

# Nuclear Fuel Performance

NE-591-010

Spring 2021

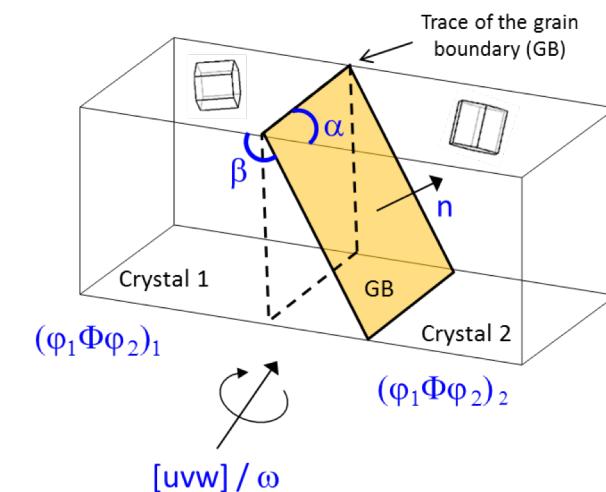
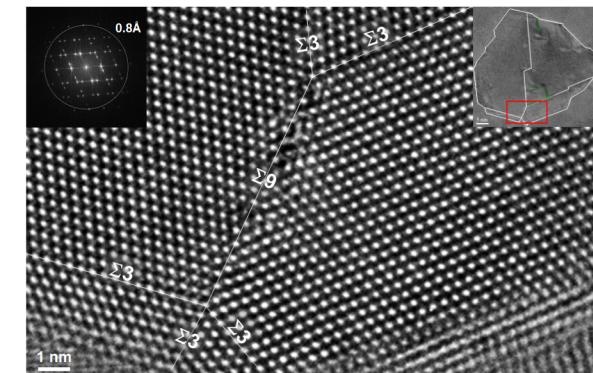
## Last Time

- Even during steady state operation, fuel and pellet conditions change with time due to microstructure evolution
- The crystal lattice is never perfect; it has defects
- However, the ratio of oxygen to uranium can change. We call this the stoichiometry and abbreviate it as O/M ratio
- The O/M ratio impacts many properties of the fuel

# GRAIN GROWTH

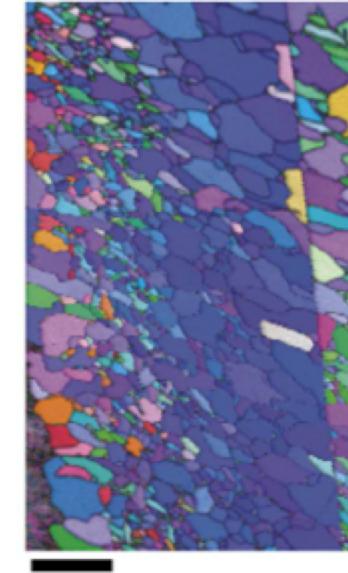
# Grain Boundaries

- Materials are typically composed of various regions where the crystal lattice is oriented differently
- When two grains meet, there is a plane of atoms that do not follow the crystal lattice called a **grain boundary**
- Grain boundaries add energy to the material that is a function of their structure
- A grain boundary's energy is determined by its:
  - **Inclination** – the orientation of the 2D grain boundary plane (2 degrees of freedom)
  - **misorientation** – the rotation required to align one grain with the other (3 degrees of freedom)

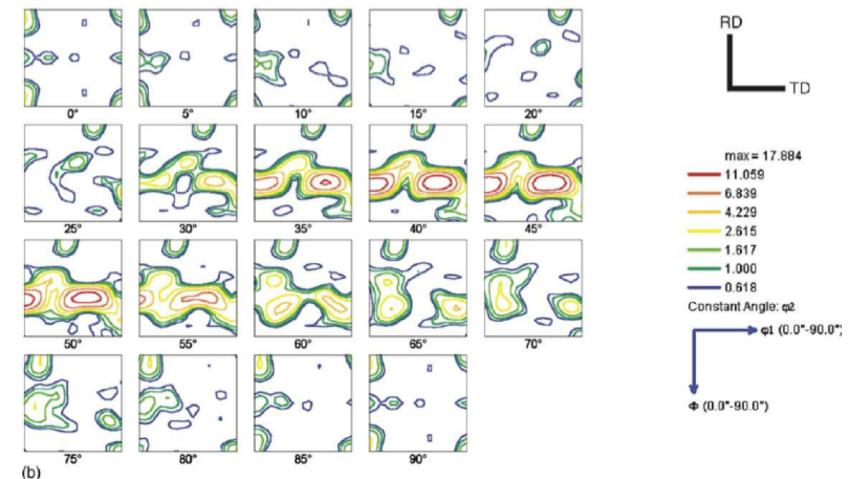


# Grain Texture

- The orientation of all the grains in a material is called the texture
- The degree of texture is dependent on the percentage of crystals having a preferred orientation
  - randomly oriented grains equals no texture
- Texture is seen in almost all engineered materials, and can have a great influence on materials properties



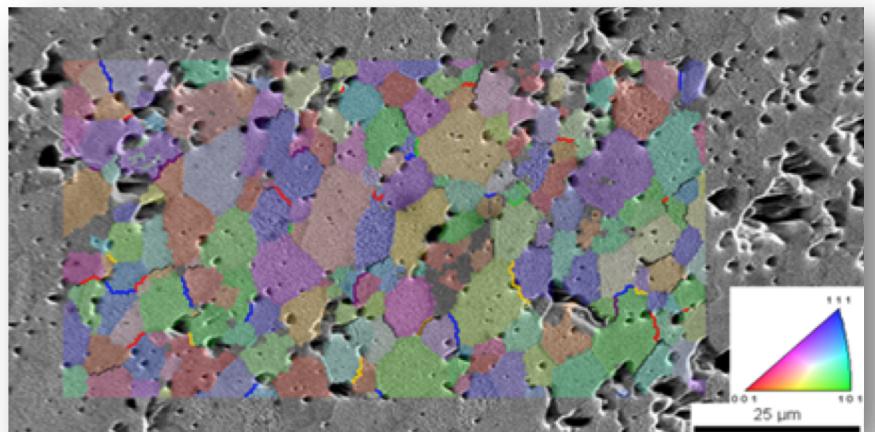
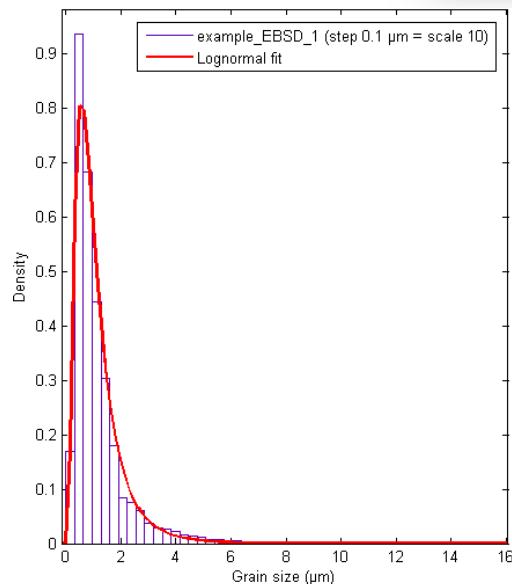
Electron back scatter diffraction map  
for cubic crystal structure



Orientation distribution function

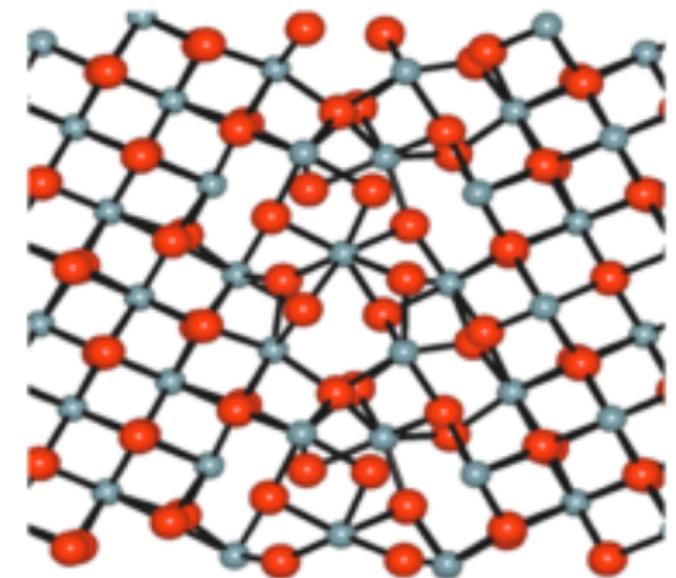
# Grain size

- In a polycrystal, there is also a distribution of grain sizes
- The average grain size of the fuel has a significant impact on its behavior
- Typical LWR fuel has an initial average grain size of about 10 microns
- The average grain size impacts
  - Fission gas release
  - Swelling
  - Thermal conductivity
  - Creep



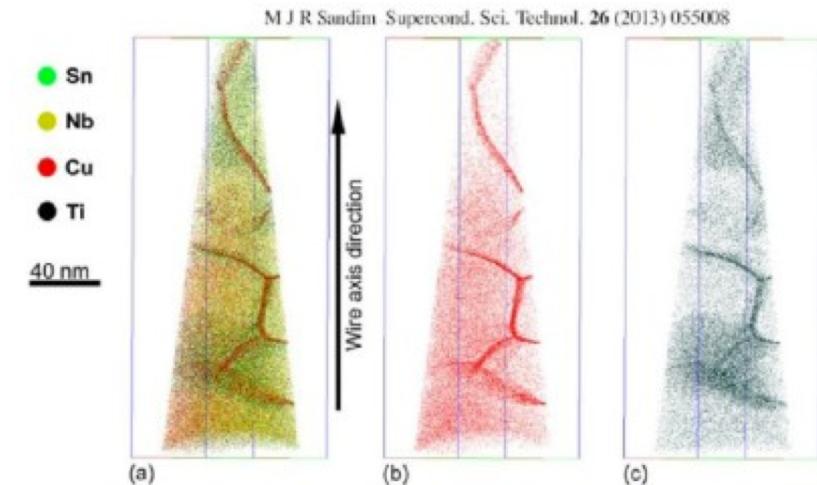
# Grain boundary diffusion

- Diffusion often occurs faster along grain boundaries than in the perfect crystal
- Grain boundaries have more space than the perfect lattice
- So, atoms diffuse faster along grain boundaries than through the perfect lattice
- This means that grain boundary, or intergranular, diffusion increasingly dominates bulk (intragranular) diffusion as the temperature is reduced
- Grain boundary vacancy diffusion has a large impact on creep

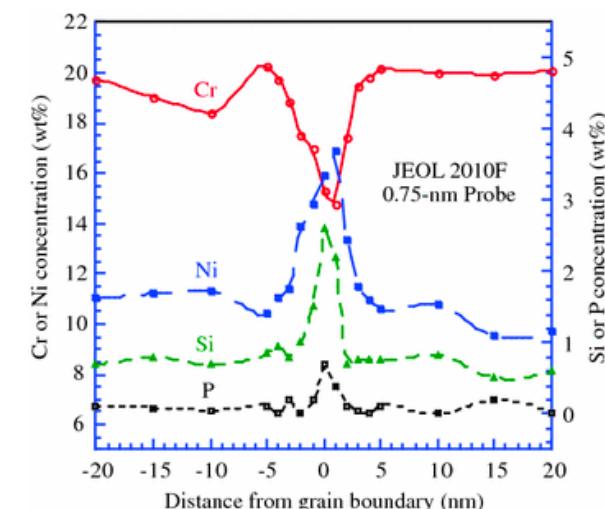


# Grain boundary segregation

- Impurity atoms (such as fission gas) and other defects move to grain boundaries
- Due to grain boundaries having more open space, impurity atoms have a lower energy when they are on grain boundaries.
- This is called **grain boundary segregation**
- Radiation induced segregation (RIS) is where radiation produces defects that drive towards the grain boundary and preferentially drag solute atoms

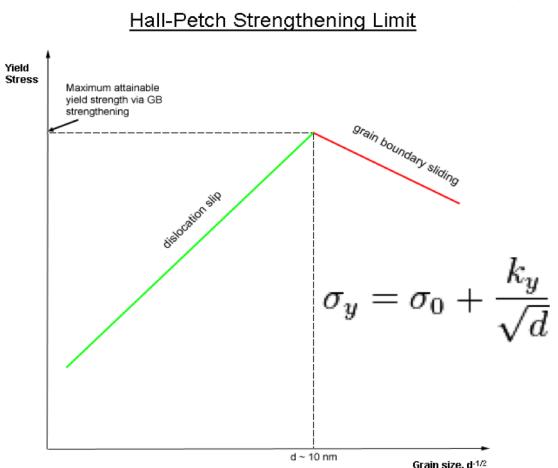
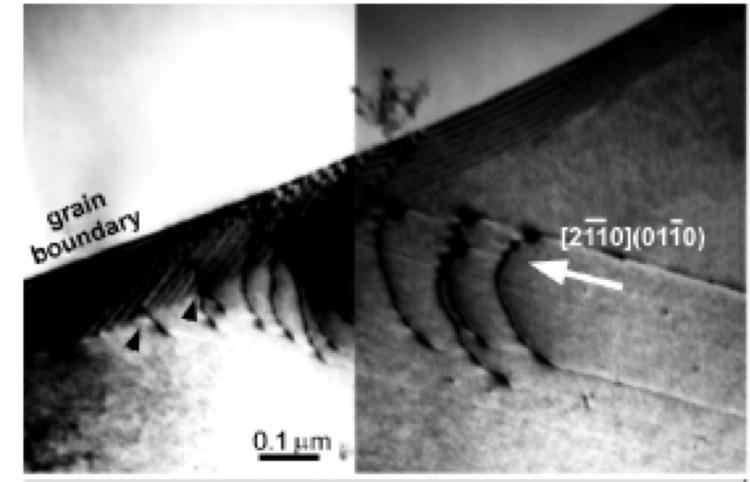


APT reconstruction : (a) elements mapped in the sample and (b) and (c) show only Cu and Ti



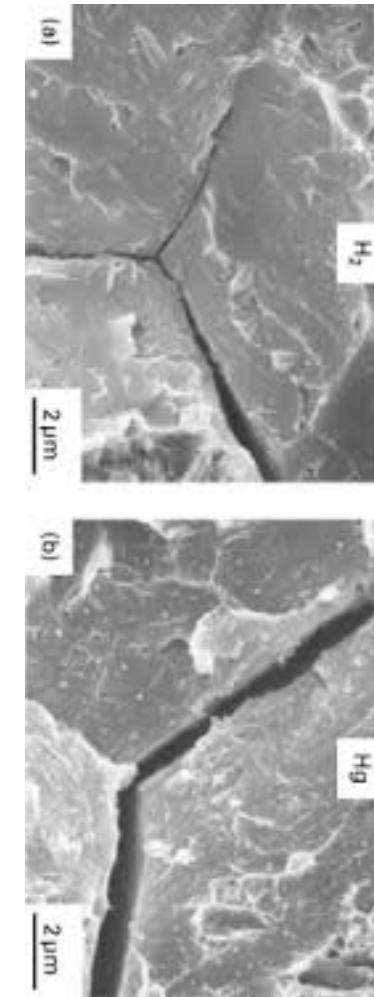
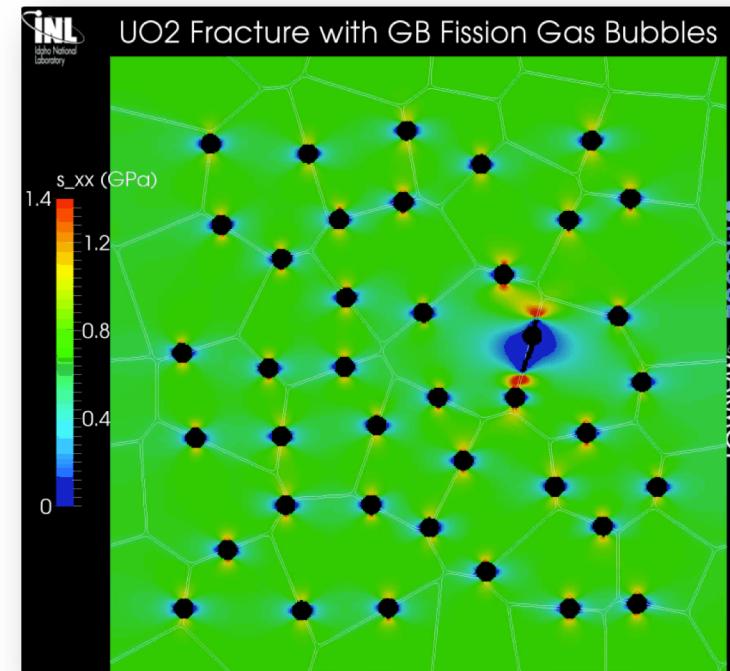
# Dislocation interaction with grain boundaries

- Grain boundaries impede dislocation motion
- The number of dislocations within a grain impacts how easily dislocations can traverse grain boundaries and travel from grain to grain.
- So, by changing grain size one can influence dislocation movement and yield strength
- This is called the Hall-Petch effect
  - where  $\sigma_y$  is the yield stress,  $\sigma_0$  is a materials constant for the starting stress for dislocation movement (or the resistance of the lattice to dislocation motion),  $k_y$  is the strengthening coefficient (a constant specific to each material), and  $d$  is the average grain diameter



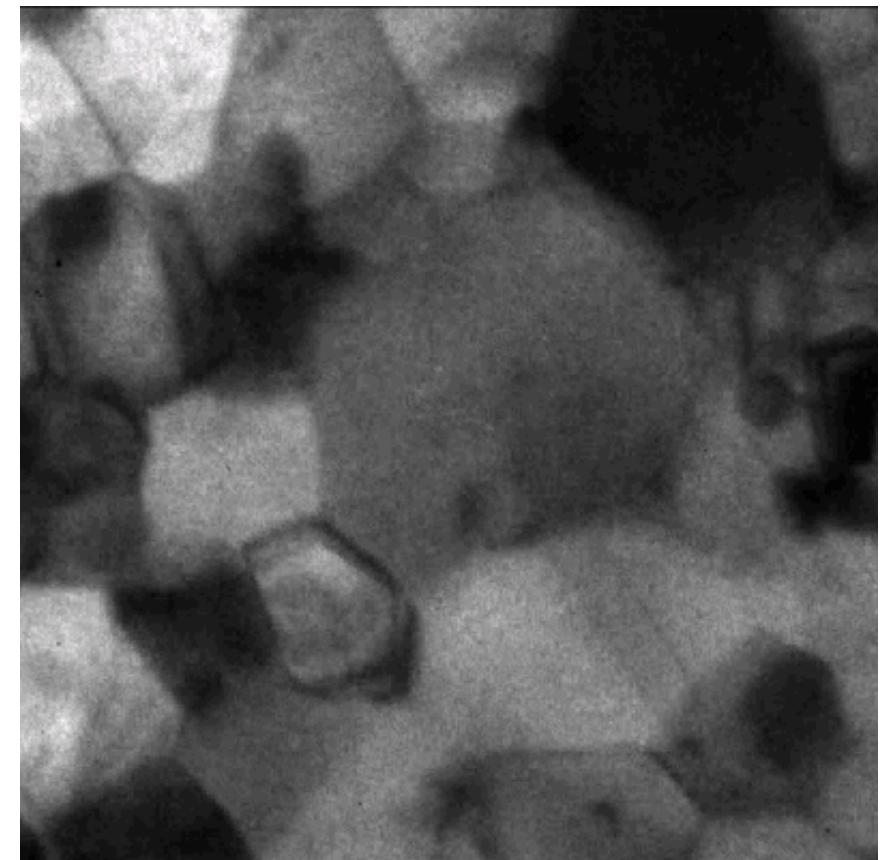
# Grain boundaries and cracks

- Materials often crack along grain boundaries
- Grain boundaries can be weaker than the perfect lattice
  - different bonding environment, increased extrinsic particles, etc.
- Thus, fracture will often occur along grain boundaries (intergranular fracture)



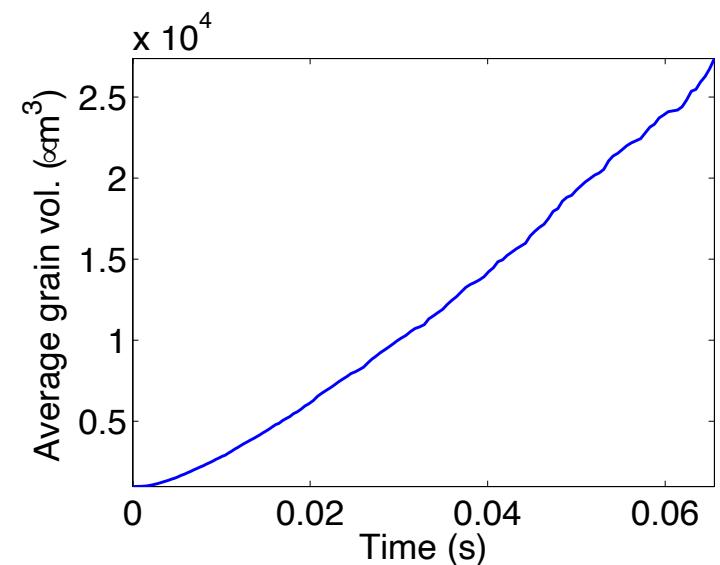
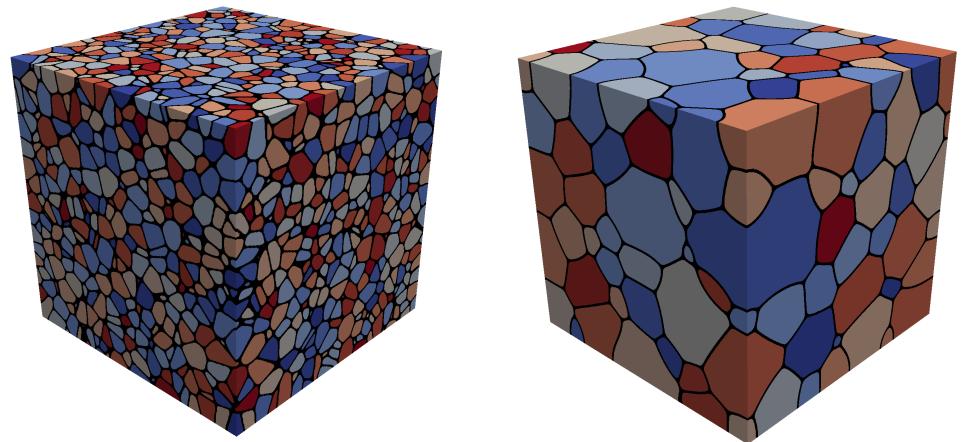
# Grain size change

- A single crystal has a lower energy state than a polycrystal, due to the grain boundary energy
- So, grain boundaries migrate to reduce the energy
- $v_{GB} = M_{GB} (P_d - P_r)$ 
  - $M_{GB}$  is the grain boundary mobility
  - $P_d$  is the driving force (pressure) for grain growth
  - $P_r$  is the pressure resisting grain growth



# Grain growth

- Due to grain boundary migration, the average grain size goes up with time during annealing/heat treating
- As grain boundaries migrate, some grains grow and some shrink
- Shrinking grains eventually disappear
- The average grain volume  $\bar{V}_{gr} = V_{mat}/N_{grains}$
- Therefore, as  $N_{grains}$  decreases due to grain disappearance, the average grain volume goes up



# Grain boundary mobility

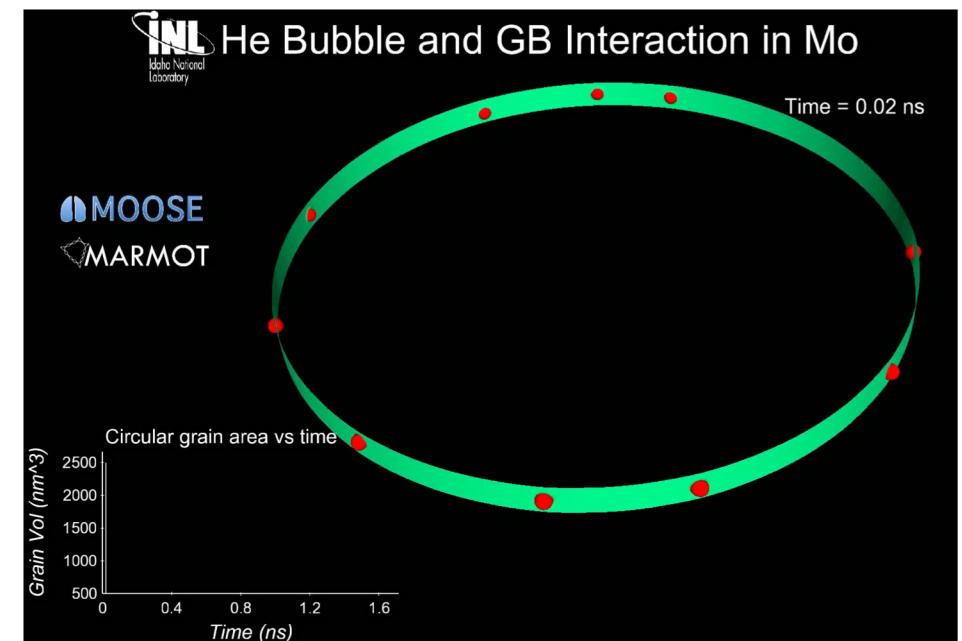
- The grain boundary mobility is a function of temperature and GrB type
- The grain boundary mobility is determined according to
  - $M_{GB} = M_0 e^{-\frac{Q}{k_b T}} \text{ m}^4/(\text{J s})$
  - $k_b = 8.6173e-5 \text{ eV/K}$  is the Boltzmann constant
- Both the pre-factor  $M_0$  and the activation energy  $Q$  change as a function of the grain boundary misorientation
- We often use an average grain boundary for a material, taken from polycrystal measurements-> UO<sub>2</sub>:  $M_0 (\text{m}^4 / \text{J-s}) = 4.6e-09$ ,  $Q (\text{eV}) = 2.77$
- What is the mobility of UO<sub>2</sub> at 1600 K?
  - $M_{GB} = 4.6e-9 * \exp(-2.77/(1600 * 8.6173303e-5)) = 8.7e-18 \text{ m}^4/\text{J-s}$

# Grain growth

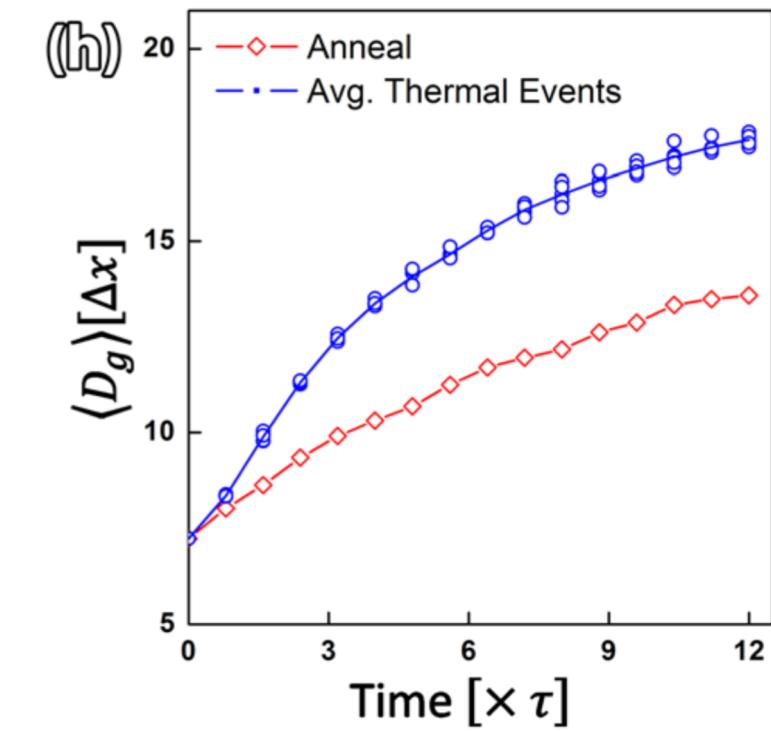
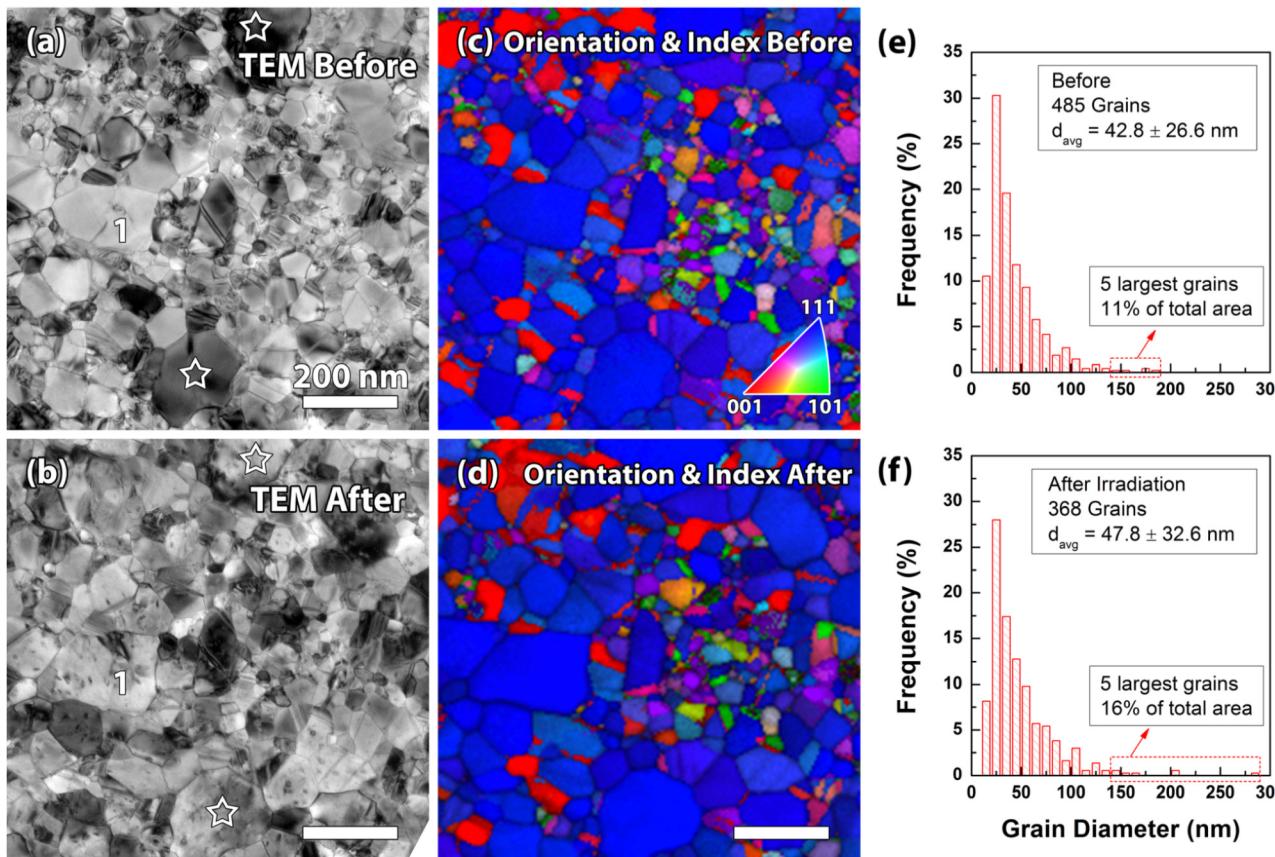
- There are various driving forces for grain growth
- The most common driving force is the reduction of grain boundary energy
  - $P_d = \frac{2\gamma_{GB}}{R}$ , where  $\gamma_{GB}$  is the GB energy and R is the radius of curvature
  - It is often called the curvature driving force, because it drives grain boundaries to be straight
  - It also causes larger grains to grow at the expense of smaller ones
- Other driving forces include
  - Temperature gradients
  - Elastic energy gradients
  - Dislocation energy gradients
- Velocity of a spherical grain:  $v = M \frac{2\gamma_{GB}}{R}$

# Grain growth

- Grain boundary motion is inhibited by pores, precipitates, solute atoms, etc.
- Solute atoms (whether in interstitial sites or vacancies) can decrease the grain boundary mobility.
  - This is called **Solute Drag**
  - Even a small concentration of impurities can decrease the mobility by 3 to 4 orders of magnitude
- Particles and pores resist grain boundary motion



# Irradiation can accelerate grain growth, but it is only significant with small grains and low temperature



# Maximum grain size

- Consider a material with an average grain size D
- The change in D can be written as

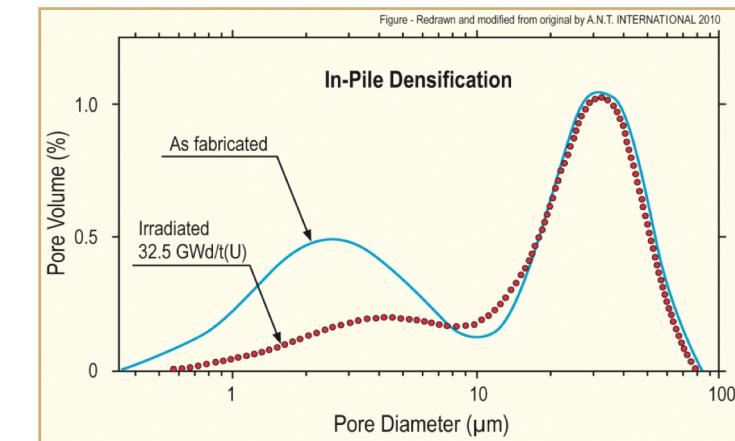
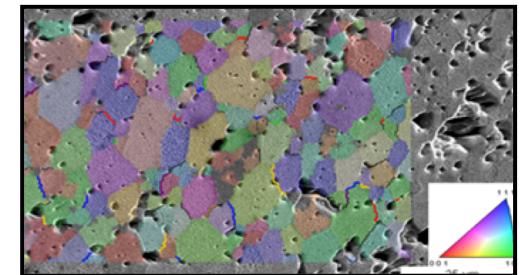
$$\frac{dD}{dt} = k \left( \frac{1}{D} - \frac{1}{D_m} \right)$$

- $k = 2 M_{GB} \gamma_{GB}$  is a rate constant that can be determined from experiments
- $D_m$  is the grain size at which the driving force equals the resistive pressure
- For UO<sub>2</sub>:

Material	$M_0$ (m <sup>4</sup> J/s)	Q (eV)	$\gamma_{GB}$ (J/m <sup>2</sup> )
UO <sub>2</sub>	4.6e-09	2.77	1.58
- $D_m$  is a function of temperature     $D_m = 2.23 \cdot 10^3 \exp(-7620/T)$     microns

# Fuel Densification

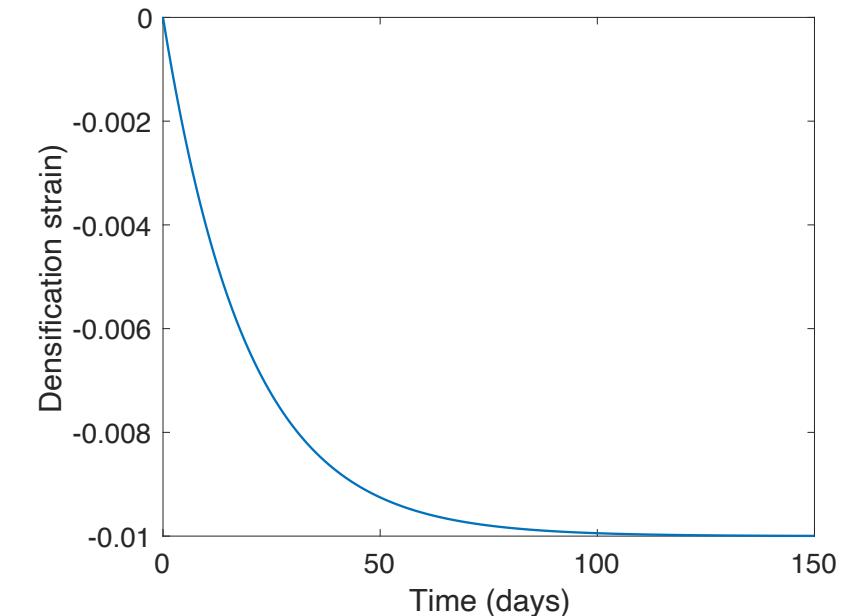
- Remember that fresh fuel pellets are not fabricated to be 100% dense, more like 95-99% dense
- Early in life, fuel pellets shrink and the initial porosity is largely eliminated
- In some ways, densification is a continuation of the sintering process
- However, irradiation accelerates the process
- Small pores close due to effects of fission spikes and vacancy diffusion
- Large pores stable (in absence of large hydrostatic stress)
- Pellets with higher initial density, densify less



# Densification

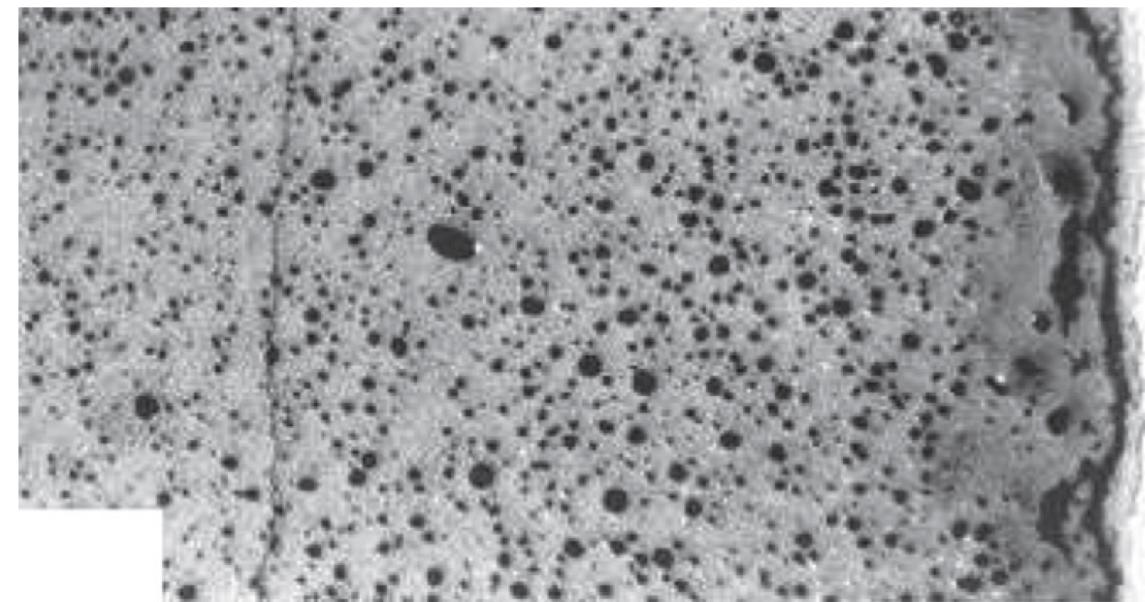
- The driving force for densification is the change in free energy from the decrease in surface area of pores and lowering of the surface free energy
- An empirical relation to describe densification has been built as a function of
  - $\beta$  - Burnup (in FIMA)
  - $\Delta\rho_0$  – Total densification that can occur ~ 0.01
  - $\beta_D$  – Burnup at which densification stops ~5 MWD/kgU, ~0.005 FIMA
  - $C_D = 7.235 - 0.0086(T - 25)$  for  $T < 750C$  and  $C_D = 1$  for  $T \geq 750C$

$$\epsilon_D = \Delta\rho_0 \left( e^{\frac{\beta \ln 0.01}{C_D \beta_D}} - 1 \right)$$



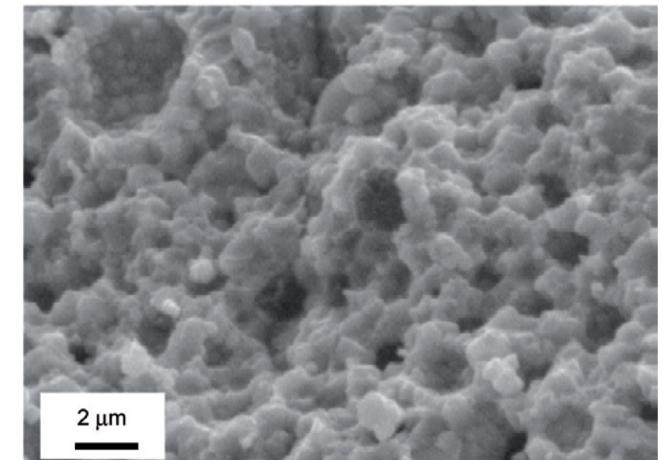
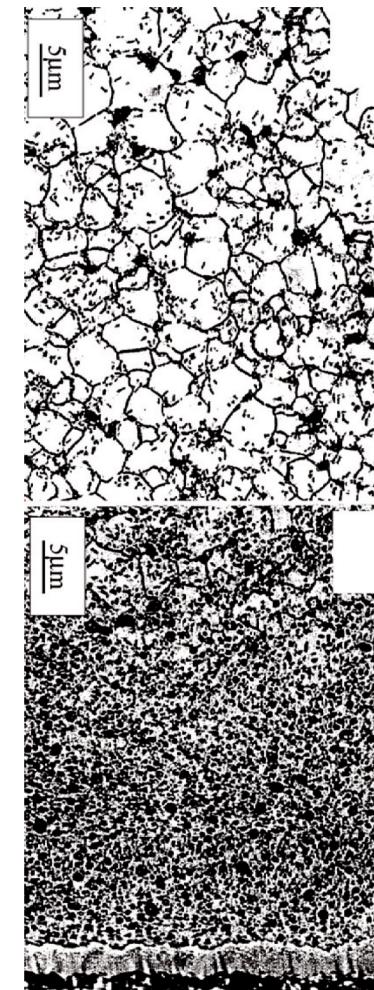
# High Burnup Structure (HBS)

- 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
- In UO<sub>2</sub>, grains subdivide (to 100-500 nm size) and a densely porous structure is formed



# 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

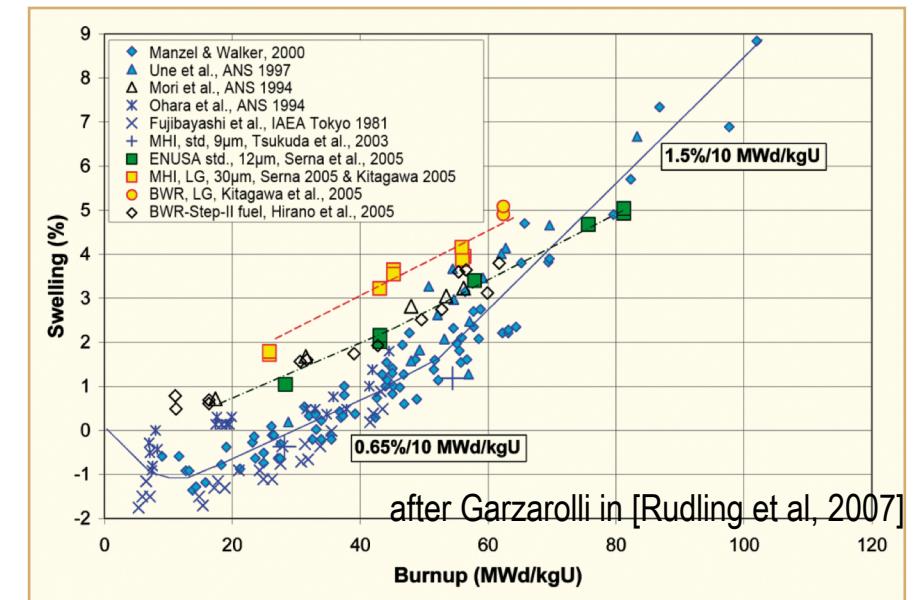


# Empirical models

- Many models of reactor fuel behavior are fits to experimental data
- They are typically correlated to burnup
- Burnup is a measure of how much fissioning has taken place. Typical units are:
  - MWD/kgU
  - FIMA
- It can be calculated from the volumetric fission rate

$$\beta = \frac{\dot{F}t}{N_U}$$

- Units of this equation are FIMA
- To convert from FIMA to MWD/kgU, multiply by 950



# Summary

- The average grain size in  $\text{UO}_2$  impacts fuel behavior and performance:
  - Fission gas release
  - Swelling
  - Thermal conductivity
  - Creep
- The material wants to reduce its energy by having large grains grow at the expense of small grains
- Grain growth is reduced due to other defects reducing the grain boundary migration
- Fuel densification is driven by reduction in surface area of pores – continuation of sintering process
- Empirical models describe densification as a function of burnup

# Notes

- End of second section
- Presentations Next Week
- Problem Session 3/16
- Exam 3/18