

Nuclear Fuel Performance

NE-533
Spring 2023

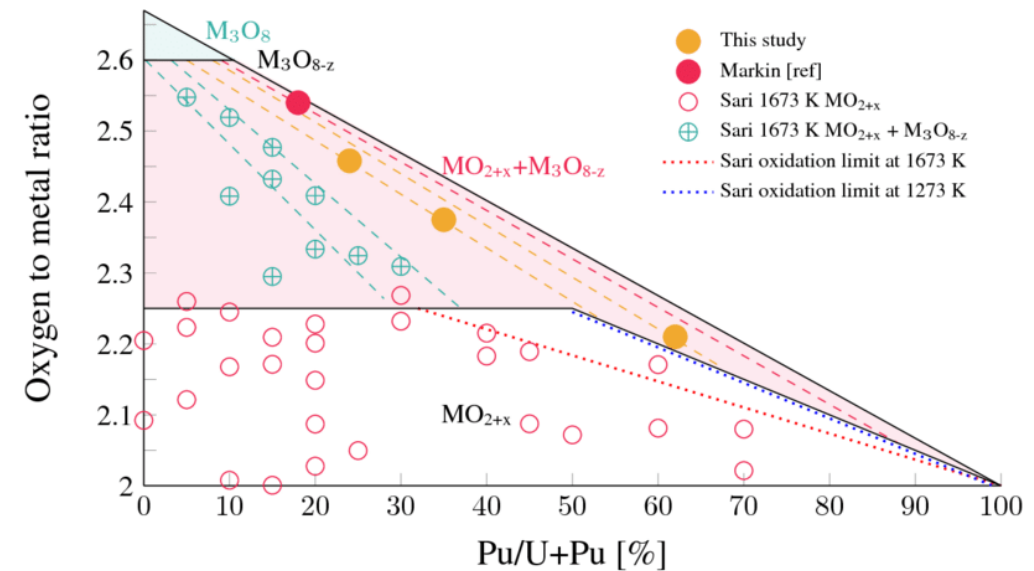
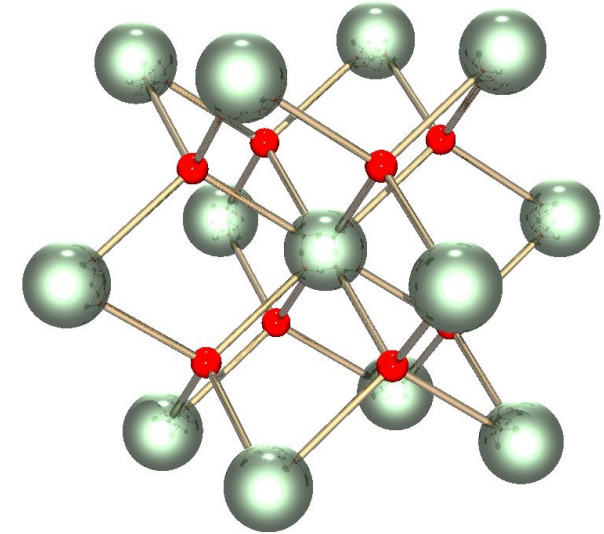
Last Time

- LOCA
 - increases temperature with a decrease in coolant pressure
 - High temperatures lead to increased oxidation, producing additional hydrogen
 - Degrades cladding, increasing likelihood for failure
- 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

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

	<i>BN350</i>	<i>Phénix</i>	<i>PFR</i>	<i>BN600</i>	<i>FFTF^a</i>	<i>Super-Phénix</i>	<i>MONJU</i>
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.%) (GWd t ⁻¹)	9.0	16.9	23.5	11.8	24.5	Not relevant	Not relevant
Maximum dose (dpa)	60	156	155	90		–	–

MOX Designs

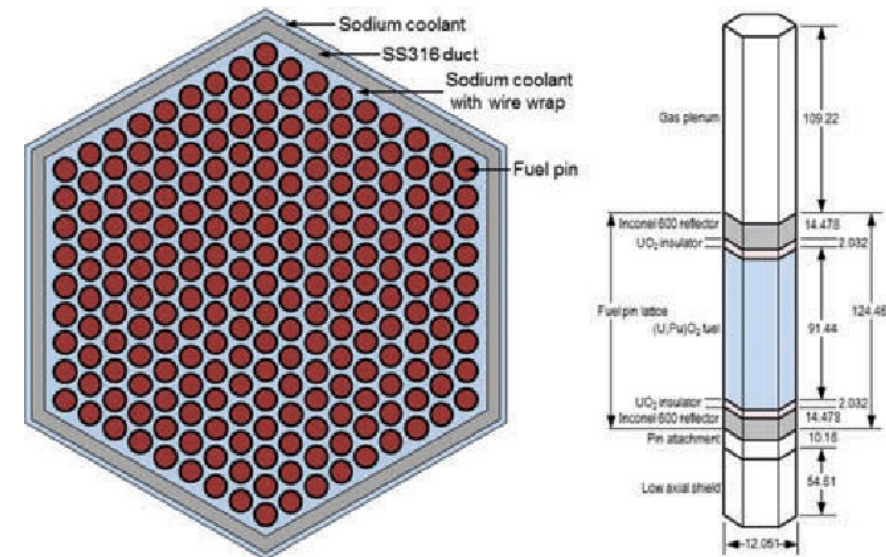
- The fuel pin is a long cylinder (2–3m long, 5–10mm diameter), clad in a steel tube (0.4–0.6mm thick) closed in both ends by welded plugs, preventing direct contact between the radioactive material and the sodium coolant
- The oxide fissile column (~1 m long) consists of a stack of conventionally pressed 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 ($\sim 7 \times 10^{15}$ n/cm²/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



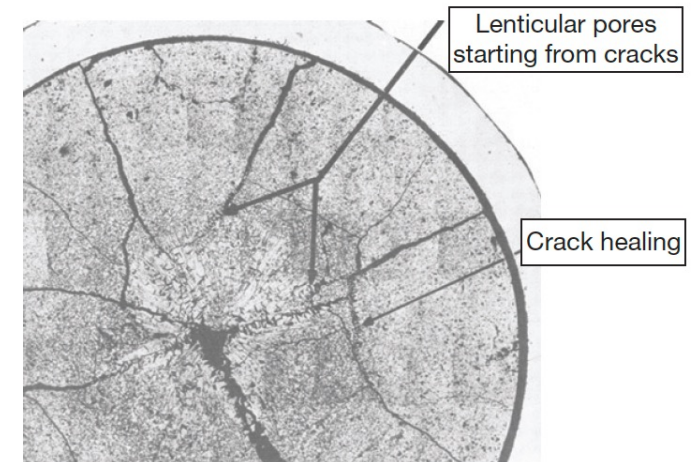
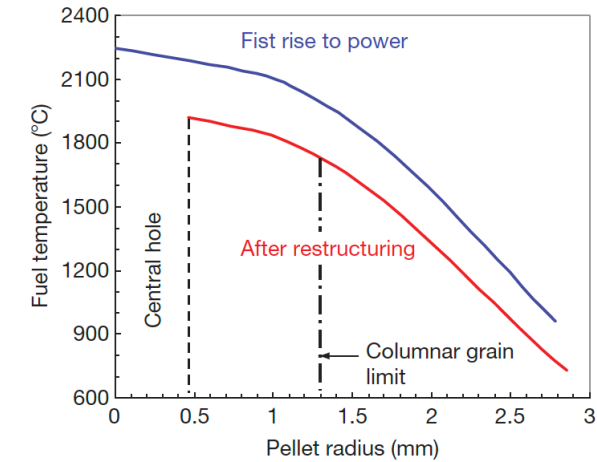
FFTF Assembly

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

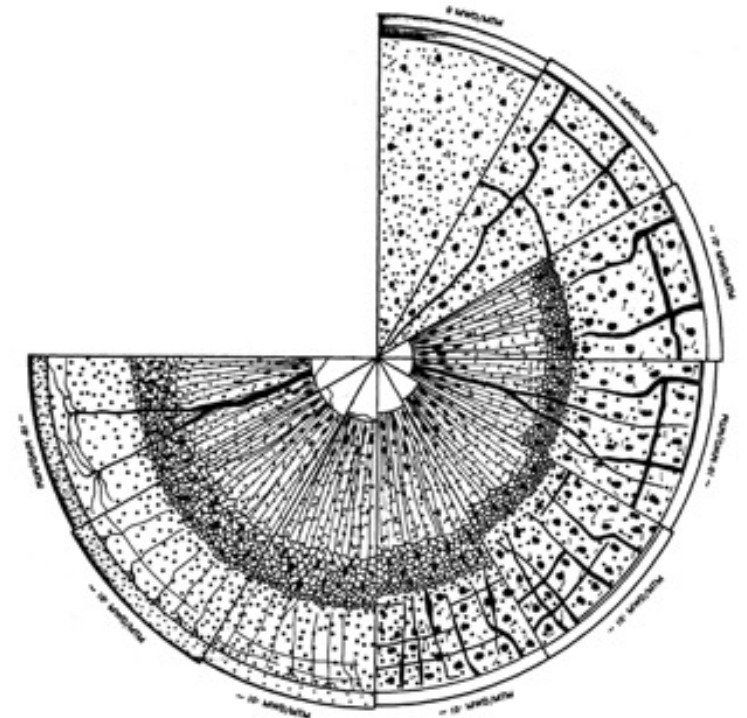
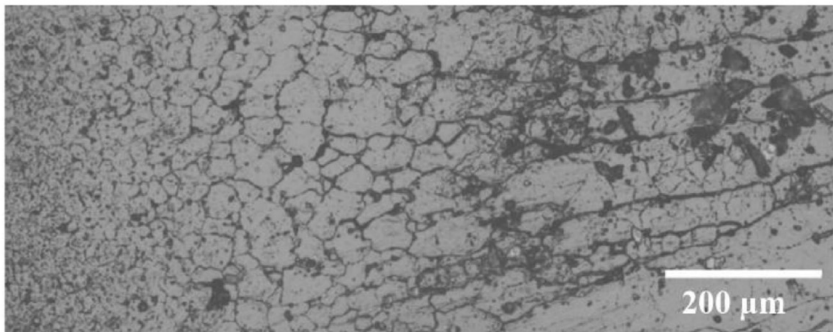
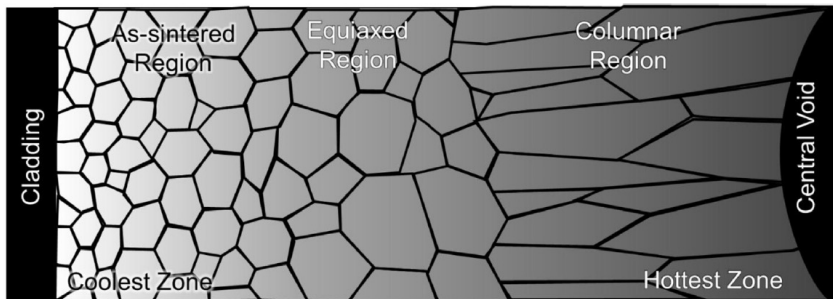
MOX Fuels

- 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



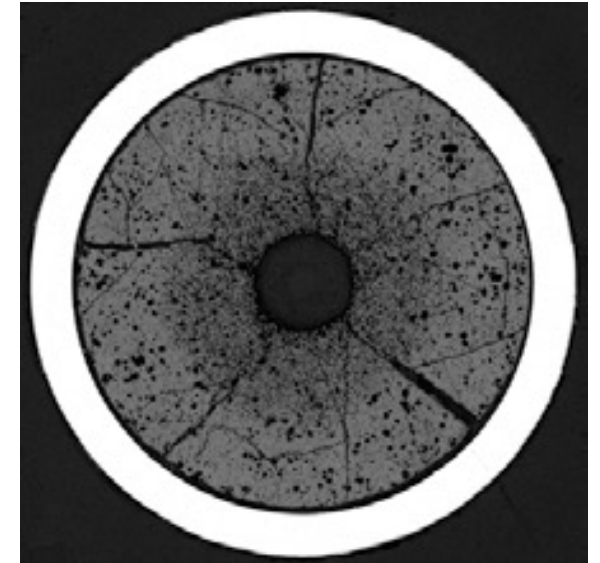
MOX Restructuring

- 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



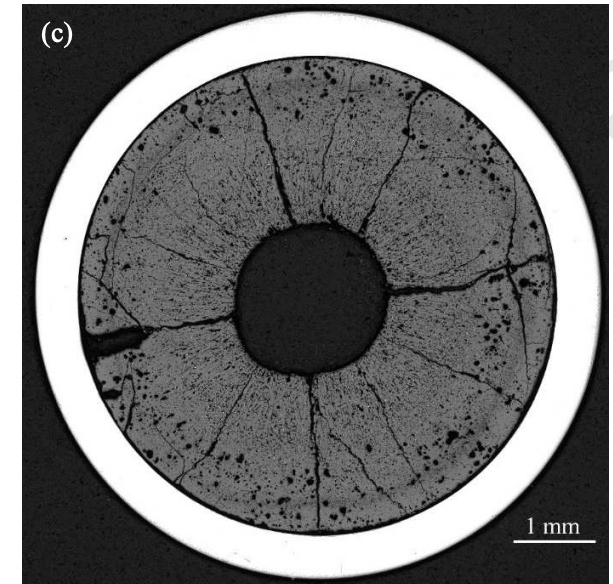
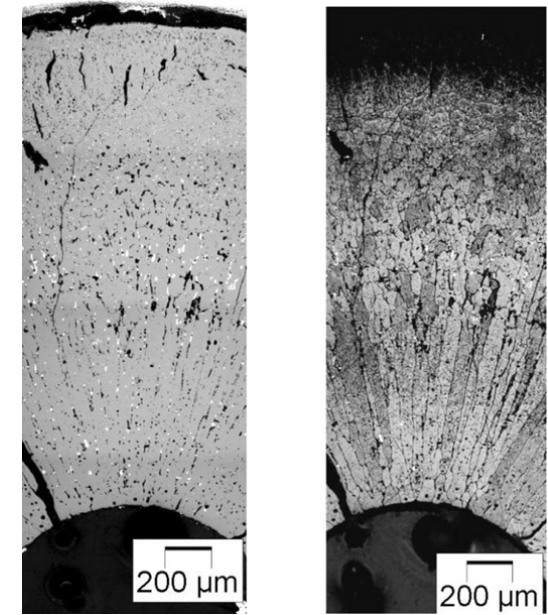
MOX Restructuring

- 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 $T \sim 2000^\circ\text{C}$



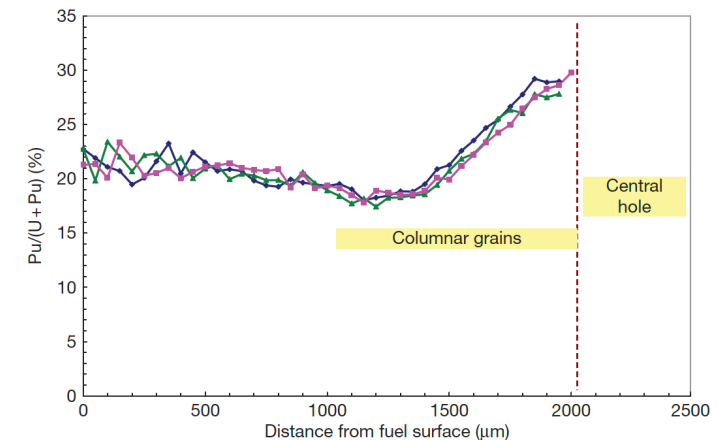
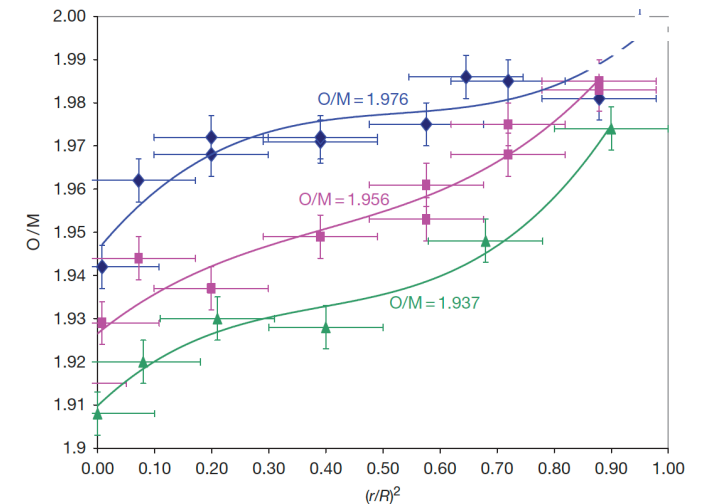
MOX Restructuring

- 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 $\sim 1800^{\circ}\text{C}$
- Below 1800°C , 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

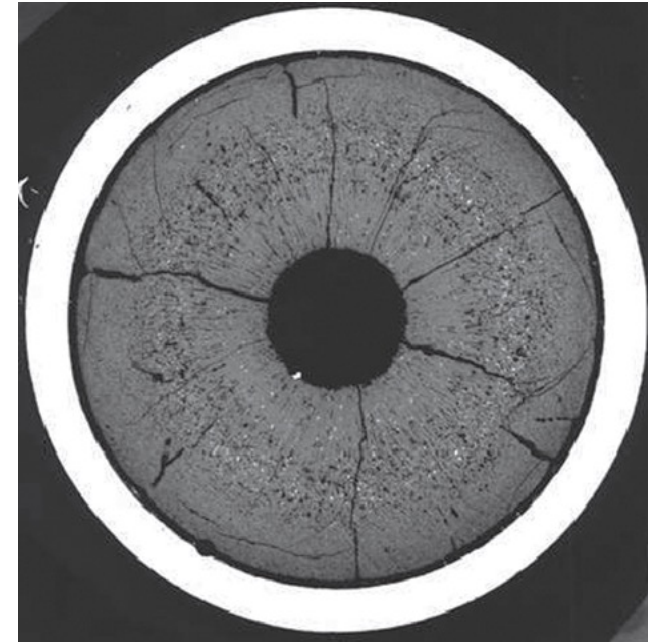
- Although the thermal expansion coefficient is lower in oxide fuel than in austenitic stainless steel cladding the temperatures in the fuel pellets are much higher than in the cladding and induce a higher thermal expansion in the fuel pellet
- At high linear powers gap closure is completed after a burnup 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
- 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 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 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

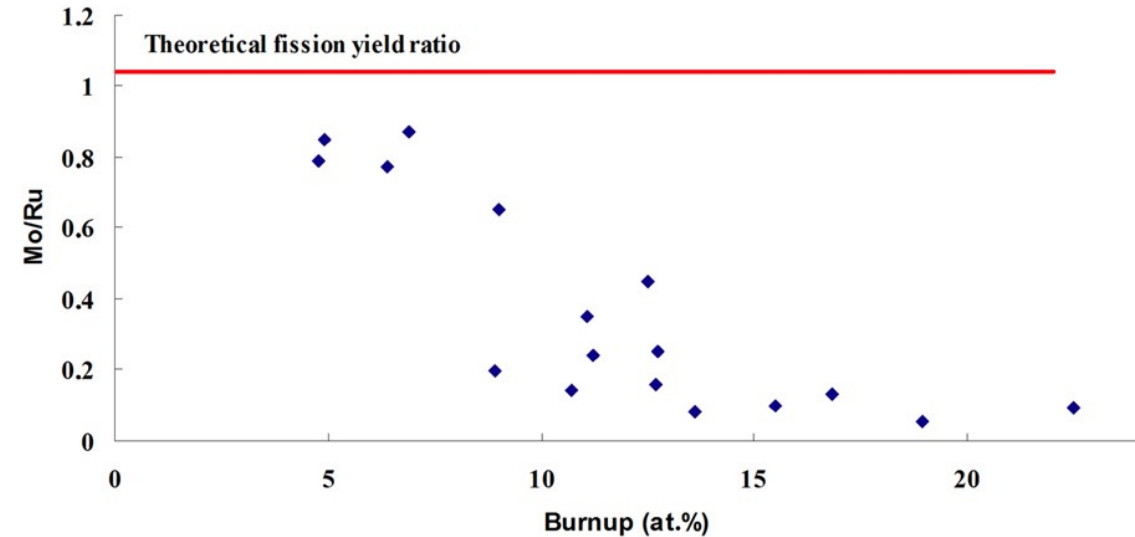
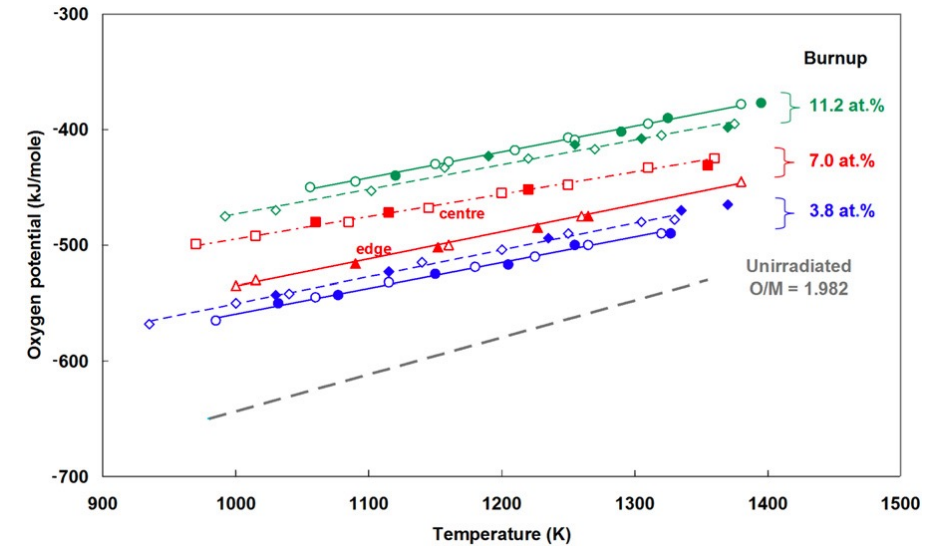
Fission Products

- 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



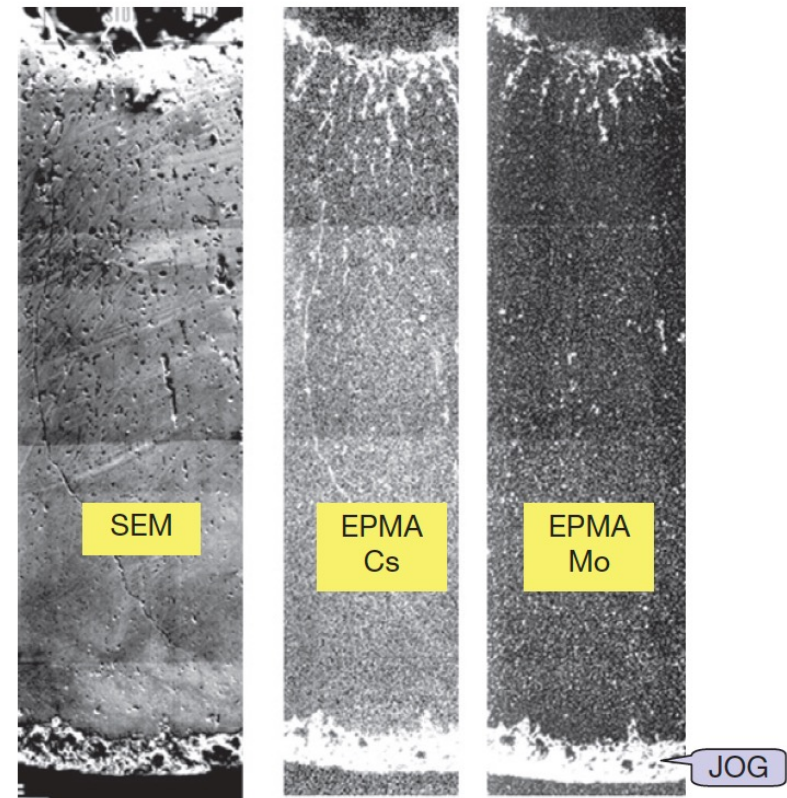
O/M Ratio

- 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 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
- JOG is probably Cs_2MoO_4 , 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 $150\text{ }\mu\text{m}$



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)O₂ 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
- Unique features include restructuring, central pore formation, and JOG formation