Fuels for Fast Spectrum Reactors

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June 12, 2010

INL/MIS-10-18898

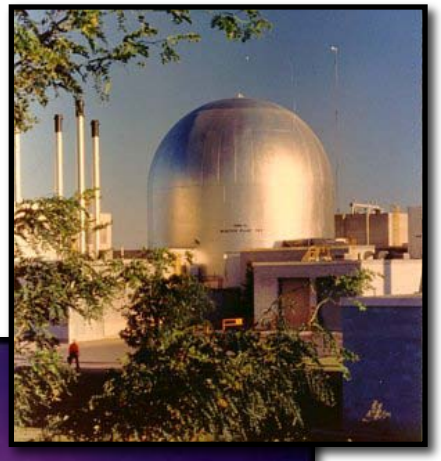
Outline of Presentation

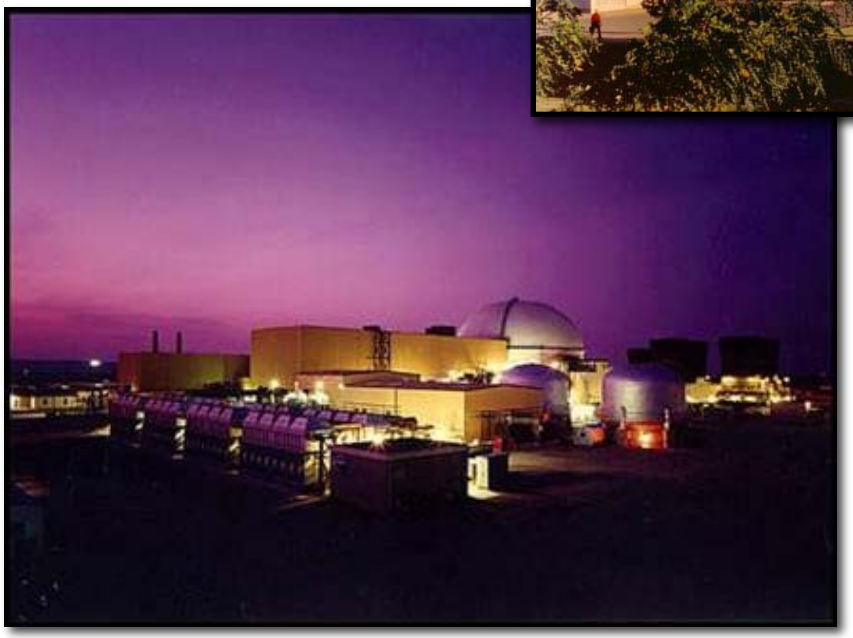
- **Introduction**
- **SFR Fuels Experience in the US**
  - Fuel Types
  - Fuel Performance Issues
  - Experience/Testing
- **Experience with Fuels Containing Minor Actinides**
- **Summary**

SFR Fuels Experience in the US

SFR Fuels Experience in the US

- **Metallic Fuels**
  - EBR-I, Fermi-1, EBR-II, FFTF
  - U-Fs, U-Mo, U-Zr, U-Pu-Fs U-Pu-Zr, others
- **Mixed Oxide Fuels (MOX)**
  - EBR-II, FFTF
  - $(U,Pu_{0.2\text{-}0.3})O_2$
- **Mixed Carbide Fuels (MC)**
  - EBR-II, FFTF
  - (U,Pu)C w/15% $(U,Pu)_2C_3$

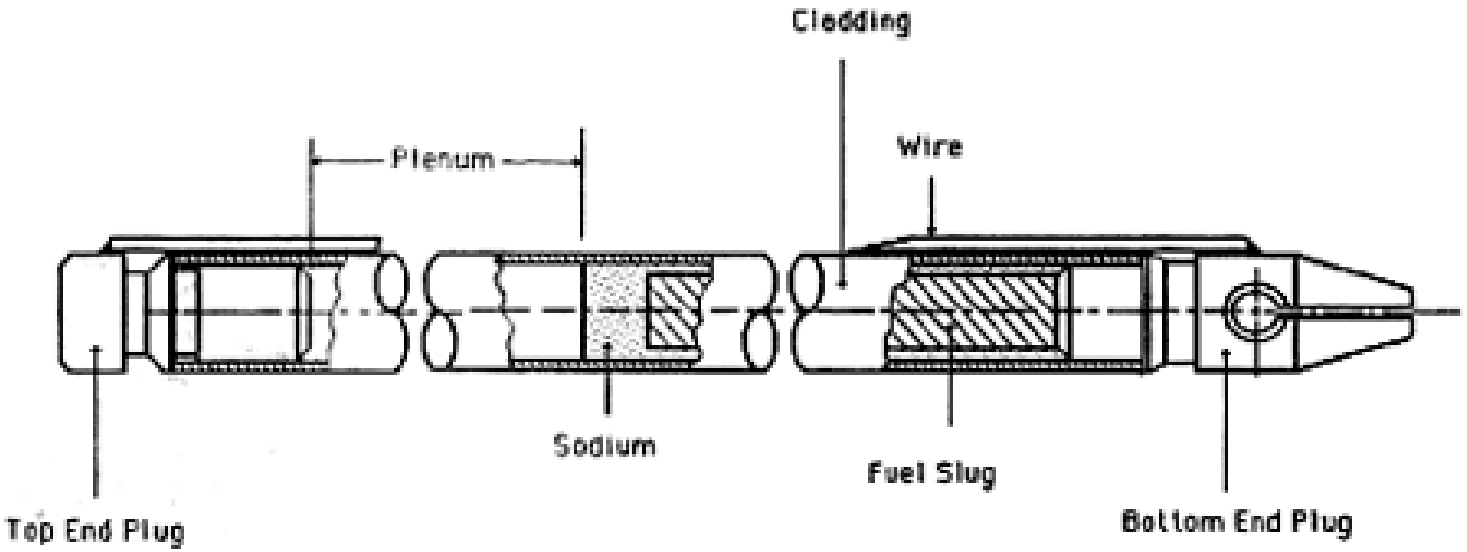




Comparative fuel designs

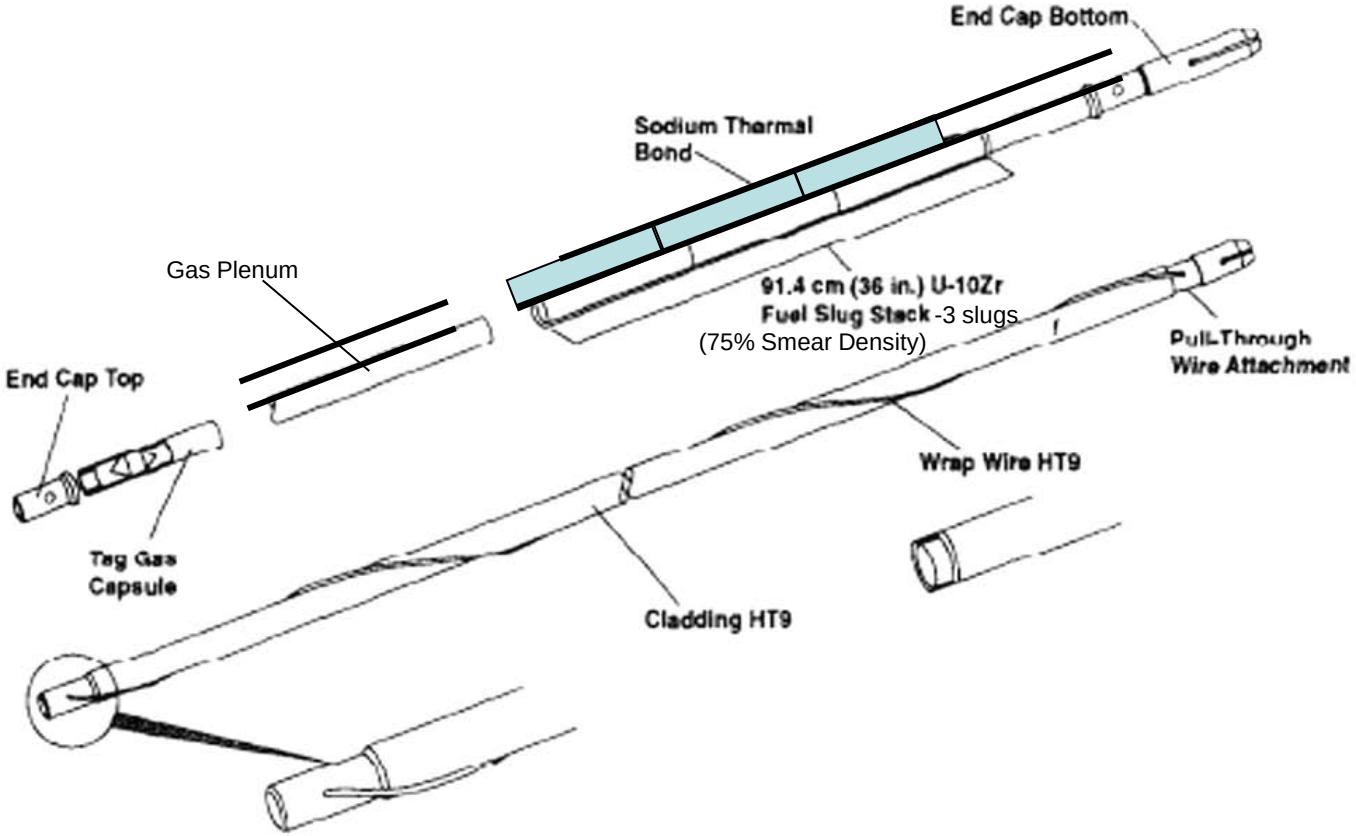
	Ceramic fuel			Metallic fuel
	Oxide	Carbide	Nitride	
Fuel form	$(U,TRU)O_2$	(U,TRU)C	(U,TRU)N	U-TRU-10Zr
Theoretical density (TD), g/cm^3	11.1	13.6	14.3	15.7
Smeared density, %TD	85	75	85	75
Smeared heavy metal density, g/cm^3	8.3	9.7	11.5	10.6
Mass fraction of non-metal, %	11.8	4.8	5.5	10.0
Melting temperature, K	2930	2750	3070	1000
Thermal conductivity, W/m-K	1.70	18.8	15.8	~27
Crystal structure	b) SC+FCC	FCC	FCC	c) α, β, γ
Bond material	He	Na or He	Na or He	Na
Achieved burnup, % atom	~15	~10 or ~15	~15	~19

Metallic Fuel Design (EBR-II)



Features of a Metallic Fuel Pin (from Pahl, et al, 1990)

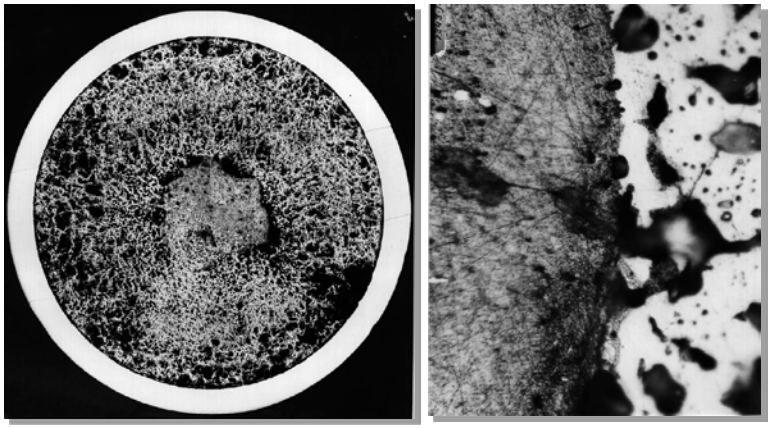
Metallic Fuel Design (FFTF)

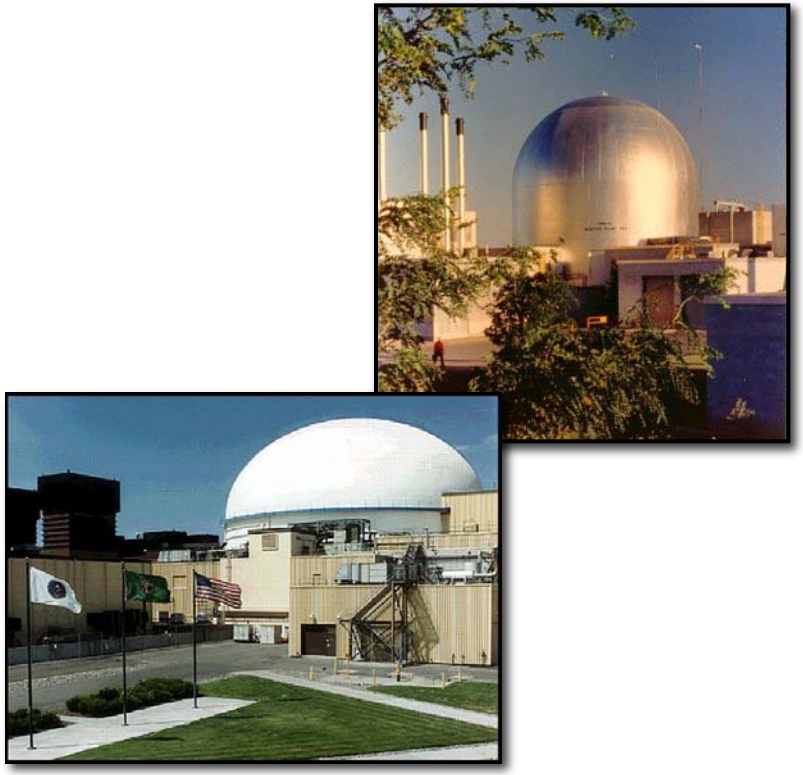


FFTF Series III.b Metallic Driver Fuel Design (from Pitner and Baker, 1993)

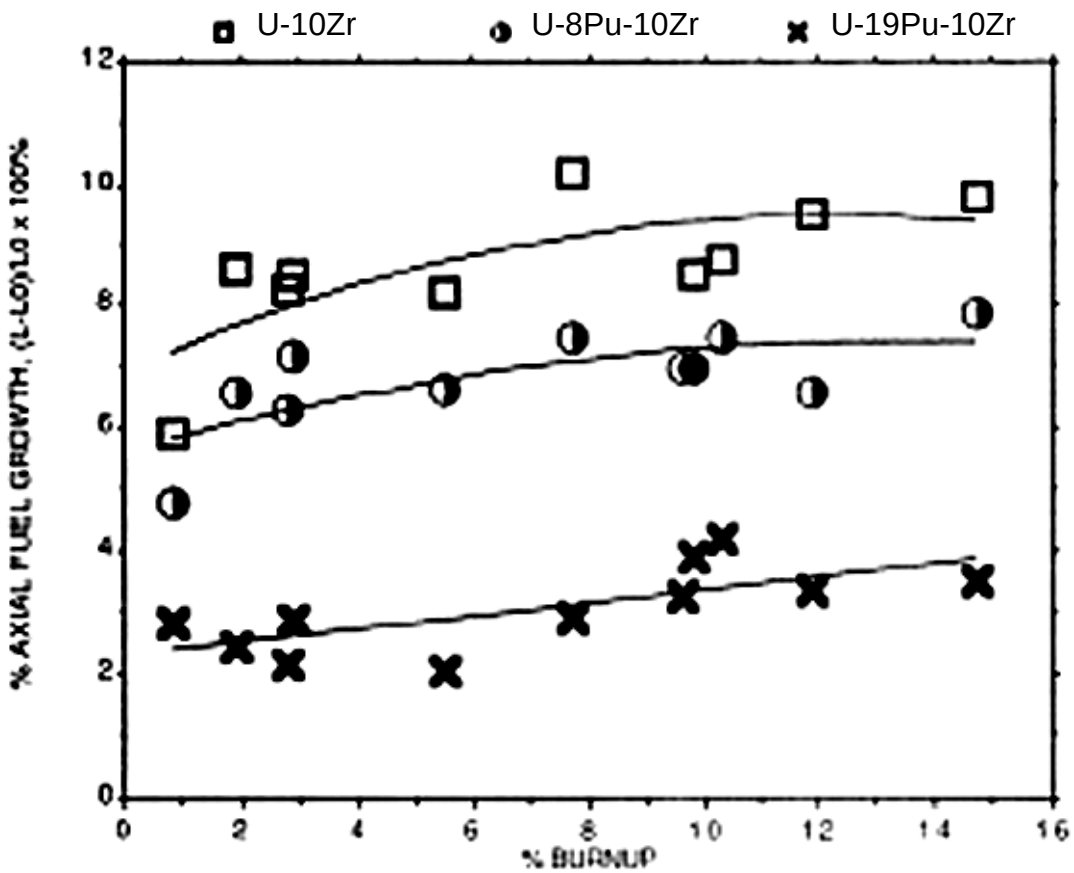
Important Metallic Fuel Performance Phenomena

- Irradiation growth
- Fuel swelling and fuel-cladding mechanical interaction (FCMI)
- Gas release
- Fuel constituent redistribution
- Fuel-cladding chemical interaction (FCCI)



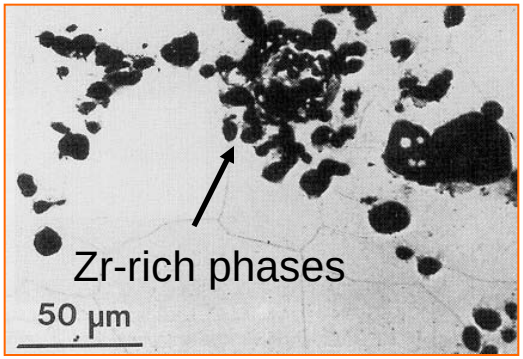


Metallic Fuel Behavior—Axial Growth

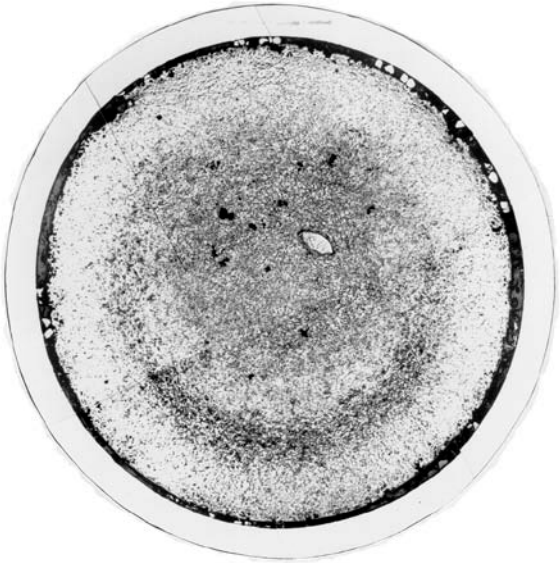


Axial Fuel Growth, from Pahl et al, 1990

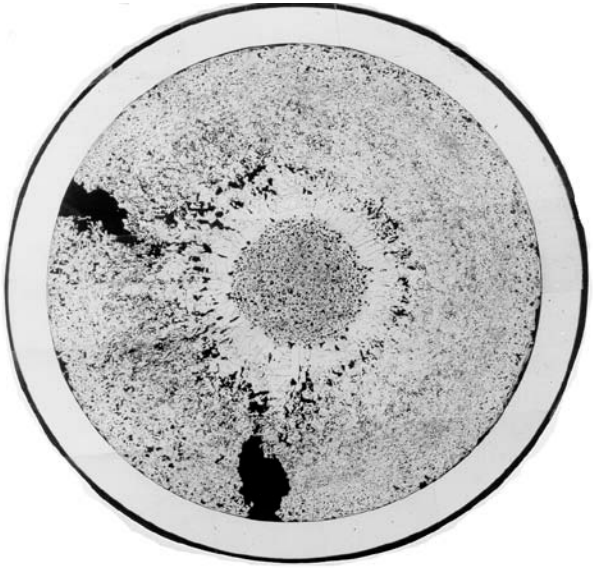
Metallic Fuel Behavior—Swelling & Restructuring



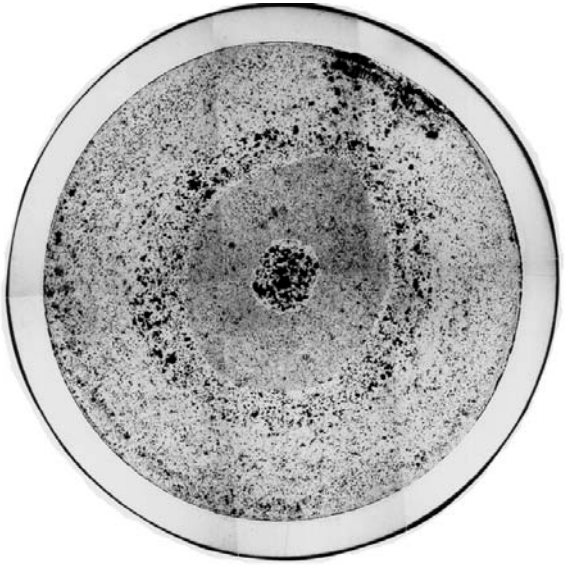
As fabricated U-20Pu-10Zr



X423A at 0.9% BU



X419 at 3% BU

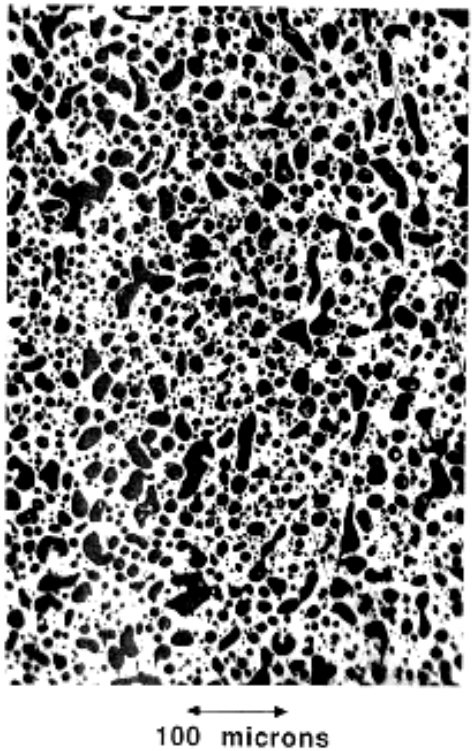


X420B at 17% BU

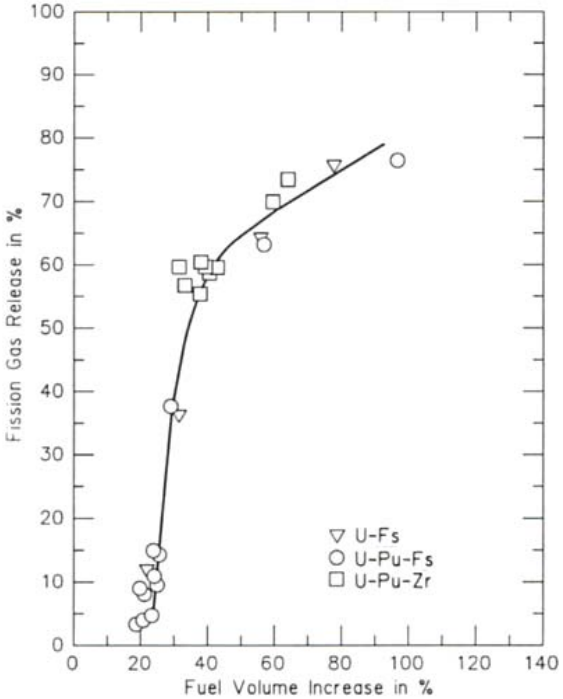
- Redistribution of U and Zr occurs early
- Inhomogeneity doesn't affect fuel life

Metallic Fuel Behavior—Swelling & Gas Release

- **Swelling**
  - Low smear density fuels
  - Rapid swelling to 33 vol% at ~2 at.% burnup
- **Gas Release**
  - Inter-linkage of porosity at 33 vol% swelling results in large gas release fraction
  - Decreases driving force for continued swelling

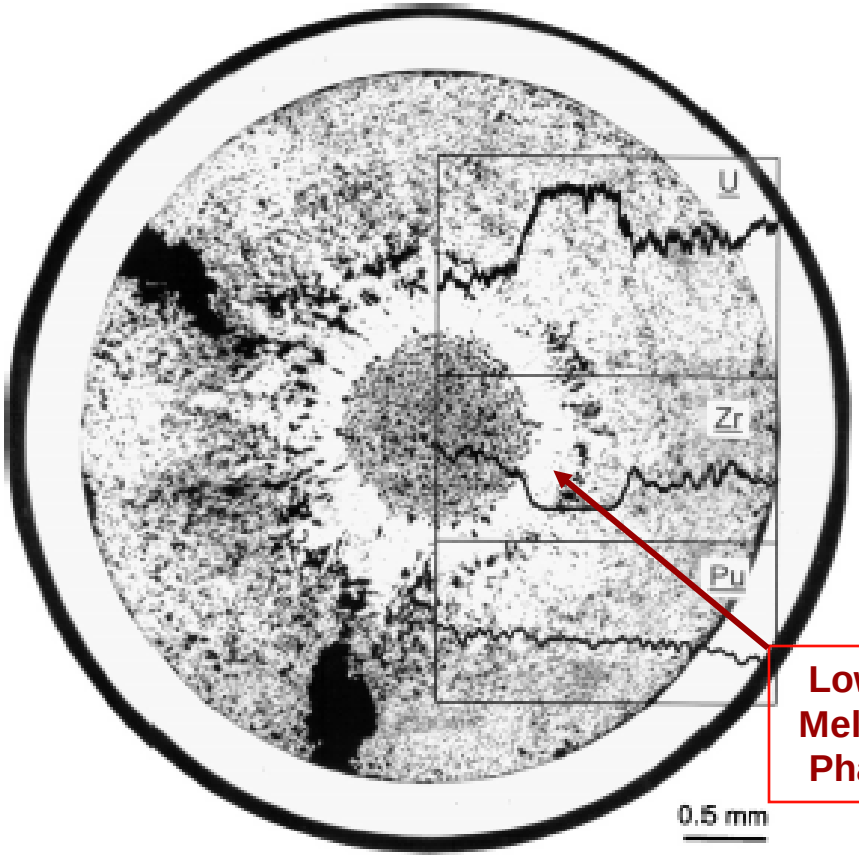


U-19Pu-10Zr (©-phase) at 2 at.% burnup

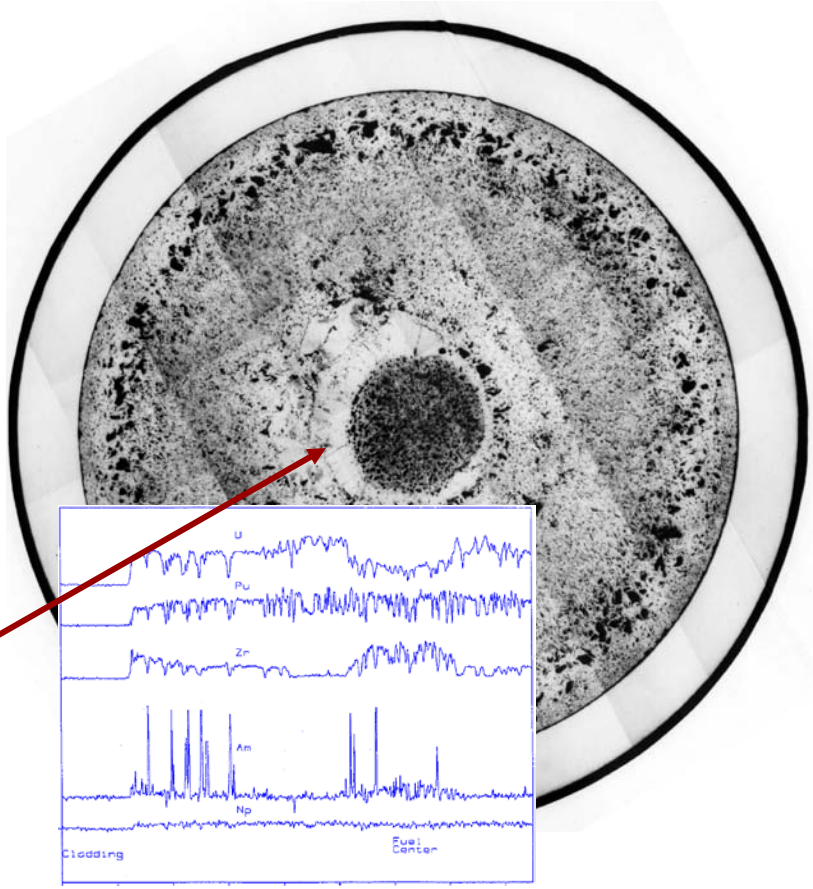


Metallic Fuel Behavior—Fuel Constituent Redistribution

U-Pu-Zr



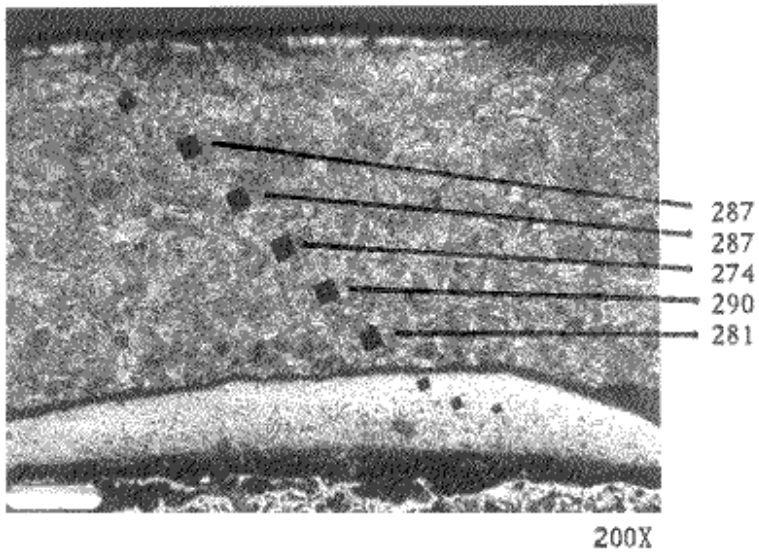
U-Pu-Am-Np-Zr



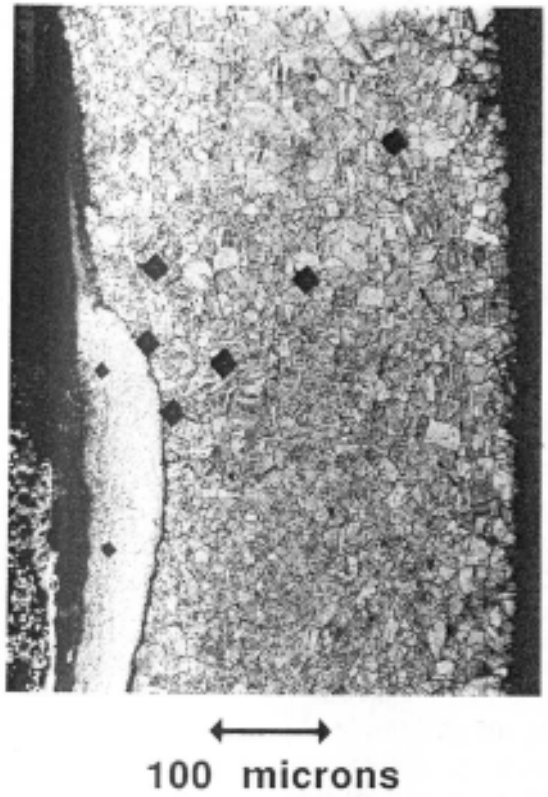
Lower Melting Phase

Metallic Fuel Behavior—Steady-state FCCI

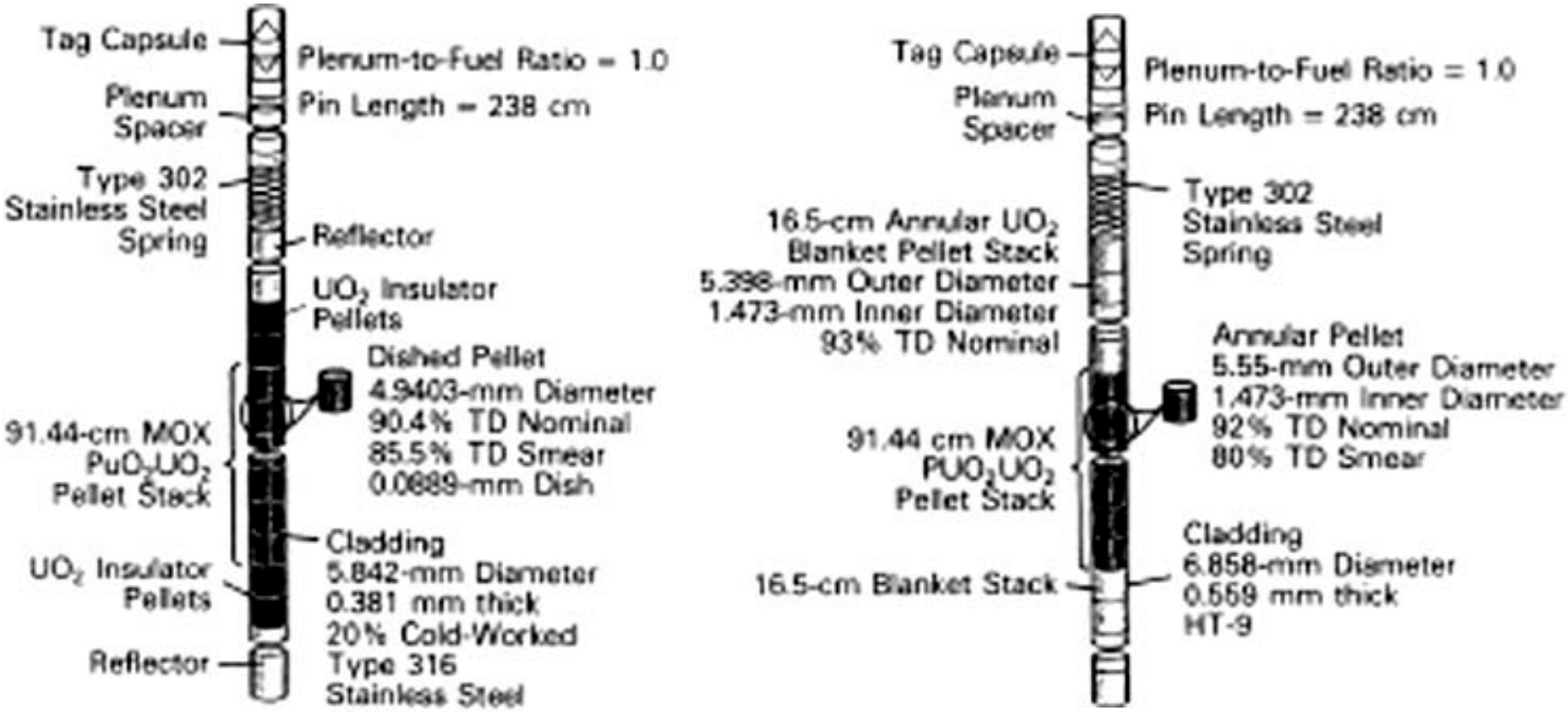
- **Fuel-Cladding Inter-diffusion**
  - RE fission products (La, Ce, Pr, Nd) and some Pu reacts with SS cladding
  - Interaction product brittle
  - Considered as cladding wastage



U-19Pu-10Zr with D9;
12 at.% burnup
(from Pahl, et al, 1990)



MOX Fuel Design (FFTF)

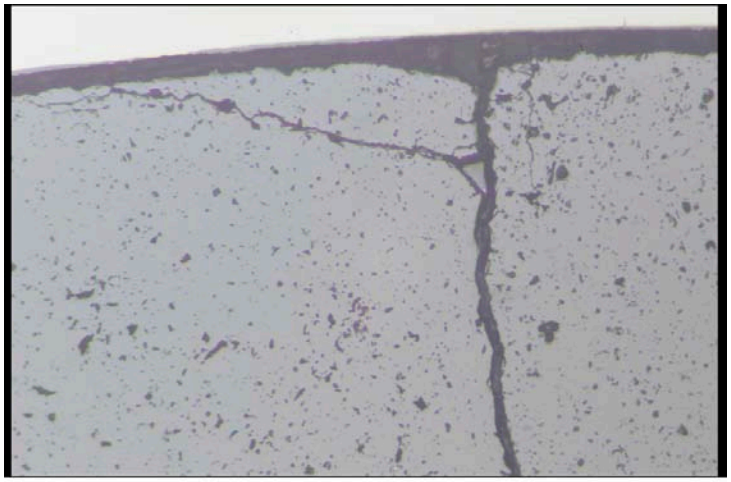


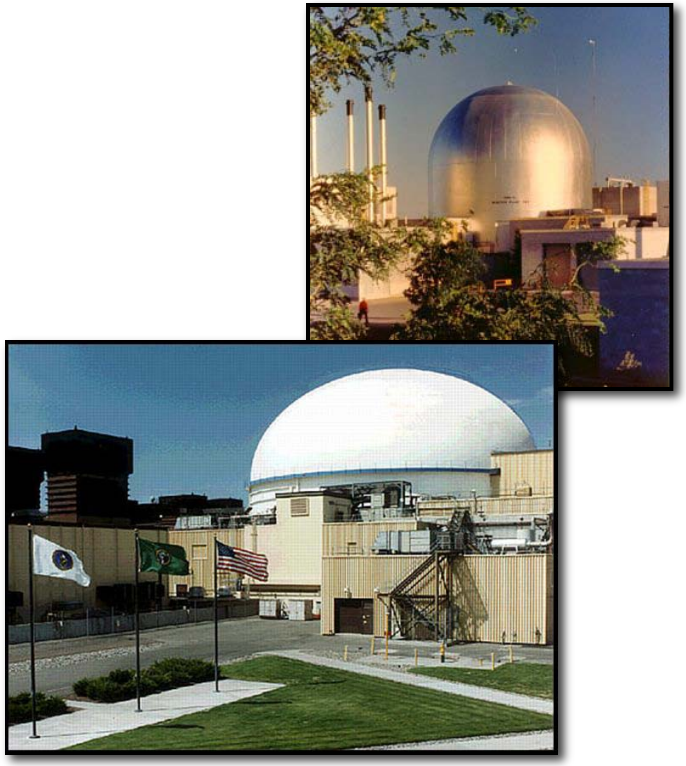
a) b)

FFTF He-bonded MOX Fuel: a) Driver Fuel and b) Core Demonstration Experiment Fuel
(from Bridges et al, 1993)

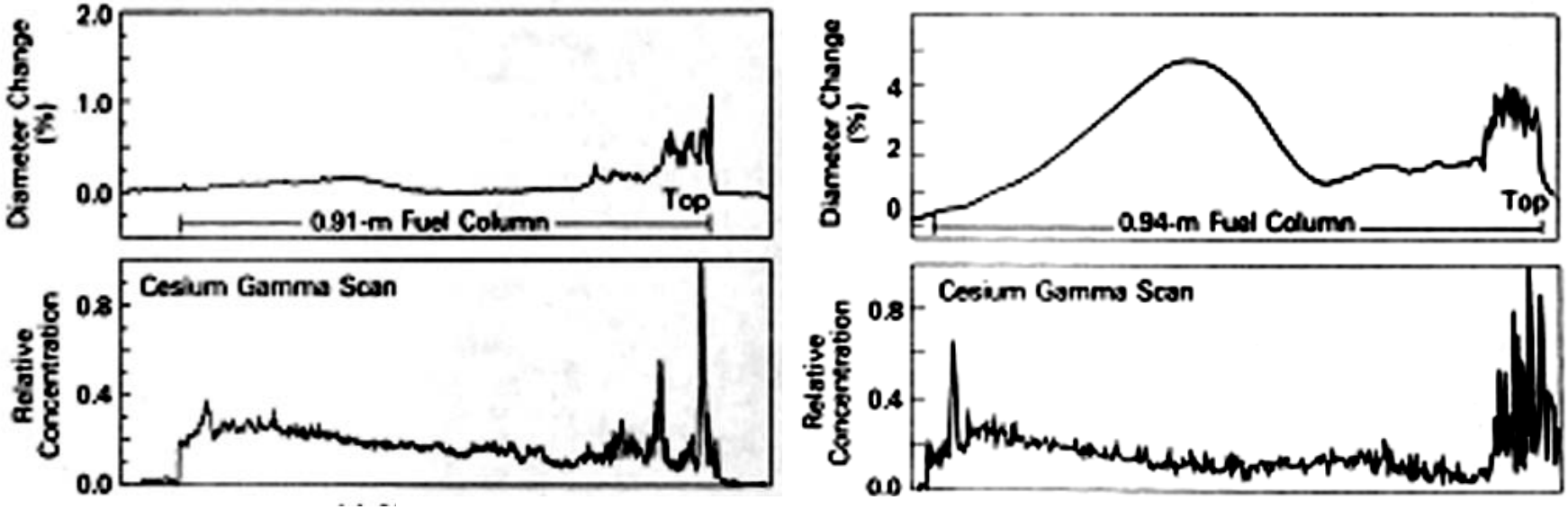
Important MOX Fuel Performance Phenomena

- Fuel swelling and FCMI
- Fuel restructuring
- Gas release
- FCCI
- Fuel-coolant compatibility





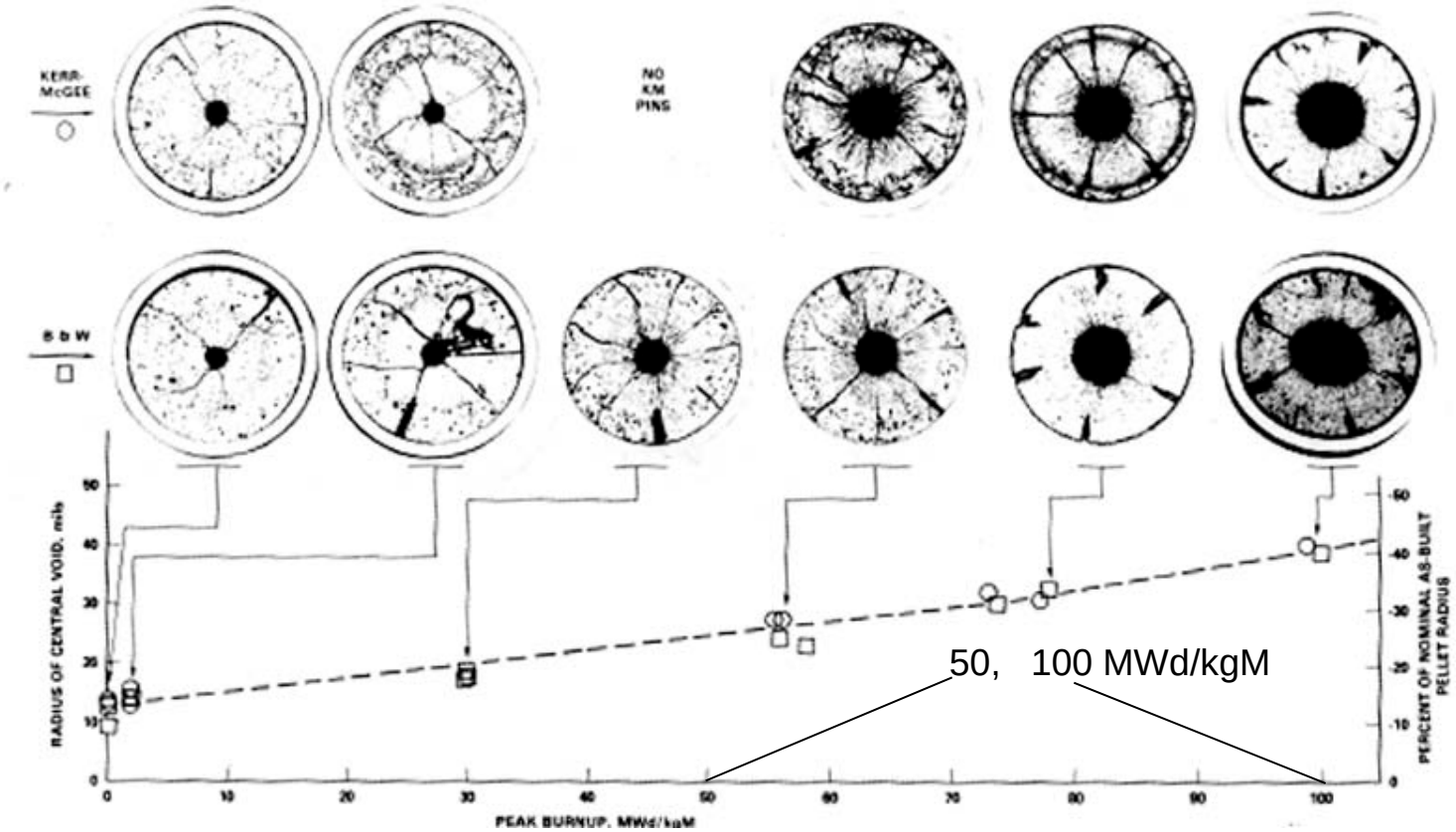
MOX Fuel Behavior—Fuel Swelling and FCMI



(a) (b)

Diameter and cesium fission product accumulation in high-temperature MOX pins, HT9-clad (a) and D9-clad (b). Cs interacted with MOX fuel causing FCMI.
(from Bridges, et al ,1993)

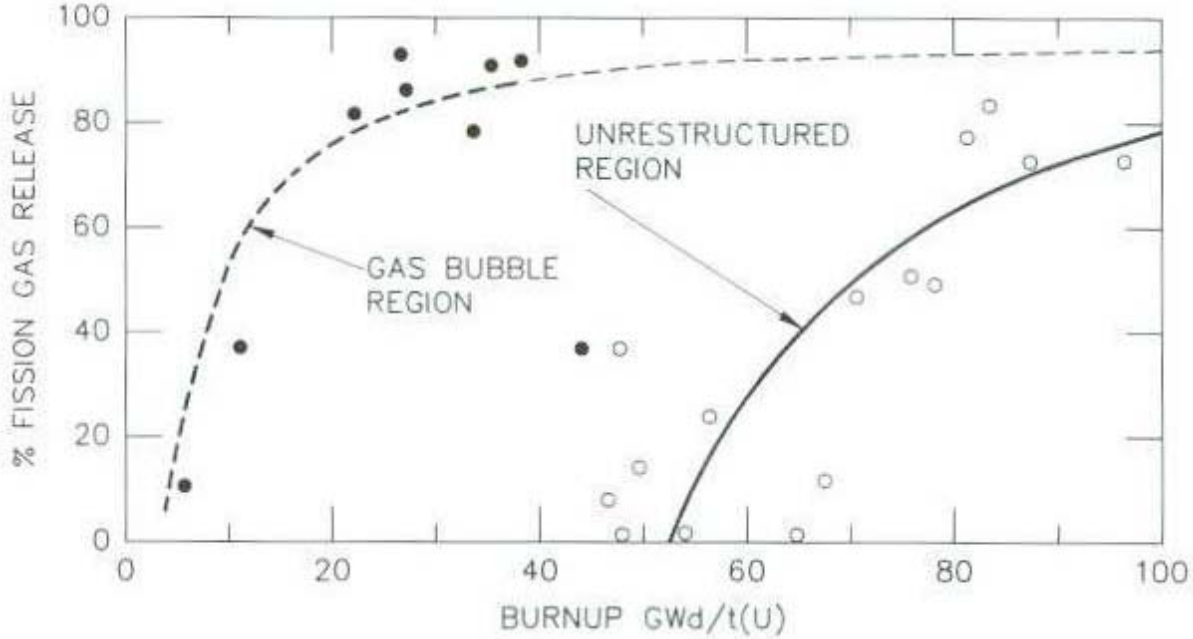
MOX Fuel Behavior—Restructuring



MOX fuel ceramography of FFTF driver fuel produced by Kerr-McGee and Babcock and Wilcox, showing restructuring as a function of burnup. **(from Hales, et al, 1986)**

MOX Fuel Behavior—Gas Release

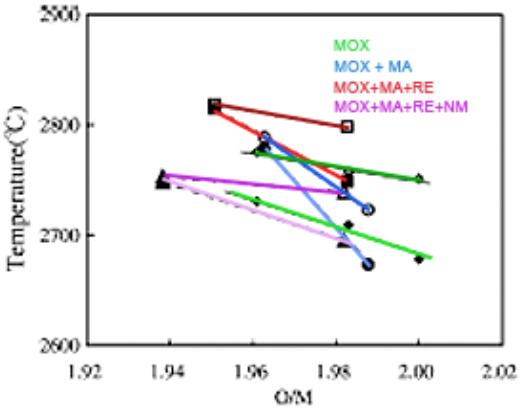
- **MOX fuel operated at high temperature and undergoing restructuring exhibits high gas release.**



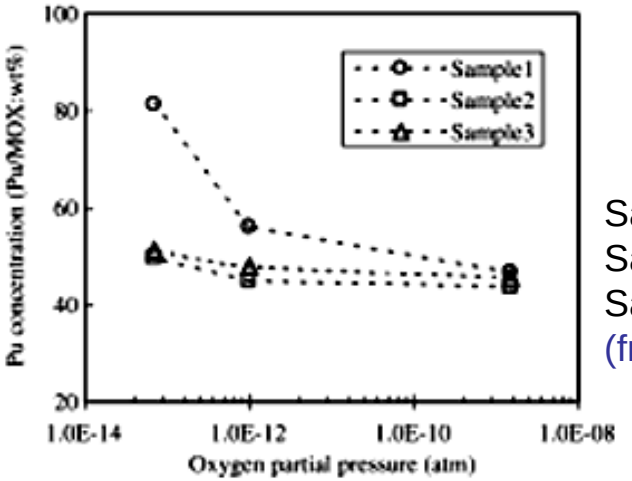
(from Lambert, et al, 1994)

MOX Fuel Behavior—FCCI

- **Hypostoichiometric MOX for SFRs**
  - As-fabricated O/M < 2.00 to suppress free oxygen at high burnup, mitigate FCCI
  - O/M ratio affects fabrication
  - O/M ratio affects properties



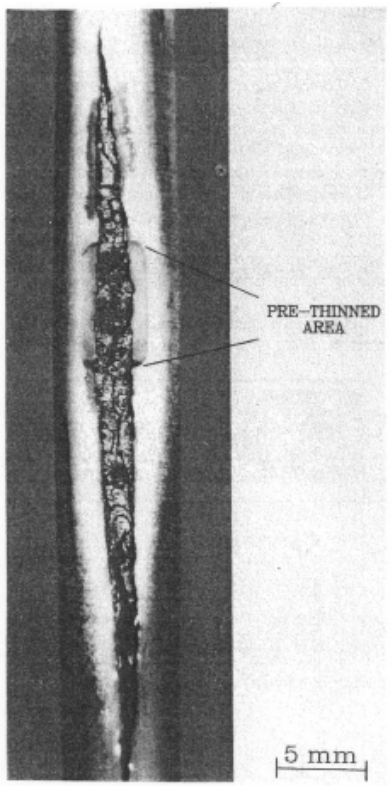
Melting T vs O/M
(from Morimoto, et al, 2005)



Sample 1 – MOX + MAs
Sample 2 – MOX+MAs+REs
Sample 3 – MOX+MAs+REs+NMs
(from Morimoto, et al, 2005)

MOX Fuel Behavior—Fuel-coolant Compatibility

- **Run-beyond-cladding-breach (RBCB) of MOX accompanied by fuel/Na reaction and initial crack extension**
- **Fuel loss can be related to degree of interaction.**
- **Reactant layer becomes coherent and inhibits further reaction with coolant.**



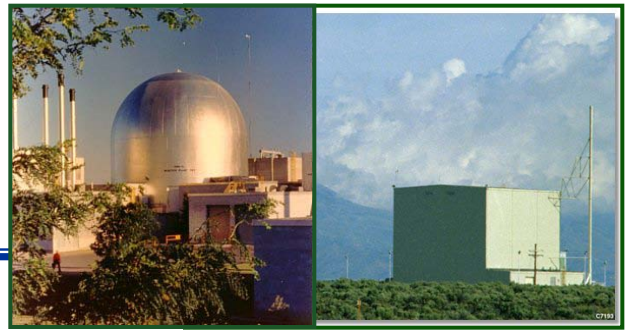
Typical breach extension in induced midlife failure, EBR-II K2B test.
(from Lambert, et al, 1990)

Base Fuel Technology: US Experience

Crawford, Porter, Hayes, **Journal of Nuclear Materials**, **371**: 202-231 (2007).

	Metallic	Mixed Oxide	Mixed Carbide
Driver Fuel Operation	≥ 120,000 U-Fs rods in 304LSS/316SS 1-8 at.% bu ~13,000 U-Zr rods in 316SS 10 at.% bu	>48,000 MOX rods in 316SS (Series I&II) 8 at.% bu	None applicable
Through Qualification	U-Zr in 316SS, D9, HT9 ≥ 10at.% bu in EBR-II & FFTF	MOX in HT9 to 15-20 at.% bu (CDE) MOX in 316SS to 10 at.% bu	None applicable
Burnup Capability & Experiments	600 U-Pu-Zr rods; D9 & HT9 to > 10 - 19 at.% in EBR-II & FFTF	4300 MOX rods in 316SS to 10 at.%; fab var's; CL melt 3000 MOX rods in EBR-II; peak at 17.5at.% bu 2377 MOX rods in D9 to 10-12 at.% bu; some at 19 at.% bu	18 EBR-II tests with 472 rods in 316SS cladding; 10 rods up to 20 at.% w/o breach 5 of which experienced 15% TOP at 12 at.% 219 rods in FFTF, incl 91 in D9, 91 with pellet & sphere-pac fuel
Safety & Operability	6 RBCB tests U-Fs & U-Pu-Zr/U-Zr(5) 6 TREAT tests U-Fs in 316SS (9rods) & U-Zr/U-Pu-Zr in D9/HT9 (6 rods)	18 RBCB tests; 30 breached rods 4 slow ramp tests 9 TREAT tests MOX in 316SS (14 rods) & HT9 (5 rods)	10 TREAT tests (10 rods; Na or He bond); ≤ 3-6 times TOP margins to breach Loss-of-Na bond test; RBCB for 100 EFPD; Centerline melting test

Transient Fuel Phenomena



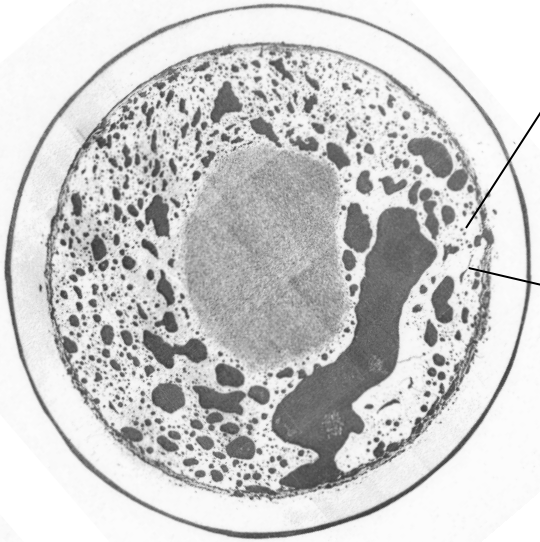
- **Metallic Fuels**
  - Pre-failure Behavior
    - *Substantial axial expansion*
    - *Cladding strain due to gas pressure*
    - *Possible fuel-cladding liquefaction*
  - Failure Behavior
    - *Failure generally near top of fuel column*
    - *Stress rupture due to gas pressure in cladding thinned by eutectic-like penetration and weakened at high temperature*
  - Post-failure Behavior
    - *Possible fuel injection into coolant*
    - *Low stored energy, no reaction with coolant, some local sodium voiding*

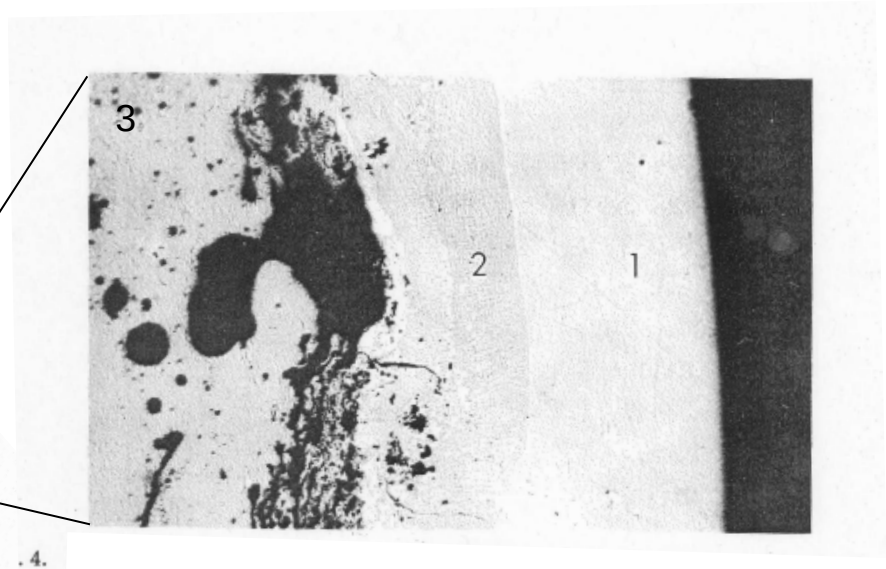
- **Oxide Fuels**
  - Pre-failure Behavior
    - *Axial relocation (apparently, upward axial motion)*
    - *Cladding strain due to FCMI and gas pressure*
  - Failure Behavior
    - *Failure generally in upper 1/3 of fuel column*
    - *Cladding melt-through with gas pressure and FCMI, cladding weakened at high temperature*
  - Post-failure Behavior
    - *Fuel dispersal into coolant*
    - *Relatively high stored energy, reaction with coolant, local sodium voiding*

Transient Phenomena—Metallic Fuels
Fuel/Cladding 'Eutectic' Formation

U-10Zr / HT9 at 800°C, 1 hr

(from H. Tsai, et al, 1990)





1 - Unaffected cladding
2 - Fuel/cladding solid-state interaction
3 - Fuel/cladding liquid phase

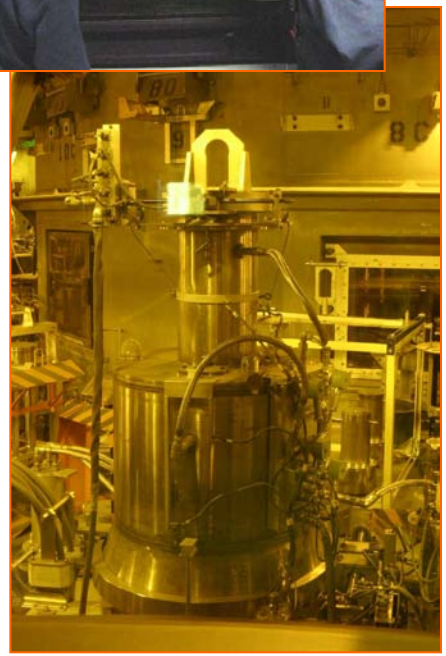
Metallic and MOX Fuels—Summary

- **Metallic Fuels (U-Zr or U-Pu-Zr)**
  - Acceptable performance and reliability demonstrated up to 10 at.% burnup, with capability demonstrated to 20 at.% burnup
  - Robust overpower capability demonstrated in TREAT tests: ~ 4 to 5x's nominal power; failures near top of fuel column; pre-failure axial expansion
  - Performance issues typically creep rupture at high burnup, accelerated due to FCCI.
  - Performance phenomena with U-Fs, U-Zr & U-Pu-Zr are the same. Burnup, temperature and cladding performance are key variables
- **MOX Fuels**
  - Acceptable performance and reliability demonstrated up to 10 at.% burnup, with capability demonstrated to 20 at.% burnup
  - Robust overpower capability demonstrated in TREAT tests: ~ 3 to 4x's nominal power; well above primary and secondary FFTF trips; failures near core mid-plane; pre-failure axial fuel motion
  - Performance issues typically creep rupture at high burnup, accelerated due to FCMI (and FCCI if O/M not controlled).
- **Metallic and MOX fuel performance in SFRs are both well known, with good experience in the US (MOX fuel in France, Japan)**

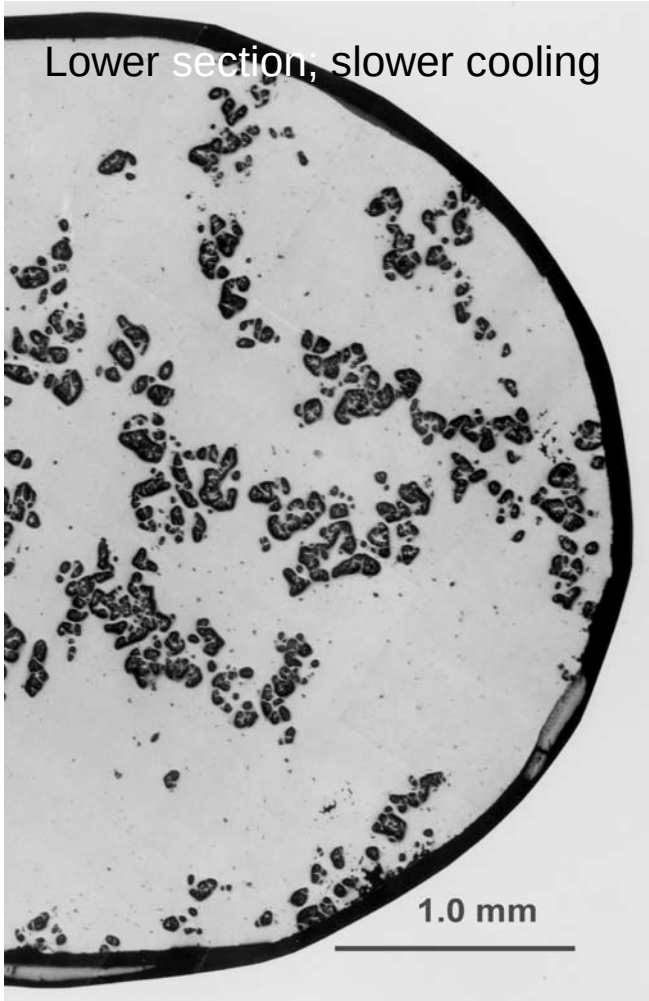
Experience with Fuels Containing Minor Actinides

SFR Transmutation Fuels with Minor Actinides (MAs) and Rare Earth (RE) Fission Products

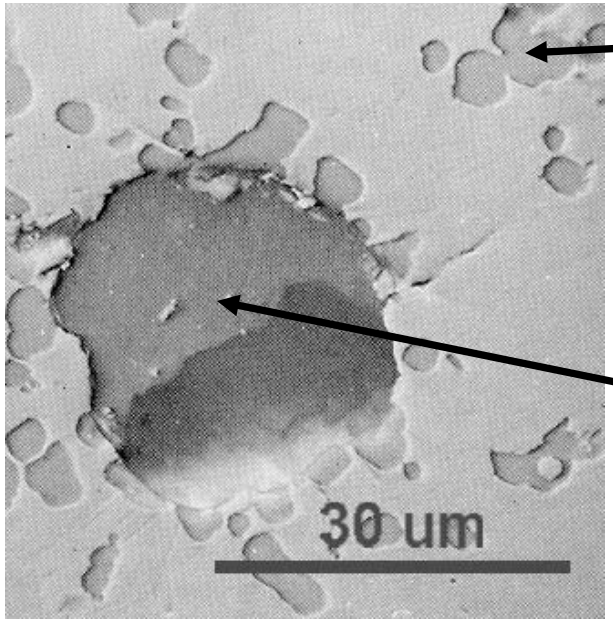
- **Unique Features of SFR Transmutation Fuels**
  - Pu content, which depending on CR selected my be higher than historic database (with corresponding decrease in U content)
  - Minor actinides (Am, Np, Cm) present in significant quantities
  - Rare earth fission product (La, Ce, Pr, Nd) carry-over from recycle step may be non-trivial
- **Gives Rise to Challenges and Unknowns**
  - Need for remote fuel fabrication
  - Likely need for new fabrication methods (e.g., due to Am volatility; waste minimization, etc.)
  - Effects on fuel performance must be determined



Metallic Fuel with MA—X501 Fabrication



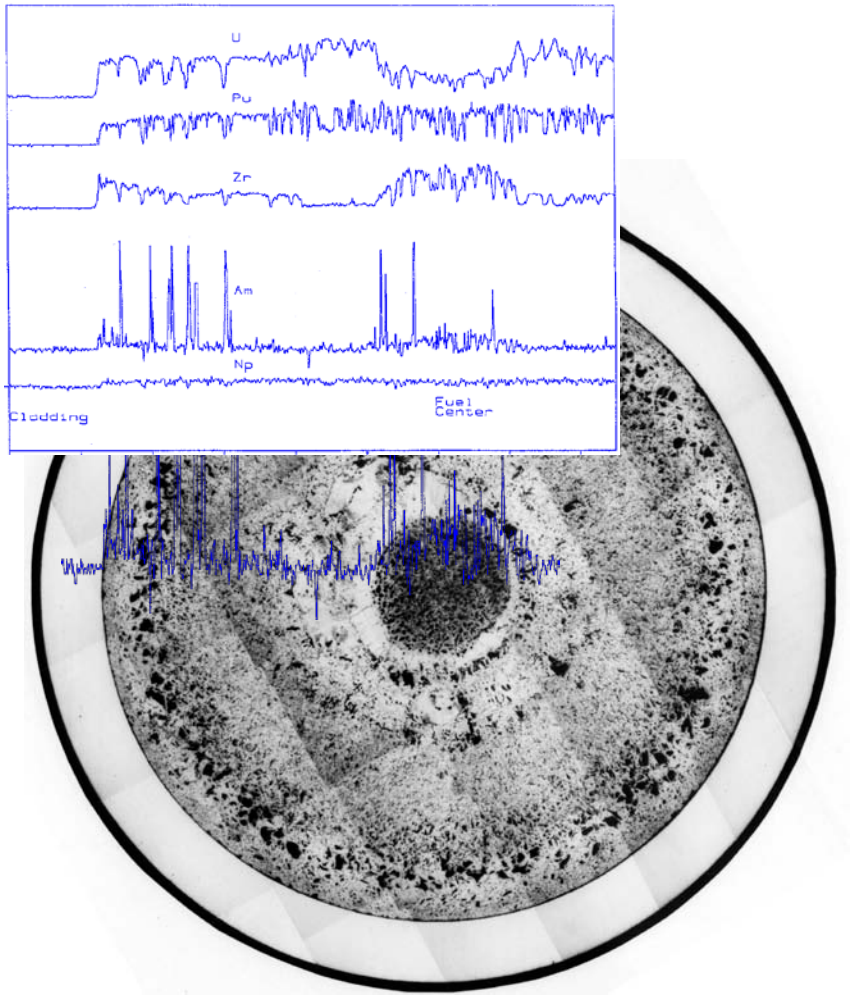
- U-20.2Pu-9.1Zr-1.2Am-1.2Np
- Injection cast at 1450°C
- Inhomogeneous microstructure
- Am and Np segregate to phases with variable composition

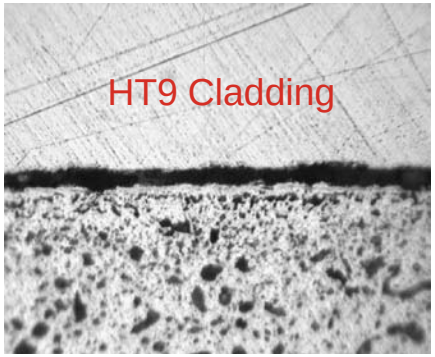


6 U
3 Pu
6 Np
86 Zr

21-47 U
14-49 Pu
9-19 Zr
0-25 Am
0-18 Np
Impurities

Metallic Fuel with MA—X501 Irradiation

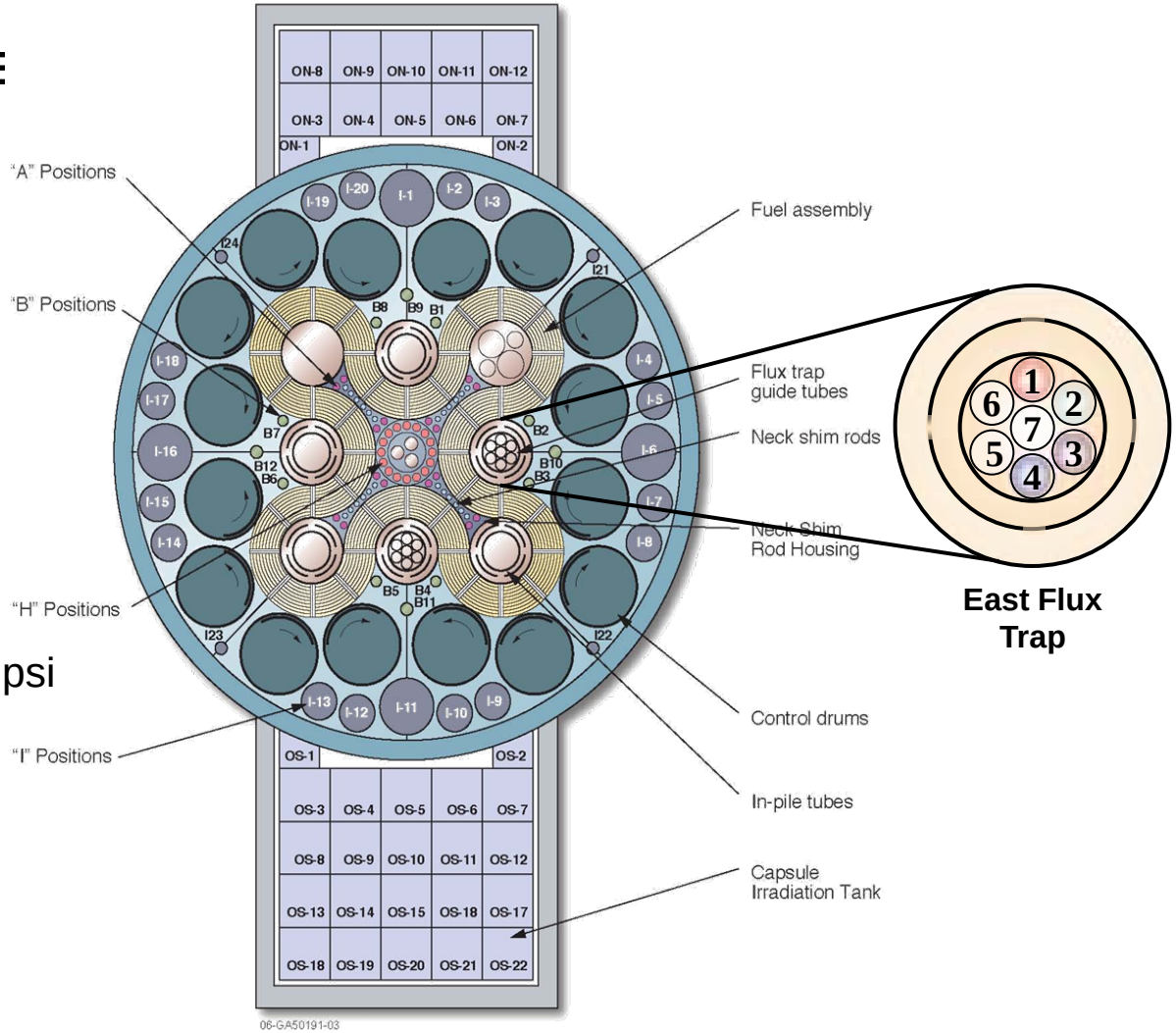




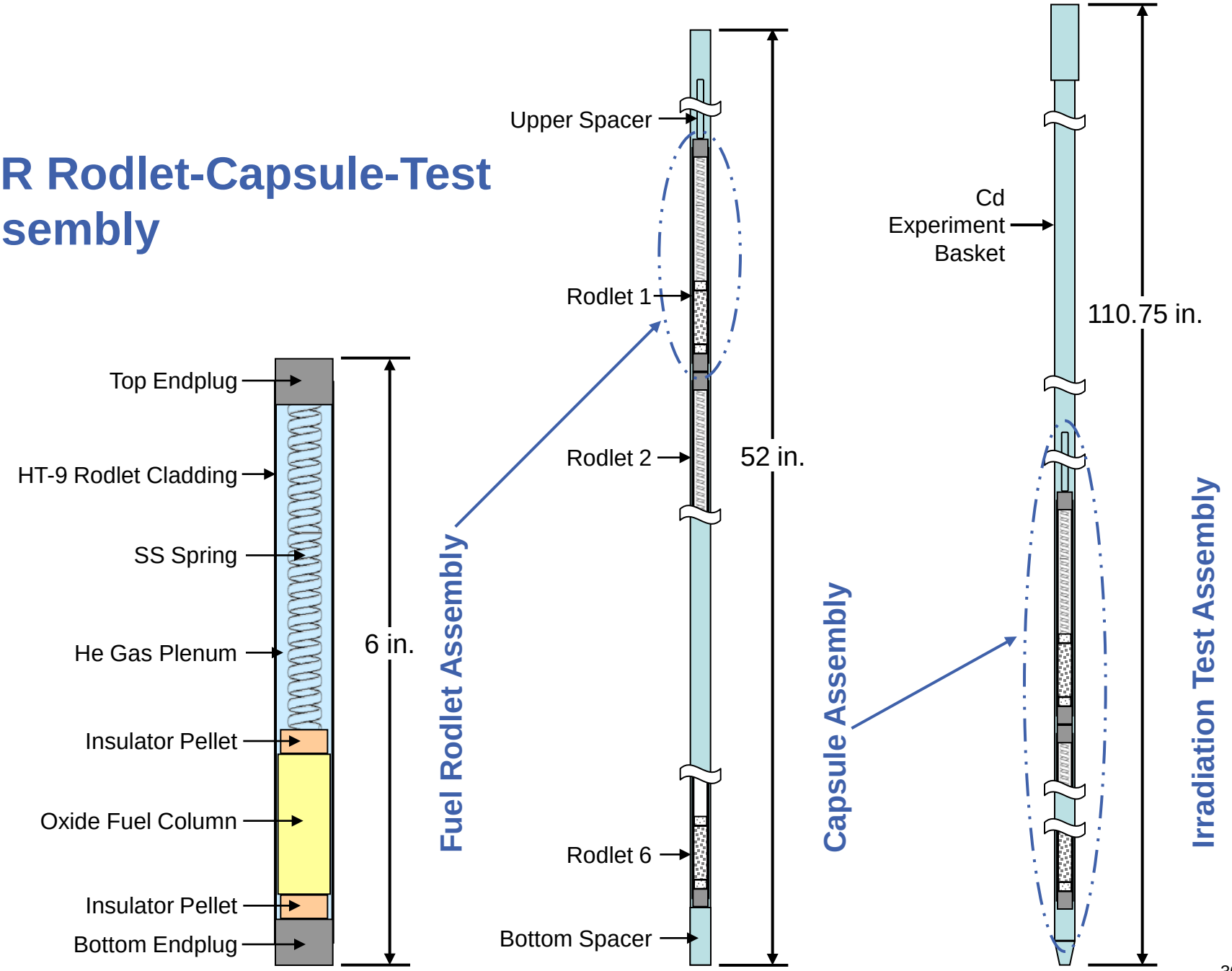
- LHGR = 450 W/cm
- PICT = 540°C
- Burnup = 7.6%
- ^{241}Am transmutation = 9.1%
- Gas Release
  - Fission gas = 79%
  - Helium = 90%

AFCI Fuels Testing in the ATR East Flux Trap

- **4 Capsule Positions in E**
  - Cd shrouds in 1,2,3,4
  - 6 rodlets per capsule
  - 24 rodlets irradiated simultaneously
- **Capsule Limits**
  - LHGR ≤ 500 W/cm
  - PICT ≤ 650°C
  - Capsule pressure ≤ 975 psi



ATR Rodlet-Capsule-Test Assembly

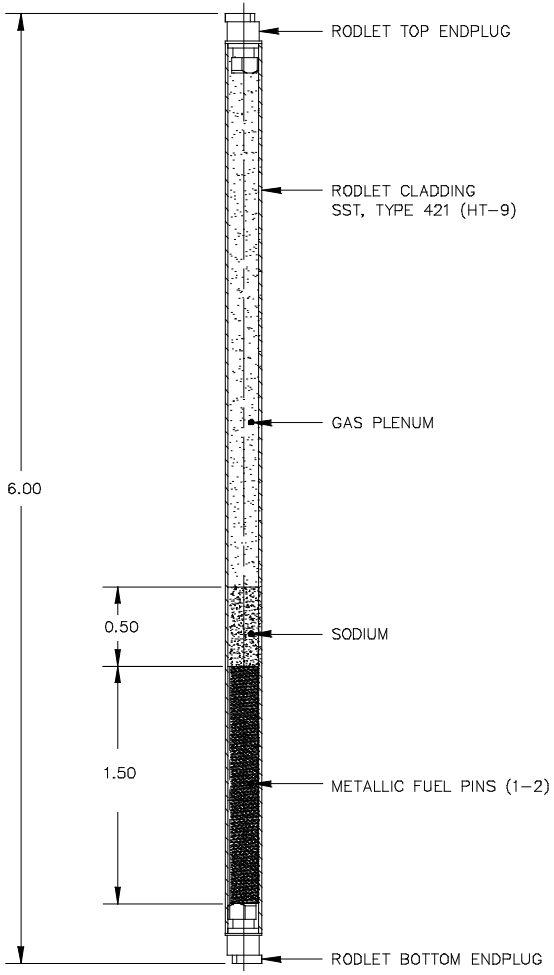


AFC-2A,B Currently Under Irradiation in the ATR

AFC-2A,B Test Matrix

Rodlet	AFC-2A&B
1	U-20Pu-3Am-2Np-15Zr
2	U-20Pu-3Am-2Np-1.0RE-15Zr
3	U-20Pu-3Am-2Np-1.5RE-15Zr
4	U-30Pu-5Am-3Np-1.5RE-20Zr
5	U-30Pu-5Am-3Np-1.0RE-20Zr
6	U-30Pu-5Am-3Np-20Zr

RE=6% La, 16% Pr, 25% Ce, 53% Nd



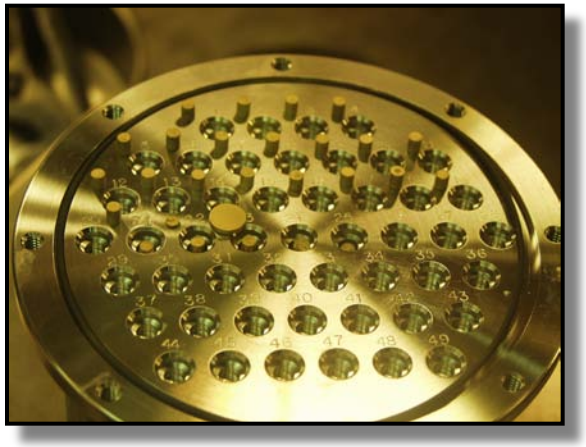
AFC-2A,B Test Objectives

- LHGR = 350 W/cm; PICT = 550°C
- Burnups of 10 at.% (2A) and 25 at.% (2B)
- Group recovery of 30 year-cooled PWR TRU
- Effect of RE fission product carry-over on FCCI

AFC-2C,D Currently Under Irradiation in the ATR

- **AFC-2C,D Test Matrix**

Rodlet	AFC-2C&D
1	$(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.95}$
2	$(U_{0.80},Pu_{0.20})O_{1.98}$
3	$(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.98}$
4	$(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.95}$
5	$(U_{0.80},Pu_{0.20})O_{1.98}$
6	$(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.98}$



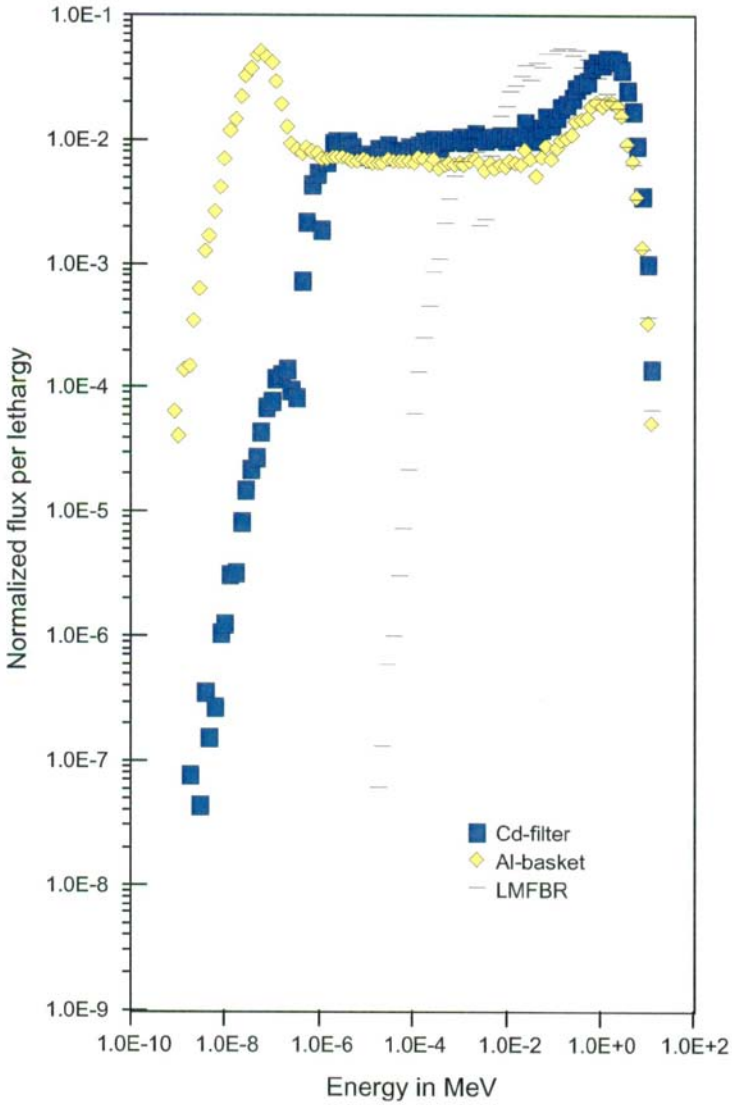
- **Test Conditions**
  - LHGR = 350 W/cm
  - PICT = 550°C
  - Group recovery of 30 year-cooled PWR TRU
- **Test Objectives**
  - Study effect of O/M on FCCI
  - Include MOX as control
  - High CR (20% Pu) for initial oxide test
  - Discharge criteria

    2C: ≥ 10 at.% burnup

    2D: ≥ 25 at.% burnup

Comparison of Spectra (ATR vs. LMFBR)

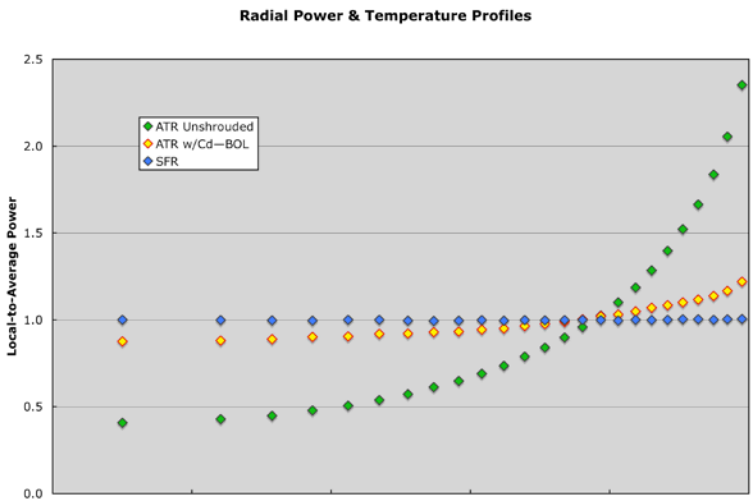
- **ATR Neutron Energy Spectrum**
  - Highly thermal spectrum in EFT with no neutron filter
  - Unaltered spectrum will result in significant self-shielding in dense, highly-enriched fuels
- **Cd-shroud Integral with Experiment Basket**
  - Efficient removal of neutrons with energies below cadmium cut-off
- **Resulting Spectrum**
  - Filtered spectrum in experiment does not have prototypic fast neutron component
  - Epi-thermal component responsible for most fissions; much more penetrating than thermal neutrons
  - Test fuels are free of gross self-shielding



Radial Flux Depression and Temperature Profiles in Test Fuels

- **How prototypic are AFC rodlets irradiated in the ATR?**
  - Assessed by analysis
  - Radial power profiles calculated w/MCNP
  - Depletion in fuel and Cd shroud calculated w/ORIGEN (MCWO)
  - 1-D thermal analysis using radial powers

- **Resulting temperatures for AFC-2C,D oxide rodlets**
  - 3 cases: SFR, unshrouded ATR, ATR w/Cd shroud
  - w/Cd shroud, peak-to-avg power at fuel periphery is 1.22; fuel central temperature 58°C less than SFR (~400°C less for unshrouded case)

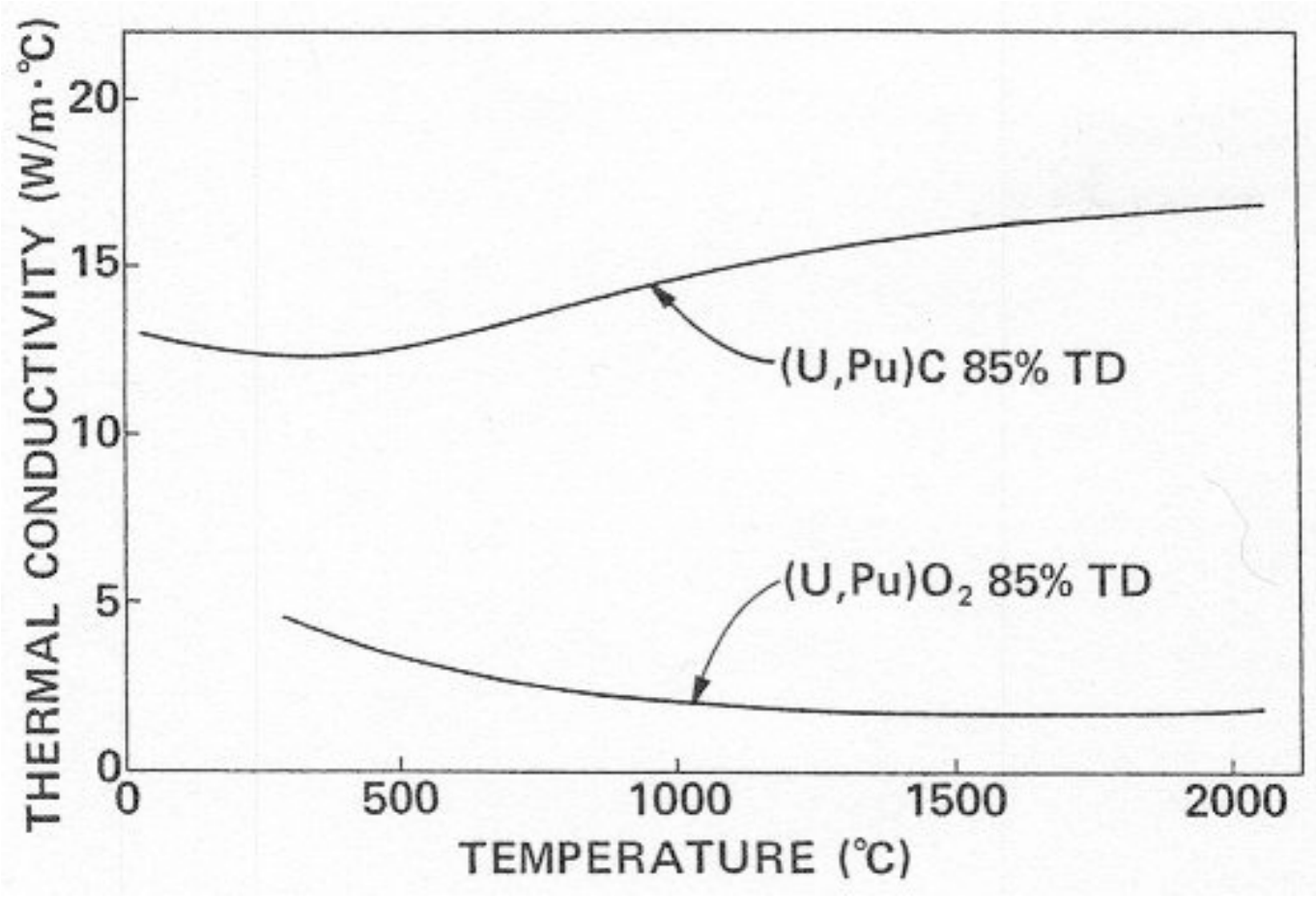


Experience with Carbides and Nitrides

U.S. Carbide/Nitride Fuel Experience

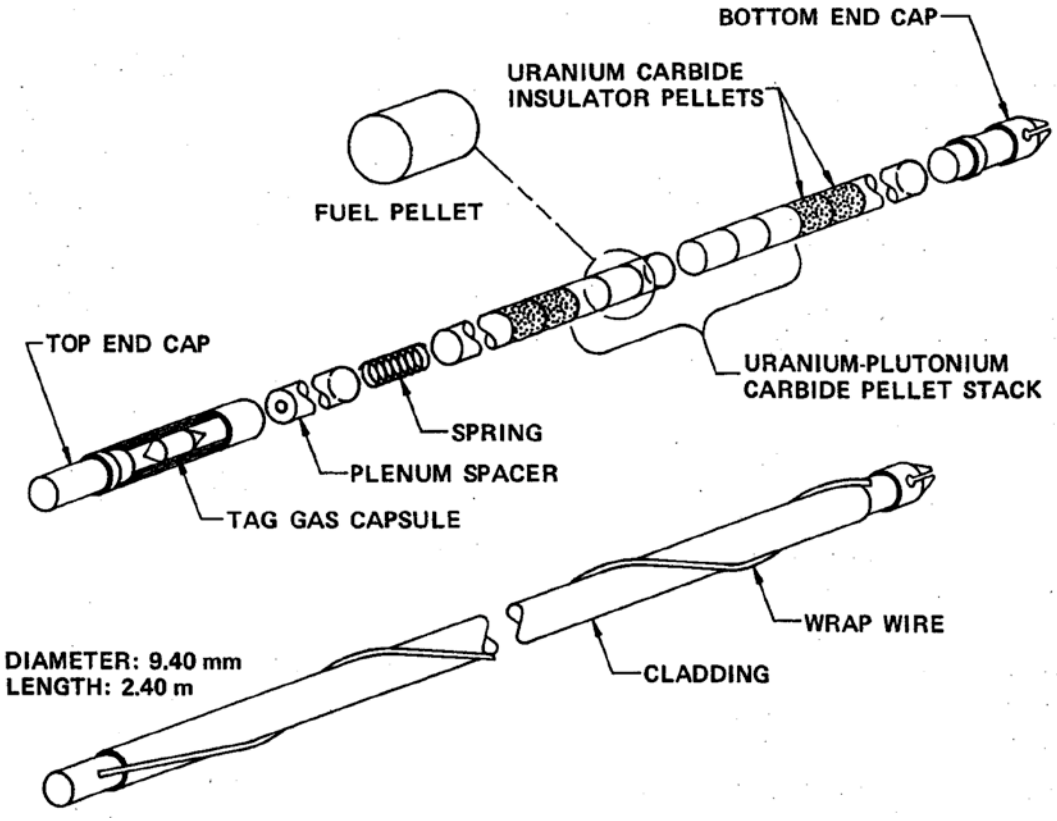
- **EBR-II Testing: ~470 (U,Pu)C rods, ~100 (U,Pu)N rods**
- **FFTF: ~200 (UC-PuC) rods –pellets and sphere-pac, >50 UN rods**
- **AAA/AFCI: (U,Pu,MA)N, (Zr,Pu,MA)N – ATR and Phenix tests**
- **GenIV GFR: (U,Pu)C Dispersion**
- **ROVER: (U, Zr)C in graphite**
- **SP-100: UN**
- **JIMO: UN**
- **Other: UN, UC, UC-UC_2, BISO, TRISO**

Carbide Thermal Conductivity



Matthews, R.B. and R.J. Herbst, Uranium-Plutonium Carbide Fuel for Fast Breeder Reactors. Nuclear Technology, 1983. **63: p. 9-22.**

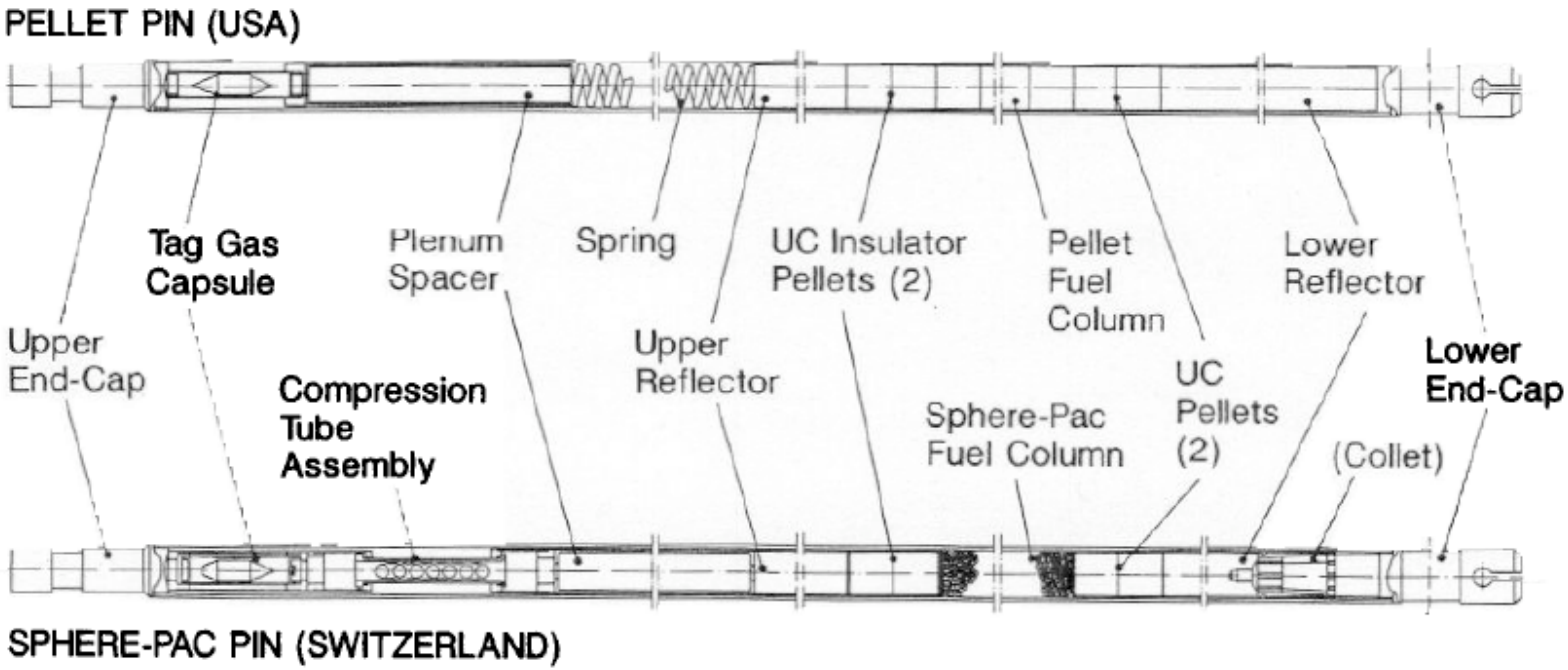
(U, Pu)C_x Fuel Design (FFTF)



FFTF He-bonded Carbide Fuel

Matthews, R.B. and R.J. Herbst, Uranium-Plutonium Carbide Fuel for Fast Breeder Reactors. Nuclear Technology, 1983. **63: p. 9-22.**

Sphere pac alternative to traditional pellet design



Stratton, R.W., et al., Fabrication Processes, Design and Experimental Conditions of the Joint U.S.-Swiss Mixed Carbide Test in FFTF (AC-3 Test). Journal of Nuclear Materials, 1993. **204: p. 39-49.**

Challenge 1: Fabrication

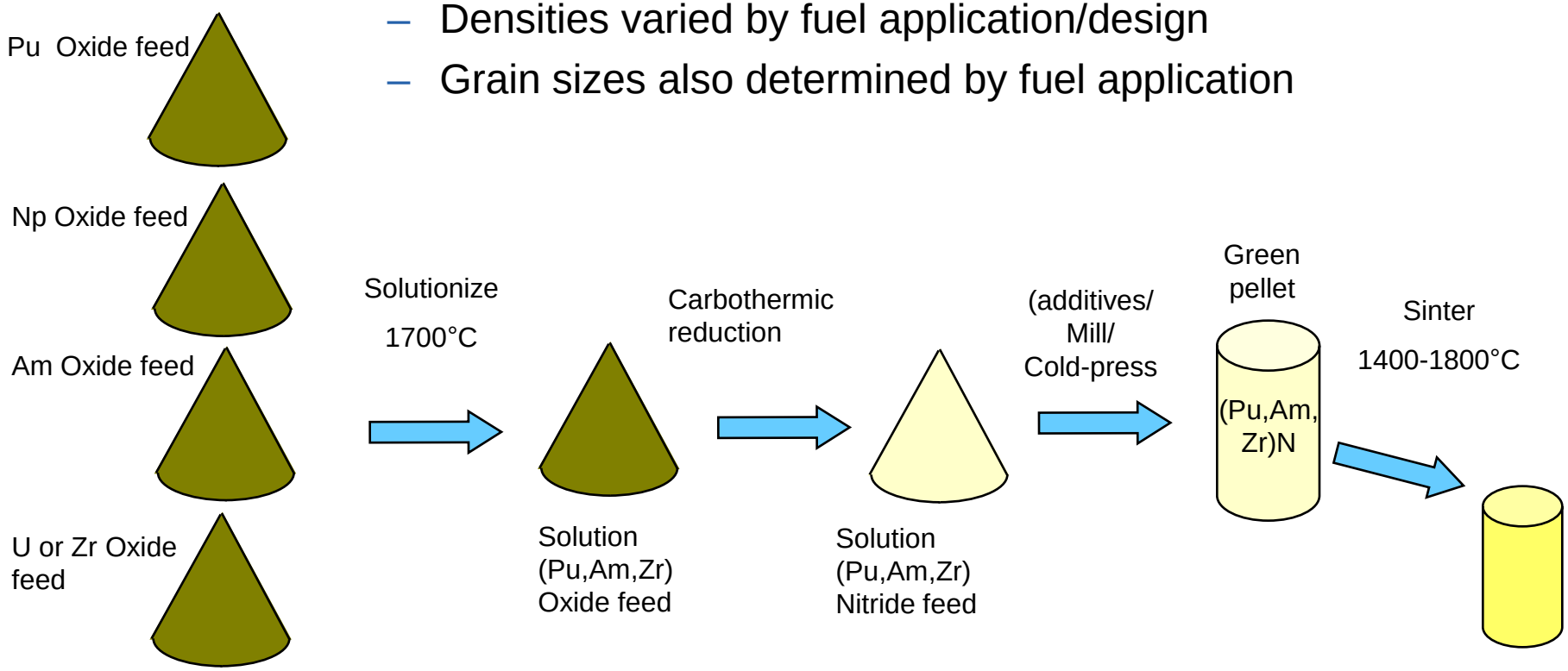
- **Carbides**
  - Casting (UC)
    - *Arc casting*
    - *Skull-melting*
  - Reaction sintering (UC)
    - *$UO_2 + 3C \rightarrow UC + 2CO$ (+U+C)*
  - Direct carborization (UC)
    - *$U + C \rightarrow UC$*
  - Carbothermic reduction (U,Pu)C
    - *$UO_2 + 3C \rightarrow UC + 2CO$*
  - Microspheres (U,Pu)C
    - *Sol-gel*
    - *Electrode atomization*

- **Nitrides**
  - Direct nitration
    - *UN, PuN*
  - Hydride-dehydride-nitride
    - *UN, PuN*
  - Low Temperature
    - *UN via fluoride/ammonia*
  - Carbothermic reduction
    - *(U,Pu,MA)N, (Zr,Pu,MA)N*

Typical Nitride Feed Synthesis and Pellet fabrication with the minor actinides

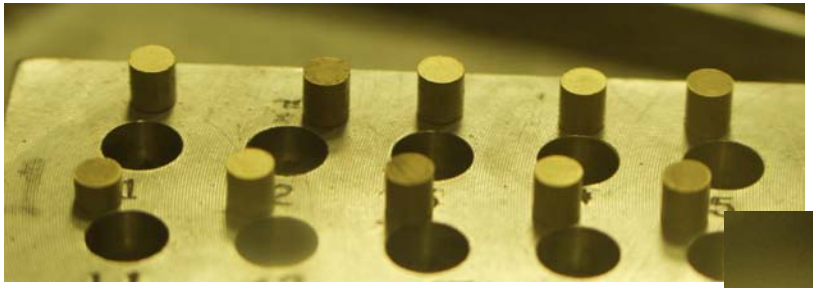
Pellet fabrication typically by dry process using cold press/sinter approach

- Densities varied by fuel application/design
- Grain sizes also determined by fuel application



Examples of fabrications for recent actinide bearing nitride fuels.

AFC-1G Pellets

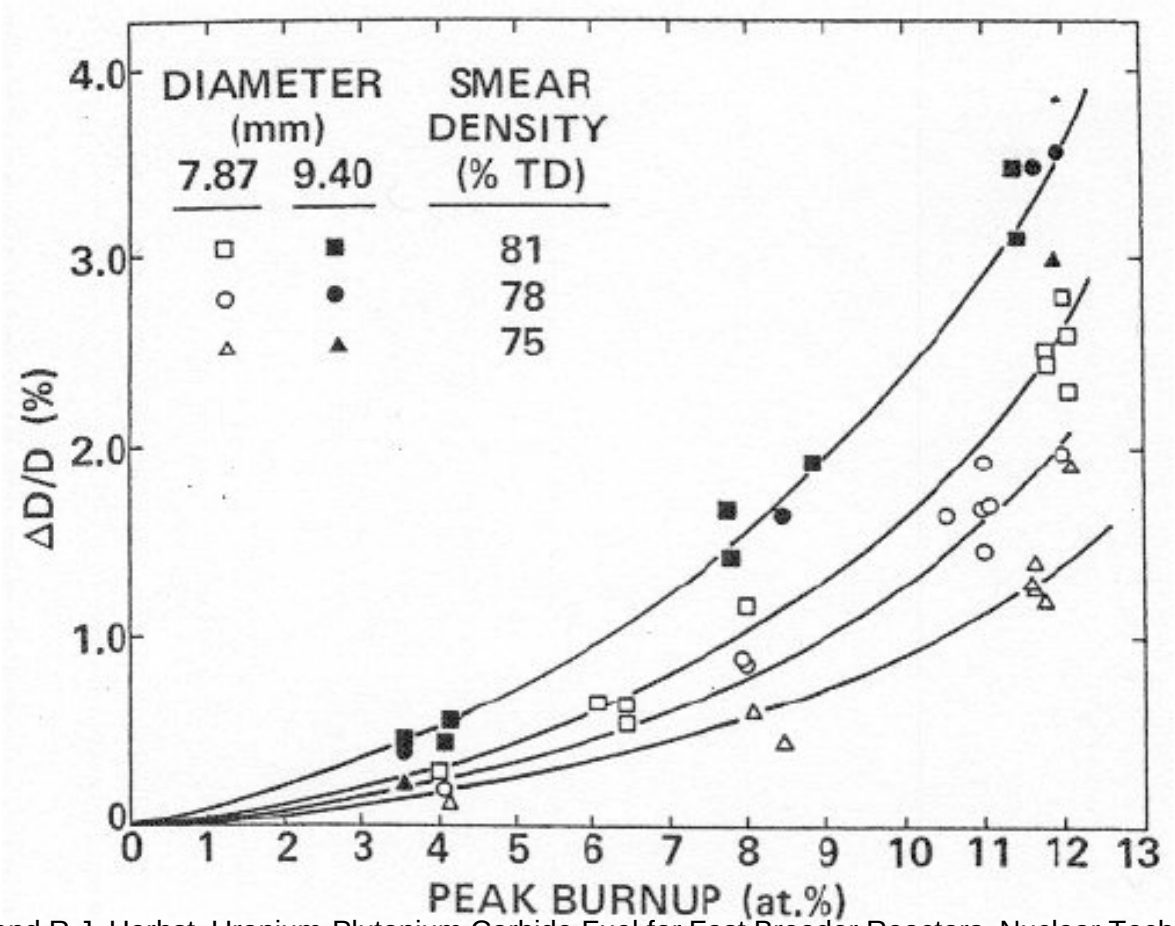


Non-Fertile:
$(Pu_{0.5}Am_{0.5})N$-36ZrN

Low-Fertile:
$(U_{0.5}Pu_{0.25}Am_{0.15}Np_{0.10})N$

Challenge 2: Swelling in carbides

Maximum cladding strain as a function of peak burnup for helium-bonded carbide fuel pins.



Matthews, R.B. and R.J. Herbst, Uranium-Plutonium Carbide Fuel for Fast Breeder Reactors. Nuclear Technology, 1983. **63: p. 9-22.**

Challenge 2 for nitrides in fast spectrums

Natural nitrogen: 99.6% ^{14}N and 0.4% ^{15}N

- generation of the radiotoxic long life ^{14}C (which has to be managed in spent fuel or during recycling) from $^{14}N(n,p)$ reaction,
- production of ^{11}B (which provides a neutronic penalty)
- production of He (which may lead to a larger fuel swelling and pin pressurisation) from $^{14}N(n,a)$ reaction.

^{15}N Enrichment Processes

NO/HNO_3 chemical exchange

$$^{15}NO_{gas} + H\,^{14}NO_{3aq} \xrightleftharpoons{} {}^{14}NO_{gas} + H\,^{15}NO_{3aq}$$

Cryogenic distillation

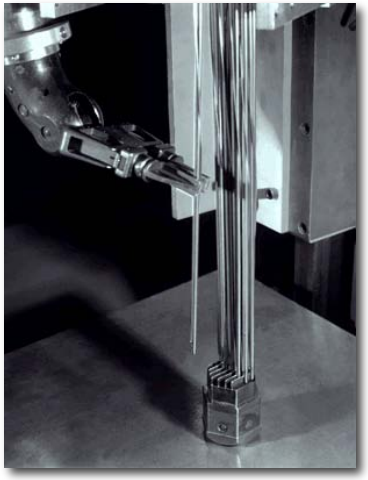
$$^{14}NO_{liq} + {}^{15}NO_{gas} \xrightleftharpoons{} {}^{15}NO_{liq} + {}^{14}NO_{gas}$$

Chromatographic separation

$$^{14}NH_4^+R^- + {}^{15}NH_4OH_{aq} \xrightleftharpoons{} {}^{15}NH_4^+R^- + {}^{14}NH_4OH_{aq}$$

Stainless-Steel Cladding & Duct Performance

- **Performance Issues**
  - Cladding dilation
  - Duct dilation, bowing, or twisting
- **Irradiation Behavior**
  - Void swelling (AS)
  - Irradiation creep (AS & FMS)
  - Irradiation embrittlement (AS & FMS)
- **Alloys to Address Issues**
  - Advanced austenitic stainless steels
  - Ferritic & tempered-martensitic stainless steels
  - Oxide-dispersion strengthened steel alloys



Summary

- **SFR Fuels Experience in the US**
  - Fuel Types
  - Fuel Performance Issues
  - Experience/Testing
- **Experience with Fuels Containing Minor Actinides**
- **Carbide and Nitride fuels are considered attractive relative to oxides, but there are several materials challenges that must be better understood and addressed.**