GEN IV International Forum℠

METALLIC FUELS FOR FAST REACTORS

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August 22, 2017

NEA Nuclear Energy Agency

cea

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INL Idaho National Laboratory

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NPS PRAESTANTIA PER SCIENTIAM

Meet the presenter

Dr. Steven Hayes is a Fellow of the Nuclear Science & Technology Directorate at Idaho National Laboratory. During his career, he has been engaged in the development, testing and modeling of a variety of nuclear fuels, including metallic, oxide, and nitride fuels for liquid metal reactors and high-density dispersion fuels for research reactors. He led numerous fuels and materials irradiation experiments in the Experimental Breeder Reactor II prior to its shutdown, and today he maintains an active fuel testing program in the Advanced Test Reactor. Dr. Hayes is a national leader in the development and testing of metallic fuels for the US-DOE’s Advanced Fuels Campaign and in the development of multiscale, multiphysics fuel performance codes for the US-DOE’s Nuclear Energy Advanced Modeling and Simulation program.

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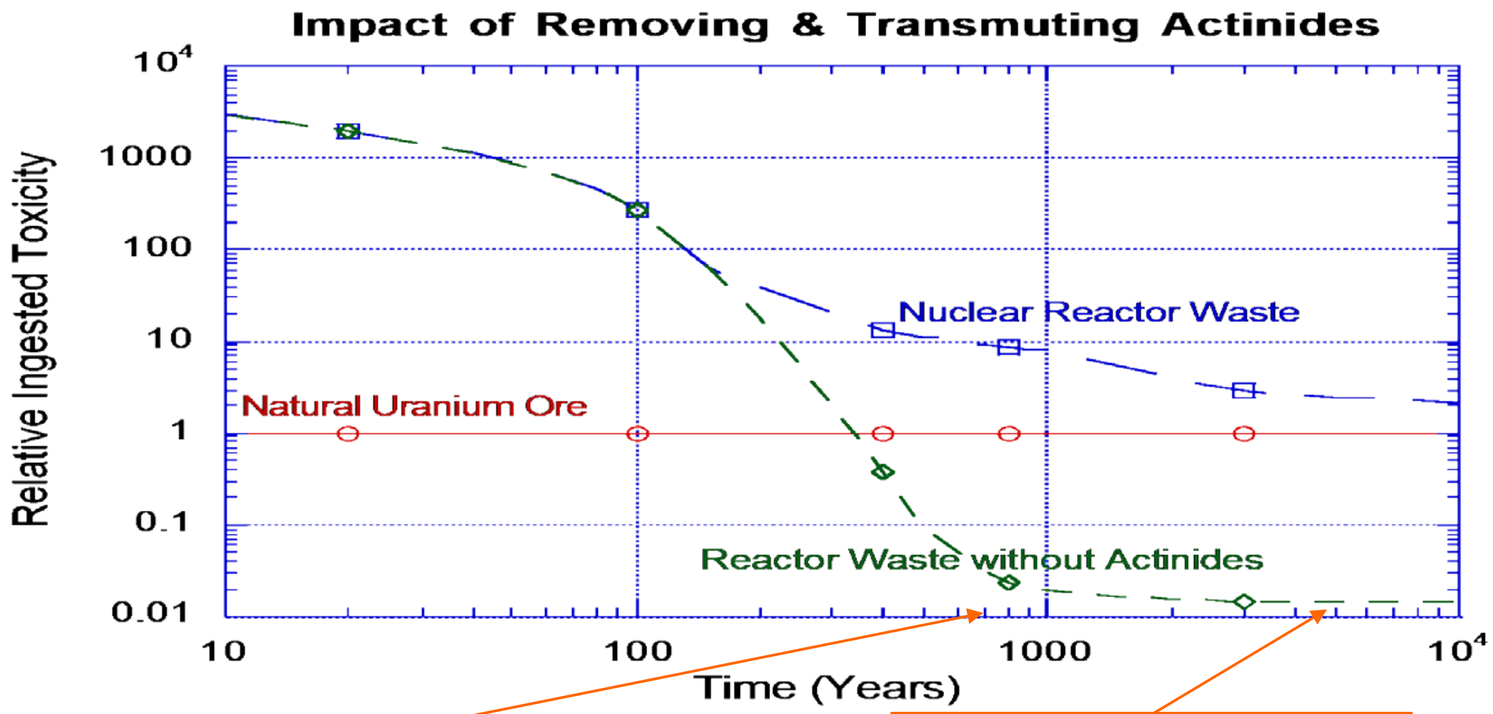
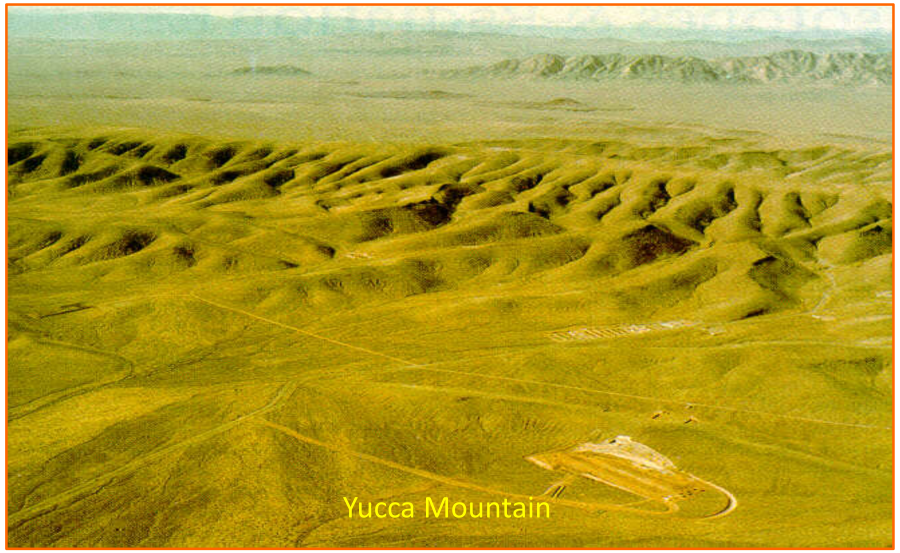
Outline of Presentation

- Background: Motivation for Actinide Transmutation
- Metallic Fuels: History & Benefits
- Casting Process Development
- Irradiation Performance
- Future Directions: Innovative “Advanced Metallic Fuel Concept”
- Summary & Conclusions

Background

Motivation for Actinide Transmutation

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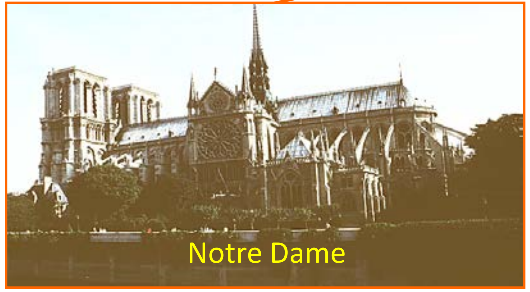
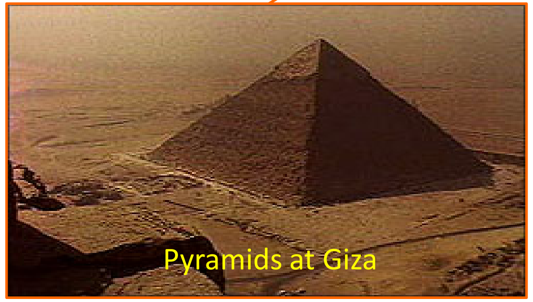

Notre Dame


Pyramids at Giza

- Plutonium and minor actinides are responsible for most of repository hazard beyond ~400 years
- Many examples of man-made structures > 400 years old

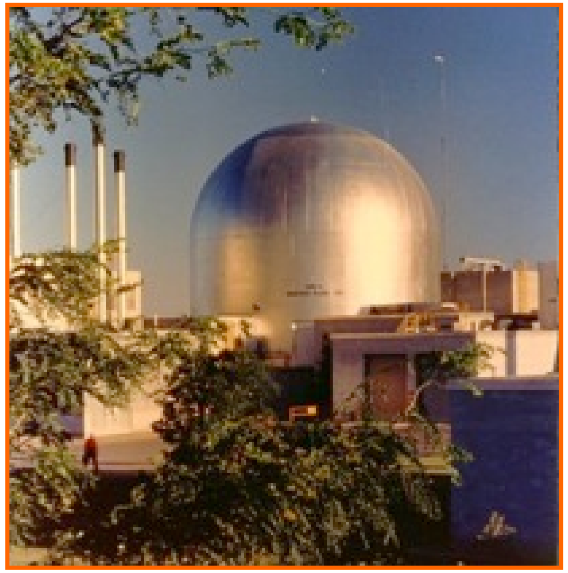
No examples > 10,000 years old

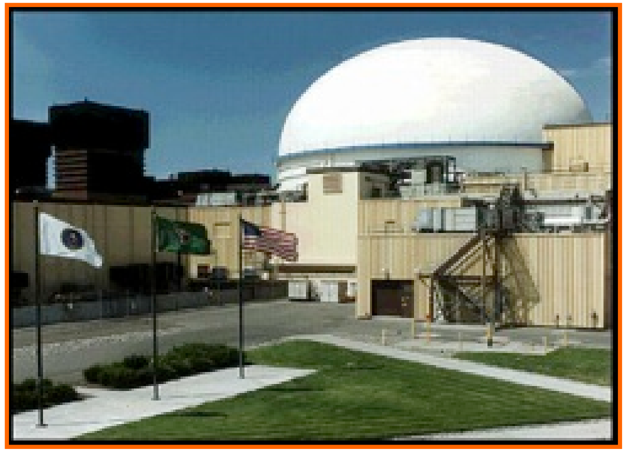
Actinide Management in Fast Reactors

- **Fast Reactors Key to an Actinide Transmutation Mission**
  - Large number of excess neutrons
  - Neutrons of high energy
  - Allow for variety of actinide management strategies
    - High actinide loading and throughput

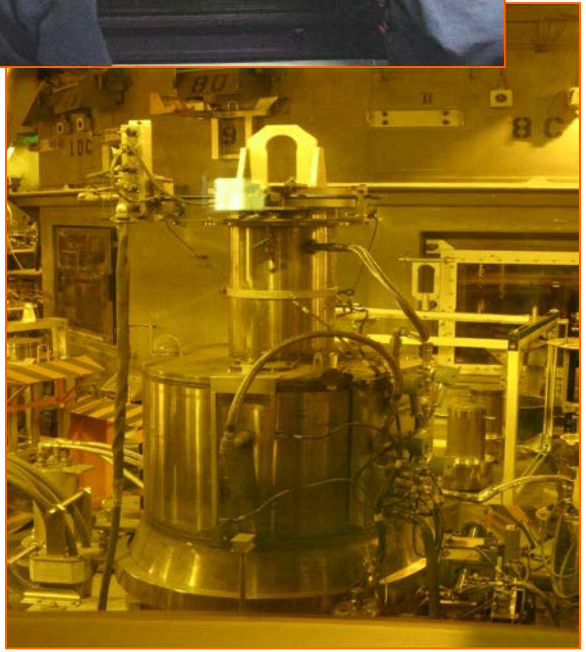
  ① Resource extension (i.e., breeding), and/or

  ② Waste management (i.e., actinide burning)





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

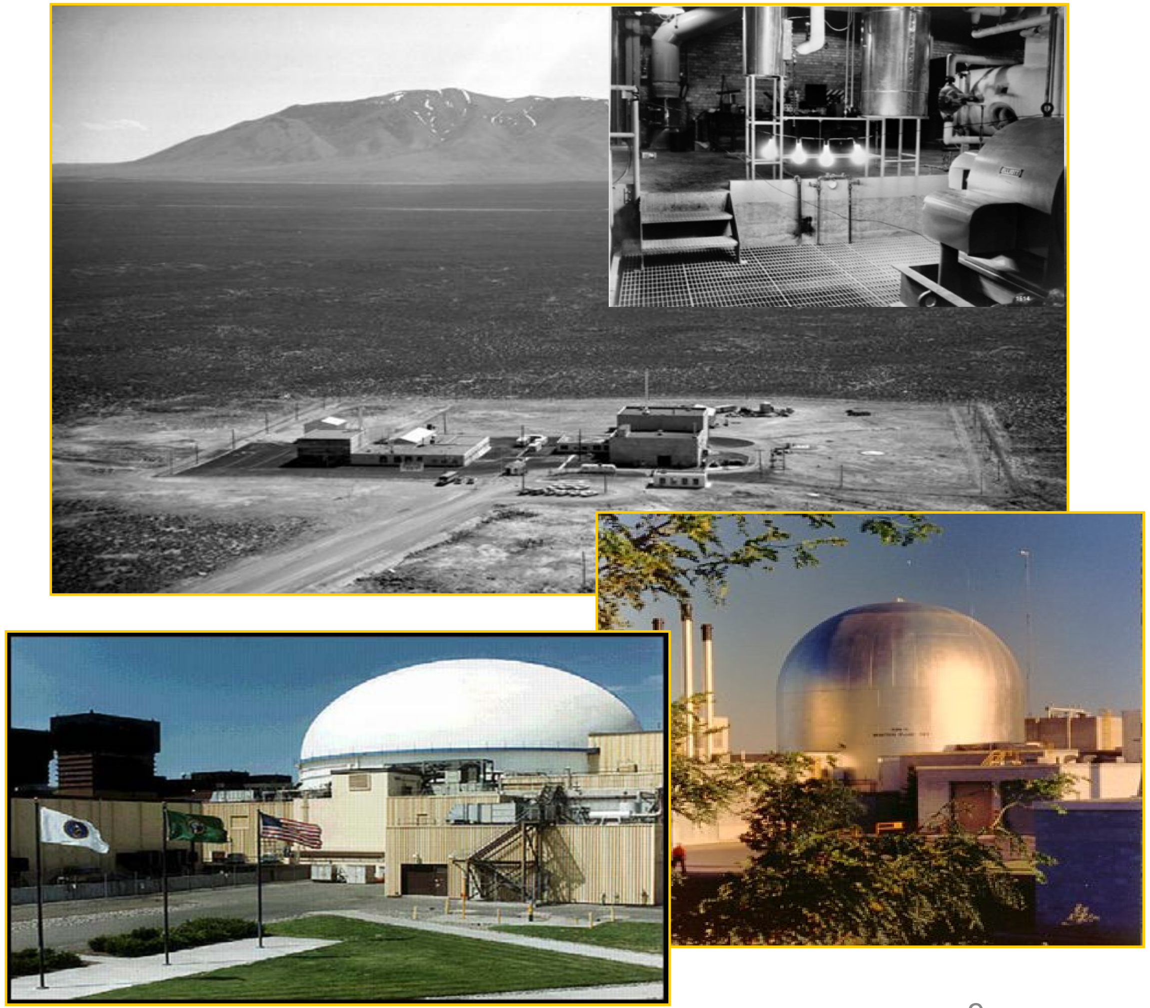


- **Unique Features of SFR Transmutation Fuels**
  - Pu content, which depending on CR selected my be higher than historic database (with corresponding decrease in U content)
  - Minor actinides (Am, Np, Cm) present in significant quantities
  - Rare earth fission product (La, Pr, Ce, Nd) carry-over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

Metallic Fuels: History & Benefits

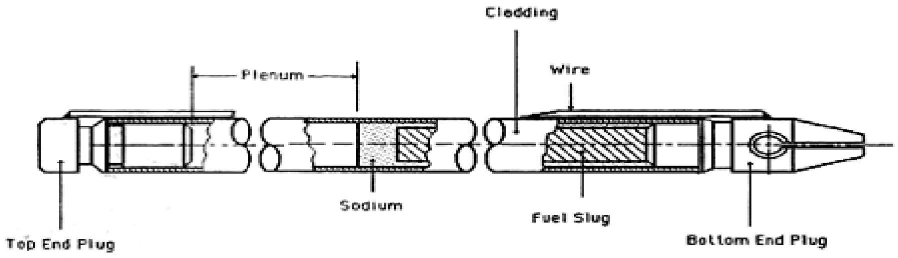
History of Metallic Fuels in Fast Reactors

- **EBR-I** (1951)
  - Unalloyed U
  - U-2Zr
  - Pu-1.25Al
- **UK Dounreay Fast Reactor** (1963)
  - U-0.1Cr
  - U-7Mo
  - U-9Mo
- **Enrico Fermi FBR** (1963)
  - U-10Mo
- **EBR-II** (1964)
  - U-5Fs
  - U-10Zr
  - U-20Pu-10Zr
- **FFTF** (1982)
  - Assembly testing of U-10Zr
  - Assembly testing of U-20Pu-10Zr



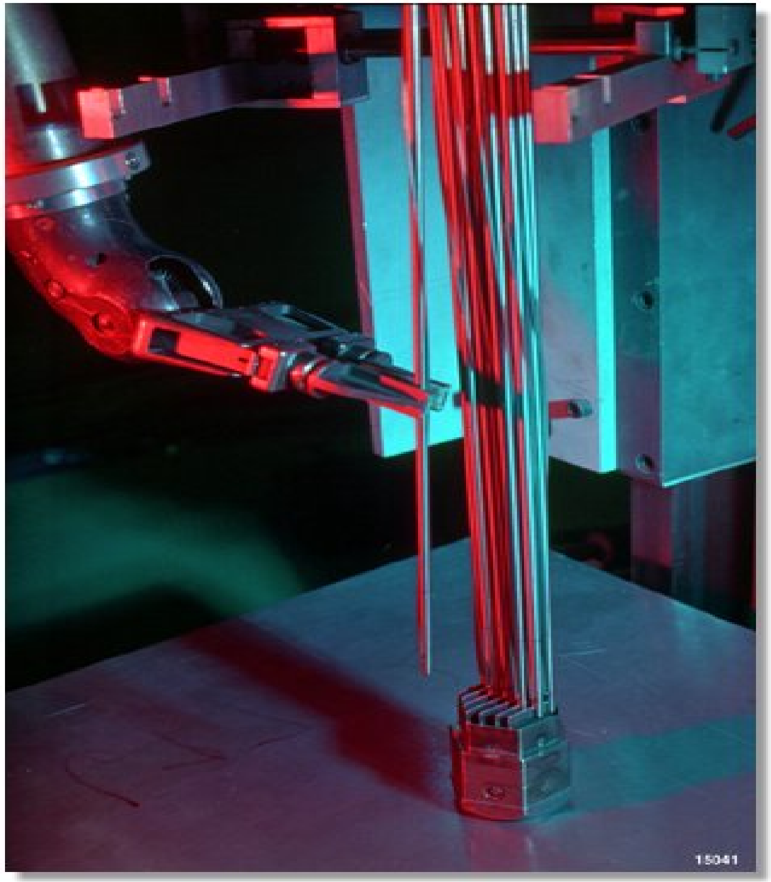
Key Features of Metallic Fuels

- **Metal fuel characteristics**
  - U-Pu-Zr alloy base (good irradiation stability)
  - 75% smeared density (accommodate fuel swelling, mitigate FCMI)
  - Large fission gas plenum (accommodate high gas release)
  - Na bond in fuel-cladding gap (keep fuel temperatures low)
  - Low-swelling FMS cladding (minimize cladding/duct dimensional changes)



- **Historic benefits**
  - Outstanding fuel reliability to high burnup (~20 at.-%)
  - Compatibility with proliferation-resistant electrochemical recycle
  - Simple, compact (demonstrated remote) fabrication processes
  - Synergistic with passive approach to reactor safety

Inherent Benefits of Metallic Fuels for Actinide Transmutation



- **Fabrication**
  - Historic ease of fabrication on large scales, remote environments
  - Process not sensitive to fuel composition (exception: Am)
  - Na-bonding allows for:
    - Loose tolerances on fuel diameters
    - Large fuel pin thermal margins
- **Irradiation Performance**
  - Consistent over wide range of compositions
  - Large fuel swelling/high gas release accommodated by design
  - Compatible with sodium coolant
  - Demonstrated high-burnup reliability; lower-density alloys for transmutation offer even higher burnup potential

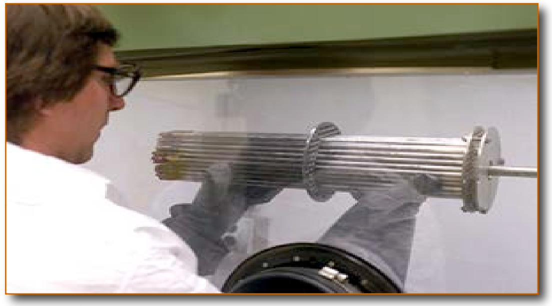
Casting Process Development

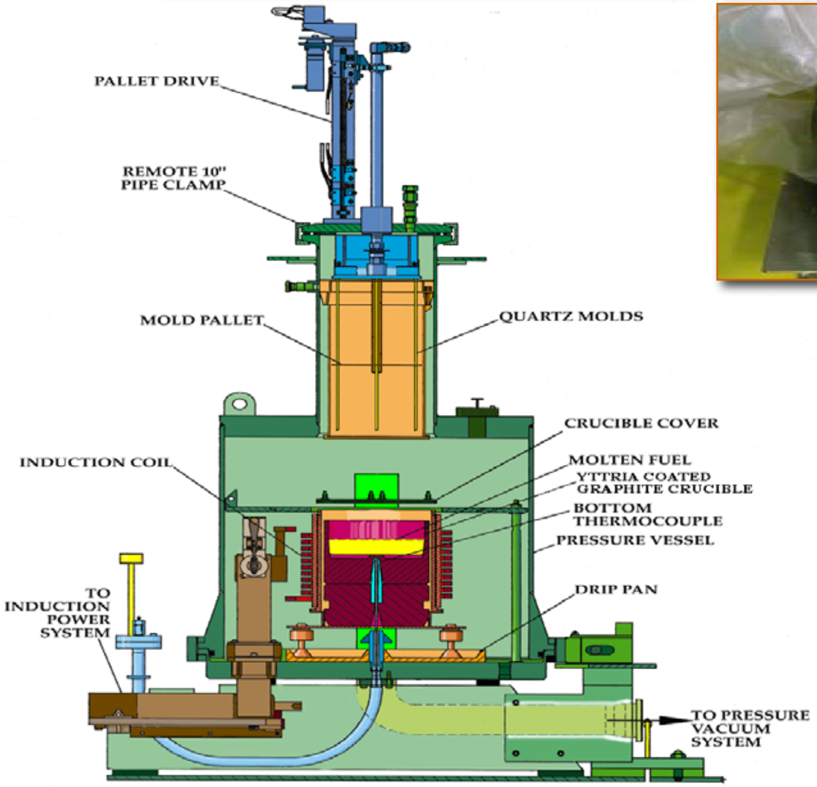
Traditional Metallic Fuel Fabrication: Injection Casting (counter-gravity)

Demonstrated Injection Casting Method

1. Fuel is melted and stirred in Y_2O_3 wash-coated graphite crucible
2. Furnace is evacuated and ZrO_2 wash-coated SiO_2 molds are submerged
3. Pulse pressurization of vessel rapidly injects and freezes fuel in molds
4. Molds are removed and shattered to release fuel slugs
5. Crucible is cleaned by wire brush and recoated

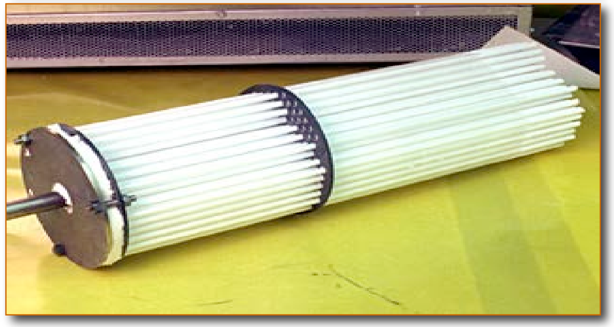
Used to remotely fabricate 39,000 metallic fuel pins for EBR-II over a 3-year period in 1960's.



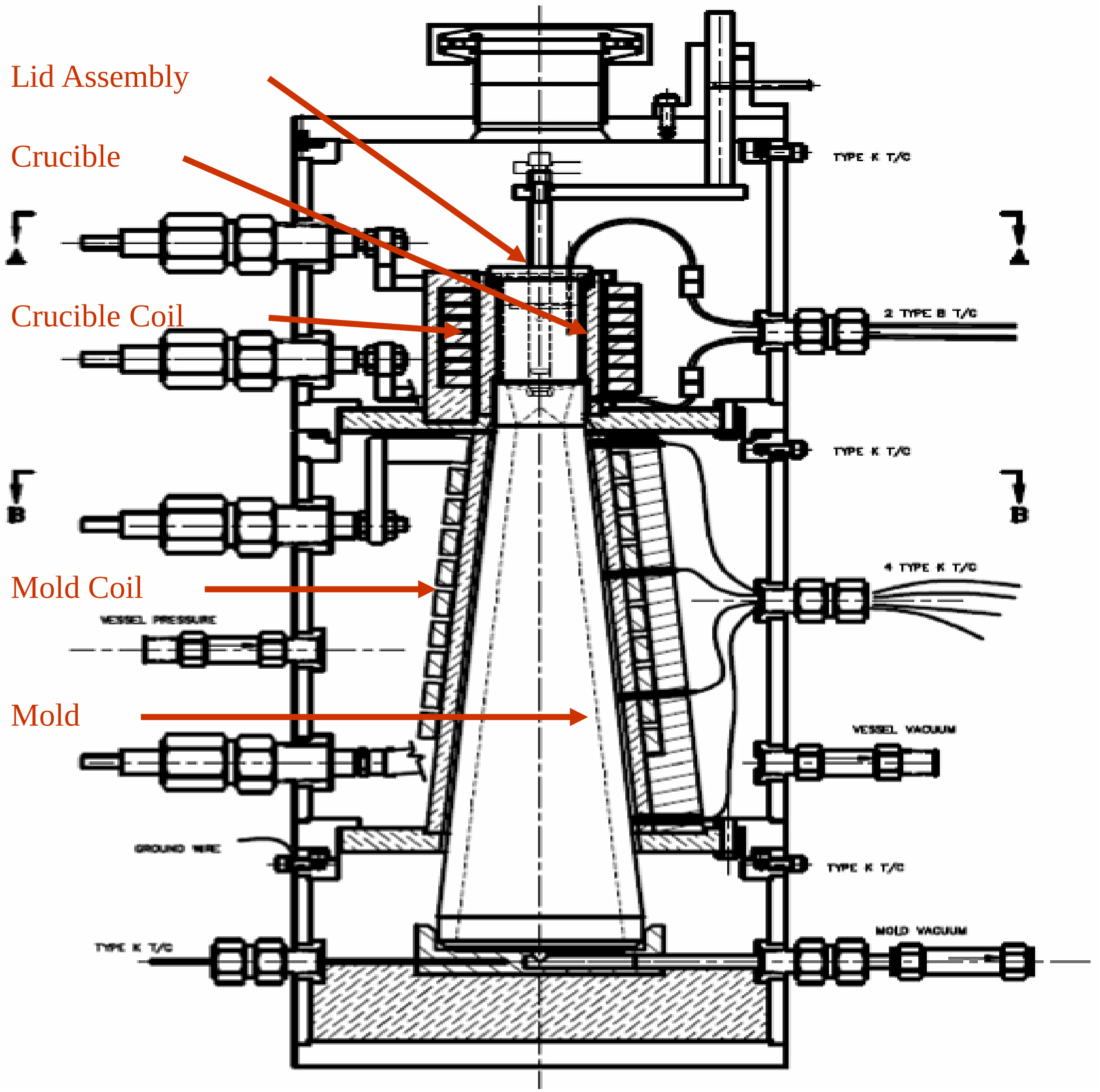


Issues for Metallic Transmutation Fuels using Traditional Casting

- **Residual fuel heel and slug end crops result in only ~33% utilization of melted charge**
- **Fuel losses**
  - Y_2O_3 crucible coating (0.2-0.3%)
  - SiO_2 mold pieces (2-5%, reduced to <0.5% w/ magnetic separation process)
  - Loss of volatile constituents (i.e., Am)
- **High level wastes**
  - Graphite crucible and Y_2O_3 coating
  - SiO_2 mold pieces
- **Crucible cleaning and coating**
  - Accomplished by operators at window with manipulators
  - Dust from crucible cleaning drives hot cell contamination levels



Development of New Casting Process



- **Rapid cycle time, advanced crucible and mold materials**
  - Minimize fuel losses to coatings and single-use molds
  - Minimize high level wastes
  - Eliminate crucible cleaning and coating
  - Minimize contamination or reaction of melt
- **Bottom casting process**
  - Increase charge utilization (up to 100%) and throughput
  - Eliminate volatile loss mechanism
    - Melt pool covered
    - Not exposed to vacuum

Casting Process Development

- **Minor actinide bearing fuel fabrication**
  1) Bottom pour crucible configuration
  2) Casting under pressure

Charge Material	Mass	Wt%
Uranium	80.64	80.9%
Zirconium	3.77	10.0%
Plutonium	7.36	7.4%
Americium	1.66	1.7%
Zirconium	6.20	
Total	99.63	100%

Casting Product	Mass	Wt% (of total charge)
Cast Material	71.215	71.5%
Casting Heel	26.14	26.2%
Dross	2.231	2.2%
Total	99.589	99.96%

1) Greatly improved melt utilization, and
2) Near-zero Am loss during fabrication.

Performance of Metallic Fuels with MAs

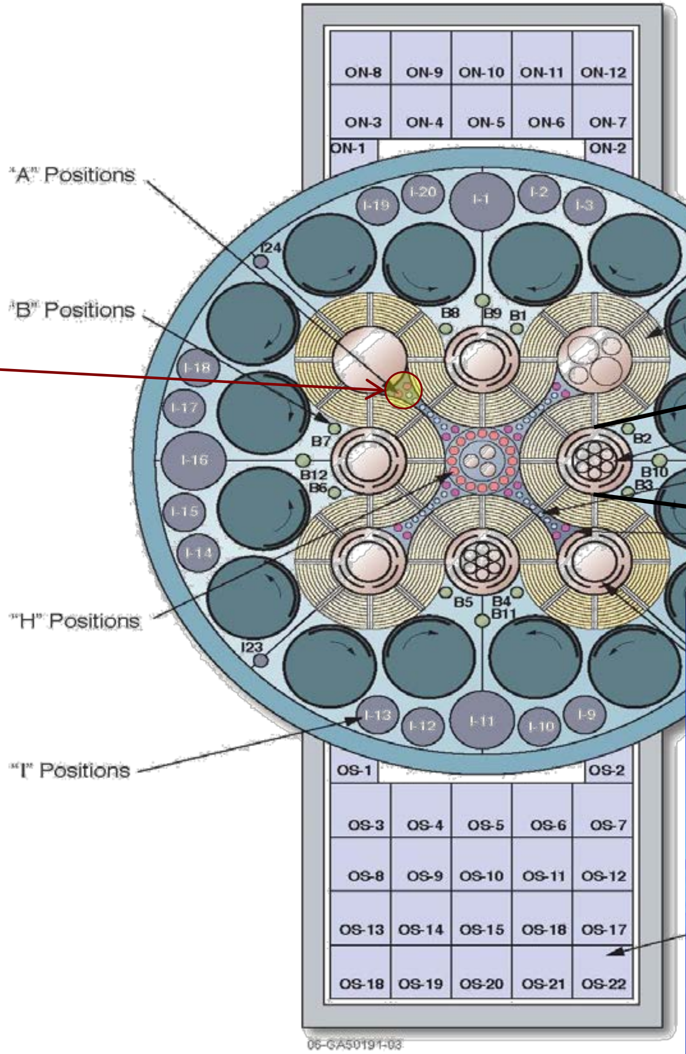
Irradiation Testing Program in the ATR

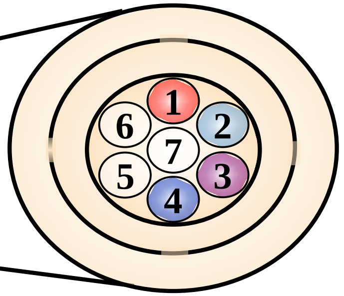
	AFC-1	AFC-2	AFC-3/4	IRT
Test Strategy	Scoping – Many compositions	Scoping – Focused compositions	Focused compositions	Focused compositions
	Nominal conditions	Nominal conditions	Nominal+ conditions	Nominal+ conditions
Capsule Type	Drop-in	Drop-in	Drop-in	Drop-in
Fuel Types	Metallic Nitrides	Metallic Oxides	Advanced Metallic Concepts	Metallic
Key Features	Baseline + MA	Baseline + MA + RE	FP control, annular fuel, FCCI barriers, ultra-high burnup	Recycle feed Remote fabrication
Time Frame	FY 2003 – FY 2008	FY 2008 – FY 2012	FY 2011 – FY 2017 +	FY 2018 – 2020

Past test series   Test series in progress   Future test series

AFC Series Irradiation Testing in the ATR

- **AFC-1,2 Testing in EFT**
  - Cd shrouds in 1, 2, 3, 4
  - 6 rodlets per capsule
  - Up to 24 rodlets irradiated simultaneously
- **AFC-3,4 Testing in 4 OA's**
  - Cd shrouds in up to 4 OA's
  - 5 rodlets individually encapsulated per OA
  - Up to 20 rodlets irradiated simultaneously; cycle-by-cycle shuffling of rodlets
- **Capsule Limits**
  - LHGR ≤ 500 W/cm
  - PICT ≤ 650°C
  - Capsule pressure ≤ 975 psi

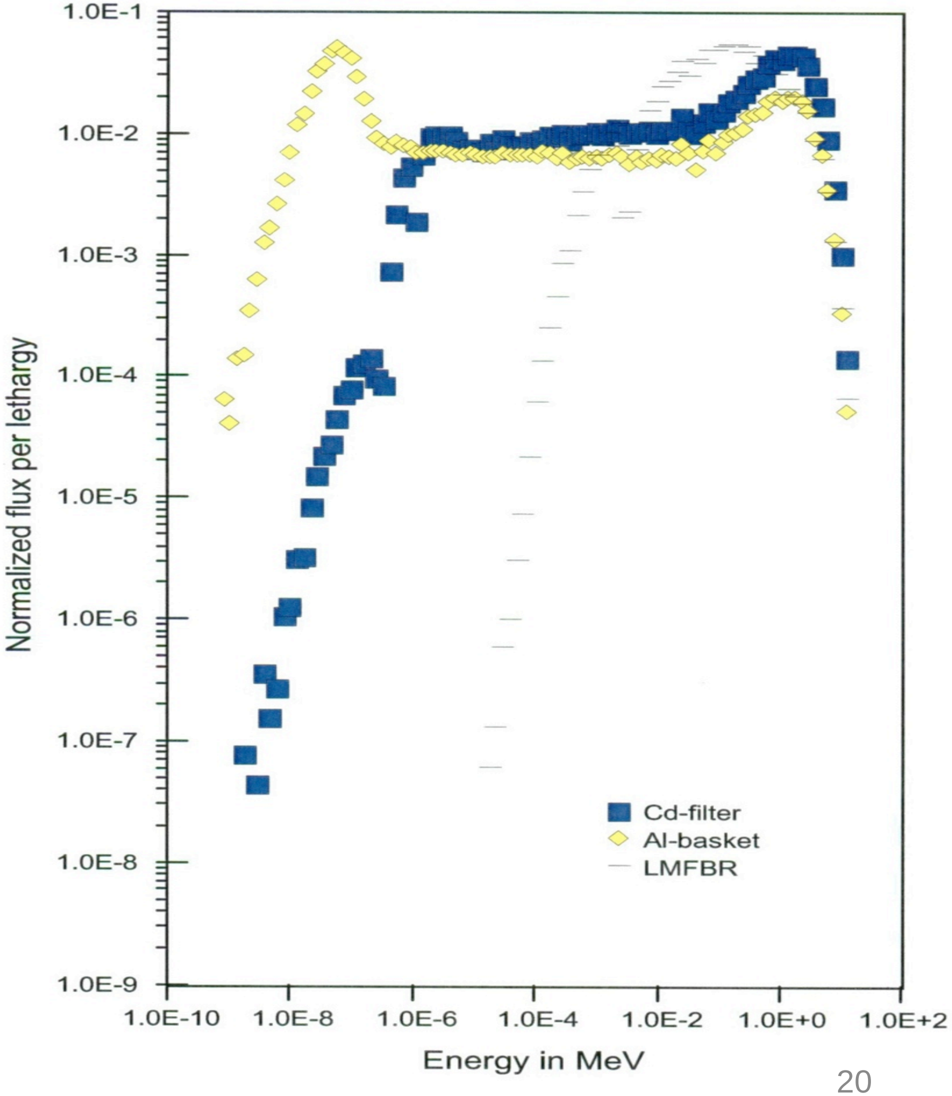




East Flux Trap

Comparison of Spectra (ATR vs. SFR)

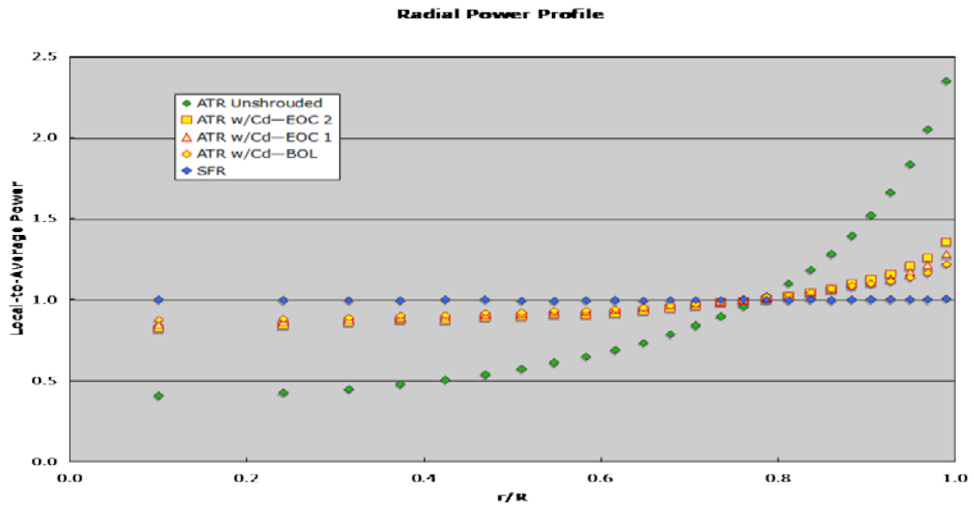
- **ATR Neutron Energy Spectrum**
  - Highly thermal spectrum in EFT with no neutron filter
  - Unaltered spectrum will result in significant self-shielding in dense, highly-enriched fuels
- **Cd-shroud Integral with Experiment Basket**
  - Efficient removal (>99%) of neutrons with energies below cadmium 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

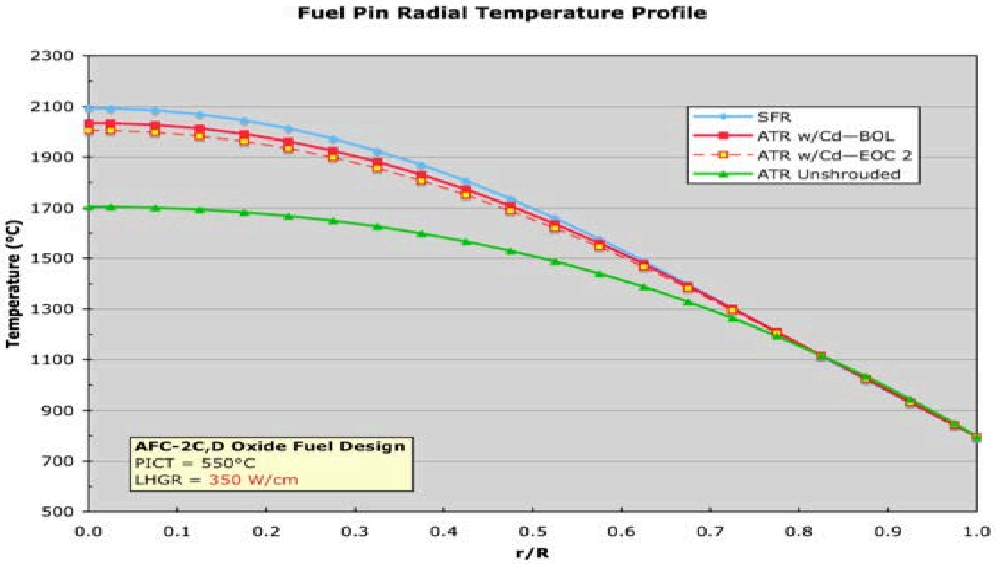


Radial Flux Depression and Temperature Profile in Test Fuels

- **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)
  - Thermal analysis using radial powers

- **Resulting temperatures for AFC-2C,D mixed-oxide rodlets**
  - <u>3 cases</u>: 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)
  - Temperature profile near-prototypic of fast reactor conditions





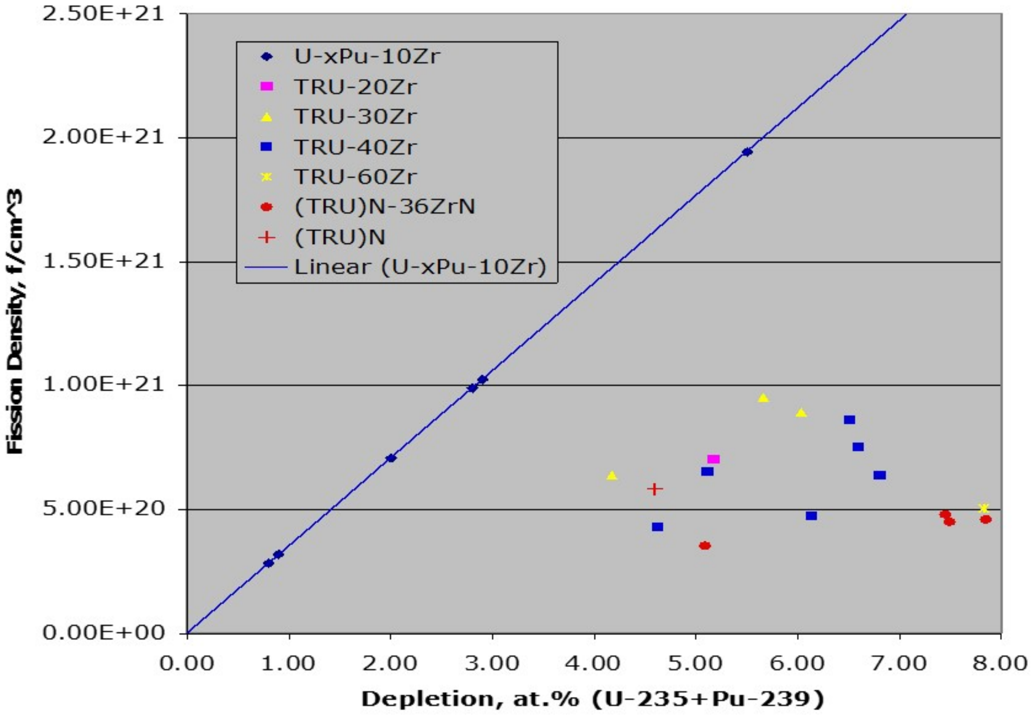
Metallic Fuel Alloys Tested in ATR

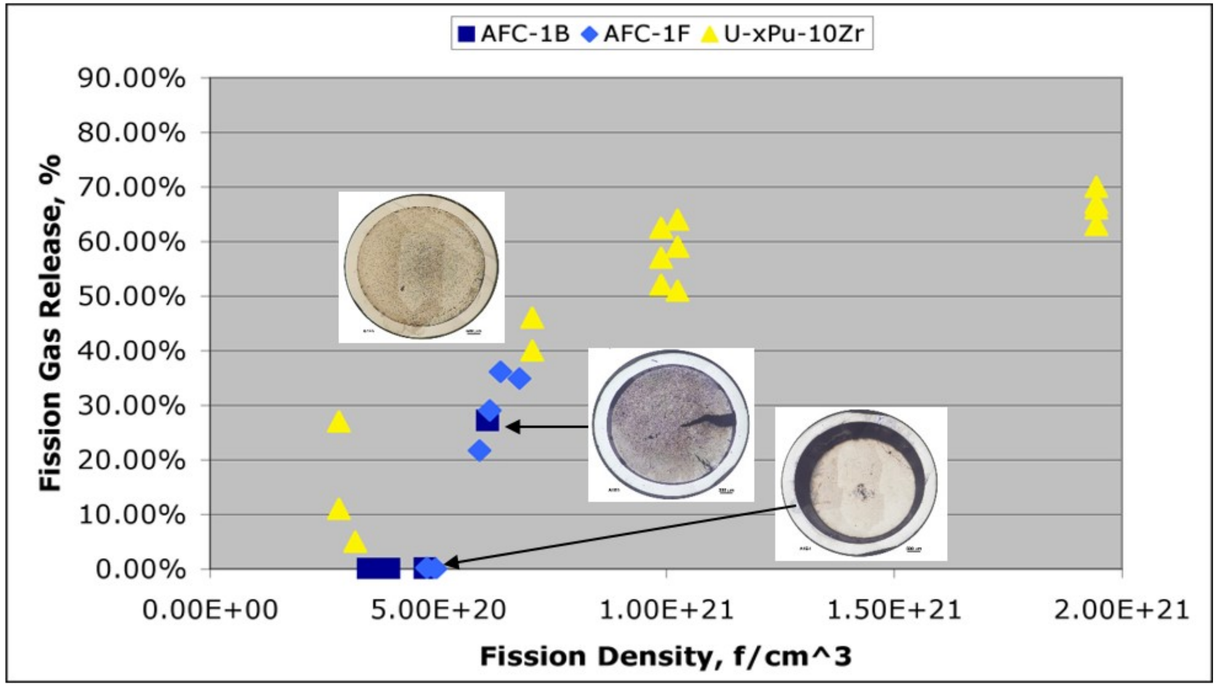
Metallic Fuel Alloy	ATR Experiment	Irradiation Time (EFPDs)	Peak LHGR (W/cm)	Peak Fission Density (fiss/cm3)	Peak Burnup (% HM)
Pu-40Zr	AFC-1B	93	300	5.26E+20	3.5
	AFC-1D	582	300	3.30E+21	**22.6**
Pu-60Zr	AFC-1B	93	300	3.51E+20	4.1
	AFC-1D	582	300	2.25E+21	**26.7**
Pu-12Am-40Zr	AFC-1B	93	300	4.27E+20	4.3
	AFC-1D	582	300	2.84E+21	**22.6**
Pu-10Np-40Zr	AFC-1G	644	300	2.91E+21	**20.3**
Pu-10Am-10Np-40Zr	AFC-1B	93	300	3.43E+20	3.5
	AFC-1D	582	300	2.24E+21	17.7
U-25Pu-3Am-2Np-40Zr	AFC-1F	94	330	5.89E+20	5.0
	AFC-1H	653	330	3.54E+21	**30.2**
U-28Pu-7Am-30Zr	AFC-1F	94	330	6.38E+20	4.6
	AFC-1H	653	330	3.96E+21	**26.1**
U-29Pu-4Am-2Np-30Zr	AFC-1F	94	330	6.38E+20	4.4
	AFC-1H	653	330	3.91E+21	**26.7**
U-34Pu-4Am-2Np-20Zr	AFC-1F	94	330	5.35E+20	2.8
	AFC-1H	653	330	3.48E+21	17.2
U-20Pu-3Am-2Np-15Zr	AFC-2A	214	350	1.33E+21	6.4
	AFC-2B	364	350	2.30E+21	10.4
U-20Pu-3Am-2Np-1.0Ln*-15Zr	AFC-2A	214	350	1.43E+21	7.3
	AFC-2B	364	350	2.43E+21	11.5
U-20Pu-3Am-2Np-1.5Ln*-15Zr	AFC-2A	214	350	1.28E+21	6.0
	AFC-2B	364	350	2.22E+21	9.6
U-30Pu-5Am-3Np-20Zr	AFC-2A	214	350	1.19E+21	7.2
	AFC-2B	364	350	2.07E+21	11.5
U-30Pu-5Am-3Np-1.0Ln*-20Zr	AFC-2A	214	350	1.29E+21	7.0
	AFC-2B	364	350	2.21E+21	11.5
U-30Pu-5Am-3Np-1.5Ln*-20Zr	AFC-2A	214	350	1.37E+21	6.8
	AFC-2B	364	350	2.49E+21	11.1
U-20Pu-10Zr	AFC-2E	483	350	3.25E+21	11.3
U-20Pu-3Am-2Np-10Zr	AFC-2E	483	350	3.09E+21	11.6

*Ln=6% La, 16% Pr, 25% Ce, 53% Nd

Irradiation Performance Results

- **Consistent Performance for Diverse Spectrum of Metallic Fuel Alloys**
  - Performance correlates with fission density (<u>not</u> burnup in %HM)
  - Lower density alloys offer potential for ultra-high burnups

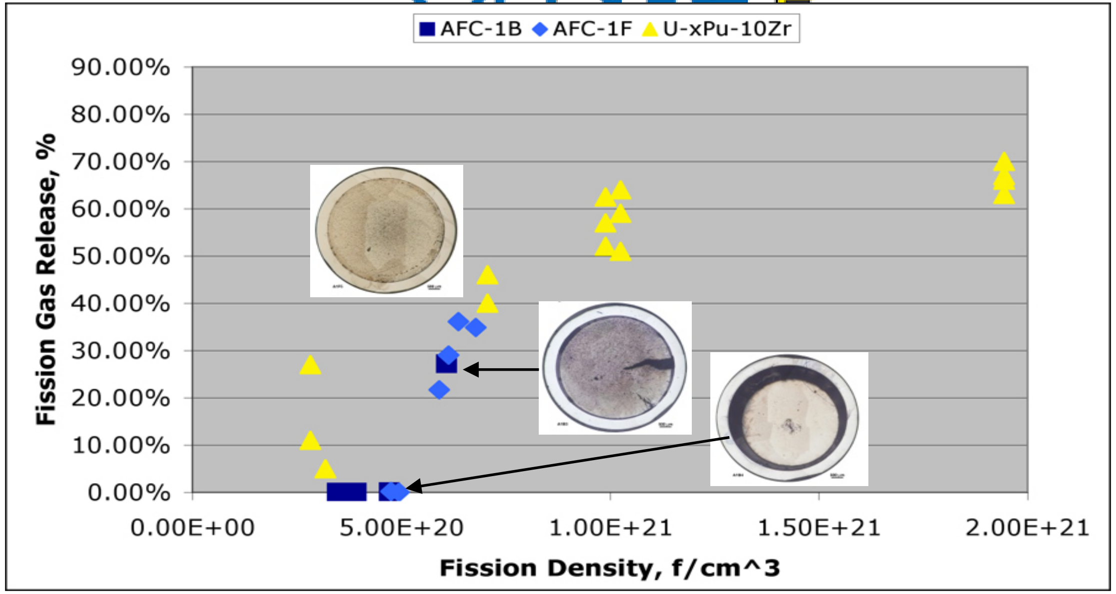




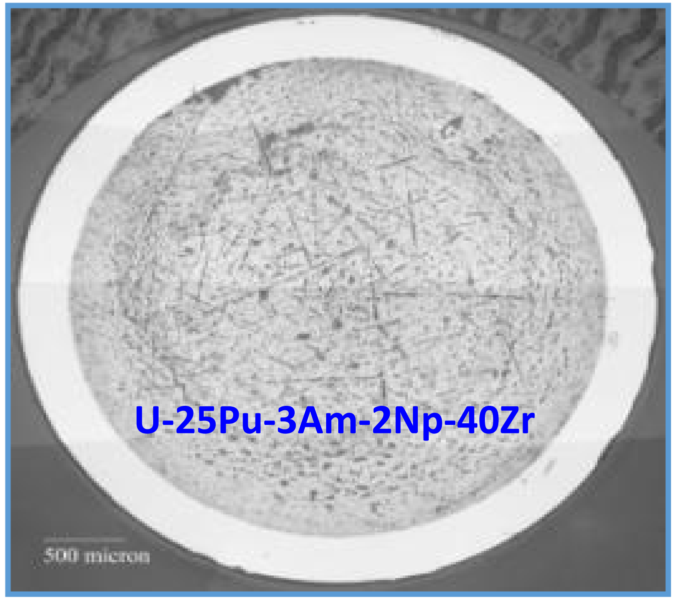
Performance of Metallic Fuels

- **U-Pu-Am-Np-Zr-RE alloys**
  - 10 < Zr < 40% (60%)
  - Pu < 34% (60%)
  - Am < 12%
  - Np < 10%
  - RE < 1.5%

  (RE = La, Pr, Ce, Nd)
- **Performance Characteristics**
  - Fuel Swelling
  - Fission Gas & He Release
  - Fuel-cladding Interaction



Irradiation performance of AFC metallic fuels has been shown to be "typical" of historic understanding for wide variations of U, Pu, Zr, & MA contents.



Future Direction

Advanced Metallic Fuel Concept

Innovations proposed to deliver multifold advances in fuel reliability and performance, actinide utilization, and fabrication efficiency, while retaining all the historic benefits of the metallic fuel system.

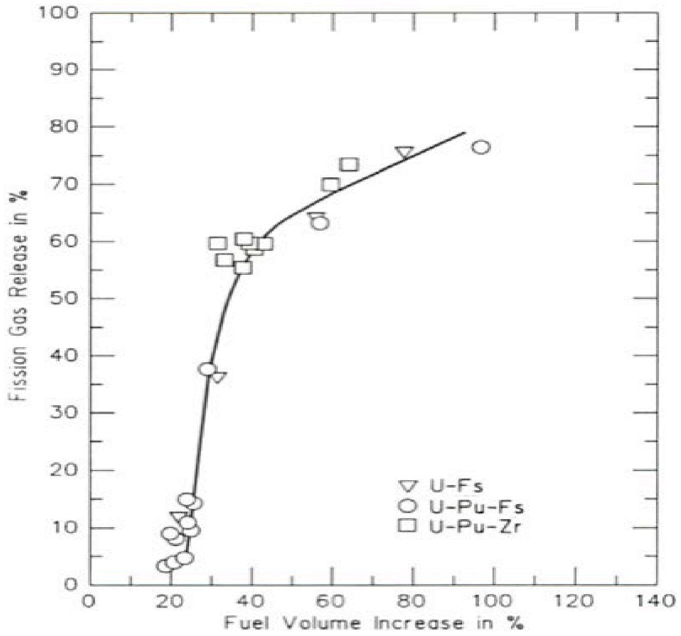
- **Proposed innovations:**
  1. **Decreased fuel smeared density (w/annular fuel)**
  2. Coating or liner on cladding inner diameter
  3. Vented fuel pin
  4. Optimization of alloy base
  5. **Targeted fuel alloy additions**
  6. Advanced fabrication method (i.e., extrusion)
  7. Compatibility with electrochemical recycling

Development of the "Advanced Metallic Fuel Concept" initiated as award under **innovative fuel competition**, but has become main line of metallic fuel development.

Annular Fuel with Decreased Smeared Density

■ Decreased fuel smeared density

- Reduction of fuel smeared density (<75%) will allow accommodation of swelling to ultra-high burnups without FCMI

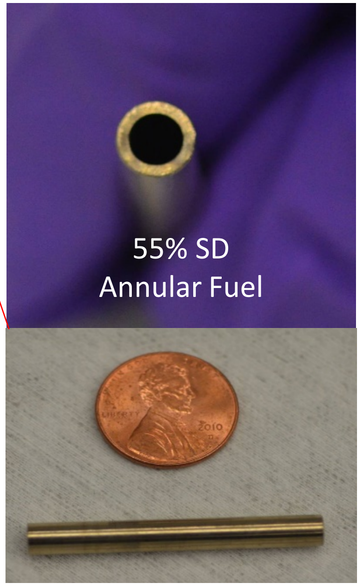


- Fuel smeared density of **55%** needed to achieve 40% burnup

■ Annular fuel form

- Annular metallic fuel in contact with cladding may be more geometrically stable
- Could eliminate need for sodium bond

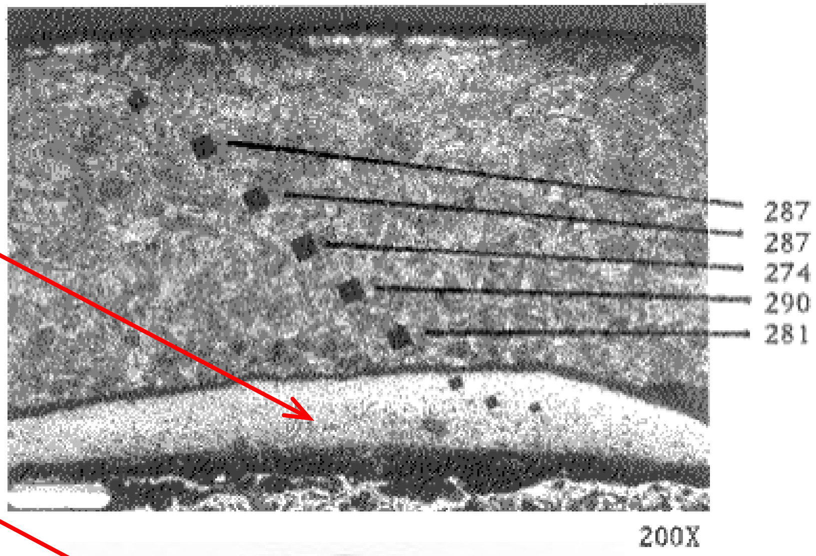
Fabrication by casting and machining for testing purposes, investigating extrusion



Will fuel swell into central annulus, or strain cladding?

Targeted Fuel Alloy Additions

- **Fuel-cladding Chemical Interaction**
  - Lanthanide fission products (La, Ce, Pr, Nd) transport thru fuel and react with SS cladding
  - Forms brittle interaction product
  - Results in cladding wastage
  - Low melting (650-750°C); limits transient overpower
  - Cladding coating/liners as diffusion barriers could mitigate reaction,



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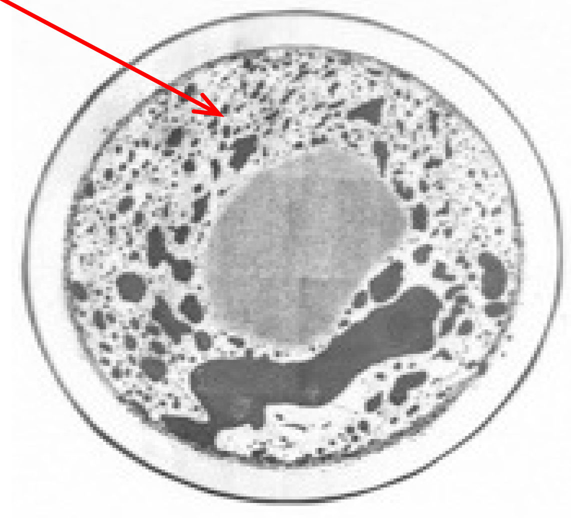
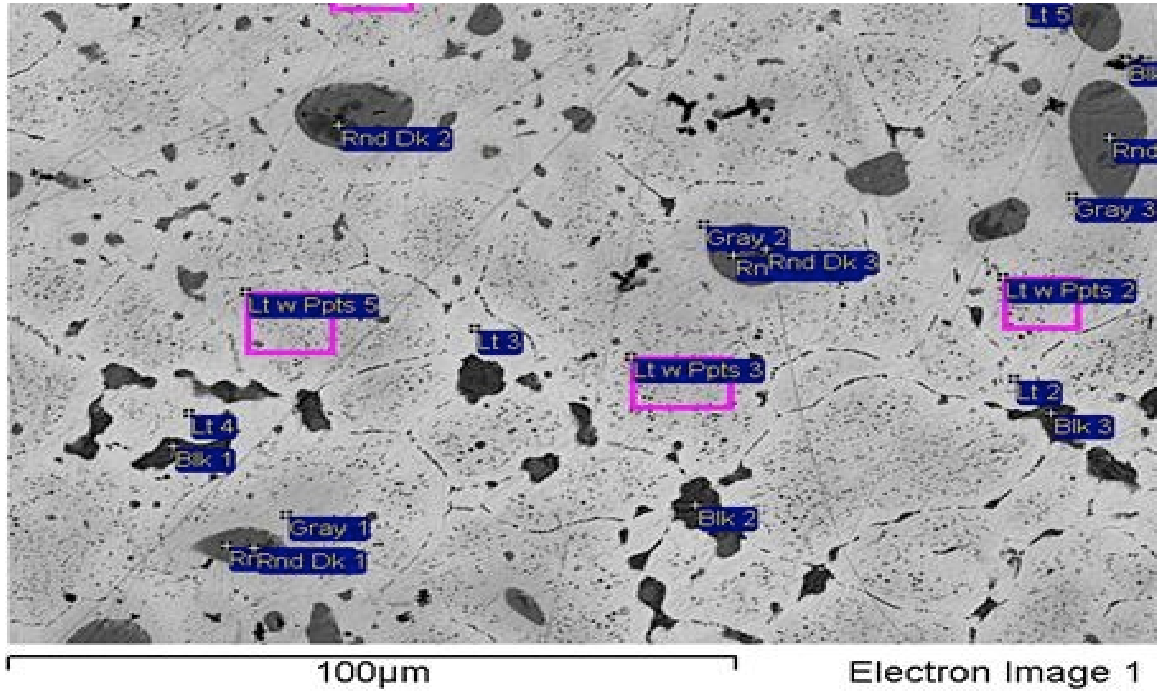


- **Targeted fuel alloy additions**
  - Minor alloying additions that chemically bind with lanthanide fission products could immobilize them in the fuel matrix, reducing or eliminating FCCI.

Immobilizing Lanthanide Fission Products

- Lanthanide fission products migrate to fuel surface and interact with cladding, the primary source of FCCI
- Use minor alloy additions to chemically bind the lanthanides and immobilize in fuel matrix
- Double Benefit:
  1) Homogenize/stabilize Ln's carried over from recycle
  2) Mitigate FCCI during irradiation

R.D. Mariani, D.L. Porter, T.P. O'Holleran, S.L. Hayes, J.R. Kennedy, *Journal of Nuclear Materials*, **419** (2011) 263.

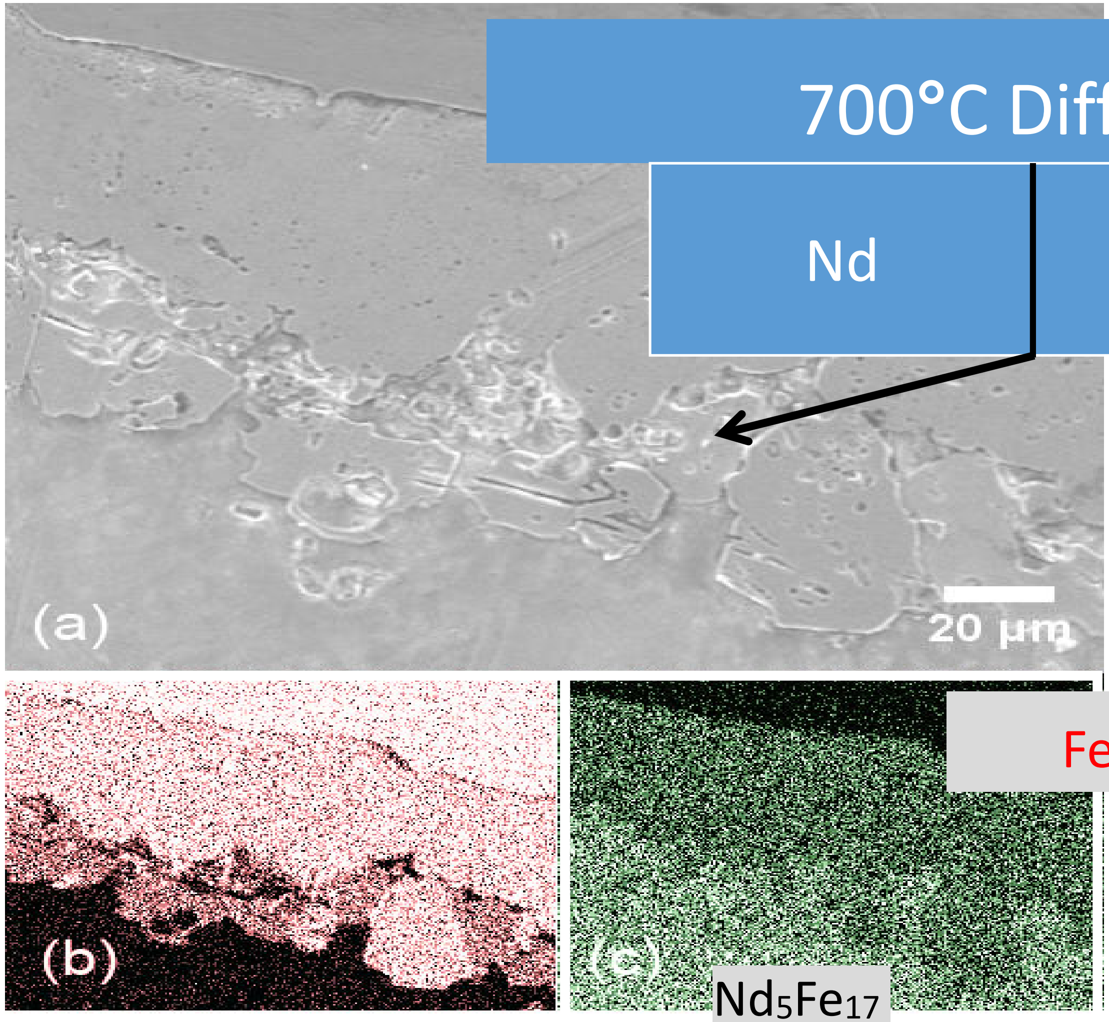


Back scattered electron image of
U-15Zr-3.86Pd-4.3Ln
(Ln = 53Nd-25Ce-16Pr-6La, in wt%)

Palladium (Pd) combines with the lanthanides, as expected from analysis of thermodynamic and PIE data.

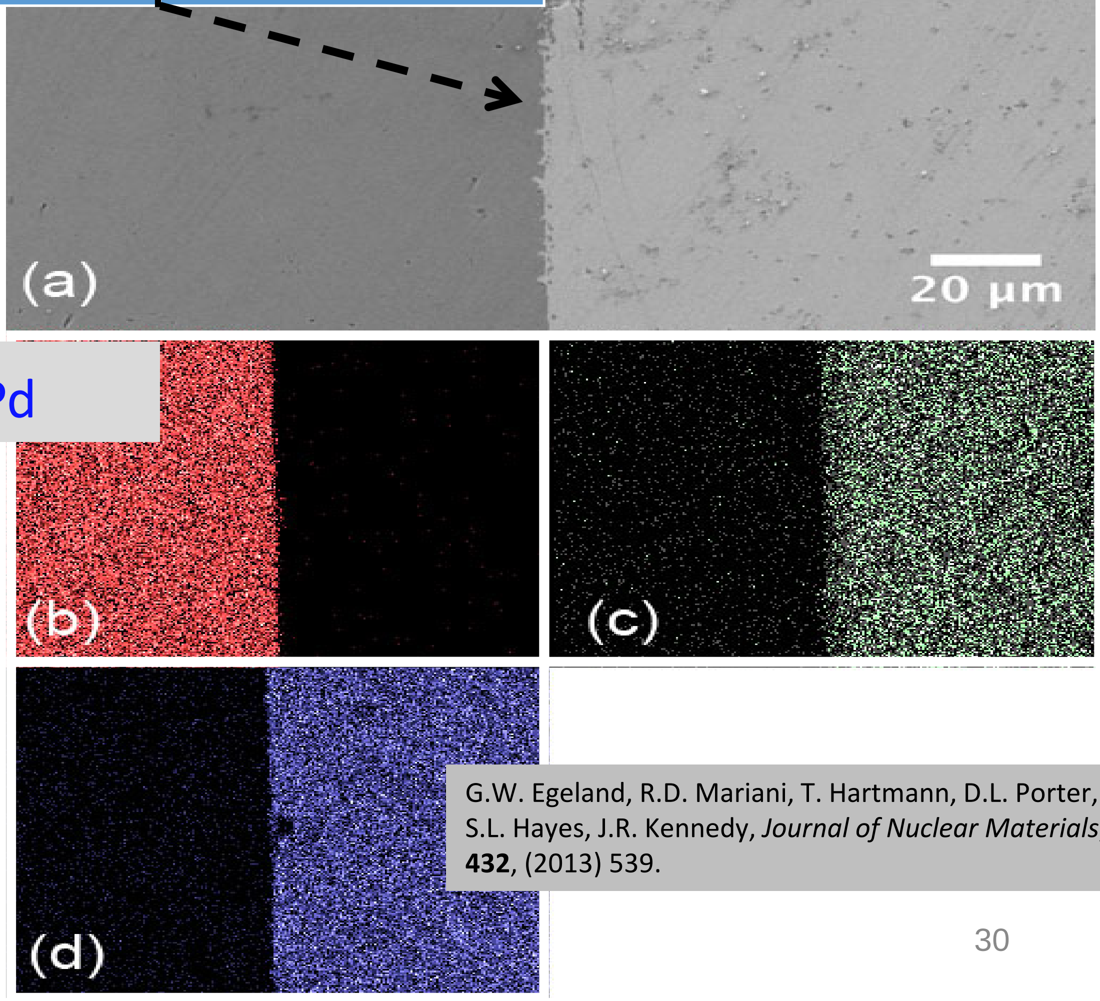
700°C Diffusion Couples

Nd | Fe | Nd/Pd



Nd$_5$Fe$_{17}$
Nd$_2$Fe$_{17}$

Fe, Nd, Pd



At 700°C, the Fe-Nd interface experienced liquefaction, while the Fe-(Nd/Pd) interface experienced no reaction.

G.W. Egeland, R.D. Mariani, T. Hartmann, D.L. Porter, S.L. Hayes, J.R. Kennedy, *Journal of Nuclear Materials*, **432**, (2013) 539.

Advanced Metallic Fuels Concept:
Irradiation Testing Strategy

Fuel	Fuel Alloy	Geometry & Bond	Smear Density	Alloy Additions	Burnup (HM)	Cladding
U U+Pu U+Pu+MA	Zr Ta-Zr Mo-Ti-Zr	Solid w/Na Annular w/He	55% 65% 75%	none Pd In Ga other combos	8-10% 15-20% 30-40%	HT-9 liner coating

- Short-term irradiations: early PIE to show feasibility
- Long-term irradiations: demonstrate ultra-high burnup
- Early materials testing of lined/coated cladding for incorporation in subsequent short-term irradiations
- Investigate alloy additions to immobilize lanthanide fission products

Summary & Conclusions

Summary & Conclusions (Metallic Fuels with MA Additions)

- **Fabrication**
  - Issue of Am volatility during casting has been resolved at bench-scale using surrogate systems; validation testing with Am underway
- **Irradiation Performance**
  - Wide spectrum of U-Pu-Am-Np-Zr fuel alloys have been irradiated in ATR
  - Performance is good, “typical” of historic metallic fuel behavior
  - **Comparison Report** (FY17) will validate ATR Cd-shrouded test results vs. data from EBR-II, FFTF, and Phénix
- **Future Direction**
  - Development of the “Advanced Metallic Fuel Concept”
    - Additives for Ln FP stabilization and immobilization
    - Cladding coatings/liners
    - Low SD annular fuel, fabrication by extrusion
  - Goal: Demonstrate reliable performance to ultra-high burnups (30-40%)

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Upcoming webinars

21 September 2017	Energy Conversion	Dr. Richard Stainsby, NNL, UK
25 October 2017	Economics of the Nuclear Fuel Cycle	Dr. Geoffrey Rothwell, NEA/OECD
29 November 2017	Feedback from Phenix and SuperPhenix	Dr. Joel Guidez, CEA, France