Nuclear Fuel Performance

NE-533

Spring 2024

Housekeeping

- Last lecture of the semester
- Exam next Tuesday, covers all material since exam 3
- MOOSE project final due on April 26
- Will be sending out a google form survey after exam 4 to get feedback on the class, completion of which will give a +5 bonus on exam 4, will be anonymous

MOOSE Project Part 2

- Everyone received grades + comments
- One mistake that almost everyone made, so i didn't discount for it
  - specifying the coolant temperature as the boundary condition on the surface of the fuel
  - need to have heat transport into the coolant to use the equation for coolant temperature change
  - peak cladding temperature is NOT at the top of the fuel column
  - peak coolant temperature IS at the top of the fuel column

```
    [Tco]
        type = ParsedFunction ## Tco = Tcool + delta(Tco-Tcool)
        expression = (1/1.2)*(350*50)/(4200*0.1)*(sin(1.2)+sin(1.2*(y/50-1)))+500+350*cos(1.2*(y/50-1))/(2*3.14159*0.5*2.65)
    []
```

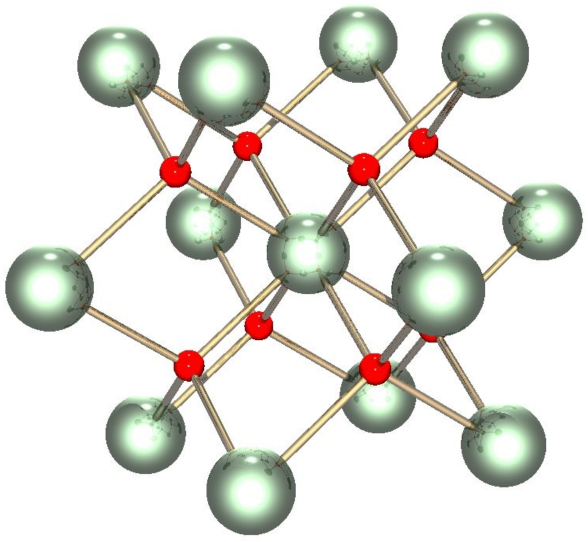
```
        [surface] # on the right
                type = ADFunctionDirichletBC
                variable = temperature
                boundary = 1
                function = Tco # Kelvin
        []
```

Last Time

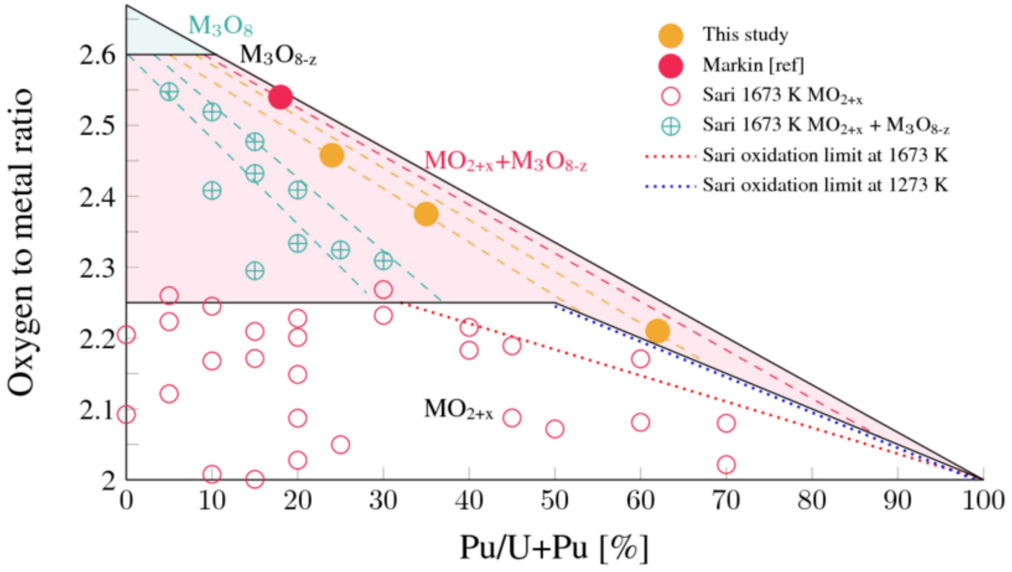
- Steam oxidation can lead to breakaway oxidation, leading to thick, cracked oxides and high H generation
- Advanced cladding materials and coatings are being explored to improve accident tolerance
- There are a variety of limiting phenomena in LWR fuel systems that provide the boundaries of operation 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, primary control through addition of hydrogen

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 couldn't achieve high burnup
- By the 1960s, mixed uranium and plutonium oxide (U,Pu)O2=MOX was known to be highly radiation tolerant and began to be considered as a reference fuel for fast reactors



Mixed Oxides

- MOX fuel 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
- Can also serve as a breeder

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

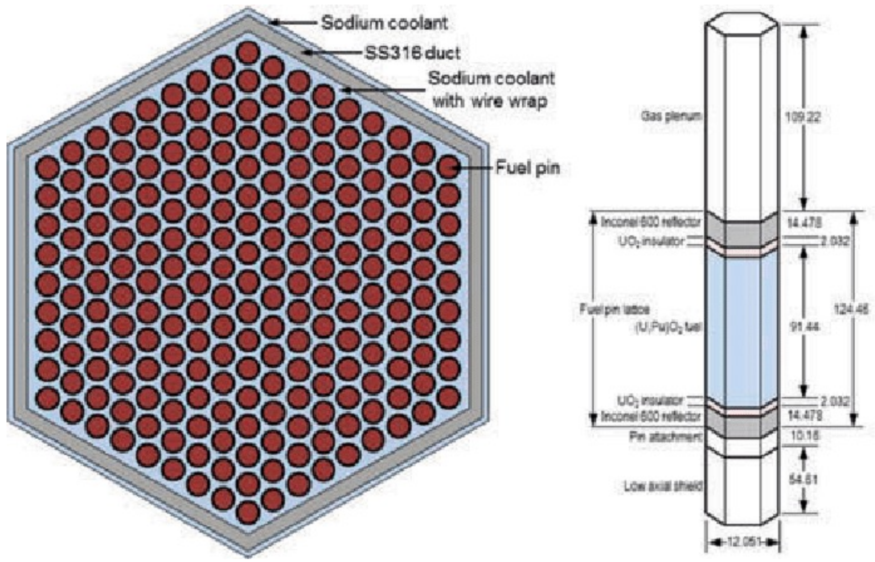
- The fuel pin is a long cylinder (2–3m long, 5–10mm diameter), clad in a steel tube (0.4–0.6mm thick) closed in both ends by welded plugs, preventing direct contact between the radioactive material and the sodium coolant
- The oxide fissile column (~1 m long) consists of a stack of conventionally pressed 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 2x-3x 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 SFRs 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

UO2 to MOX Design Differences

- Shorter fuel column/rod in MOX
- Smaller fuel diameter in MOX
- Steel cladding vs. zircaloy cladding
- Much higher heating rates in MOX
- Larger gas plenum
- Sodium coolant in SFRs
- Extended burnup for MOX
- Hexagonal packing of fuel pins



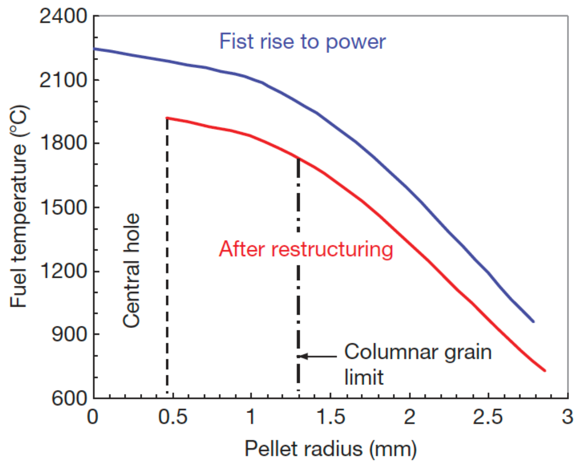
FFTF Assembly

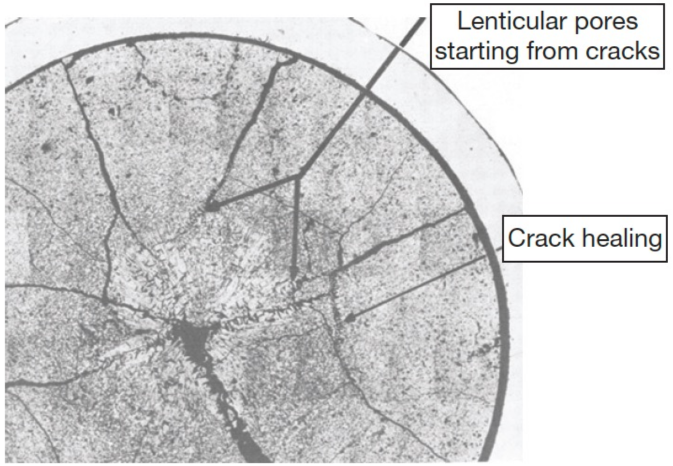
MOX Fuel

- 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

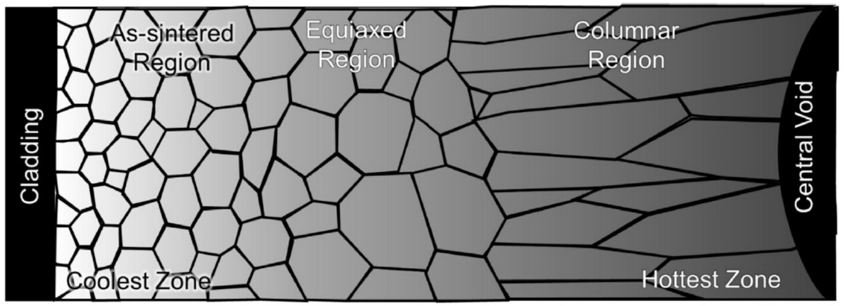
- Most phenomena occurring inside oxide fuel pellets are thermally activated, and a good knowledge of the thermal field inside the fuel stack is key
- Centerline temperatures can reach above 2000C, significantly higher than thermal LWRs
- Thermal conductivity is low and degrades with irradiation
- Due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

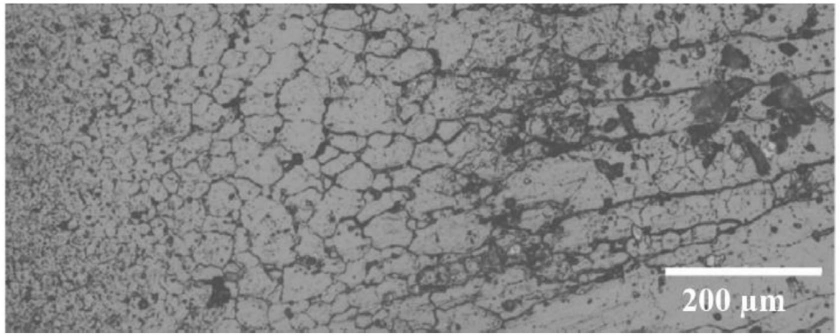


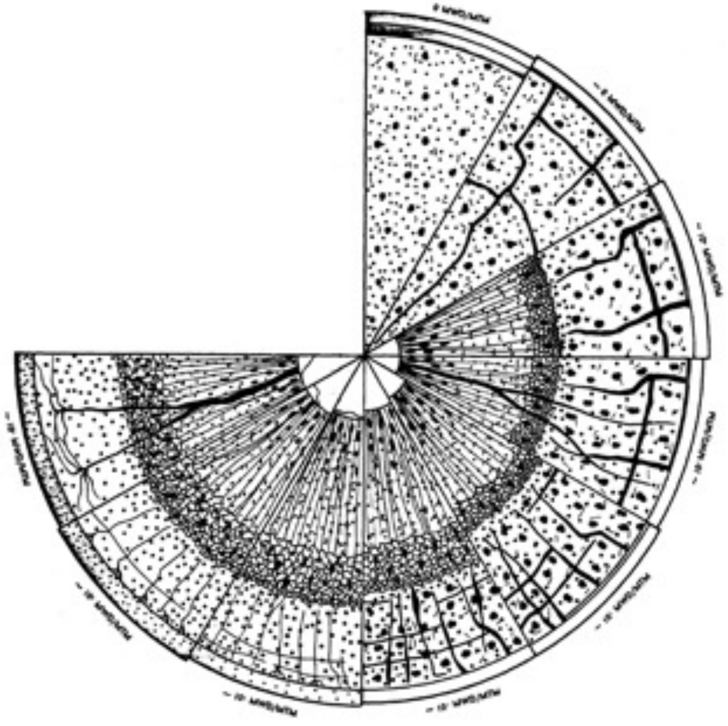


MOX Restructuring

- 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

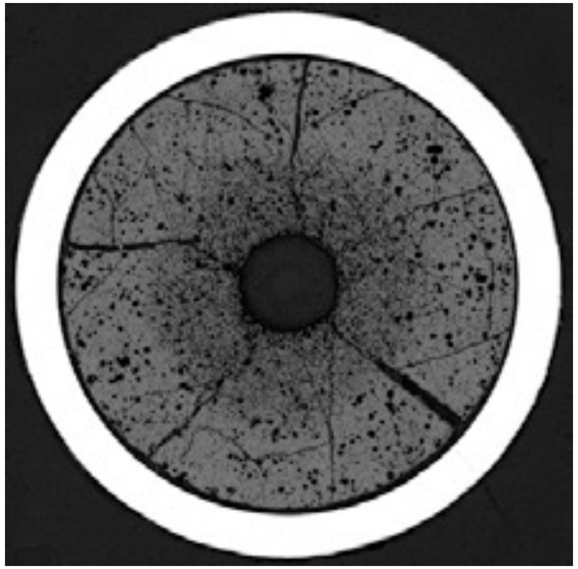


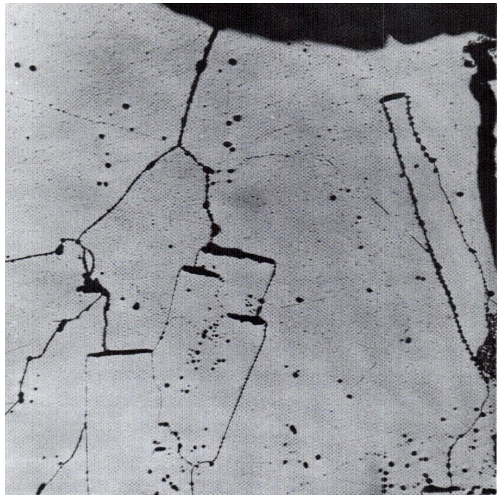




MOX Restructuring

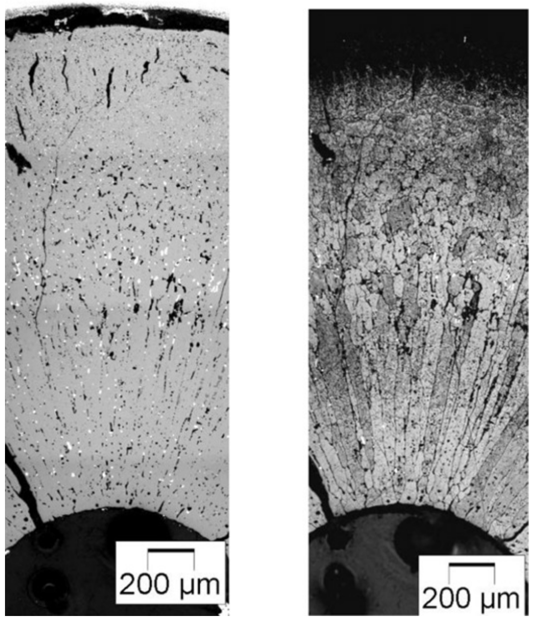
- The central void forms from the accumulation of voids and pores present in the fuel along a thermal gradient
- The pores orient in a lenticular shape normal to the temperature gradient
- The pores are filled with gas, and thus have a higher temperature gradient than that across the fuel
- The difference in fuel vapor pressures from hot to cold induces an evaporation–condensation mechanism: fuel evaporating from the hot face and condensing on the cold face travels down the thermal gradient
- This induces an inverse displacement of the lenticular pores that climb the thermal gradient toward the center of the pellet
- Vapor transport requires T~1800C

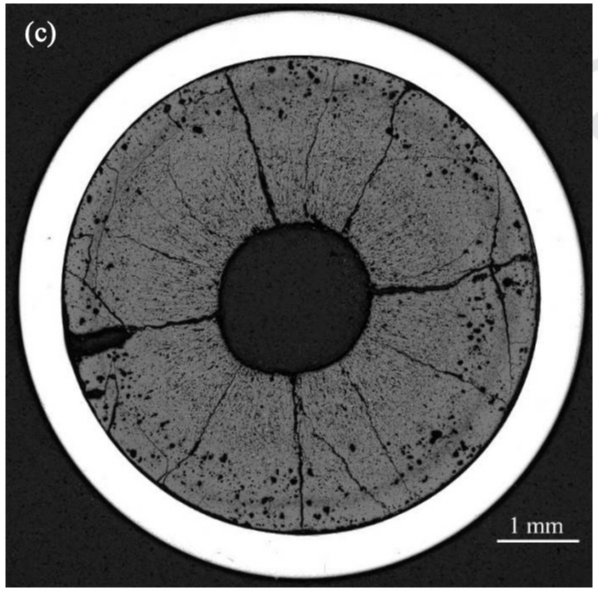




MOX Restructuring

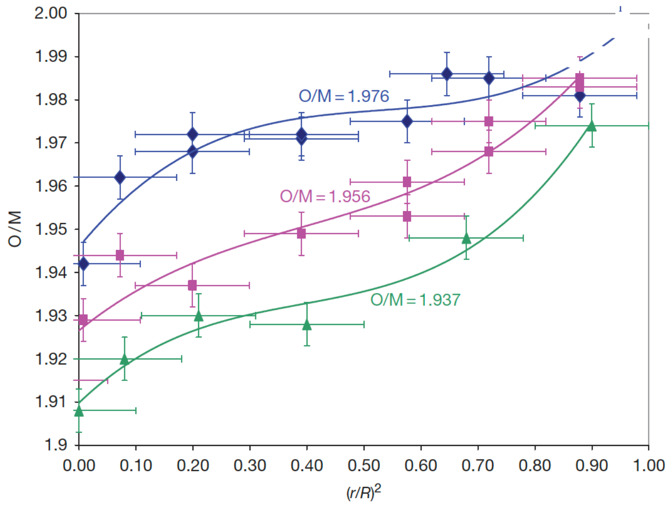
- While moving toward the pellet center, the lenticular pores destroy the initial fuel microstructure and leave behind them a dense fuel in elongated ‘columnar grains’ that appear clearly in micrographs
- The velocity of the pores varies rapidly with temperature, and the columnar grains only form at temperatures above ~1800C
- Below 1800C, an equiaxed region consists of grains that have undergone significant growth when compared to the un-irradiated samples
- At the periphery of the fuel pellet, temperatures are sufficiently low to limit grain growth, and thus the microstructure doesn’t undergo rapid changes

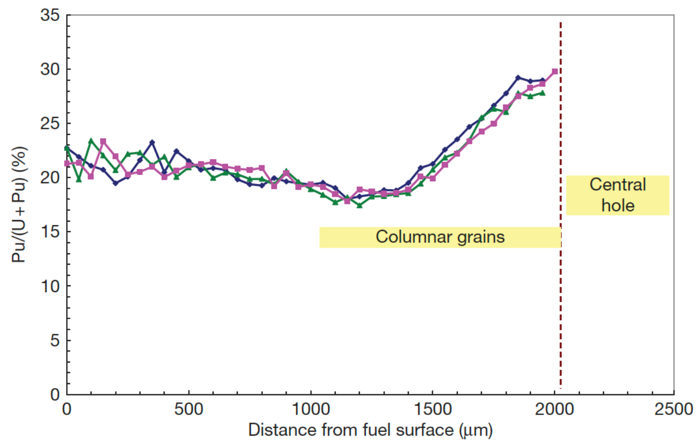




Constituent Redistribution

- The as-fabricated oxide pellets 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
- Oxygen and plutonium transport may be either via vapor transport, or Soret diffusion – not known





Gap Closure

- Although the thermal expansion coefficient is lower in oxide fuel than in austenitic SS 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
- Cladding creep down occurs relatively quickly
- The force exerted on the fuel column by the spring located in the upper rod (in addition to the weight of the fuel column) of causes an axial compression creep outwards of the fuel
- The main cause of swelling and gap closure is the fission product swelling of the fuel

Fission Products

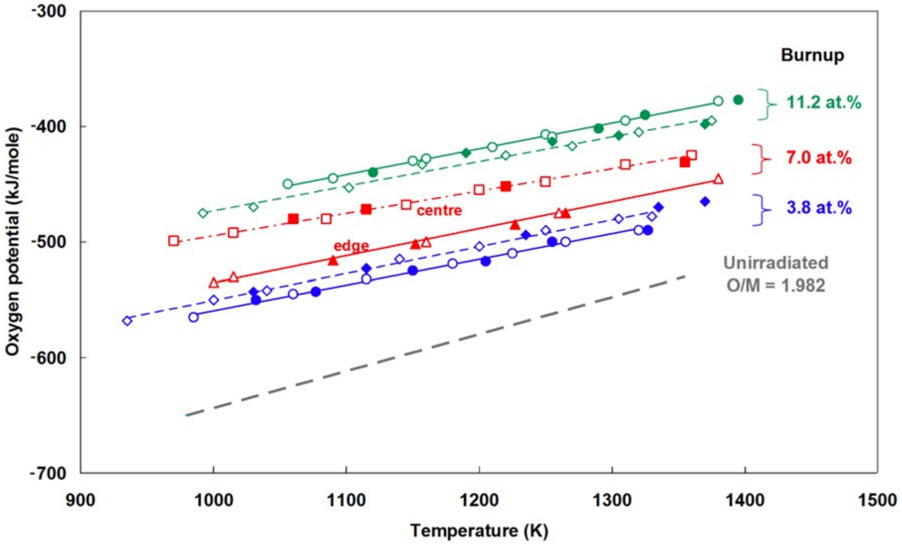
- The main objective of oxide fuels in fast reactors is to achieve very high burnup; 15 at.% or even more is typically considered as a reference target
- This means that at the end of irradiation, 15% of the initial actinide atoms (U and Pu) have disappeared and ~26% atoms present in the fuel are FPs
- All physical and chemical properties of oxide fuel will continuously evolve during its lifetime in the reactor; in particular, fission products will decrease the thermal conductivity and the 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

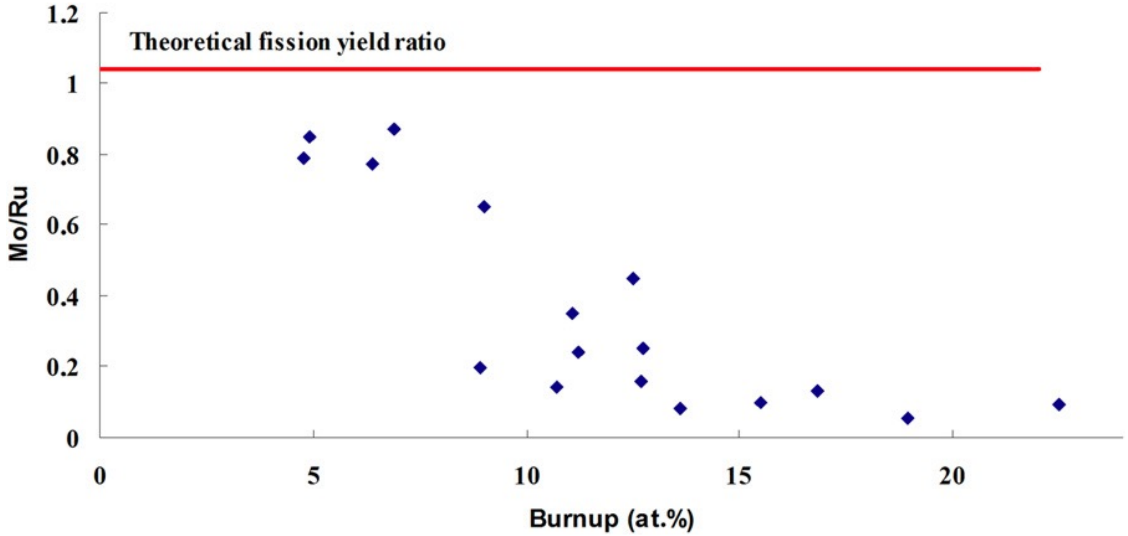
Fission Products

- 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

O/M Ratio

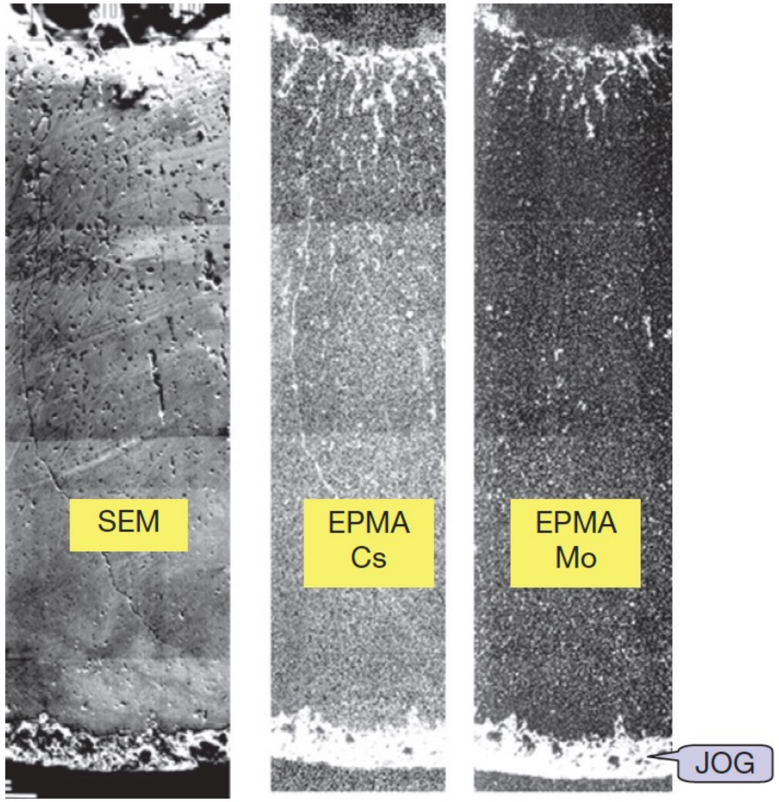
- Fission releases two oxygen atoms, which cannot be fully bound by the fission products, this increases the O potential in the fuel as dissolved O
- The O potential increases until the point where Mo starts to oxidize, leaving the metallic inclusions
- The O/M ratio increases until Mo starts to oxidize, then maintains at approximately 2.0 or thereabouts





Joint Oxyde Gaine (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
- JOG is probably Cs2MoO4, but includes lots of other fission product species
- 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



JOG Effects

- The thermal conductivity of Cs_2MoO_4 is about 10x lower than MOX fuel, but much higher than gases
- JOG acts as a mechanical buffer, limiting the amount of PCMI, or stresses in the cladding
- JOG reduces fuel swelling, by allowing fission products to leave the fuel into a preferable chemical state
- Fission products in the JOG play a predominant role on FCCI and on the resulting strong corrosion

FCCI

- FCCI appears as one of the potential life-limiting factors for high burnup fuel elements
- 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

Accidents/Transients in MOX SFRs

- For sodium fast reactors three main types of transient and accident scenarios are considered as references for the study of fuel transient and accident behavior:
  - the slow power transient representative of one control-rod withdrawal
  - the loss of cooling due to blockage of a subassembly
  - the unprotected loss of flow (ULOF) accident that might lead to a core disruptive accident

Control Rod Withdrawal Accident (CRWA)

- Slow power transients due to control-rod withdrawal are the most common transients occurring during reactor operation as they are used for regulation of the core power
- In case of an inadvertent control-rod withdrawal due to operator action together with a failure of various protection systems, this transient might lead to severe consequences
- During such a transient, nominal power can increase by 1-3% per second, and may lead to partial fuel melting inside the pins of the subassemblies surrounding the control rod

Local blockage of a subassembly

- Local blockage formation in a fuel assembly due to ingression of some external material into the bundle may lead to pin failure with subassembly degradation and melting
- The hypothetical total instantaneous inlet blockage (TIB) of a subassembly at nominal power has been suggested as a potential initiator for a core melt accident
- Owing to the rapid and complete loss of flow in the faulted subassembly, the usual detection systems are not operating, so core power can not be shut down in time
- The accident is characterized by overheating and melting of the fuel pins, degradation of the subassembly, wall failure, and possible propagation of molten materials into neighboring subassemblies
- The main safety issue is the risk of propagation of the accident beyond the neighboring subassemblies that might lead to critical events and generalized core melting
- Such an accident caused the Fermi-1 core meltdown in 1966

Unprotected loss of flow accident (ULOF)

- The ULOF accident is the result of loss of primary pump flow due to an initiating event such as electrical break-down without reactor scram
- The first phase leads to sodium flow reduction and to associated power reduction linked to reactivity feedback
- The power to flow ratio increases so that the temperature causes sodium boiling
- Due to the positive ‘sodium void effect’ in the central core regions, the ULOF leads to sodium boiling and channel voiding and may result in a core disruptive accident (CDA)
- This can lead to generalized core degradation: fuel melting, clad failure and/or melting, fuel ejection into coolant, fuel dispersal and relocation into the channels, and possible mechanical energy release
- Although this accident is initiated by a loss of coolant flow, it may rapidly evolve toward a fast reactivity insertion accident

Fuel Behavior Under Slow Power Transients

- Despite high fission gas release rate in MOX fuel due to high operation temperature (60-80% release), power increases resulting in higher temperatures also activate fission gas-related phenomena that influence thermal and mechanical pin behavior
- Intragranular gas migration toward grain boundary related to the thermal gradient
- Gas bubble growth due to coalescence via vacancy diffusion
- Fission gas-induced fuel swelling driven by an increase in the bubble pressure
- Saturation and interconnection of grain boundary bubbles leading to additional gas release
- Under slow power transients, fuel heat generation and heat removal by the sodium coolant are almost in thermal equilibrium, and these ‘quasi’ steady-state conditions allow the evaluation of the power level at which fuel melting occurs, providing operating limits

Fuel Melting

- SFRs with MOX are sensitive to any fuel compaction effect that might result from fuel motion (for instance, due to melting) with the potential risk of prompt-critical events and mechanical energy release to the vessel structure
- Fuel-melting occurrence depends on pin thermal behavior that is governed by thermal conductivity, heat exchange between fuel and clad through the pellet–clad gap, and melting temperature
- Melting temperature and thermal conductivity depend on stoichiometry and burnup
- Heat exchange in the fuel system is governed by evolution of gap thickness and composition
- High T plastic fuel creep may lead to central hole reduction and to increase of the macroscopic fuel porosity with subsequent reduction of the thermal conductivity and thus higher fuel temperature in the center that results in a lower power at melting onset
- High peaking factors (~1.3, compared to ~1.1 in LWRs), can increase the local temperatures in high power regions and increase probability of fuel melting

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
- Unique features include restructuring, central pore formation, and JOG formation
- Three main types of transient of interest in MOX SFRs
  - slow power transient (CRWA); blockage of a subassembly; unprotected loss of flow accident
- Fuel melting is one of the most critical concerns