

Nuclear Fuel Performance

NE-533

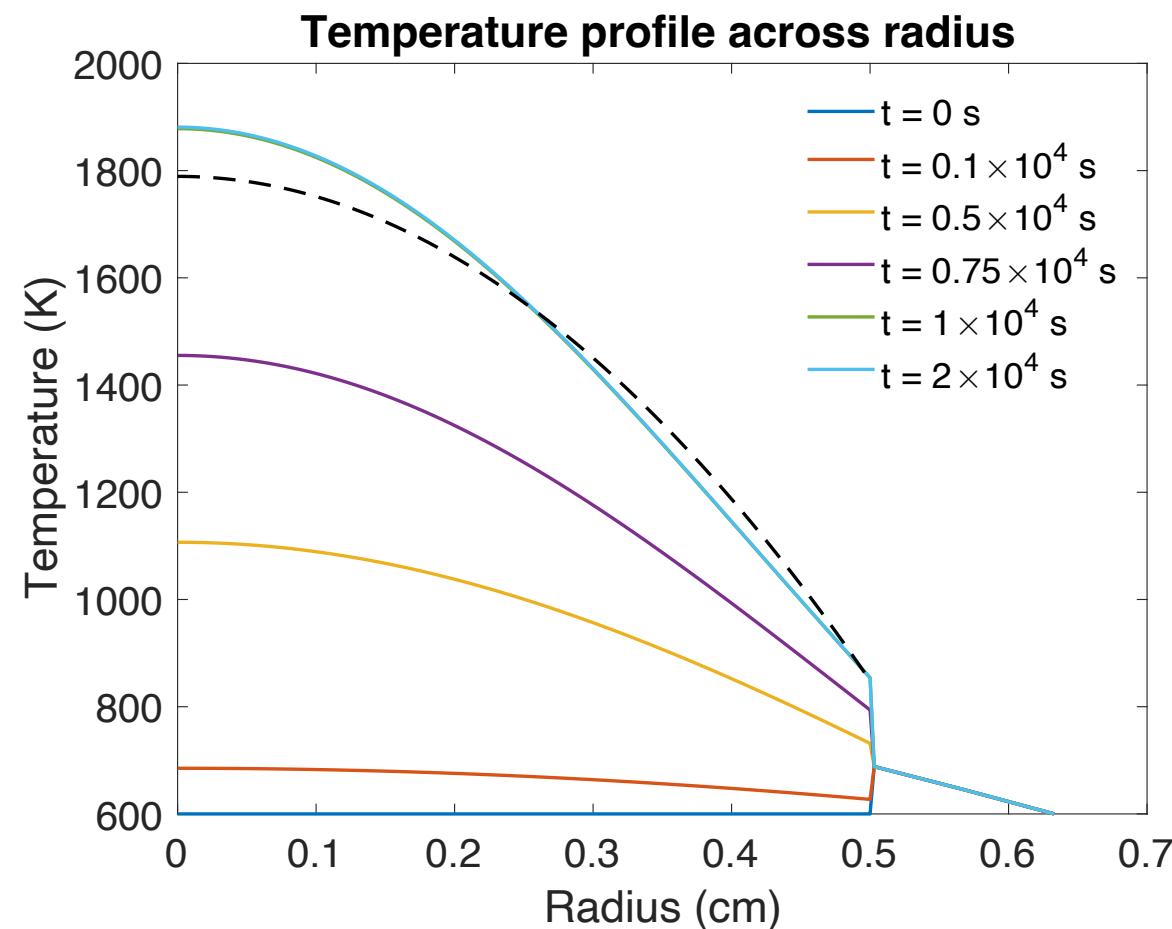
Spring 2022

Last Time

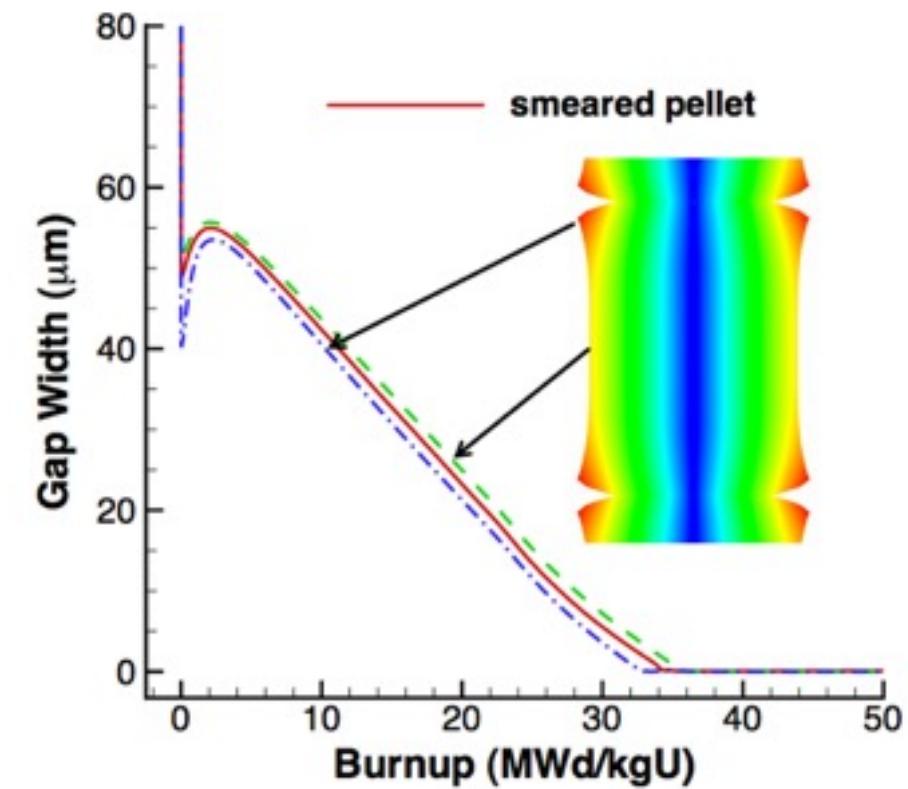
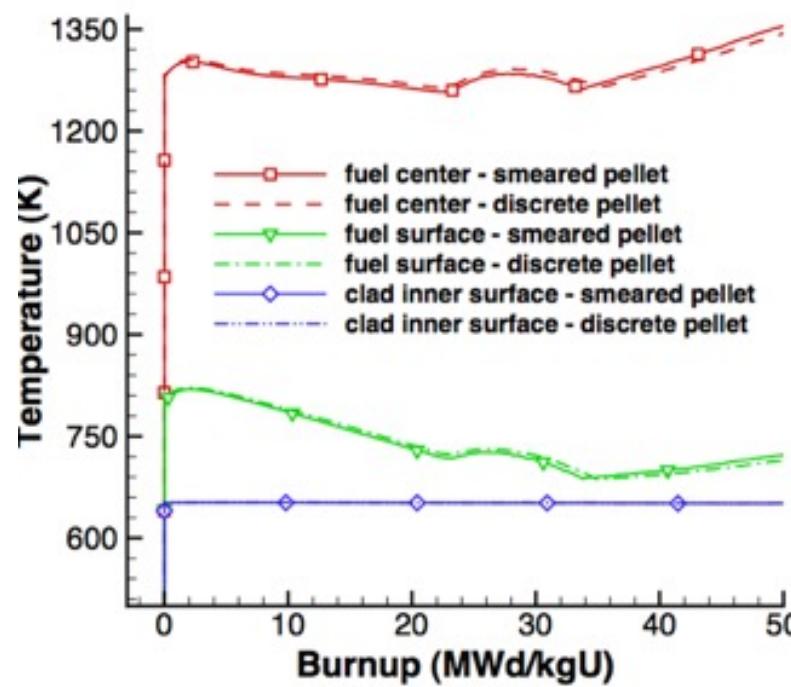
- Wrapped up thermomechanics
- Overviewed US fuel performance codes

MATERIAL PROPERTY EVOLUTION

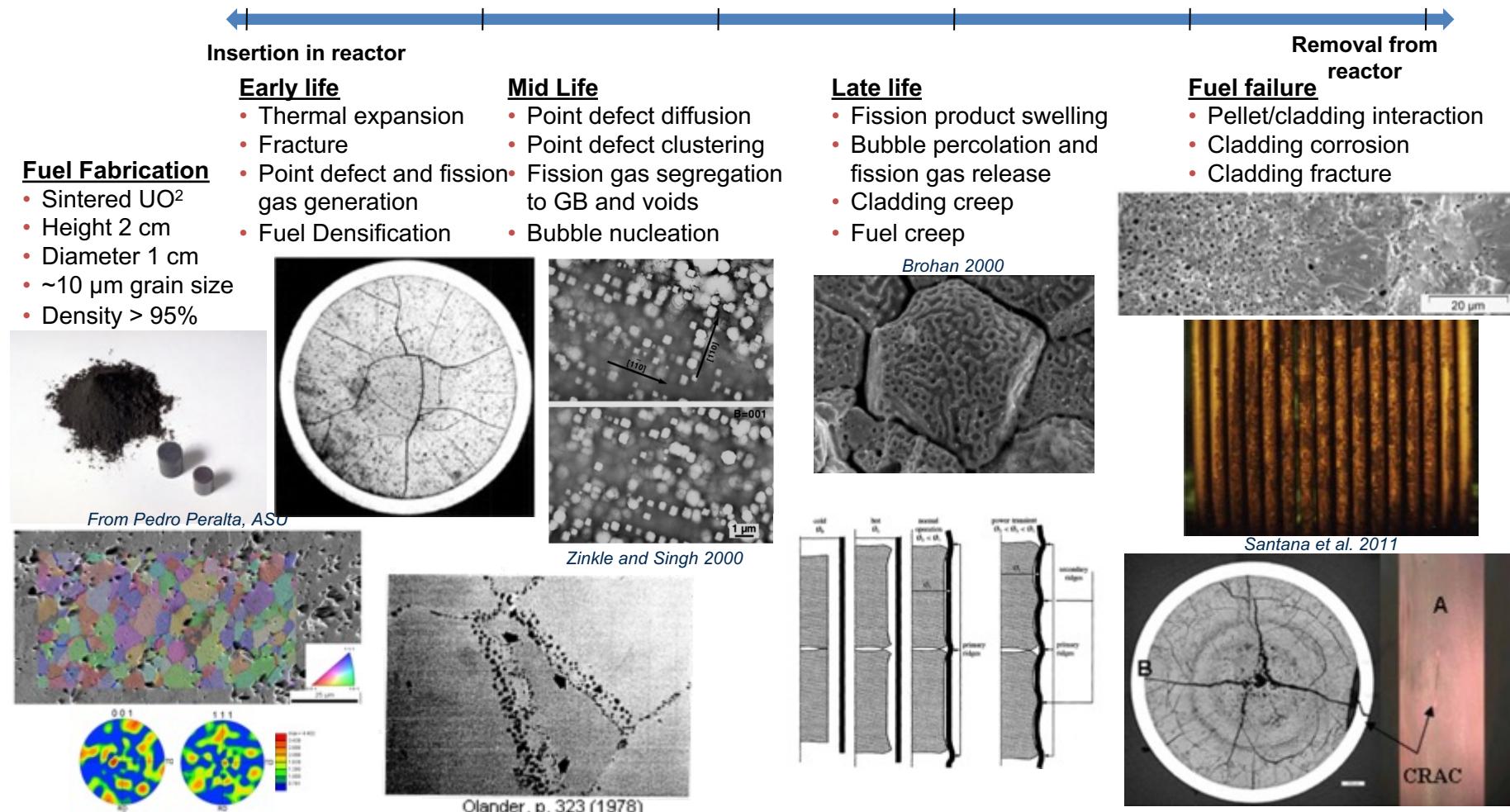
Pretty much everything we have learned so far, nothing changes once the fuel and cladding reach steady state



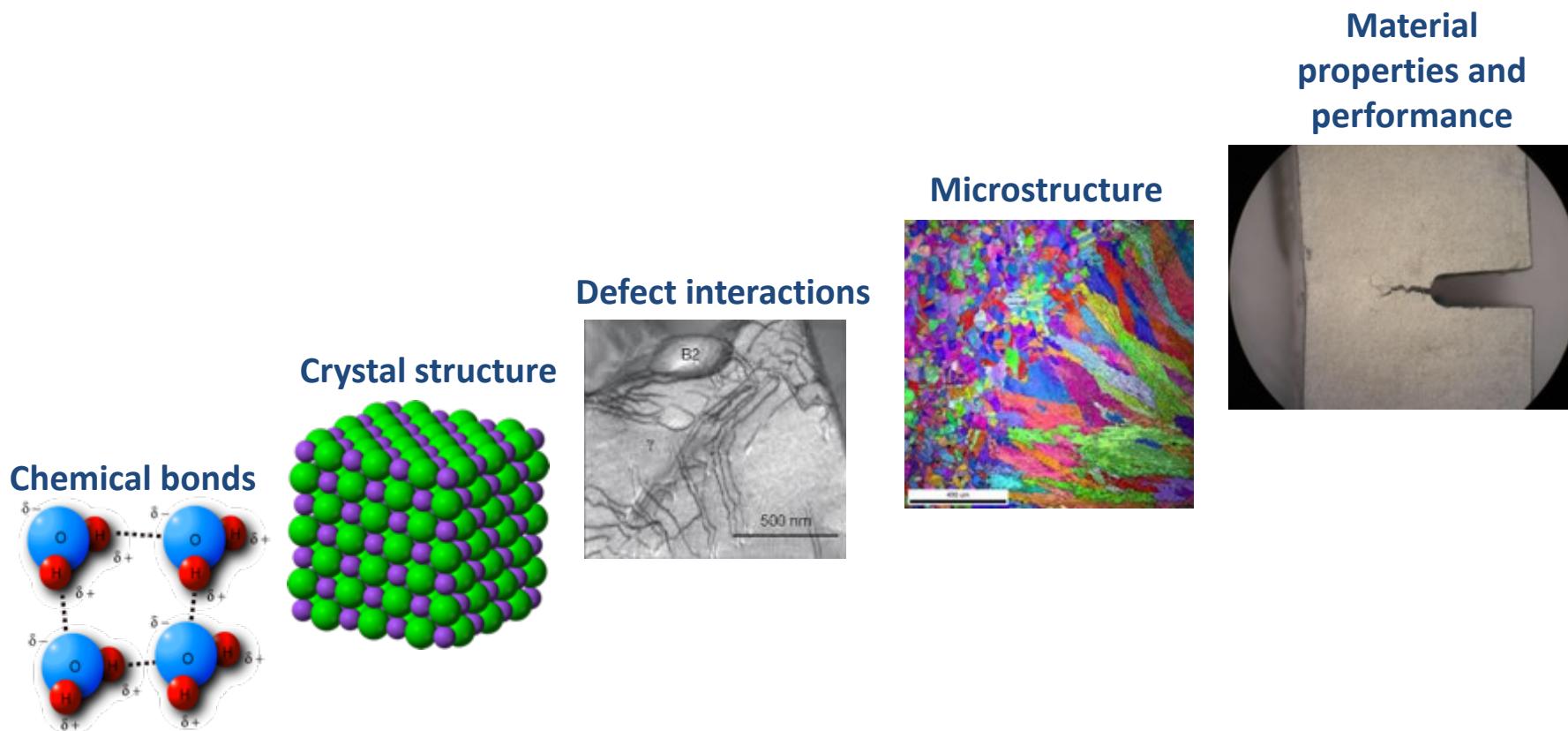
Fuel is dynamic!



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

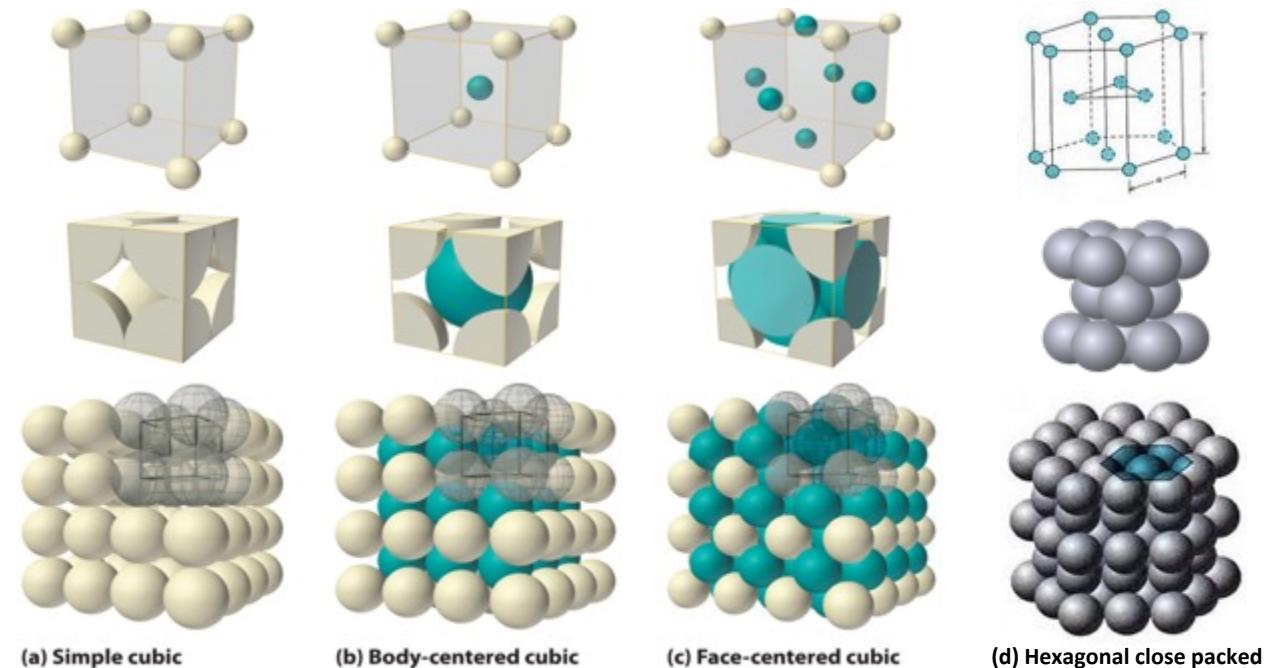


The properties and performance of a material are a result of factors across various length scales



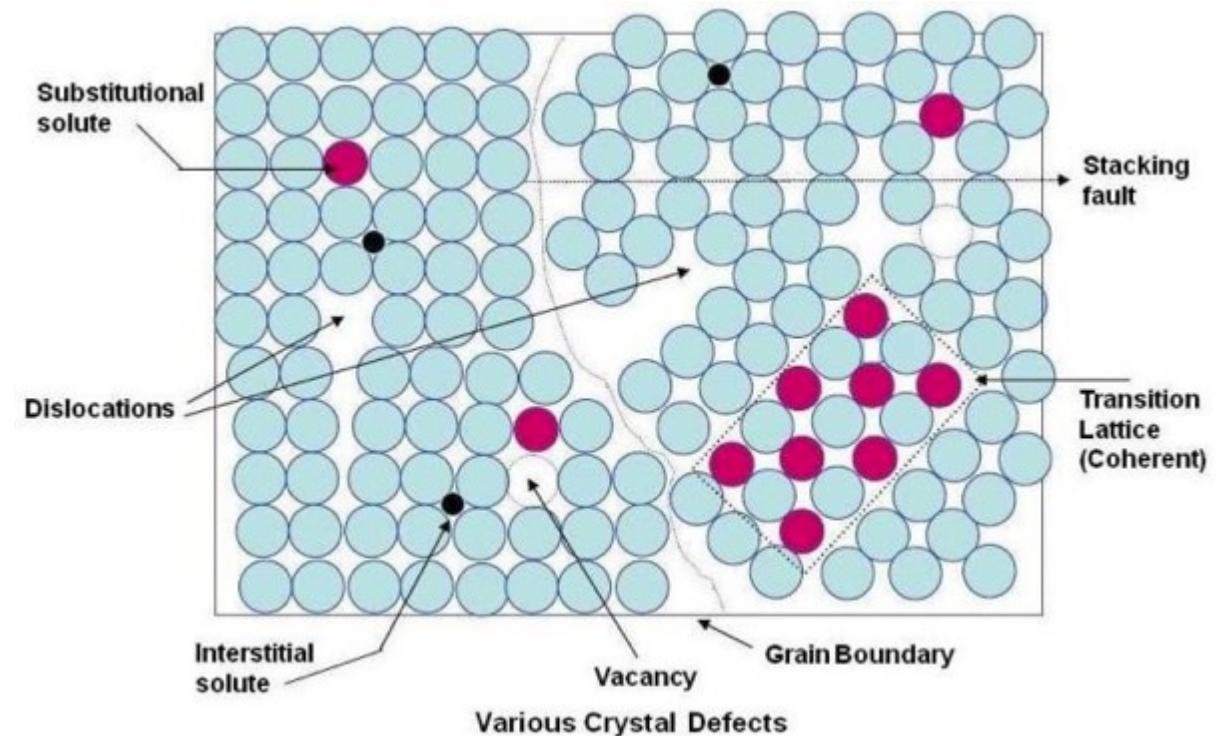
The crystal structure of a material is the shape of the ordered structure of the atoms

- 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



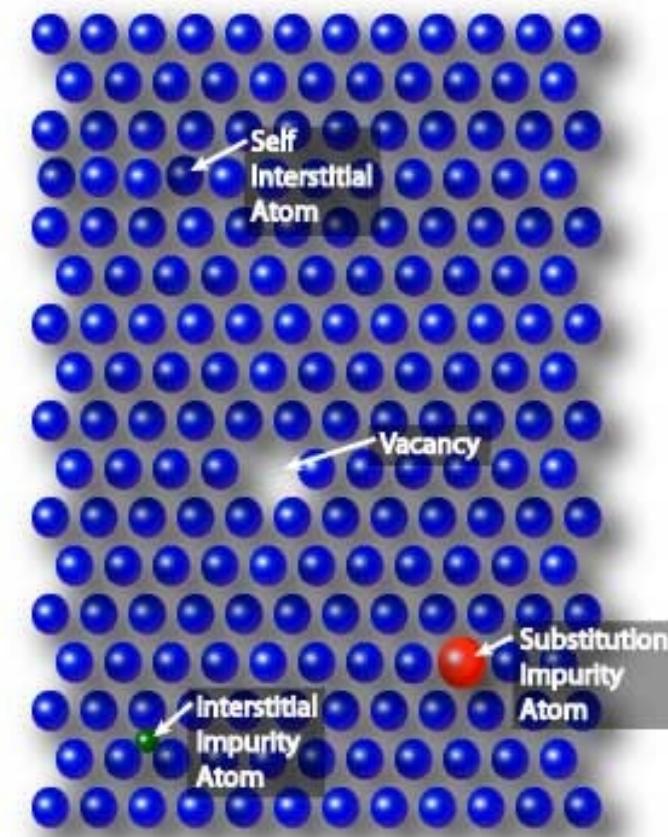
Crystalline materials

- 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



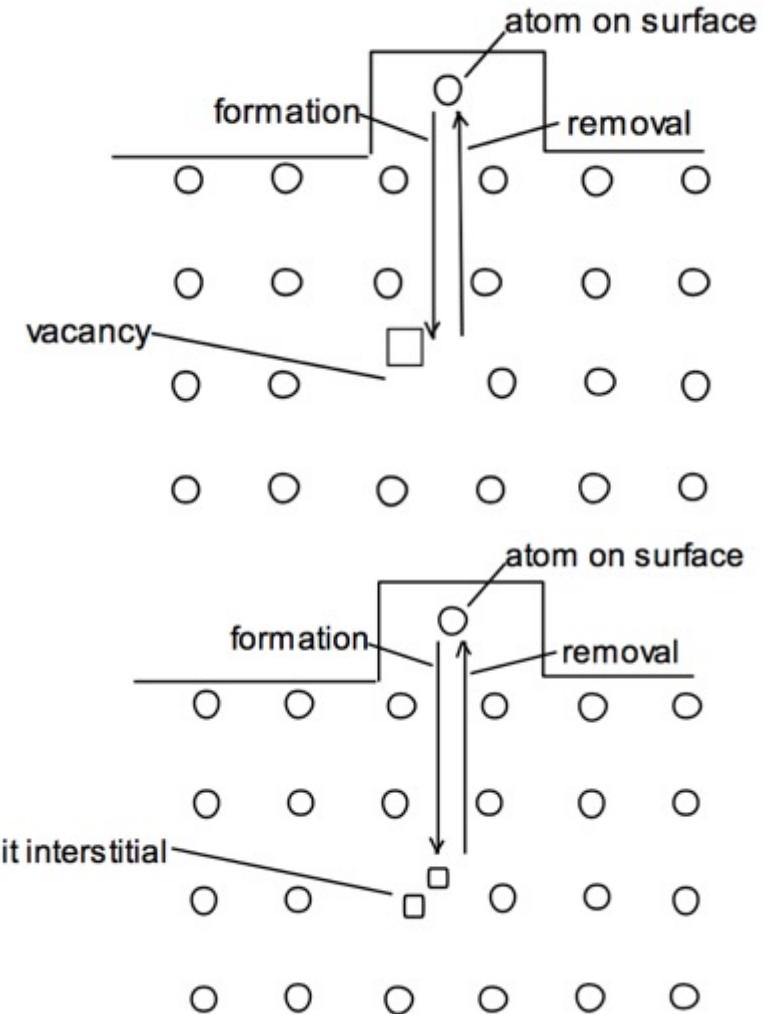
Point defects

- 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

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

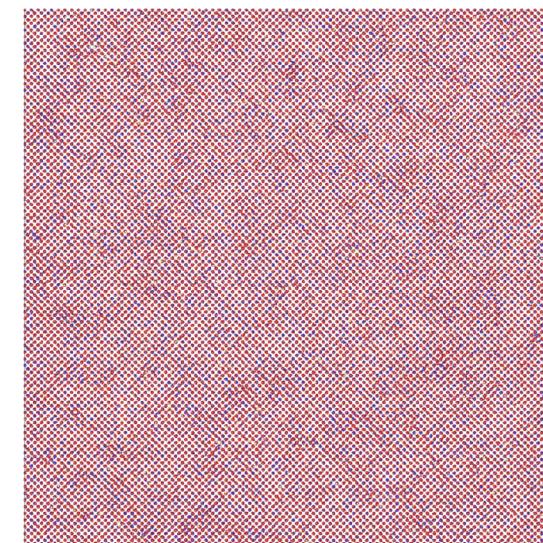
- There exists an equilibrium concentration of point defects

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



Radiation damage

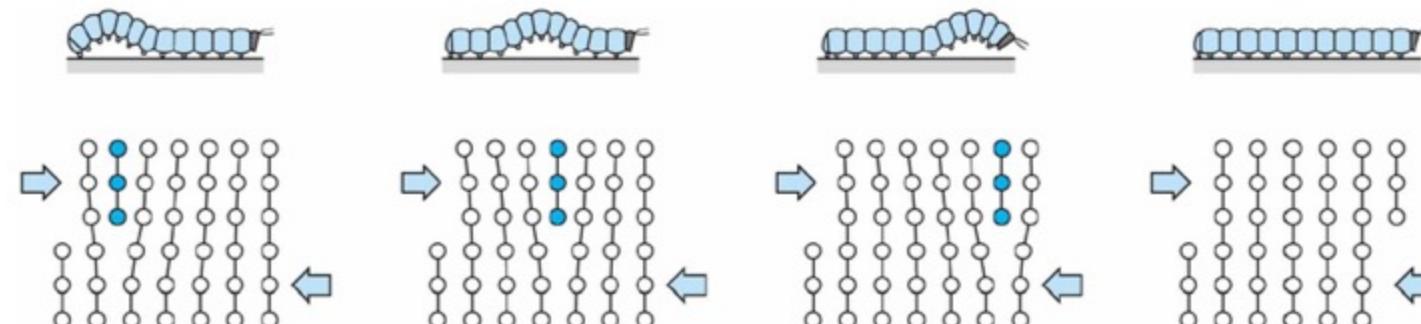
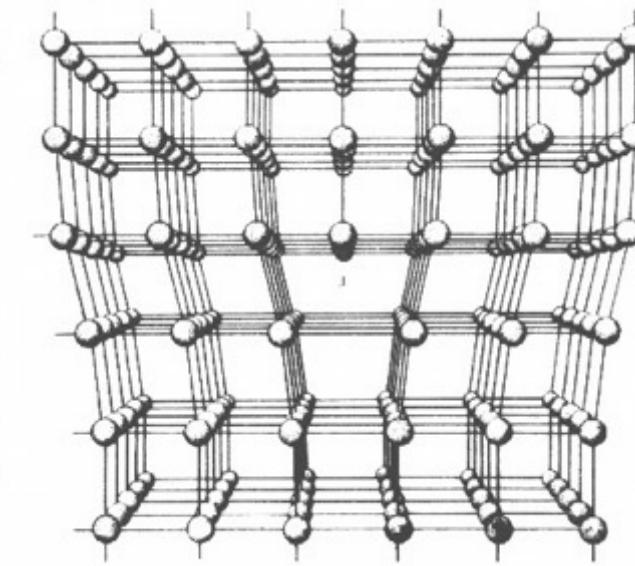
- Ionizing radiation can introduce defects into the crystal lattice, called radiation damage
- PKA collisions create a collision cascade, which results in the generation of defects (point and clusters)



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

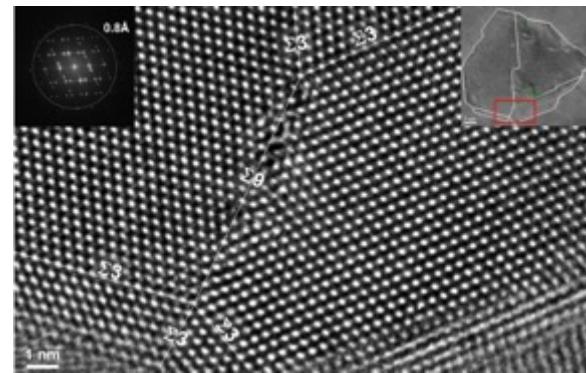
Dislocations are imperfections associated with a line of lattice sites (1D defect)

- 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

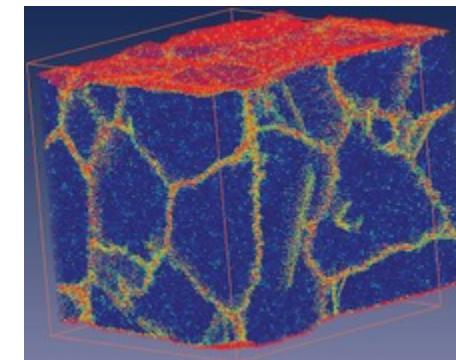


Grain boundaries

- 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

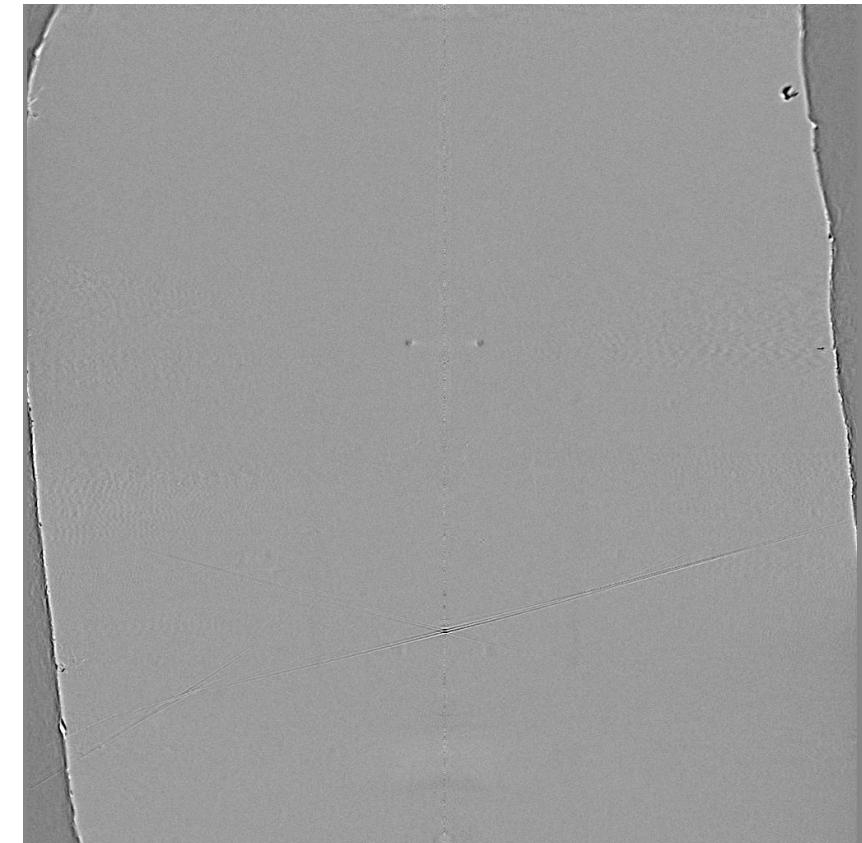


High-res transmission electron microscopy can also show individual atoms (palladium)
www.knmf.kit.edu/T_EM.php



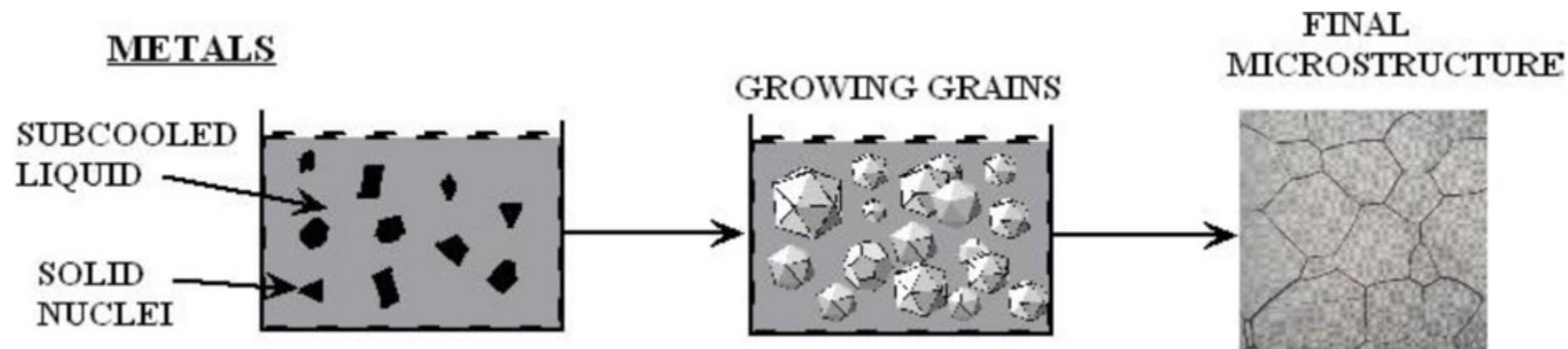
Atomistic simulation of grain boundaries in 3D

Metals are often cast, and polycrystals naturally form during casting



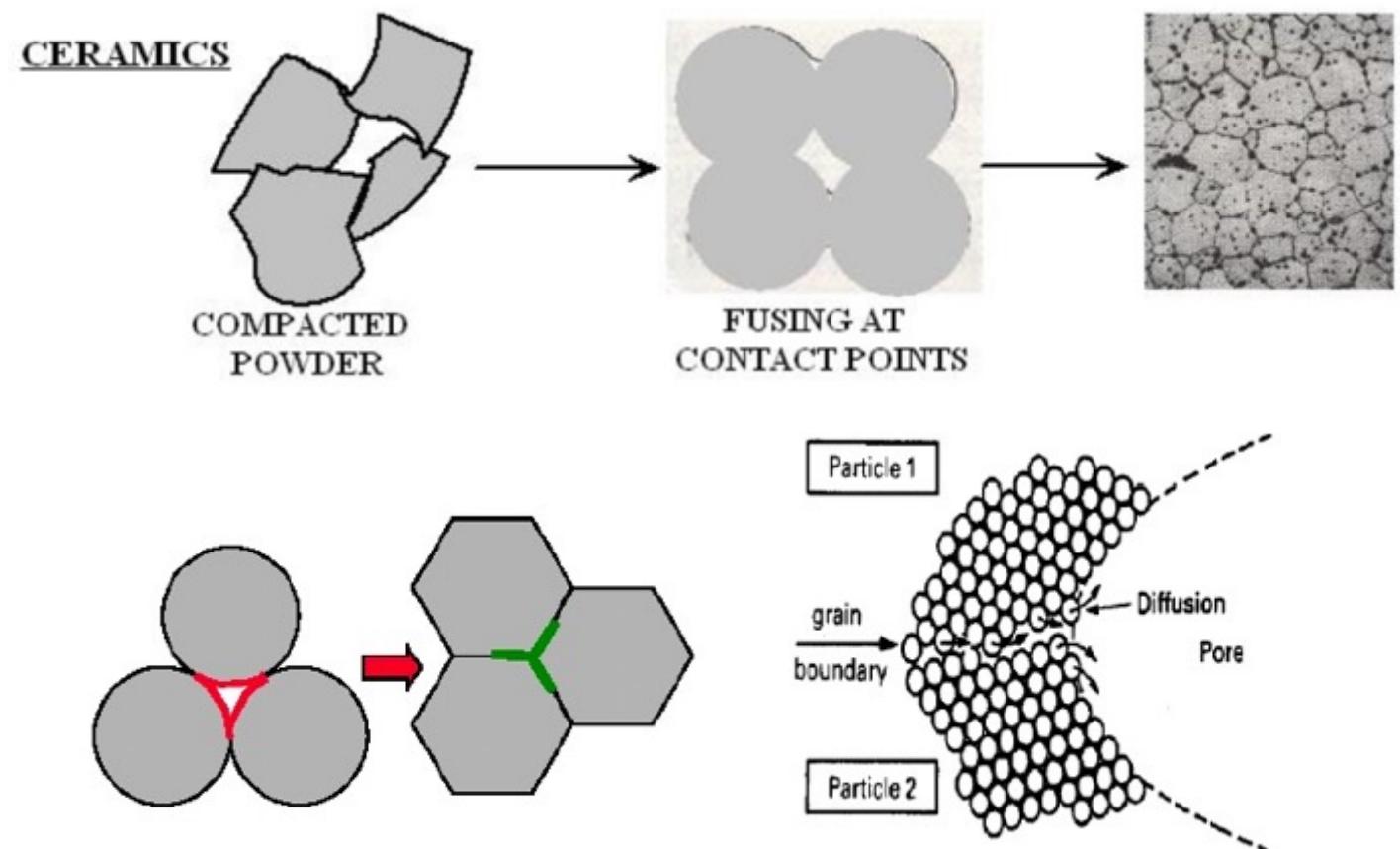
Polycrystals naturally form during casting

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



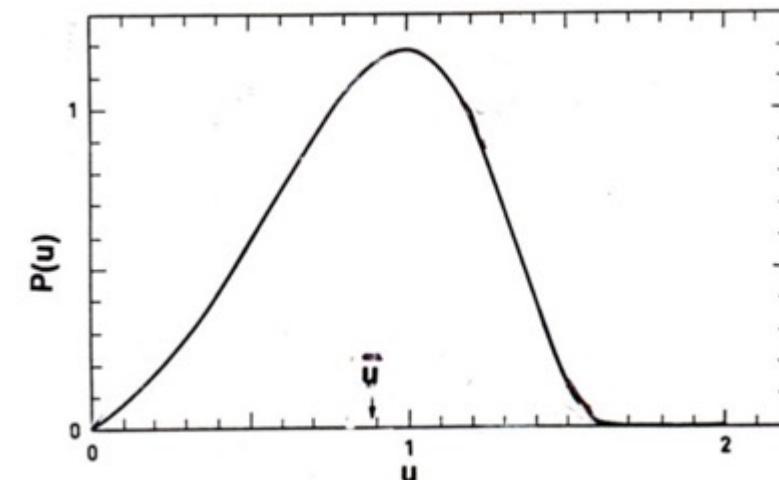
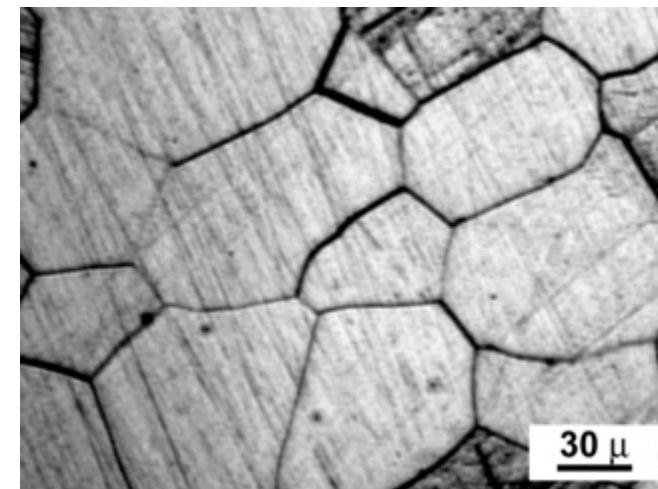
Ceramic sintering

- 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



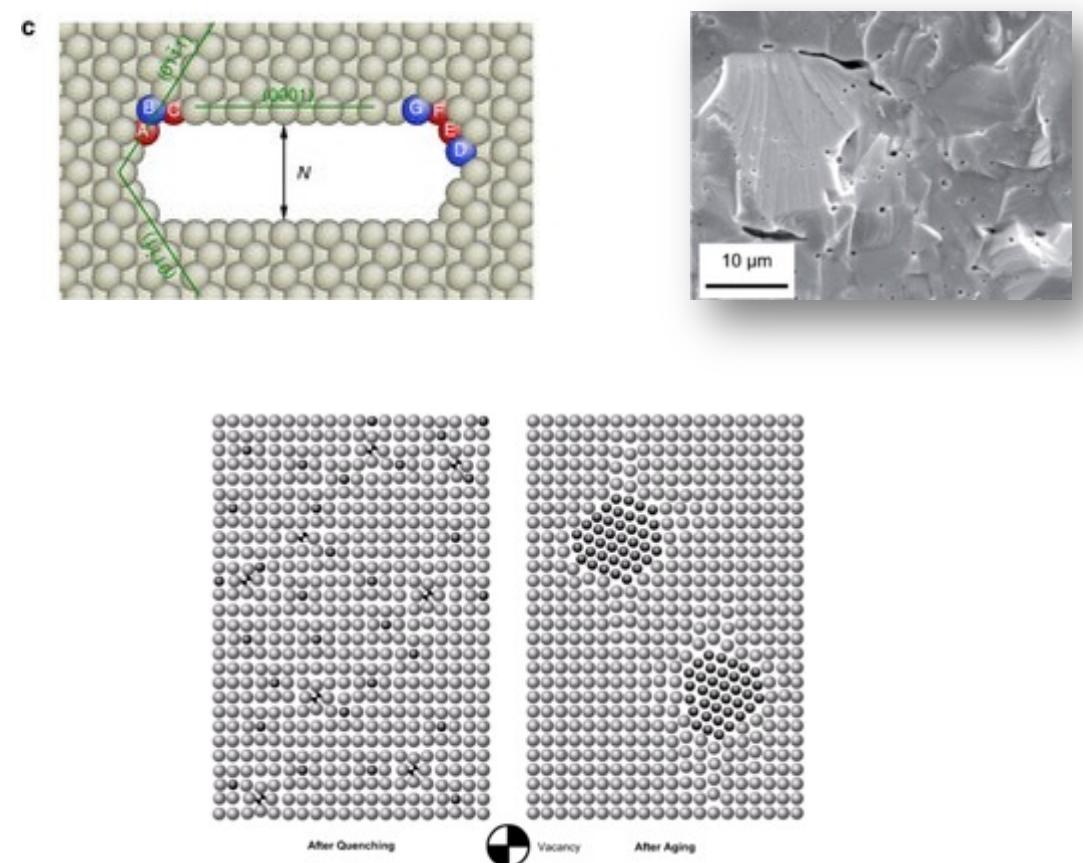
Distribution of grain sizes

- 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



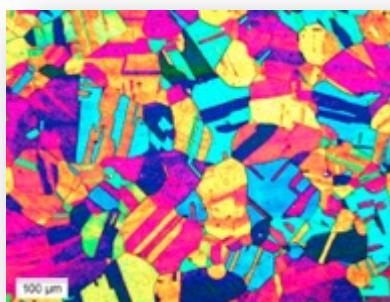
3D Defects

- 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

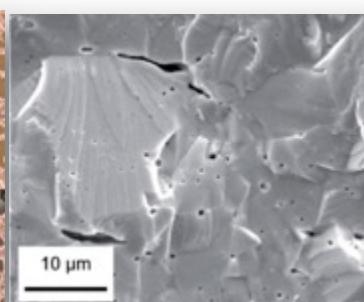
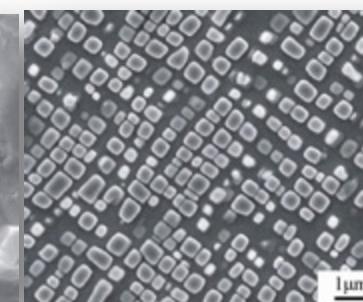
- Material microstructure is the structure observable with 25x magnification
- The microstructure includes grain structure, secondary phases, porosity, and more
- The microstructure can strongly influence physical properties such as strength, toughness, ductility, hardness, corrosion resistance, high/low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



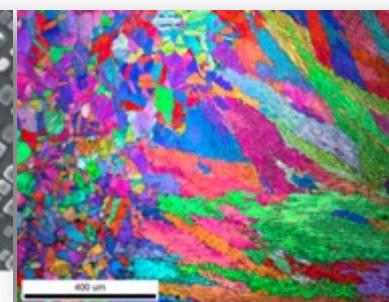
Annealed Stainless Steel



Cast Bronze

Sintered UO₂

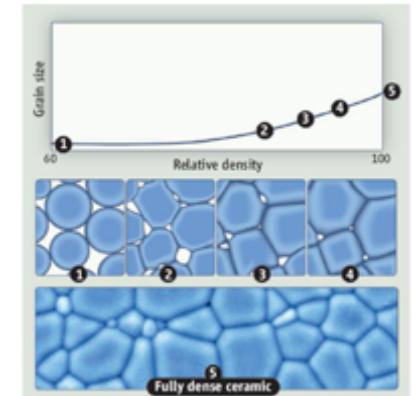
Co-Al-W Superalloy



Friction stir welded stainless steel

Material processing

- The process we use to make a material has 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

UO₂ RADIATION EFFECTS

Thermal Conductivity Degradation

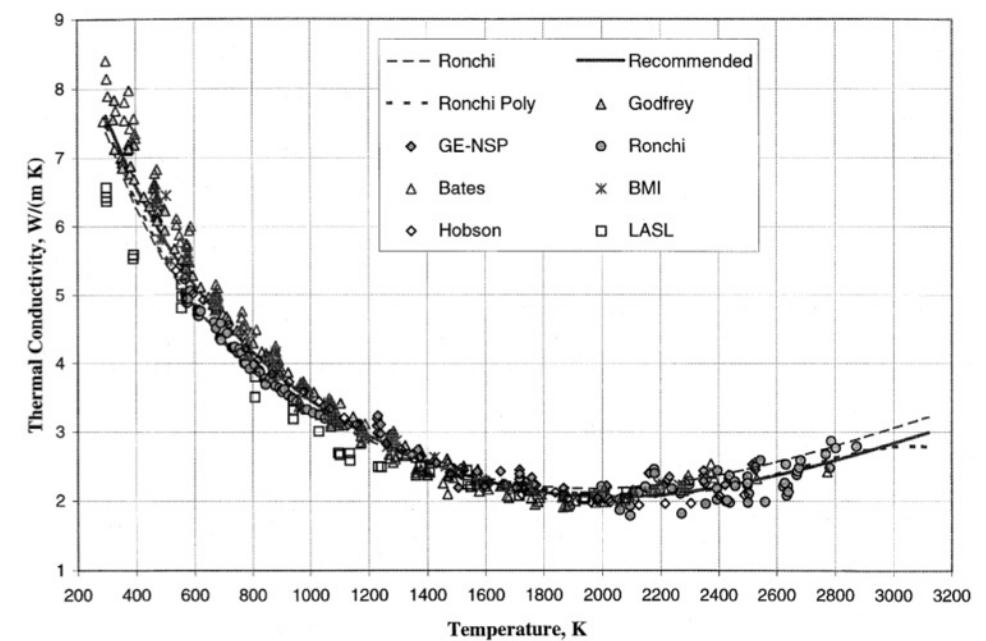
- The physical effects caused by atomic displacements are rather complex and depend on the relative sink strengths of a given material for interstitials and vacancies and on the temperature
- The defects that survive migrate through the crystal lattice where they cluster to form extended defects like dislocation loops, lines and networks, which can potentially be absorbed in grain boundaries, gas bubbles or precipitates, which act as sinks
- Damage by fission also leads to changes in chemistry due to production of fission products
- 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

UO₂ Thermal Conductivity

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

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

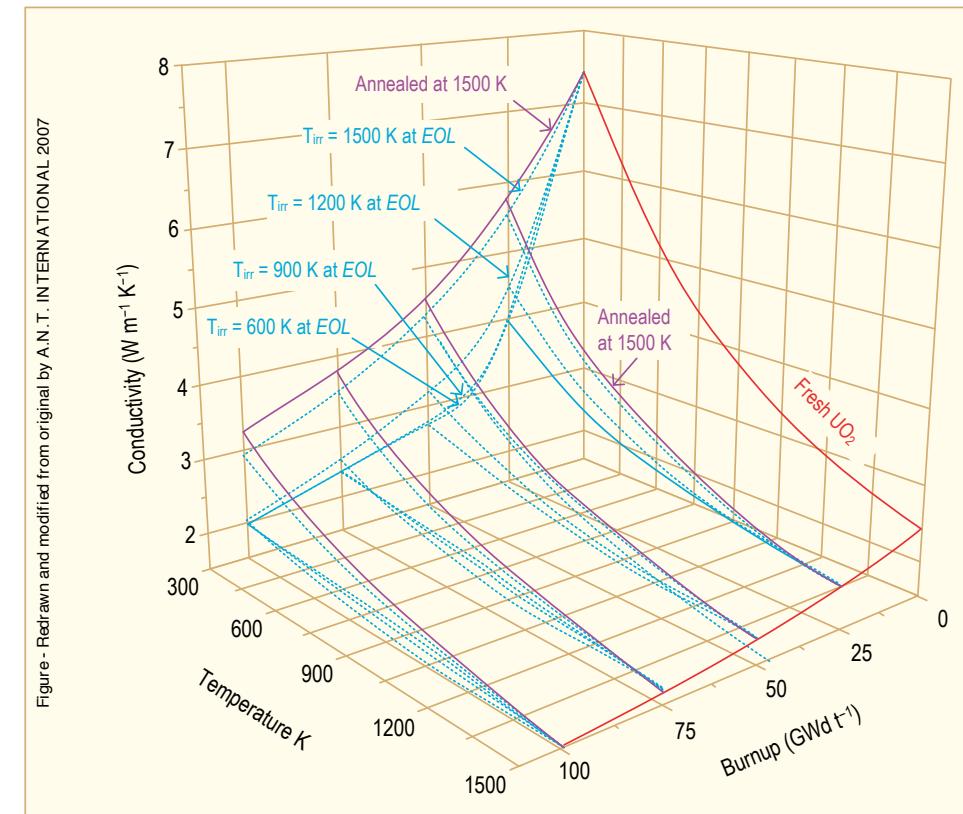
- Where t = T/1000
- The first part of the equation describes the phonon interactions
- The second part describes electronic transport which becomes significant at high temperature



UO₂ Thermal Conductivity

- UO₂ 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

- The primary 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)

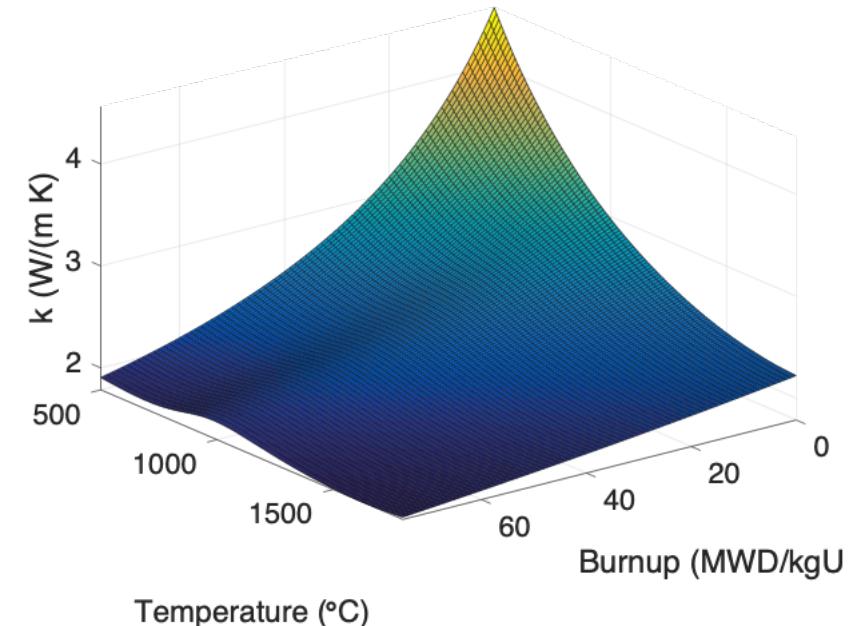
$$k = (1 - R_f(T))k_{ph1}(T, \beta) + R_f(T)k_{ph2}(T, \beta) + k_{el}(T)$$

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

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

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

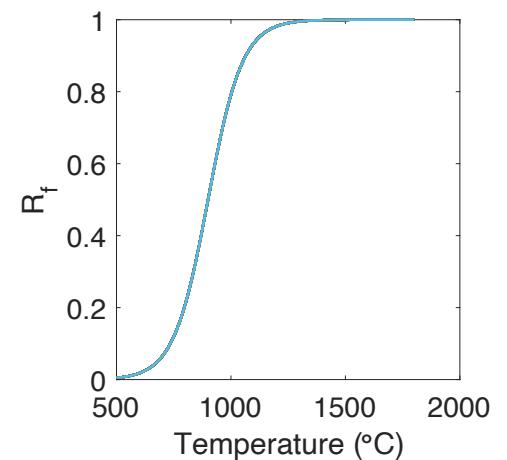
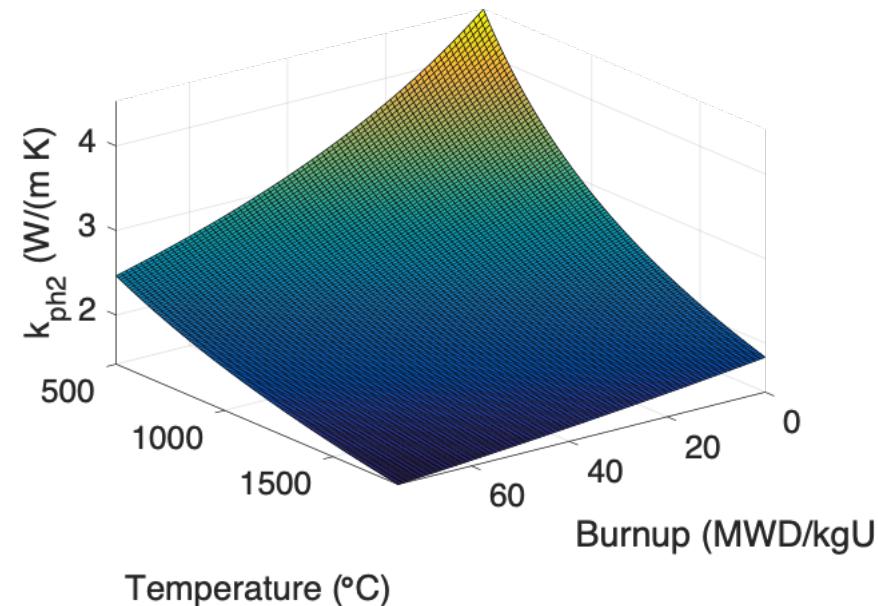
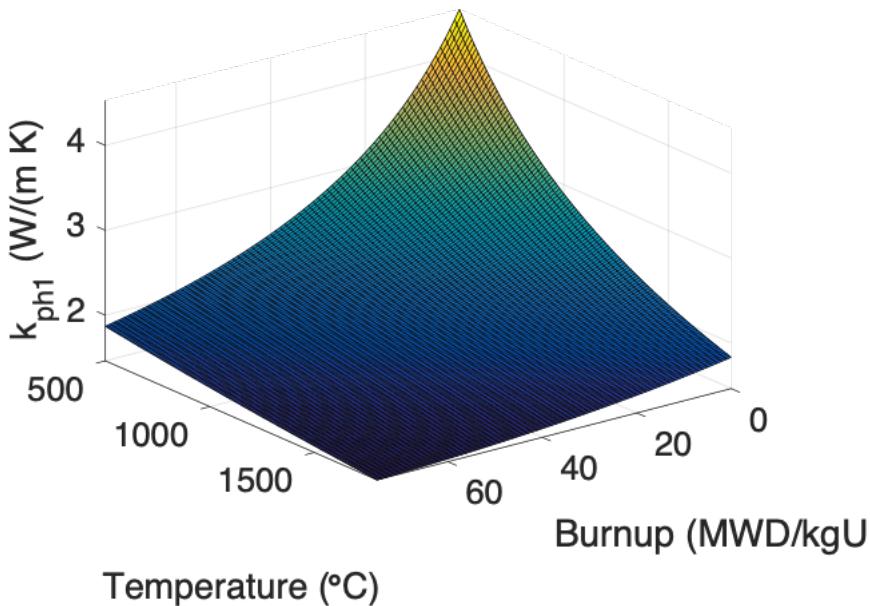
$$k_{el} = 1.32 \times 10^{-2} e^{1.88 \times 10^{-3}T}$$



UO₂ Thermal Conductivity

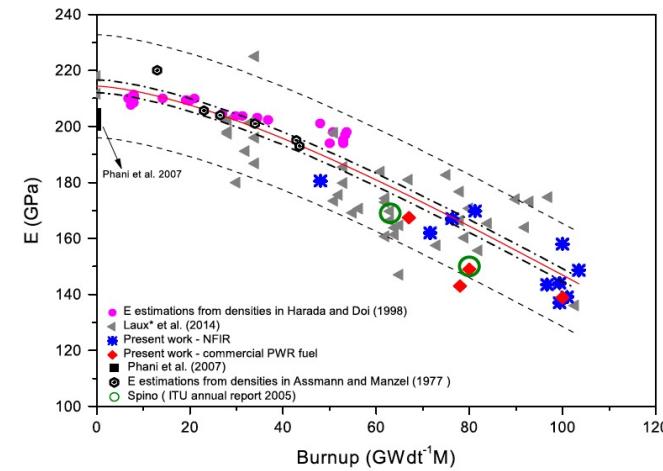
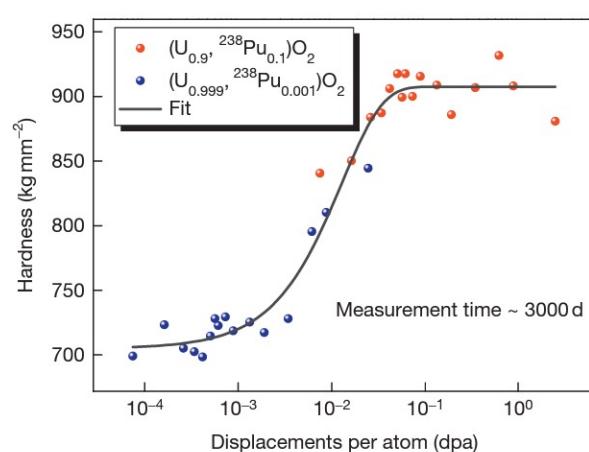
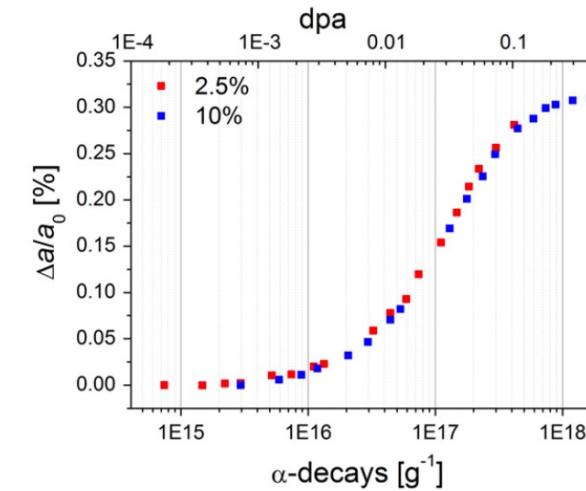
- The R_f function switches between two k_{ph} functions

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



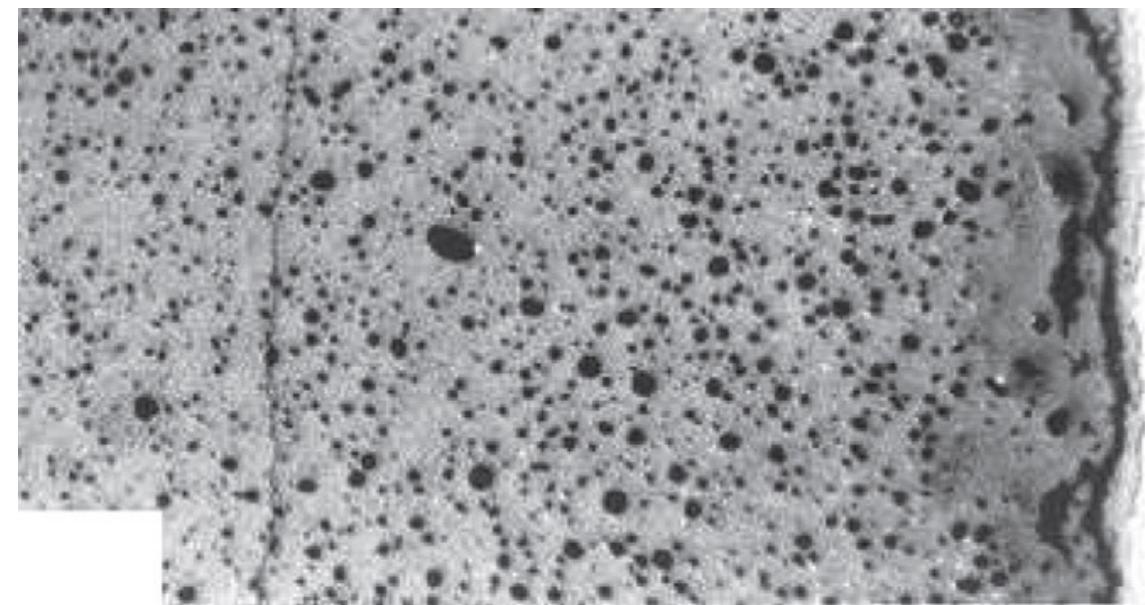
Physical Changes

- 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, increasing with dpa
- UO_2 does not become amorphous, but damage accumulation can eventually cause polygonization: transformed into a material consisting of very small grains



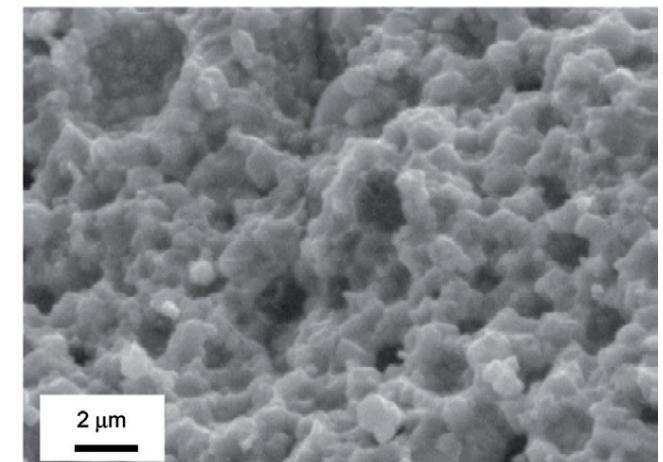
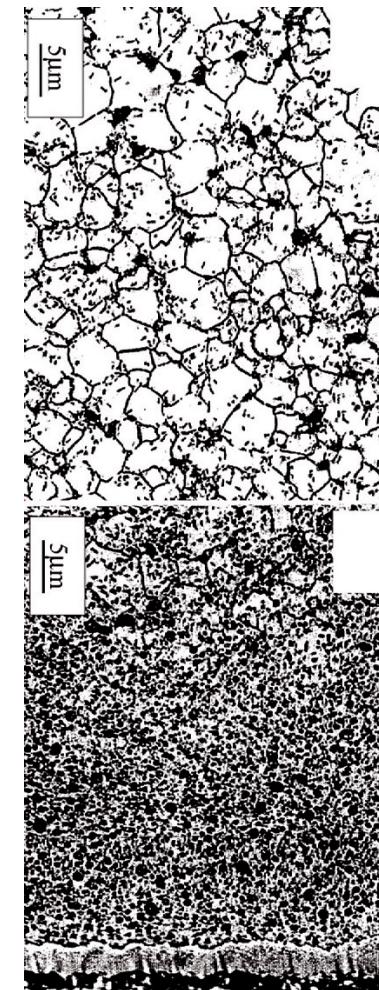
High Burnup Structure (HBS)

- 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
- Strong capture occurs in the periphery of the fuel pellet, leading to Pu production and an increase in fissile density
- In UO₂, grains subdivide from 10mm 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 understood



Summary

- Radiation damage leads to accumulation of defects in UO₂, changing properties and microstructure
- Thermal conductivity degrades with time
- High Burnup Structure can form along the rim

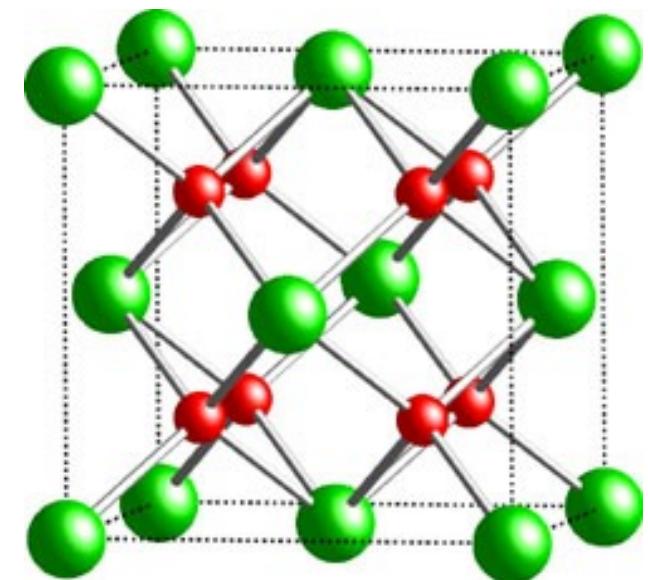
Next Time

- MOOSE overview
- Good to bring a laptop, or to work remotely, so we can move towards getting an active MOOSE build you have access to

FUEL CHEMISTRY

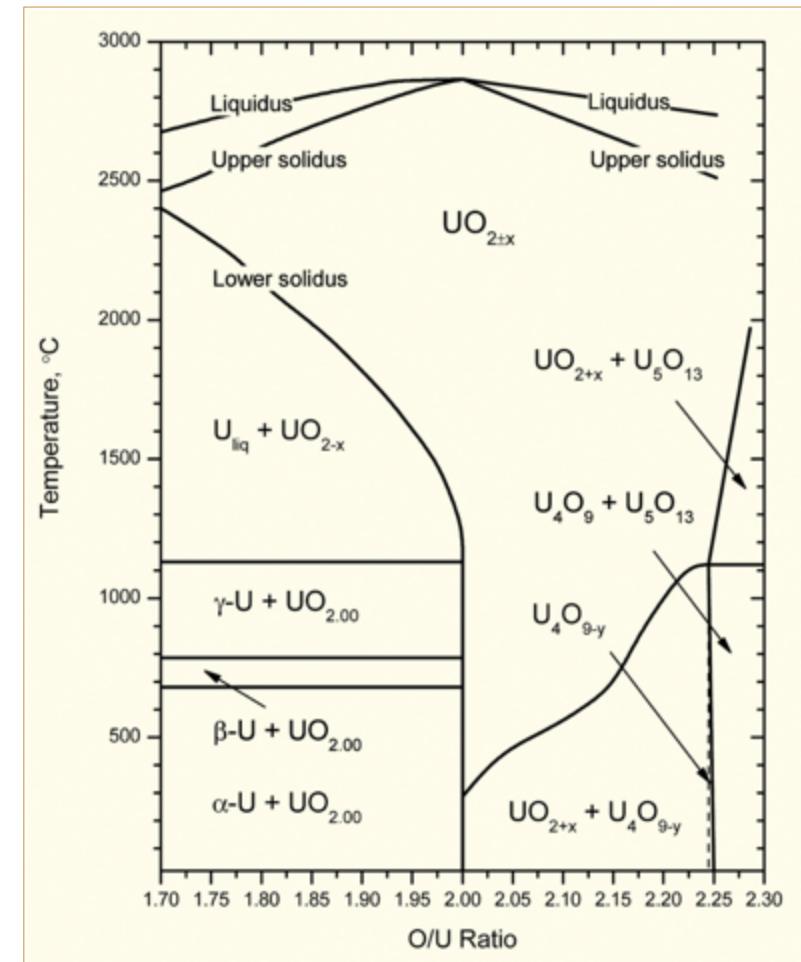
Fuel Chemistry

- UO_2 is an ionic compound that must have balanced charges
- What is the charge of a typical oxygen ion?
 - O^{2-}
- Uranium valence states
 - Possible: U^{3+} , U^{4+} , U^{5+} , U^{6+}
 - Most stable: U^{4+} , U^{5+} , U^{6+}
 - Beyond UO_2 , can have U_4O_9 , U_3O_8 , UO_3
- The structure is very stable all the way up to the melting temperature and down to extremely low temperatures, even with irradiation damage
- There is space in the uranium lattice that can accommodate fission products



O/M ratio

- The ratio of oxygen to uranium metal (O/M ratio) can vary
- This is the stoichiometry
- The homogeneity range of uranium dioxide extends to both hypo- and hyperstoichiometric compositions in oxygen
- The minimum and maximum oxygen contents in the dioxide correspond to the compounds with the formula of respectively $\text{UO}_{1.67}$ at 2720 K and $\text{UO}_{2.25}$ at approximately 2030 K
- Will the O/M ratio go up or down during reactor operation?
 - It is complicated, because of the formation of fission products that also react with the oxygen, and O interaction with Zr cladding



Excess O

- The crystal structure of UO_2 can accommodate extra oxygen
- Excess oxygen resides at interstitial locations
- Oxygen in neighboring sites is displaced
- Cation valence increases to maintain electrical neutrality
- Fuel fabricated to be nearly stoichiometric; i.e., $\text{UO}_{2.00 \pm}$ because:
 - It is the most stable
 - It has the highest melting temperature

