

Nuclear Fuel Performance

NE-533

Spring 2023

Housekeeping

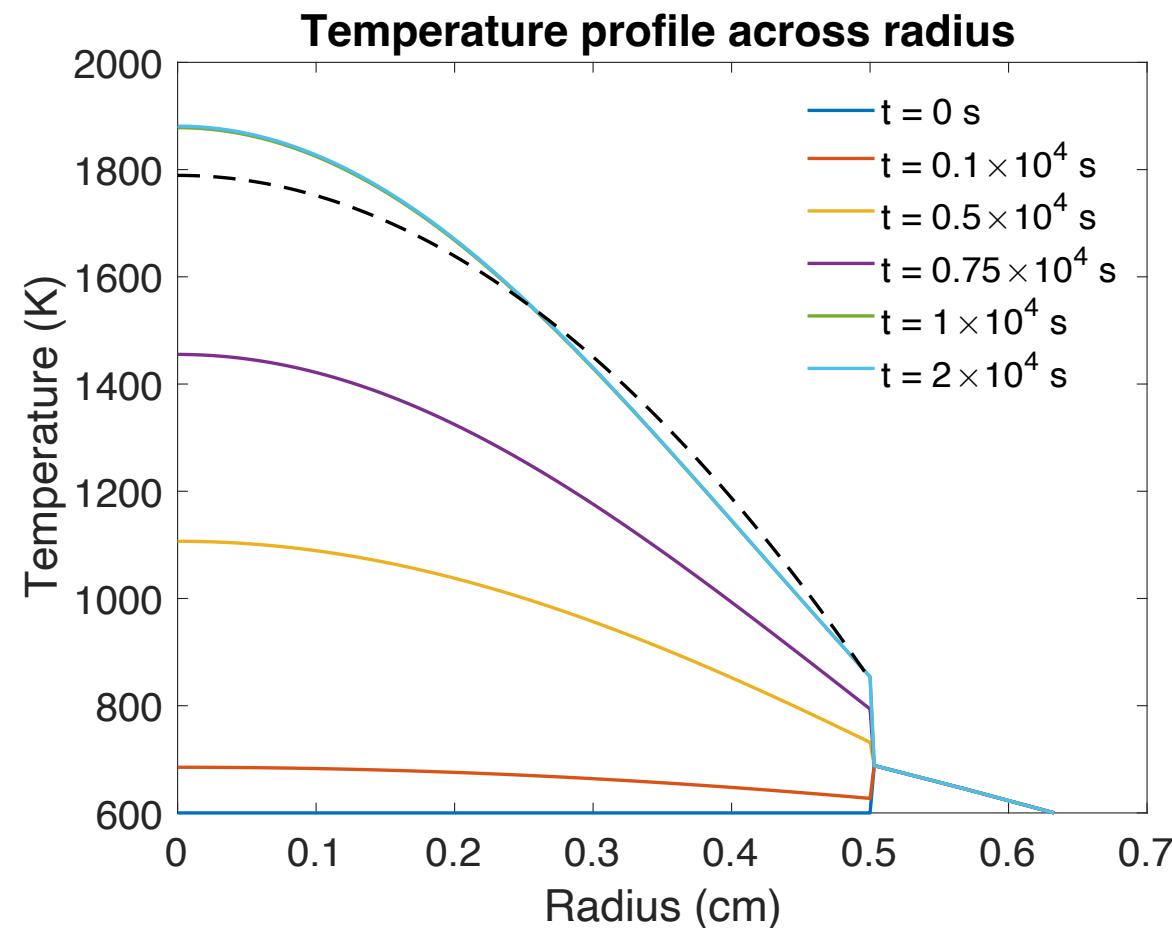
- Gone on Wed-Fri, will have a guest lecture via zoom on Thursday from Dr. Larry Aagesen (INL) – will be in class, also, your choice
- MOOSE project part 1 due on Friday
 - submission is available on Moodle
 - closes 11:59 pm on Feb 24
- This is the last lecture of the second module, topics up to Thursday's lecture will be covered on the exam, which will be on March 2 (next Thursday)
- Problem session next Tuesday, plus start module 3

Last Time

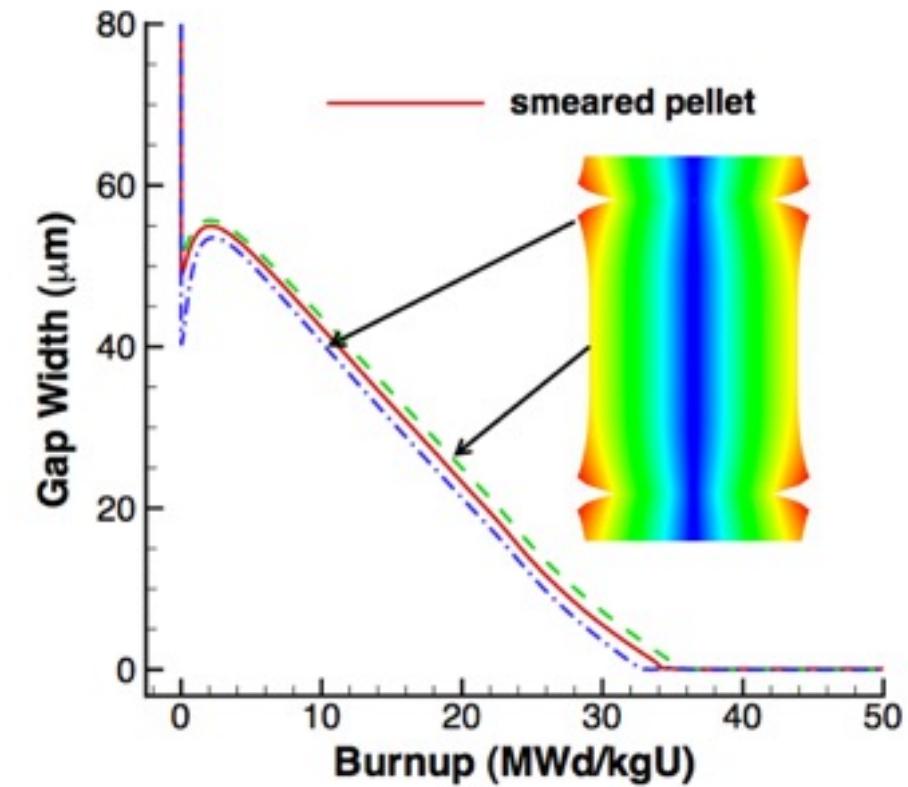
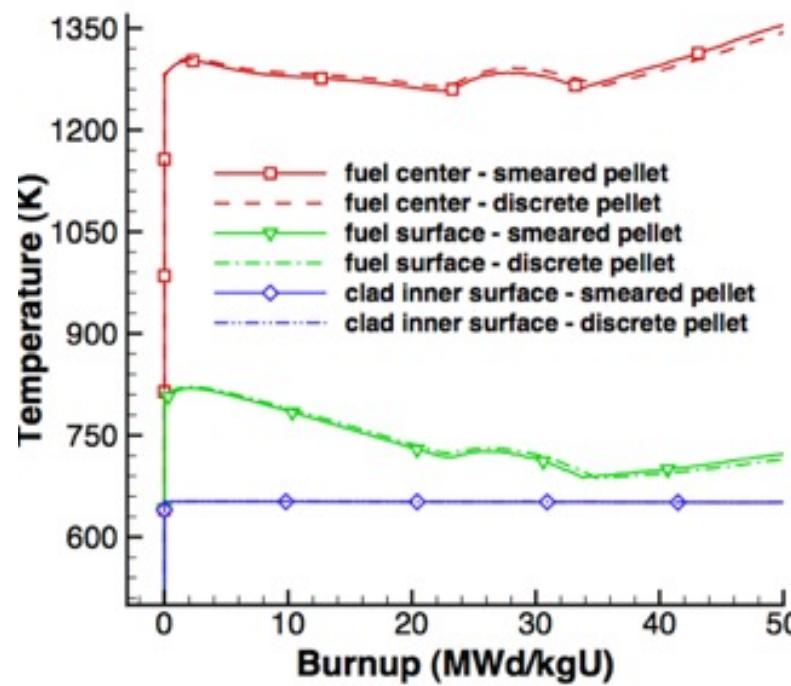
- Gap size changes due to thermal expansion
- Often have displacements instead of strains, and can solve for stress via displacements
- All fuel performance codes must:
 - Numerically model the temperature in the fuel
 - Numerically model the stress in the cladding
 - And consider gap pressure, closure, and heat transfer in some way
- Identified and talked through the primary US fuel performance codes

MATERIAL PROPERTY EVOLUTION

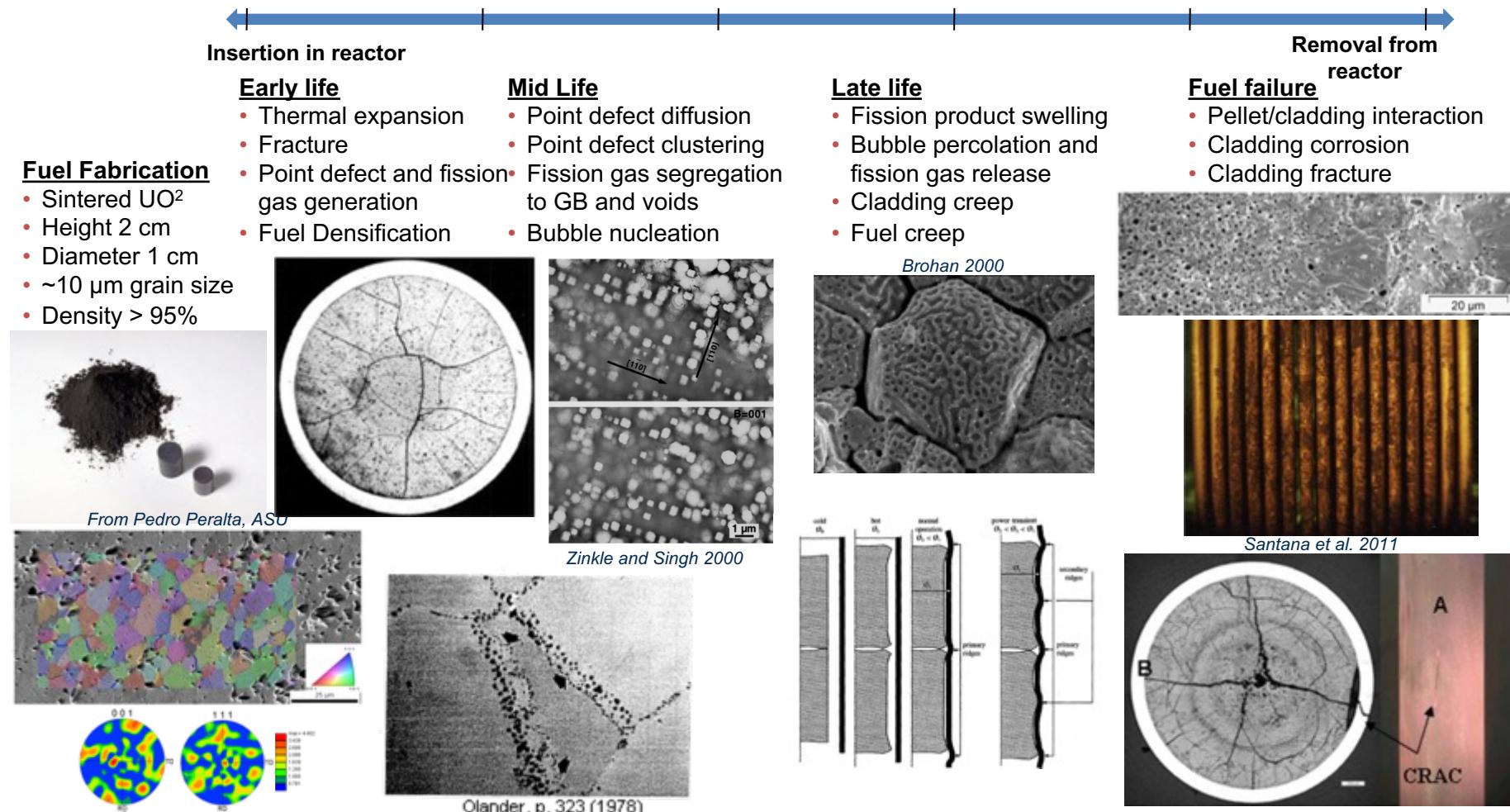
Pretty much everything we have learned so far, nothing changes once the fuel and cladding reach steady state



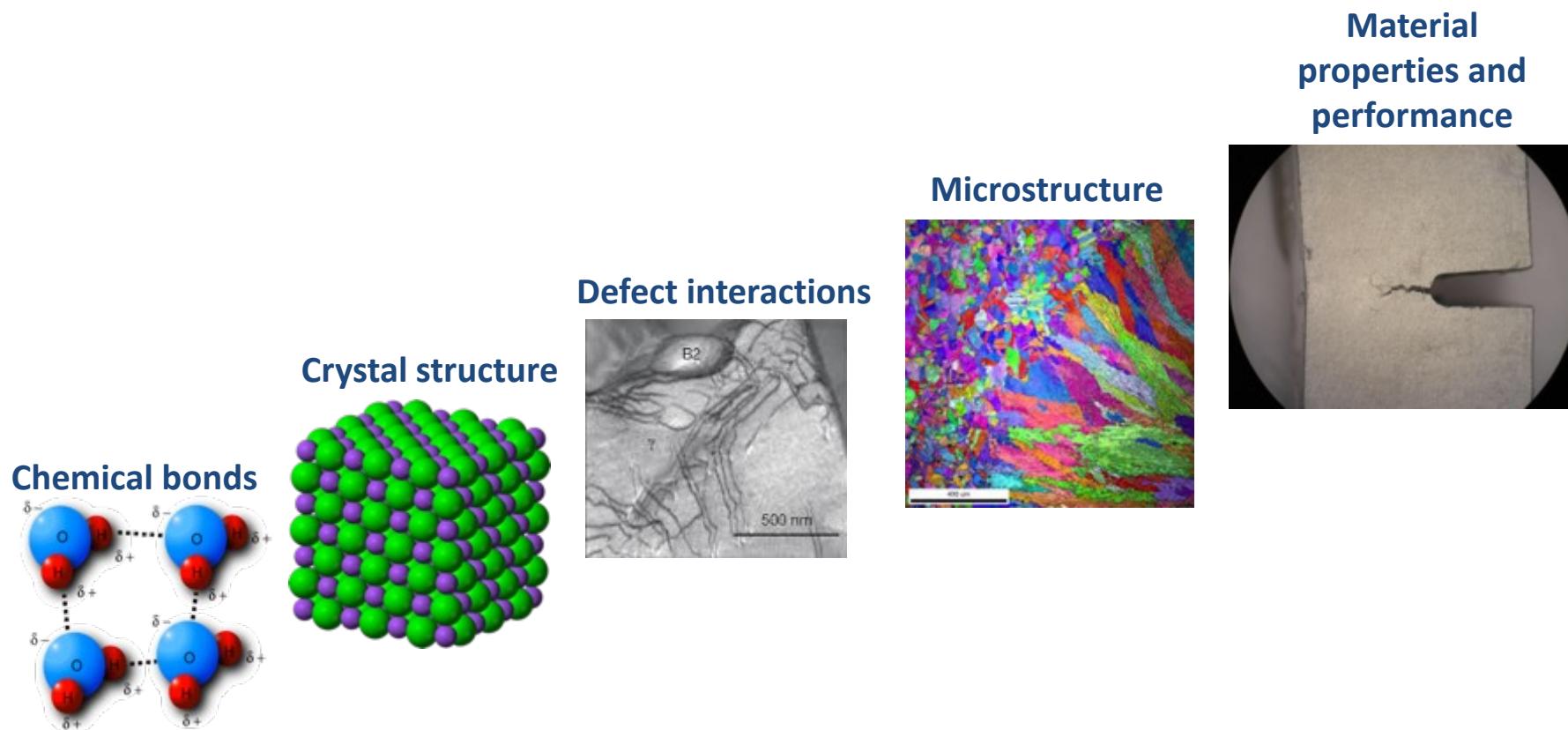
Fuel is dynamic!



Property changes due to microstructure evolution during reactor operation must be considered

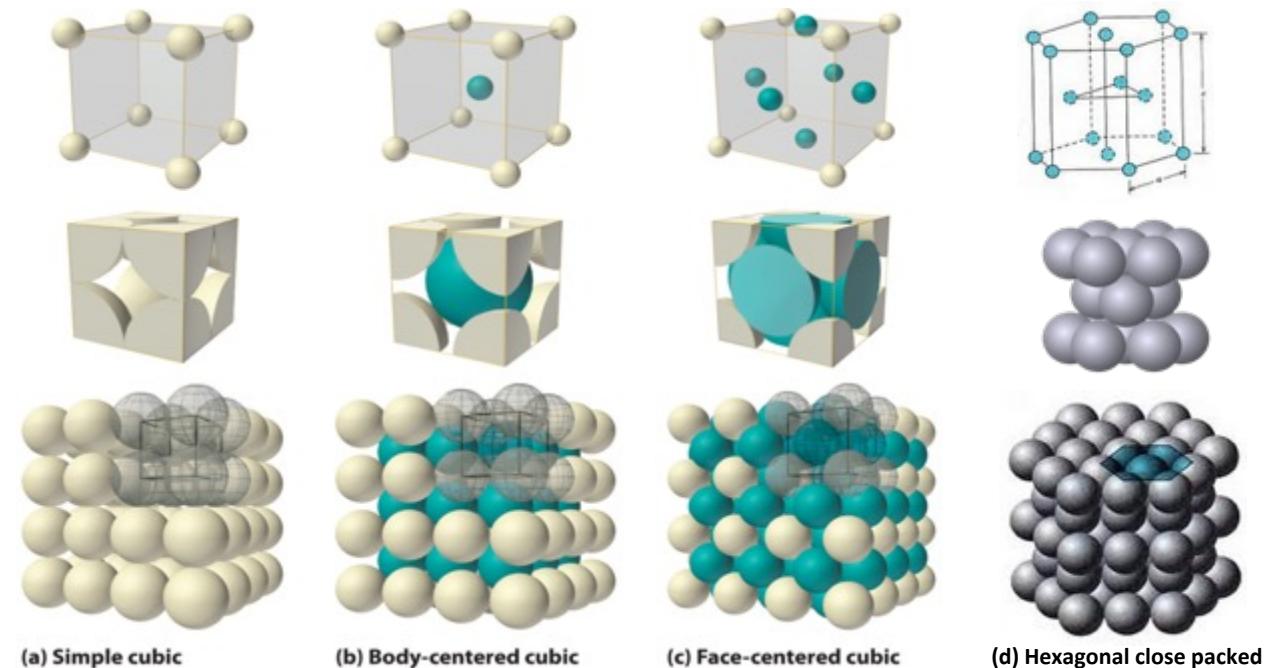


The properties and performance of a material are a result of factors across various length scales



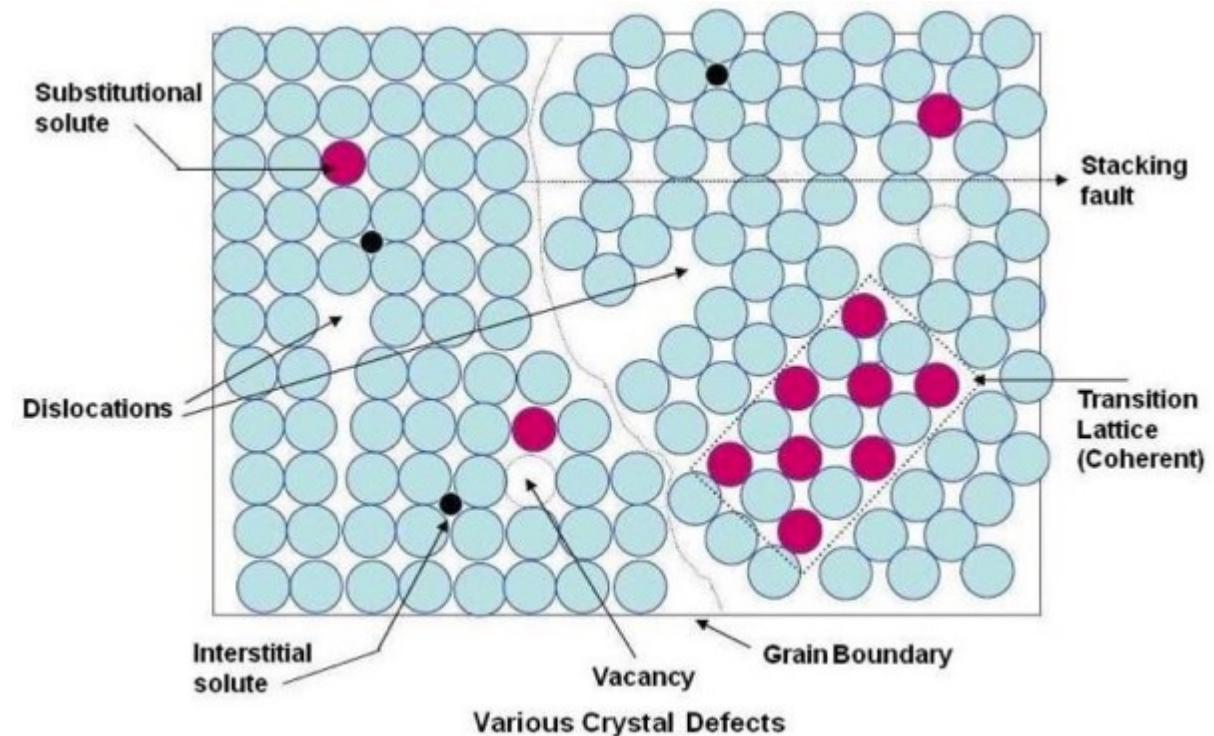
The crystal structure of a material is the shape of the ordered structure of the atoms

- The atoms of many materials form an organized lattice, these are called **crystalline materials**. All reactor materials are crystalline.
- The configuration of the atoms in the lattice impacts the properties of the material (density, thermal expansion coefficient, elastic modulus, etc.)
- All major light water nuclear materials are either cubic or hexagonal



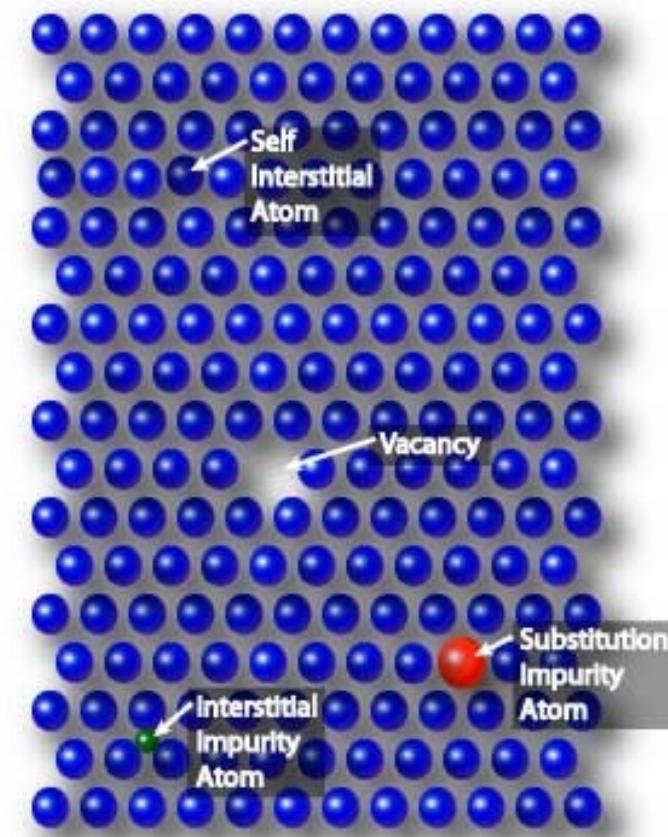
Crystalline materials

- In crystalline materials, the lattices are not perfect, they have defects
- Defects can be 0D, 1D, 2D, 3D
- The interactions between defects seriously impact material properties and how they perform



Point defects

- Point defects (zero-dimensional defects) are lattice imperfections related to one or two lattice sites
- There are several types of point defects
 - **Vacancies**
 - **Self interstitial atoms (SIA)**
 - **Interstitial impurity atoms**
 - **Substitutional impurity atoms**
- Point defects control the mobility of atoms and, therefore influence all processes that depend on diffusion



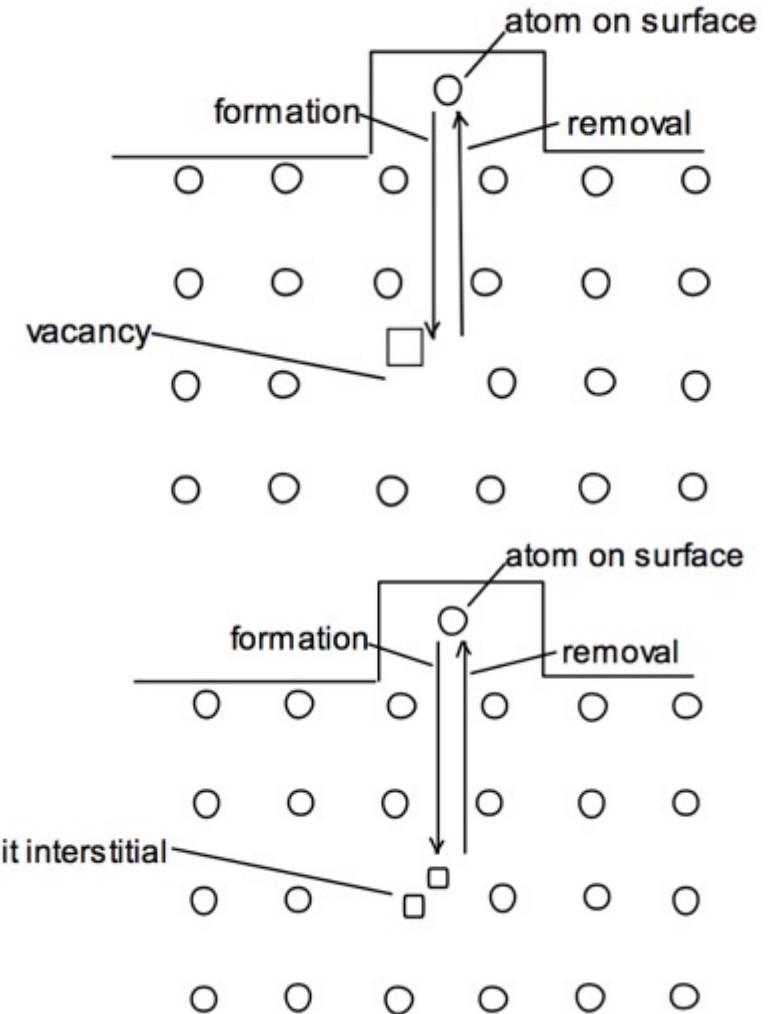
Point defects

- Vacancies and interstitials form naturally in materials
- Extra atoms can sit on a surface, creating a vacancy, or a surface can have a missing atom, creating a self interstitial atom (SIA)
- We define the number of vacancies or interstitials in terms of a concentration

$$C_v = \frac{N_v}{N_s} = \frac{\text{number of empty sites}}{\text{total number of sites}}$$

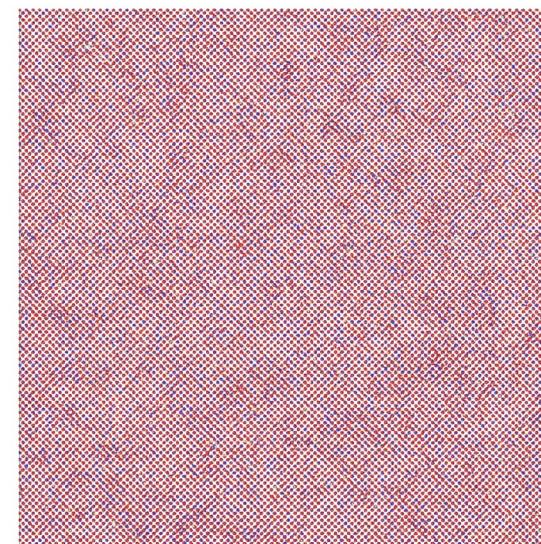
- There exists an equilibrium concentration of point defects

$$C_v = \exp\left(\frac{S_f^v}{k}\right) \exp\left(\frac{-E_f^v}{kT}\right)$$



Radiation damage

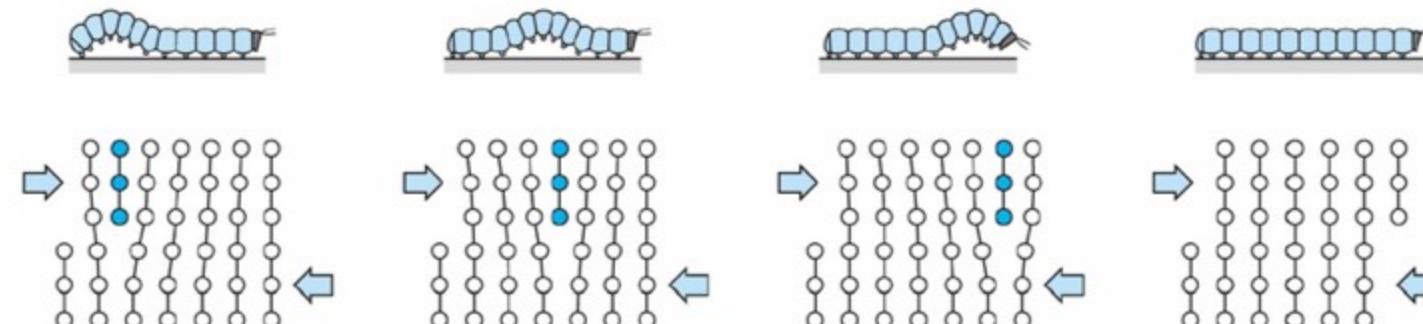
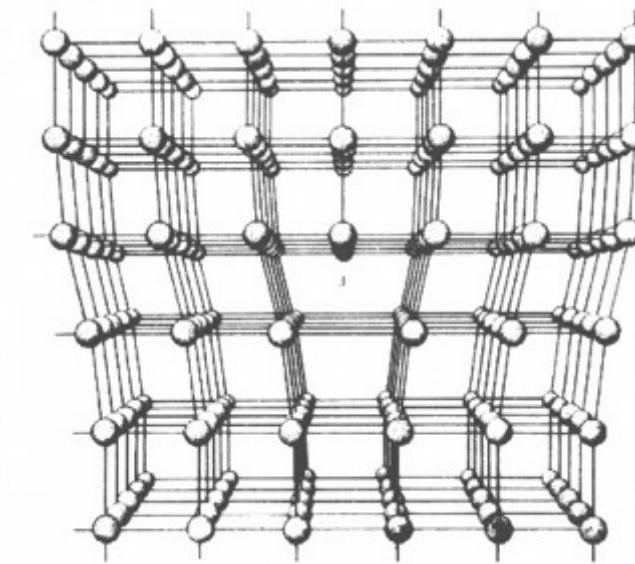
- Ionizing radiation can introduce defects into the crystal lattice, called radiation damage
- PKA collisions create a collision cascade, which results in the generation of defects (point and clusters)



Duration (ps)	Event	Result	Parameters
10^{-6}	Transfer of energy from energetic particle	Creation of PKA	$\Sigma_n(E_n, E)$
10^{-6} to 0.2	Slowing down of PKA, generation of displacement cascade	Recoil atoms Vacancies Subcascades	E_d = energy V_{NRT} = # atoms T = energy transferred
0.2 – 0.3	Thermal spike cool down	Stable interstitials, clusters, atomic mixing	$V(T)$ = # stable defects F = clustering fraction
3 – 10	Cascade cooling to bulk solid temperature	SIA depleted zone in cascade core	Loop collapse probability
> 10	Diffusion of defects and interaction with sinks	Microstructure evolution	Many

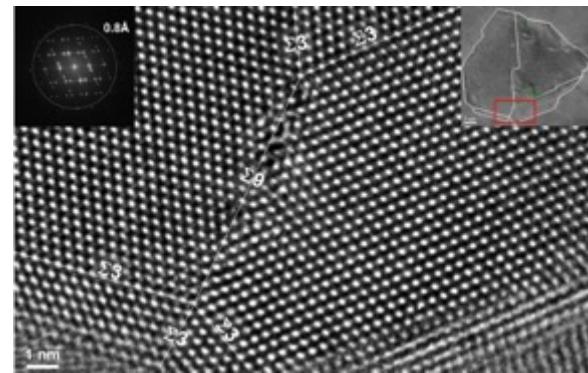
Dislocations are imperfections associated with a line of lattice sites (1D defect)

- In a dislocation, an extra half plane of atoms is inserted into the lattice
- When it moves, only a small number of bonds are broken at a time
- Dislocation motion controls the plastic (permanent) deformation of crystalline materials

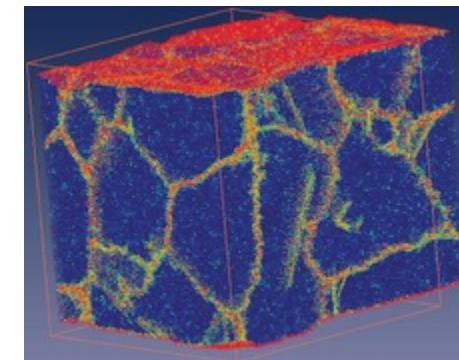


Grain boundaries

- Materials are typically composed of various regions where the crystal lattice is oriented differently. These regions are called **grains**
- When two grains meet, there is a plane of atoms that do not follow the crystal lattice called a **grain boundary**
- Most crystalline materials are polycrystalline, not single crystal

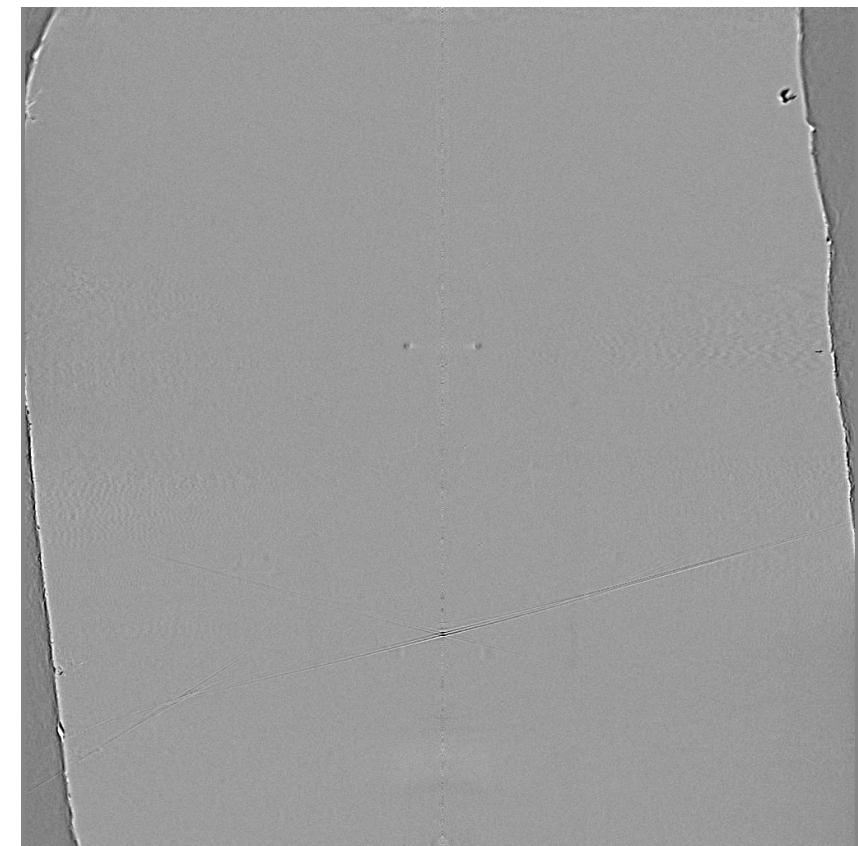


High-res transmission electron microscopy can also show individual atoms (palladium)
www.knmf.kit.edu/TEM.php



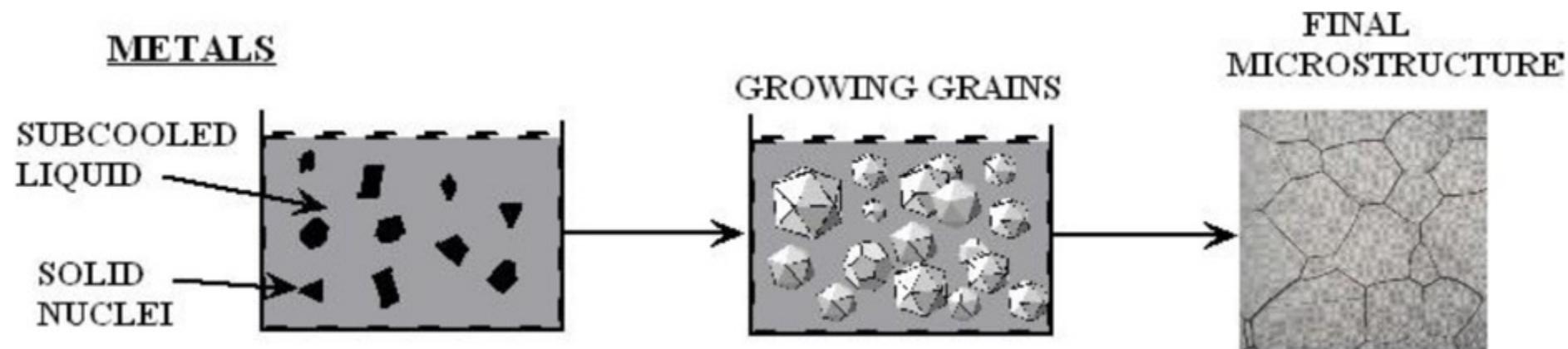
Atomistic simulation of grain boundaries in 3D

Metals are often cast, and polycrystals naturally form during casting



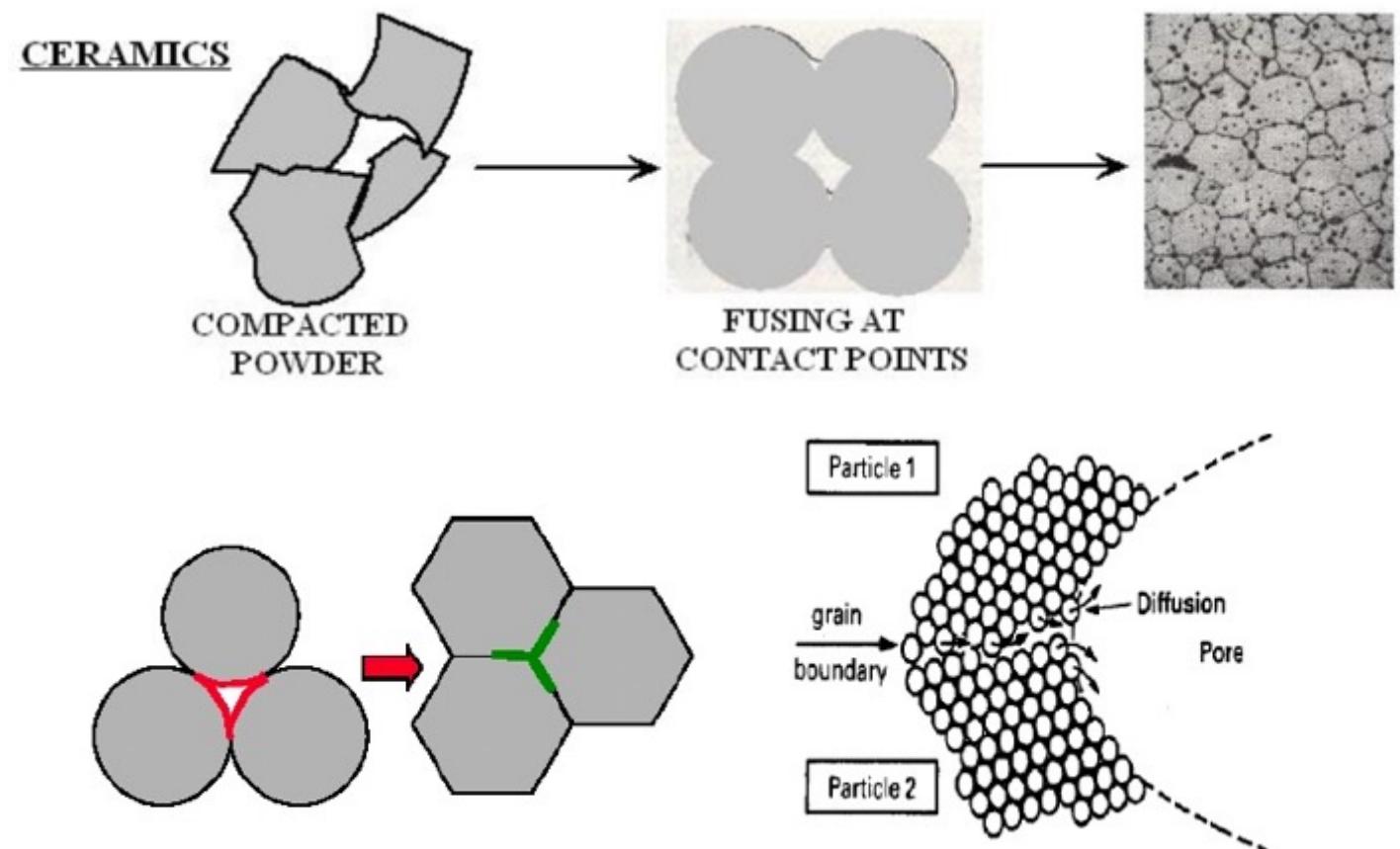
Polycrystals naturally form during casting

- Solidification begins in different regions of the melt, each with a different orientation
- Once the different regions meet, grain boundaries form between them



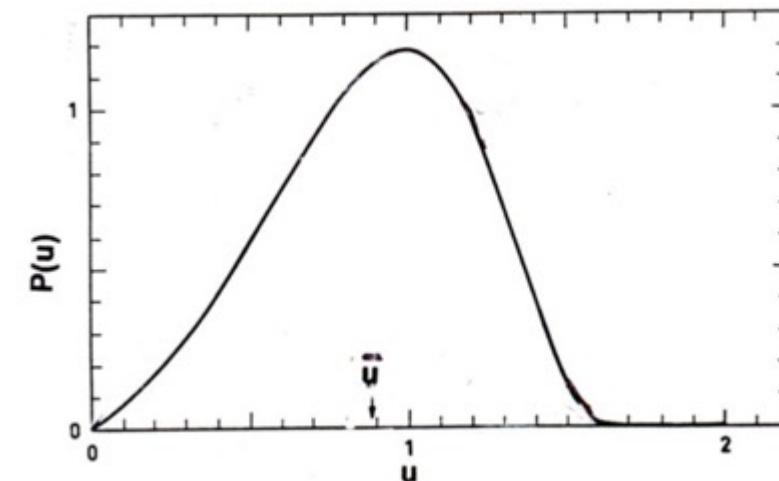
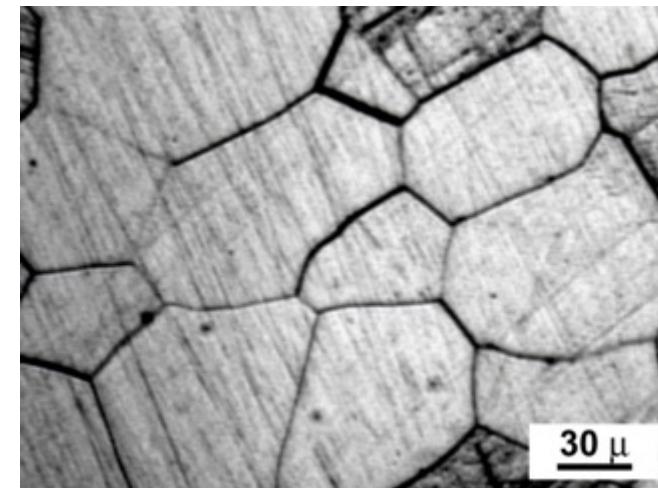
Ceramic sintering

- Ceramics are typically sintered, and polycrystals also naturally form from sintering
- In sintering, powders are compacted at high temperature
- The particles are each oriented differently, and as they fuse, grain boundaries form
- The differences between the grain orientations result in the grain boundary



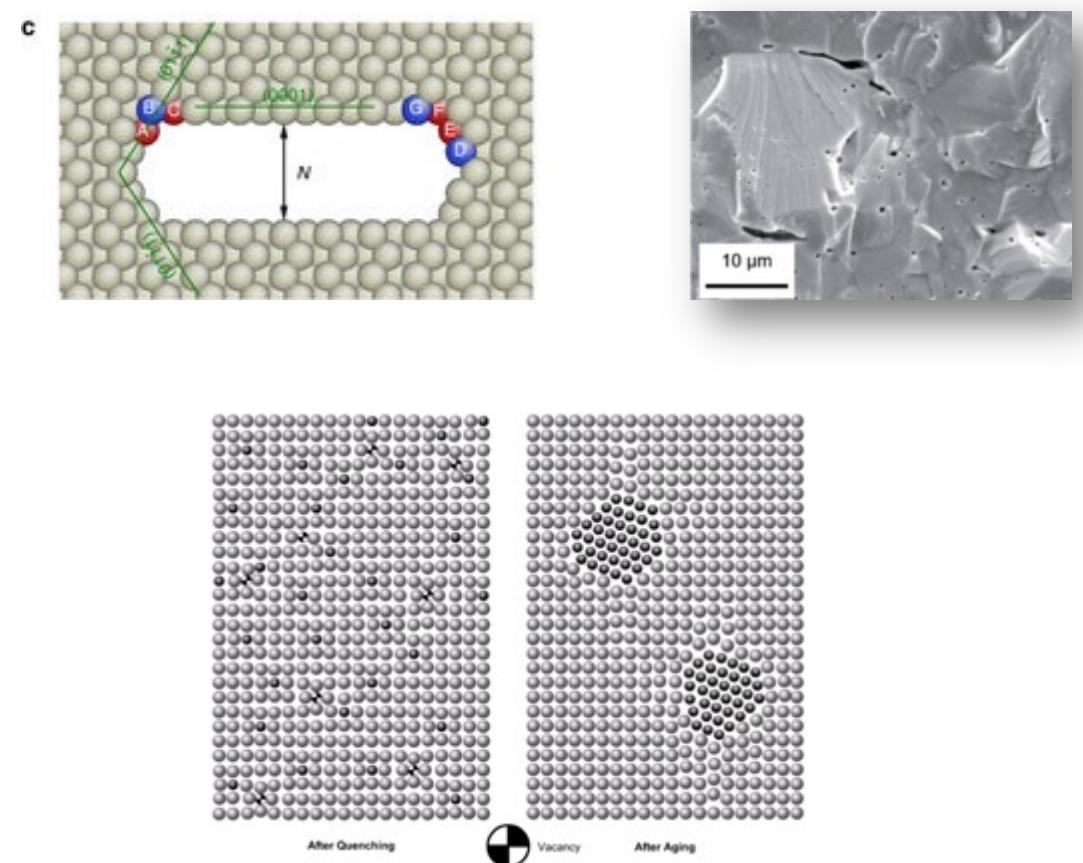
Distribution of grain sizes

- In a polycrystal, there is also a distribution of grain sizes
- Therefore, we commonly refer to the grain size distribution and the **average grain size**
- The Hillert distribution is an analytical distribution for grain size
- Real materials often vary from this behavior



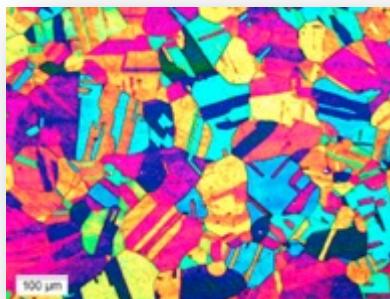
3D Defects

- When point defects cluster, they form three dimensional defects
- The energy of a point defects is reduced when several point defects cluster together
- Larger clusters of vacancies are called **voids**
- Clusters of impurity atoms are called **precipitates**

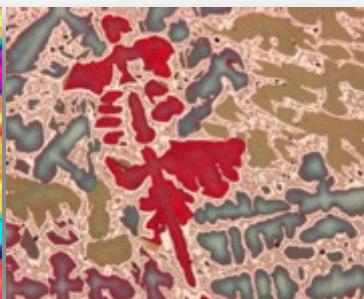


Microstructure

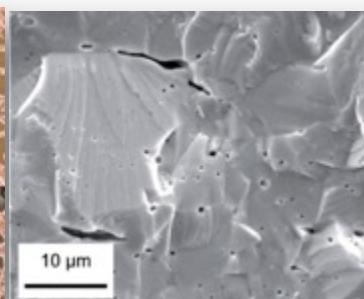
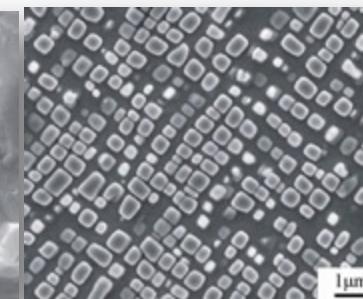
- Material microstructure is the structure observable with 25x magnification
- The microstructure includes grain structure, secondary phases, porosity, and more
- The microstructure can strongly influence physical properties such as strength, toughness, ductility, hardness, corrosion resistance, high/low temperature behavior or wear resistance.
- These properties in turn govern the application of these materials in industrial practice



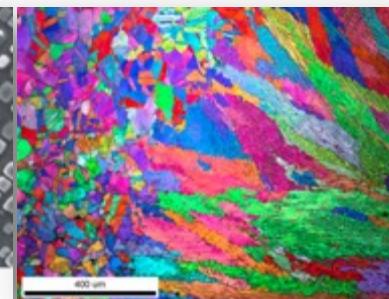
Annealed Stainless Steel



Cast Bronze

Sintered UO₂

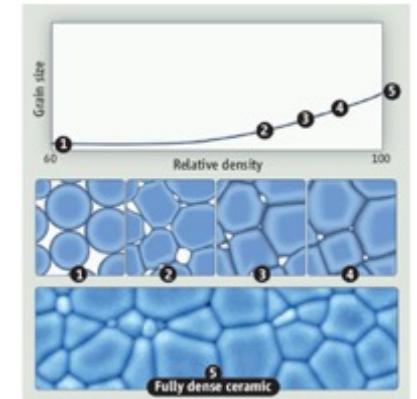
Co-Al-W Superalloy



Friction stir welded stainless steel

Material processing

- The processes we use to make a material have a huge impact on its microstructure and properties
- **Casting** – manufacturing process in which a liquid material is poured into a mold and then allowed to solidify
 - Can be used to make complex shapes
 - The solidified microstructure typically has properties that are far from ideal
- **Sintering** – Forming a solid from a powder using heat and/or pressure without melting the material
 - Applicable to metals and ceramics
 - Difficult to obtain a material that is fully dense
 - Used to make fuel pellets
- **Heat treatment** – heating or cooling a material to extreme temperatures to get desired microstructure and properties
 - Used to control the rate of microstructure change, including diffusion, grain growth, or phase change
 - Use on many types of materials (metals, ceramics, glasses)



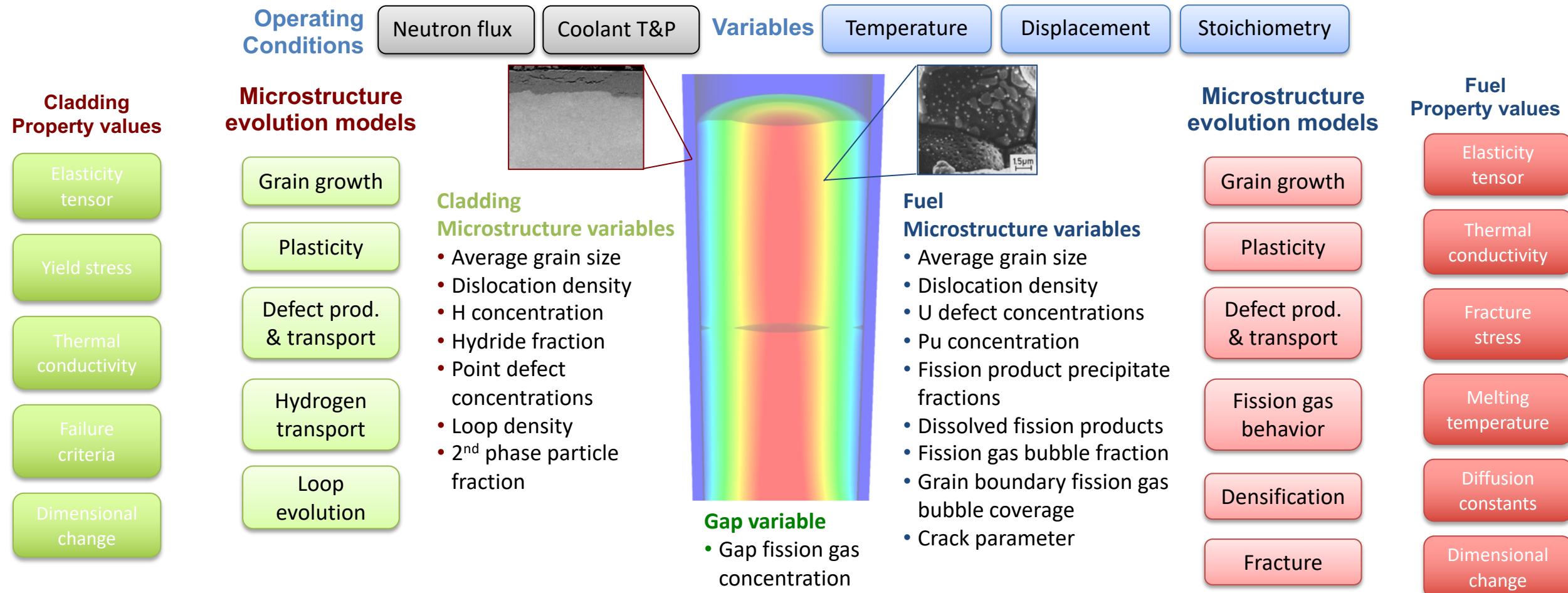
Summary

- Even during steady state operation, fuel and pellet conditions change with time due to microstructure evolution
- Atoms in the fuel and cladding materials are arranged in a crystal lattice
- The crystal lattice is never perfect; it has defects
 - Point defects include vacancies, interstitials, and impurity atoms
 - Dislocations are line defects
 - Grain boundaries are planar defects
 - Voids and precipitates are volume defects
- All materials have defects, radiation damage causes many more defects
- Microstructure can be tailored through processing

MECHANISTIC MODELING

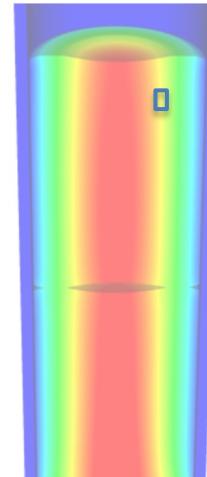
Microstructure-based fuel performance modeling

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

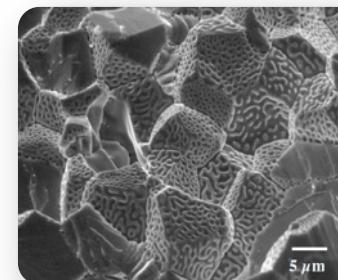


Variables

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

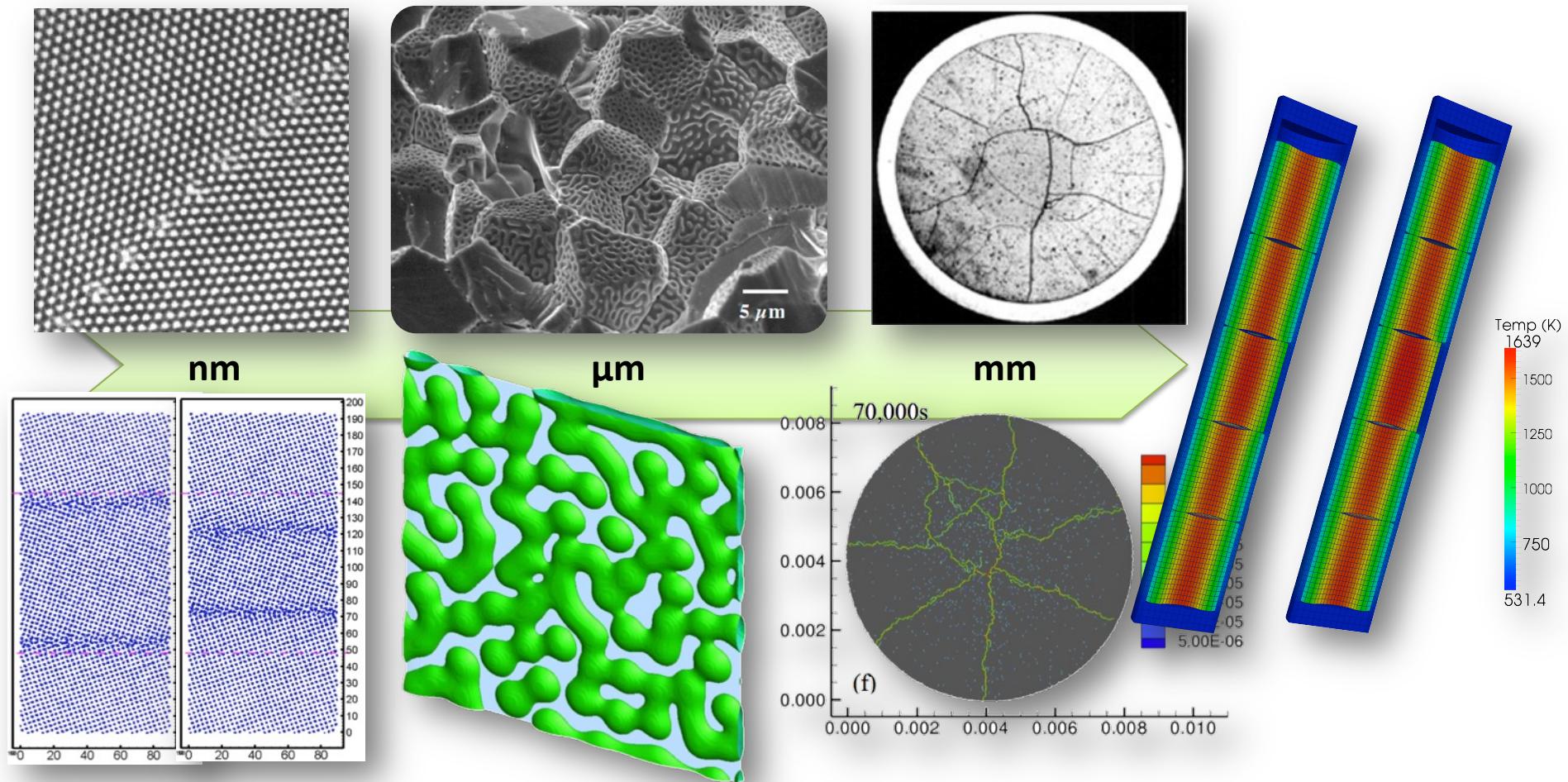
Fracture stress

Dimensional change

Gap conductance

Gap pressure

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



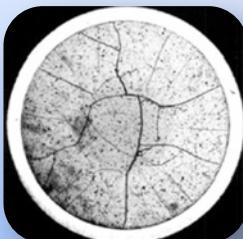
Microstructure-based models

- 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

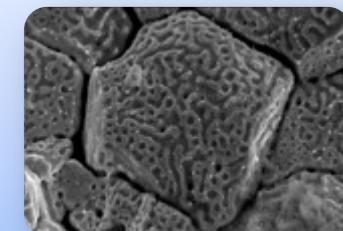
Grain boundary
and bubbles

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

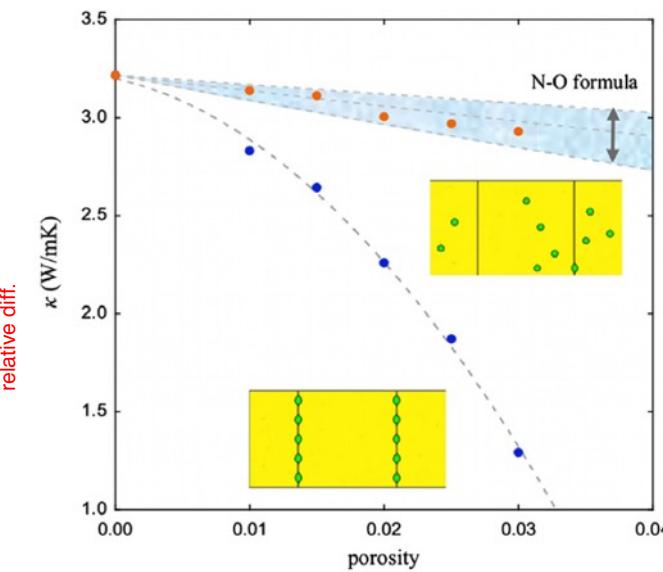
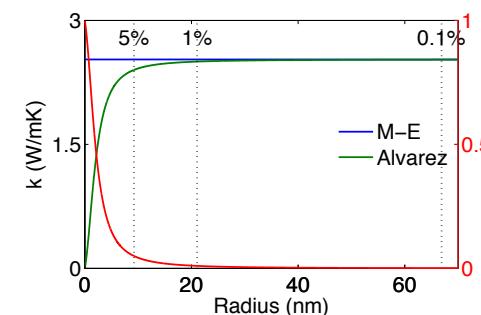
Parametrizing the mechanistic model

- 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 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

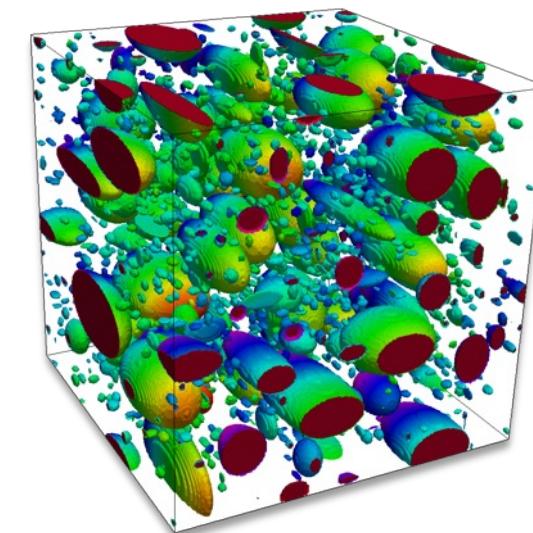
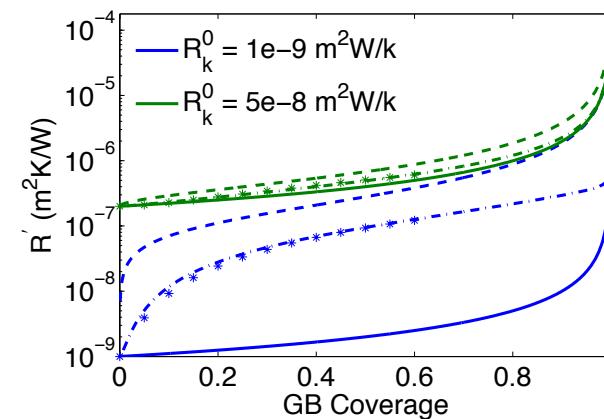
Maxwell-Eucken (no phonon scattering)

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



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



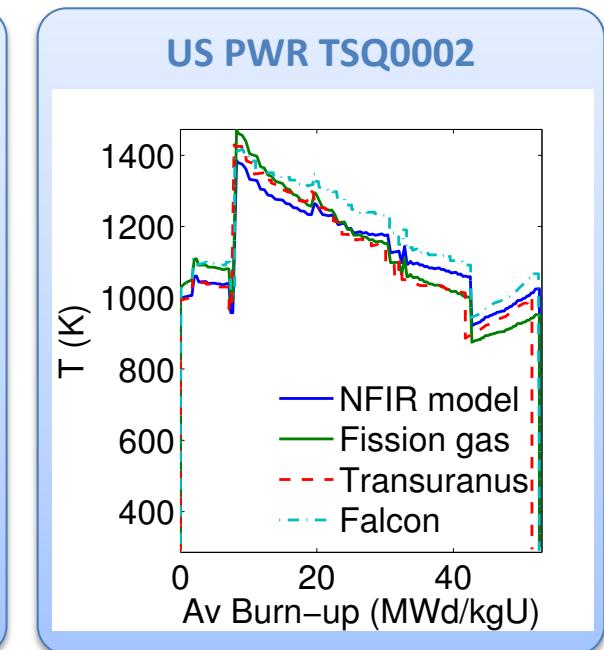
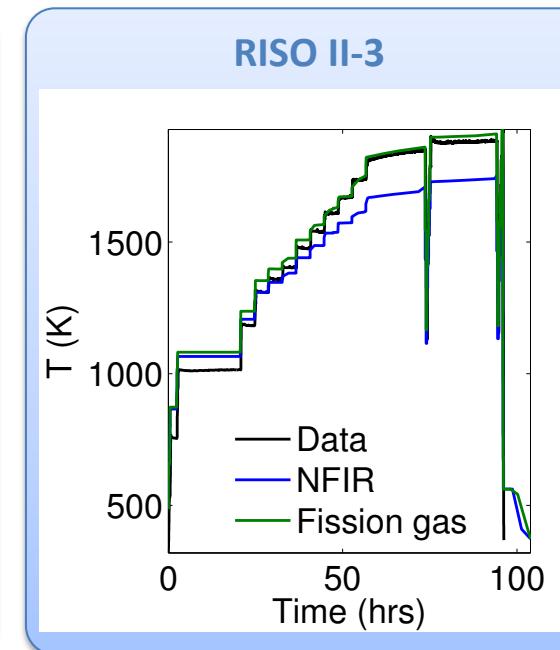
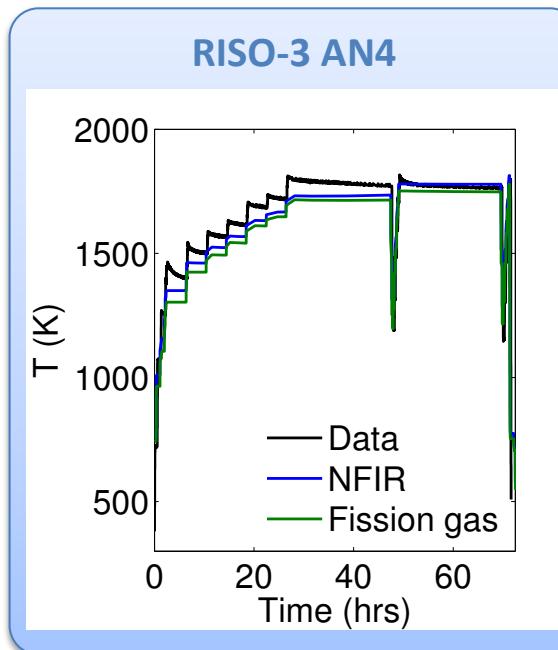
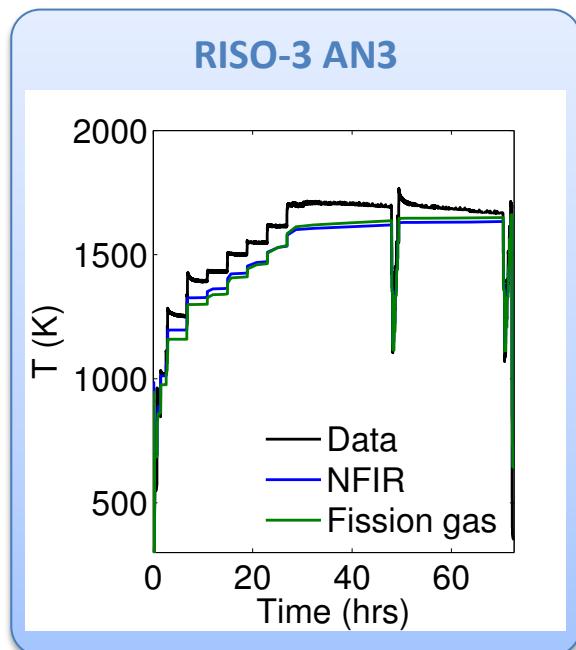
Parametrizing the mechanistic model

- 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

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

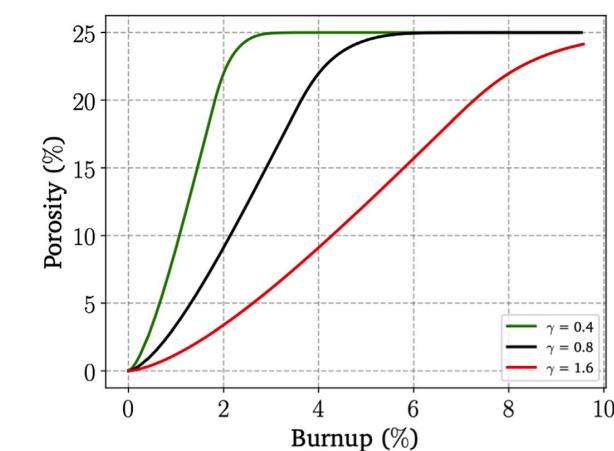
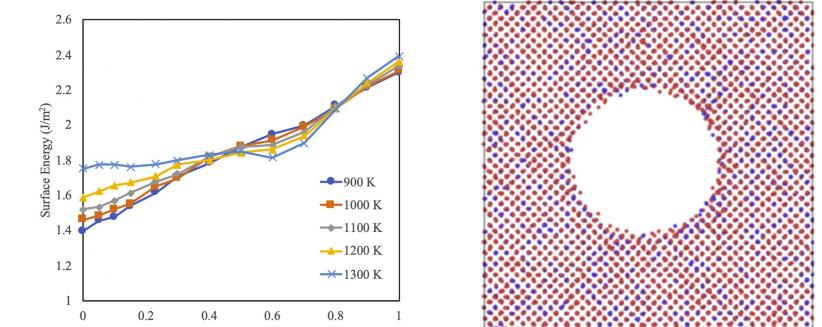
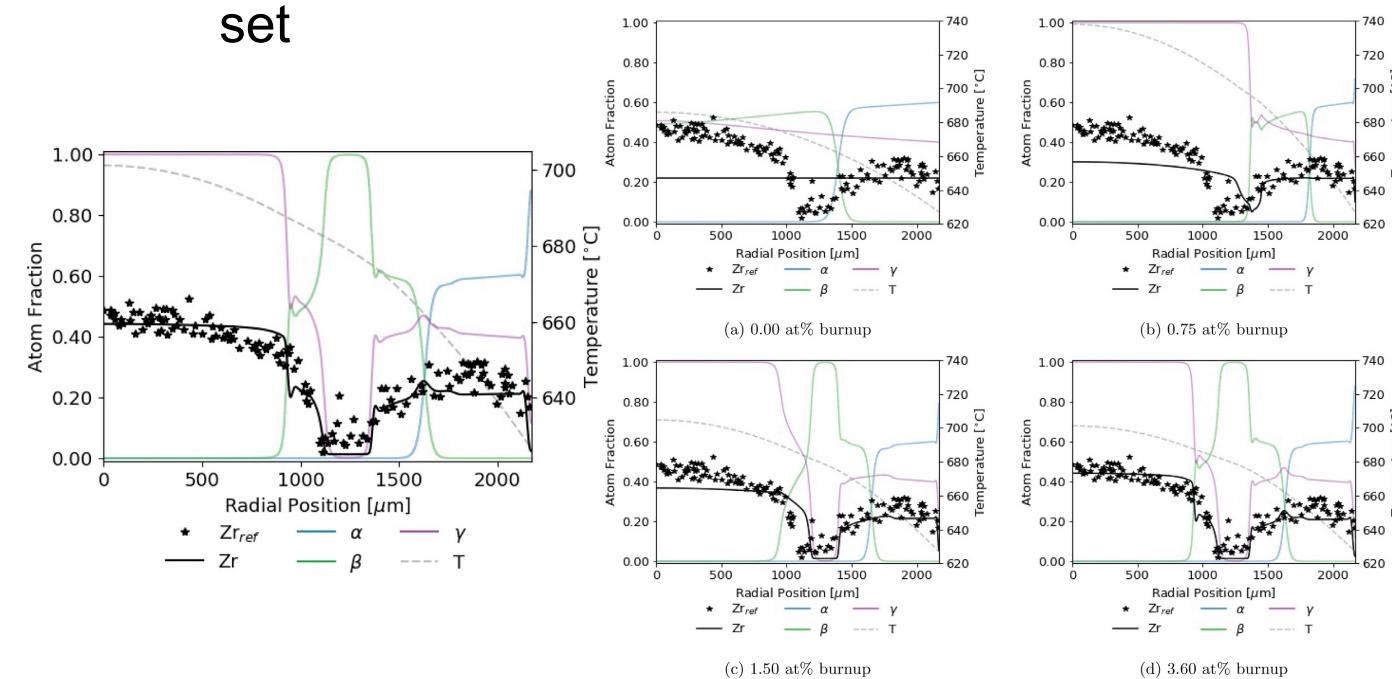
Comparing with experiments...

- 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



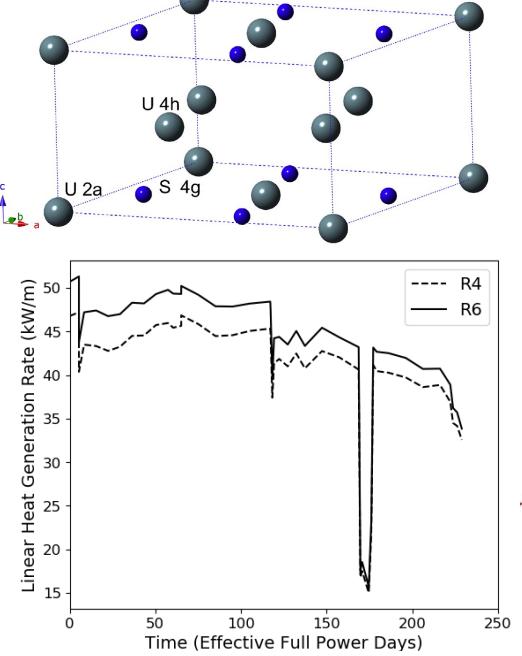
UZr Fuel Performance Modeling

- 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

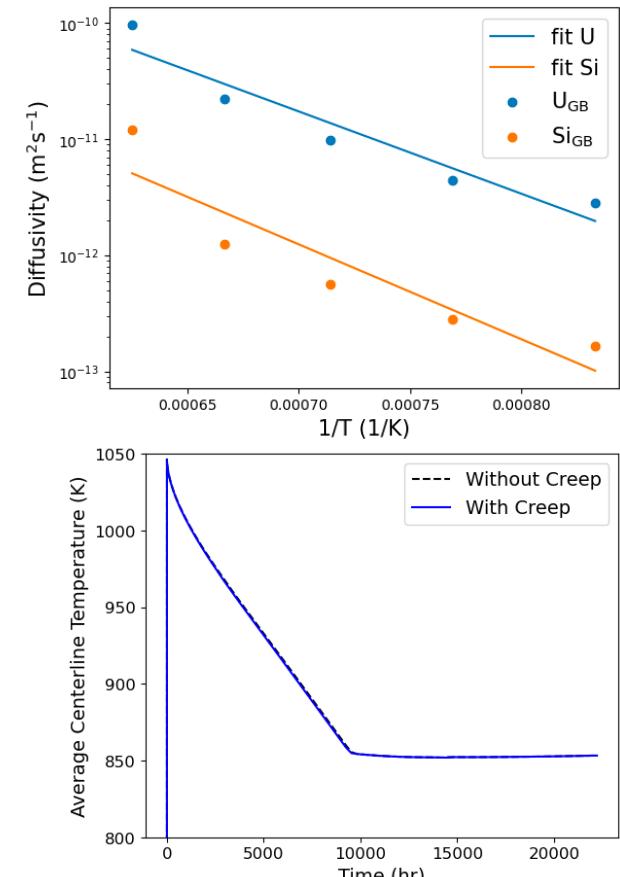
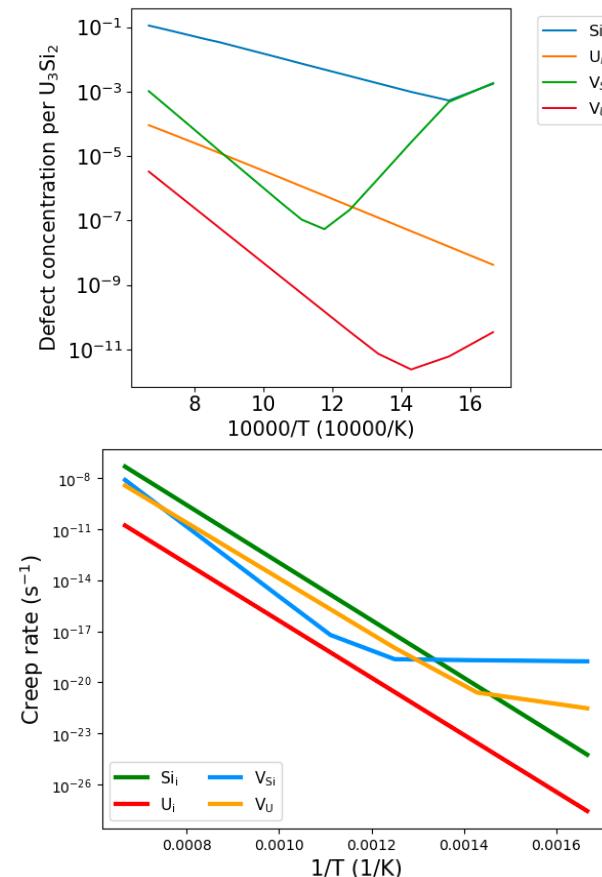


U₃Si₂ Fuel Performance Modeling

- 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

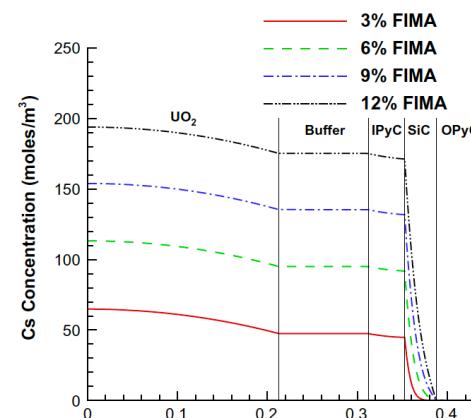
- TRISO particle modeling is still under development from a mechanistic model standpoint
- Imperfect data for many critical fission products through different layers
- Data typically for UO₂ only, not for UC or UCO fuel kernels

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

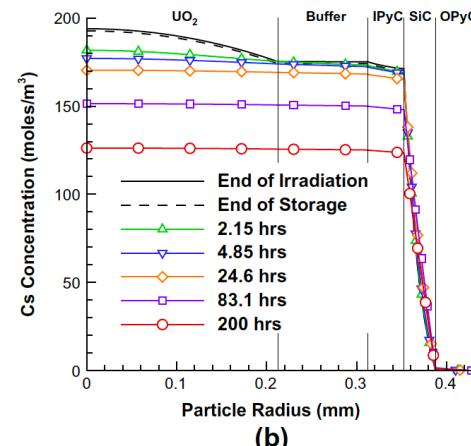
$$\mathbf{J} = -D \nabla C,$$

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

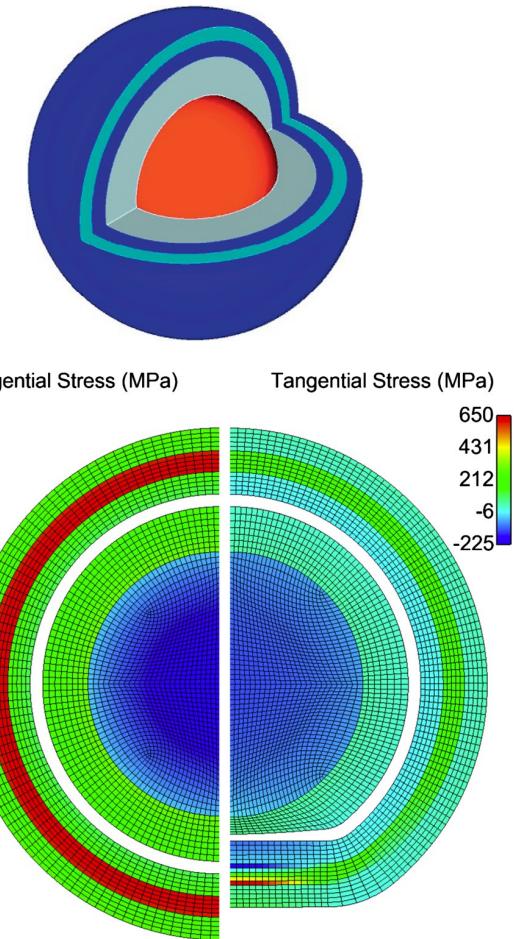
Material	D_1 (m ² /s)	Q_1 (kJ/mol)	D_2 (m ² /s)	Q_2 (kJ/mol)
UO ₂	5.6×10^{-8}	209	5.2×10^{-4}	362
Buffer	1×10^{-12}	0	0	0
PyC	6.3×10^{-8}	222	0	0
SiC	$5.5 \times 10^{-14}(e^{4.75})$	125	1.6×10^{-2}	514



(a)

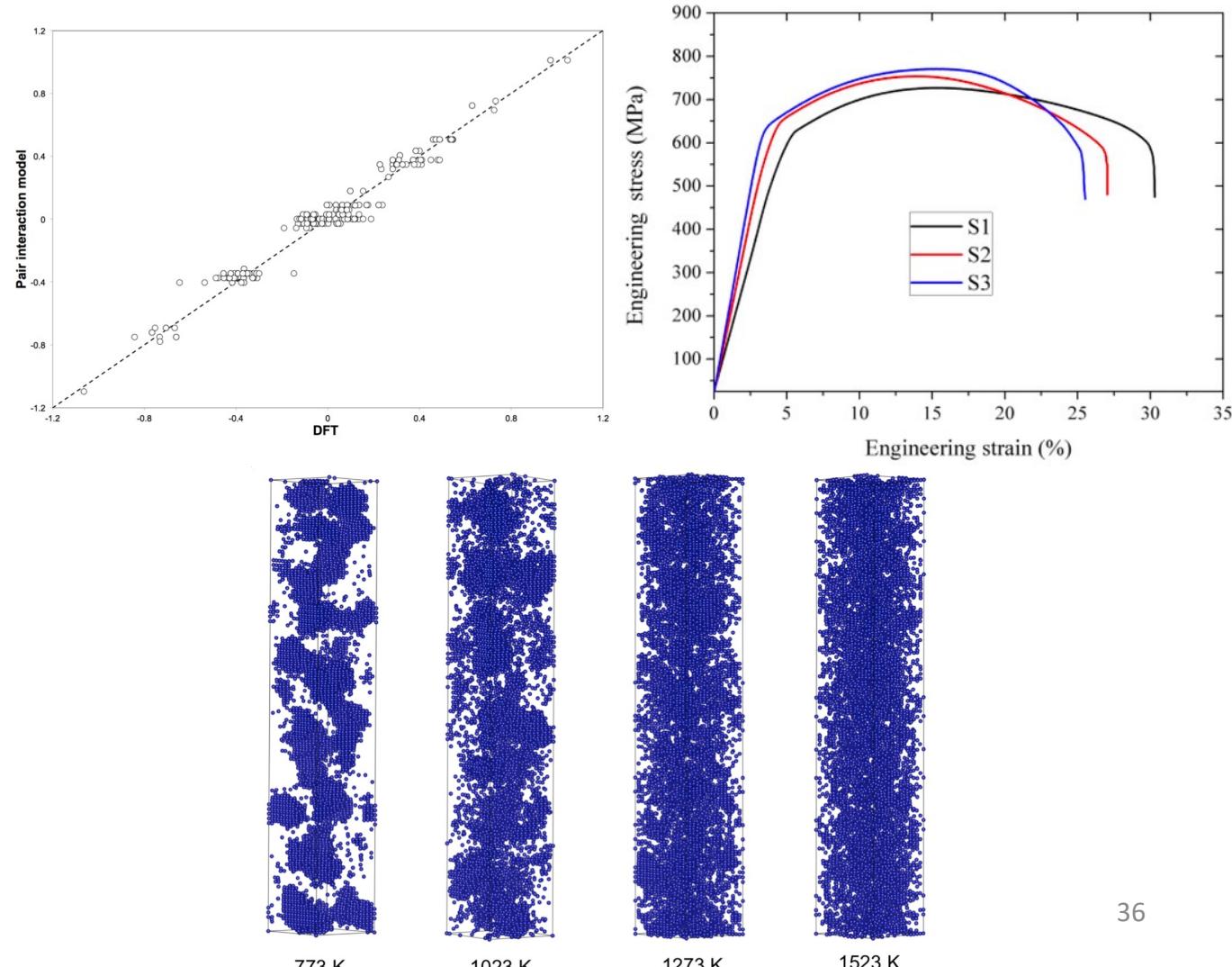


(b)



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.