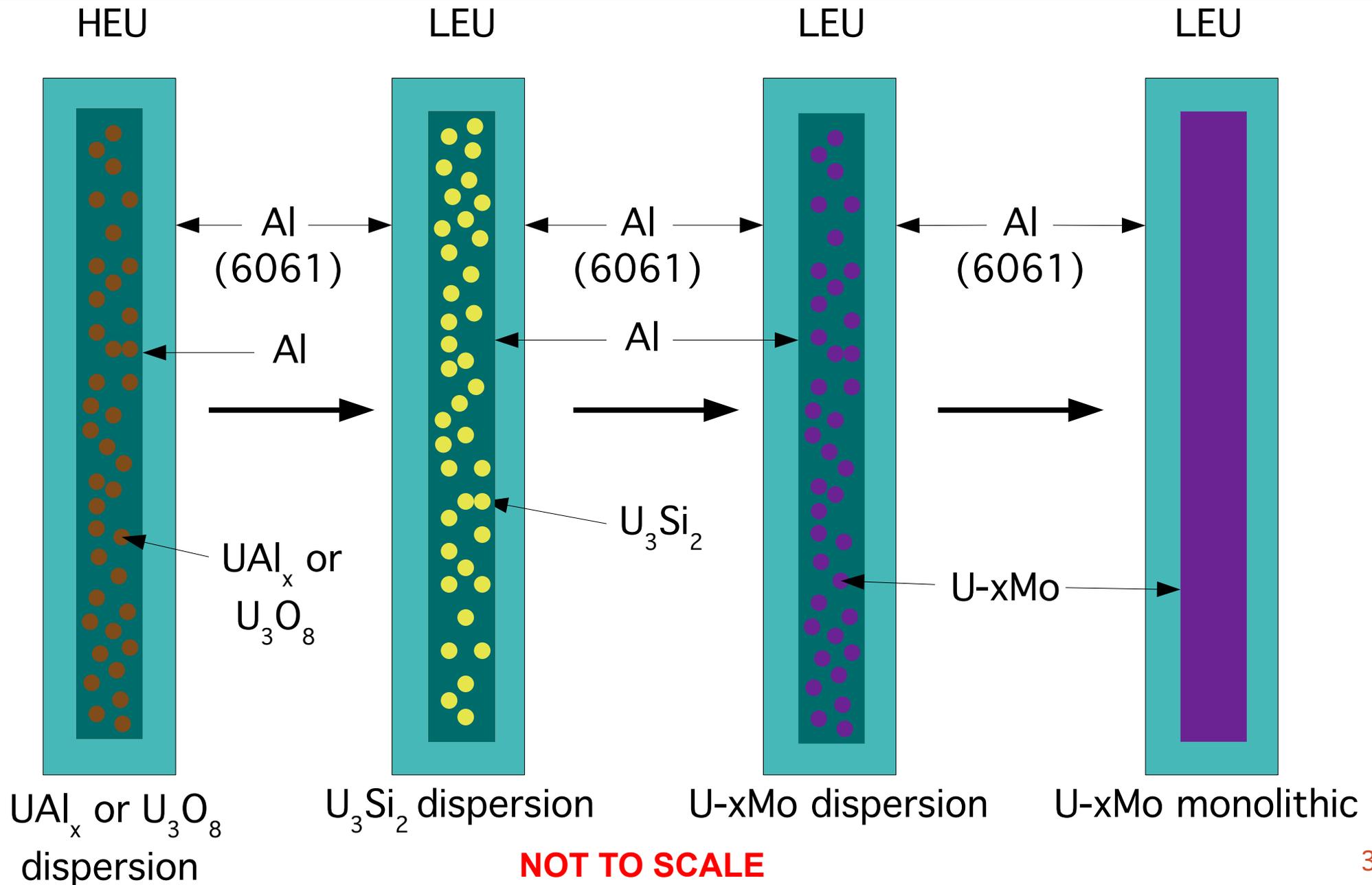
# Future Work

- change number of ionic iterations
- use output from previous calculations for refinement
- compare results from different functionals
- set up different paths
- do same calculations with RSPt
- determine activation energies, compare to literature
- add more vacancies, use different defects
- add strain
- repeat with Kr

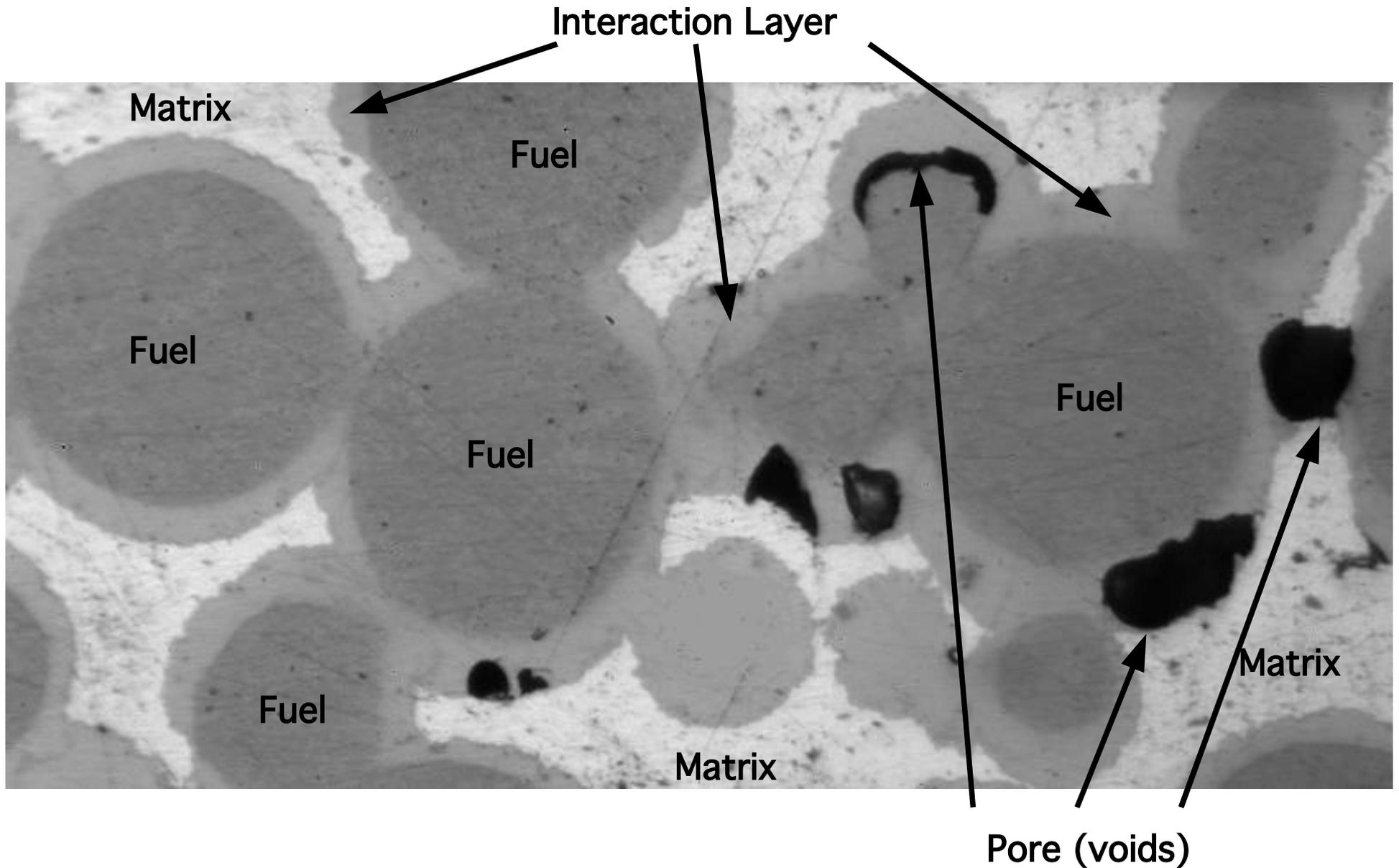
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# MTR-type Reactor Fuel Evolution



# Irradiated U-xMo Fuel Microstructure



# Fuel Performance Modeling

- Qualifying a new reactor fuel and gaining NRC approval is a complex, many year (think decades) process
- One key aspect of this process is developing fuel performance codes to predict fuel behavior as a function of time, temperature and burn-up
  - Fuel performance codes are, in some ways, the ultimate repositories for what we know about a fuel
- Each major fuel type has an associated fuel performance code(s)
  - LWR – FRAPCON
  - TRISO – PARFUME, et al.
  - RERTR – PLATE, MAIA, et al.

# PLATE Fuel Performance Code

- Plate Lifetime Accurate Thermal Evaluation
- Developed at the Idaho National Laboratory
- Uses empirical correlations for the growth of the interface layers with time and burnup
- Effective thermal conductivity of the fuel is predicted by a simple analytical model for multiphase material
  - Modified Hashin & Shtrikman equation

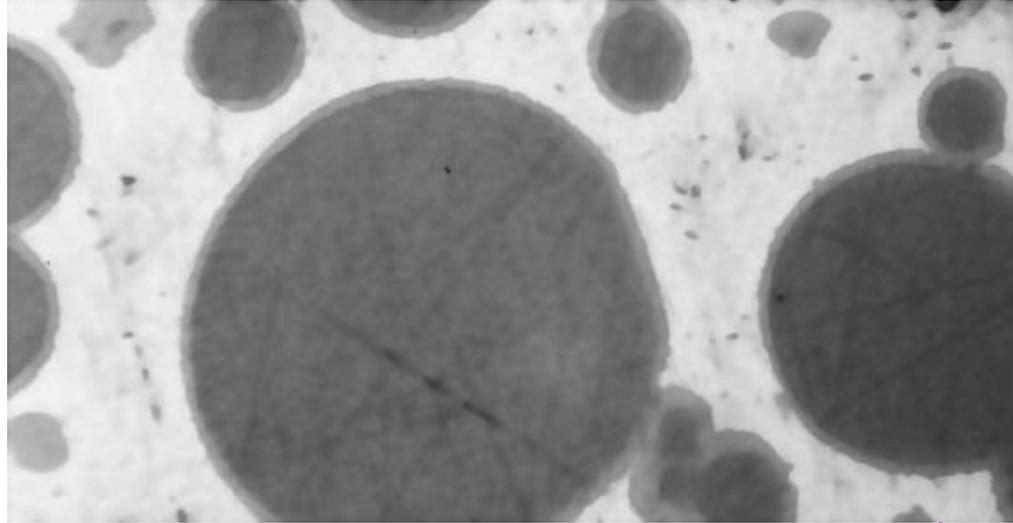
# Current PLATE Assumptions

- Fuel particles are perfectly spherical & interaction layer formed around particle is uniform
- Model doesn't take into account packing arrangement of fuel particles in matrix
- **Validation of effective thermal conductivity model is warranted**

# Micro-structural FEA Technique

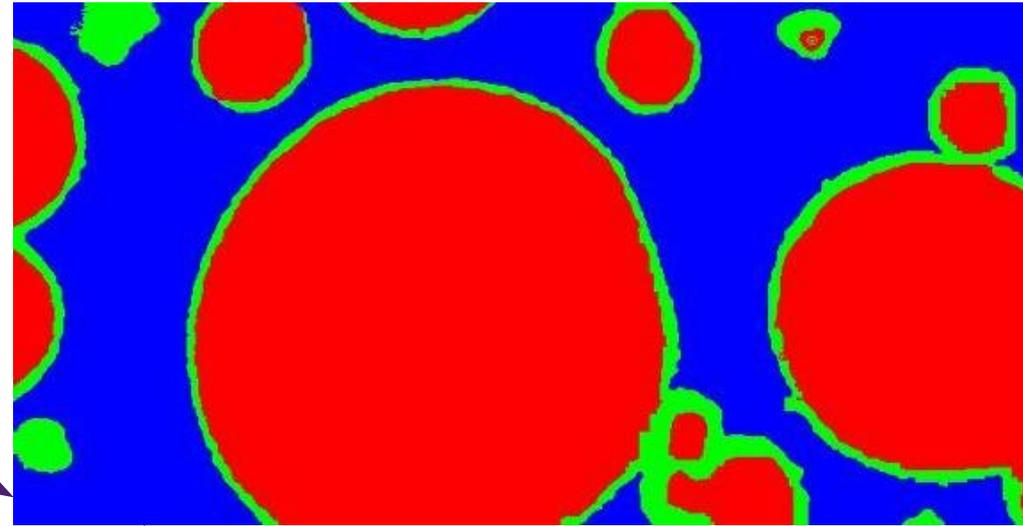
- Based on the Object Oriented Framework (OOF2)
  - Developed by NIST (open-source)
  - Based on open-source tools available in linux or OS X environments
- Converts a 2-D image to an FEA mesh
- Resulting mesh can be analyzed with the built-in solver or exported to a more advanced FEA package (e.g. ABAQUS)

# Validation Flowpath

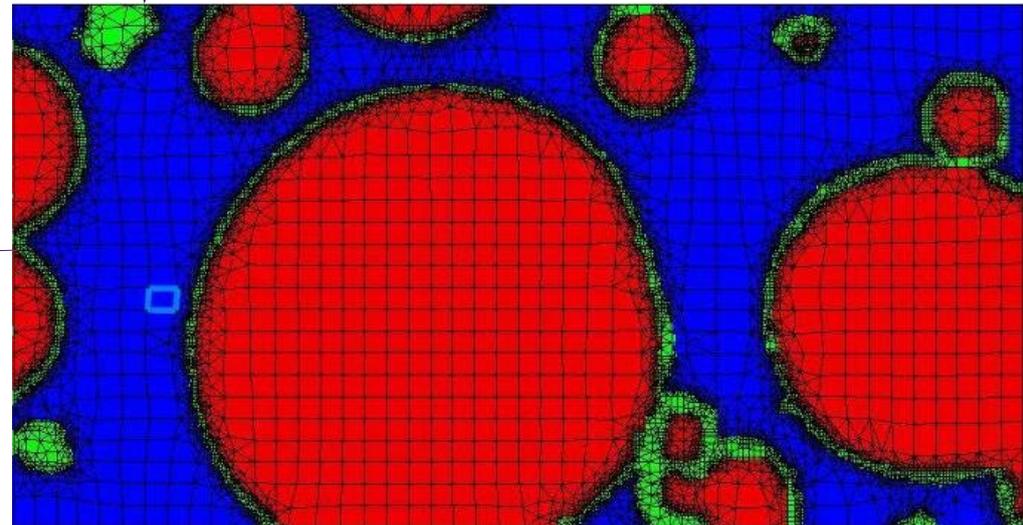


Original image (optical micrograph)

Processed Image (GIMP)



FEA Mesh (OOF)



$$V_c = ?$$

(based on pixel count)

$$k_{\text{FEA-x}} = ? \text{ W/m-K}$$

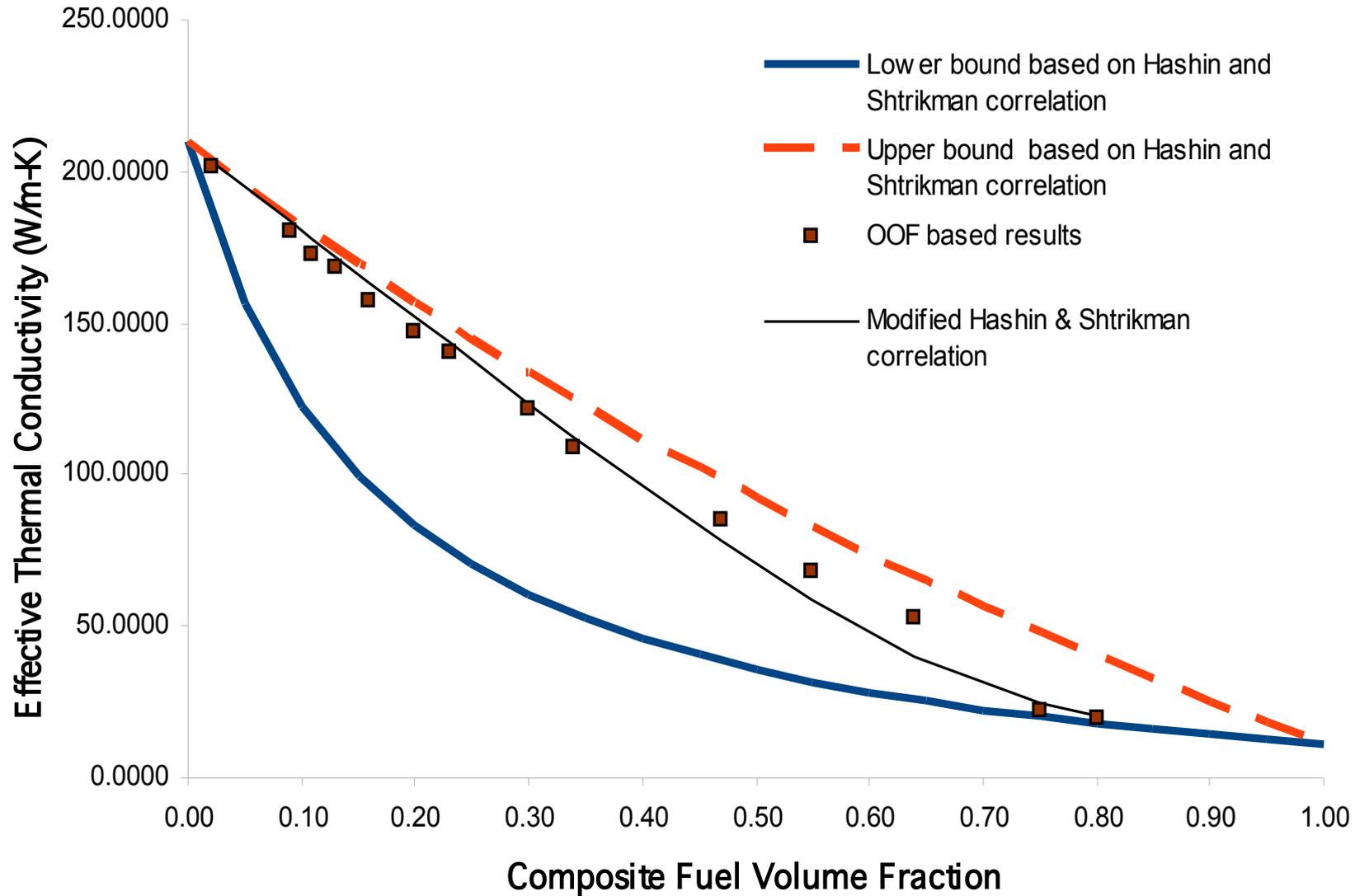
$$K_{\text{FEA-y}} = ? \text{ W/m-K}$$

(OOF-FEA)

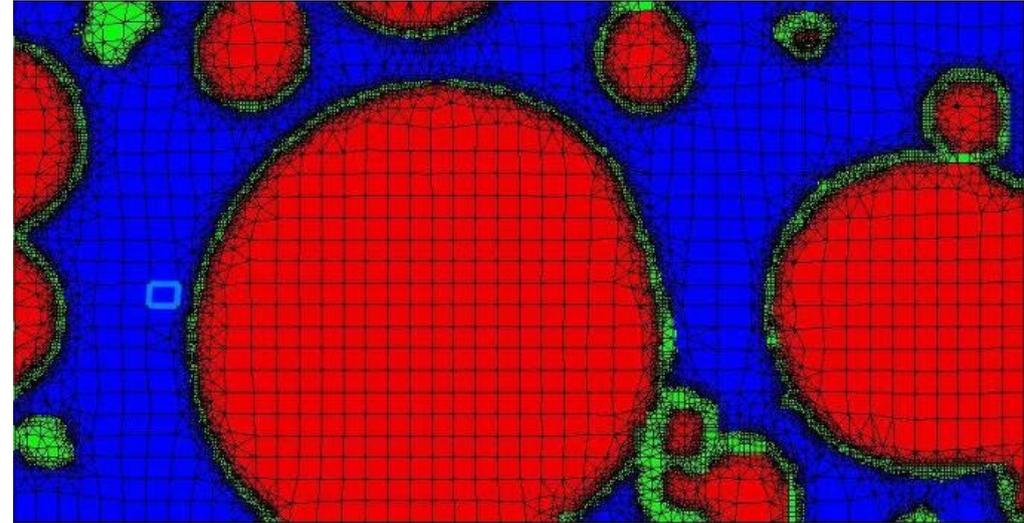
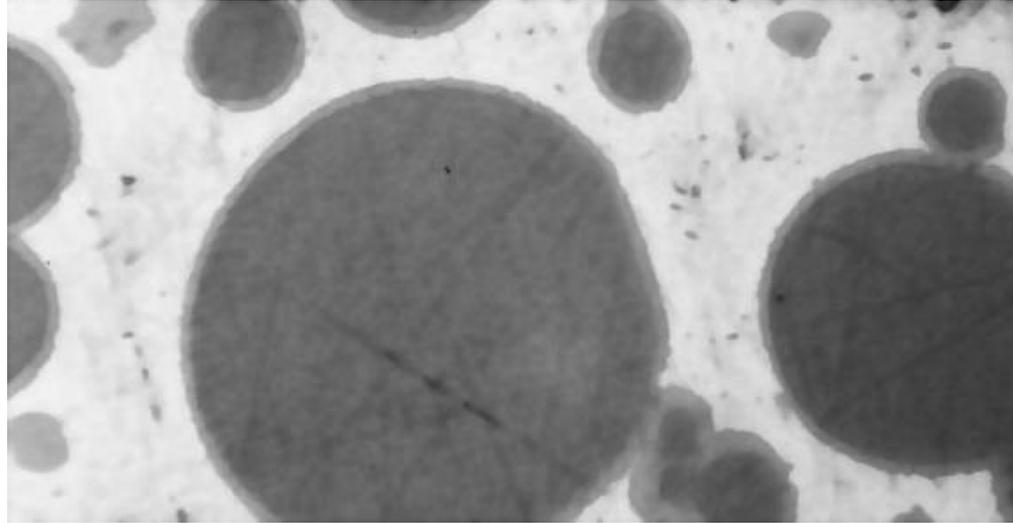
$$K_{\text{H\&S}} = ? \text{ W/m-K}$$

(modified H & S equation)

# Prediction Comparison



# 6-V1R010 Micrograph



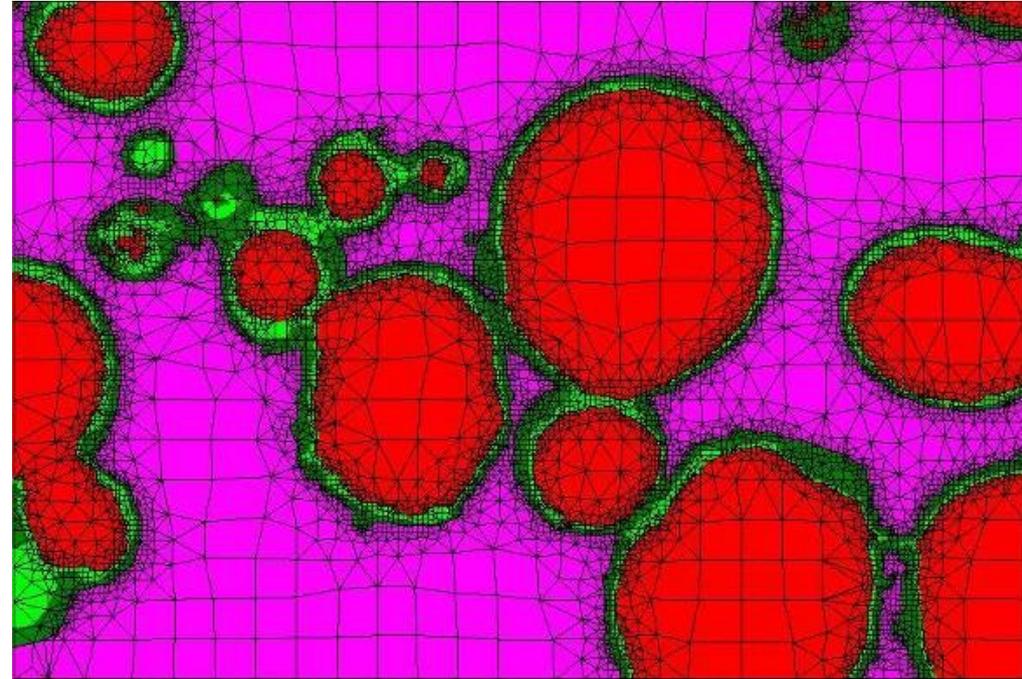
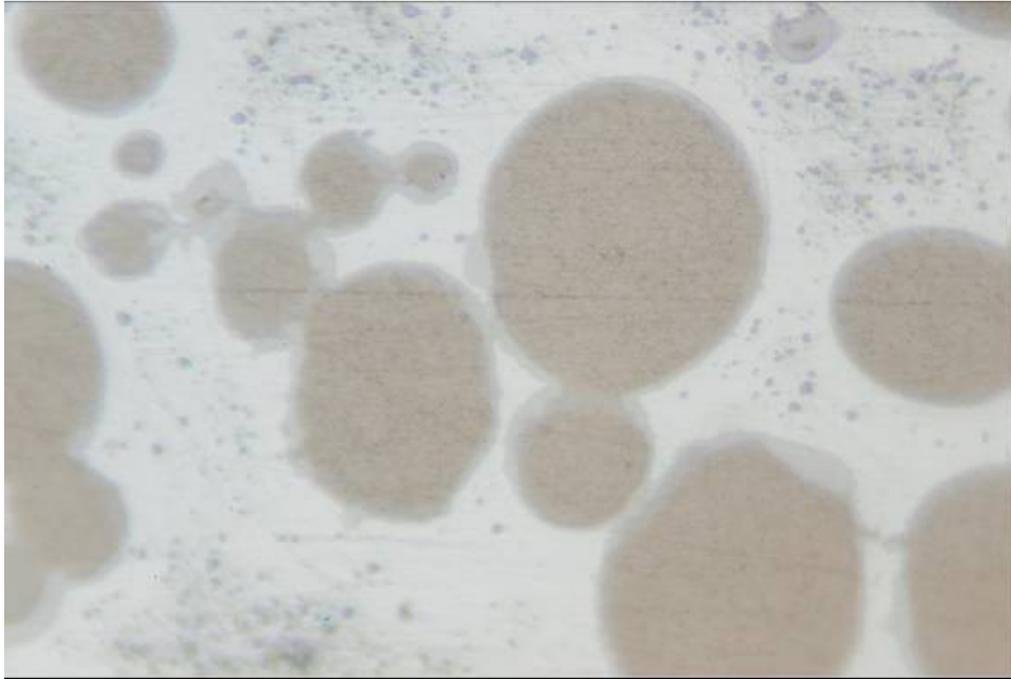
$$V_c = 0.62$$

$$k_{\text{FEA-x}} = 41.9 \text{ W/m-K}$$

$$k_{\text{FEA-y}} = 43.1 \text{ W/m-K}$$

$$k_{\text{H\&S}} = 44.2 \text{ W/m-K}$$

# 6-R3R030 Micrograph



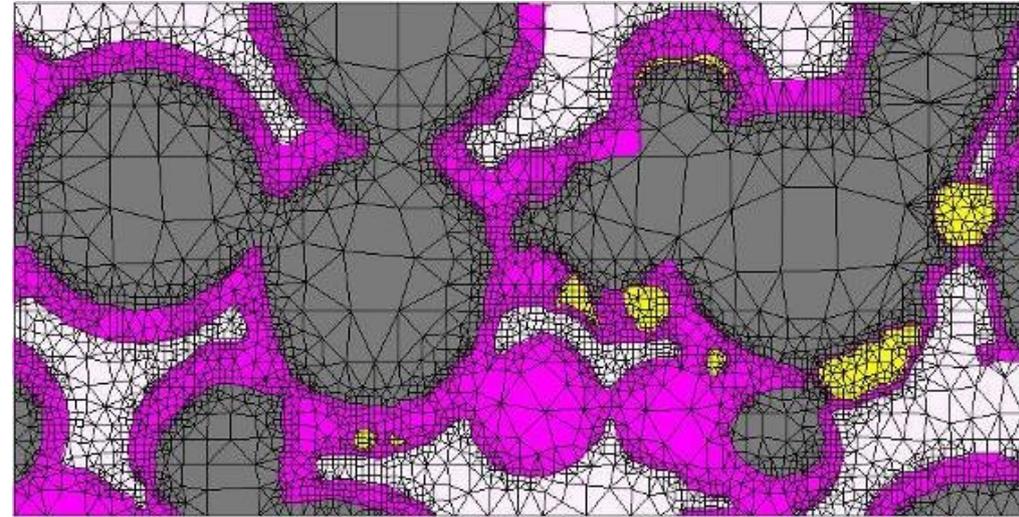
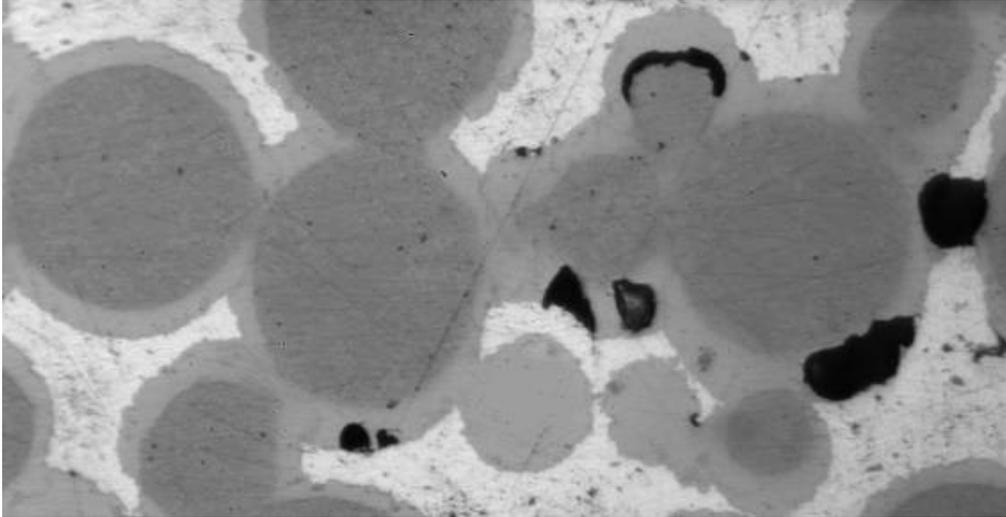
$$V_c = 0.51$$

$$k_{\text{FEA-x}} = 54.2 \text{ W/m-K}$$

$$K_{\text{FEA-y}} = 47.8 \text{ W/m-K}$$

$$K_{\text{H\&S}} = 66.7 \text{ W/m-K}$$

# 6-V1R010 Micrograph (with pores)



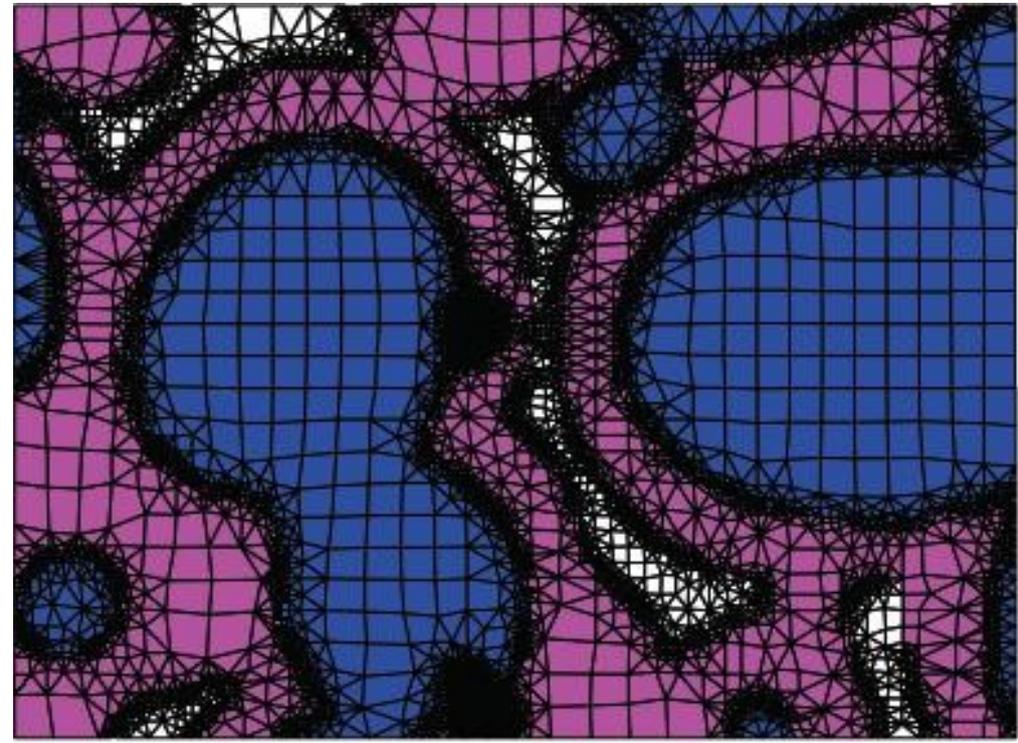
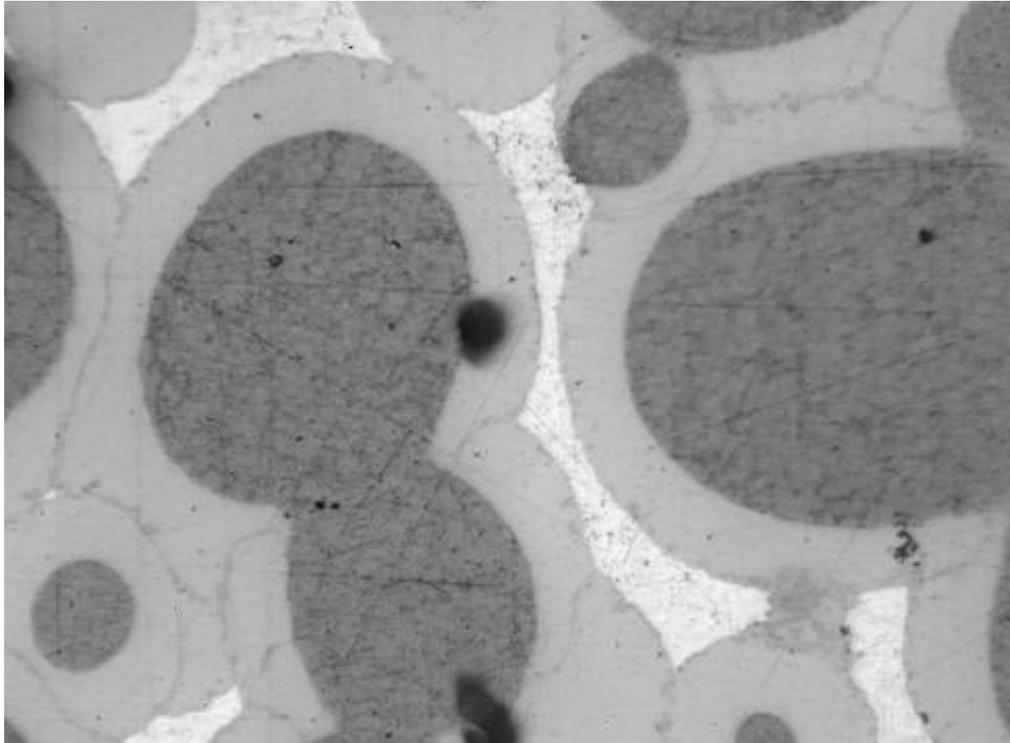
$$V_c = 0.78$$

$$k_{\text{FEA-x}} = 26.7 \text{ W/m-K}$$

$$K_{\text{FEA-y}} = 27.1 \text{ W/m-K}$$

$$K_{\text{H\&S}} = 24.8 \text{ W/m-K}$$

# 6-R5R020 Micrograph (with pores)

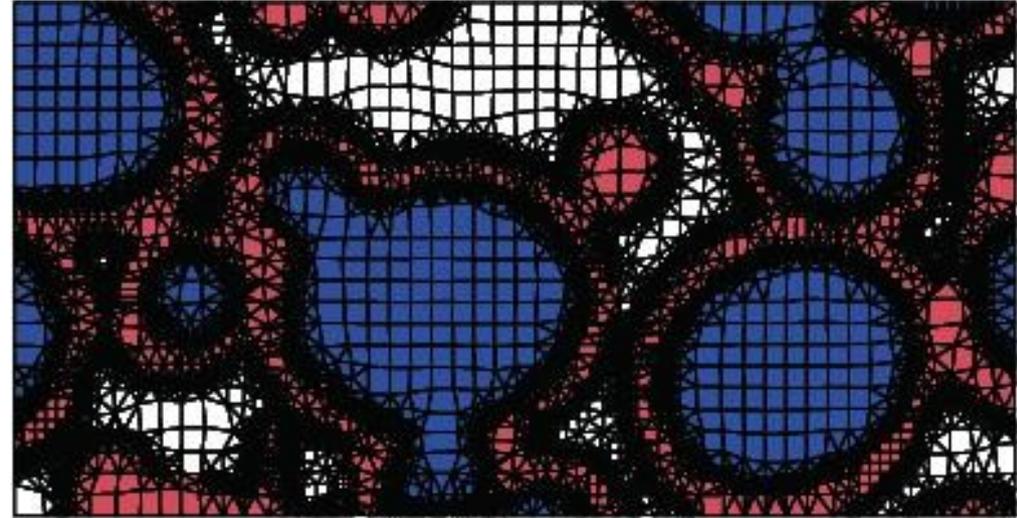
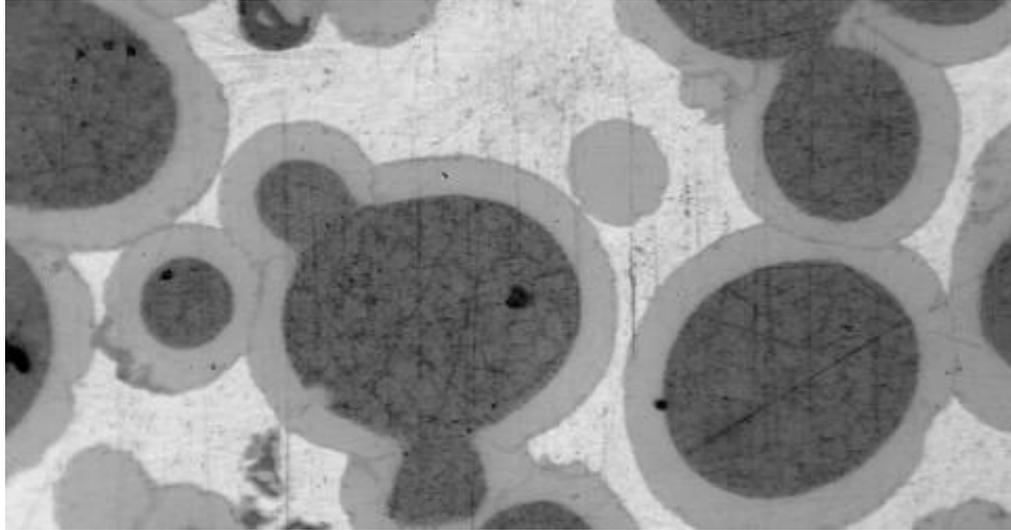


$$V_c = 0.92$$

$$k_{\text{FEA-x}} = 13.4 \text{ W/m-K}$$

$$K_{\text{H\&S}} = 12.4 \text{ W/m-K}$$

# 6-R5R020-38N Micrograph



$$V_c = 0.77$$

$$k_{\text{FEA-x}} = 20.6 \text{ W/m-K}$$

$$K_{\text{H\&S}} = 19.3 \text{ W/m-K}$$

# RERTR Results

Source of microstructure	Volume fraction ( $V_c$ )	FEA results (W/m-K)			Analytical model results (W/m-K)	Difference (%)	
		x-dir	y-dir	x/y		x-dir	y-dir
6-V1R010 plate	0.62	41.9	43.1	0.97	44.2	+5.2	+2.5
6-R3R030 plate	0.51	54.2	47.8	1.13	66.7	+18.7	+28.3
6-V1R010 plate (with pores)	0.78	26.7	27.1	0.99	24.8	-7.7	-9.2
6-R5R020 plate (with pores)	0.92	13.4	--	--	12.4	+8.1	--
6-R5R020-38N	0.77	20.6	--	--	19.3	+6.7	--

# Observations

- Finite Element Analysis (FEA) of the microstructure of irradiated uranium/molybdenum fuel plates offers the opportunity to validate the results of the PLATE fuel performance code
- The Hashin and Shtrikman correlation used in PLATE assumes a uniform distribution of spherical particles in the matrix
  - The effect of the interface layer is considered in the composite thermal conductivity of the particles
  - Shape and arrangement effects are neglected
- Image-based FEA of actual fuel plates can account for the interface layer directly, as well as including the shape and arrangement effects

# Observations, cont.

- Generally speaking, the thermal conductivity model in the PLATE code produces satisfactory results
  - Some sensitivity to large particles and non-regular particle arrangements
    - Differences of up to 32% for high particle volume fractions
      - Both under- and over-predictions occur
  - At low volume fractions, the effect of pores on the effective thermal conductivity is minor
  - No strong evidence of anisotropy

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