Nuclear Fuel Performance

NE-533

Spring 2025

Housekeeping

- Exam Thursday
- Problem session after lecture
- MOOSE project due 2/28
- Snowstorm coming…
  - Assuming that class is cancelled on Thursday, we will do the exam via zoom
  - Cameras on, I will be a virtual proctor
  - Will submit your scanned or photographed exam and cheat sheet and email them to me

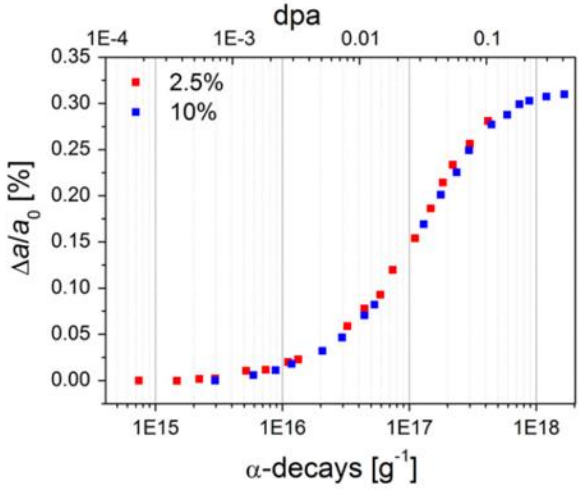
Last Time

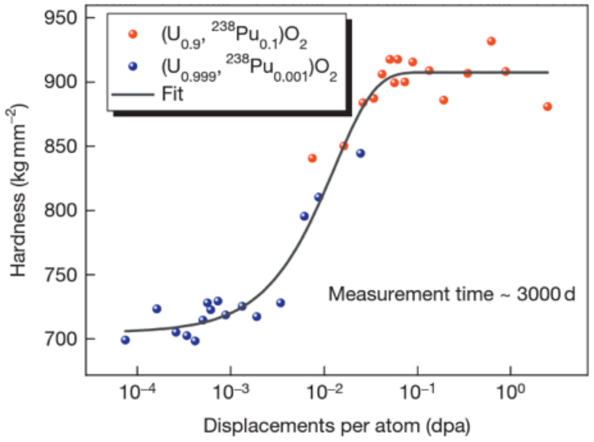
- The crystal lattice is never perfect; it has defects
  - Point defects, dislocations, grain boundaries, voids and precipitates
- Microstructure can be tailored through processing, during fabrication or post-fabrication
- Started UO2 radiation effects
  - interstitial loops and voids
  - changes in thermal conductivity due to irradiation and chemistry changes
  - have various empirical models which take into account phonon and electron kth, as well as burnup

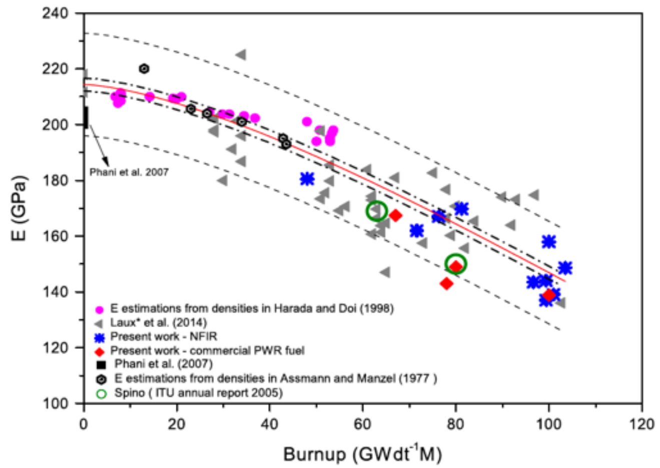
UO2 RADIATION EFFECTS CONTINUED

Other Physical Changes

- An important effect of radiation damage is the volume increase of the crystal lattice, leading to macroscopic swelling of the fuel material
- Alpha particle damage alone can cause approximately 0.4% swelling
- Mechanical properties also vary with damage
- Hardness increases as a function of dpa
- Young’s modulus decreases with dpa

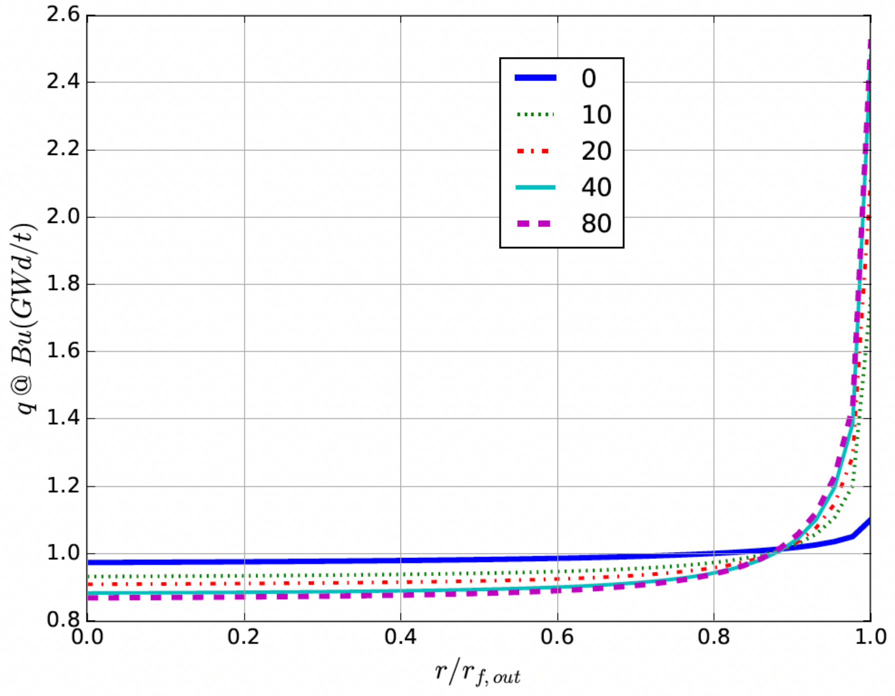






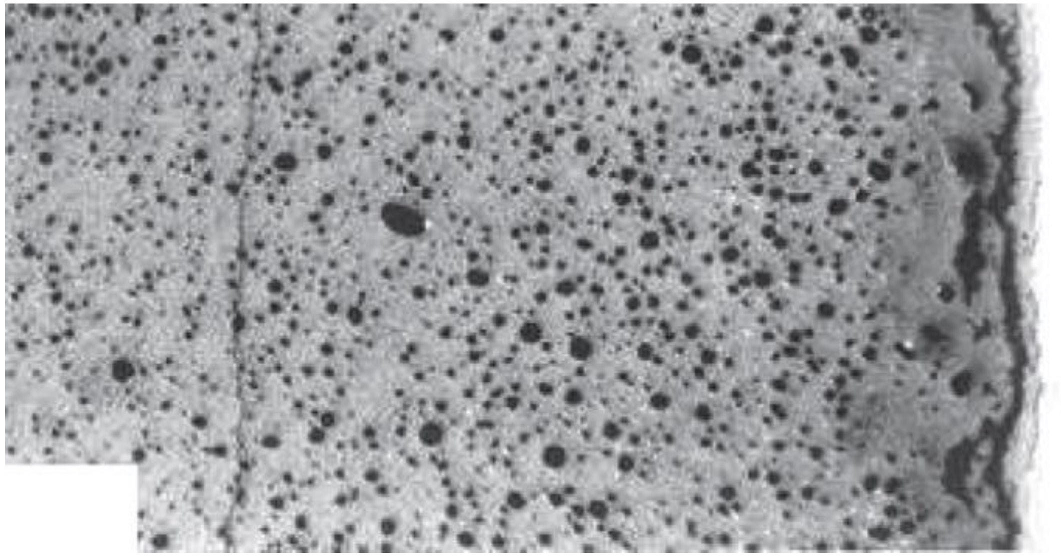
High Burnup Structure (HBS)

- At the beginning of the irradiation, the local power is ~10% higher on the pellet periphery, due to self-shielding effect in 235U fissions driven by neutrons entering the rods after interactions in the water
- With increasing burnup, a strong capture of neutrons by U-238 in the resonance range occurs at the periphery of the fuel
- This leads to the production of Pu-239, and leads to a higher fissile density and thus a further increase in the local power/burnup
- This high local burnup area is on the outer ring of the fuel and is about 200 microns thick, which represents about 8% of the fuel



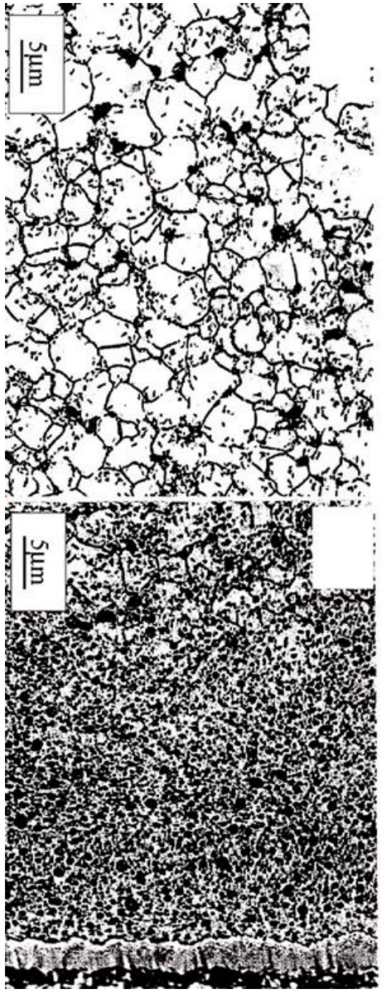
High Burnup Structure (HBS)

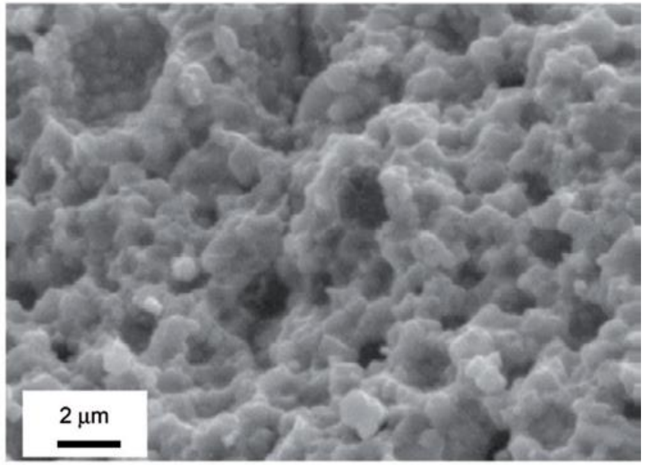
- When thermal recovery is not sufficiently efficient (at lower temperatures), the accumulation of defects at high burnup can lead to the instability of the crystalline structure, initiating a restructuring driven by the energy stored in the material
- This can be amorphization (not observed in UO2) or recrystallization/polygonization
- Polygonization is the rearrangement of dislocations into sub-domain walls with slightly misaligned grains
- In UO2, grains subdivide from 10 microns in size to 100-200 nm size, and a densely porous structure is formed (~20% porosity)



High Burnup Structure (HBS)

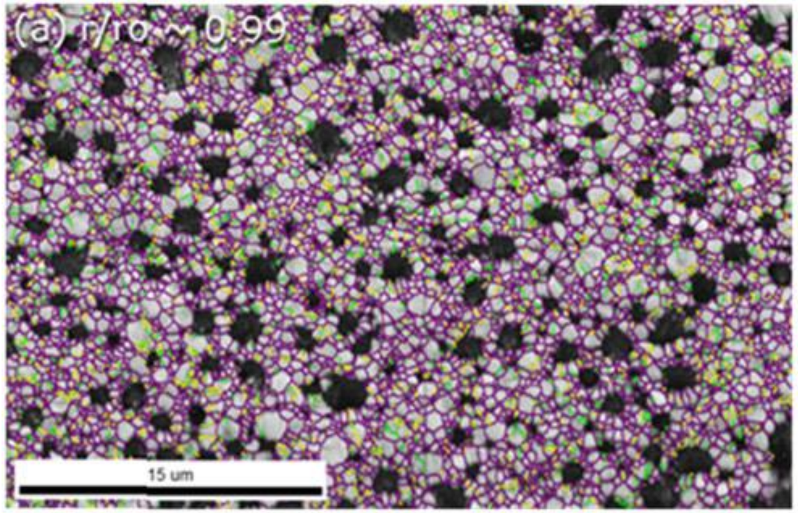
- The increase of the relative porosity volume degrades the material conductivity and reduces the mean grain size
- On the other hand, the intragranular irradiation defect-cleaning improves the fuel intrinsic thermal conductivity
- Fission gas in HBS bubble is retained, not released
- Exact mechanism of HBS formation is not fully understood, believe to be polygonization, interacting with fission gas bubbles, dislocation growth, and high levels of energy deposition



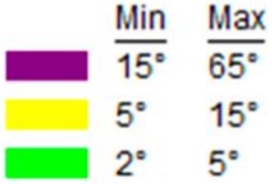


High Burnup Structure (HBS)

- HBS has a high fission gas retention capacity, and does not evolve toward an open system of interconnected channels, even when porosity reaches very high values
- Thus, the HBS is a relatively stable structure, limiting pressure increase in the plenum
- While burnup is higher in the rim region, the HBS polygonization removes/sweeps defects to new grain boundaries, removing some phonon scatterers
- Despite high porosity, thermal conductivity increases, slightly decreasing centerline temperatures in high burnup fuel



2 - 5° LAGBs shown in green,
5 - 15° LAGBs in yellow,
15 - 65° LAGBs in purple



Radiation Effects Summary

- Formation of loops of voids as primary defect clusters
- Radiation effects include both chemistry and physical property changes
- One of the key physical properties modified is the thermal conductivity
  - have several empirical (and mechanistic) models to account for burnup and temperature
- HBS forms towards the end of life, generating a fine-grained structure with high porosity
- HBS retains fission gas and slightly increases thermal conductivity

MECHANISTIC MODELING

Microstructure-based fuel performance modeling

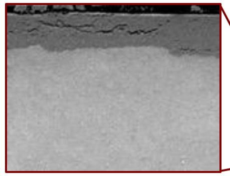
Structure/property relationships connect the microstructure variables to the property values

Operating Conditions: Neutron flux | Coolant T&P

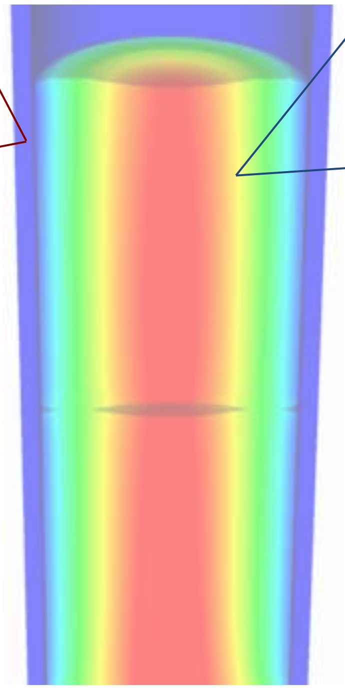
Variables: Temperature | Displacement | Stoichiometry

Cladding Property values
- Elasticity tensor
- Yield stress
- Thermal conductivity
- Failure criteria
- Dimensional change

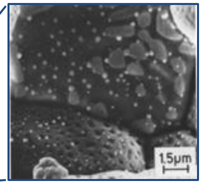
Microstructure evolution models
- Grain growth
- Plasticity
- Defect prod. & transport
- Hydrogen transport
- Loop evolution



Cladding Microstructure variables
- Average grain size
- Dislocation density
- H concentration
- Hydride fraction
- Point defect concentrations
- Loop density
- 2^{nd} phase particle fraction



Gap variable
- Gap fission gas concentration



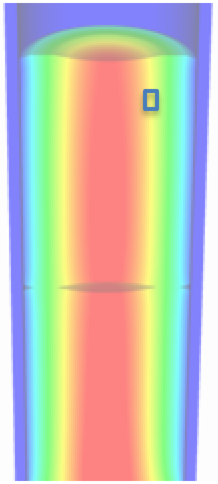
Fuel Microstructure variables
- Average grain size
- Dislocation density
- U defect concentrations
- Pu concentration
- Fission product precipitate fractions
- Dissolved fission products
- Fission gas bubble fraction
- Grain boundary fission gas bubble coverage
- Crack parameter

Microstructure evolution models
- Grain growth
- Plasticity
- Defect prod. & transport
- Fission gas behavior
- Densification
- Fracture

Fuel Property values
- Elasticity tensor
- Thermal conductivity
- Fracture stress
- Melting temperature
- Diffusion constants
- Dimensional change

Example: fission gas behavior in the fuel

- Take into account a finite set of variables to describe the state of the material
- Utilize a mechanistic model of fission gas behavior to predict the evolution of the microstructure
- Utilize this updated microstructure to inform a number of structure/property relationships



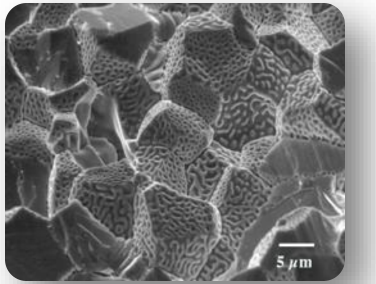
Variables

Temperature

Displacement

Stoichiometry

Model of fission gas behavior



- Dissolved fission products
- Fission gas bubble fraction
- Grain boundary fission gas bubble coverage
- Gap fission gas concentration

Structure/property relationships

Elasticity tensor

Thermal conductivity

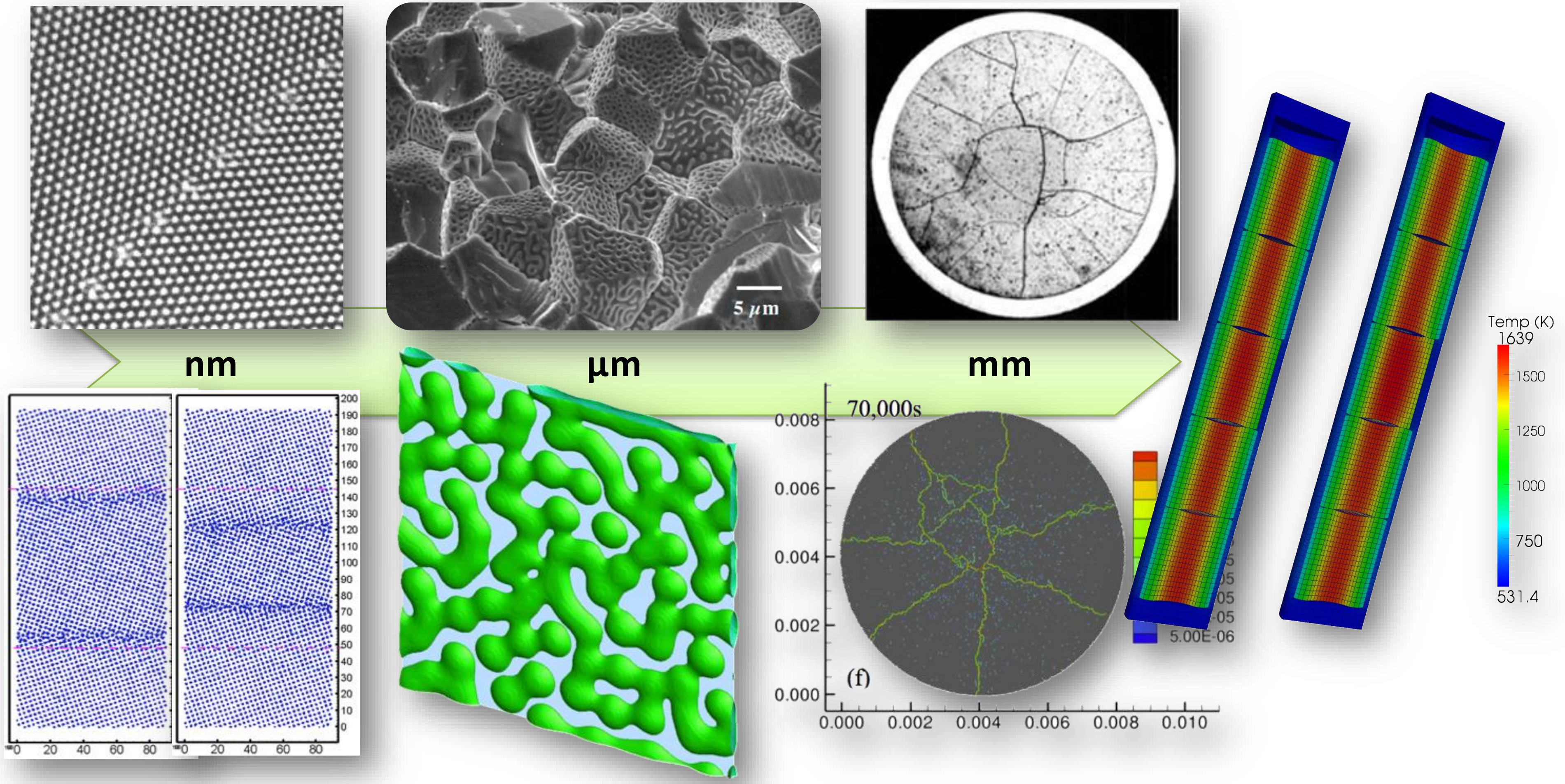
Fracture stress

Dimensional change

Gap conductance

Gap pressure

Multiscale separate effects experiments and simulations inform the development of the models



Microstructure-based models

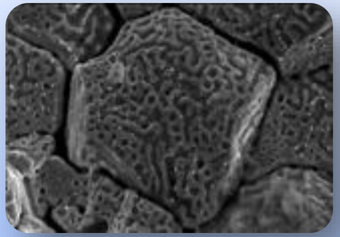
- Can provide a structure/property relationship to replace the existing burnup dependent model
- For example, thermal conductivity, taking into account microstructural features and their evolution

Grain boundary and bubbles

Intragranular porosity

Precipitated fission products

$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$



Bulk conductivity

Vacancies and interstitials

Fission gas

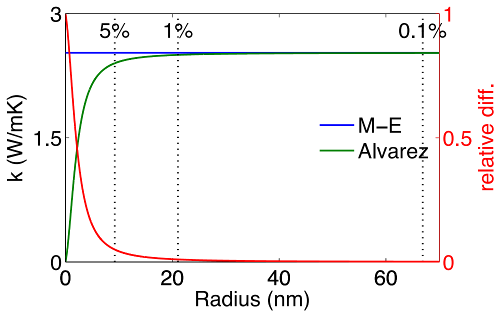
Parametrizing the mechanistic model

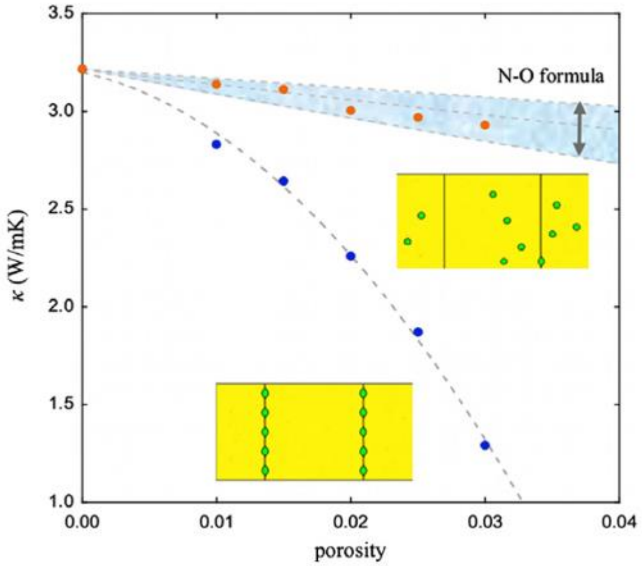
- We employ multiscale modeling and simulation to determine the various parameters for the model
- MD simulations conducted at LANL have been used to determine the coefficients for various point defects
- MD simulations have shown that phonon scattering must be accounted for to accurately represent small bubbles
- Mesoscale simulations have shown that GrB bubbles have a larger impact on the thermal conductivity

Defect	a_i	Defect	a_i
O interstitial	12.63	Xe atom	33.9
O vacancy	21.74	La atom	3.97
U interstitial	29.98	Zr atom	2.23
U vacancy	23.78	Pu atom	0.08

Maxwell-Eucken (no phonon scattering)

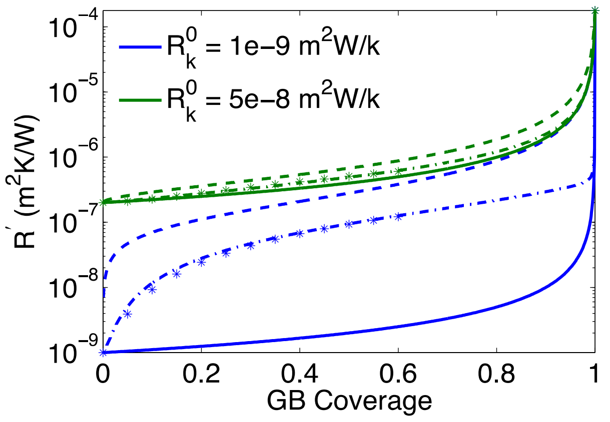
$$\kappa_{ME} = \frac{1-p}{1+p/2}$$



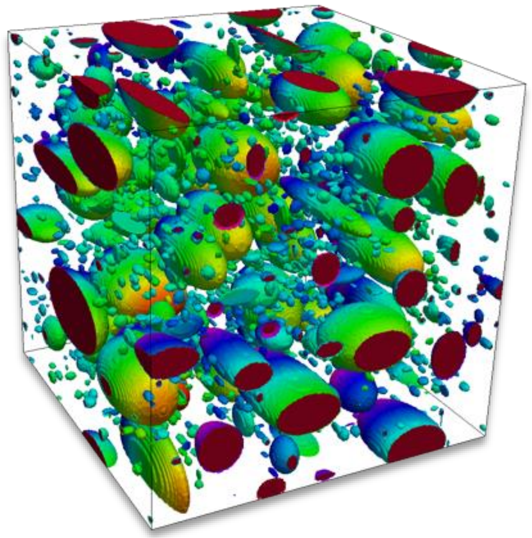


Parametrizing the mechanistic model

- A thermal resistor model is created to describe the impact of GrB bubbles on the thermal conductivity



- MARMOT simulations are currently being used to inform the development of the precipitate multiplier



Parametrizing the mechanistic model

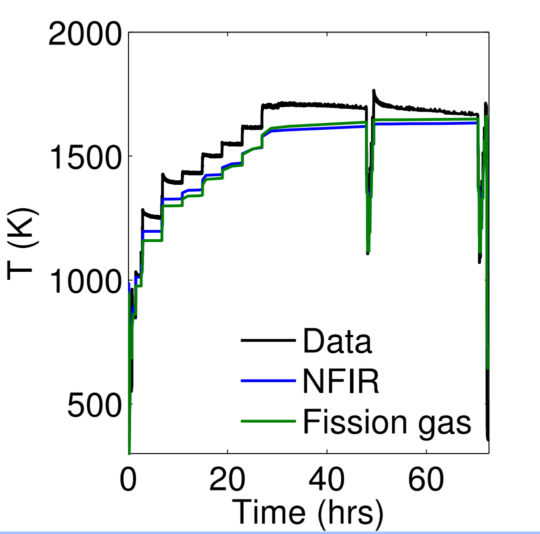
- Each term in the expression must be coupled to a corresponding state variable
- The full model calculates the thermal conductivity as a function of:
  - Temperature
  - Point defect concentrations
  - Intragranular bubble density and average radius
  - Fractional coverage of bubbles on GBs and average radius
  - Precipitate volume fractions and average sizes
- Currently effects of precipitate fission products and individual point defects are neglected in the model, as they are not tracked or predicted in BISON

$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_g c_g}$$

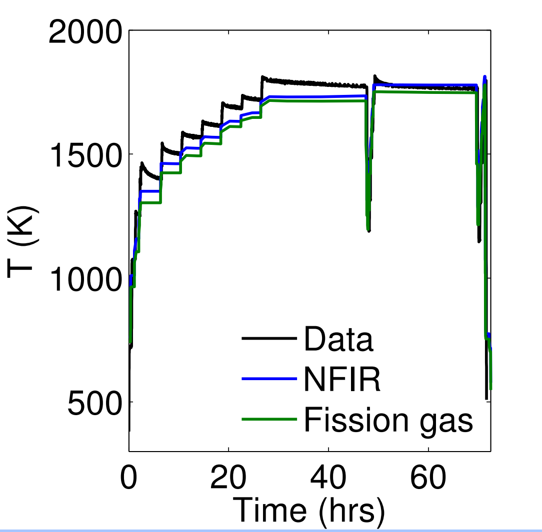
Comparing with experiments…

- The model under-predicts the temperature in most cases, but not all
- Thus, the model is neglecting some resistive effects from the microstructure
- Generally performs as well, and in some cases better, than the burnup based model

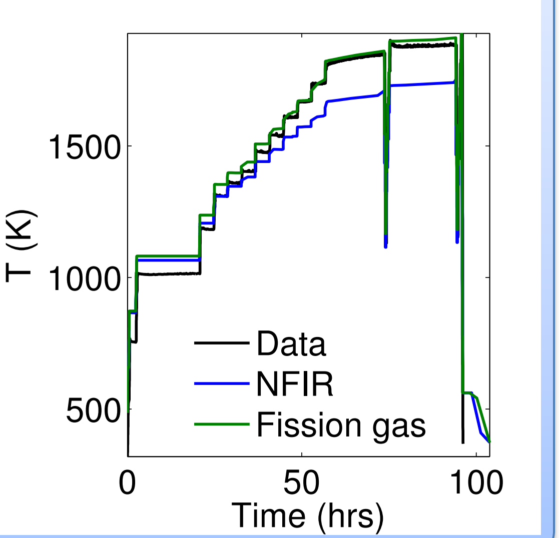
RISO-3 AN3



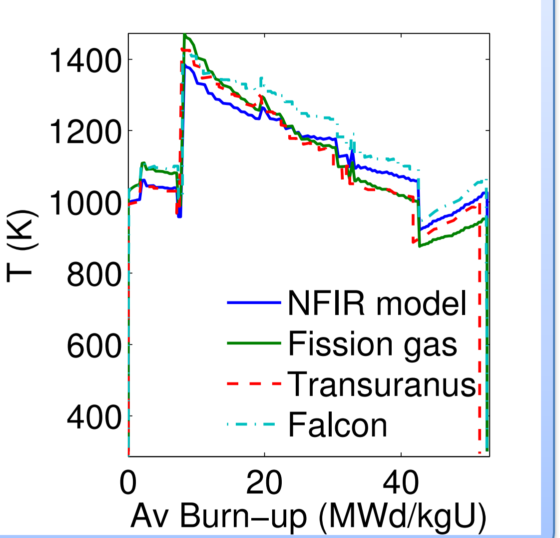
RISO-3 AN4



RISO II-3



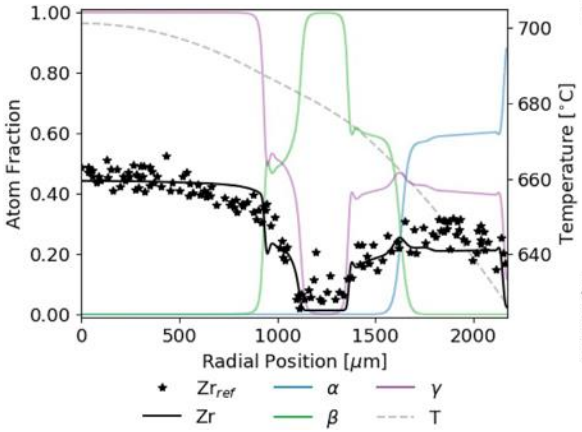
US PWR TSQ0002

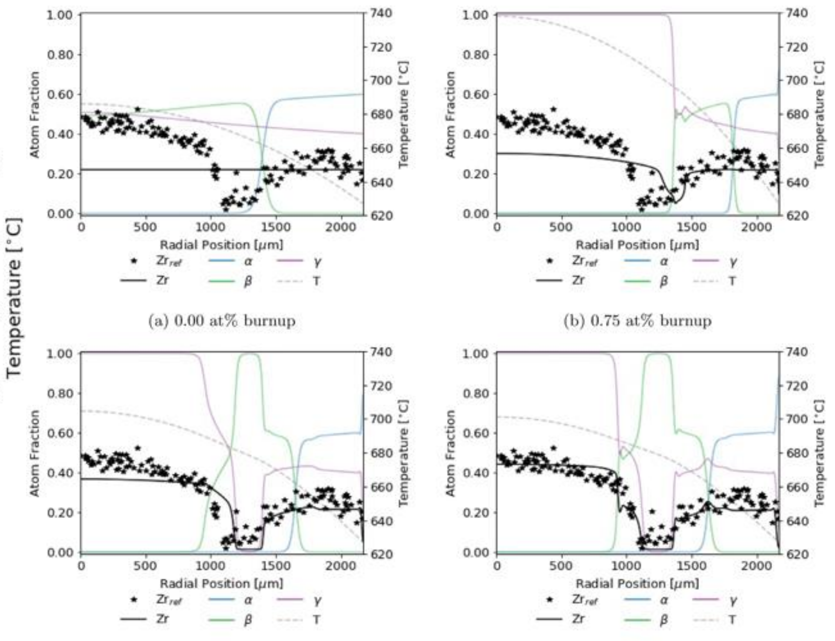


UZr Fuel Performance Modeling

- Development of a quantitative phase-field model of macroscale constituent redistribution in the U-Zr system, where model parameters were optimized, and the model validated against an independent data set

- Calculation of surface tension based on molecular dynamics, which is used in the BISON gaseous metallic fuel swelling model



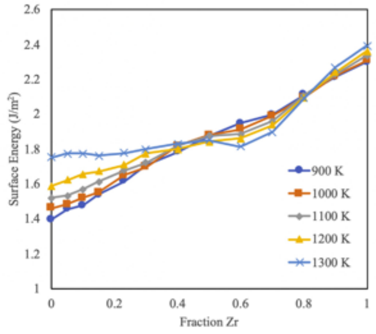


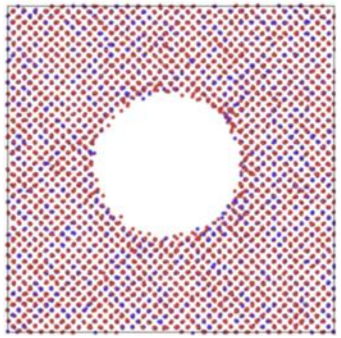
(a) 0.00 at% burnup

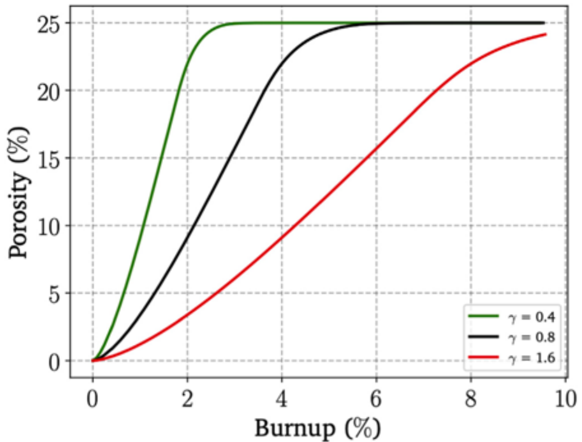
(b) 0.75 at% burnup

(c) 1.50 at% burnup

(d) 3.60 at% burnup

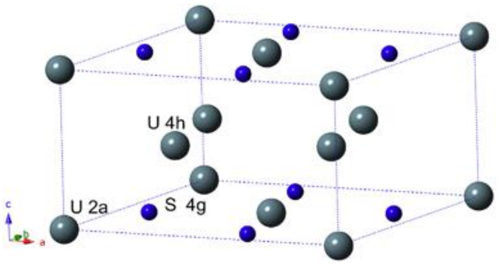


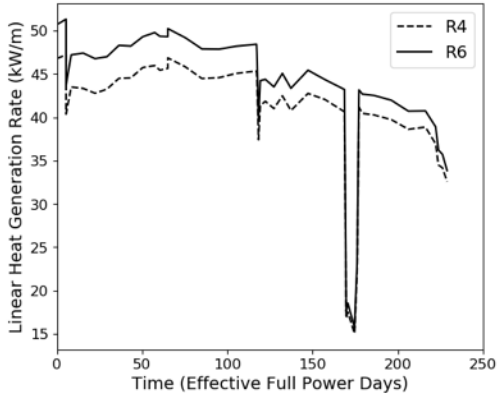


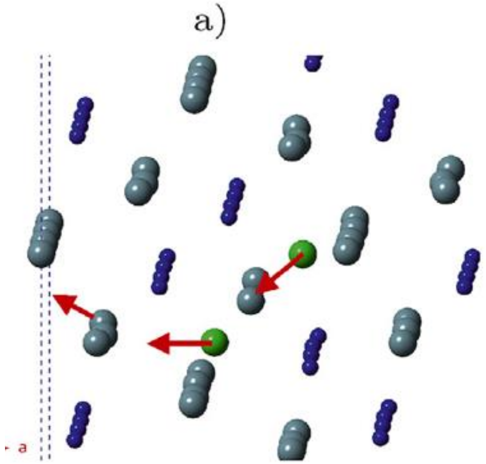


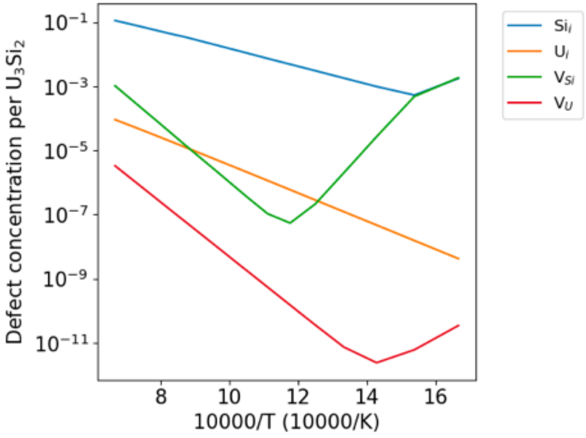
U3Si2 Fuel Performance Modeling

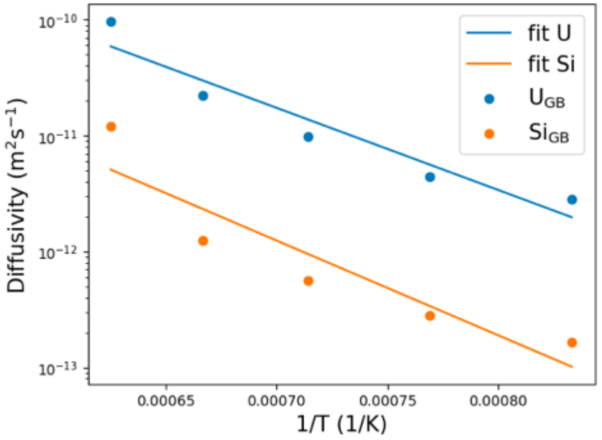
- Point Defect Diffusion, Fission gas swelling, thermal and irradiation creep

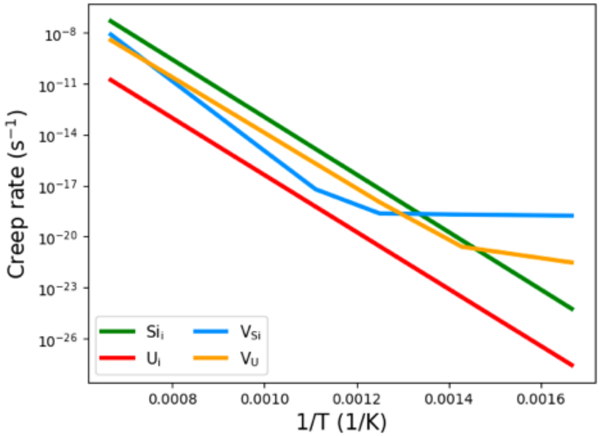


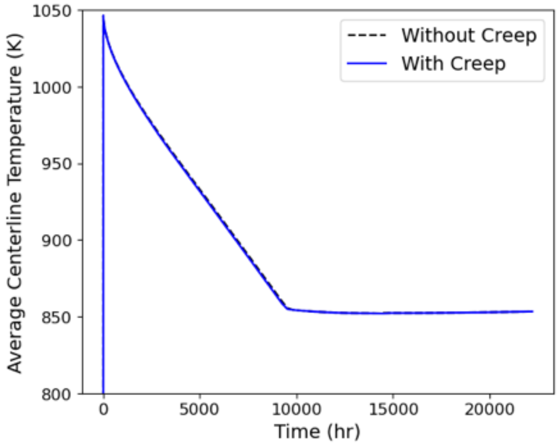












	BISON				Experiment	
	R4		R6		R4	R6
	Stoichiometric	Si-Rich	Stoichiometric	Si-Rich		
Fuel elongation (mm)	0.099 to 0.163	0.088 to 0.154	0.135 to 0.223	0.112 to 0.225	0.0	0.0
Fission gas release (/)	0.0 to 0.007	0.0 to 0.002	0.0 to 0.014	0.0 to 0.011	0.0006	0.0006

TRISO Particle Fuel Performance Modeling

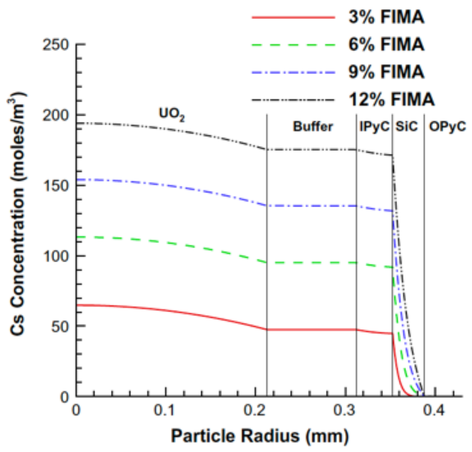
- TRISO particle modeling is still under development from a mechanistic model standpoint
- Imperfect data for many critical fission products through different layers
- Data typically for UO_2 only, not for UC or UCO fuel kernels

$$\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{J} + \lambda C - S = 0,$$

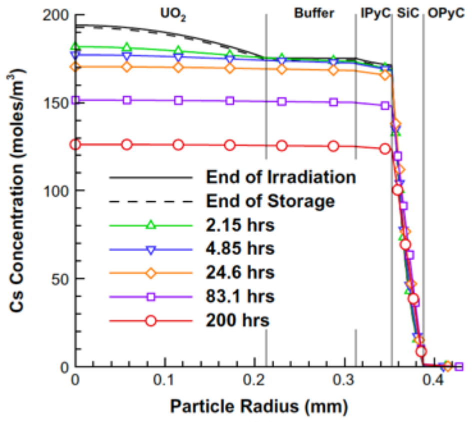
$$\mathbf{J} = -D \nabla C,$$

Table 6
Cs diffusion coefficient parameters from [11] for use in Eqn. (5). Note that Γ is the fast neutron fluence ($\times 10^{25} n/m^2$).

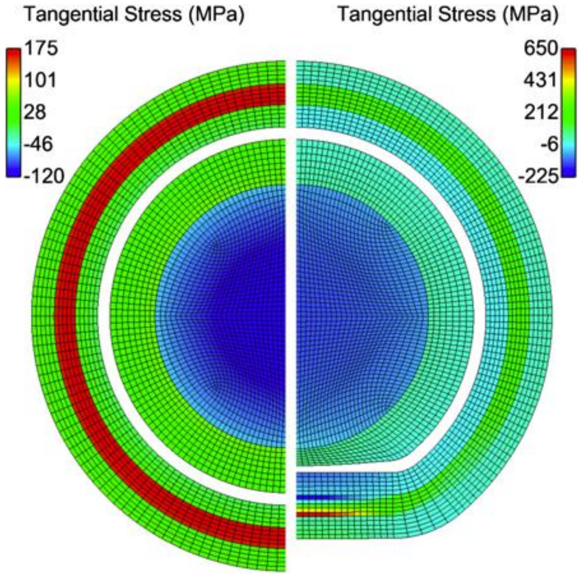
Material	D_1 (m^2/s)	Q_1 (kJ/mol)	D_2 (m^2/s)	Q_2 (kJ/mol)
UO_2	5.6×10^{-8}	209	5.2×10^{-4}	362
Buffer	1×10^{-12}	0	0	0
PyC	6.3×10^{-8}	222	0	0
SiC	$5.5 \times 10^{-14}(e^{\frac{\Gamma}{4.5}})$	125	1.6×10^{-2}	514



(a)

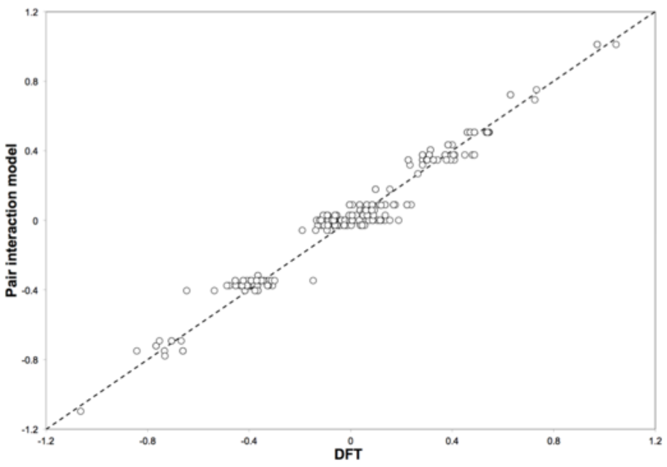


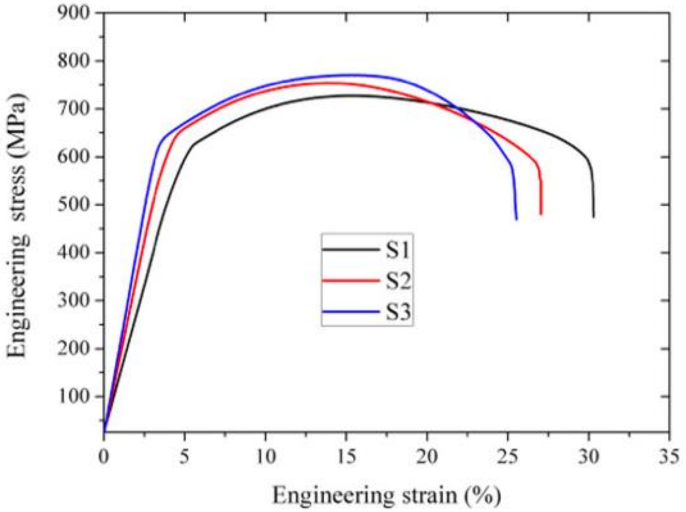
(b)

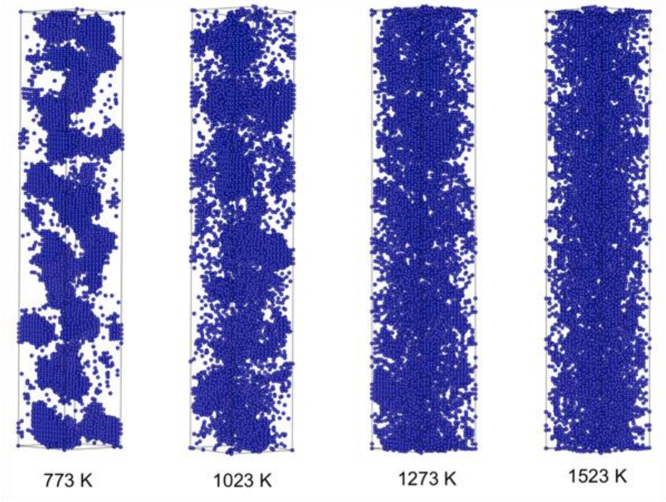


FeCrAl

- Evolution of compositional randomness (precipitation) will affect the mechanical properties of FeCrAl
- Kinetic Monte Carlo simulations modeled the precipitation of alpha' phase from bcc Fe-Cr alloys under thermal aging at various temperatures
- Fe-Al alloys do not show any phase separation
- Can determine critical temperatures for precipitation and the resulting microstructures to investigate potential hardening







Summary

- Researchers are working to develop materials models for the fuel and cladding that are mechanistic rather than empirical and that are based on the evolution of the microstructure rather than the burnup