PennState
College of Engineering

NucE 497: Reactor Fuel Performance

Lecture 27: High burnup structure and thermal conductivity

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Michael R Tonks
Mechanical and Nuclear Engineering

Content taken from slides from ANT international

Today we will discuss dimensional changes in UO_2 due to densification, swelling, creep, and fracture

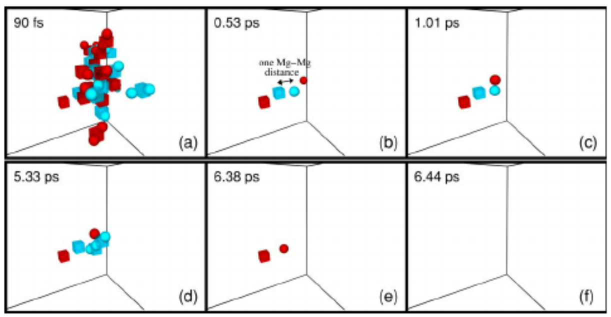
- Module 1: Fuel basics
- Module 2: Heat transport
- Module 3: Mechanical behavior
- Module 4: Materials issues in the fuel
  - Property evolution and Intro to materials science
  - Chemistry
  - Grain growth
  - Fission products and fission gas
  - Densification, swelling, and creep
  - Fracture
  - **HBS**
  - **Thermal conductivity**
- Module 5: Materials issues in the cladding
- Module 6: Accidents, used fuel, and fuel cycle

Here is some review from last time

- During the lifetime of the fuel, its volume
  a) Decreases and then increases
  b) Increases and then decreases
  c) Decreases, increases, and then decreases
  d) Increases, decreases, and then increases
- Which statement is true
  a) Creep causes the fuel volume to increase
  b) Pellet fracture decreases the size of the gap
  c) Creep is shape change that occurs without stress but with irradiation
  d) Creep and fracture do not occur in the fuel

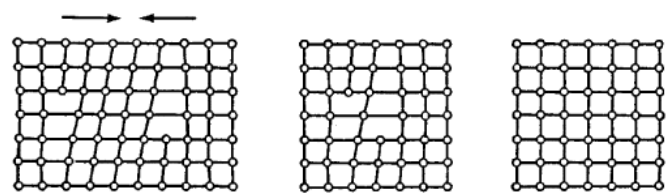
Materials reduce the free energy contributed by defects in various ways

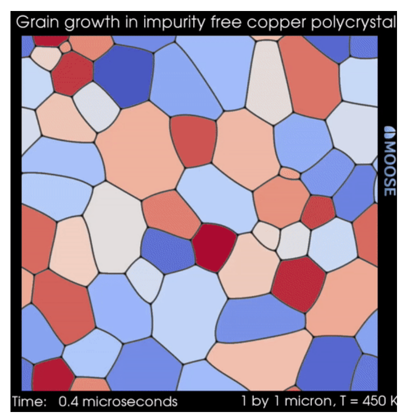
- Vacancies and interstitials recombine or are absorbed by other defects



Uberuaga, B. P., et al. "Dynamical simulations of radiation damage and defect mobility in MgO." Physical Review B 71.10 (2005): 104102.

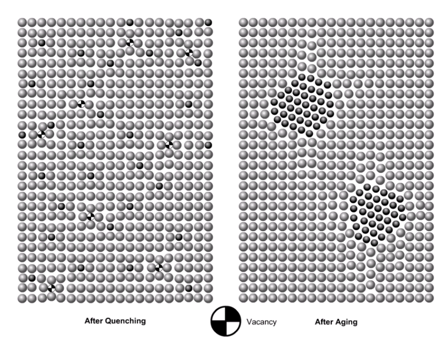
- Dislocations move and annihilate





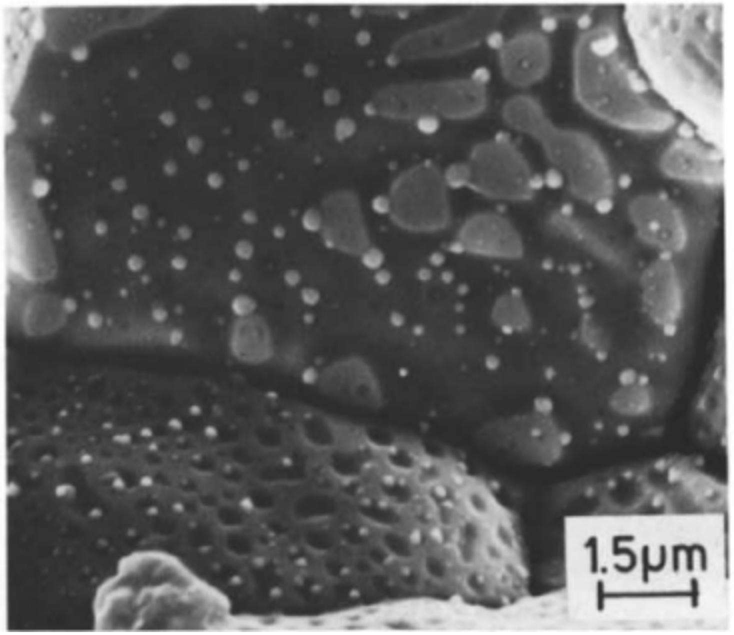
- Grain boundaries migrate

- Impurity atoms cluster to form precipitates

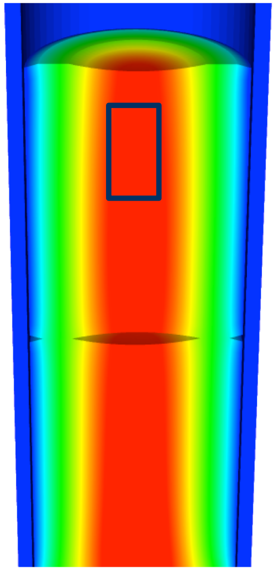


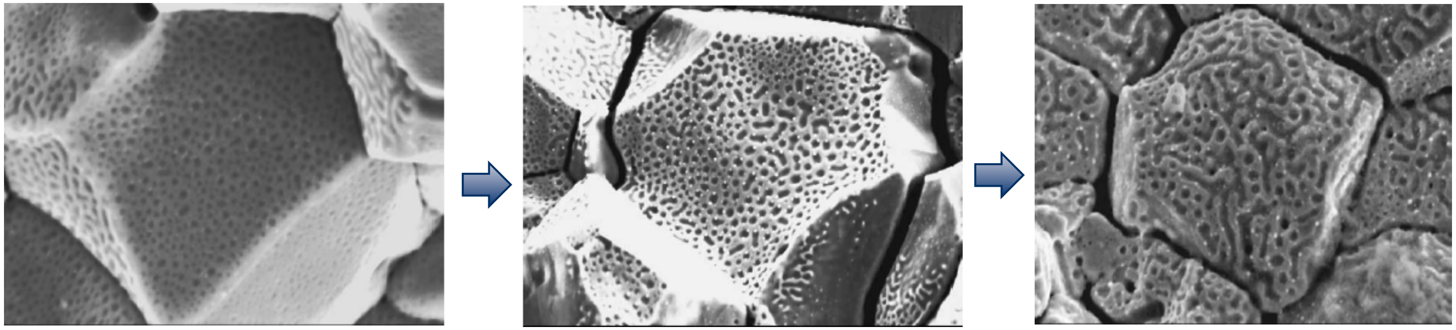
However, all these mechanisms are thermally activated

- When the temperature is low, the material must take more drastic measures

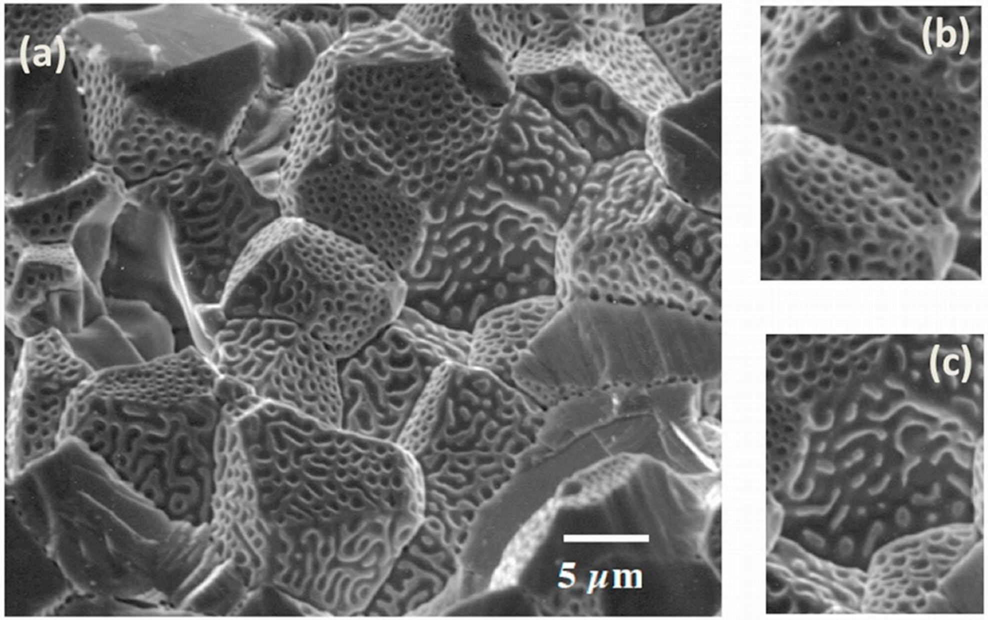


At the hottest regions of the fuel, defects and fission products cluster or segregate to interfaces

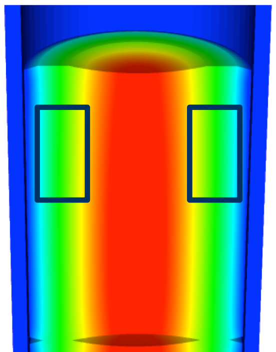




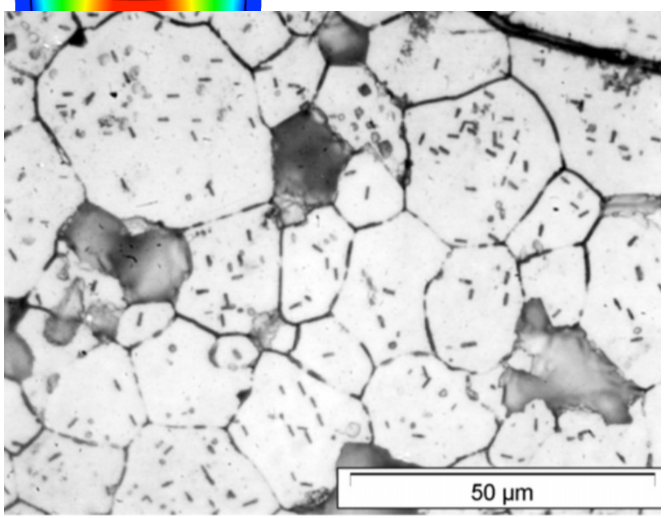
Nucleation Growth Percolation



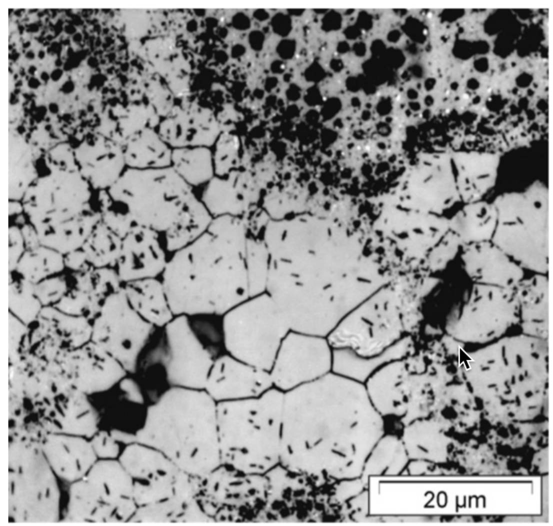
In the cool regions of the fuel, the diffusion is too slow for defects to arrive at the interfaces



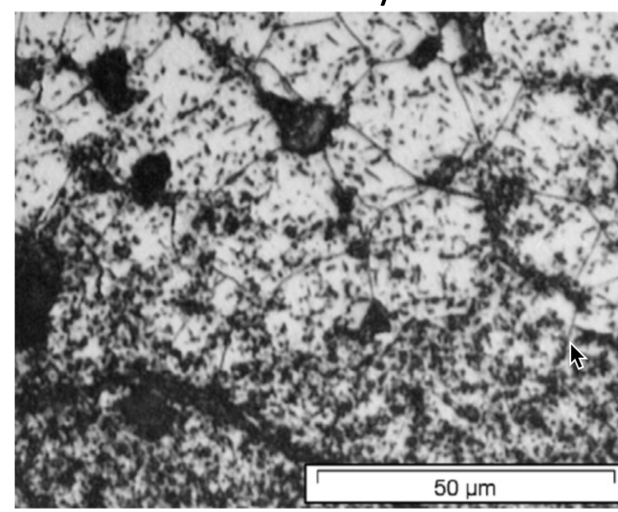
30 GWd/t



57 GWd/t

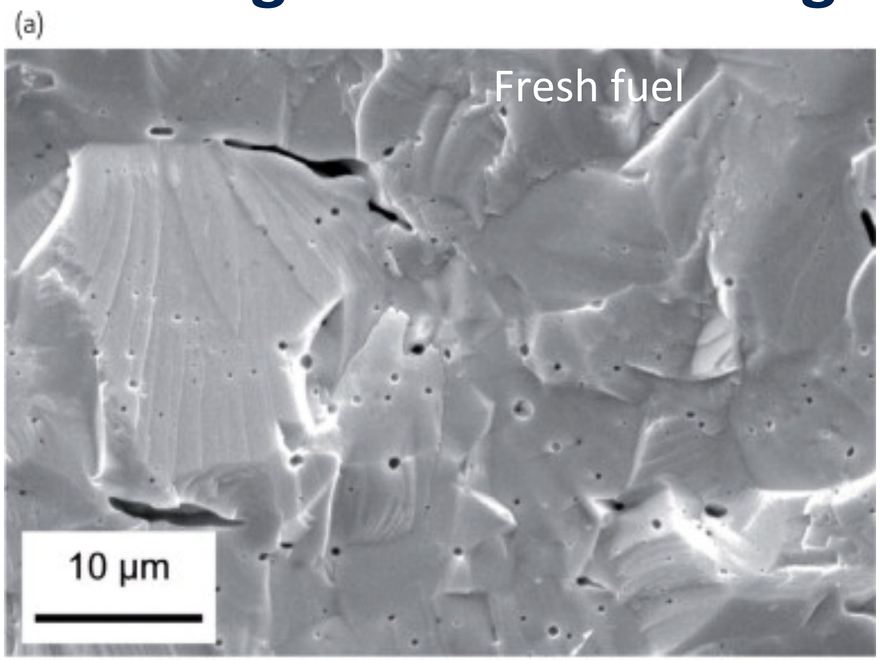


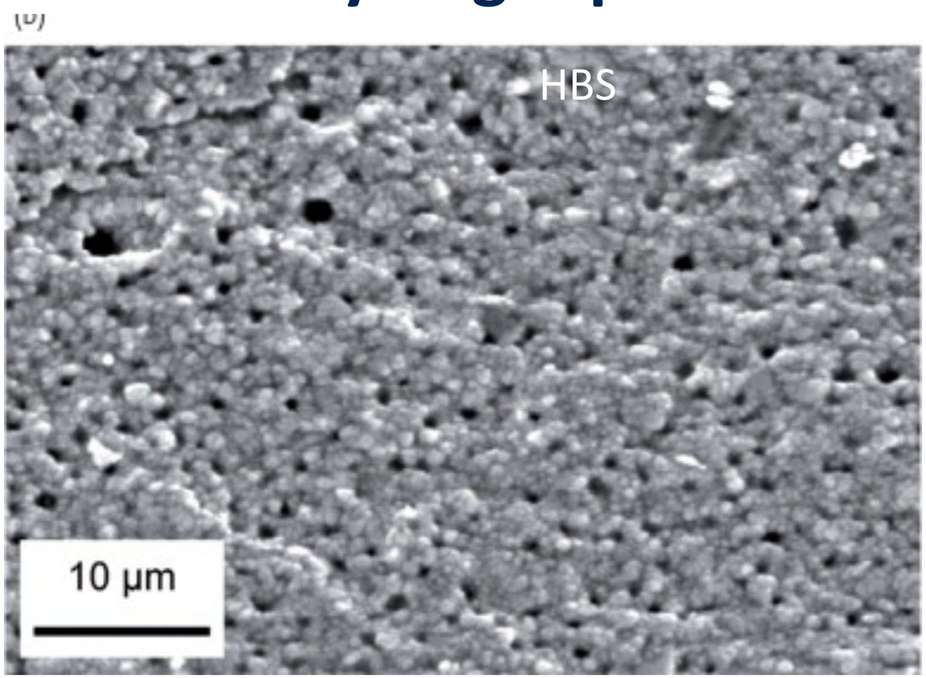
69 GWd/t

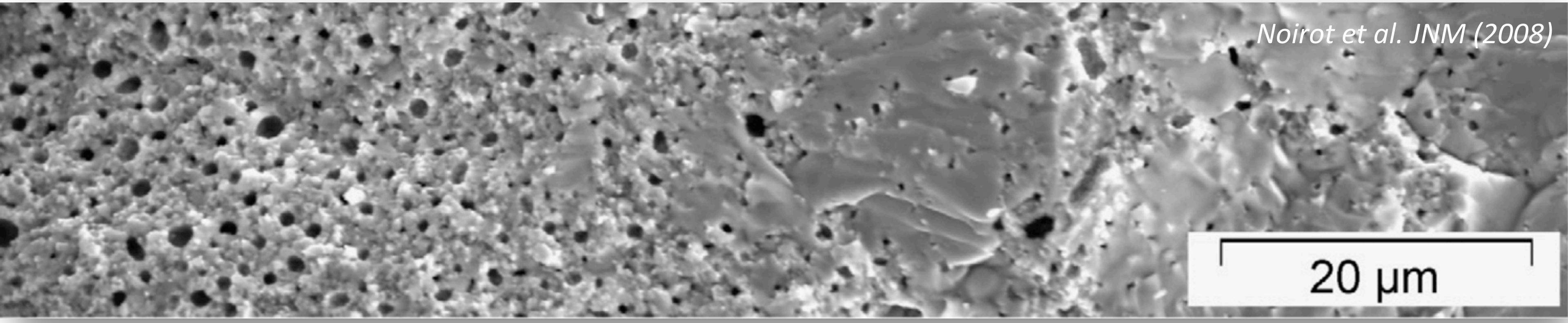


Noirot et al. JNM (2008)

In order to annihilate the defects, the structure changes to have small grains and many larger pores

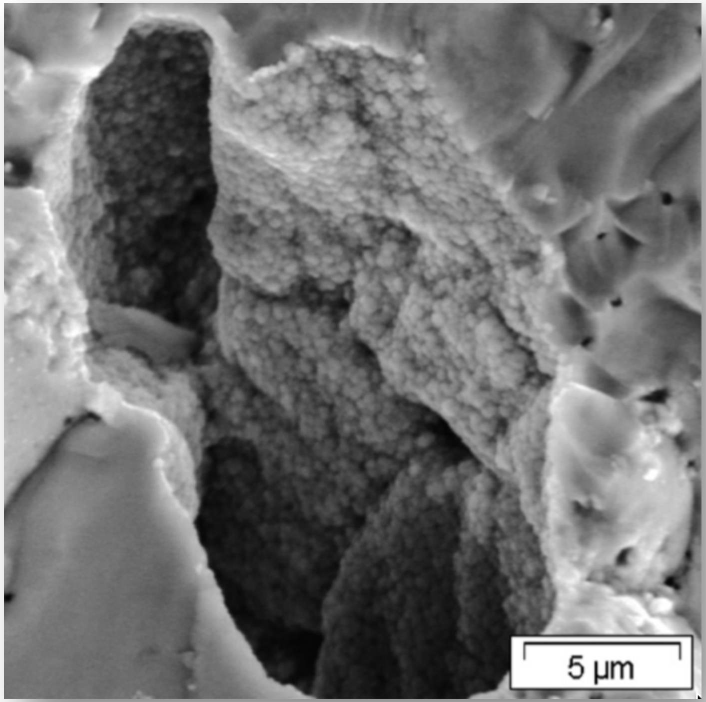


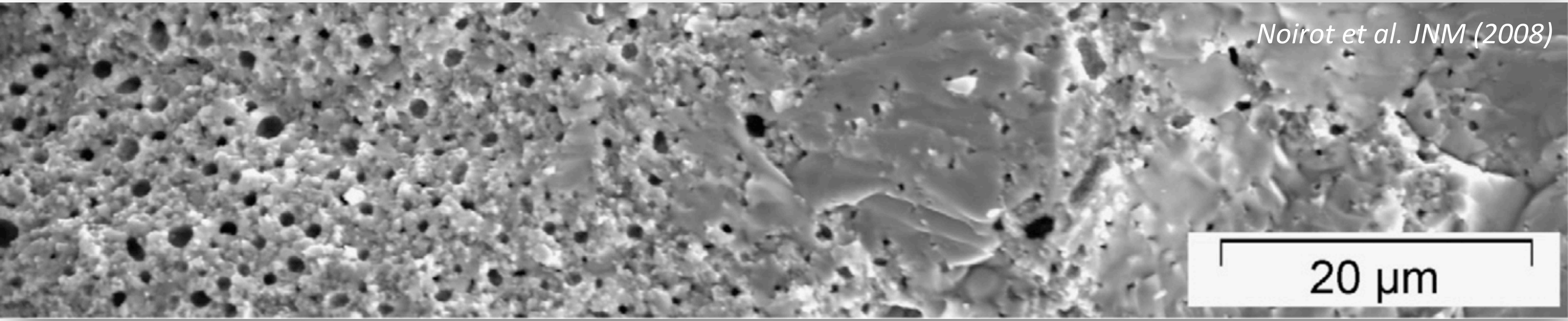




The mechanism for the formation of HBS is still up for debate

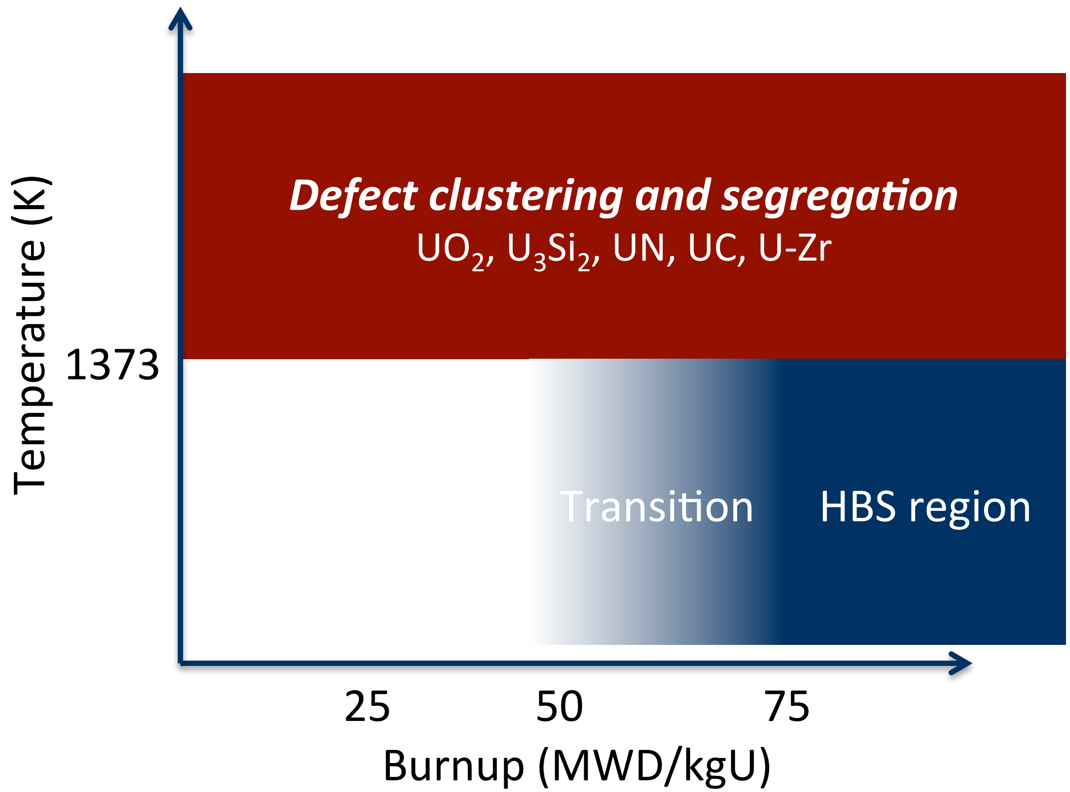
- One theory is that dislocation networks cause grain subdivision
- Another is that dislocation networks cause polyganization
- Neither of these account for the importance of the planar defects





Noirot et al. JNM (2008)

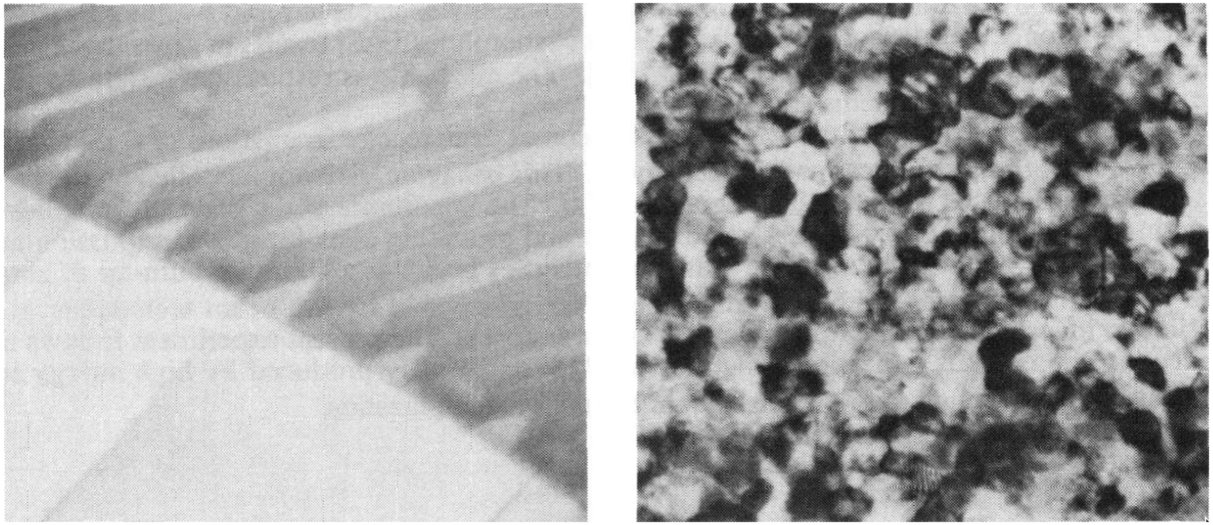
In UO_2, their appears to be a burnup and temperature threshold for this restructuring



This grain restructuring also occurs in uranium silicides, nitrides, carbides, and metal alloys

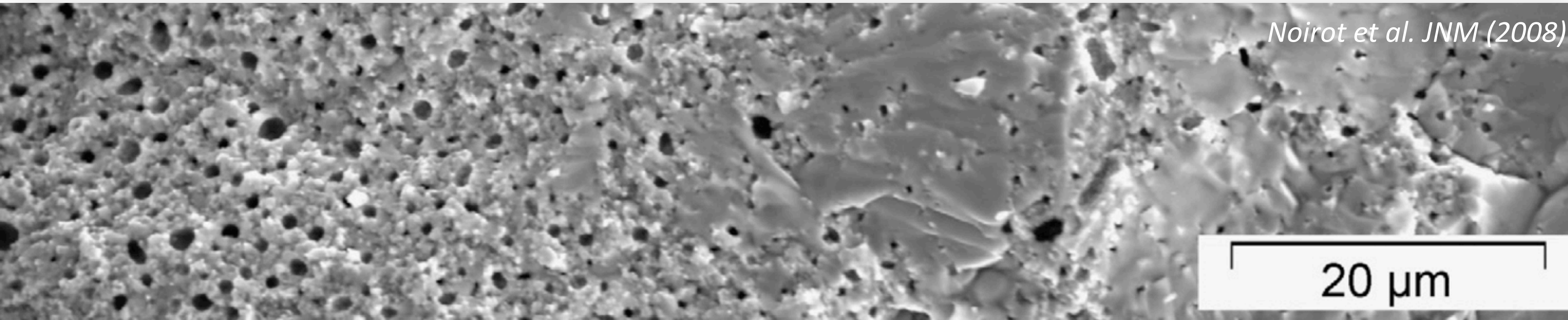
- The formation of high burnup structure is not unique to UO_2, but occurs in all fuel types at high burnup and lower temperature

Grain subdivision in U_3Si_2 under ion irradiation

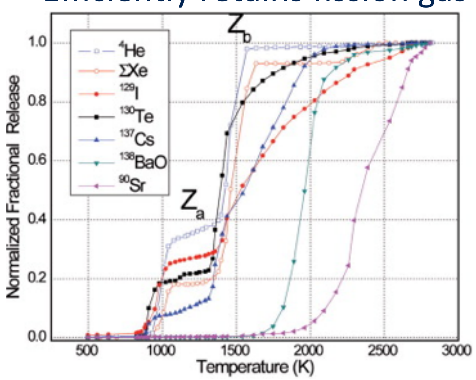


Birtcher and Wang, 1991

Originally it was assumed to hurt fuel performance, but recent research shows it has a positive effect

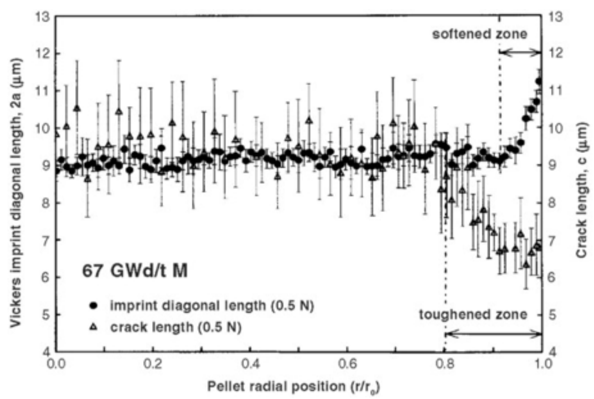


Efficiently retains fission gas



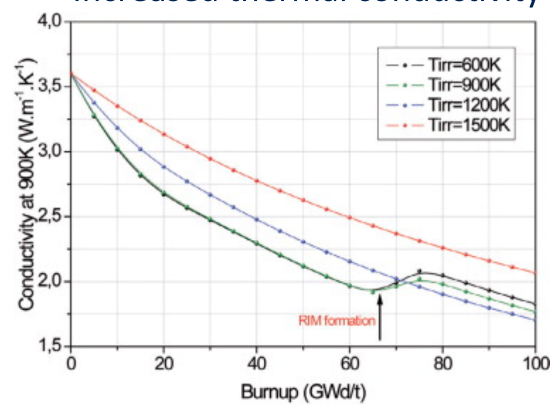
From Hiernaut et al. JNM (2008)

Increased toughness and softness



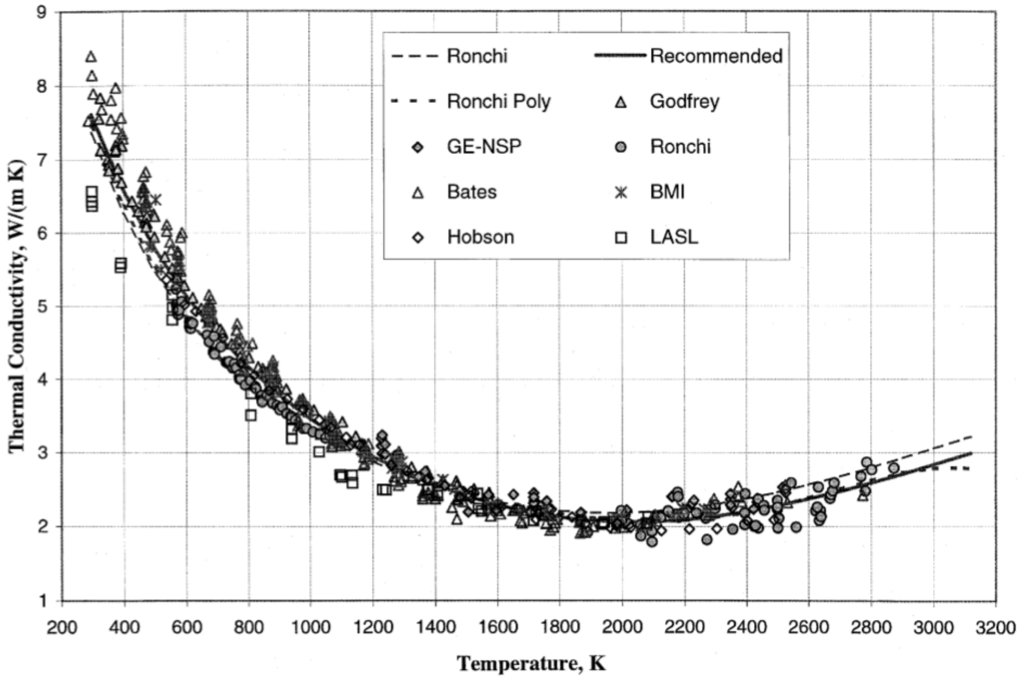
From Spino et al. JNM (2003)

Increased thermal conductivity



From Ronchi et al. JNM (2004)

We've already discussed the thermal conductivity of fresh fuel



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

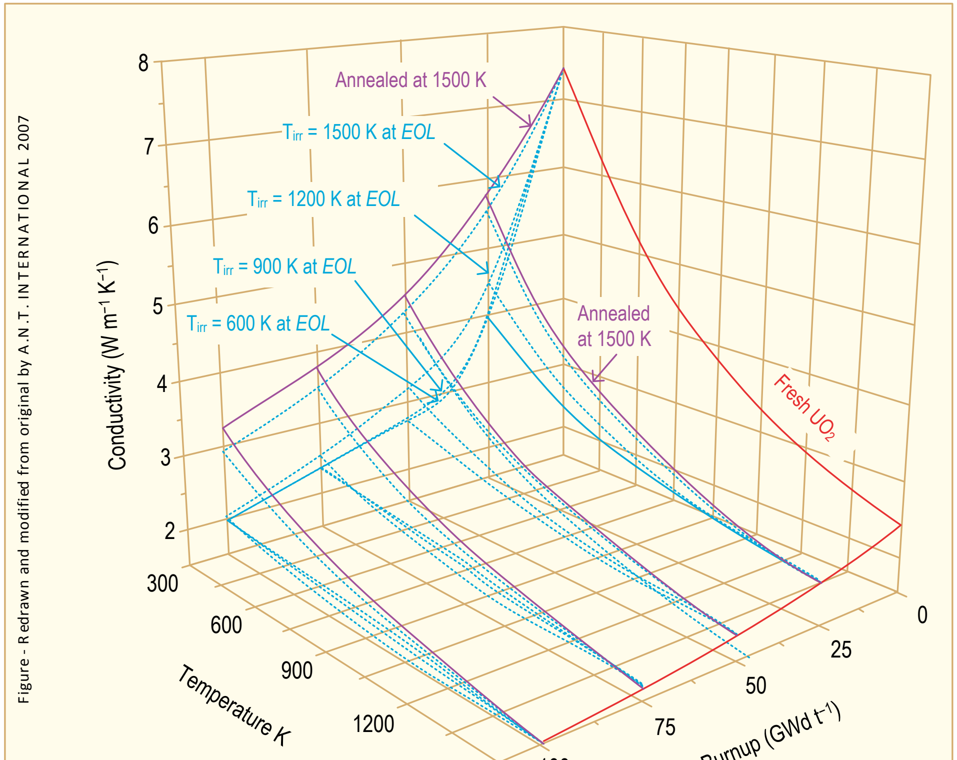
- Where t = T/1000
- The first part of the equation describes the phonon interactions
- The second part describes electronic transport which becomes significant at high temperature

J.K. Fink, J. Nucl. Mater. 279 (2000) 1-18

UO_2 thermal conductivity is low and decreases more during reactor life.

- The thermal conductivity has been collected after various amounts of burnup to make empirical fits

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



after [Ronchi et al, 2004]

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).

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

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

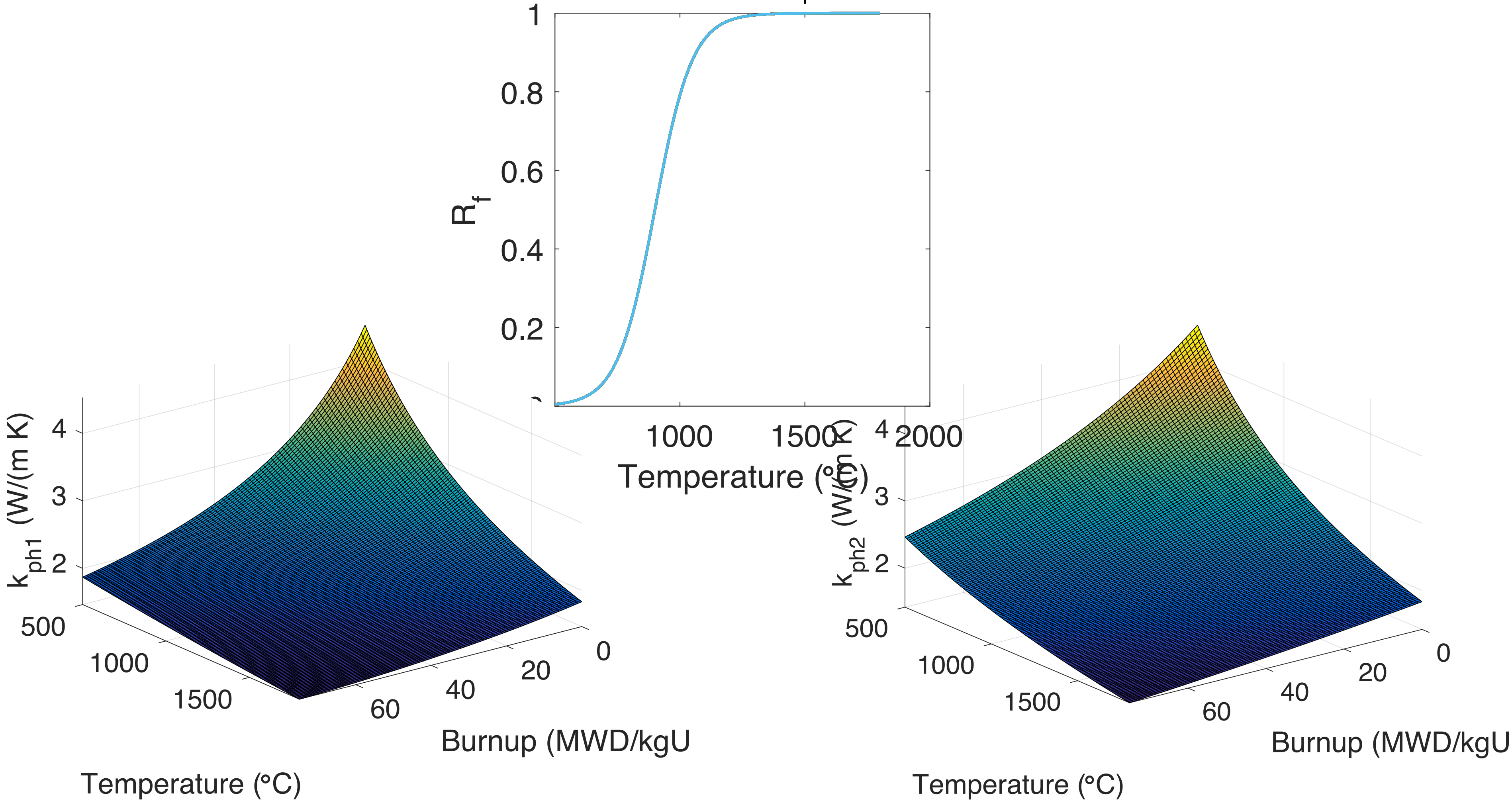
$$k_{ph1} = \frac{1}{(9.592\times10^{-2} + 6.14\times10^{-3}\beta - 1.4\times10^{-5}\beta^2 + (2.5\times10^{-4} - 1.81\times10^{-6}\beta)T}$$

$$k_{ph2} = \frac{1}{(9.592\times10^{-2} + 2.6\times10^{-3}\cdot\beta + (2.5\times10^{-4} - 2.7\times10^{-7}\beta)T}$$

$$k_{el} = 1.32\times10^{-2}\,e^{1.88\times10^{-3}T}$$

The primary empirical model used in BISON is the NFIR model

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



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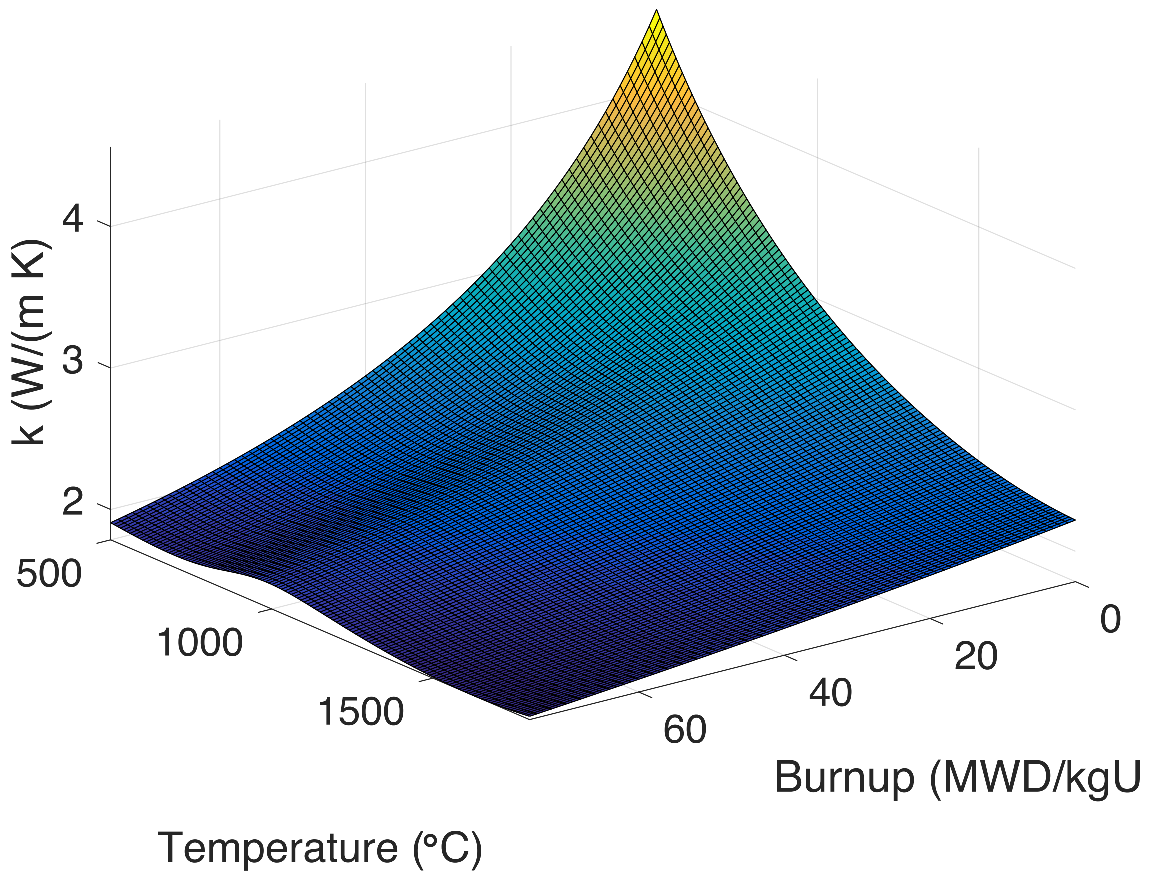


Now, let's work a problem

- What is the thermal conductivity of a fuel predicted by the NFIR model at a temperature of 1200 K and a burnup of 5% FIMA?
- First, we need to convert T to °C and burnup to MWD/kgU
  - T = 1200 K – 273.15 = 926.85 °C, β = 0.05 FIMA*950 = 47.5 MWD/kgU
- Next, we need to calculate R_f $R_f(T) = \frac{1}{2}\left(1 + \tanh\left(\frac{T - 900}{150}\right)\right)$
  - R_f = 0.5*(1 + tanh((926.86 – 900)/150)) = 0.5886
- Now, we need k_{ph1} $k_{ph1} = \frac{1}{(9.592 \times 10^{-2} + 6.14 \times 10^{-3}\beta - 1.4 \times 10^{-5}\beta^2 + (2.5 \times 10^{-4} - 1.81 \times 10^{-6}\beta)T}$
  - k_{ph1} = 1/(9.592e-2 + 6.14e-3*47.5 – 1.4e-5*47.5^2 + (2.5e-4 – 1.81e-6*47.5)*926.85) = 1.9685 W/(m K)
- Then, k_{ph2} $k_{ph2} = \frac{1}{(9.592 \times 10^{-2} + 2.6 \times 10^{-3} \cdot \beta + (2.5 \times 10^{-4} - 2.7 \times 10^{-7}\beta)T}$
  - k_{ph2} = 1/(9.592e-2 + 2.6e-3*47.5 + (2.5e-4 – 2.7e-7*47.5)*926.85) = 2.2766 W/(m K)
- Finally, k_{el} $k_{el} = 1.32 \times 10^{-2}\, e^{1.88 \times 10^{-3} T}$
  - k_{el} = 1.32e-2*exp(1.88e-3*926.85) = 0.0754 W/(m K)
- Then, we put it all together $k = (1 - R_f(T))k_{ph1}(T,\beta) + R_f(T)k_{ph2}(T,\beta) + k_{el}(T)$
  - k = (1 – 0.5886)*1.9685 + 0.5886*2.2766 + 0.0754 = 2.23 W/(m K)

Now you do a problem

- What is the thermal conductivity of a fuel predicted by the NFIR model at a temperature of 1200 K and a burnup of 0% FIMA?
  - Remember T = 1200 K – 273.15 = 926.85 °C, R_f = 0.5886, and k_{el} = 0.0754 W/(mK)

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

$$k_{ph1} = \frac{1}{(9.592\times10^{-2} + 6.14\times10^{-3}\beta - 1.4\times10^{-5}\beta^2 + (2.5\times10^{-4} - 1.81\times10^{-6}\beta)T}$$

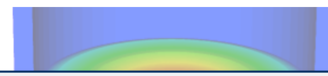
$$k_{ph2} = \frac{1}{(9.592\times10^{-2} + 2.6\times10^{-3}\cdot\beta + (2.5\times10^{-4} - 2.7\times10^{-7}\beta)T}$$

- So, we need to calculate k_{ph1} and k_{ph2}
- First, we need to calculate k_{ph1}
  - k_{ph1} = 1/(9.592e-2 + 6.14e-3*0 – 1.4e-5*0^2 + (2.5e-4 – 1.81e-6*0)*926.85)
  - k_{ph1} = 1/(9.592e-2 + 2.5e-4*926.85) = 3.0522 W/(m K)
- Next, we need to calculate k_{ph2}
  - k_{ph2} = 1/(9.592e-2 + 2.6e-3*0 + (2.5e-4 – 2.7e-7*0)*926.85)
  - Without the burnup terms, this is the same as k_{ph2}!
- So, the final expression for k is
  - k = k_{ph1} + k_{el} = 3.0522 + 0.0754 = 3.13 W/(m K)

Fuel performance codes historically rely on materials models correlated to temperature and burn-up

Operating Conditions: Neutron flux | Coolant T&P

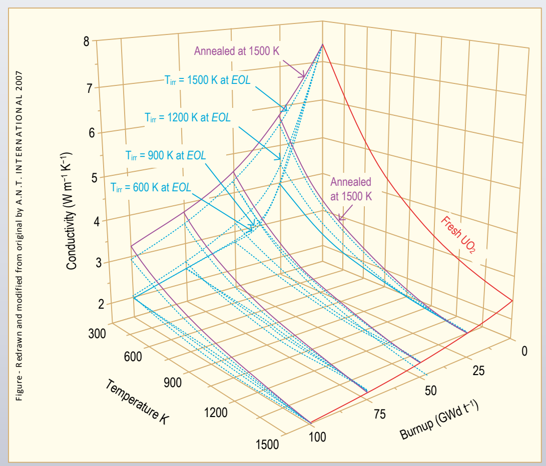
Variables: Temperature | Displacement



Cladding Property va

- Elasticit tenso
- Yield str
- Therma conducti
- Failure criteri
- Dimension change

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



after [Ronchi et al, 2004]

Fuel ty values

- sticity ensor
- ermal ductivity
- acture tress
- elting perature
- ffusion constants
- Dimensional change

Gap conductance | Gap pressure

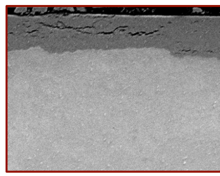
We have begun development of models based on microstructure rather than burn-up

Operating Conditions: Neutron flux | Coolant T&P

Variables: Temperature | Displacement | Stoichiometry

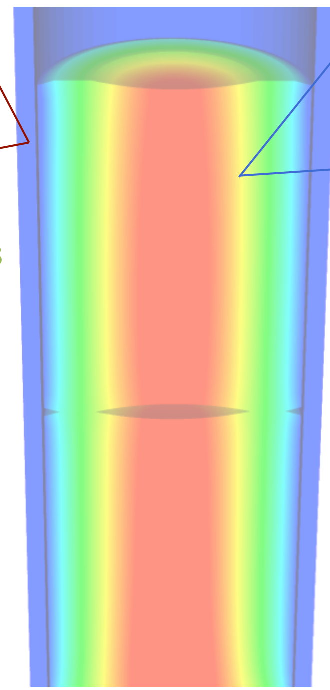
Microstructure evolution models

- Grain growth
- Plasticity
- Defect prod. & transport
- Hydrogen transport
- Loop evolution



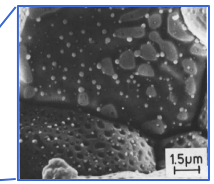
Cladding Microstructure variables

- Average grain size
- Dislocation density
- H concentration
- Hydride fraction
- Point defect concentrations
- Loop density
- 2nd 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

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

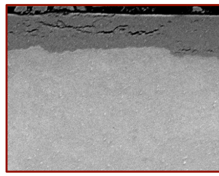
Structure/property relationships connect the microstructure variables to the property values

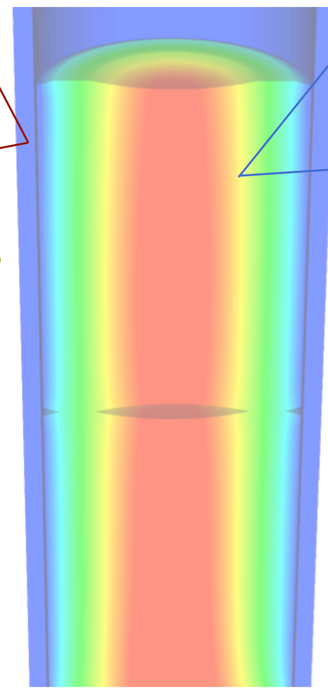
Operating Conditions: Neutron flux | Coolant T&P

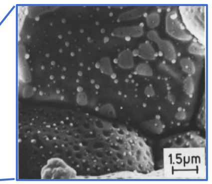
Variables: Temperature | Displacement | Stoichiometry

Cladding Property values

- Elasticity tensor
- Yield stress
- Thermal conductivity
- Failure criteria
- Dimensional change







Cladding Microstructure variables

- Average grain size
- Dislocation density
- H concentration
- Hydride fraction
- Point defect concentrations
- Loop density
- 2nd phase particle fraction

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

Gap variable

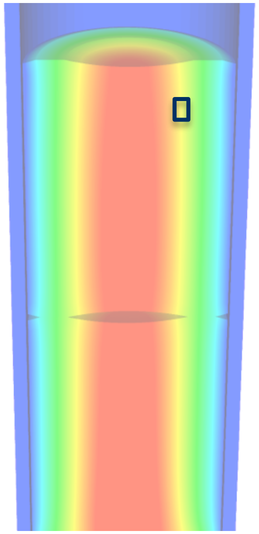
- Gap fission gas concentration

Gap conductance | Gap pressure

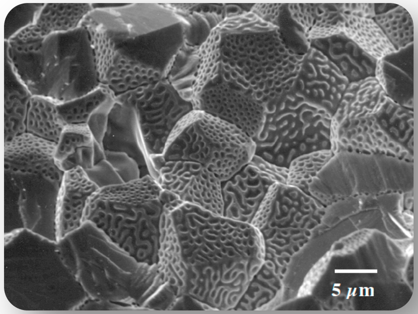
Fuel Property values

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

Example: Fission gas behavior in the fuel



Variables Temperature | Displacement | Stoichiometry



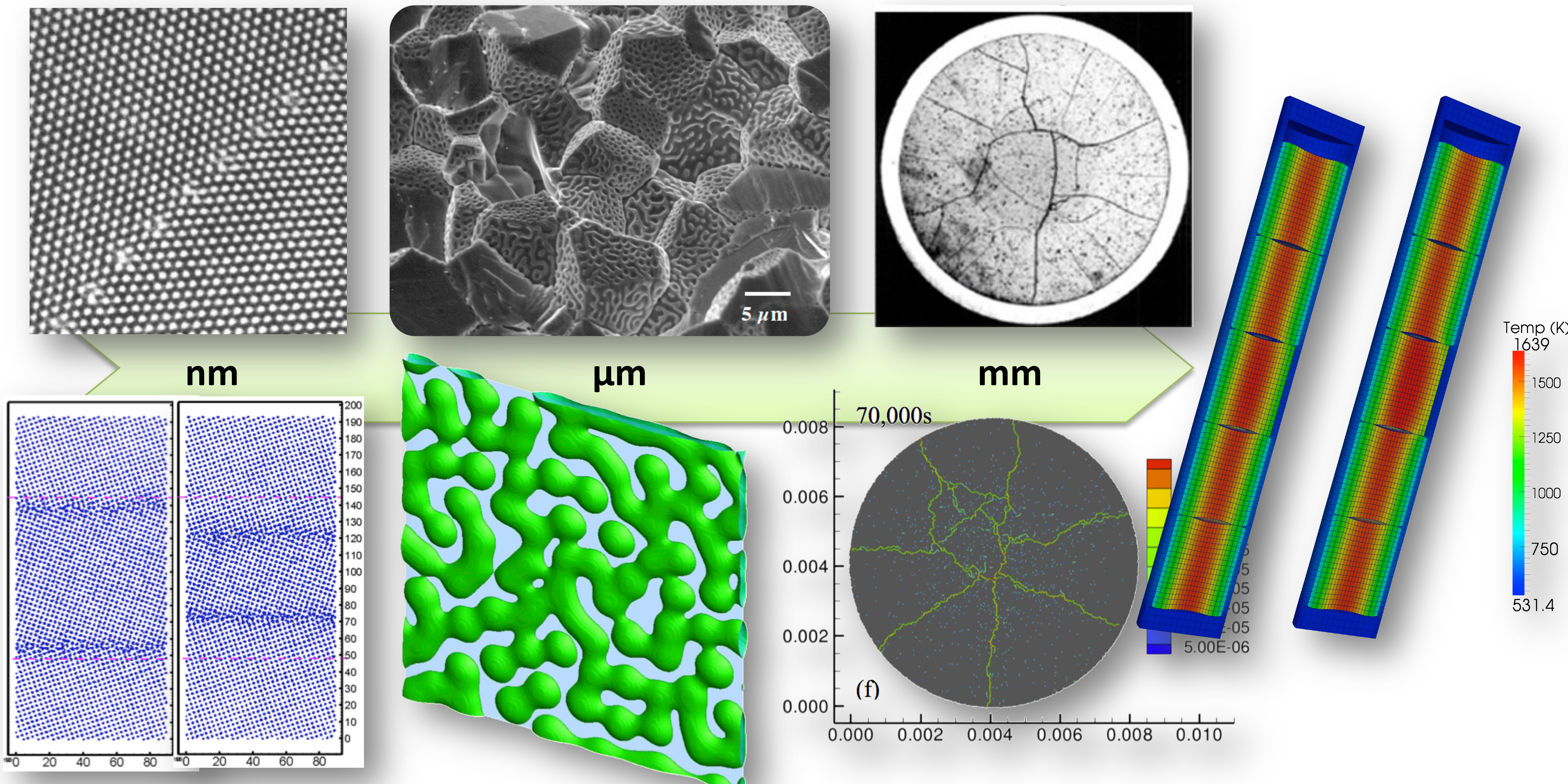
Model of fission gas behavior

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

Structure/property relationships

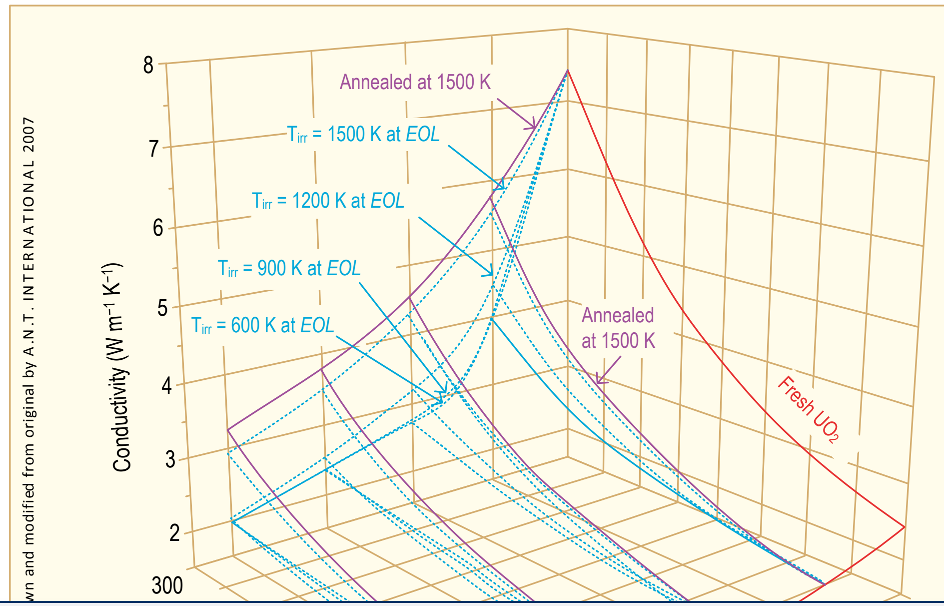
- Elasticity tensor
- Thermal conductivity
- Fracture stress
- Dimensional change
- Gap conductance
- Gap pressure

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



A critical structure/property relationship is between the microstructure and the thermal conductivity

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



Grain boundary and bubbles

Intragranular porosity

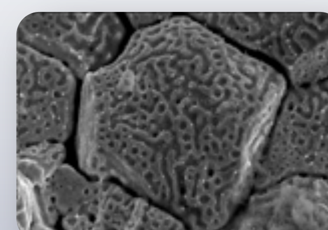
Precipitated fission products

$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

Bulk conductivity

Vacancies and interstitials

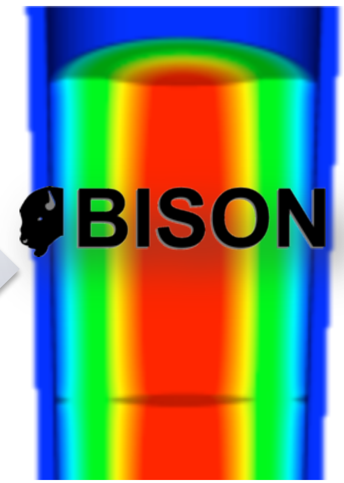
Fission gas



We employ multiscale modeling and simulation to determine the various parameters for the model

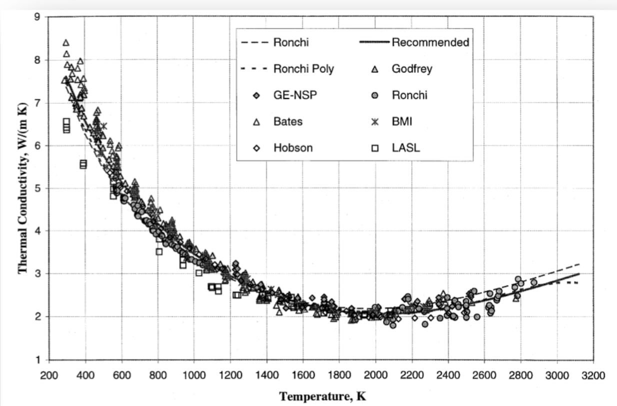
microns
Mesoscale
- Solves Fourier's law with microstructure dependent thermal conductivity

$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

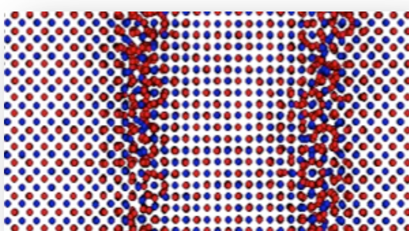


millimeters and up
Engineering scale
- Solves Fourier's Law

Experimental data



J.K. Fink, J. Nucl. Mater. 279 (2000) 1-18

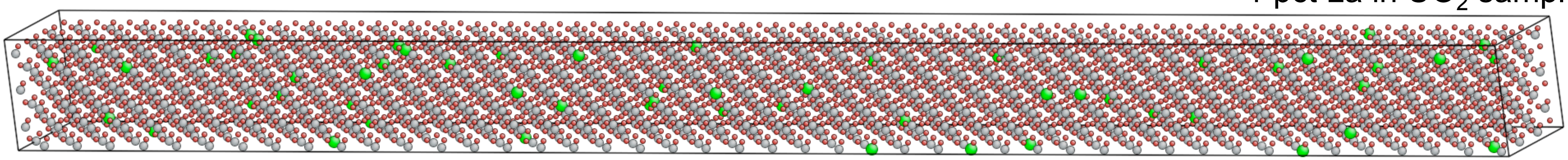


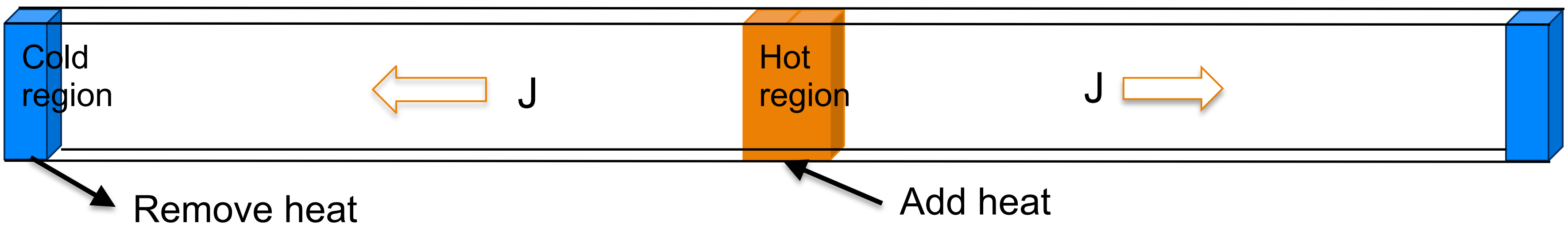
100's of nanometers
Molecular Dynamics
- Explicitly model phonon transport

$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

MD simulations conducted at LANL have been used to determine the coefficients for various point defects

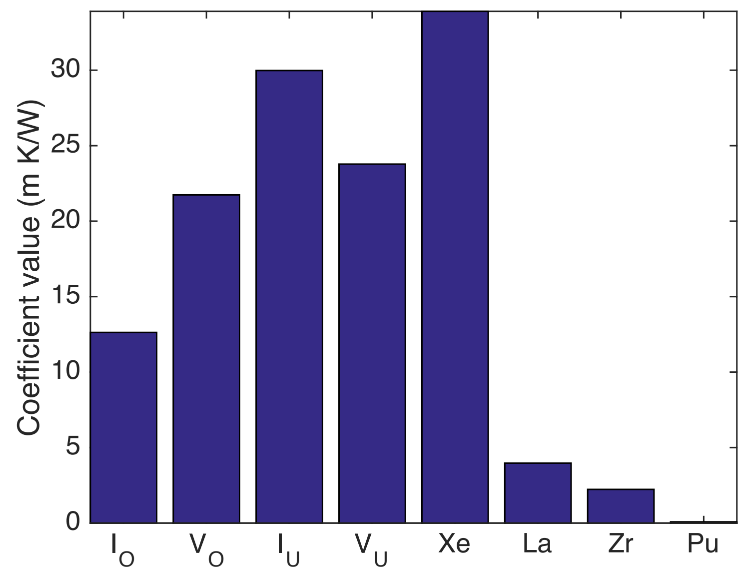
1 pct La in UO_2 sample





$$k = \frac{100}{7.5408 + 17.629t + 3.6142t^2 + 100\sum_i a_i C_i}$$

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



$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

Phonon scattering must also be included when modeling the impact of intragranular bubbles

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- Intragranular bubbles in the fuel are small and spherical, $r_b < 5$ nm.
- Various analytical models exist that represent porosity using a linear thermal resistor approximation but do not consider phonon scattering
- MD simulations have shown that phonon scattering must be accounted for to accurately represent small bubbles

Maxwell-Eucken (no phonon scattering)

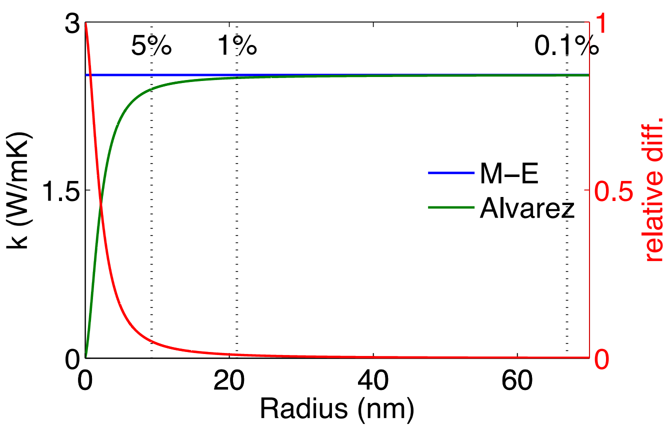
$$\kappa_{ME} = \frac{1-p}{1+p/2}$$

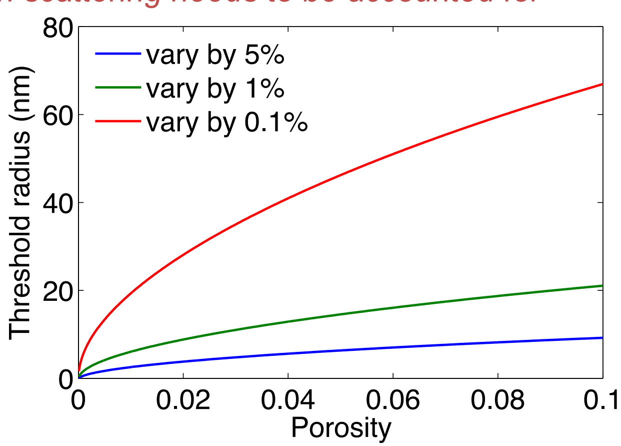
vs

Alvarez (phonon scattering)

$$\kappa_A = \left(\frac{1}{k_{ME}} + \frac{9l^2}{2r^2} \frac{p(1+3\sqrt{p/2})}{1+(0.864+0.29e^{-1.25\frac{r}{l}})\frac{l}{r}} \right)^{-1}$$

Phonon scattering needs to be accounted for

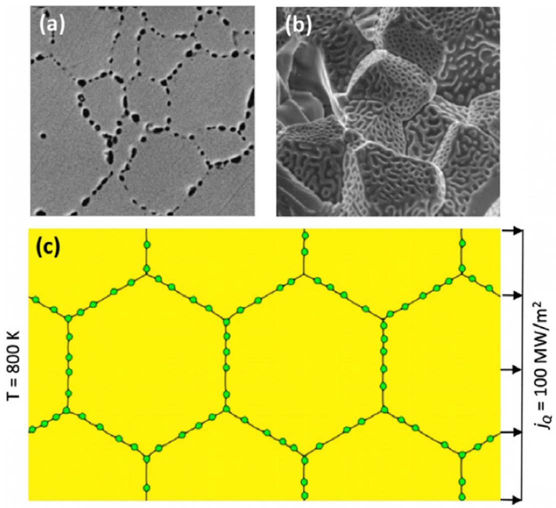


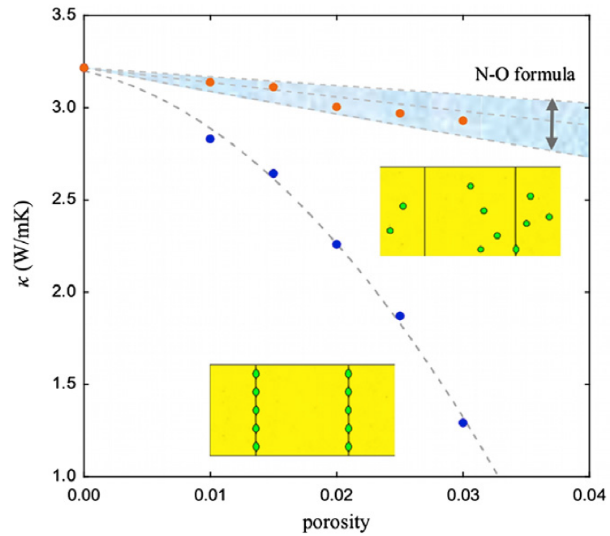


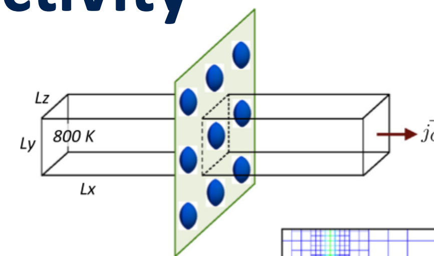
C.-W. Lee, et al., J. Nucl. Mater., 456 (2015) 253 – 259

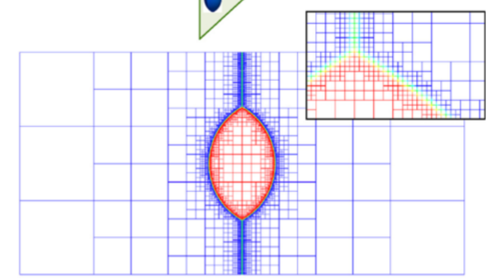
$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

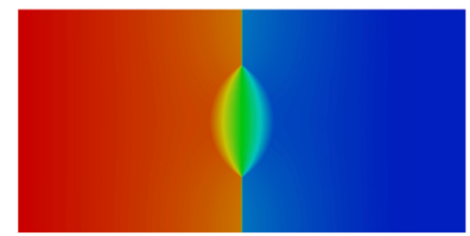
Mesoscale simulations have shown that GB bubbles have a larger impact on the thermal conductivity











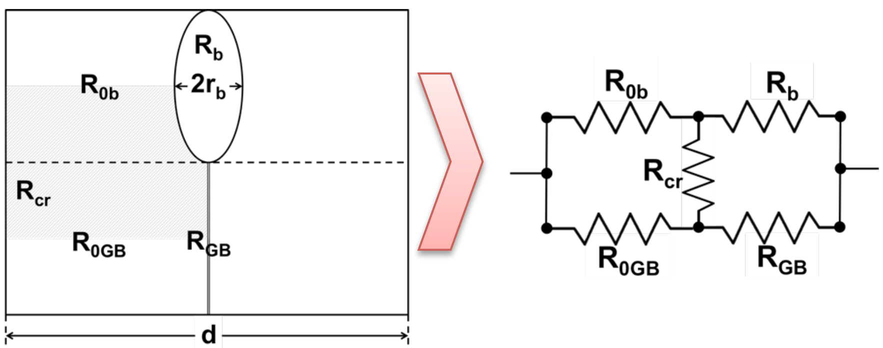
- They can be accounted for by changing the effective GB thermal resistance R' to be a function of the fractional coverage
- However, a mechanistic model is needed that accounts for this effect.

Millett and Tonks, J. Nucl. Mat., 412:3 (2011) 281–86

Millett, et al., J. Nucl. Mater. 439 (2013) 117–122.

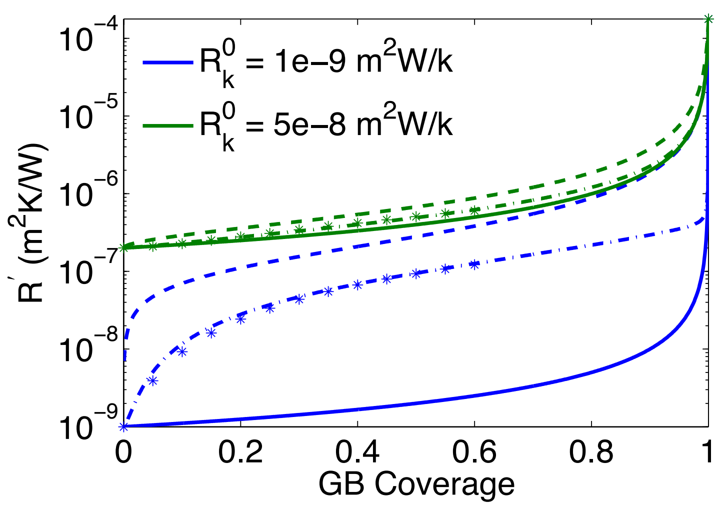
$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

A thermal resistor model is created to describe the impact of GB bubbles on the thermal conductivity



- System is represented by one bubble and one section of GB
- It is approximated by five thermal resistors
  - Three for bulk UO_2
  - One for bubble
  - One for GB

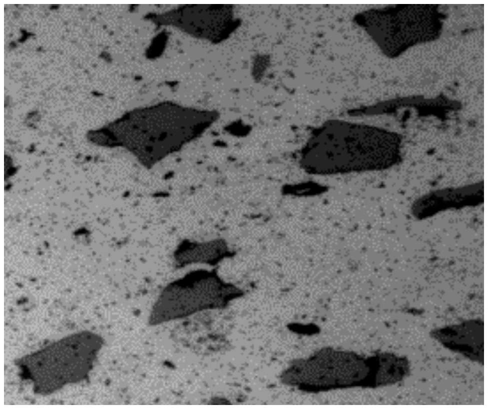
- $R_{cr} = 0$ gives a lower bound of the GB thermal resistance
- $R_{cr} = large$ gives an upper bound
- By fitting to 3D MARMOT simulation results we obtain:
  - $R_{cr} = 5\times10^{-7}$ m²W/K

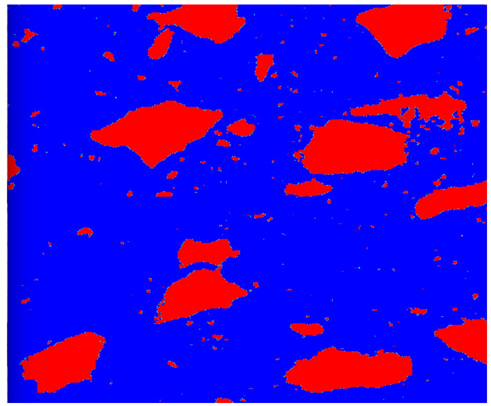


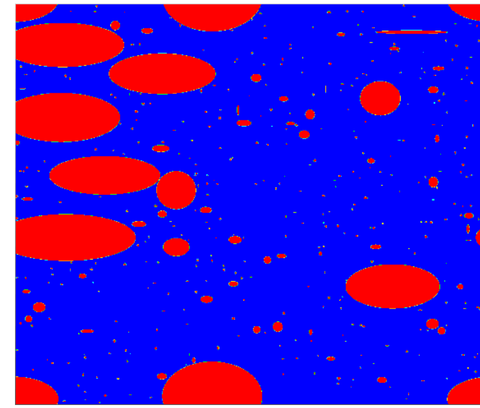
Tonks et al., J. Nucl. Mat., 469 (2016) 89–98

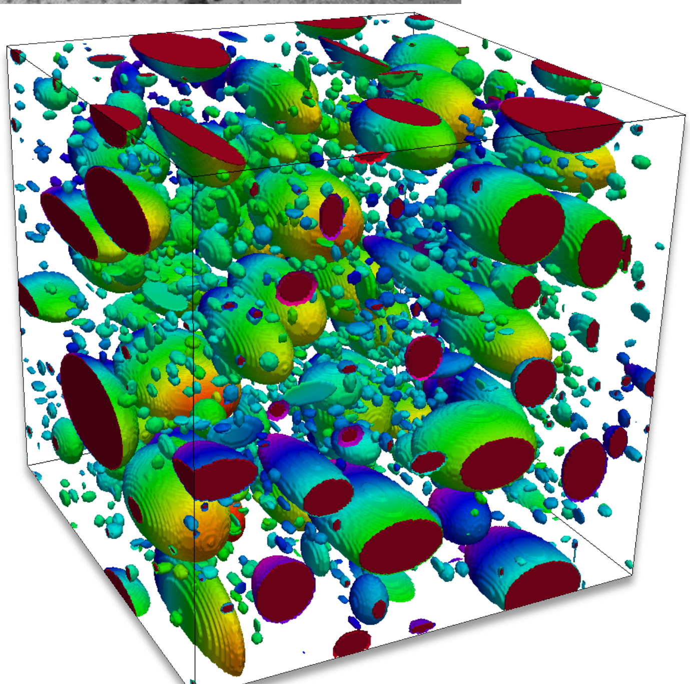
$$k = \frac{\kappa_{GB}\kappa_{p}\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

MARMOT simulations are currently being used to inform the development of the precipitate multiplier







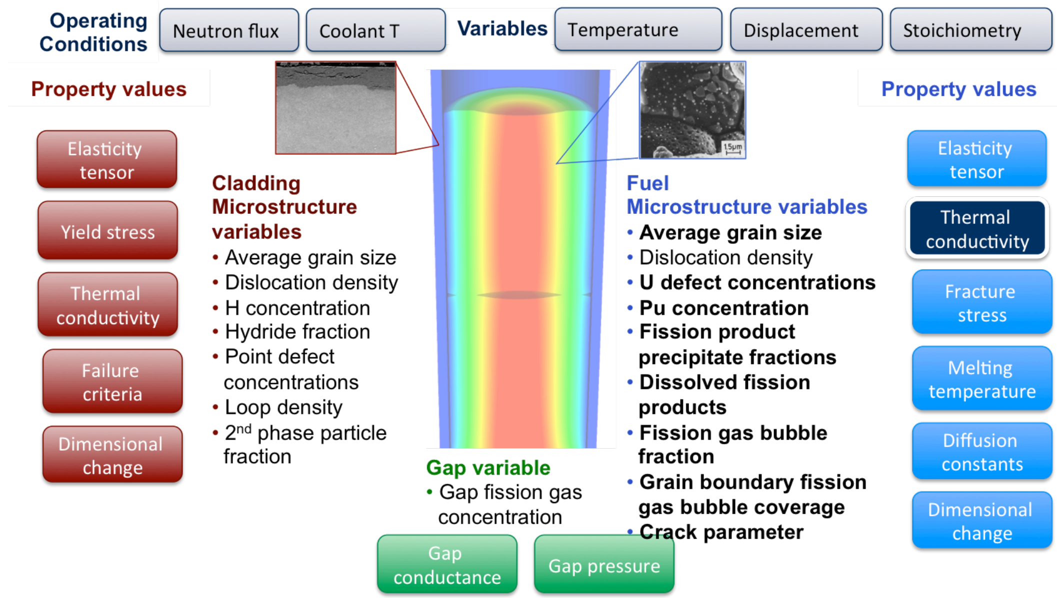


- Both generated microstructures and microstructures reconstructed from data are being used to inform the model
- The final value of κ_{pr} for metallic precipitates will be > 1.0 and will be a function of the precipitate concentrations and their average size

$$k = \frac{\kappa_{GB}\kappa_p\kappa_{pr}}{A + BT + CT^2 + C_v c_v + C_i c_i + C_g c_g}$$

Each term in the expression must be coupled to a corresponding state variable

- Our 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



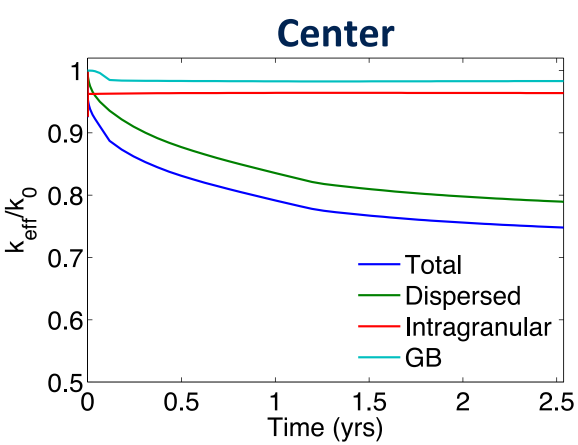
- Currently **only the portion of the model related to fission gas** has been implemented in the BISON fuel performance code because the microstructure model is in place.

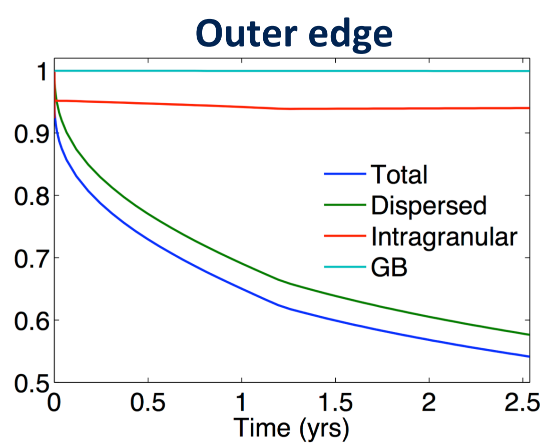
$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_g c_g}$$

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

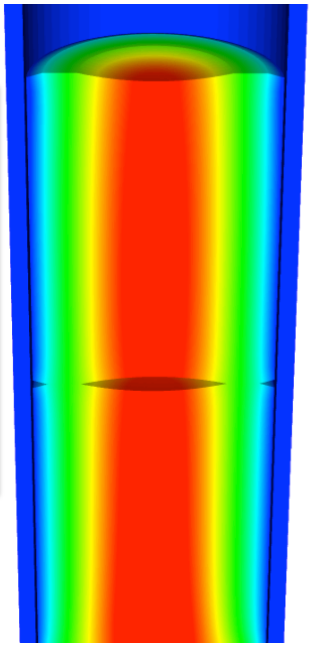
The effect of dispersed fission gas is much higher than the intragranular bubbles and GB bubble effects

- To investigate the relative importance of the three distributions of fission gas, we outputted the fission gas variables from a BISON simulation from a ten pellet rodlet at two locations:
  - Pellet center
  - Outer edge



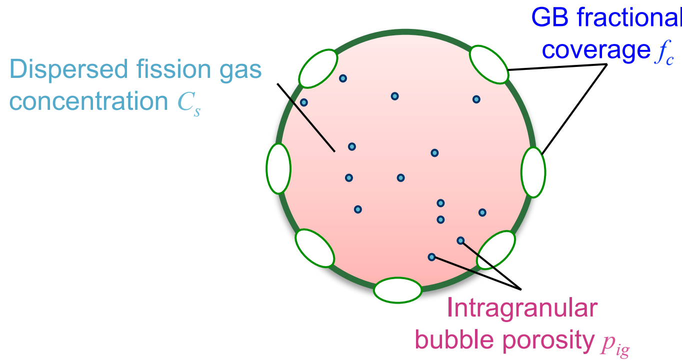


Outputs:
- Grain size
- Dispersed fission gas c_g
- Intragranular porosity p_{ig}
- Fractional coverage f_c
- Temperature T



We have implemented our fission gas thermal conductivity model in the BISON code

Fission gas behavior

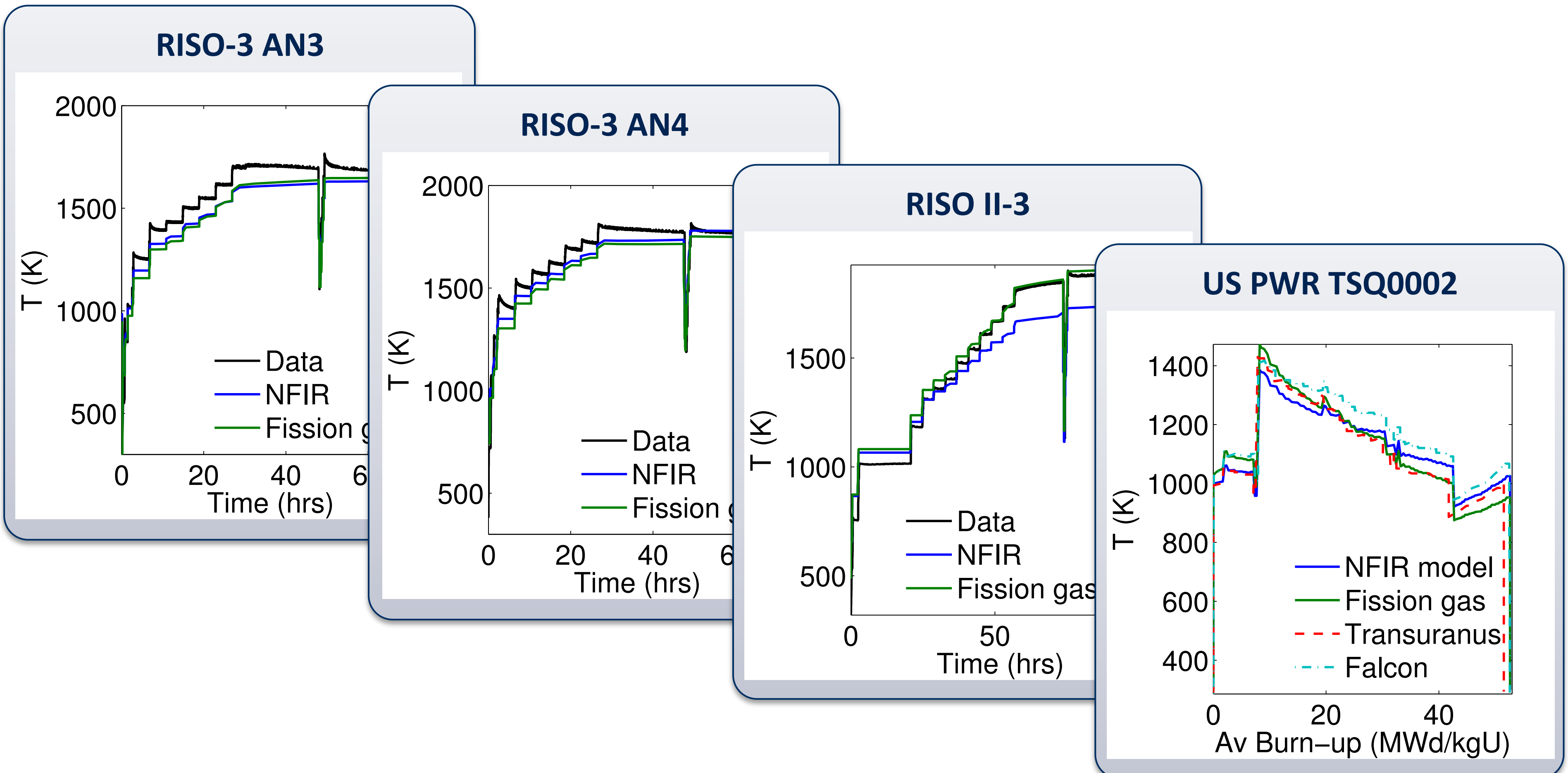


$$k = \frac{\kappa_{GB}\kappa_p}{A + BT + CT^2 + C_g c_g}$$

- To begin evaluating the model, we compare against data and results from other codes
  - Since the model is missing other defects besides fission gas, it should under-predict the temperature.

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

The model under-predicts the temperature in most cases, but not all



Summary

- At high burnup but mid to low temperature the fuel restructures to have a small grain size and large bubbles
  - The structure is called High Burnup Structures (HBS)
  - HBS may actually improve many fuel properties
- The fuel thermal conductivity decreases with burnup
- The NFIR model is a fairly accurate emprical model of the fuel thermal conductivity
- We 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.