Nuclear Fuel Performance

NE-533

Spring 2022

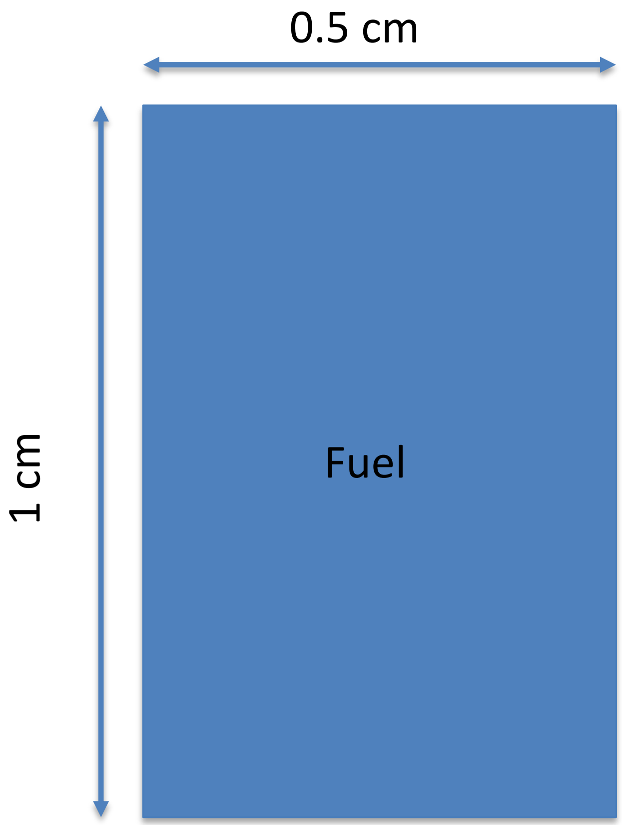
Last Time

- There are a variety of limiting phenomena in LWR fuel systems that provide the boundaries of operation and lifetime
- These limits include phenomena in the fuel, gap, cladding, corrosion, and assembly levels
- Primary system water chemistry affects fuel performance through the deposition of corrosion products on fuel pin surfaces
- Beginning of mechanistic modeling

MOOSE Project Part 3

- 1-D problem, set up in 2D-RZ
- Given a uniform heat generation rate of LHR = 175 W/cm, what are the stresses due to thermal expansion in the fuel?
- Will need tensor mechanics and heat conduction

- 1) Assume a constant fuel surface temperature and constant thermal conductivity
- 2) Assume a constant fuel surface temperature but temperature dependent thermal conductivity



Microstructure-based fuel performance modeling

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

Operating Conditions: Neutron flux | Coolant T&P

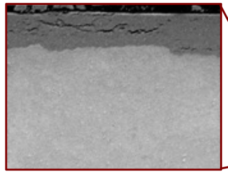
Variables: Temperature | Displacement | Stoichiometry

Cladding Property values

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

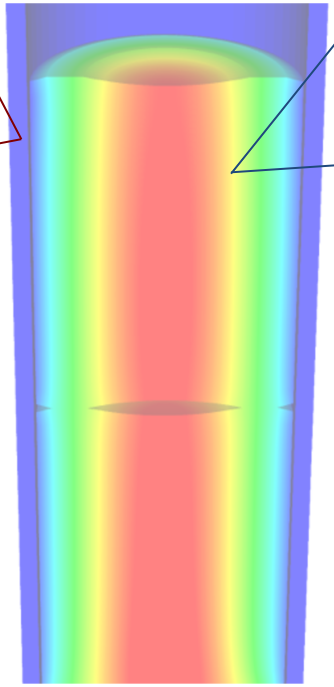
Microstructure evolution models

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



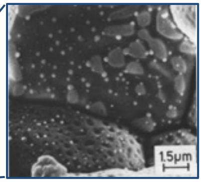
Cladding Microstructure variables

- 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

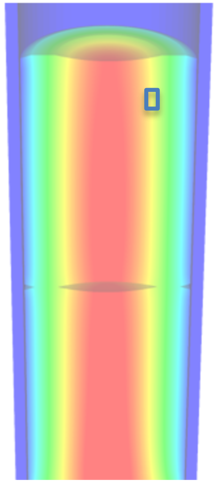
- Grain growth
- Plasticity
- Defect prod. & transport
- Fission gas behavior
- Densification
- Fracture

Fuel Property values

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

Example: fission gas behavior in the fuel

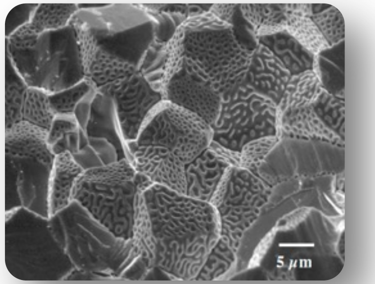
- Take into account a finite set of variables to describe the state of the material
- Utilize a mechanistic model of fission gas behavior to predict the evolution of the microstructure
- Utilize this updated microstructure to inform a number of structure/property relationships



Variables

Temperature | Displacement | Stoichiometry

Model of fission gas behavior

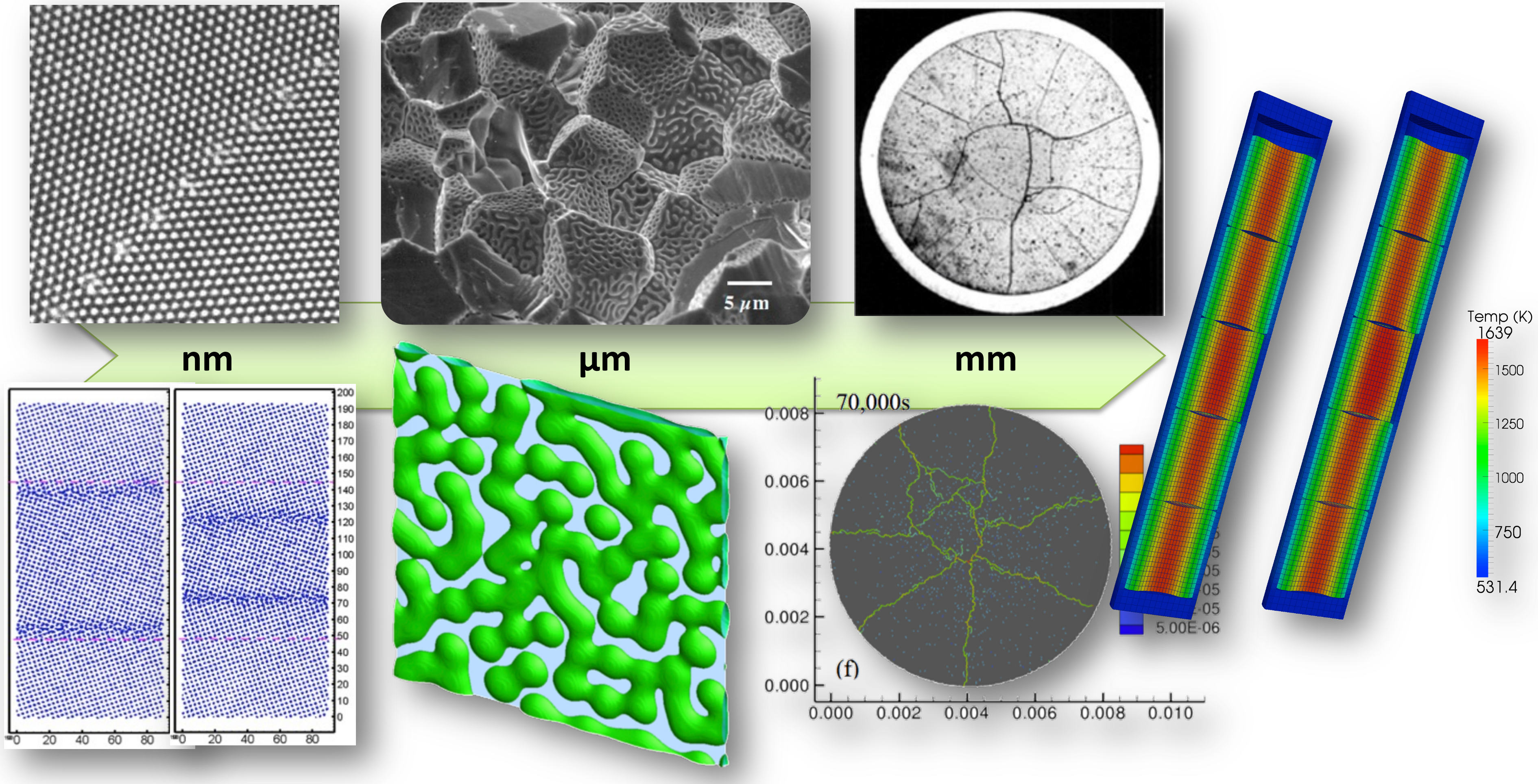


- Dissolved fission products
- Fission gas bubble fraction
- Grain boundary fission gas bubble coverage
- Gap fission gas concentration

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

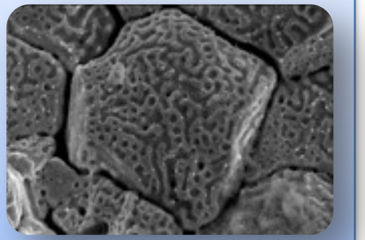
- Can provide a structure/property relationship to replace the existing burnup dependent model
- For example, thermal conductivity, taking into account microstructural features and their evolution

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

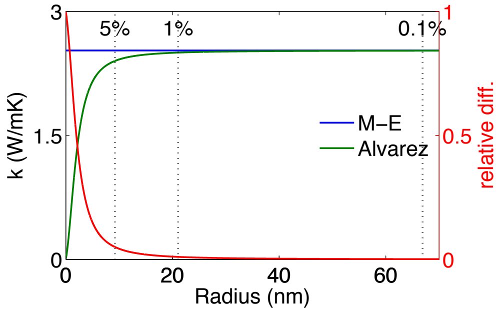
Parametrizing the mechanistic model

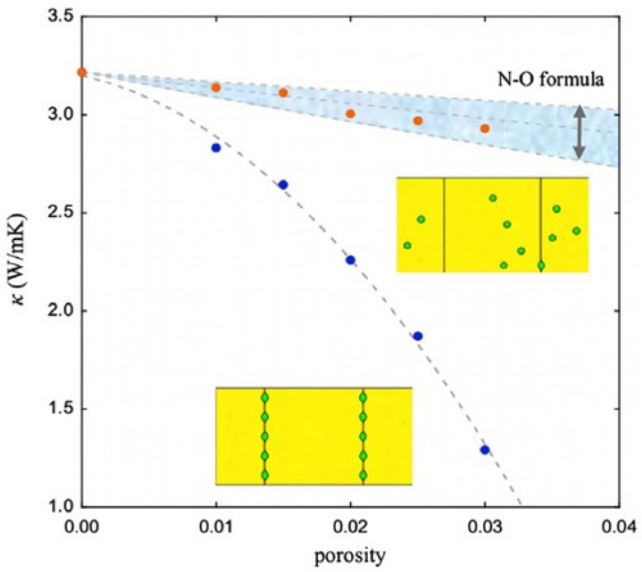
- We employ multiscale modeling and simulation to determine the various parameters for the model
- MD simulations conducted at LANL have been used to determine the coefficients for various point defects
- MD simulations have shown that phonon scattering must be accounted for to accurately represent small bubbles
- Mesoscale simulations have shown that GrB bubbles have a larger impact on the thermal conductivity

Defect	a_i	Defect	a_i
O interstital	12.63	Xe atom	33.9
O vacancy	21.74	La atom	3.97
U interstitial	29.98	Zr atom	2.23
U vacancy	23.78	Pu atom	0.08

Maxwell-Eucken (no phonon scattering)

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

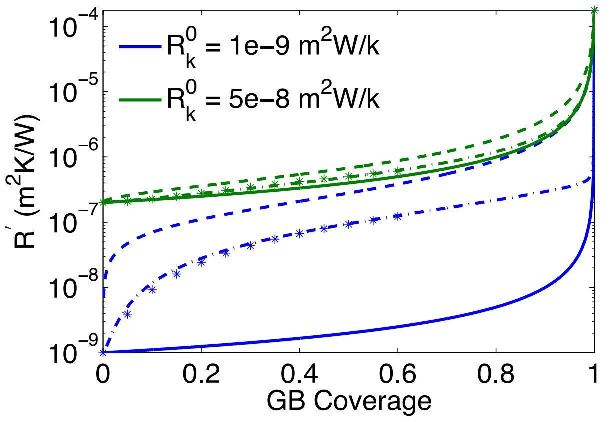


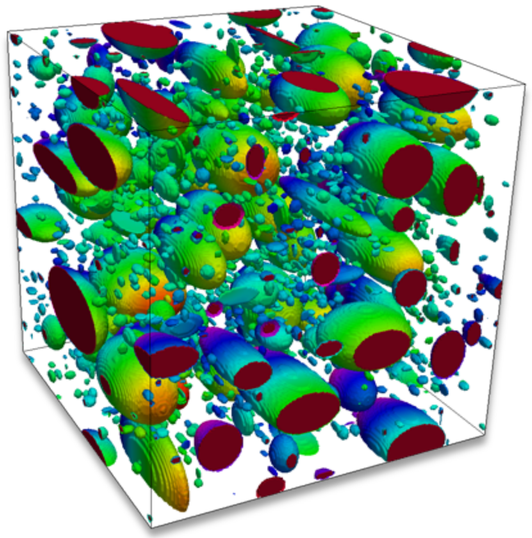


Parametrizing the mechanistic model

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

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





Parametrizing the mechanistic model

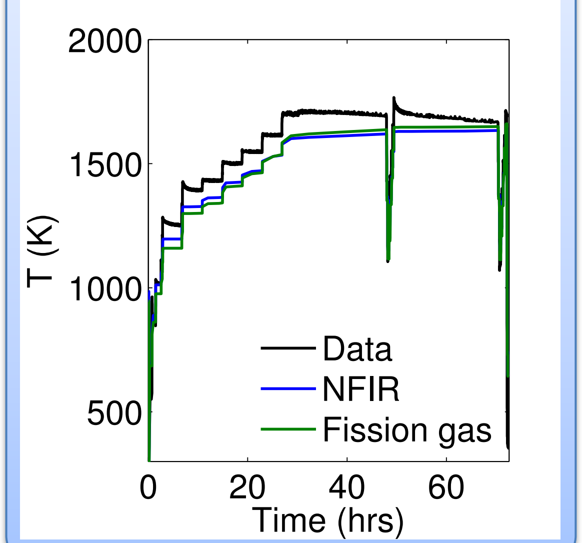
- Each term in the expression must be coupled to a corresponding state variable
- The full model calculates the thermal conductivity as a function of:
  - Temperature
  - Point defect concentrations
  - Intragranular bubble density and average radius
  - Fractional coverage of bubbles on GBs and average radius
  - Precipitate volume fractions and average sizes
- Currently effects of precipitate fission products and individual point defects are neglected in the model, as they are not tracked or predicted in BISON

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

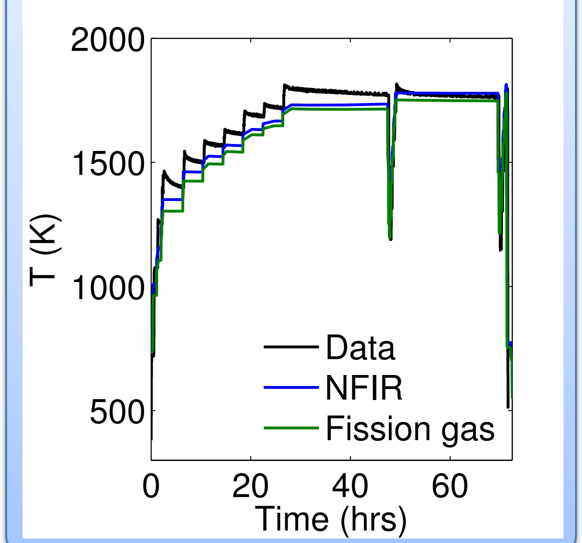
Comparing with experiments…

- 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

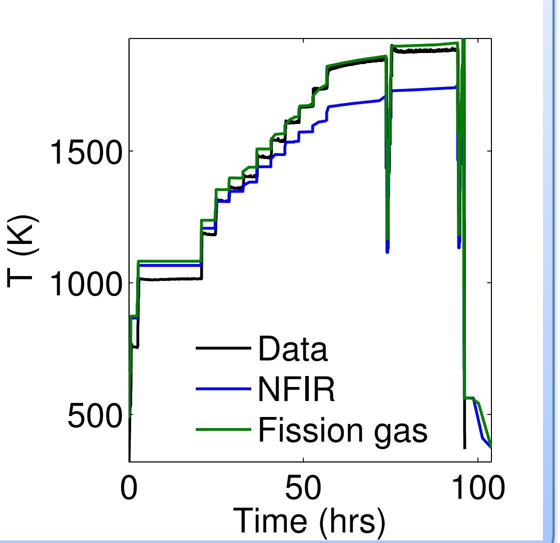
RISO-3 AN3



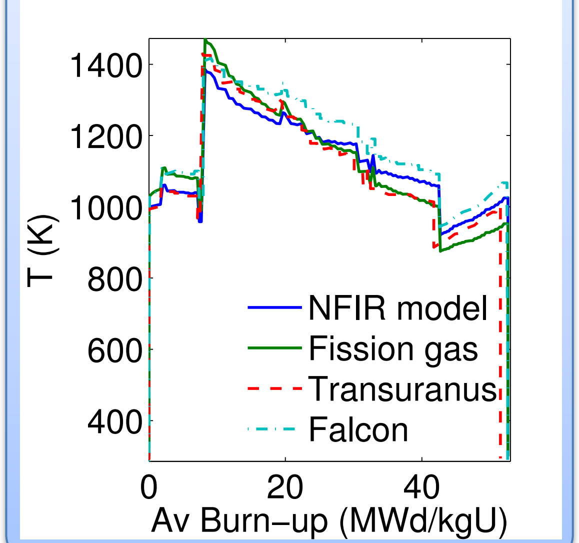
RISO-3 AN4



RISO II-3

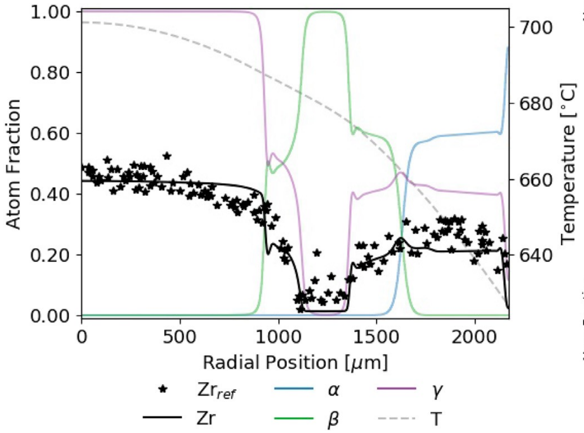


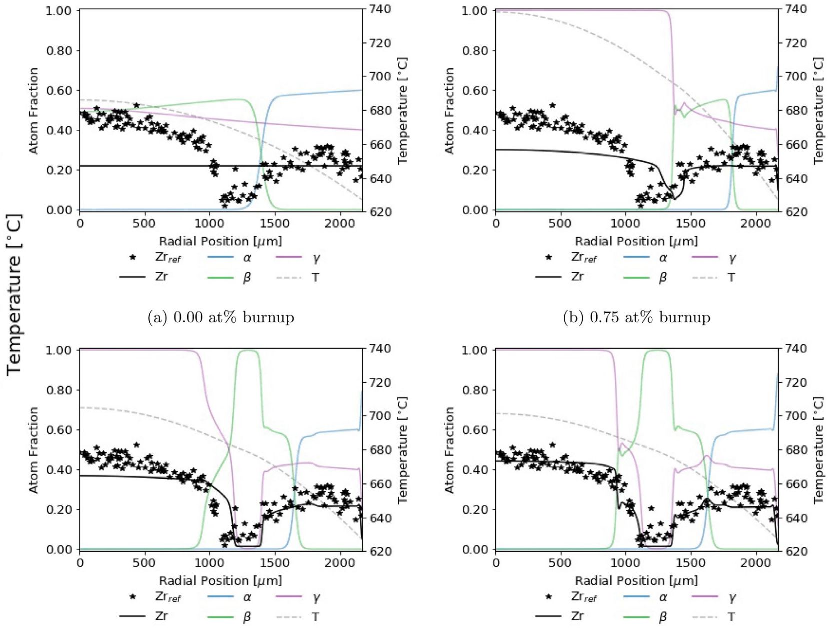
US PWR TSQ0002



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





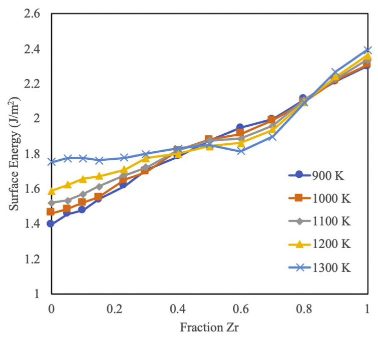
(a) 0.00 at% burnup

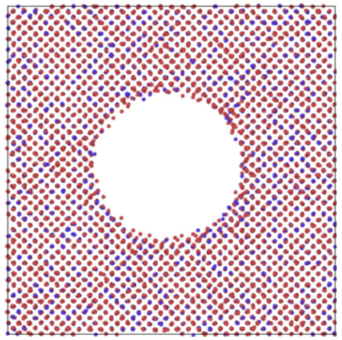
(b) 0.75 at% burnup

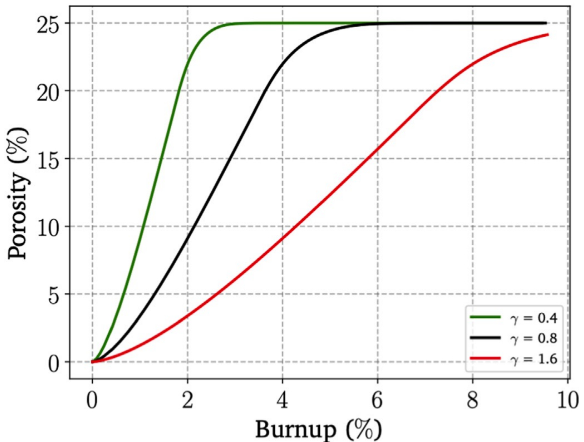
(c) 1.50 at% burnup

(d) 3.60 at% burnup

- Calculation of surface tension based on molecular dynamics, which is used in the BISON gaseous metallic fuel swelling model

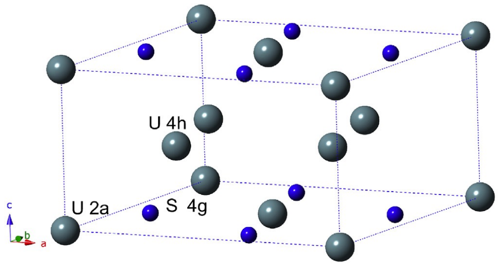


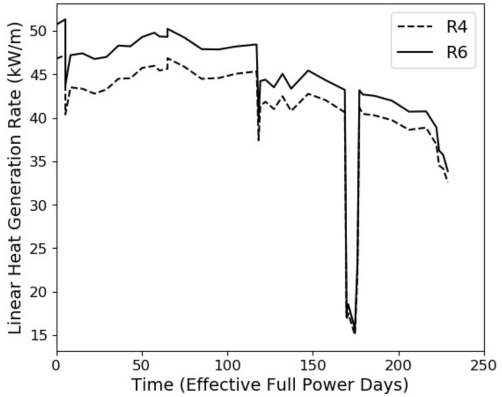


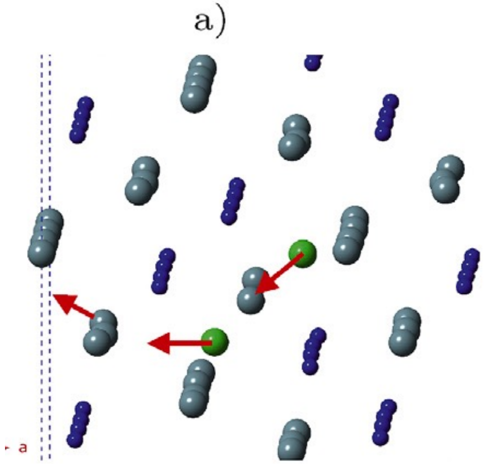


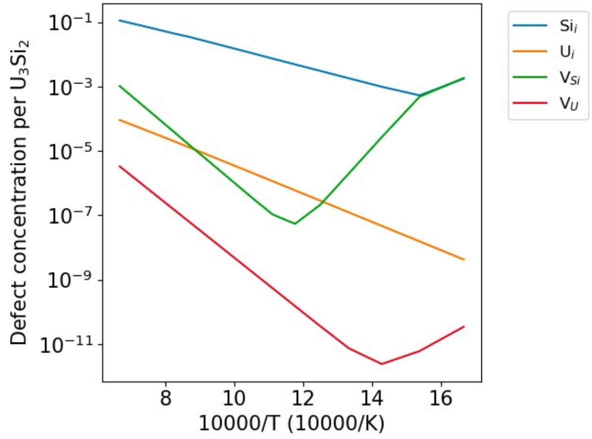
U3Si2 Fuel Performance Modeling

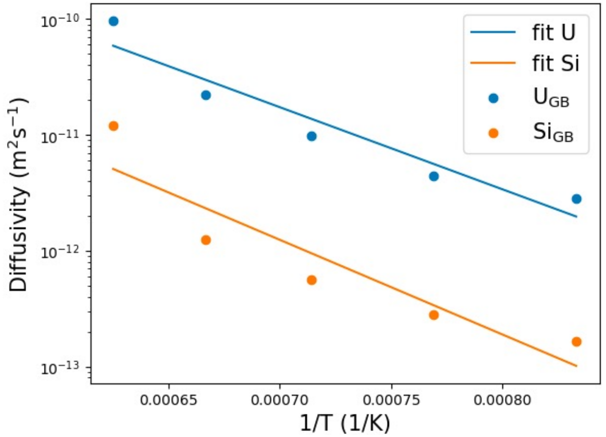
- Point Defect Diffusion, Fission gas swelling, thermal and irradiation creep

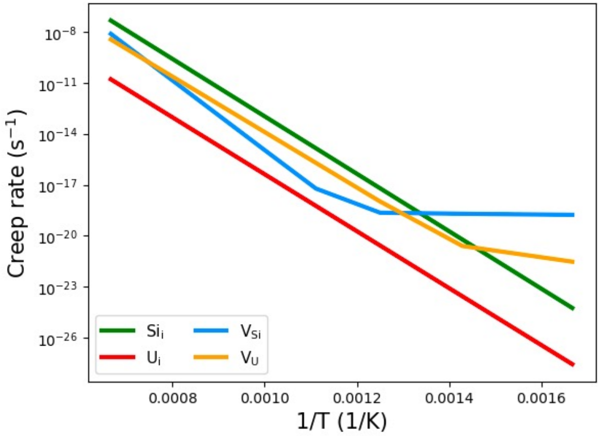


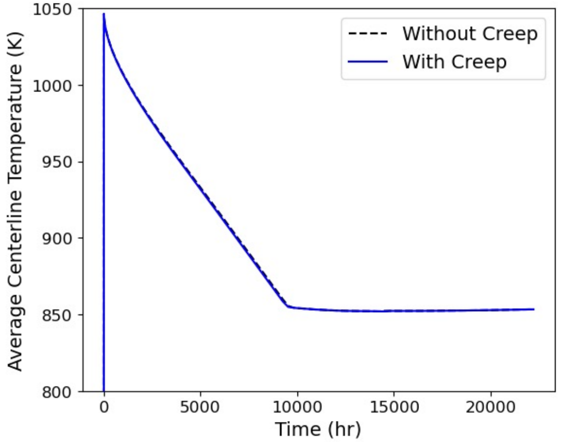












	BISON				Experiment	
	R4		R6		R4	R6
	Stoichiometric	Si-Rich	Stoichiometric	Si-Rich		
Fuel elongation (mm)	0.099 to 0.163	0.088 to 0.154	0.135 to 0.223	0.112 to 0.225	0.0	0.0
Fission gas release (/)	0.0 to 0.007	0.0 to 0.002	0.0 to 0.014	0.0 to 0.011	0.0006	0.0006

TRISO Particle Fuel Performance Modeling

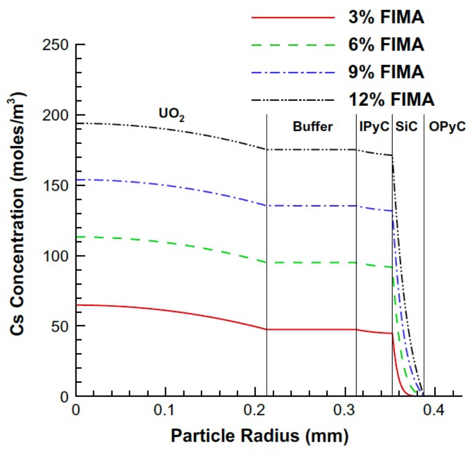
- TRISO particle modeling is still under development from a mechanistic model standpoint
- Imperfect data for many critical fission products through different layers
- Data typically for UO2 only, not for UC or UCO fuel kernels

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

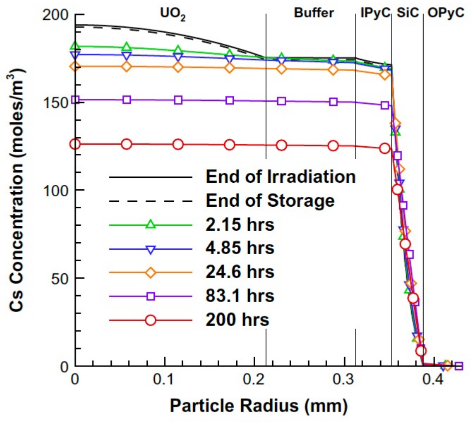
$$\mathbf{J} = -D \nabla C,$$

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

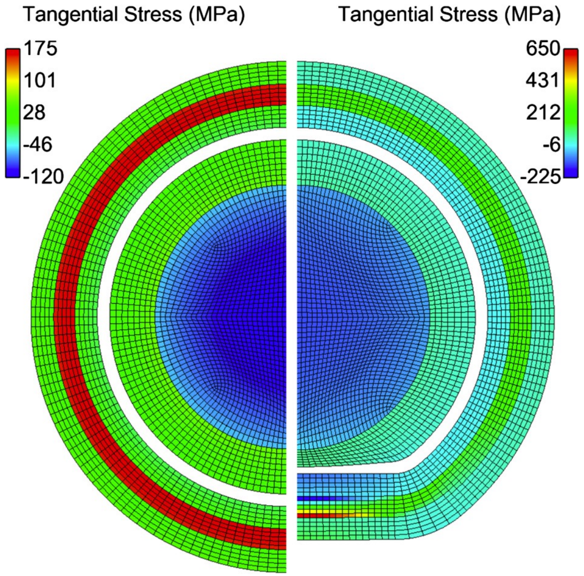
Material	D_1 (m^2/s)	Q_1 (kJ/mol)	D_2 (m^2/s)	Q_2 (kJ/mol)
UO_2	5.6×10^{-8}	209	5.2×10^{-4}	362
Buffer	1×10^{-12}	0	0	0
PyC	6.3×10^{-8}	222	0	0
SiC	$5.5 \times 10^{-14}(e^{\frac{\Gamma}{1.5}})$	125	1.6×10^{-2}	514



(a)

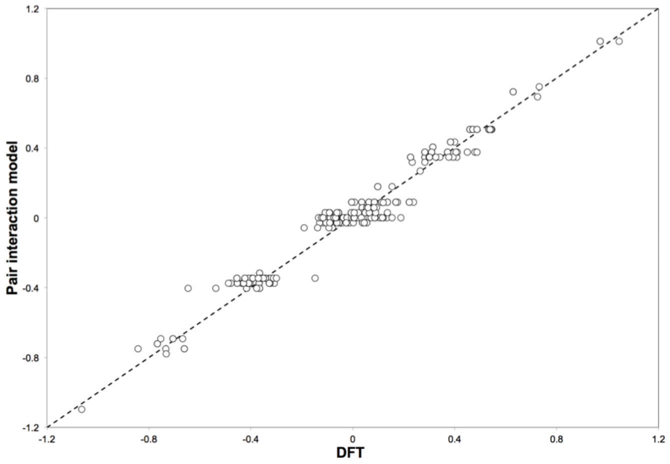


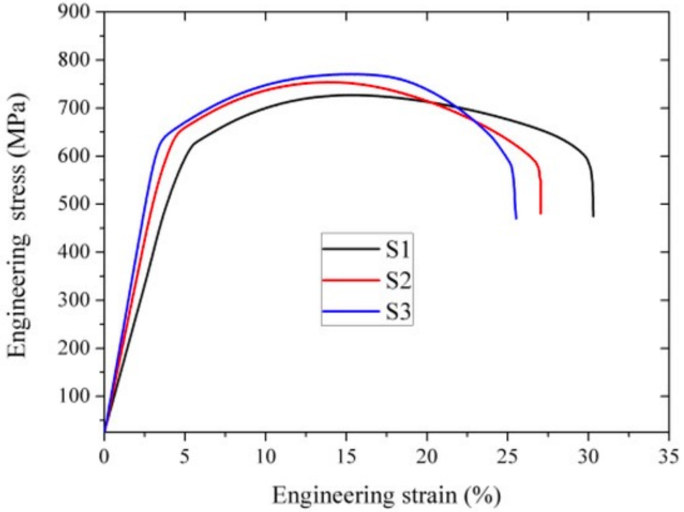
(b)

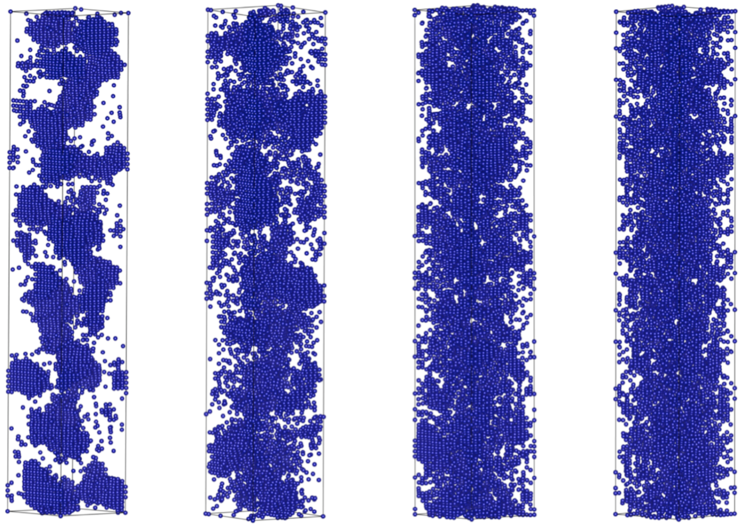


FeCrAl

- 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







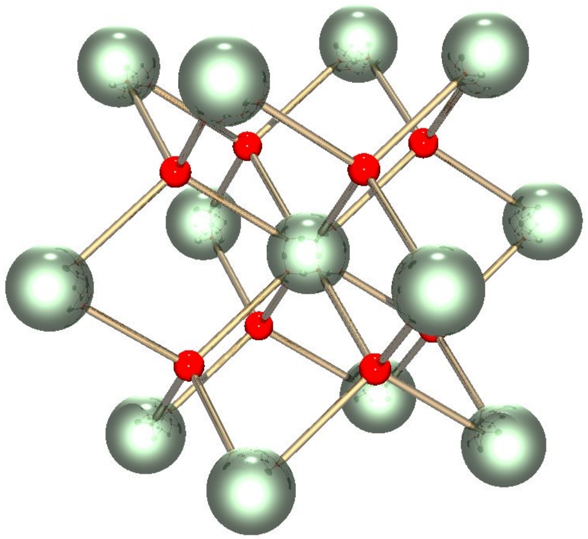
773 K 1023 K 1273 K 1523 K

Summary

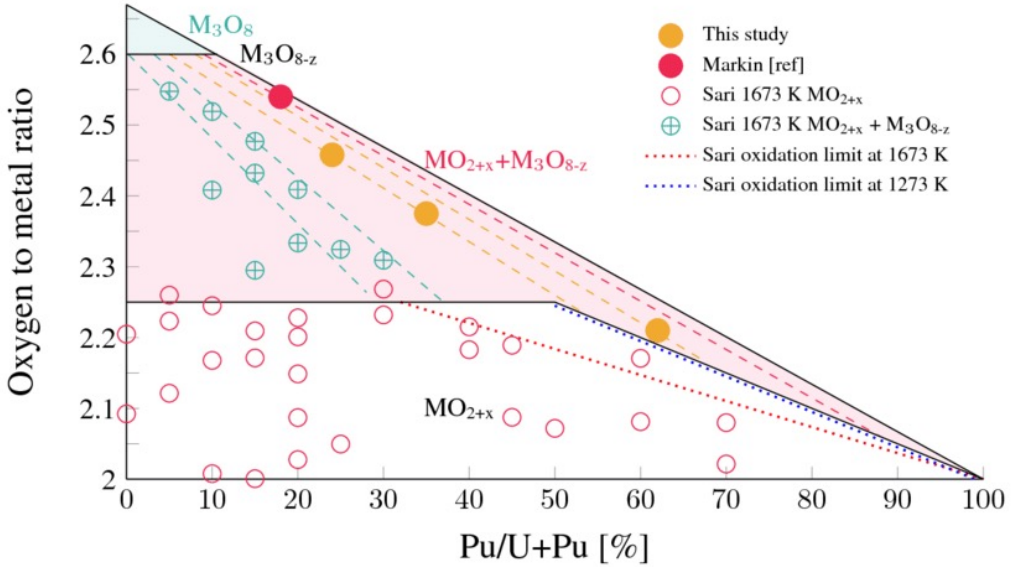
- Researchers are working to develop materials models for the fuel and cladding that are mechanistic rather than empirical and that are based on the evolution of the microstructure rather than the burnup.

MIXED OXIDE (MOX) FUELS

Why MOX?



- The first fast breeder reactors built in the 1950s used metallic fuel (plutonium and uranium), as metals offer the highest heavy metal density and therefore the highest breeding ratio
- Because of dimensional instability due to swelling and growth, metal fuels could hardly achieve high burnup
- By the 1960s, mixed uranium and plutonium oxide (U,Pu)O2 was known to be highly radiation tolerant and began to be considered as a reference fuel for fast reactors



Mixed Oxides

- UO2 can be combined with PuO2 for a mixed oxide (MOX) fuel for use in fast reactors
- Allows to burn excess weapons grade plutonium
- About 30 reactors in Europe currently utilize a partial MOX core
- Similar behavior to UO2, but different neutronics, fission gas release, thermal conductivity, etc.
- Less common is inclusion of minor actinides in MOX to burn waste

Table 1 Main characteristics of standard fuel pins irradiated in the prototype and commercial fast reactors ($p > 200$ MWth)

	BN350	*Phénix*	*PFR*	*BN600*	*FFTF*[a]	*Super-Phénix*	*MONJU*
First criticality	1972	1973	1974	1979	1980	1985	1994
Thermal power (MWth)	750	563	600	1470	400	3000	714
Electric power (MWe)	350[b]	250	250	600	–	1200	280
Type of fuel	UO_2	$(U,Pu)O_2$	$(U,Pu)O_2$	UO_2	$(U,Pu)O_2$	$(U,Pu)O_2$	$(U,Pu)O_2$
No. of subassemblies (inner/outer core)	109/117	55/48	28/44	209/160	28/45	193/171	108/90
No. of pins per assembly	127	217	325	127	217	271	169
Type of spacer	Wire	Wire	Grids	Wire	Wire	Wire	Wire
Length of pin (m)	1.8	1.793	2.25	2.445	2.38	2.7	2.813
Height of fissile column (m)	1.06	0.85	0.914	1.0	0.914	1.0	0.93
Lower fertile column length (m)	0.4	0.3	0.45	0.4	–	0.3	0.35
Upper fertile column length (m)	0.57	0.31	0.45	0.4	–	0.3	0.3
Clad outer diameter (mm)	6.9	6.55	5.8	6.9	5.84	8.5	6.5
Clad thickness (mm)	0.4	0.45	0.38	0.4	0.38	0.565	0.47
Helical wire diameter (mm)		1.15			1.42	1.2	1.32
Pellet diameter (mm)		5.42				7.14	5.4
Fuel clad diametral gap (mm)		0.23			0.14	0.23	0.16
Central hole diameter (mm)	0	0	1.5	0		2.0	0
Fissile atoms/(U + Pu) (%) (inner core/outer core)	17/26	18/23	22/28	17/26	20/25	15/22	16/21
Fuel density (% TD)	95	95.5	97	95	91	95.5	85
Smeared density (%)	75	88	78	77	86	83	80
Plenum volume (cm^3)	8	13	14	21	19	43	28
Maximum linear power ($W\ cm^{-1}$)	400	450	420	472	413	470	360
Peak cladding temperature (°C)	570	650	670	700	660	620	675
Maximum neutron flux ($10^{15}\ n\ cm^2\ s^{-1}$)	7	7.1	7.6	7.7	7	6	6.0
Maximum burnup (at.%)	9.0	16.9	23.5	11.8	24.5	Not relevant	Not relevant
($GWd\ t^{-1}$)	65	144	200				
Maximum dose (dpa)	60	156	155	90		–	–

MOX Designs

- The fuel pin is a long cylinder (2–3m long, 5–10mm diameter), clad in a steel tube (0.4–0.6mm thick) closed in both ends by welded plugs, preventing direct contact between the radioactive material and the sodium coolant
- The oxide fissile column (~1 m long) consists of a stack of conventionally pressed and sintered pellets with an outer diameter slightly smaller than the inner diameter of the clad, providing a gap ~100 micron gap
- Both full pellets and annular pellets have been used
- A He gap with a pressure ~1 atm is used
- Fuel pins of fast reactors are designed to operate at a high linear heat generation rate: between 400 and 500 W/cm, about twice higher than standard linear power in light water reactors (LWRs)

MOX Fuel

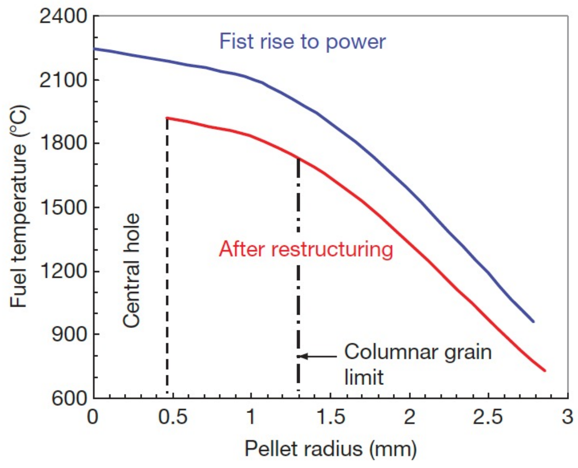
- As the fast reactor fuel pin diameters are generally smaller than classical rod diameters of LWRs, the power density and heat fluxes are much higher in fast reactors than in LWRs
- For example, in a Phenix fuel pin at 450W/cm, the power density in the pellet reaches almost 2000 W/cc
- Sodium enters the bottom part of the core at about 400 C and the average coolant temperature above the core is typically about 550 C
- The neutron flux is very intense (~7×10^{15} n/cm^2/s in the core center) and the assembly materials suffer high damage, more than 100 dpa, at high burnup
- Qualifying metallic materials able to withstand such high damage while keeping their shape and mechanical properties was/is one of the key challenges for fast reactors
- In order to reduce fuel cycle costs, the main objective of oxide fuels R&D for fast reactors has been to reach high burnup, typically around 150 GWd/ton, about twice the burnup achieved in LWRs

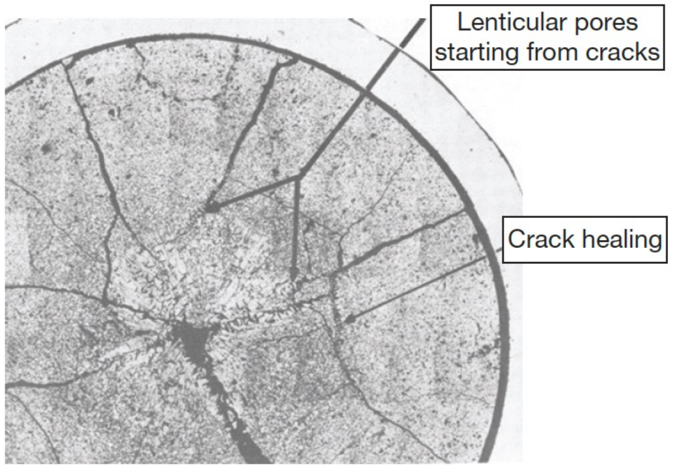
MOX Fuel

- The main requirements and design criteria are the following:
  - Guaranteeing the absence of fuel melting, both in nominal conditions and during off-normal events
  - Keeping cladding integrity and fuel pin tightness
  - Cooling of the fuel pin bundle must be ensured up to high burnup in all operating conditions
  - Loading and unloading of subassemblies have to be guaranteed, which induces a limitation on the deformation of the hexagonal wrapper tubes

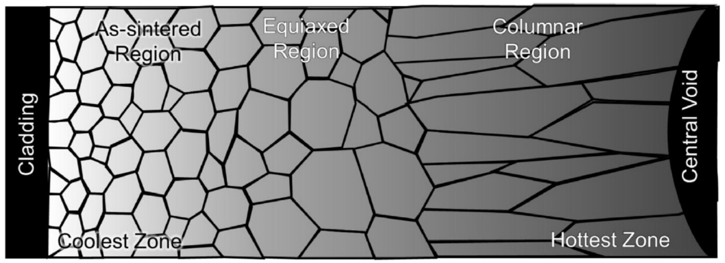
MOX Fuels

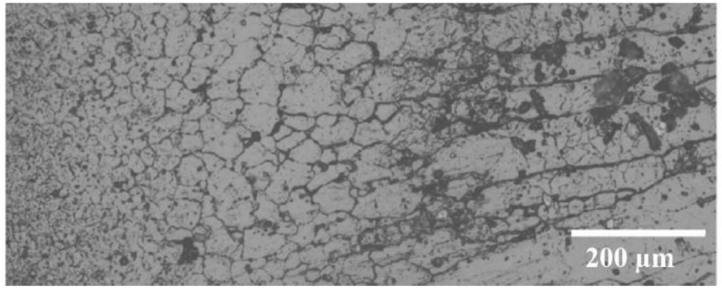
- Most phenomena occurring inside oxide fuel pellets are thermally activated, and a good knowledge of the thermal field inside the fuel stack is key
- Centerline temperatures can reach above 2000C, significantly higher than thermal LWRs
- Thermal conductivity is low and degrades with irradiation
- Due to the steep thermal gradient, thermal stress cracks form
- Restructuring takes place due to the high temperatures, leading to distinct regions in the fuel

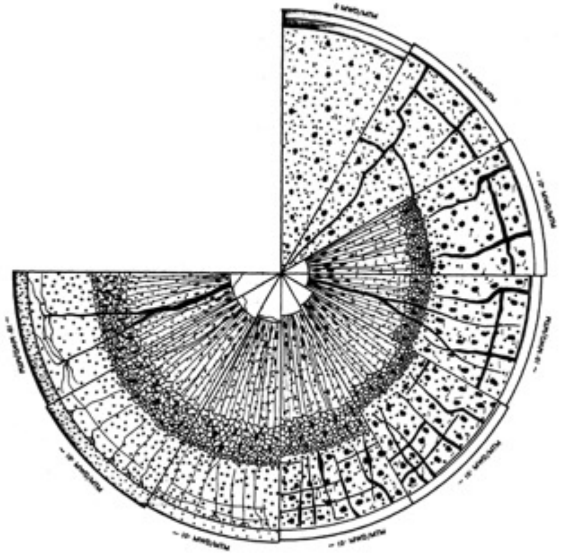




MOX Restructuring



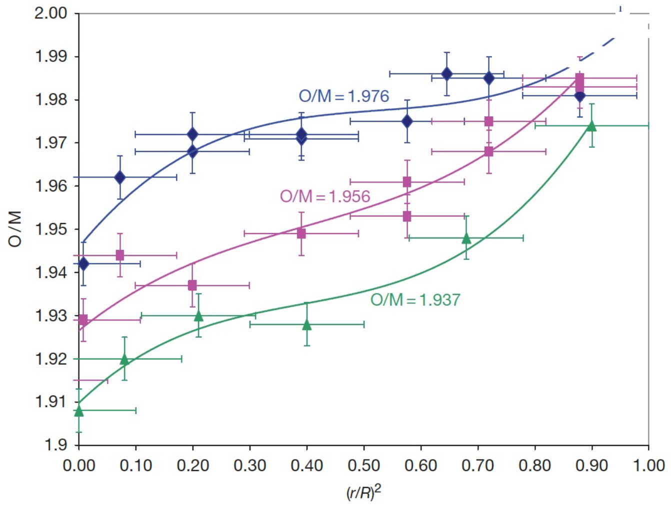


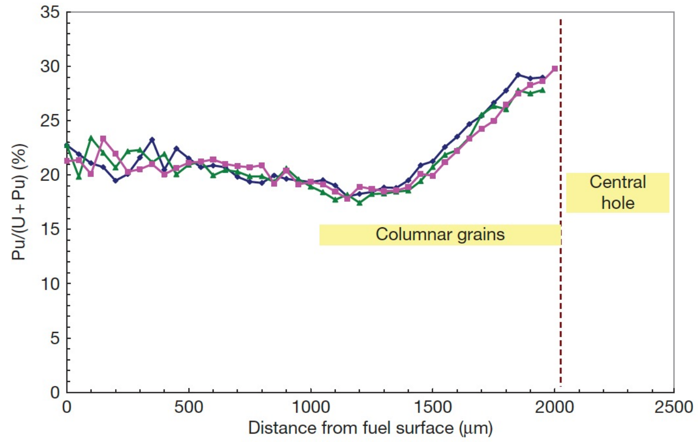


- Pu bearing fast reactor oxide fuels display four defining regions of a restructured pellet:
  - the central void, the columnar grain growth region, the equiaxed grain growth region, and the as-sintered region
- The higher temperatures and heating rates form coarse, elongated grains that grow radially toward the outer rim of the fuel
- The equiaxed region consists of grains that have undergone significant growth when compared to the un-irradiated samples
- The central void forms from the accumulation of voids and pores present in the fuel along a thermal gradient

Constituent Redistribution

- The as-fabricated oxide pellets to be used as fuel in fast reactors are always hypostoichiometric with an initial O/M typically in the range 1.93–2.00
- Oxygen is redistributed radially, migrating down the thermal gradient, thus bringing the composition close to stoichiometry near the periphery, whereas the O/M ratio becomes very low in the hottest area
- Irradiated oxide pellets generally exhibit a plutonium enrichment in the central area near the central hole, and a slight plutonium depletion in a ring located near the periphery of columnar grains





Gap Closure

- Although the thermal expansion coefficient is lower in oxide fuel than in austenitic stainless steel cladding the temperatures in the fuel pellets are much higher than in the cladding and induce a higher thermal expansion in the fuel pellet
- At high linear powers gap closure is completed after a burnup of about 1% or even less
- Fuel pellets are broken into several fragments at the end of first rise to power, resulting in a small average displacement of matter toward the cladding
- The force exerted on the fuel column by the spring located in the upper part of causes an axial compression creep
- The main cause of gap closure is probably the gaseous swelling of the fuel

Fission Products

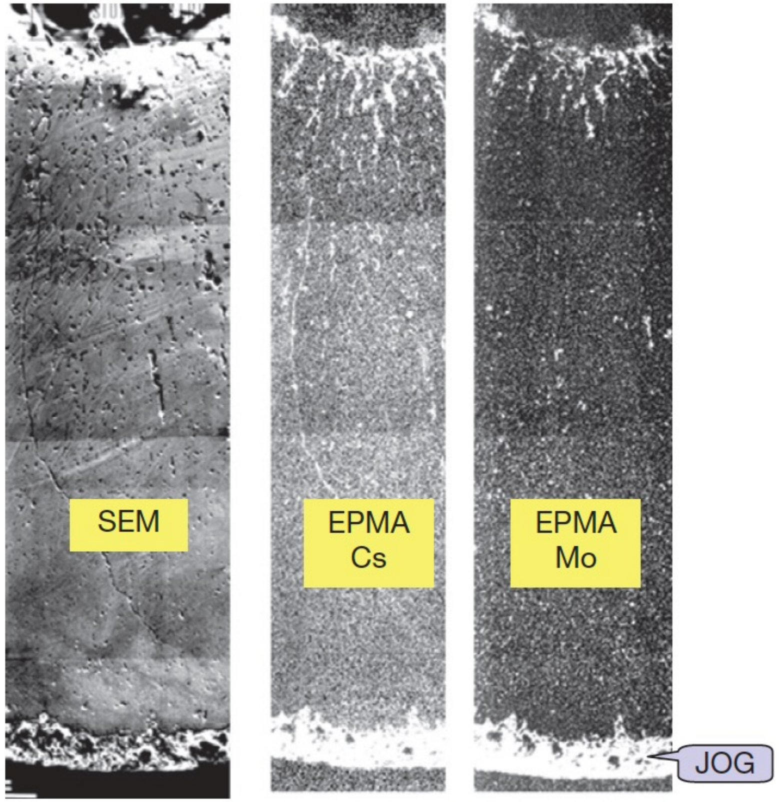
- The main objective of oxide fuels in fast reactors is to achieve very high burnup; 15 at.% or even more is typically considered as a reference target
- This means that at the end of irradiation, 15% of the initial actinide atoms (U and Pu) have disappeared and 30% new atoms are present in the fuel
- All physical and chemical properties of oxide fuel will continuously evolve during its lifetime in the reactor; in particular, fission products will induce a decrease of thermal conductivity as well as a decrease of melting point, thus reducing the margin to fuel melting
- Most phenomena occurring in the fuel pins will be a direct consequence of these fission products
- This large amount of fission products is one of the specificities of fast oxide fuel
- The fission products effects depend upon the chemical state of the fission product, which is influenced by the oxygen potential of the fuel

Fission Products

- Fission products lie in the main five families:
  - solid solution, oxide precipitates, metallic precipitates, volatile gases, noble gases
- On the metallographs of fuel irradiated at high burnup, white inclusions are systematically observed
- The higher the burnup and the temperature, the larger these precipitates
- In most cases, EPMA on these precipitates shows essentially five elements: Mo, Ru, Tc, Rh, and Pd.
- They are the five ‘noble metal’ fission products with the highest yield

Joint Oxide Gain (JOG)

- Even after gap closure, there remains a residual gap a couple of micrometers wide due to surface roughness
- At high burnup, radial micrographs show a reopening of the gap, however, this gap is no longer filled with gas, but with fission product compounds
- All the fission products migrating toward the cold region of the pellet accumulate first in the oxide fuel, and then escape the fuel and accumulate between the fuel and the cladding where they form a bonding layer
- At high burnup, this JOG reaches a diameter width of about 150 μm
- Serves as mechanical buffer; has low thermal cond.



FCCI

- FCCI appears as one of the potential life-limiting factors for high burnup fuel elements
- Fission products in the JOG play a predominant role on FCCI and on the resulting strong corrosion
- The $(U,Pu)O_2$ fuel itself does not directly react with the cladding, but it provides the oxygen needed for some of the reactions
- Volatile fission products, tellurium and cesium, are the corrosive species able to overcome the passivation of stainless steel and therefore to induce clad corrosion
- Several types of corrosion reactions and mechanisms are possible, occurring at different stages of irradiation, sometimes successively in the same pin, and resulting in different attack features
- Thus, a qualitative understanding of corrosion mechanisms has been achieved, but it is not yet possible to give a complete physical description of FCCI and to predict corrosion depths

Summary

- Oxide fuel for fast reactors has proved to be a mature, quite reliable, and very robust fuel concept
- SFRs with MOX operate at much higher power and temperature than LWRs
- Despite the low thermal conductivity, fuel loadings to the cladding remain low, and oxide fuel pins have demonstrated an ability to reach extremely high burnup (>15 at%)
- O/M ratio is one of the most significant factors in determining the nature of actinide redistribution, fission product precipitate chemistries, and FCCI formation