Nuclear Fuel Performance

NE-533

Spring 2024

Housekeeping

- MOOSE project due end of next week
- Exam next Thursday: 2/29
- Problem session next Tuesday: 2/27

Last Time

- Dr. Jiang covered an intro to numerical thermomechanics
- Fuel performance codes are focused on predicting the temperature of the pellet and the stress in the cladding
- All fuel performance codes
  - Numerically model the temperature in the fuel
  - Numerically model the stress in the cladding
  - And consider gap pressure, closure, and heat transfer in some way
- Last-Last Time:
  - Gap size change due to thermal expansion
  - Intro into defects and materials processing

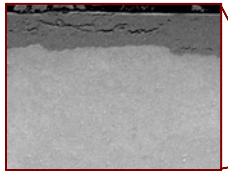
MECHANISTIC MODELING

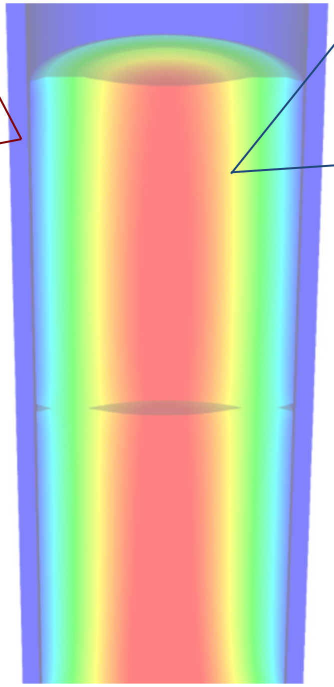
Microstructure-based fuel performance modeling

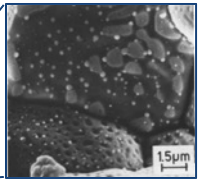
Structure/property relationships connect the microstructure variables to the property values

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 (Cladding)

- 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 (Fuel)

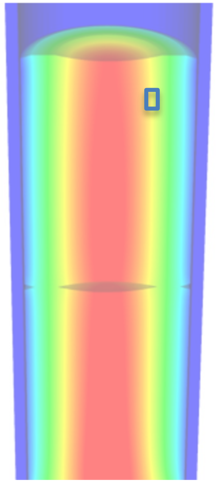
- Grain growth
- Plasticity
- Defect prod. & transport
- Fission gas behavior
- Densification
- Fracture

Fuel Property values

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

Example: fission gas behavior in the fuel

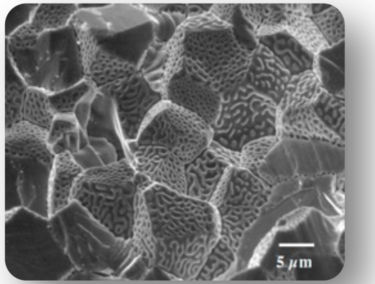
- Take into account a finite set of variables to describe the state of the material
- Utilize a mechanistic model of fission gas behavior to predict the evolution of the microstructure
- Utilize this updated microstructure to inform a number of structure/property relationships



Variables

Temperature | Displacement | Stoichiometry

Model of fission gas behavior

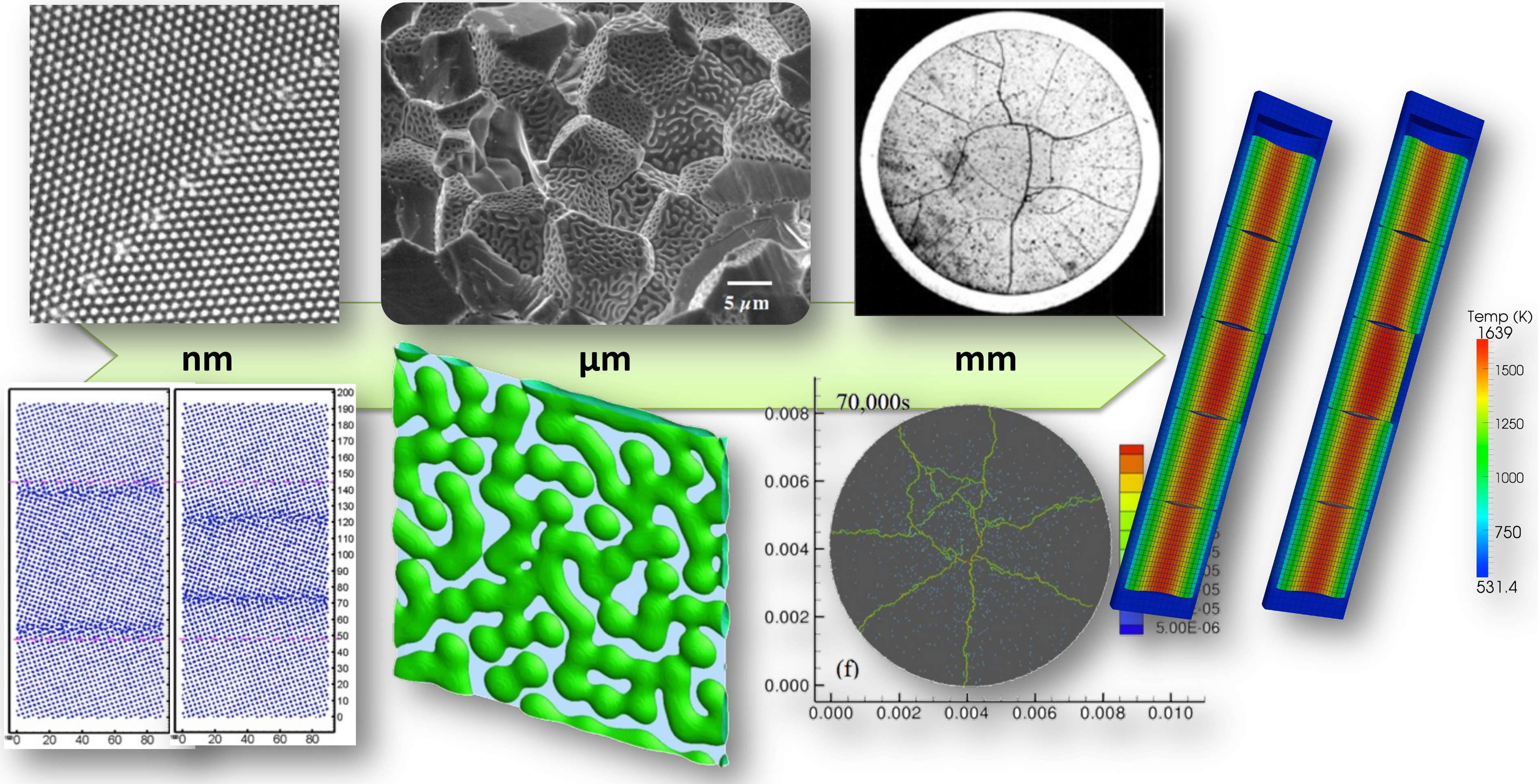


- Dissolved fission products
- Fission gas bubble fraction
- Grain boundary fission gas bubble coverage
- Gap fission gas concentration

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

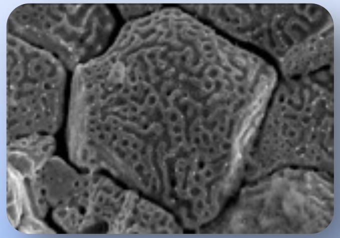
- Can provide a structure/property relationship to replace the existing burnup dependent model
- For example, thermal conductivity, taking into account microstructural features and their evolution

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

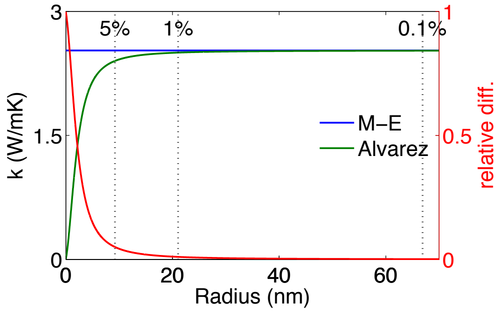
Parametrizing the mechanistic model

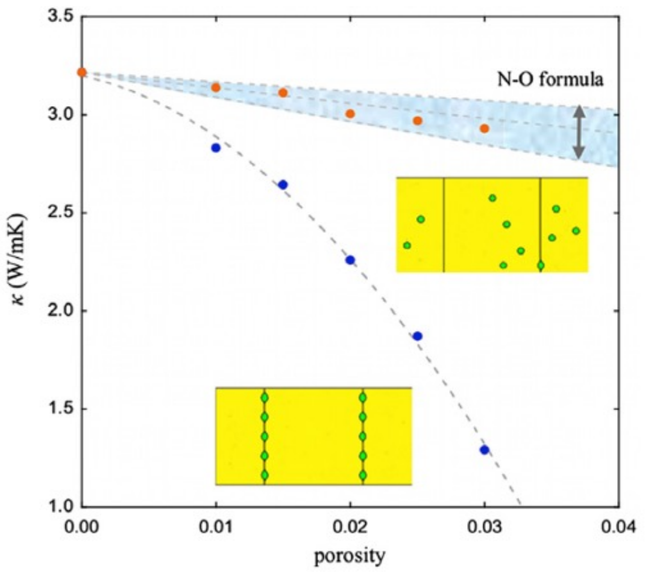
- We employ multiscale modeling and simulation to determine the various parameters for the model
- MD simulations conducted at LANL have been used to determine the coefficients for various point defects
- MD simulations have shown that phonon scattering must be accounted for to accurately represent small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

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

Maxwell-Eucken (no phonon scattering)

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

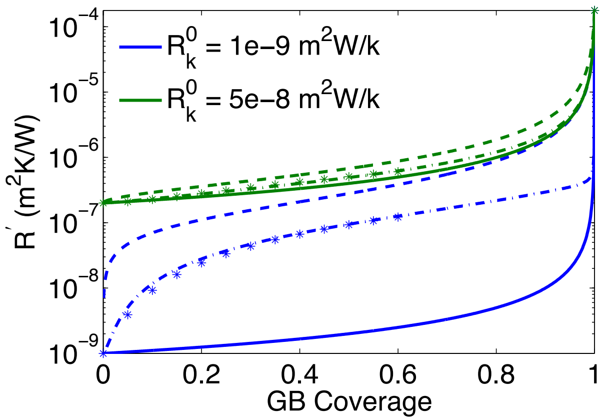


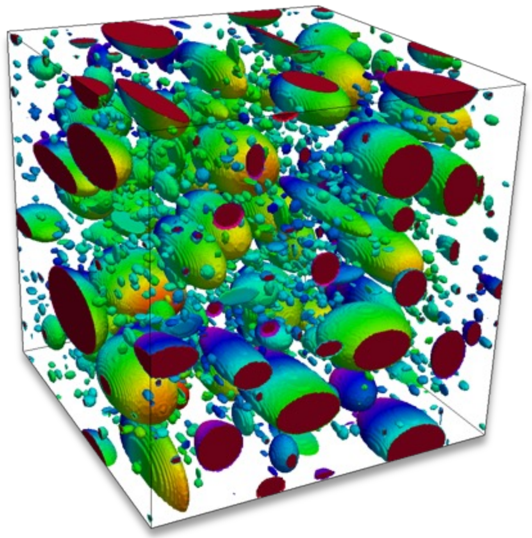


Parametrizing the mechanistic model

- 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

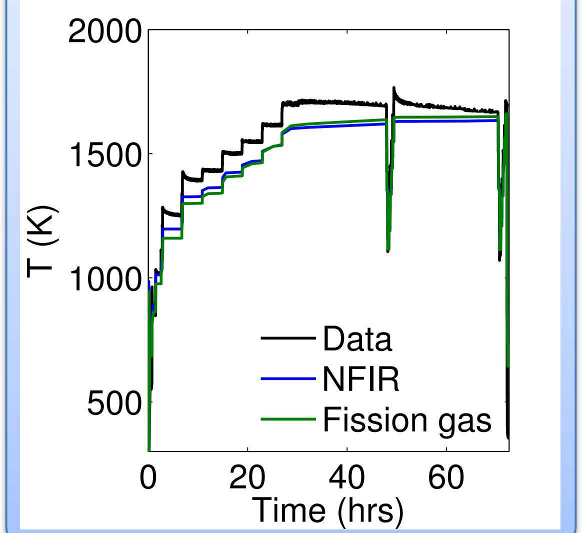
- Each term in the expression must be coupled to a corresponding state variable
- The full model calculates the thermal conductivity as a function of:
  - Temperature
  - Point defect concentrations
  - Intragranular bubble density and average radius
  - Fractional coverage of bubbles on GBs and average 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

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

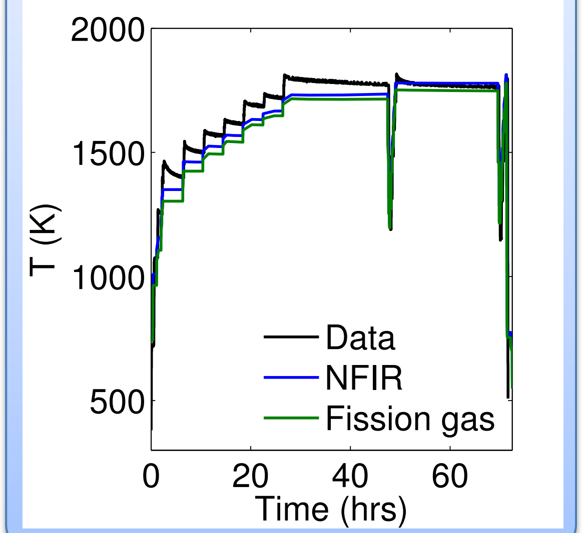
Comparing with experiments…

- 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

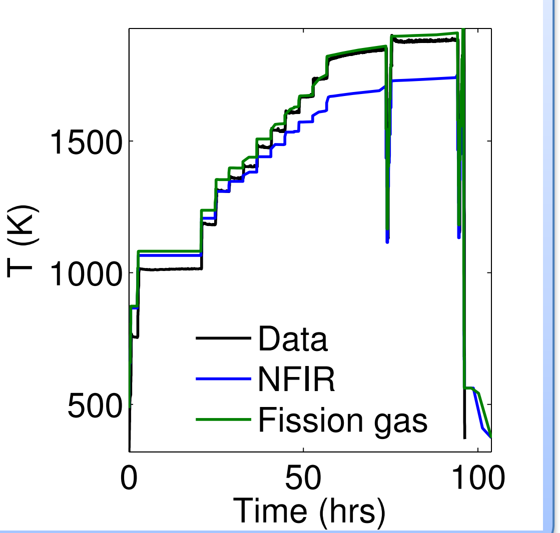
RISO-3 AN3



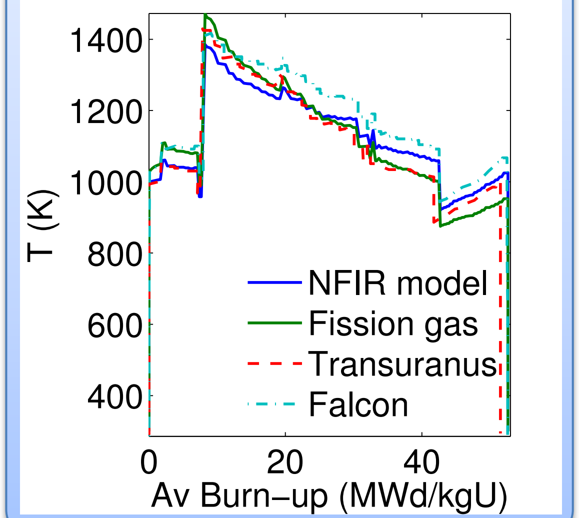
RISO-3 AN4



RISO II-3

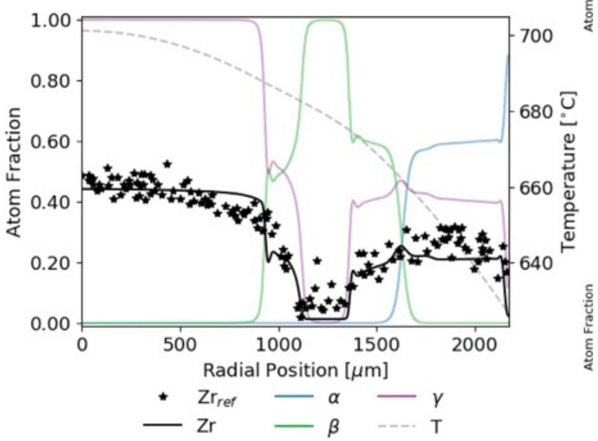


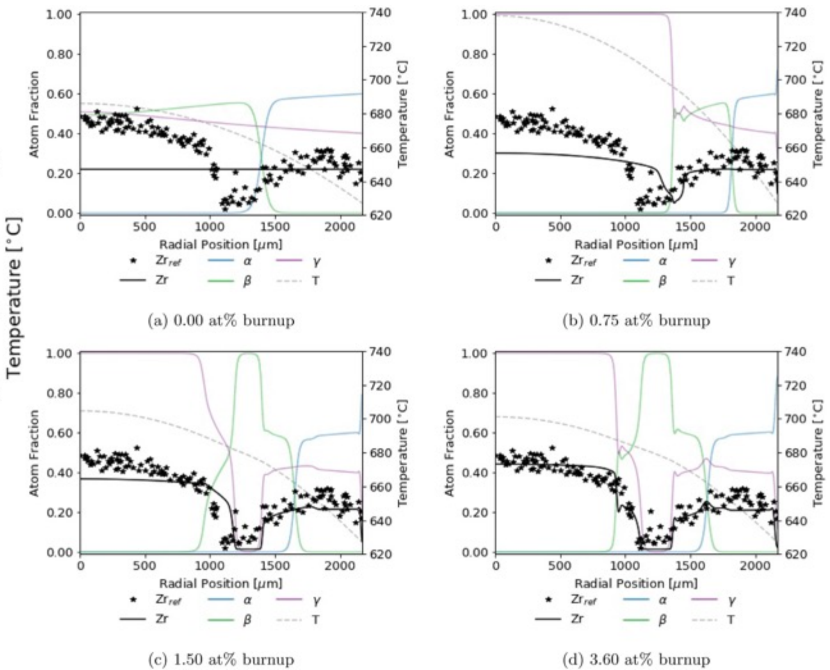
US PWR TSQ0002



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





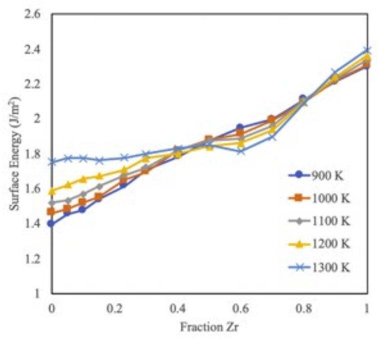
(a) 0.00 at% burnup

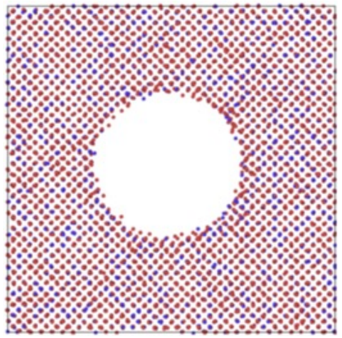
(b) 0.75 at% burnup

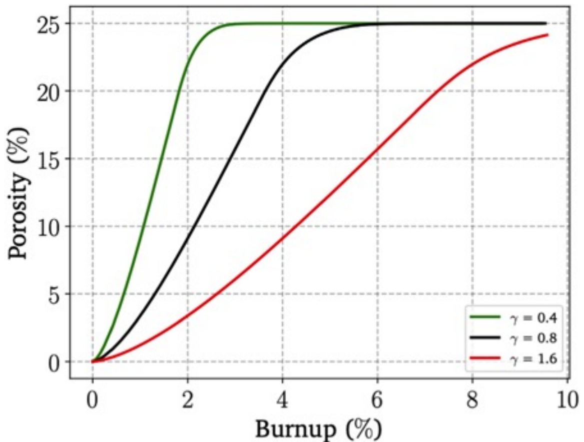
(c) 1.50 at% burnup

(d) 3.60 at% burnup

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

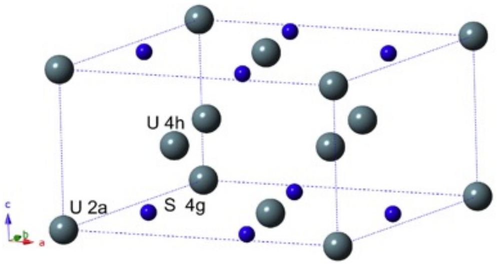


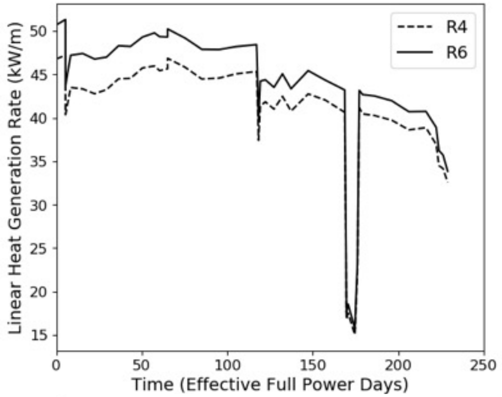


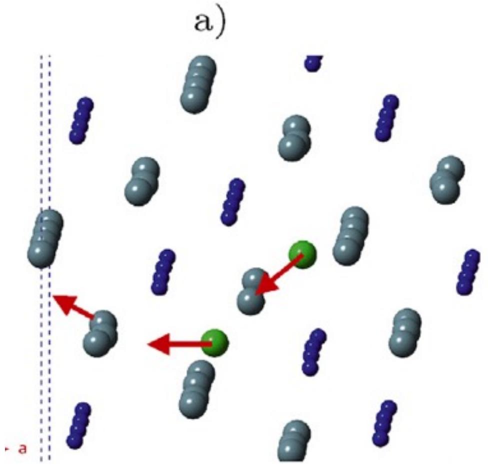


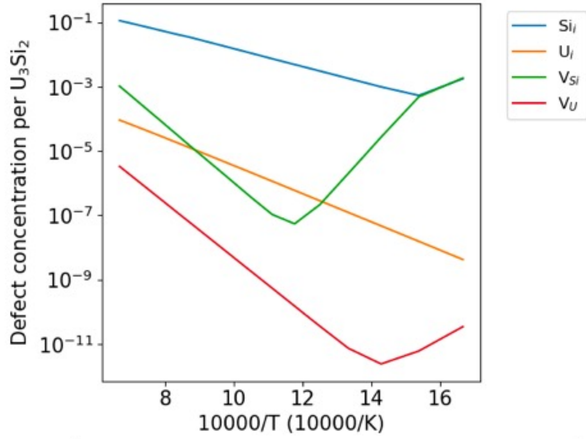
U3Si2 Fuel Performance Modeling

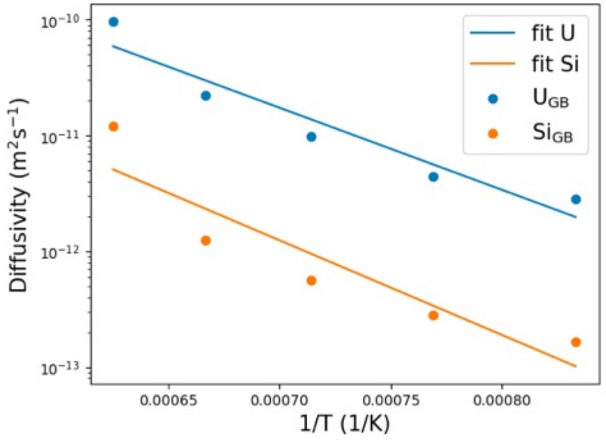
- Point Defect Diffusion, Fission gas swelling, thermal and irradiation creep

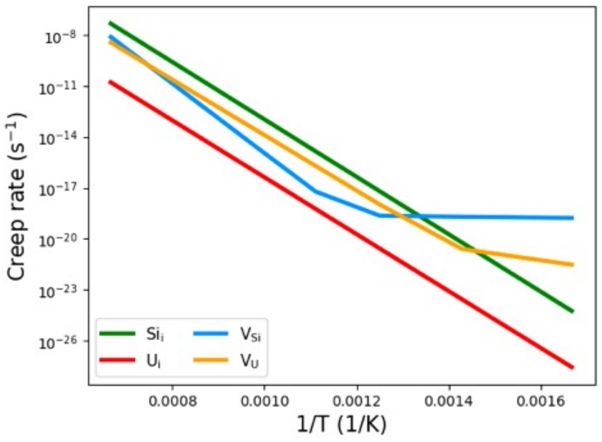


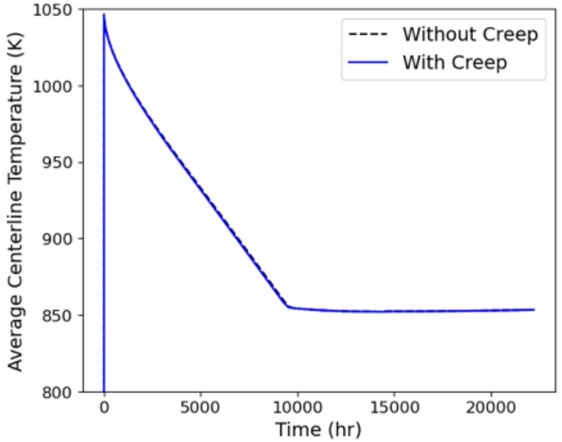








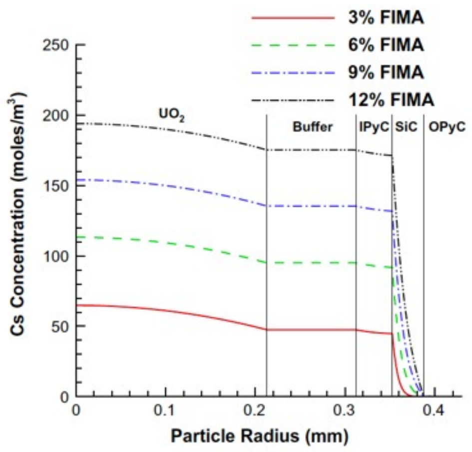




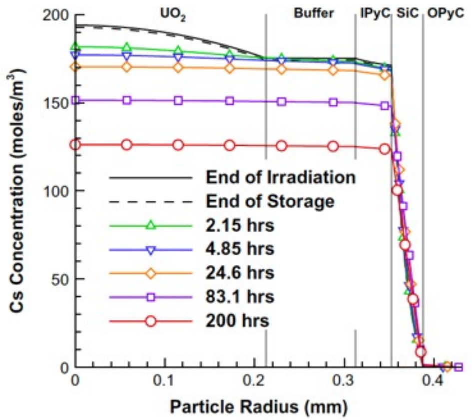
	BISON				Experiment	
	R4		R6		R4	R6
	Stoichiometric	Si-Rich	Stoichiometric	Si-Rich		
Fuel elongation (mm)	0.099 to 0.163	0.088 to 0.154	0.135 to 0.223	0.112 to 0.225	0.0	0.0
Fission gas release (/)	0.0 to 0.007	0.0 to 0.002	0.0 to 0.014	0.0 to 0.011	0.0006	0.0006

TRISO Particle Fuel Performance Modeling

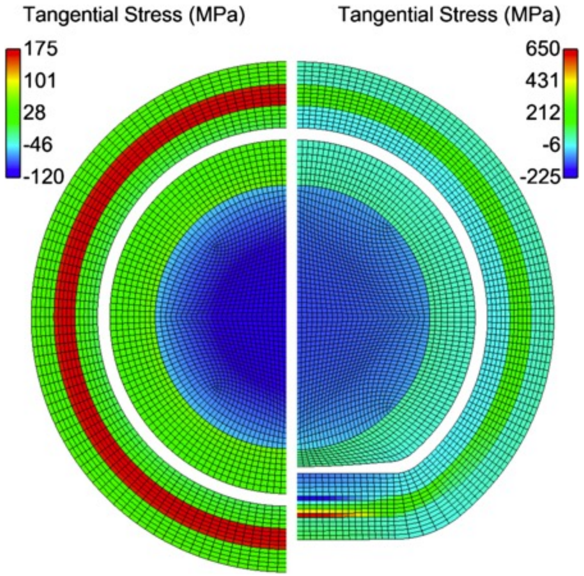
- TRISO particle modeling is still under development from a mechanistic model standpoint
- Imperfect data for many critical fission products through different layers
- Data typically for UO2 only, not for UC or UCO fuel kernels



(a)



(b)



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

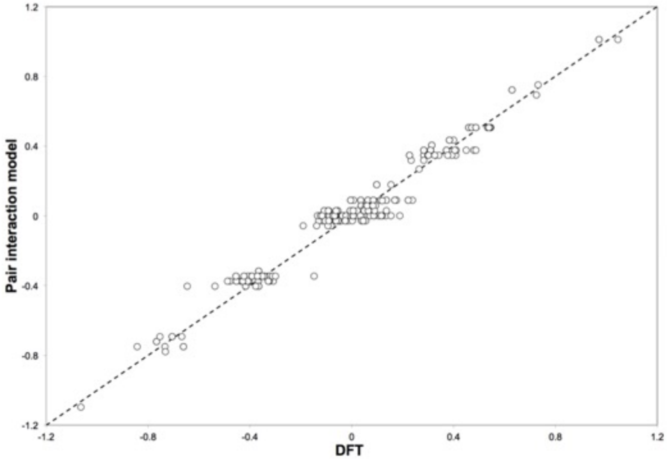
$$\mathbf{J} = -D\nabla C,$$

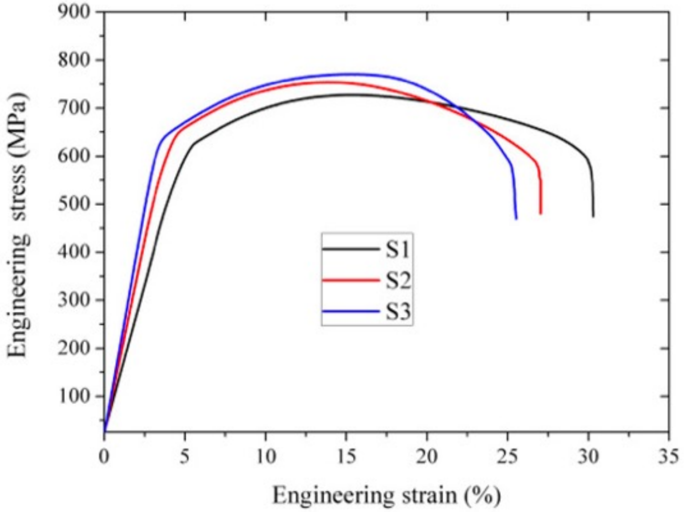
Table 6
Cs diffusion coefficient parameters from [11] for use in Eqn. (5). Note that Γ is the fast neutron fluence ($\times 10^{25} n/m^2$).

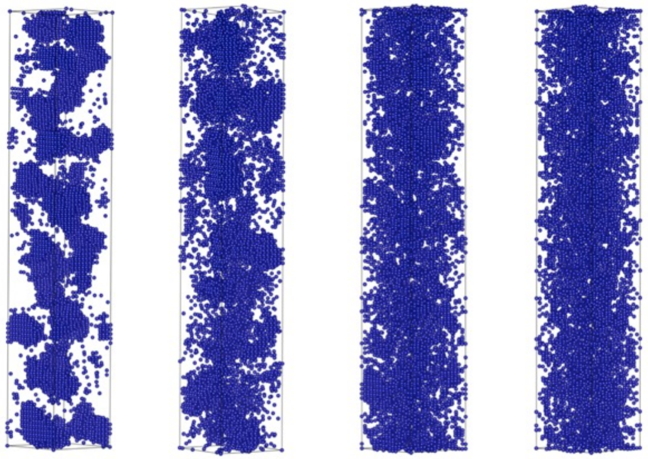
Material	D_1 (m^2/s)	Q_1 (kJ/mol)	D_2 (m^2/s)	Q_2 (kJ/mol)
UO_2	5.6×10^{-8}	209	5.2×10^{-4}	362
Buffer	1×10^{-12}	0	0	0
PyC	6.3×10^{-8}	222	0	0
SiC	$5.5 \times 10^{-14}(e^{\frac{\Gamma}{4.5}})$	125	1.6×10^{-2}	514

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

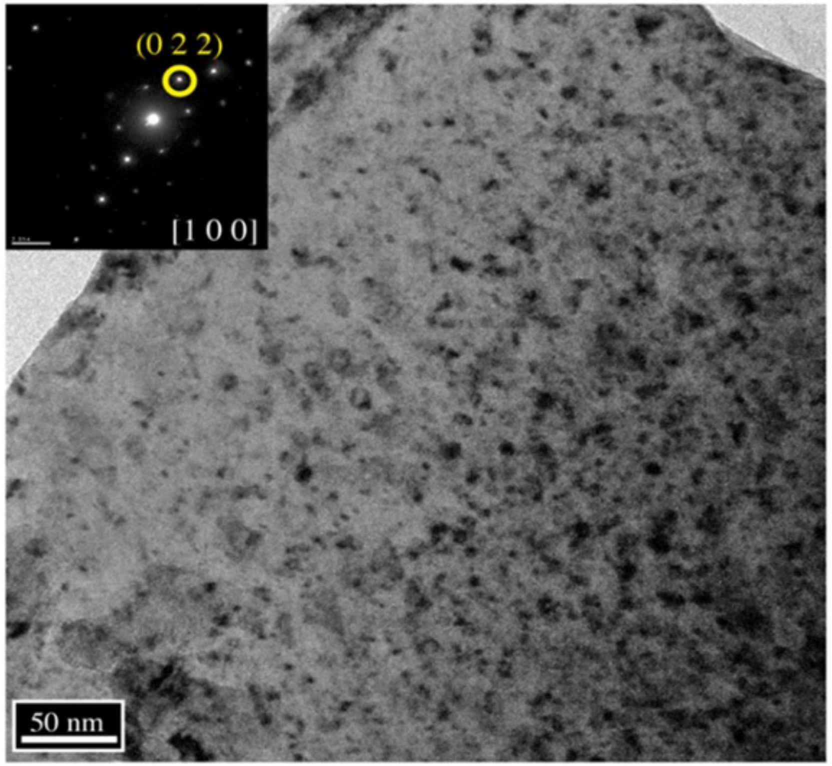
UO_2 RADIATION EFFECTS

Radiation Damage

- The macroscopic and observable results of exposure of solids to energetic particles are collectively known as radiation effects
- Heat production from the nuclear fuel to generate electricity ensues mostly from the slow down of the fission products by nuclear or electronic interactions
- As a direct consequence, there are also defects created along the path of the fission fragments leading to modification of the physical properties of the fuel
- Most Frenkel defects (interstitial-vacancy pair) are produced by secondary collision cascades
- Local charge neutrality prevents the formation of interstitials and vacancies as independent processes, as the cations and anions are charged species
- In UO2, two anion vacancies need to be created for each cation vacancy, forming a Schottky trio
- The dominant defect type is anion Frenkel pairs

Loop Formation

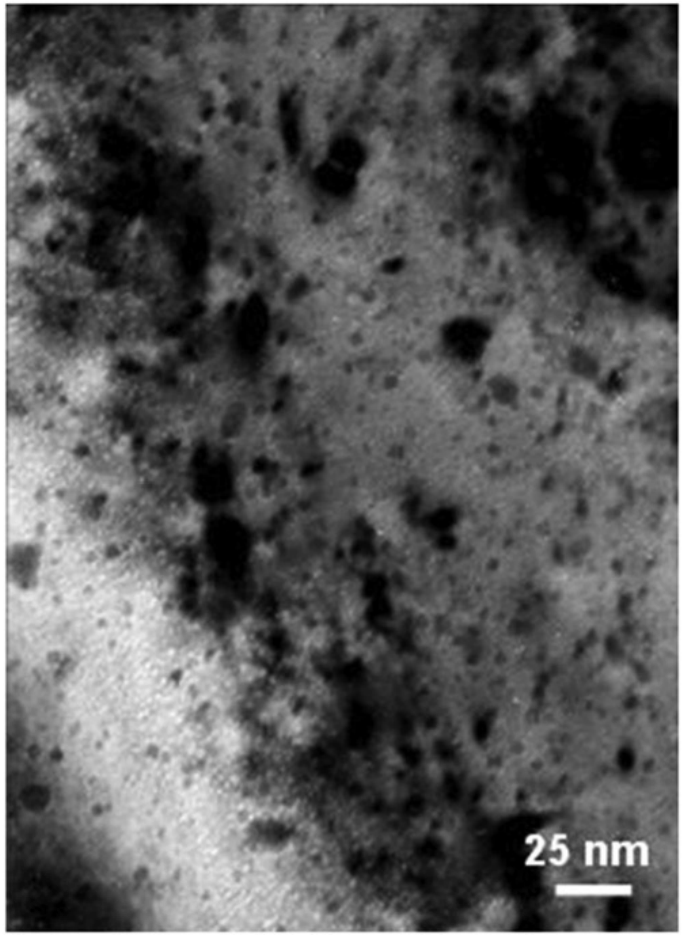
- Dislocations generally a mixture of screw and edge components in the shape of a loop
- The most important slip system in UO2 is the one which minimizes the electrostatic energy, the easy slip along {100}<110> tends to minimize the intense repulsive force between cations
- Prismatic loops are of interstitial type and consist of a disc-shaped layer of defects in a cluster
- This defect cluster is more energetically favorable than dispersed defects in the bulk
- Vacancy clusters will form into voids



Prismatic loops in UO2 from alpha particle irradiation

Voids

- Due to preferential absorption of interstitials at dislocations, we are left with a slight excess of vacancies in the bulk to nucleate and grow voids
- Void nucleation can occur homogeneously by the collection of vacancies in the matrix, or heterogeneously at structural features (precipitates, grain boundaries, etc.)
- The vacancy-rich radiation damage core is considered to be an important void nucleation mechanism in UO2
- Vacancy diffusion then allows void growth
- Gas atoms can often serve to stabilize small voids



Voids (bright spots) from alpha irradiation in UO2

Radiation Effects

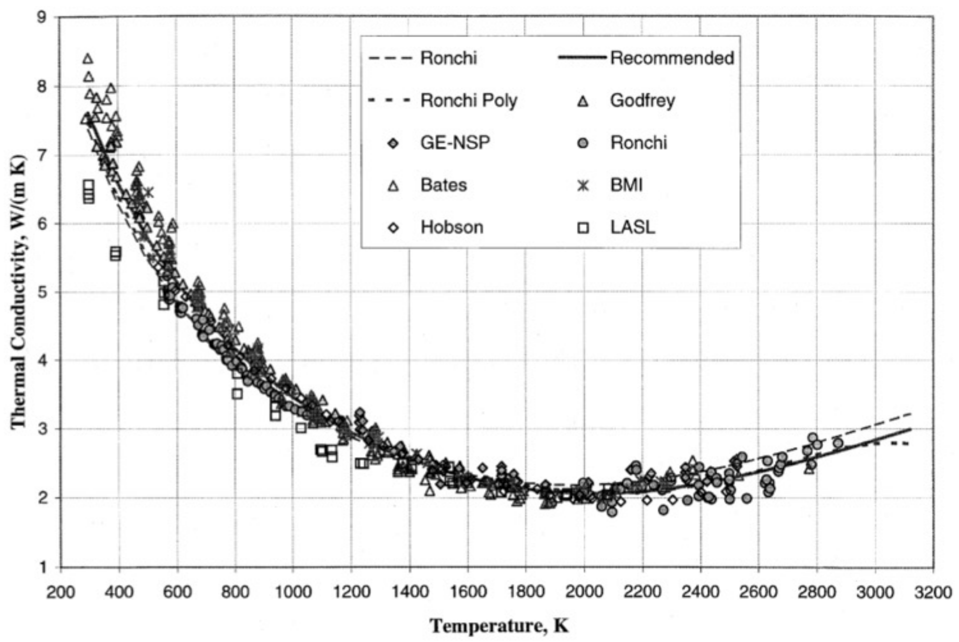
- Chemistry changes
  - two fission products are generated from each fission, resulting up to about 3% of the chemical species present at the end of life for commercial fuel
  - the amount of fission products change both chemical and physical properties
- Physical property changes
  - The physical effects caused by atomic displacements are rather complex and depend on the relative sink strengths of a given material for interstitials and vacancies and on the temperature
  - lattice parameter, thermal conductivity, elasticity, formation of the high burnup structure

Thermal Conductivity Degradation

- One of the major effects of defect formation is the degradation of thermal diffusivity, due to phonon scattering
- Results obtained on irradiated UO2 show that the thermal conductivity is decreasing with increasing burn-up, and that at equal burn-up, samples with higher irradiation temperatures have higher thermal conductivity
- There are mesoscopic and atomistic features that affect the thermal conductivity
- The porosity existing in fresh fuels evolves during irradiation and has a strong impact on heat transport, because voids/bubbles have significantly lower kth than the lattice
- Precipitates are formed by fission products that are insoluble in the UO2 lattice, forming oxide or metallic inclusions: can use composite theory to get effective thermal conductivities; typically, these precipitates INCREASE the thermal conductivity of the fuel

Thermal Conductivity Degradation

- Actinides, rare earths, and transition metals can form mixed oxides with UO2 (soluble fission products): these atoms act as phonon scattering centers as a result of the differences in bonding, radii, or mass, and thus decrease kth
- Volatile fission products/gases are partially in solution and partially in bubbles, where they act as phonon scatterers or as low kth composite fractions
- Point defects and extended defects contribute to the degradation of the thermal conductivity by scattering or limiting the mean free path of the phonons

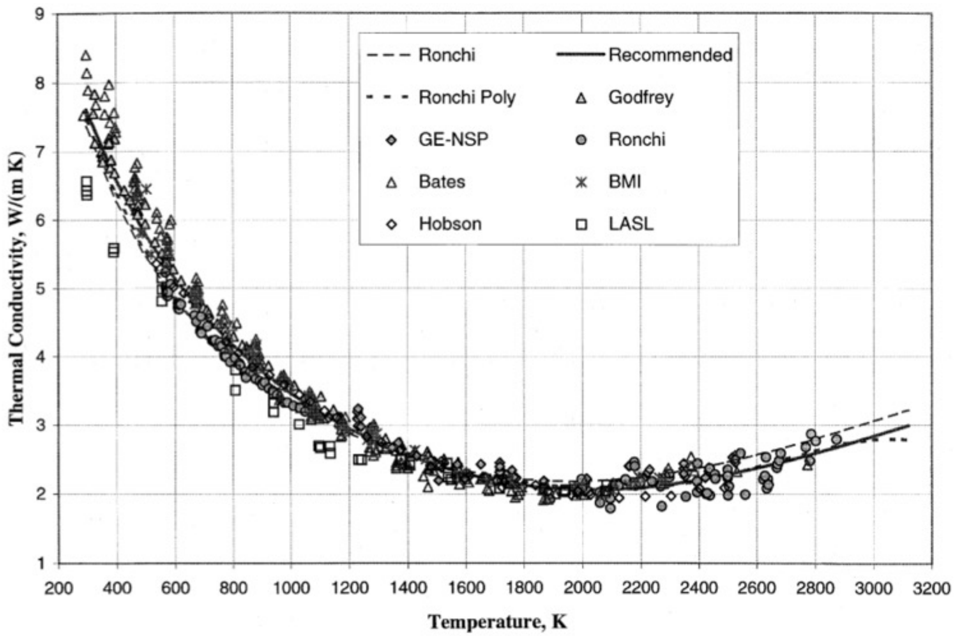


UO2 Thermal Conductivity

- We have established correlations for thermal conductivity of fresh UO2 fuel as a function of temperature

$$k_0 = \frac{100}{7.5408 + 17.629t + 3.6142t^2} + \frac{6400}{t^{5/2}} \exp\left(\frac{-16.35}{t}\right)$$

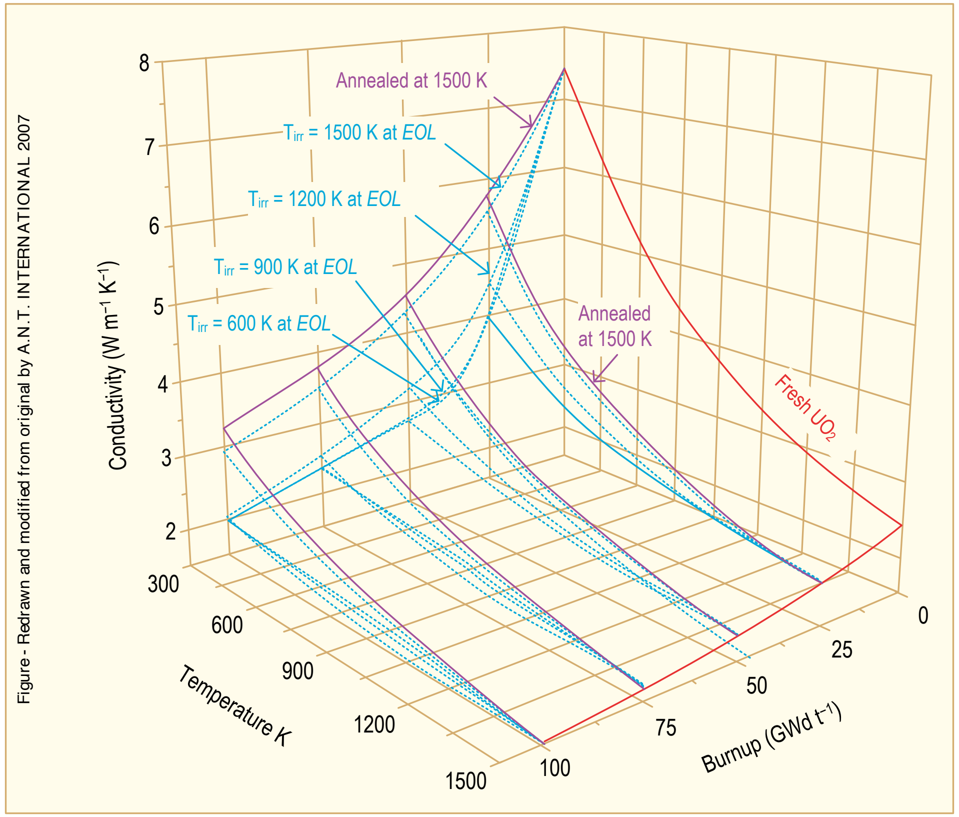
  - Where t = T/1000
  - The first part of the equation describes the phonon interactions
  - The second part describes electronic transport which becomes significant at high temperature



UO2 Thermal Conductivity

- UO_2 thermal conductivity is low and decreases more during reactor operation
- The thermal conductivity has been collected after various amounts of burnup to make empirical fits

$$\lambda_{95} = \frac{1}{A(x) + aG + B(x)T + f_1(Bu) + f_2(Bu)g(Bu)h(T)} + \frac{C}{T^2}\exp\left(\frac{-D}{T}\right)$$



UO2 Thermal Conductivity

- One empirical model used in BISON is the NFIR model
- The model is a function of the temperature T (in °C) and the burnup β (in MWD/kgU

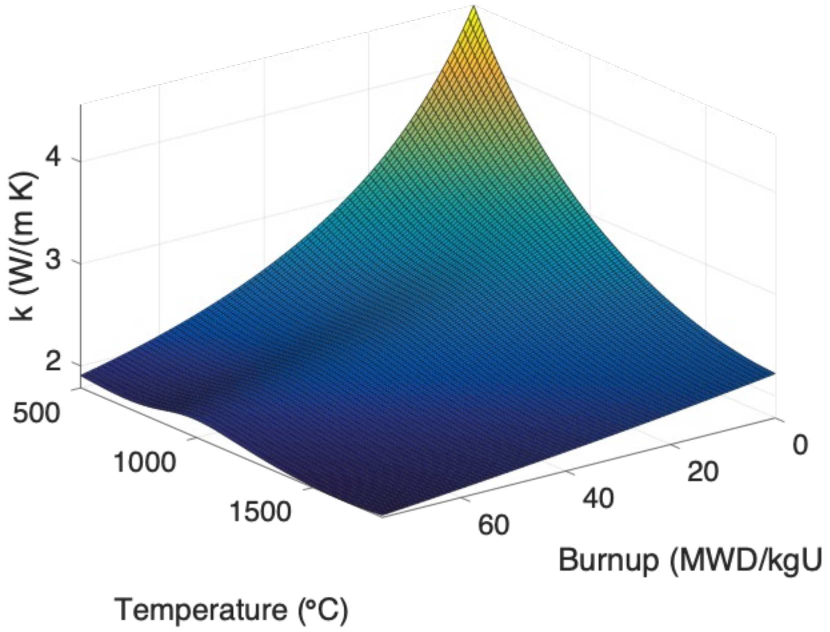
$$k = (1 - R_f(T))k_{ph1}(T, \beta) + R_f(T)k_{ph2}(T, \beta) + k_{el}(T)$$

$$R_f(T) = \frac{1}{2}\left(1 + \tanh\left(\frac{T - 900}{150}\right)\right)$$

$$k_{ph1} = \frac{1}{(9.592 \times 10^{-2} + 6.14 \times 10^{-3}\beta - 1.4 \times 10^{-5}\beta^2 + (2.5 \times 10^{-4} - 1.81 \times 10^{-6}\beta)T}$$

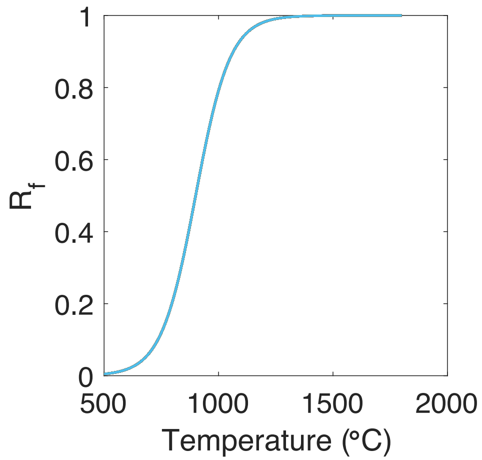
$$k_{ph2} = \frac{1}{(9.592 \times 10^{-2} + 2.6 \times 10^{-3} \cdot \beta + (2.5 \times 10^{-4} - 2.7 \times 10^{-7}\beta)T}$$

$$k_{el} = 1.32 \times 10^{-2}\, e^{1.88 \times 10^{-3} T}$$



UO2 Thermal Conductivity

- The R_f function switches between two k_{ph} functions, accounting for thermal recovery



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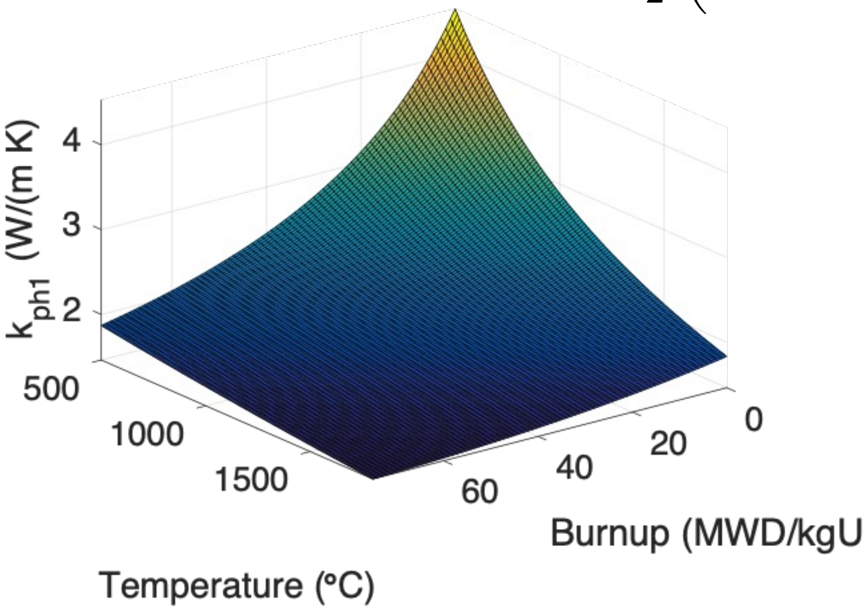
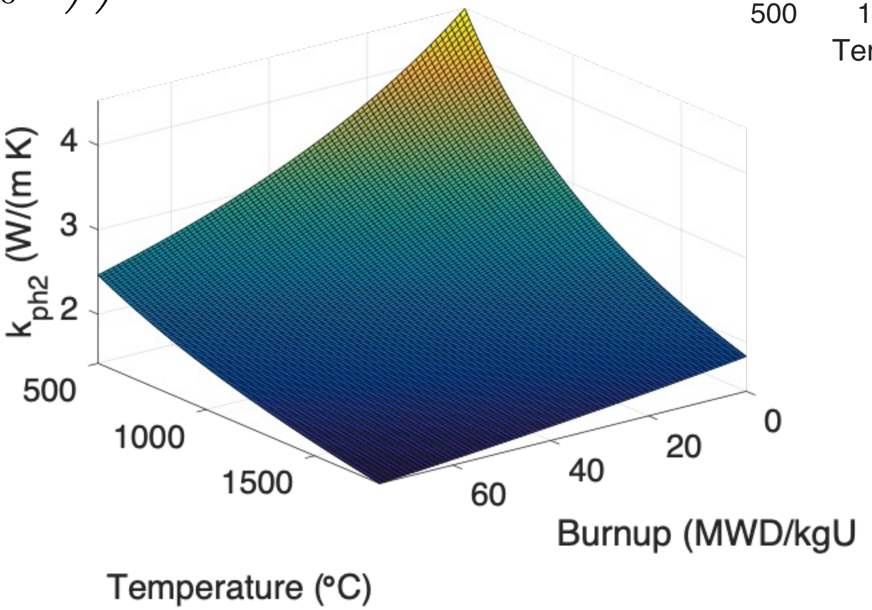
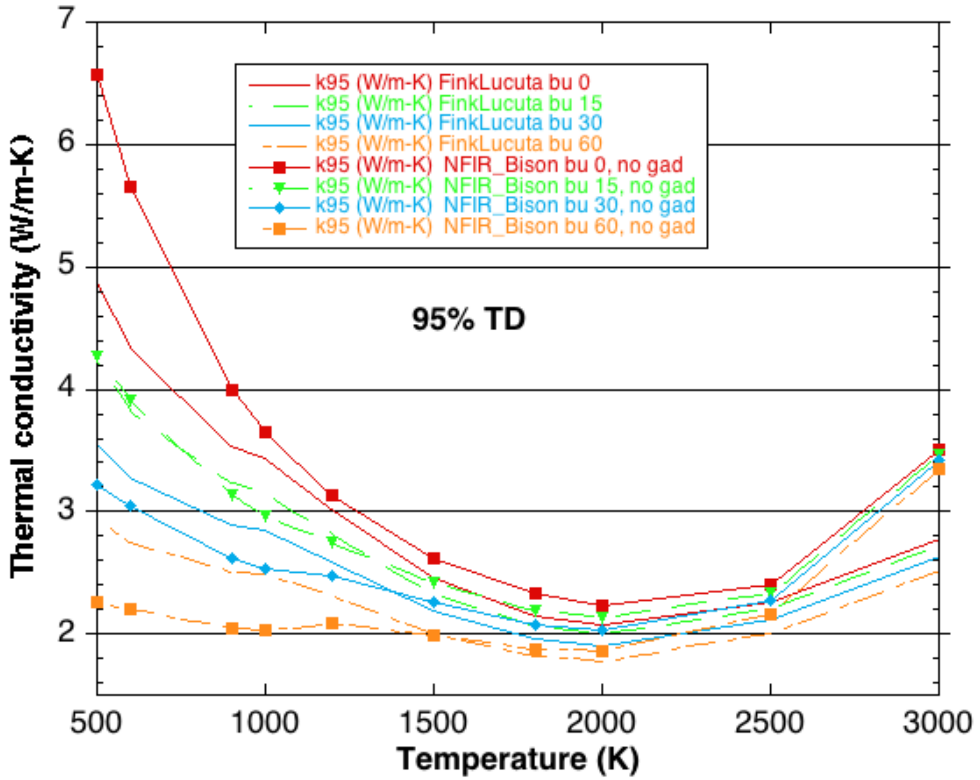




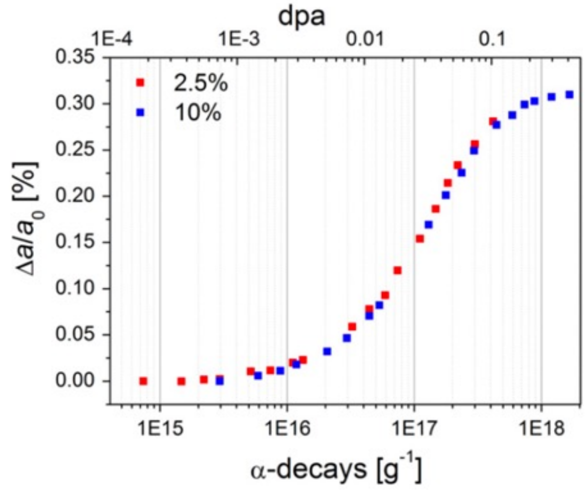
MOOSE/BISON Models

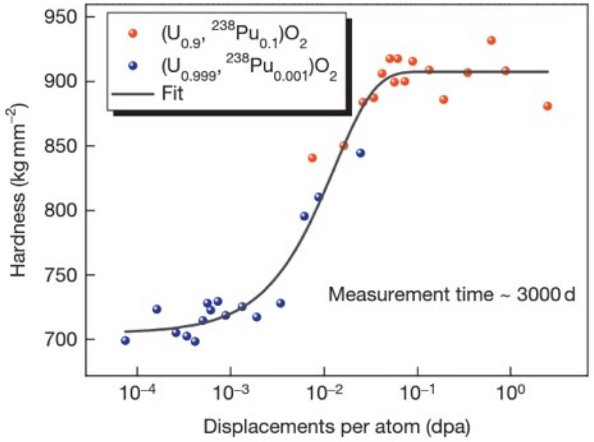
- https://mooseframework.inl.gov/bison/source/materials/UO2Thermal.html
- Fink-Lucuta
- Halden
- NFIR
- NFI
- Ronchi-Staicu
- Toptan (NCSU alum)

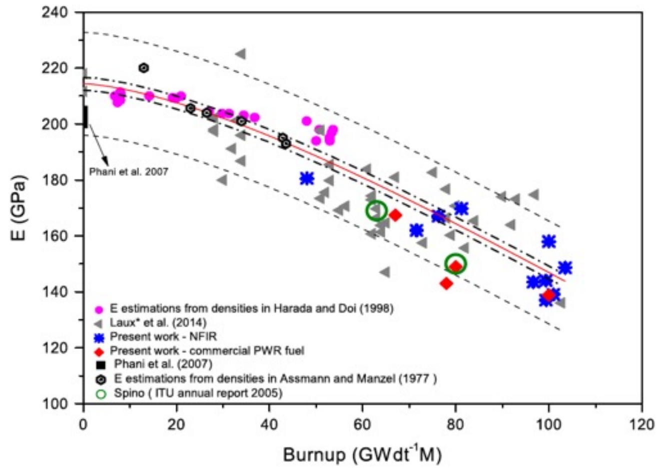


Other Physical Changes

- 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

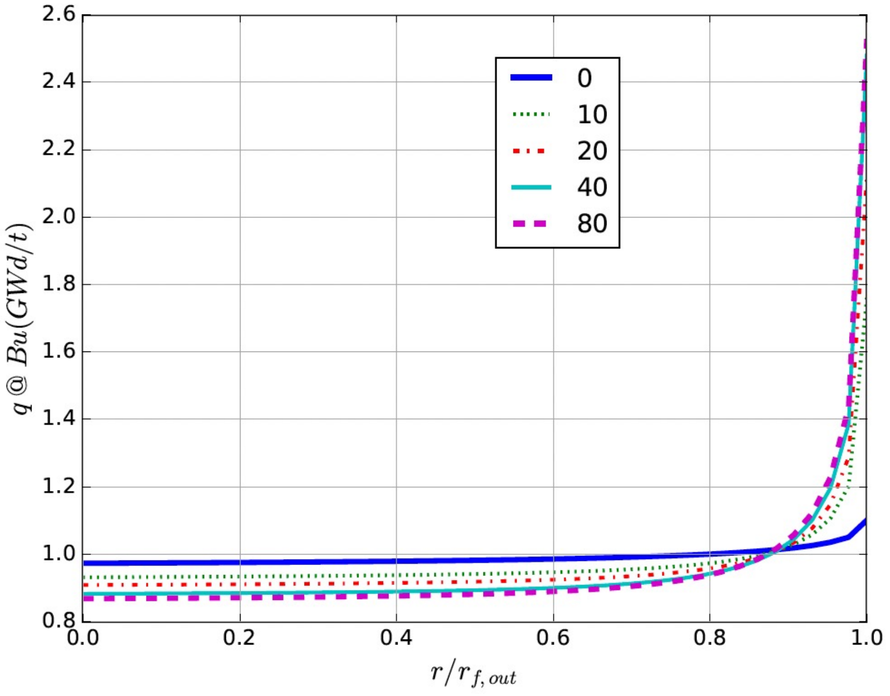






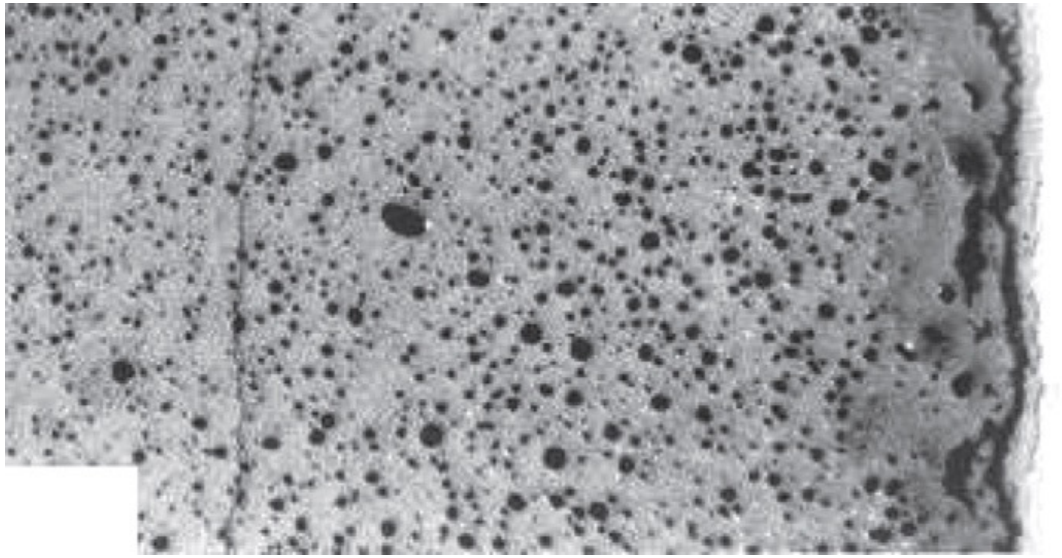
High Burnup Structure (HBS)

- 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



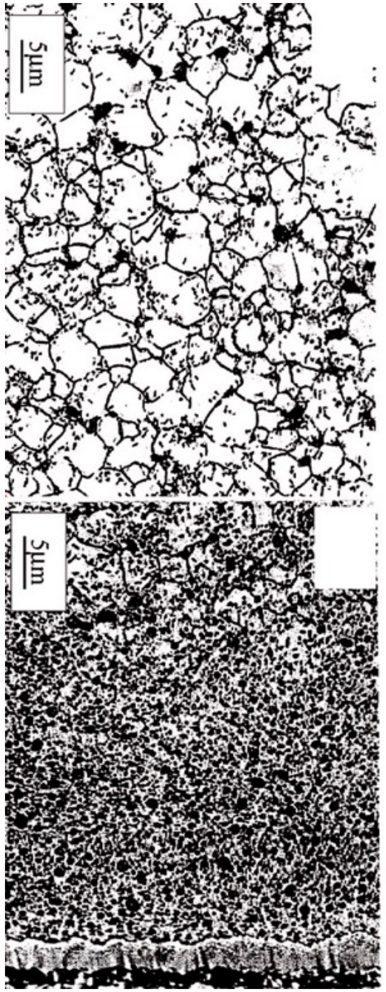
High Burnup Structure (HBS)

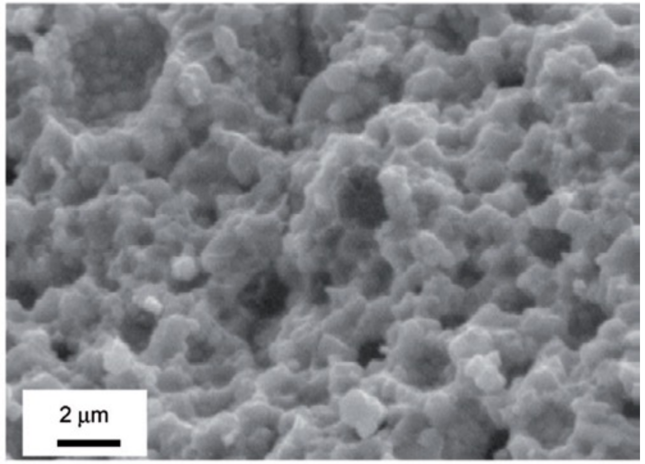
- When thermal recovery is not sufficiently efficient (at lower temperatures), the accumulation of defects at high burnup can lead to the instability of the crystalline structure, initiating a restructuring driven by the energy stored in the material
- This can be amorphization (not observed in UO2) 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)

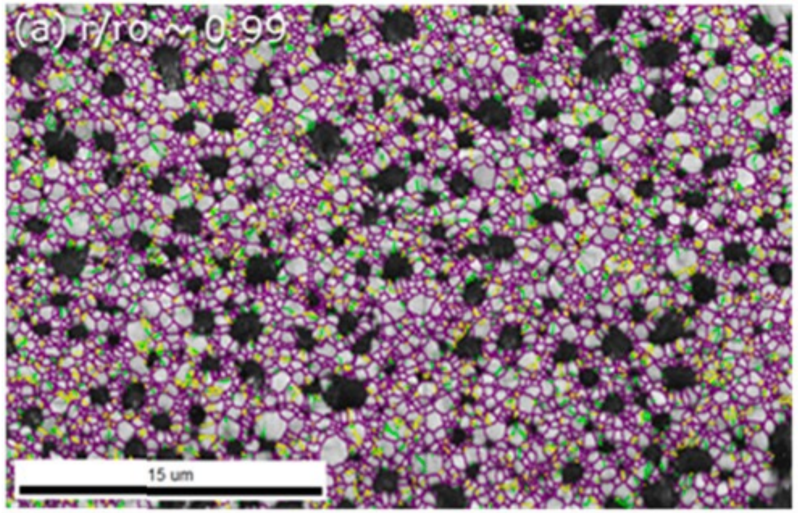
- The increase of the relative porosity volume degrades the material conductivity and reduces the mean grain size
- On the other hand, the intragranular irradiation defect-cleaning improves the fuel intrinsic thermal conductivity
- Fission gas in HBS bubble is retained, not released
- Exact mechanism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



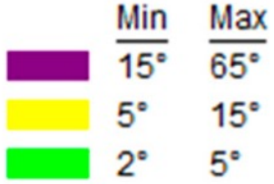


High Burnup Structure (HBS)

- 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