

# Nuclear Fuel Performance

NE-591-010  
Spring 2021

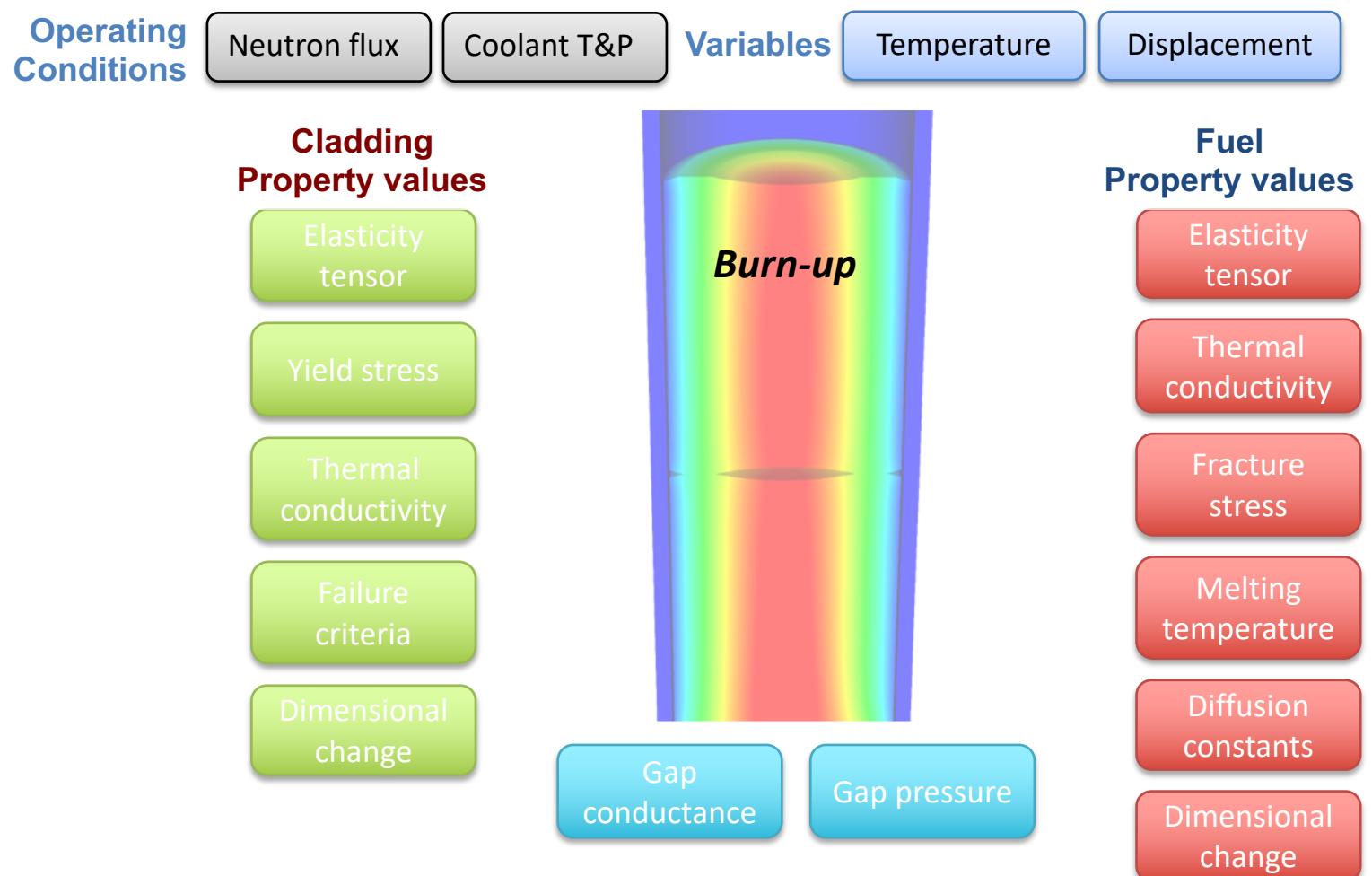
## Last Time

- Reactivity insertion accident
  - often in PWR/BWR dependent upon control rod ejection/drop
- Loss of coolant accidents (LOCA)
- The primary negative effects are:
  - Embrittlement and ballooning of the cladding, relocation and fission gas release in the fuel, cladding can burst and release fuel fragments, etc.

# MECHANISTIC FUEL PERFORMANCE MODELING

# Going beyond burnup...

- Fuel performance codes historically rely on materials models correlated to temperature and burn-up
- Development has begun on models based on microstructure rather than burn-up

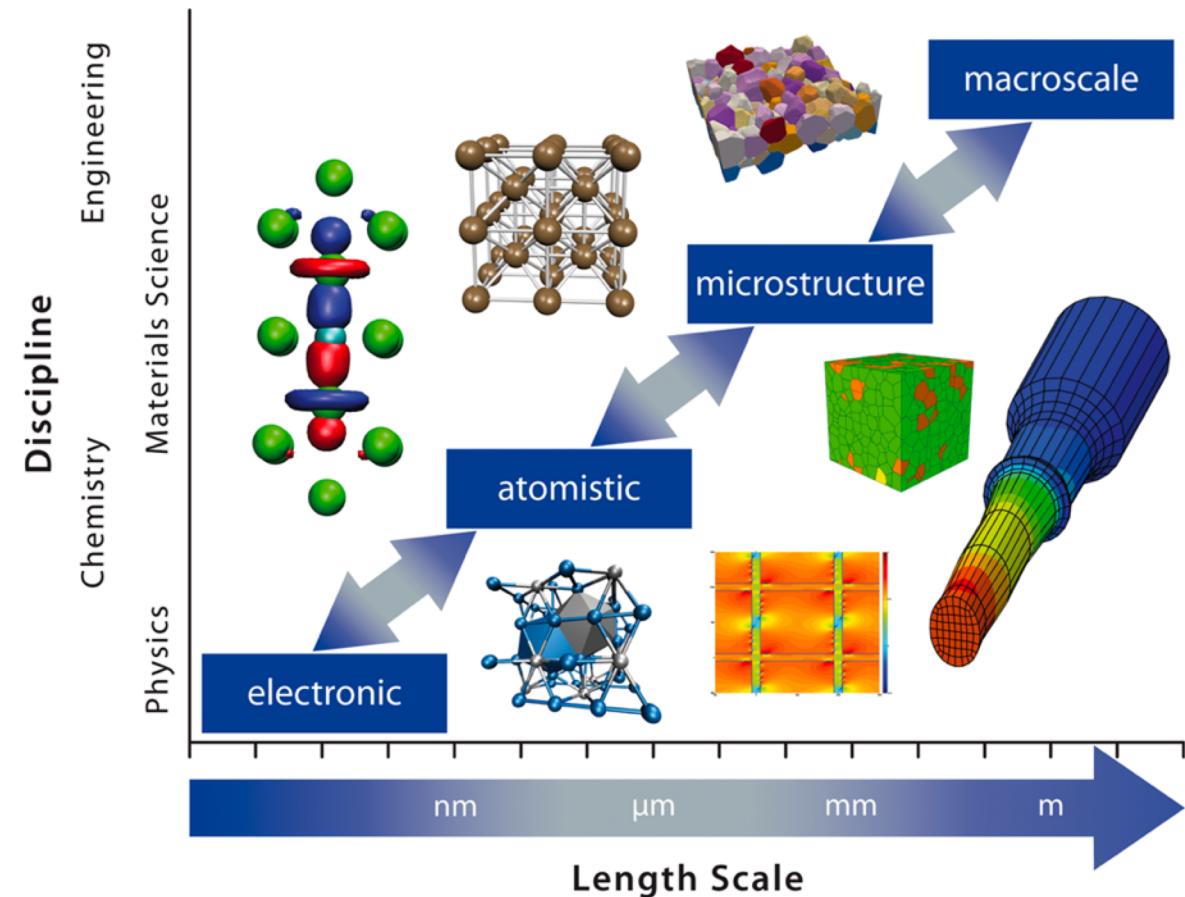


# Microstructure/Mechanistic/Multiscale

- Mechanistic: relating to theories which explain phenomena in purely physical or deterministic terms
- There is a drive for fuel performance codes to employ mechanistic materials models that are based on the current state of the evolving microstructure rather than burn-up
- A series of state variables define the current state of the microstructure, and the evolution of these state variables is defined by mechanistic models that are functions of fuel conditions and other state variables
- The material properties of the fuel and cladding are determined from microstructure/property relationships that are functions of the state variables and the current fuel conditions
- Multiscale modeling and simulation is being used in conjunction with experimental data to inform the development of these models
- This mechanistic, microstructure-based approach has the potential to provide a more predictive fuel performance capability

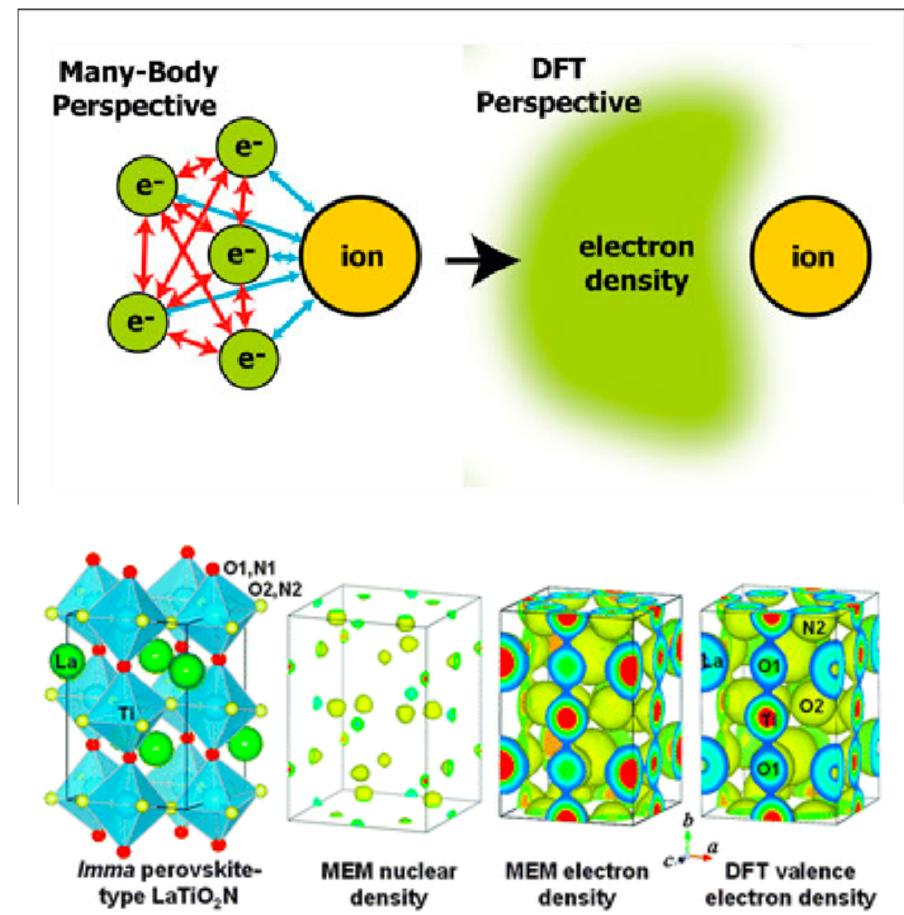
# Multiscale Modeling

- Modeling and simulation is the essential bridge from good science to good engineering, spanning from fundamental understanding of materials behavior to deliberate design of new materials technologies leveraging new properties and processes
- Lower length scale modeling can provide insight, properties, mechanisms, behaviors, etc., that can be input into higher length scale modeling tools to describe, mechanistically, macroscale behavior



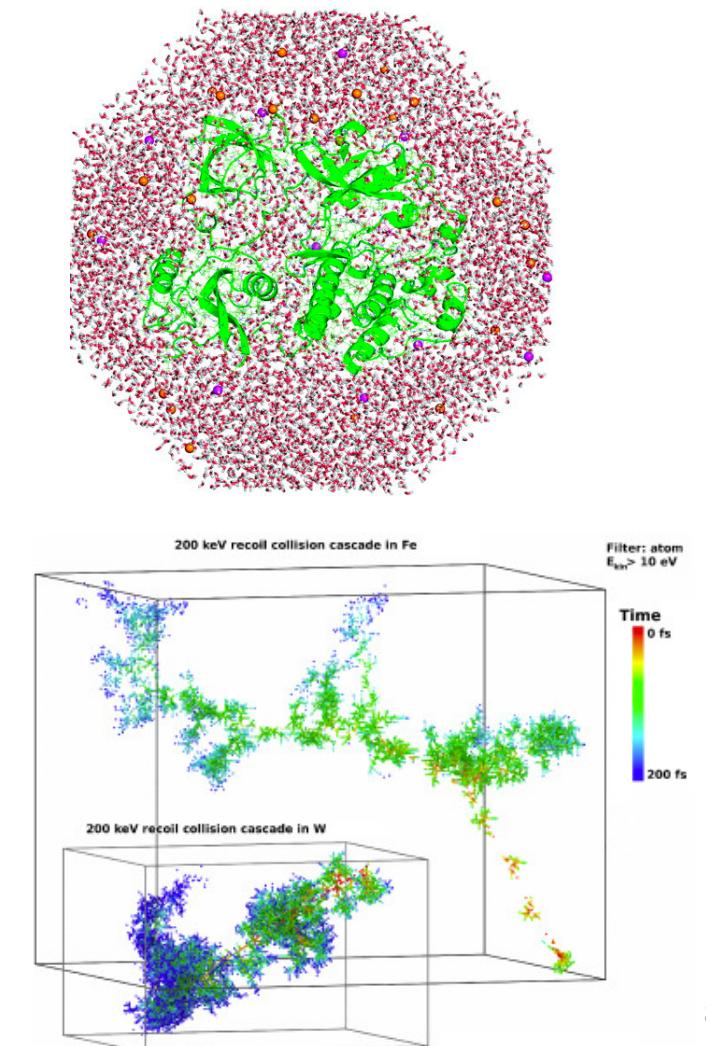
# Density Functional Theory

- DFT is a computational quantum mechanical modelling method used to investigate the electronic structure of many-body systems, in particular atoms, molecules, and the condensed phases
- DFT is primarily utilized to investigate electronic structure, cohesive energy, elastic constants, phonons, entropies, etc.



# Molecular Dynamics

- MD is a computer simulation method for analyzing the physical movements of atoms and molecules, determined by numerically solving Newton's equations of motion for a system of interacting particles
- MD is still atomistic, and is often utilized to calculate transport properties, melting temperature, clustering, etc., that are properties just beyond DFT capabilities



# Rate Theory/Cluster Dynamics

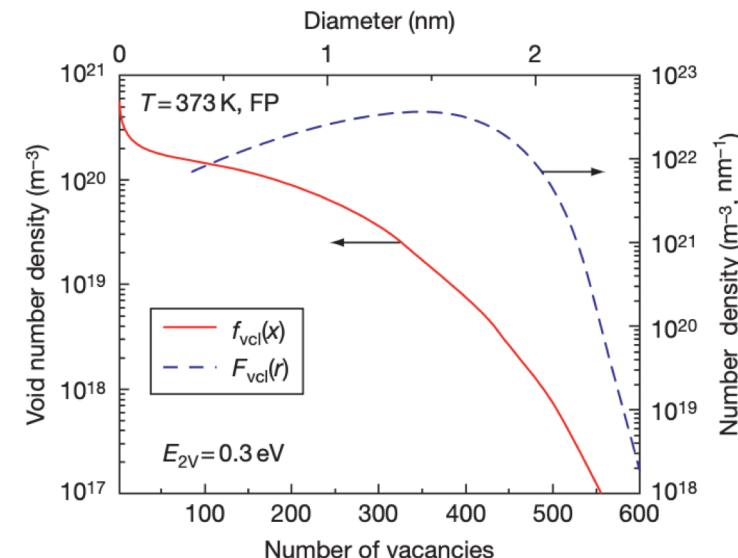
- Provides recipes for calculating reaction rates between individual species of the types which are ubiquitous in chemistry and physics
- Is typically a mean-field approach which uses transition state theory as a tool for describing reactions involving radiation-produced defects
- Can be utilized to describe clustering, absorption, emission, growth, resolution, etc. to describe microstructural phenomena on an intermediate time scale

$$v(\tilde{E}) = 0.8 \frac{E^{\text{PKA}}(\tilde{E})}{2E_d} \quad G_v = G^{\text{NRT}}(1 - \varepsilon_r)(1 - \varepsilon_v)$$

$$G_i = G^{\text{NRT}}(1 - \varepsilon_r)(1 - \varepsilon_i)$$

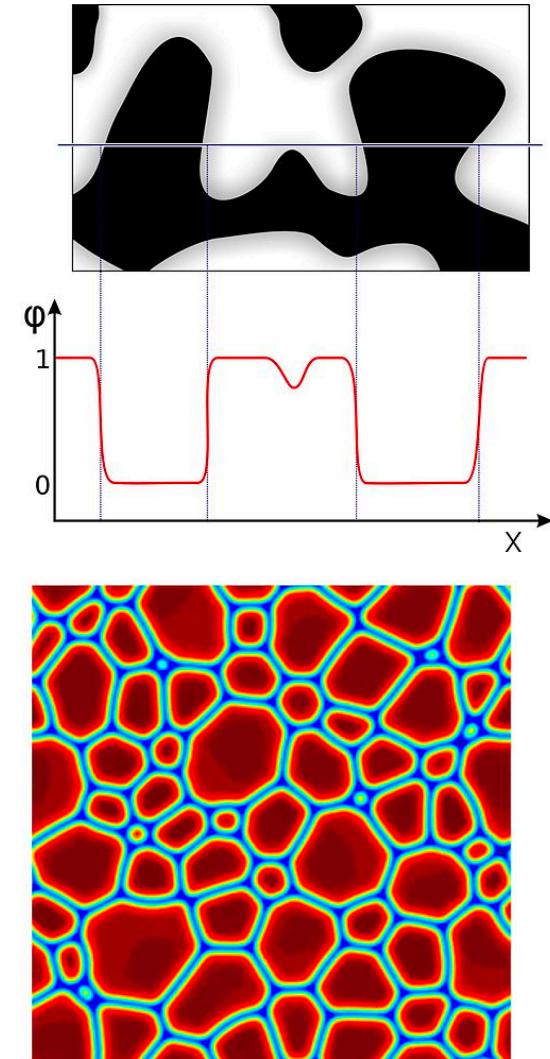
$$\frac{dC_v}{dt} = G^{\text{NRT}}(1 - \varepsilon_r)(1 - \varepsilon_v) + G_v^{\text{th}} - k_v^2 D_v C_v - \mu_R D_i C_i C_v$$

$$\frac{dC_i}{dt} = G^{\text{NRT}}(1 - \varepsilon_r)(1 - \varepsilon_i) - k_i^2 D_i C_i - \mu_R D_i C_i C_v$$



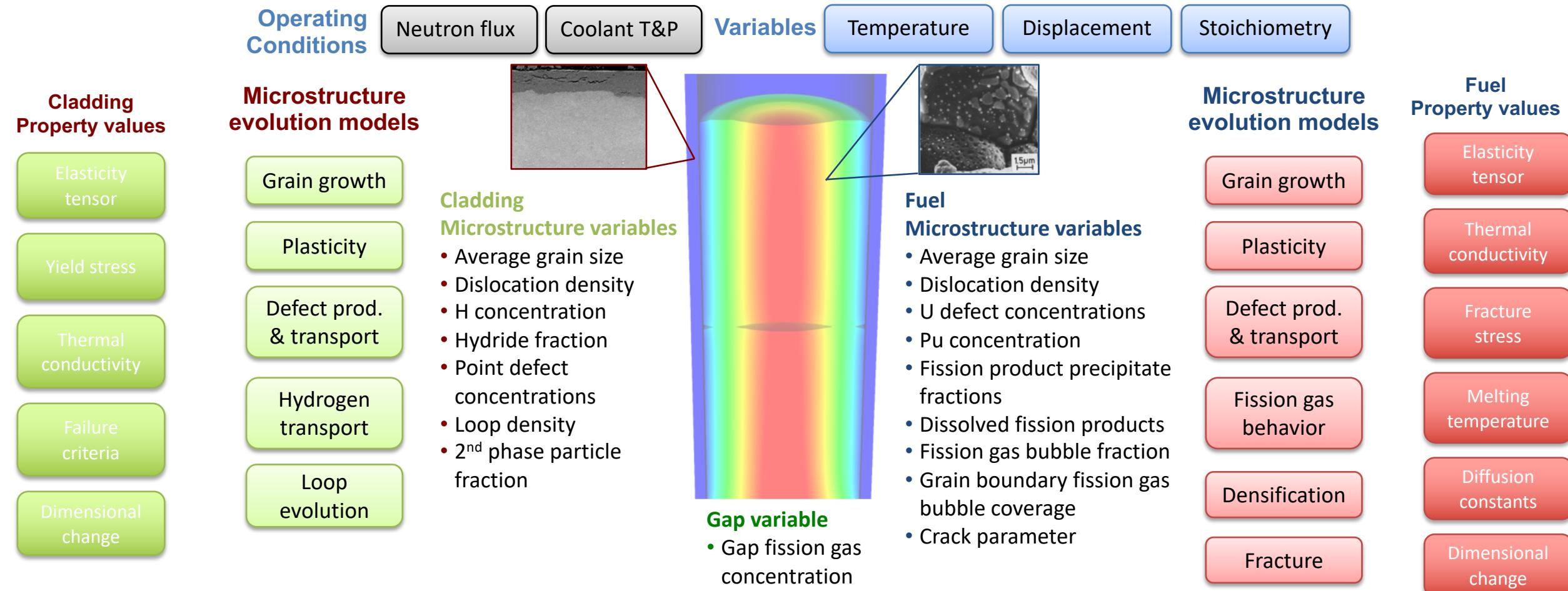
# Phase-Field

- PF is mathematical model for solving interfacial problems that substitutes boundary conditions at the interface by a partial differential equation for the evolution of an auxiliary field (the phase field) that takes the role of an order parameter
- The order parameter takes two distinct values (for instance 1 and 0) in each of the phases, with a smooth change between both values in the zone around the interface, which is then diffuse with a finite width
- PF is commonly used for grain growth, phase separation, bubble coalescence, recrystallization, etc.



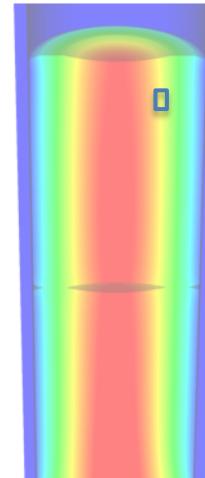
# Microstructure-based fuel performance modeling

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



# 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

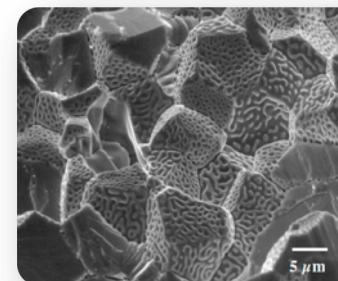


Variables

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

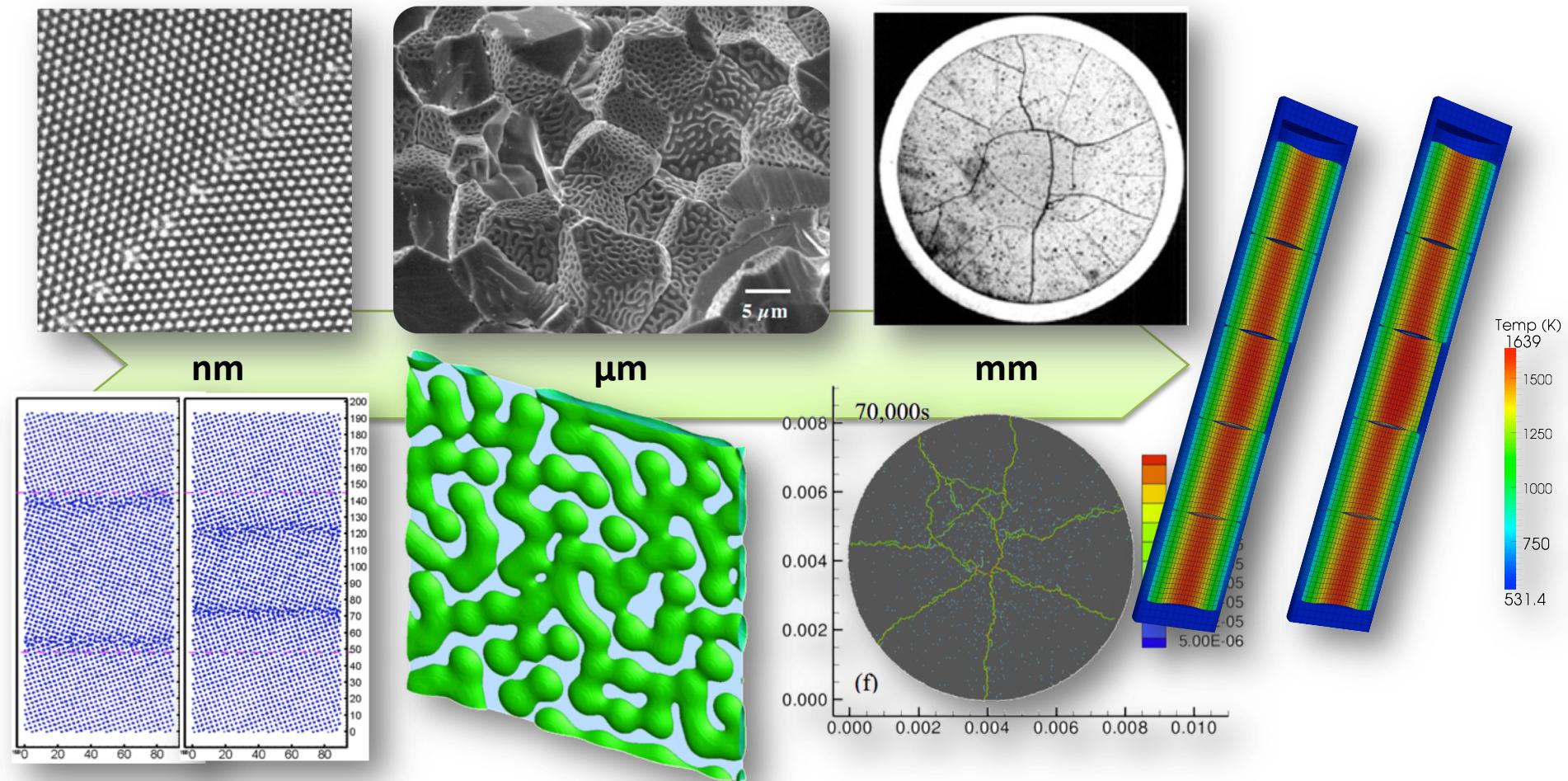
Fracture stress

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

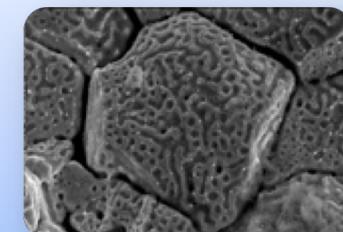
Grain boundary  
and bubbles

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

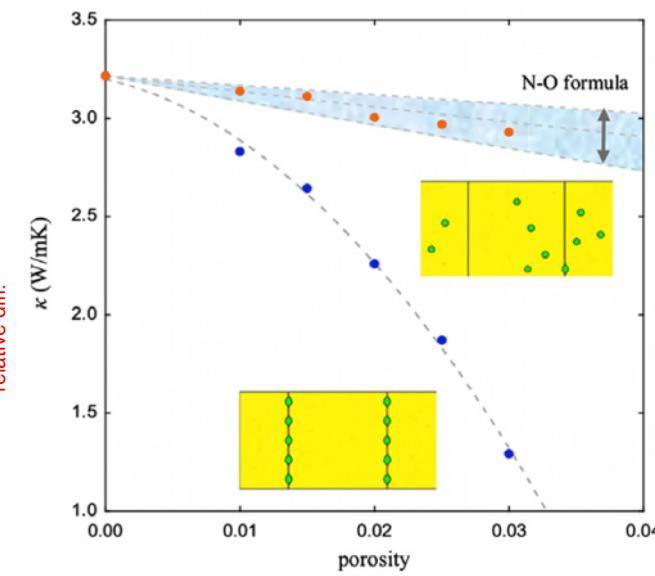
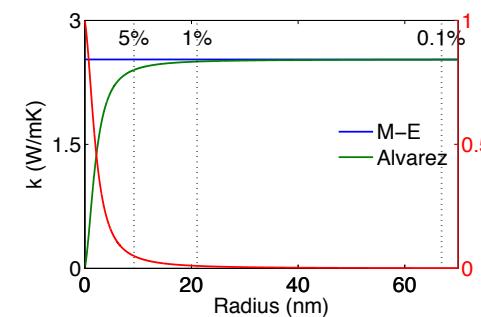
# Parametrizing the mechanistic model

- 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 GB 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

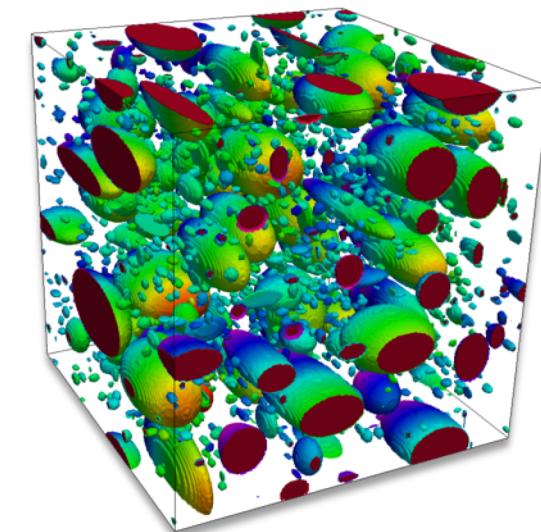
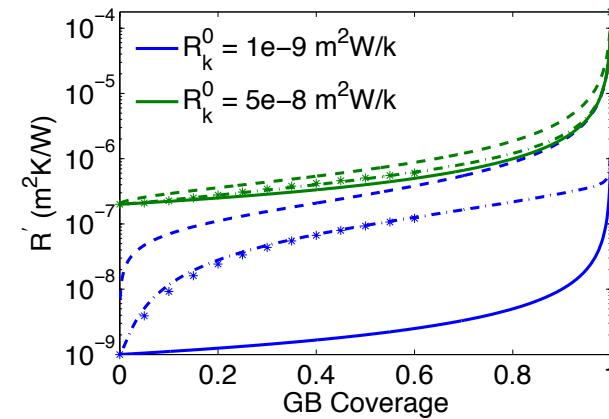
Maxwell-Eucken (no phonon scattering)

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



# Parametrizing the mechanistic model

- A thermal resistor model is created to describe the impact of GB bubbles on the thermal conductivity
- MARMOT simulations are currently being used to inform the development of the precipitate multiplier



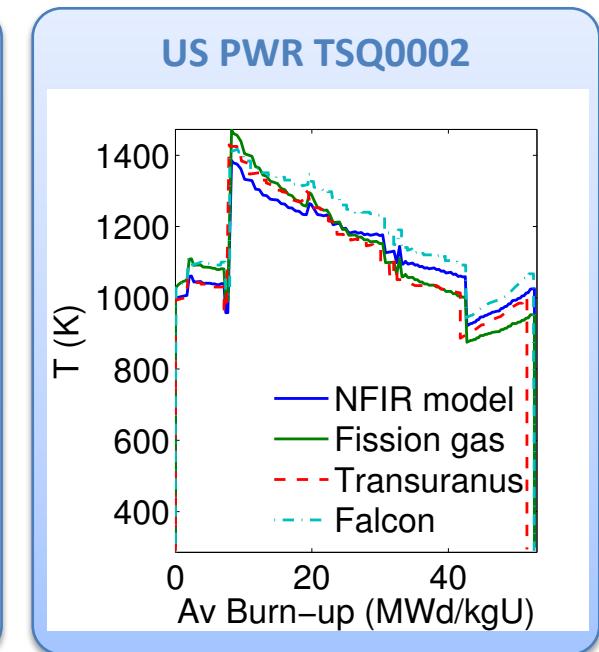
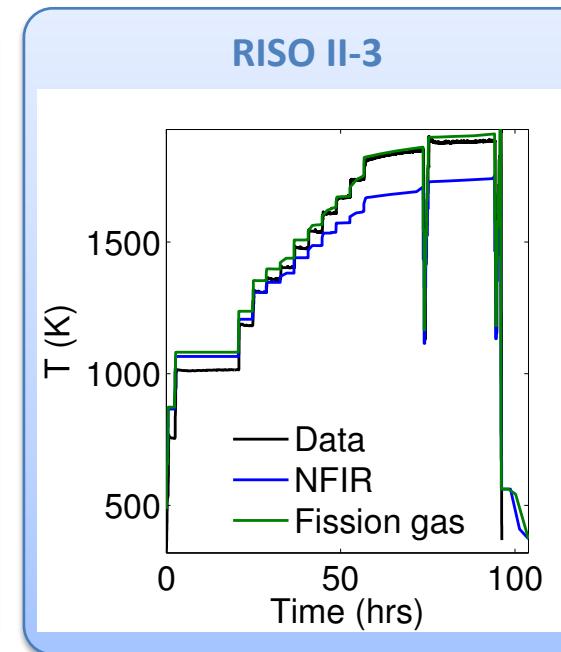
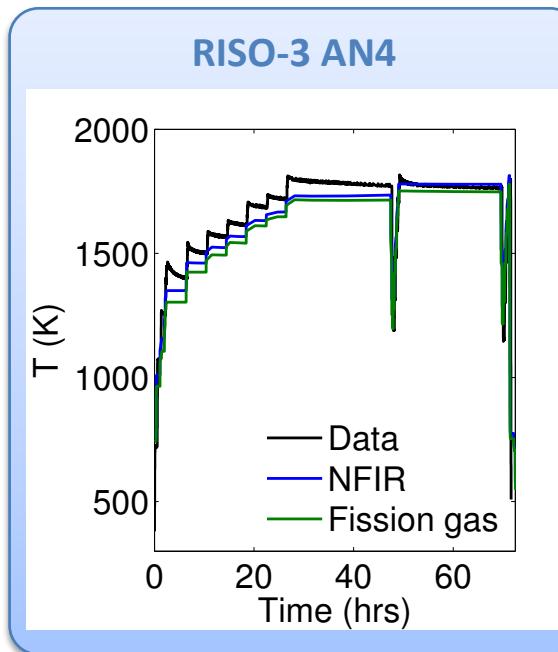
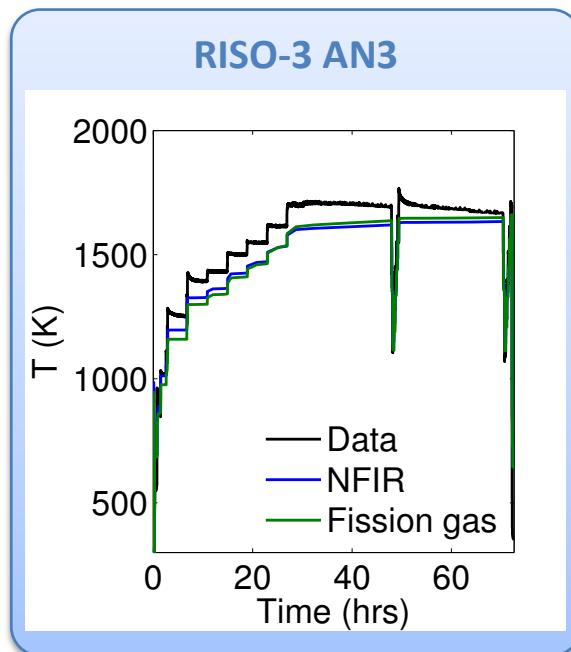
# Parametrizing the mechanistic model

- 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 precipitates fission products and individual point defects are neglected in the model, as they are not tracked or predicted in BISON

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

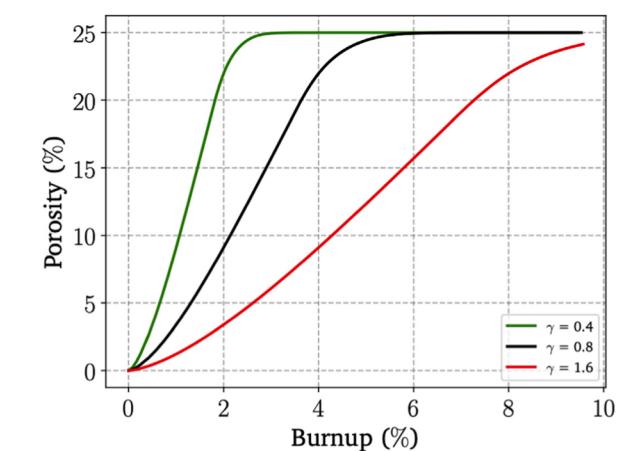
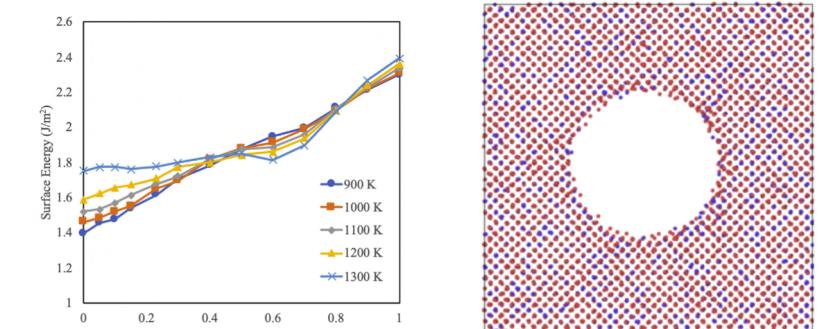
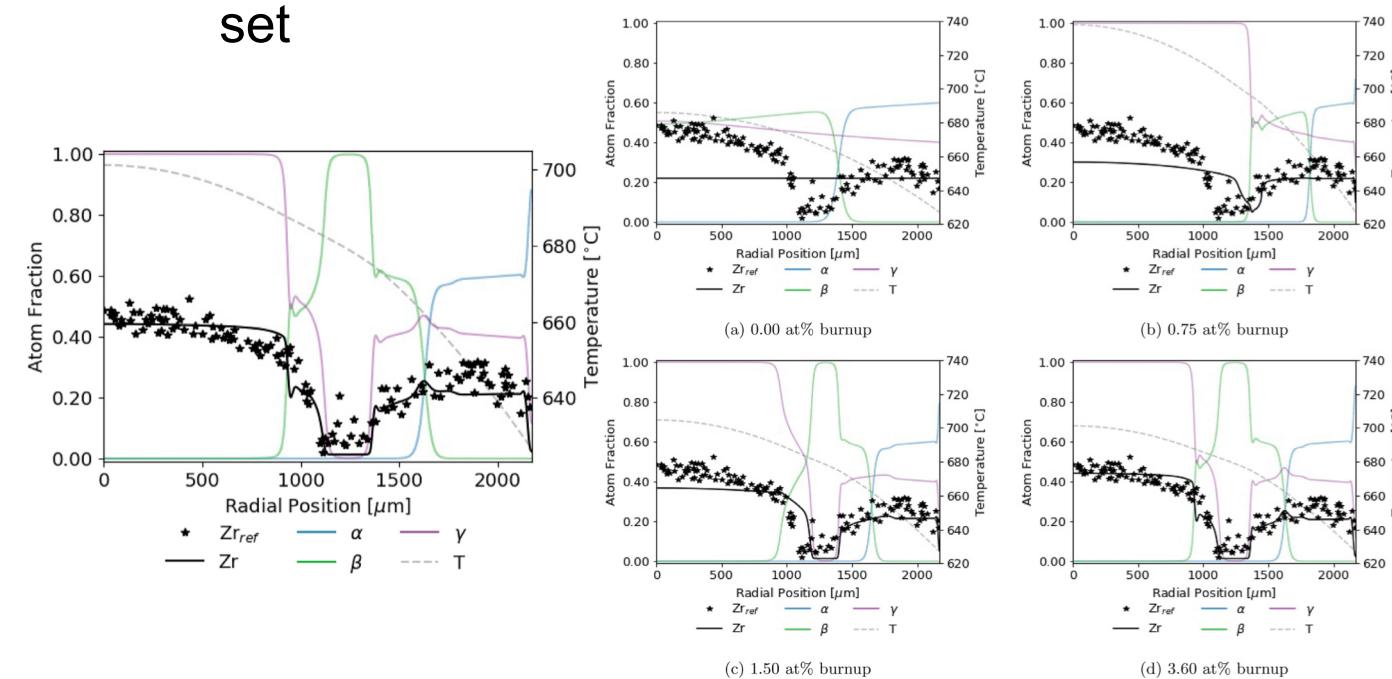
# Comparing with experiments...

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



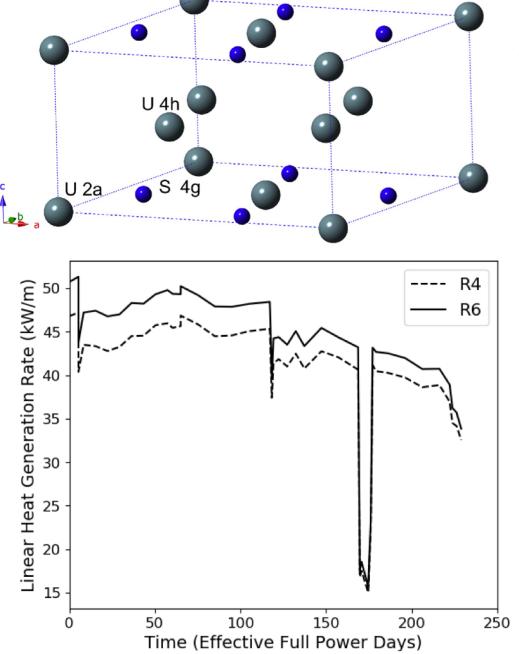
# UZr Fuel Performance Modeling

- 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

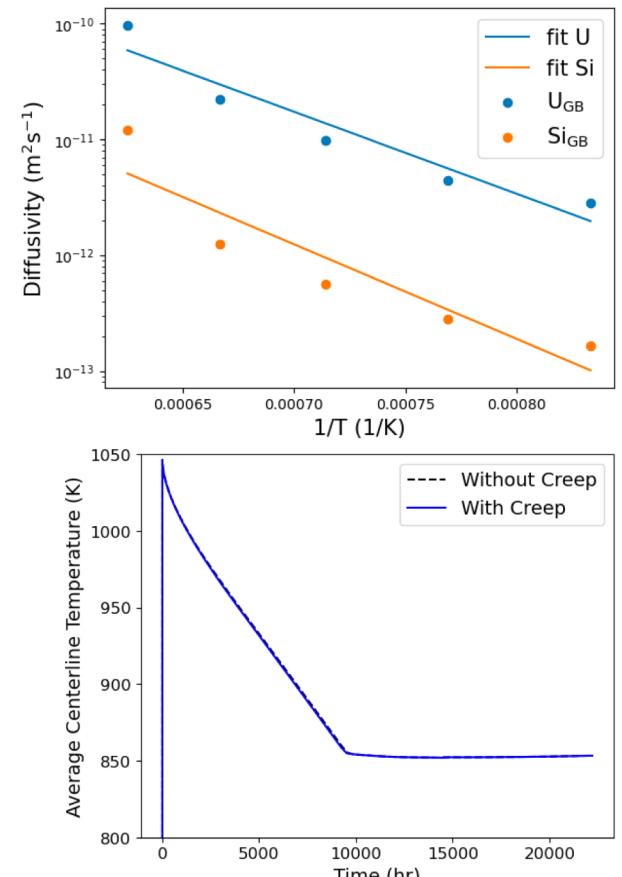
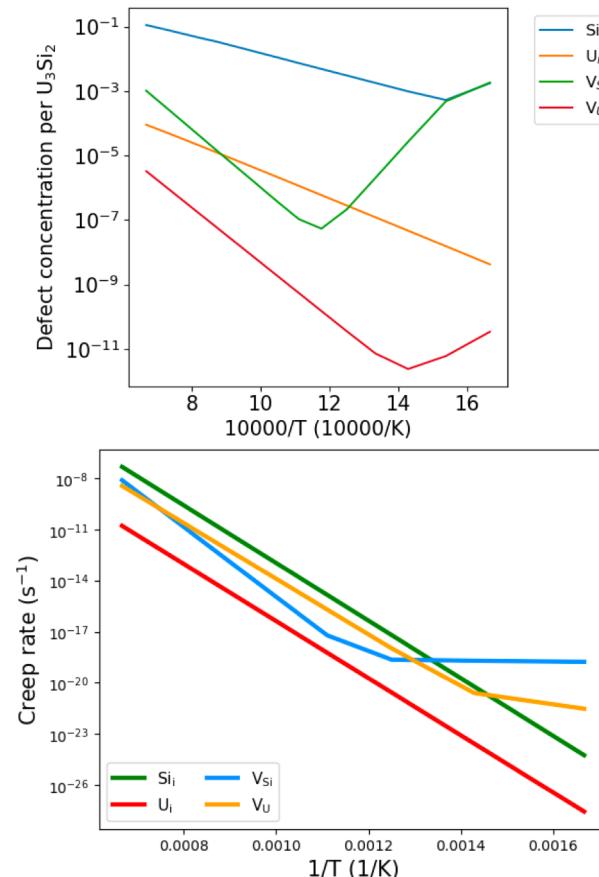


# U<sub>3</sub>Si<sub>2</sub> Fuel Performance Modeling

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



	BISON				Experiment	
	R4		R6		R4	R6
Fuel elongation (mm)	Stoichiometric	Si-Rich	Stoichiometric	Si-Rich	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<sub>2</sub> only, not for UC or UCO fuel kernels

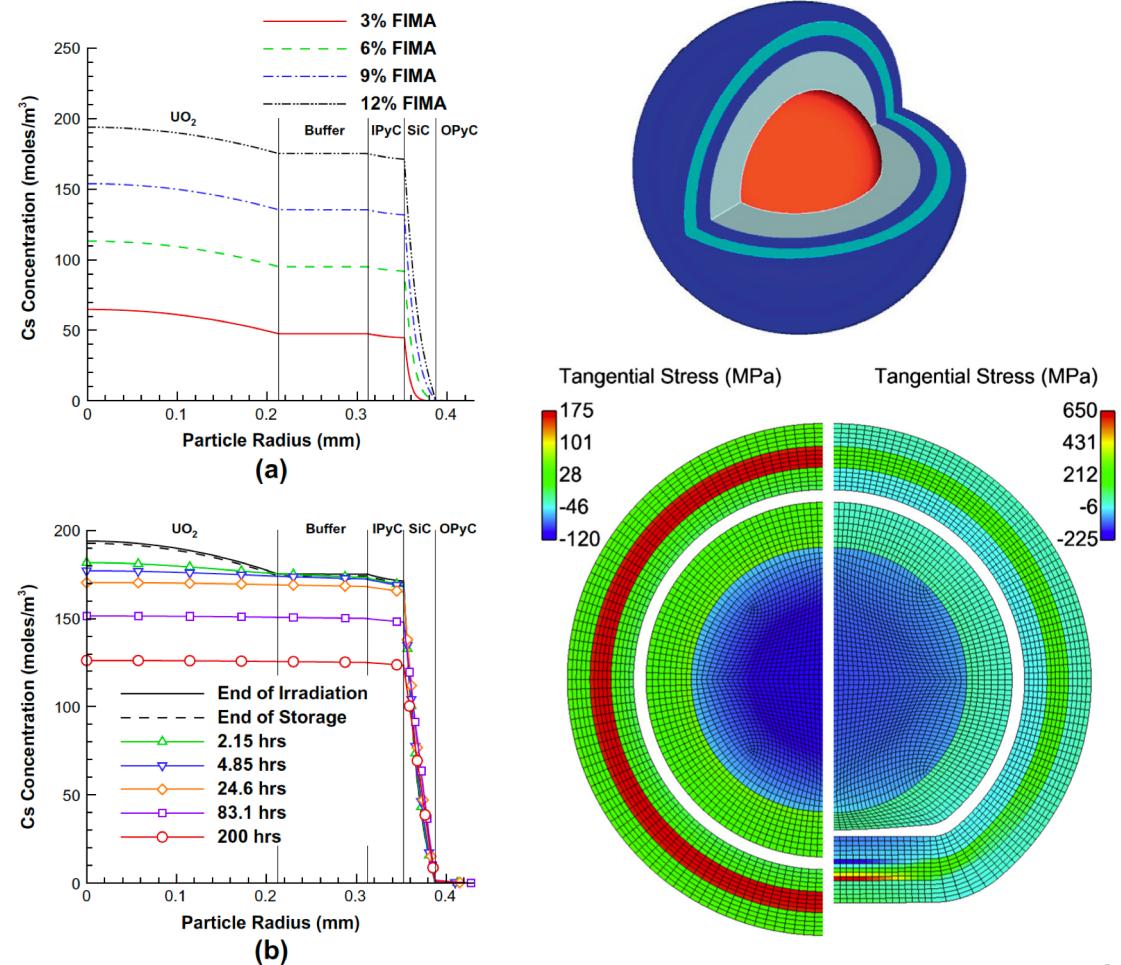
$$\frac{\partial C}{\partial t} + \nabla \cdot \mathbf{J} + \lambda C - S = 0,$$

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

**Table 6**

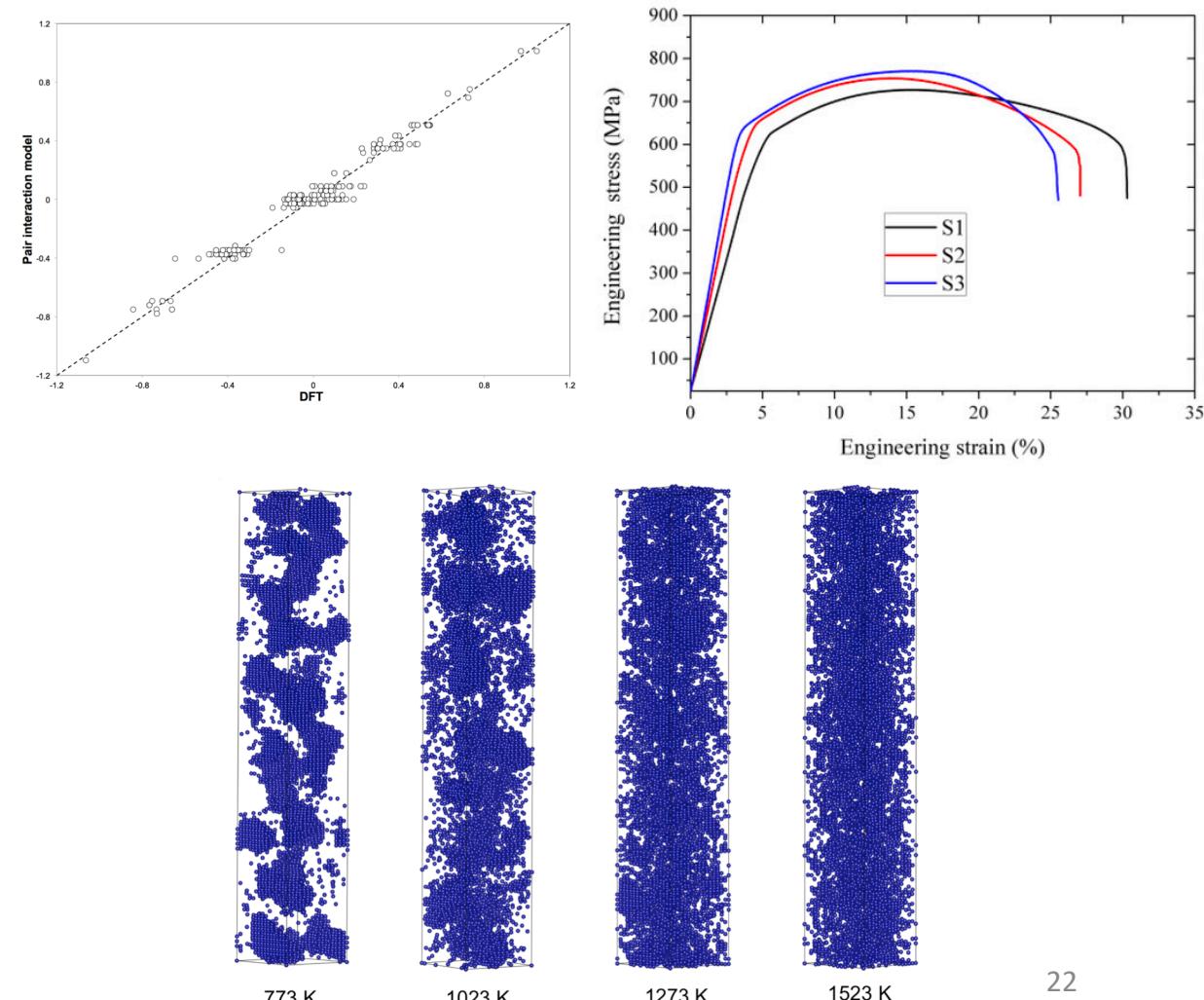
Cs diffusion coefficient parameters from [11] for use in Eqn. (5). Note that  $\Gamma$  is the fast neutron fluence ( $\times 10^{25} n/m^2$ ).

Material	$D_1$ (m <sup>2</sup> /s)	$Q_1$ (kJ/mol)	$D_2$ (m <sup>2</sup> /s)	$Q_2$ (kJ/mol)
UO <sub>2</sub>	$5.6 \times 10^{-8}$	209	$5.2 \times 10^{-4}$	362
Buffer	$1 \times 10^{-12}$	0	0	0
PyC	$6.3 \times 10^{-8}$	222	0	0
SiC	$5.5 \times 10^{-14}(e^{\frac{f}{T}})$	125	$1.6 \times 10^{-2}$	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 investigation 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
- By incorporating underlying physics, models can be predictive, even outside of existing experimental data

# Class Wrap-up

- Optional project due tonight
- Will have exam on final day of class: April 29
- Problem session on Tuesday April 27
- MOOSE project due April 30
- Please complete class evaluation
  - if there is information or comments that you would like to contribute, and that are not covered in the class eval, please send me an email
- Hope that you all have gotten an overview of fuel performance: the underlying thermo-mechanics, some key microstructural issues, core concepts required for fuel performance modeling, and hand on experience with a state-of-the-art code
- Thanks for bearing with me through this all-virtual course!